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U. Schollwöck J. Richter D.J.J. Farnell R.F. Bishop (Eds.)

Quantum Magnetism



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Preface

Putting the quantum into magnetism might, at first sight, seem like stating the obvious; the exchange interactions leading to collective magnetic behavior are, after all, a pure quantum effect. Yet, for many phenomena in magnetism this underlying quantum nature may be safely ignored at least on the qualitative level. The investigation of magnetic systems where quantum effects play a dominant role and have to be accounted for in detail has, over the last decades, evolved to be a field of very active research. On the experimental side, major boosts have come from the discovery of high-temperature superconductivity in the mid-eighties and the increasing ability of solid state chemists to fashion magnetic systems of restricted dimensionality. While hightemperature superconductivity has raised the question of the link between the mechanism of superconductivity in the cuprates and spin fluctuations and magnetic order in one- and two-dimensional spin-1/2 antiferromagnets, the new magnetic materials have exhibited a wealth of new quantum phenomena of interest in their own. In one-dimensional systems, the universal paradigm of Luttinger liquid behavior has come to the center of interest; in all restricted geometries, the interplay of low dimension, competing interactions and strong quantum fluctuations generates, beyond the usual long range ordered states, a wealth of new states of condensed matter, such as valence bond solids, magnetic plateaux, spin liquid states or spin-Peierls states, to name but a few.

The idea for this book arose during a Hereaus seminar on "Quantum Magnetism: Microscopic Techniques For Novel States of Matter" back in 2002, where it was realized that a set of extensive tutorial reviews would address the needs of both postgraduate students and researchers alike and fill a longstanding gap in the literature.

The first three chapters set out to give an account of conceptual problems and insights related to classes of systems, namely one-dimensional (Mikeska and Kolezhuk), two-dimensional (Richter, Schulenburg and Honecker) and molecular (Schnack) magnets.

The following five chapters are intended to introduce to methods used in the field of quantum magnetism, both for independent reading as well as a backup for the first chapters: this includes time-honored spin wave analysis (Ivanov and Sen), exact diagonalization (Laflorencie and Poilblanc), quantum field theory (Cabra and Pujol), coupled cluster methods (Farnell and Bishop) and the Bethe ansatz (Klümper).

To close, a more unified point of view is presented in a theoretical chapter on quantum phase transitions (Sachdev) and an experimentally oriented contribution (Lemmens and Millet), putting the wealth of phenomena into the solid state physics context of spins, orbitals and lattice topology.

Aachen, Magdeburg, Liverpool, Manchester March 2004

Ulrich Schollwöck Johannes Richter Damian Farnell Ray Bishop

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1 One-Dimensional Magnetism

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Abstract. We present an up-to-date survey of theoretical concepts and results in the field of one-dimensional magnetism and of their relevance to experiments and real materials. Main emphasis of the chapter is on quantum phenomena in models of localized spins with isotropic exchange and additional interactions from anisotropy and external magnetic fields.

Three sections deal with the main classes of model systems for 1D quantum magnetism: S = 1/2 chains, spin chains with S > 1/2, and S = 1/2 Heisenberg ladders. We discuss the variation of physical properties and elementary excitation spectra with a large number of model parameters such as magnetic field, anisotropy, alternation, next-nearest neighbour exchange etc. We describe the related quantum phase diagrams, which include some exotic phases of frustrated chains discovered during the last decade.

A section on modified spin chains and ladders deals in particular with models including higher-order exchange interactions (ring exchange for S=1/2 and biquadratic exchange for S=1 systems), with spin-orbital models and mixed spin (ferrimagnetic) chains.

The final section is devoted to gapped one-dimensional spin systems in high magnetic field. It describes such phenomena as magnetization plateaus and cusp singularities, the emergence of a critical phase when the excitation gap is closed by the applied field, and field-induced ordering due to weak three-dimensional coupling or anisotropy. We discuss peculiarities of the dynamical spin response in the critical and ordered phases.

1.1 Introduction

The field of low-dimensional magnetism can be traced back some 75 years ago: In 1925 Ernst Ising followed a suggestion of his academic teacher Lenz and investigated the one-dimensional (1D) version of the model which is now well known under his name [1] in an effort to provide a microscopic justification for Weiss' molecular field theory of cooperative behavior in magnets; in 1931 Hans Bethe wrote his famous paper entitled 'Zur Theorie der Metalle. I. Eigenwerte und Eigenfunktionen der linearen Atomkette' [2] describing the 'Bethe ansatz' method to find the exact quantum mechanical ground state of the antiferromagnetic Heisenberg model [3], for the 1D case. Both papers were actually not to the complete satisfaction of their authors: The 1D Ising model failed to show any spontaneous order whereas Bethe did not live up to the expectation expressed in the last sentence of his text: 'In einer folgenden Arbeit soll die Methode auf räumliche Gitter ausgedehnt ... werden' ('in a subsequent publication the method is to be extended to cover 3D lattices').

In spite of this not very promising beginning, the field of low-dimensional magnetism developed into one of the most active areas of today's solid state physics. For the first 40 years this was an exclusively theoretical field. Theorists were attracted by the chance to find interesting exact results without having to deal with the hopelessly complicated case of models in 3D. They succeeded in extending the solution of Ising's (classical) model to 2D (which, as Onsager showed, *did* exhibit spontaneous order) and in calculating excitation energies, correlation functions and thermal properties for the quantum mechanical 1D Heisenberg model and (some of) its anisotropic generalizations. In another line of research theorists established the intimate connection between classical models in 2D and quantum mechanical models in 1D [4,5]. An important characteristic of low-dimensional magnets is the absence of long range order in models with a continuous symmetry at any finite temperature as stated in the theorem of Mermin and Wagner [6], and sometimes even the absence of long range order in the ground state [7].

It was only around 1970 when it became clear that the one- and twodimensional models of interest to theoretical physicists might also be relevant for real materials which could be found in nature or synthesized by ingenious crystal growers. One of the classical examples are the early neutron scattering experiments on TMMC [8]. Actually, magnets in restricted dimensions have a natural realization since they exist as real bulk crystals with, however, exchange interactions which lead to magnetic coupling much stronger in one or two spatial directions than in the remaining ones. Thus, in contrast to 2D lattices (on surfaces) and 2D electron gases (in quantum wells) low D magnets often have all the advantages of bulk materials in providing sufficient intensity for experiments investigating thermal properties (e.g. specific heat), as well as dynamic properties (in particular quantum excitations) by e.g. neutron scattering.

The interest in low-dimensional, in particular one-dimensional magnets developed into a field of its own because these materials provide a unique possibility to study ground and excited states of quantum models, possible new phases of matter and the interplay of quantum fluctuations and thermal fluctuations. In the course of three decades interest developed from classical to quantum mechanics, from linear to nonlinear excitations. From the theoretical point of view the field is extremely broad and provides a playground for a large variety of methods including exact solutions (using the Bethe ansatz and the mapping to fermion systems), quantum field theoretic approaches (conformal invariance, bosonization and the semiclassical nonlinear σ -model (NLSM)), methods of many-body theory (using e.g. Schwinger bosons and hard core bosons), perturbational approaches (in particular high order series expansions) and finally a large variety of numerical methods such as exact diagonalization (mainly using the Lanczos algorithm for the lowest eigenvalues but also full diagonalization), density matrix renormalization group (DMRG) and Quantum Monte Carlo (QMC) calculations.

The field of one-dimensional magnets is characterized by strong interactions between theoretical and experimental research: In the early eighties, the seminal papers of Faddeev and Takhtajan [9] who revealed the spinon nature of the excitation spectrum of the spin- $\frac{1}{2}$ antiferromagnetic chain, and Haldane [10] who discovered the principal difference between chains of integer and half-integer spins caused an upsurge of interest in new quasi-1D magnetic materials, which substantially advanced the corresponding technology. On the other hand, in the mid eighties, when the interest in the field seemed to go down, a new boost came from the discovery of high temperature superconductors which turned out to be intimately connected to the strong magnetic fluctuations which are possible in low D materials. At about the same time a new boost for experimental investigations came from the new energy range opened up for neutron scattering experiments by spallation sources. Further progress of material science triggered interest in spin ladders, objects staying "in between" one and two dimensions [11]. At present many of the phenomena which turned up in the last decade remain unexplained and it seems safe to say that low-dimensional magnetism will be an active area of research good for surprises in many years to come.

It is thus clear that the field of 1D magnetism is vast and developing rapidly. New phenomena are found and new materials appear at a rate which makes difficult to deliver a survey which would be to any extent complete. Our aim in this chapter will be to give the reader a proper mixture of standard results and of developing topics which could serve as an advanced introduction and stimulate further reading. We try to avoid the overlap with already existing excellent textbooks on the subject [12–14], which we recommend as complementary reading. In this chapter we will therefore review a number of issues which are characteristic for new phenomena specific for one-dimensional magnets, concentrating more on principles and a unifying picture than on details.

Although classical models played an important role in the early stage of 1D magnetism, emphasis today is (and will be in this chapter) on models where quantum effects are essential. This is also reflected on the material side: Most investigations concentrate on compounds with either Cu^{2+} -ions which realize spin- $\frac{1}{2}$ or Ni²⁺-ions which realize spin 1. Among the spin- $\frac{1}{2}$ chain-like materials, $CuCl_2 \cdot 2NC_5H_5$ (Copperpyridinchloride = CPC) is important as the first quantum chain which was investigated experimentally [15]. Among today's best realizations of the spin- $\frac{1}{2}$ antiferromagnetic Heisenberg model we mention KCuF₃ and Sr₂CuO₃. Another quasi-1D spin- $\frac{1}{2}$ antiferromagnet which is widely investigated is CuGeO₃ since it was identified in 1992 as the first inorganic spin-Peierls material [16]. The prototype of ladder materials with spin- $\frac{1}{2}$ is SrCu₂O₃; generally, the SrCuO materials realize not only chains and two-leg ladders but also chains with competing interactions and ladders with more than two legs. Of particular interest is the material

 $Sr_{14}Cu_{24}O_{41}$ which can be easily synthesized and consists of both CuO_2 zigzag chains and Cu_2O_3 ladders. A different way to realize spin- $\frac{1}{2}$ is in chains with Co^{++} -ions which are well described by a pseudospin $\frac{1}{2}$. The free Coion has spin $\frac{3}{2}$, but the splitting in the crystal surrounding is so large that for the interest of 1D magnetism only the low-lying doublet has to be taken into account (and then has a strong tendency to Ising-like anisotropy, e.g. in $CsCoCl_3$). Among the spin-1 chain-like materials, $CsNiF_3$ was important in the classical era as a ferromagnetic xy-like chain which allowed to demonstrate magnetic solitons; for the quantum S=1 chain and in particular the Haldane gap first $(Ni(C_2H_8N_2)_2NO_2(ClO_4) = NENP)$ and more recently $(Ni(C_5H_{14}N_2)_2N_3(PF_6) = NDMAP)$ are the most important compounds. It should be realized that the anisotropy is usually very small in spin- $\frac{1}{2}$ chain materials with Cu^{2+} -ions whereas S=1 chains with Ni²⁺-ions, due to spinorbit effects, so far are typically anisotropic in spin space. An increasing number of theoretical approaches and some materials exist for alternating spin-1 and $\frac{1}{2}$ ferrimagnetic chains and for chains with V²⁺-ions with spin $\frac{3}{2}$ and Fe²⁺-ions with spin 2, however, to a large degree this is a field for the future. Tables listing compounds which may serve as 1D magnets can be found in earlier reviews [17, 18]; for a discussion of the current experimental situation, see the Chapter by Lemmens and Millet in this book.

We will limit ourselves mostly to models of localized spins S_n with an exchange interaction energy between pairs, $J_{n,m}$ ($S_n \cdot S_m$) (Heisenberg model), to be supplemented by terms describing (spin and lattice) anisotropies, external fields etc., when necessary. Whereas for real materials the coupling between the chains forming the 1D system and in particular the transition from 1D to 2D systems with increasing interchain coupling is of considerable interest, we will in this chapter consider only the weak coupling limit and exclude phase transitions into phases beyond a strictly 1D character. With this aim in mind, the most important single model probably is the S = 1/2 ($S^{\alpha} = \frac{1}{2}\sigma^{\alpha}$) XXZ model in 1D

$$\mathcal{H} = J \sum_{n} \left\{ \frac{1}{2} \left(S_n^+ S_{n+1}^- + S_n^- S_{n+1}^+ \right) + \Delta S_n^z S_{n+1}^z \right\}.$$
 (1.1)

We have decomposed the scalar product into longitudinal and transverse terms

$$\boldsymbol{S}_1 \cdot \boldsymbol{S}_2 = S_1^z S_2^z + \frac{1}{2} \left(S_1^+ S_2^- + S_1^- S_2^+ \right)$$
(1.2)

 $(S^{\pm} = S^x \pm iS^y)$ and we note that the effect of the transverse part for S = 1/2 is nothing but to interchange up and down spins, $|\uparrow\downarrow\rangle \iff |\downarrow\uparrow\rangle$ $\uparrow\rangle$ (apart from a factor of $\frac{1}{2}$). The Hamiltonian of (1.1), in particular for antiferromagnetic coupling, is one of the important paradigms of both manybody solid state physics and field theory. Important for the discussion of its properties is the presence of symmetries leading to good quantum numbers such as wave vector \boldsymbol{q} (translation), S_{tot}^z (rotation about z-axis), S_{tot} (general rotations, for $|\Delta| = 1$) and parity (spin inversion).

This chapter will present theoretical concepts and results, which, however, are intimately related to experimental results. The most important link between theory and experiment are the spin correlation functions or resp. dynamical structure factors which for a spin chain are defined as follows:

$$S^{\alpha,\alpha}(q,\omega) = \sum_{n} \int dt e^{i(qn-\omega t)} \langle S_n^{\alpha}(t) S_0^{\alpha}(t=0) \rangle$$
(1.3)

$$S^{\alpha,\alpha}(q) = \sum_{n} e^{iqn} \langle S_n^{\alpha} S_0^{\alpha} \rangle = \frac{1}{2\pi} \int d\omega S^{\alpha,\alpha}(q,\omega).$$
(1.4)

 $S(q,\omega)$ determines the cross section for scattering experiments as well as line shapes in NMR and ESR experiments. A useful sum rule is the total intensity, obtained by integrating $S(q,\omega)$ over frequency and wave vector,

$$\frac{1}{4\pi^2} \int d\omega S^{\alpha,\alpha}(q,\omega) = \frac{1}{2\pi} \int dq S^{\alpha,\alpha}(q) = \langle (S_0^{\alpha})^2 \rangle$$
(1.5)

which is simply equal to $\frac{1}{3}S(S+1)$ in the isotropic case.

1.2 $S = \frac{1}{2}$ Heisenberg Chain

The $S = \frac{1}{2}$ XXZ Heisenberg chain as defined in (1.1) (XXZ model) is both an important model to describe real materials and at the same time the most important paradigm of low-dimensional quantum magnetism: it allows to introduce many of the scenarios which will reappear later in this chapter: broken symmetry, the gapless Luttinger liquid, the Kosterlitz-Thouless phase transition, gapped and gapless excitation continua. The XXZ model has played an essential role in the development of exact solutions in 1D magnetism, in particular of the Bethe ansatz technique. Whereas more details on exact solutions can be found in the chapter by Klümper, we will adopt in this section a more phenomenological point of view and present a short survey of the basic properties of the XXZ model, supplemented by an external magnetic field and by some remarks for the more general XYZ model,

$$\mathcal{H} = J \sum_{n} \left\{ (1+\gamma) S_{n}^{x} S_{n+1}^{x} + (1-\gamma) S_{n}^{y} S_{n+1}^{y} + \Delta S_{n}^{z} S_{n+1}^{z} \right\} -g \mu_{B} \boldsymbol{H} \sum_{n} \boldsymbol{S}_{n}$$
(1.6)

as well as by further typical additional terms such as next-nearest neighbor (NNN) interactions, alternation etc. We will use a representation with positive exchange constant J > 0 and we will frequently set J to unity, using it as the energy scale.

1.2.1 Ferromagnetic Phase

For $\Delta < -1$ the XXZ chain is in the ferromagnetic Ising phase: the ground state is the saturated state with all spins aligned in either z or -z direction, i.e., the classical ground state with magnetization $S_{tot}^z = \pm \frac{1}{2}N$, where N is the number of sites. This is thus a phase with broken symmetry: the ground state does not exhibit the discrete symmetry of spin reflection $S^z \to -S^z$, under which the Hamiltonian is invariant. In the limit $\Delta = -1$ this symmetry is enlarged to the full rotational symmetry of the isotropic ferromagnet.

When an external magnetic field in z-direction is considered, the Zeeman term as included in (1.6), $\mathcal{H}_Z = -g\mu_B H \sum_n S_n^z$, has to be added to the Hamiltonian. Since \mathcal{H}_{XXZ} commutes with the total spin component S_{tot}^z , the external magnetic field results in an additional energy contribution $-g\mu_B H S_{\text{tot}}^z$ without affecting the wave functions. The symmetry under spin reflection is lifted and the saturated ground state is stabilized.

The low-lying excited states in the ferromagnetic phase are magnons with the total spin quantum number $S_{tot}^z = \frac{1}{2}N - 1$ and the dispersion law (valid for general spin S)

$$\epsilon(q) = 2JS \left(1 - \cos q - (\Delta + 1)\right) + 2g\mu_B HS.$$
 (1.7)

These states are *exact* eigenstates of the XXZ Hamiltonian. In zero field the excitation spectrum has a gap at q = 0 of magnitude $|\Delta| - 1$ for $\Delta < -1$. At $\Delta = -1$ the discrete symmetry of spin reflection generalizes to the continuous rotational symmetry and the spectrum becomes gapless. This is a consequence of Goldstone's theorem: the breaking of a continuous symmetry in the ground state results in the emergence of a gapless excitation mode. Whereas the ground state exhibits long range order, the large phase space available to the low-lying excitations in 1D leads to exponential decay of correlations at arbitrarily small finite temperatures following the theorem of Mermin and Wagner [6].

Eigenstates in the subspace with two spin deviations, $S_{tot}^z = N-2$ can be found exactly by solving the scattering problem of two magnons. This results in the existence of bound states below the two magnon continuum (for a review see [19]) which are related to the concept of domain walls: In general two spin deviations correspond to 4 domains walls (4 broken bonds). However, two spin deviations on neighboring sites correspond to 2 domain walls and require intermediate states with a larger number of walls, i.e. higher energy, to propagate. They therefore have lower energy and survive as a bound state. General ferromagnetic domain wall states are formed for smaller values of S_{tot}^z The ferromagnetic one-domain-wall states can be stabilized by boundary fields opposite to each other. They contain admixtures of states with a larger number of walls, but for $\Delta < -1$ they remain localized owing to conservation of S_{tot}^z [20]. A remarkable exact result is that the lowest magnon energy is not affected by the presence of a domain wall [21]: the excitation energy is $|\Delta| - 1$ both for the uniform ground state and for the one domain wall states. We mention two trivial, but interesting consequences of (1.7) which can be generalized to any XXZ-type Hamiltonian conserving S_{tot}^z :

(i) For sufficiently strong external magnetic field the classical saturated state is forced to be the ground state for arbitrary value of Δ and the lowest excitations are exactly known. If the necessary magnetic fields are within experimentally accessible range, this can be used for an experimental determination of the exchange constants from the magnon dispersion (an example in 2D are recent neutron scattering experiments on Cs₂CuCl₄ [22]).

(ii) The ferromagnetic ground state becomes unstable when the lowest spin wave frequency becomes negative. This allows to determine e.g. the boundary of the ferromagnetic phase for $\Delta > -1$ in an external field as $H = H_c$ with $g\mu_B H_c = \Delta + 1$.

1.2.2 Néel Phase

For $\Delta > +1$ the XXZ chain is in the antiferromagnetic Ising or Néel phase with, in the thermodynamic limit, broken symmetry and one from 2 degenerate ground states, the S = 1/2 remnants of the classical Néel states. The spatial period is 2a, and states are described in the reduced Brillouin zone with wave vectors $0 \le q \le \pi/a$. The ground states have $S_{tot}^z = 0$, but finite sublattice magnetization

$$N^{z} = \sum_{n} (-1)^{n} S_{n}^{z}.$$
 (1.8)

and long range order in the corresponding correlation function. In contrast to the ferromagnet, however, quantum fluctuations prevent the order from being complete since the sublattice magnetization does not commute with the XXZ Hamiltonian. For periodic boundary conditions and large but finite N (as is the situation in numerical approaches), the two ground states mix with energy separation $\propto \exp(-\text{const} \times N)$ (for $N \to \infty$). Then invariance under translation by the original lattice constant a is restored and the original Brillouin zone, $0 \le q \le 2\pi/a$, can be used.

The elementary excitations in the antiferromagnetic Ising phase are described most clearly close to the Ising limit $\Delta \to \infty$ starting from one of the two ideal Néel states: Turning around one spin breaks two bonds and leads to a state with energy Δ , degenerate with all states resulting from turning around an arbitrary number of subsequent spins. These states have $S_{\text{tot}}^z = \pm 1$, resp. 0 for an odd, resp. even number of turned spins. They are appropriately called two-domain wall states since each of the two broken bonds mediates between two different Néel states. The total number of these states is N(N-1): there are $N^2/4$ states with $S_{tot}^z = +1$ and $S_{tot}^z = -1$ (number of turned spins odd) and $N^2/2 - N$ states with $S_{tot}^z = 0$ (number of turned spins even). These states are no more eigenstates when Δ^{-1} is finite, but for $\Delta^{-1} \ll 1$ they can be dealt with in perturbation theory, leading to the excitation spectrum in the first order in $1/\Delta$ [23]

$$\omega(q,k) = \Delta + 2\,\cos q \cos 2\Phi \tag{1.9}$$

$$=\epsilon(\frac{q}{2}+\Phi)+\epsilon(\frac{q}{2}-\Phi) \tag{1.10}$$

with

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$$\epsilon(k) = \frac{1}{2}\Delta + \cos 2k. \tag{1.11}$$

q is the total momentum and takes the values $q = 2\pi l/N$ with l = 1, 2...N/2, Φ is the wave vector related to the superposition of domain walls with different distances and for $S_{\text{tot}}^z = \pm 1$ takes values $\Phi = m\pi/(N+2)$ with m = 1, 2...N/2. Φ is essentially a relative momentum, however, the precise values reflect the fact that the two domain walls cannot penetrate each other upon propagation. The formulation of (1.10) makes clear that the excitation spectrum is composed of two entities, domain walls with dispersion given by (1.11) which propagate independently with momenta k_1, k_2 . These propagating domain walls were described first by Villain [24], marking the first emergence of magnetic (quantum) solitons. A single domain wall is obtained as eigenstate for an odd number of sites, requiring a minimum of one domain wall, and therefore has spin projection $S_{\text{tot}}^z = \pm \frac{1}{2}$. A domain wall can hop by two sites due to the transverse interaction whence the argument 2k in the dispersion.



Fig. 1.1. Domain wall picture of elementary excitations in the Néel phase of the XXZ $S = \frac{1}{2}$ chain: (a) acting with S_n^- on the Néel state, one obtains a "magnon" which decays into two domain walls (DW) under repeated action of the Hamiltonian; (b) the two-DW continuum in the first order in Δ , according to (1.9)

Figure 1.1 shows the basic states of this picture and the related dispersions. The two domain wall dispersion of (1.9) is shown in the reduced Brillouin zone; the full BZ can, however, also be used since the corresponding wave functions (for periodic boundary conditions) are also eigenstates of the translation by one site. The elementary excitations in the antiferromagnetic Ising phase thus form a continuum with the relative momentum of the two domain walls serving as an internal degree of freedom.

1.2.3 XY Phase

For $-1 < \Delta < +1$ and zero external field the XXZ chain is in the XY phase, characterized by uniaxial symmetry of the easy-plane type and a gapless excitation continuum. Whereas the full analysis of this phase for general Δ requires the use of powerful methods such as Bethe ansatz and bosonization, to be discussed in later chapters, an approach in somewhat simpler terms is based on the mapping of $S = \frac{1}{2}$ spin operators in 1D to spinless fermions via the nonlocal Jordan-Wigner transformation [25, 26]:

$$S_n^+ = c_n^{\dagger} \ e^{i\pi \sum_{p=1}^{n-1} c_p^{\dagger} c_p}, \qquad S_n^z = c_n^{\dagger} c_n - \frac{1}{2}.$$
(1.12)

When a fermion is present (not present) at a site *n*, the spin projection is $S_n^z = +\frac{1}{2} \left(-\frac{1}{2}\right)$. In fermion language the XXZ Hamiltonian reads

$$\mathcal{H}_{XXZ} = J \sum_{n} \left\{ \frac{1}{2} \left(c_n^{\dagger} c_{n+1} + c_{n+1}^{\dagger} c_n \right) + \Delta \left(c_n^{\dagger} c_n - \frac{1}{2} \right) \left(c_{n+1}^{\dagger} c_{n+1} - \frac{1}{2} \right) \right\} - g \mu_B H \sum_{n} \left(c_n^{\dagger} c_n - \frac{1}{2} \right)$$
(1.13)

For general Δ the XXZ chain is thus equivalent to an interacting 1D fermion system. We discuss here mainly the simplest case $\Delta = 0$ (XX model), when the fermion chain becomes noninteracting and is amenable to an exact analysis in simple terms to a rather large extent: For periodic boundary conditions the assembly of free fermions is fully described by the dispersion law in wave vector space

$$\epsilon(k) = J\cos k - g\mu_B H. \tag{1.14}$$

Each of the fermion states can be either occupied or vacant, corresponding to the dimension 2^N of the Hilbert space for N spins with $S = \frac{1}{2}$. The ground state as the state with the lowest energy has all levels with $\epsilon(k) \leq 0$ occupied: For $g\mu_B H > J$ all fermion levels are occupied (maximum positive magnetization), for $g\mu_B H < -J$ all fermion levels are vacant (maximum negative magnetization) whereas for intermediate H two Fermi points $k = \pm k_F$ exist, separating occupied and vacant levels. This is the regime of the XY phase with a ground state which is a simple Slater determinant. For H = 0, as assumed in this subsection, the Fermi wave vector is $k_F = \pi/2$ and the total ground state magnetization vanishes. Magnetic field effects will be discussed in Sect. 1.2.7.

We note that periodic boundary conditions in spin space are modified by the transformation to fermions: the boundary term in the Hamiltonian depends explicitly on the fermion number N_f and leads to different Hamiltonians for the two subspaces of even, resp. odd fermion number. For fixed fermion number this reduces to different sets of allowed fermion momenta k: If the total number of spins N is even, the allowed values of fermion momenta are given by $k_n = 2\pi I_n/N$, where the numbers I_n are integer (halfodd-integer) if the number of fermions $N_f = S_{\text{tot}}^z + \frac{N}{2}$ is odd (even). The total momentum of the ground state is thus $P = N_f \pi$. The same two sets of k-values are found in the Bethe ansatz solution of the XXZ chain. The complication of two different Hilbert spaces is avoided with free boundary conditions, giving up translational symmetry.

Static correlation functions for the XX model can be calculated for the discrete system (without going to the continuum limit) [26]. The longitudinal correlation function in the ground state is obtained as

$$\langle 0|S_n^z S_0^z|0\rangle = -\frac{1}{4} \left(\frac{2}{\pi n}\right)^2 \tag{1.15}$$

for n odd, whereas it vanishes for even $n \neq 0$. The transverse correlation function is expressed as a product of two $n/2 \times n/2$ determinants; an explicit expression is available only for the asymptotic behavior [27]

$$\langle 0|S_n^x S_0^x|0\rangle = \langle 0|S_n^y S_0^y|0\rangle \sim C \frac{1}{\sqrt{n}}, \qquad C \approx 0.5884...$$
 (1.16)

A discussion of these correlation functions for finite temperature has been given by Tonegawa [28]. Static correlation functions can also be given exactly for the open chain, thus accounting for boundary effects, see e.g. [29]. Dynamic correlation functions cannot be obtained at the same level of rigor as static ones since they involve transitions between states in different Hilbert spaces (with even resp. odd fermion number). Nevertheless, detailed results for the asymptotic behavior have been obtained [30] and the approach to correlation functions of integrable models using the determinant representation to obtain differential equations [31] has emerged as a powerful new method.

Quantities of experimental relevance can be easily calculated from the exact expression for the free energy in terms of the basic fermion dispersion, (1.14),

$$F = -N k_{\rm B} T \left[\ln 2 + \frac{2}{\pi} \int_0^{\frac{\pi}{2}} dk \ln \cosh\left(\frac{\epsilon(k)}{2k_{\rm B}T}\right) \right].$$
(1.17)

An important quantity is the specific heat whose low-temperature behavior is linear in T:

$$C(T) \simeq \frac{\pi T}{6v_F},\tag{1.18}$$

where $v_F = (\partial \epsilon / \partial k)|_{k=k_F} = J$ is the Fermi velocity.

Low-lying excitations are also simply described in the fermion picture: They are either obtained by adding or removing fermions, thus changing the total spin projection S_{tot}^z by one unity and adding or removing the energy $\epsilon(k)$, or particle-hole excitations which do not change S_{tot}^z . Creating a general particle-hole excitation involves moving a fermion with momentum k_i inside the Fermi sea to some momentum k_f outside the Fermi sea. It is clear that moving a fermion just across the Fermi point costs arbitrarily low energy: the excitation spectrum is gapless. It is easily seen that for a given total momentum $q = k_f - k_i$ a finite range of excitation energies is possible, thus the spectrum of particle-hole excitations is a continuum with the initial momentum $k = k_i$ as internal degree of freedom:

$$\omega(q,k) = \epsilon(k+q) - \epsilon(k). \tag{1.19}$$

The resulting continuum for $S_{tot}^z = 0$ is shown in Fig. $1.2.S_{tot}^z = \pm 1$ excitations result from the one-fermion dispersion, but develop a continuum as well by adding particle-hole excitations with appropriate momentum; those excitations involve changing the number of fermions by one which implies a change of the total momentum by π , and thus the $S_{tot}^z = \pm 1$ spectrum is the same as in Fig. 1.2 up to the shift by π along the q axis.



Fig. 1.2. Excitation spectrum of the spin- $\frac{1}{2}$ XY chain in the $S_{tot}^z = 0$ subspace

For $\Delta \neq 0$ the interacting fermion Hamiltonian can be treated in perturbation theory [32]; from this approach and more generally from the Bethe ansatz and field-theoretical methods it is established that the behavior for $-1 < \Delta < +1$ is qualitatively the same as the free fermion limit $\Delta = 0$ considered so far: the excitation spectrum is gapless, a Fermi point exists and correlation functions show power-law behavior. The Heisenberg chain in the XY regime thus is in a critical phase. This phase is equivalent to the socalled Tomonaga-Luttinger liquid [33]. The fermion dispersion to first order in Δ is obtained by direct perturbation theory starting from the free fermion limit [34] (in units of J),

$$\epsilon(k) = \Delta - \lambda + \cos q -(2\Delta/\pi) \,\theta(1-\lambda) \left\{ \arccos \lambda - (1-\lambda^2)^{1/2} \cos q \right\} \,, \qquad (1.20)$$

where $\lambda = g\mu_B H/J$, and θ is the Heaviside function.

Finally we indicate how these results generalize for $\gamma > 0$, i.e. (see (1.6)) when the rotational symmetry in the *xy*-plane is broken and a unique preferred direction in spin space exists: $\Delta = 0$ continues to result in a free fermion system, but the basic fermion dispersion acquires a gap and the ground state correlation function $\langle 0|S_n^x S_0^n|0\rangle$ develops long range order [26].

1.2.4 The Isotropic Heisenberg Antiferromagnet and Its Vicinity

The most interesting regime of the S = 1/2 XXZ chain is $\Delta \approx 1$, i.e. the vicinity of the isotropic Heisenberg antiferromagnet (HAF). This important limit will be the subject of a detailed presentation in the chapters by Cabra and Pujol, and Klümper, with the use of powerful mathematical methods of Bethe ansatz and field theory. Here we restrict ourselves to a short discussion of important results.

The ground state energy of the HAF is given by

$$E_0 = -NJ\ln 2 \tag{1.21}$$

The asymptotic behavior of the static correlation function at the isotropic point is [35–37]

$$\langle 0|\boldsymbol{S}_n \cdot \boldsymbol{S}_0|0\rangle \propto (-1)^n \frac{1}{(2\pi)^{\frac{3}{2}}} \frac{\sqrt{\ln n}}{n}.$$
 (1.22)

This translates to a weakly diverging static structure factor at $q \approx \pi$,

$$S(q) \propto \frac{1}{(2\pi)^{\frac{3}{2}}} |\ln|q - \pi||^{\frac{3}{2}}.$$
 (1.23)

The uniform susceptibility at the HAF point shows the logarithmic corrections in the temperature dependence [38]

$$\chi(T) = \frac{1}{\pi^2 J} \left(1 + \frac{1}{2 \ln(T_0/T)} + \dots \right);$$
(1.24)

this singular behavior at $T \to 0$ was experimentally observed in Sr₂CuO₃ and SrCuO₂ [39]. The elementary excitations form a particle-hole continuum $\omega(q,k) = \epsilon(q+k) - \epsilon(k)$, obtained from fundamental excitations with dispersion law

$$\epsilon(k) = \frac{\pi}{2} J |\sin k| \tag{1.25}$$

which are usually called spinons. This dispersion law was obtained by des-Cloizeaux and Pearson [40], however, the role of $\epsilon(k)$ as dispersion for the basic constituents of a particle-hole continuum was first described by Faddeev and Takhtajan [9]. When the HAF point is crossed, a phase transition from the gapless XYregime to the gapped antiferromagnetic Ising regime takes place which is of the Kosterlitz-Thouless type: the Néel gap opens up with nonanalytic dependence on $\Delta - 1$ corresponding to a correlation length

$$\xi \propto e^{\pi/\sqrt{\Delta-1}} \tag{1.26}$$

The divergence of the transverse and the longitudinal structure factors differs when the HAF is approached from the Ising side in spite of the isotropy at the HAF point itself [37].

In contrast to the behavior of the isotropic HAF, the correlation functions for $\Delta < 1$ do not exhibit logarithmic corrections and the asymptotic behavior in the ground state is given by

$$\langle 0|S_n^x \cdot S_0^x|0\rangle = (-1)^n A_x \frac{1}{n^{\eta_x}}, \quad \langle 0|S_n^z \cdot S_0^z|0\rangle = (-1)^n A_z \frac{1}{n^{\eta_z}}, \qquad (1.27)$$

where

$$\eta_x = \eta_z^{-1} = 1 - \frac{\arccos \Delta}{\pi}.$$
(1.28)

For $|\Delta| < 1$ presumably exact expressions for the amplitudes A_x , A_z have been given in [41,42].

1.2.5 The Dynamical Structure Factor of the XXZ Chain

Two-Domain Wall Picture of the Excitation Continua

The dynamical structure factor $S(q, \omega)$ of the XXZ chain for low frequencies is dominated by the elementary excitations for the HAF as well as in the Ising and XY phases. The common feature is the presence of an excitation continuum as was made explicit in the Néel phase and for the free fermion limit above and stated to be true for the HAF.

In the Néel phase a one-domain wall state was seen to have $S_{\text{tot}}^z = \pm 1/2$. The only good quantum number is S_{tot}^z and two domain walls can combine into two states with $S_{\text{tot}}^z = 0$ and two states with $S_{\text{tot}}^z = \pm 1$ with equal energies (in the thermodynamic limit) but different contributions to the DSF. When the isotropic point is approached these four states form one triplet and one singlet to give the fourfold degenerate spinon continuum.

For all phases the excitation continuum emerges from the presence of two dynamically independent constituents. The spinons of the isotropic HAF can be considered as the isotropic limit of the Néel phase domain walls. The domain wall picture applies also to the XY phase: A XY-phase fermion can be shown to turn into a domain wall after a nonlocal transformation [43] and adding a fermion at a given site corresponds to reversing all spins beyond that site. Thus the domain wall concept of the antiferromagnetic Ising regime is in



Fig. 1.3. Spinon continuum for various anisotropies Δ (reproduced from [46])

fact a general concept unifying the dynamics in the regime $+\infty > \Delta > -1$, i.e. up to the transition to the ferromagnetic regime.

The one-DW dispersion as well as the appearance of a continuum with an energy gap for $\Delta > 1$ agrees with the results obtained from Bethe ansatz calculations [44, 45] taken in lowest order in $1/\Delta$. We make use of the full Bethe ansatz results for finite values of $1/\Delta$ to show a a numerical evaluation of these results. Figure 1.3 demonstrates that the gapped, anisotropic two spinon continuum develops continuously from the antiferromagnetic Ising phase into the gapless spinon continuum of the isotropic Heisenberg antiferromagnet. To make contact with the isotropic limit, in Fig. 1.3 spectra in the Néel phase are presented using the extended Brillouin zone (the Bethe ansatz excitations can be chosen as eigenfunctions under translation by one site). Although these graphs are suggestive the precise relation between the Bethe ansatz excitation wave functions and the lowest order domain wall ones (cf. Fig. 1.1) is difficult to establish.

Frequency Dependence of $S(q, \omega)$

In the XY regime (including the limit of the HAF) the asymptotic spatial dependence of the static correlation function is generalized to the timedependent case by replacing n^2 by (n-vt)(n+vt) (v is the spin wave velocity). This leads immediately to the most important property of the dynamic structure factor, namely the appearance (at T = 0) of an edge singularity at the lower threshold of the continuum:

$$S^{\alpha,\alpha}(q,\omega) \propto \frac{1}{(\omega^2 - \omega(q)^2)^{1 - \frac{\eta_\alpha}{2}}} \theta(\omega^2 - \omega(q)^2)$$
(1.29)

(obtained by bosonization for S = 1/2 in the zero temperature and long wavelength limit, by Schulz [47]) with exponents η_{α} depending on the anisotropy Δ as given in (1.28) above. This expression is consistent with the exact result obtained for the longitudinal DSF of the XX model using the free fermion approach [48, 49]:

$$S^{zz}(q,\omega) = 2\frac{1}{\sqrt{4J^2 \sin^2\left(\frac{q}{2}\right) - \omega^2}} \Theta(\omega - J\sin q) \Theta(2J\sin\frac{q}{2} - \omega); \quad (1.30)$$

the XX model is however peculiar since there is no divergence in S^{zz} at the lower continuum boundary.

This edge singularity is of essential relevance for experiments probing the dynamics of spin chains in the XY phase including the antiferromagnetic point and we therefore give a short survey of the phenomenological, more physical approaches in order to provide an understanding beyond the formal results.

The singularity is already obtained on the semiclassical level in an expansion in 1/S. This approach served to interpret the first experimental verification of the infrared singularity by neutron scattering experiments on the material CPC [15]. In this approach the exponent to first order in 1/S is $\eta = 2/(\pi \hat{S}), \hat{S} = \sqrt{S(S+1)}$ for $\Delta = 1$ [50] and has also been obtained to second order in 1/S for chains with XY like exchange and single-ion anisotropy [51].

The semiclassical approach clearly shows the essence of this singularity: Many low-lying modes which are harmonic in simple angular variables ϕ_n , θ_n add up to produce the singularity in the spin variable $S_n \propto \exp i\phi_n$, whose correlations are actually measured in $S(q, \omega)$. The finite temperature result for $S(q, \omega)$ in this approach is identical to the result of bosonization [32] which was then generalized to the exact Bethe ansatz result with exact values $\eta = 1$ for $\Delta = 1$ (HAF) and $\eta = \frac{1}{2}$ for $\Delta = 0$ (XY). The physical understanding of the excitation continuum as domain wall continuum was finally established by Faddeev and Takhtajan [9].

The singular behavior of the dynamic structure factor was supported by numerical calculations using complete diagonalization. Combined with exact results, this lead to the formulation of the so-called Müller ansatz [49,52] for the isotropic $S = \frac{1}{2}$ chain:

$$S(q,\omega) = \frac{A}{\sqrt{\omega_1^2 - \omega(q)^2}} \Theta(\omega - \omega_1(q))\Theta(\omega_2(q) - \omega), \qquad (1.31)$$

with $\omega_1(q) = (\pi/2)J|\sin q|$ and $\omega_2(q) = \pi J|\sin(q/2)|$. This ansatz parametrizes the dynamic structure factor as in (1.29) and adds an upper limit corresponding to the maximum two spinon energy (note that for the isotropic chain there is no divergence at the upper continuum boundary). This ansatz is now frequently used for an interpretation of experimental data, neglecting the presence of small but finite excitation strength above the upper threshold frequency $\omega_2(q)$ as confirmed by detailed numerical investigations (the total intensity of the two spinon continuum has been determined as 72.89 % of the value 1/4, given by the sum rule (1.5) [53]). Experimental investigations of the excitation continuum include the Heisenberg antiferromagnet CuCl₂·2NC₅H₅ (CPC) [15] and recent work on the HAF KCuF₃ [54]. Beautiful pictures of the spinon continuum are also available for the spin-Peierls material CuGeO₃ [55].

Temperature dependence and lineshapes of the dynamic structure factor for $q \approx \pi$ have been investigated by bosonization techniques [47], conformal field theory [13] and numerical approaches [56]. Numerical calculations of all eigenvalues for chains with 16 spins [57] have shown the full picture of the spinon continuum and its variation with temperature. The functional form of the Müller ansatz found strong support when the dynamical structure factor for the Haldane-Shastry chain (Heisenberg chain on a ring geometry with long range interactions propertional to the inverse square of the distance [58]) was calculated exactly [59] and was shown to take exactly the form of (1.31).

For XXZ chains close to the Ising limit with their spectrum determined by gapped solitons the dynamic response is different: At T = 0 both $S^{xx}(q,\omega)$ and $S^{zz}(q,\omega)$ are dominated by the two-domain wall or spin wave continuum in the finite frequency range determined from (1.9) with no singularity at the edges [23] (there is just an asymmetry with a steepening at the lower frequency threshold). Upon approach to the isotropic limit the infrared singularity develops gradually starting from wave vector $\pi/2$. At finite temperature an additional central peak develops from energy transfer to a single domain wall [24]. These continua have been observed in the material CsCoCl₃ [60–62]. The two-domain wall continuum has been shown to shift its excitation strength towards the lower edge in frequency when a (ferromagnetic) NNN interaction is added to the Hamiltonian [63].

1.2.6 Modified S=1/2 Chains

In this subsection we shortly discuss a number of modifications to the ideal S = 1/2 XXZ chain which add interesting aspects to the theoretical picture and are also relevant for some real materials.

A theoretically particularly important model is the isotropic Heisenberg chain with nearest and next-nearest exchange

$$\mathcal{H} = J \sum_{n} \left(\boldsymbol{S}_{n} \cdot \boldsymbol{S}_{n+1} + \alpha \boldsymbol{S}_{n} \cdot \boldsymbol{S}_{n+2} \right)$$
(1.32)

which for $\alpha > 0$ exhibits the effects of frustration from competing interactions. In the classical limit the system develops spiral order in the ground state for $\alpha > 1/4$ whereas for S = 1/2 a phase transition to a dimerized state occurs at $\alpha = \alpha_c \approx 0.2411...$ This dimerized state is characterized by a singlet ground state with doubled lattice constant and twofold degeneracy and an excitation gap to the first excited states, a band of triplets. It is thus one of the simple examples for the emergence of an energy gap in a 1D system with rotational symmetry by dynamical symmetry breaking. This quantum phase transition was first found at $\alpha \approx 1/6$ from the bosonization approach [64]. The phase transition has been located with high numerical accuracy by Okamoto and Nomura [65] considering the crossover between the singlet-singlet and singlet-triplet gaps, a criterion which has proven rather effective also in related cases later.

For $\alpha = 1/2$, one arrives at the Majumdar-Ghosh limit [66], where the exact form of these singlet ground states $|0\rangle_{I,II}$ is known to be a product of singlets (dimers):

$$|0\rangle_{I} = |[1,2] \cdots [2p+1,2p+2] \cdots \rangle \qquad |0\rangle_{II} = |[2,3] \cdots [2p,2p+1] \cdots \rangle$$
(1.33)

with the representation of a singlet as

$$|[2p, 2p+1]\rangle = \frac{1}{\sqrt{2}} \sum_{s,s'} \chi_{2p}(s) \ \epsilon^{s,s'} \ \chi_{2p+1}(s') \tag{1.34}$$

where $\chi_m(s)$ is the spin state at site m and ϵ is the antisymmetric tensor

$$\epsilon = \begin{pmatrix} 0 & 1\\ -1 & 0 \end{pmatrix}. \tag{1.35}$$

in spin space $s=(\uparrow,\downarrow).$ This becomes easily clear by considering the following Hamiltonian

$$\widetilde{\mathcal{H}}_{MG} = \frac{1}{4}(\mathbf{S}_1 + \mathbf{S}_2 + \mathbf{S}_3)^2 + \frac{1}{4}(\mathbf{S}_2 + \mathbf{S}_3 + \mathbf{S}_4)^2 + \frac{1}{4}(\mathbf{S}_3 + \mathbf{S}_4 + \mathbf{S}_5)^2 + \dots$$

for N spins and periodic boundary conditions. $\widetilde{\mathcal{H}}_{MG}$ is identical to \mathcal{H}_{MG} apart from a constant:

$$\widetilde{\mathcal{H}}_{MG} = \sum_{n} \boldsymbol{S}_{n} \cdot \boldsymbol{S}_{n+1} + \frac{1}{2} \sum_{n} \boldsymbol{S}_{n} \cdot \boldsymbol{S}_{n+2} + \frac{3}{4} \sum_{n} \boldsymbol{S}_{n}^{2} = \mathcal{H}_{MG} + \frac{9}{16} N$$

Using

$$(\mathbf{S}_n + \mathbf{S}_{n+1} + \mathbf{S}_{n+2})^2 \ge S(S+1)|_{S=\frac{1}{2}} = \frac{3}{4}$$

we obtain

$$\widetilde{E}_0 \ge \frac{3}{16}N.$$

The two ground states obtained by covering the chain completely with singlets formed of two spins 1/2 have energy equal to this lower bound since each contribution of the type $(\mathbf{S}_n + \mathbf{S}_{n+1} + \mathbf{S}_{n+2})^2$ contains two spins which are coupled to a singlet and therefore reduces to $\mathbf{S}^2 = \frac{3}{4}$. The dimer product states are therefore ground states of the Majumdar-Ghosh Hamiltonian with energy per spin $E_0/N = -3/8$. It is evident that this ground state is completely disordered, i.e. all two-spin correlation functions vanish identically. There is however, perfect order of the singlets, expressed in the statement that the Majumdar-Ghosh ground state forms a *dimer crystal*. Quantitatively this is expressed in a finite value of the dimer-dimer (four spin) correlation function

$${}_{I}\langle 0|(\boldsymbol{S}_{1}\cdot\boldsymbol{S}_{2})(\boldsymbol{S}_{2p+1}\cdot\boldsymbol{S}_{2p+2})|0\rangle_{I}.$$

$$(1.36)$$

for arbitrary n (and the equivalent relation for $|0\rangle_{II}$).

Another variant of the Heisenberg chain is obtained by adding dimerization explicitly to the Hamiltonian, giving the alternating chain

$$\mathcal{H} = J \sum_{n} \left(1 + (-1)^n \delta \right) \left(\boldsymbol{S}_n \cdot \boldsymbol{S}_{n+1} \right)$$
(1.37)

This model was first investigated by Cross and Fisher [67]; with explicit dimerization the ground state is unique and a gap opens up immediately, $E_g \propto \delta^{2/3}$ (apart from logarithmic corrections). The ground state prefers to have singlets at the strong bonds and the lowest excitations are propagating one-triplet states. These can be considered as bound domain wall states since two domain walls of the type described above with singlets on the 'wrong' sites between them feel an attractive interaction growing with distance. The model with both NNN exchange and alternation is equivalent to a spin ladder and will be discussed in more detail in Sect. 1.4.

Models with explicit or spontaneous dimerization are now frequently used to describe spin-Peierls chains, i.e. spin chains which dimerize due to the spin phonon interaction. This field was stimulated in particular by the discovery of the inorganic spin-Peierls material CuGeO₃ [16]. Whereas the adiabatic limit when phonons follow spins without relaxation is not appropriate for this material, the flow equation approach has been used to reduce the general spin-phonon model to a spin only Hamiltonian [68, 69] and the spin Peierls gap then results from the combined action of alternation and frustration. Phonons, however, do introduce some features not covered by this simplification [70] and it is not clear at the moment whether the simplified spin model captures the physics of real spin Peierls materials, in particular of the inorganic compound CuGeO₃ (for a review see [71]).

Another variant of the simple 1D chain are decorated chains, where more complicated units are inserted in the 1D arrangement. As an example we mention the orthogonal-dimer spin chain with frustrated plaquettes inserted in the chain [72,73], see Fig. 1.4. Depending on the strength of the competing interactions, this chain can be in a dimer phase or in a plaquette phase with interesting dynamic properties. Interest in this model is motivated by its relation to the 2D orthogonal-dimer model which is realized in the compound $SrCu_2(BO_3)_2$.



Fig. 1.4. An example of decorated chains: orthogonal-dimer spin chain [72]

Interesting aspects are found in S = 1/2 chains with random couplings. Using the real space renormalization group it has been shown that the ground state of the random antiferromagnetic Heisenberg chain is the random singlet state, i.e. spins form singlets randomly with distant partners [74]. Hida has extended these studies to dimerized chains [75]. Heisenberg chains with a random distribution of ferro- and antiferromagnetic exchange constants have been shown to have a different type of ground state called the large spin state [76, 77], characterized by a fixed point distribution not only of bond strength, but also of spin magnitudes.

1.2.7 The XXZ Chain in an External Magnetic Field

An external magnetic field leads to qualitatively new phenomena in spin chains when the Zeeman energy becomes comparable to the scale set by the exchange energies. Contrary to other parameters in the Hamiltonian (e.g. chemical composition, exchange integrals) an external field is relatively easy to vary experimentally. Therefore these effects deserve particular attention; actually experimental and theoretical investigations involving high magnetic fields have developed into one of the most interesting topics in the field of low-dimensional magnetism in the last few years.

The phase diagram of the XXZ model in an external magnetic field in zdirection is shown in Fig. 1.5: The boundary between the ferromagnetic phase and the XY phase is given by $H_c = \pm J(1 + \Delta)$. For $\Delta < 1$ (XY symmetry) the XY phase extends down to H = 0. In the fermion representation the external field acts as chemical potential, and the fermion occupation number changes from zero to saturation when the XY phase is crossed at constant Δ . For $\Delta > 1$ (Ising symmetry) there is a transition from the Néel phase to the XY phase at $H = H_{c1} = E_g(\Delta)$, where $E_g(\Delta)$ is the triplet gap. In the $S = \frac{1}{2}$ chain this transition is of the second order [78] and the magnetization appears continuously as $m \propto (H - H_c)^{1/2}$, whereas for $S > \frac{1}{2}$ it acquires the features of the classical first-order spin-flop transition with a jump in m at $H = H_{c1}$ [79].



Fig. 1.5. Phase diagram of a XXZ Heisenberg $S = \frac{1}{2}$ chain in magnetic field

The effect of the external field on the excitation spectrum is calculated exactly for the XX model, i.e. in the free fermion case, with the result shown in Fig. 1.6: The Fermi points shift from $k_F = \pm \pi/2$ to $\pm (\pi/2 + \delta k)$ and gapless excitations are found for wave vectors $q = \pi \pm 2\delta k$, where δk is determined by $J\cos(\pi/2 + \delta k) + H = 0$ and implies incommensurability in the ground state. This result is representative for the XY-phase and the isotropic Heisenberg antiferromagnet. It has been confirmed in neutron scattering experiments on the $S = \frac{1}{2}$ chain material Cu-Benzoate [80]. On the theoretical side, e.g., line shapes for finite external field have been calculated from the Bethe ansatz [81].



Fig. 1.6. Excitation spectrum of the spin- $\frac{1}{2}$ XY chain in the $S_{\text{tot}}^z = 0$ subspace for finite external field, $g\mu_B H/J = 0.3$

For the Heisenberg antiferromagnet with general anisotropies a remarkable curiosity has been found by Kurmann et al [82]: For any combination of couplings and any field direction there exists a field strength H_N which renders the ground state very simple, namely factorizable, i.e. it essentially becomes identical to the classical ground state. Simple examples are the XXZ model with external field in z- resp. x-direction, where the corresponding field values are

$$H_N^{(z)} = J(1+\Delta), \quad H_N^{(x)} = J\sqrt{2(1+\Delta)}.$$
 (1.38)

An interesting situation can develop when a uniform external field via a staggered g-factor and possibly a Dzyaloshinkii-Moriya interaction induces a staggered field such as in Cu-Benzoate [80] and materials of related symmetry [83,84]: Then a staggered field is induced which is proportional to the external field and a gap opens up which for small fields behaves as [85,86]

$$E_g \propto \left(\frac{H}{J}\right)^{2/3} \ln^{1/6}\left(\frac{J}{H}\right).$$
 (1.39)

The magnetic chain in this situation is equivalent to a quantum sine-Gordon chain carrying solitons and breathers (soliton-antisoliton bound states) as excitations; these were identified in neutron scattering and ESR experiments [87] and their contributions to the dynamical structure factor were calculated from sine-Gordon field theory [88,89].

For an external *transverse* field the Ising model in a transverse field is the best known example. It is solved as free fermion model [90] and serves as one of the standard models of a quantum phase transition [91]. More interesting and much more difficult is the case of an XY chain where a transverse field breaks the rotational symmetry since in this case a simple free fermion limit does not exist and also bosonization does not go beyond establishing the existence of a gap. Such a system is of interest as the quantum analog of the standard example for classical soliton bearing magnetic chains like $CsNiF_3$ [18]. The phase diagram for the Heisenberg chain in a transverse field has been discussed already in [82] and recently again for the XX model [56] and for the XXZ model in mean-field approximation (MFA) [92] and MFA with additional field theoretic input [93]. Recent experiments on the XY spin chain Cs_2CoCl_4 in a transverse magnetic field [94] show an interesting phase diagram including a quantum spin liquid phase which extends to zero temperature and are presently stimulating theoretical investigations in this field.

1.2.8 Effects of 3D Coupling

Since the isotropic spin- $\frac{1}{2}$ chain is gapless, even a weak 3D coupling between the chains $J' \ll J$ will lead to the emergence of the long-range staggered order. The magnitude of this order as a function of J' can be calculated within the mean-field or RPA approximation [95–98]: solving the problem of an isolated chain in an external staggered field $h_{\rm st}$, one obtains for the staggered magnetization $m_{\rm st}$ the expression [86]

$$m_{\rm st} = c \left[(h_{\rm st}/J) \ln(J/h_{\rm st}) \right]^{1/3}, \quad c \simeq 0.387.$$
 (1.40)

This is then treated as a self-consistency equation for $m_{\rm st}$ after assuming the mean-field relation $h_{\rm st} = \tilde{J}'(\boldsymbol{q}_B)m_{\rm st}$, where $\tilde{J}'(\boldsymbol{q})$ is the Fourier transform of the interchain interaction and \boldsymbol{q}_B is the magnetic Bragg wave vector. This yields $m_{\rm st} \simeq 0.29 \left[(J'/J) \ln(J/J') \right]^{1/2}$ [98], where $J' \equiv \tilde{J}'(\boldsymbol{q}_B)$.

The dynamical susceptibilities of an isolated chain in a staggered field were calculated in [97]. Both the longitudinal and transverse (with respect to the ordered moment) polarization channels contain quasiparticle and continuum contributions. The transverse single mode has the gap $\Delta \simeq$ $0.842J' \ln^{1/2}(J/J')$ [98], and the gap of the longitudinal mode is $\Delta\sqrt{3}$, while the continuum in both channels starts at 2Δ . The 3D dynamical susceptibility $\chi_{3D}(\mathbf{q},\omega)$ can be obtained with the help of the RPA formula

$$\chi_{3D}^{\alpha}(\boldsymbol{q},\omega) = \frac{\chi_{1D}^{\alpha}(\boldsymbol{q}_{\parallel},\omega)}{1 - \tilde{J}'(\boldsymbol{q})\chi_{1D}^{\alpha}(\boldsymbol{q}_{\parallel},\omega)},\tag{1.41}$$

where $\alpha = \parallel, \perp$ denotes the longitudinal or transverse direction with respect to the ordered moment. This expression follows from the usual susceptibility definition $m(q,\omega) = \chi(q,\omega)h(q,\omega)$ if one replaces h with the effective mean field $h_{\text{eff}} = h(\boldsymbol{q}, \omega) + \widetilde{J}'(\boldsymbol{q})m(\boldsymbol{q}, \omega)$. Physical excitation frequencies are determined as poles of the χ_{3D} . An intrinsic flaw of this approach is that both the transverse and longitudinal modes come out gapped, while it is physically clear that there should be gapless Goldstone modes in the transverse channel at $q = q_B$. This can be fixed [96] by the renormalization $\chi_{1D}^{\perp} \mapsto Z \chi_{1D}^{\perp}$, where the renormalization factor Z is determined from the condition $Z\tilde{J}'(\boldsymbol{q}_B)\chi_{1D}^{\perp}(\boldsymbol{q}_B,0)=1$. Within this approach, the longitudinal mode remains a well-defined gapped excitation. Such a mode was successfully observed in $KCuF_3$ [99], but it was argued it cannot be distinguished from the continuum in another $S = \frac{1}{2}$ -chain material BaCu₂Si₂O₇ [98, 100]. Those results indicate that the lifetime of the longitudinal mode can be limited by the processes of decay into a pair of transverse modes with nearly zero frequency [98], which cannot be analyzed in framework of the RPA approach.

1.3 Spin Chains with S > 1/2

Antiferromagnetic Heisenberg spin chains with integer and half-integer value of spin S behave in a very different way, as was discovered by Haldane twenty years ago [10]. He has shown that the ground state of an integer-S Heisenberg AF chain should have a finite spectral gap, though exponentially small in the large-S limit. This special disordered state of isotropic integer-S chains with only short-range, exponentially decaying AF spin correlations has received the name of the Haldane phase. The most thoroughly studied example is the S = 1 chain.

1.3.1 S = 1 Haldane Chain

The isotropic S = 1 Heisenberg antiferromagnetic chain is the simplest example of a system with the Haldane phase and is thus often called *the Haldane*

chain. Following Haldane's conjecture it was the subject of numerous investigations and although an exact solution of the proper S = 1 HAF has not been found, many approximate and numerical approaches have established a coherent picture characterized by the following properties [101, 102]: The Haldane chain has the ground state energy per spin $E \simeq -1.40$ and shortrange AF spin correlations $\langle S_0^{\alpha} S_n^{\beta} \rangle \propto (-1)^n \delta_{\alpha\beta} n^{-1/2} e^{-n/\xi}$ characterized by the correlation length $\xi \simeq 6.0$. Its lowest excitations form a massive magnon triplet, the excitation spectrum has a gap $\Delta \simeq 0.41J$ at wave vector $q = \pi$, and the dispersion of the low-lying excitations with q close to π is well described by the "relativistic" law $\varepsilon(q) = \sqrt{\Delta^2 + v^2(q-\pi)^2}$, with the spin wave velocity $v \simeq 2.46J$. The single-particle energy grows fast as q moves away from π , so that the spectrum around q = 0 is dominated by the two-particle continuum whose lower boundary starts at approximately 2Δ . The second lowest excitation at $q = \pi$ belongs to the three-soliton continuum and has the energy $\approx 3\Delta$, as shown in Fig. 1.7. The gap in the spectrum translates into an activated behavior of magnetic specific heat and susceptibility, the fingerprints of gapped systems in macroscopic properties.



Fig. 1.7. Spectrum of low-lying excitations in S = 1 Haldane chain, from the QMC calculation of [103]

An important property of the S = 1 Haldane chain is the so-called string order *string order* which is a nonlocal quantity defined as the limiting value of the correlator

$$\mathcal{O}_1^{\alpha}(n,n') = \left\langle -S_n^{\alpha} e^{i\pi \sum_{j=n+1}^{n'-1} S_j^{\alpha}} S_{n'}^{\alpha} \right\rangle, \qquad \alpha = x, y, z.$$
(1.42)

at $|n - n'| \to \infty$. Presence of this order means that the ground state of the chain favors such spin states where the $|+\rangle$ and $|-\rangle$ spin-1 states alternate, "diluted" with strings of $|0\rangle$ of arbitrary length. One speaks about a "diluted AF order". This "diluted AF order" reaches its maximal value, 1, in the Néel state. In the Haldane phase, however, the Néel order vanishes, while the string order persists, its value for a *rotational invariant* state being limited

by 4/9 from above. For the Haldane chain the value of the string order is somewhat lower, $\mathcal{O}_1^{\text{Hald}} \simeq 0.37$ [104, 105].

Hidden order was originally introduced in constructing an analogy to surface phase transitions in *solid-on-solid* (SOS) models [106] and to the fractional quantum Hall effect [107]. This leads to a very visual interpretation of the hidden order: If we define a correspondence between $|+\rangle$ sites and a positive $\Delta h = +1$ step of the interface position, and respectively between $|-\rangle$ sites and a $\Delta h = -1$ step, then hidden order corresponds to the socalled "disordered flat" (or "fluid flat") phase. This *preroughening* phase is characterized by a flat surface with a *finite* average fluctuation of the surface height, but no order in the *position* of the $\Delta h = \pm 1$ steps. As shown by Kennedy and Tasaki [108], the hidden symmetry breaking by the string order parameter can be transformed into an explicit breaking of a $Z_2 \times Z_2$ symmetry by a nonlocal unitary transformation which characterizes the Haldane chain.

Importance of the string order is even more stressed by the fact that the lowest excitations of the S = 1 Haldane chain can be interpreted as *solitons* in the string order [109–112].

Experimentally the Haldane chain was most comprehensively studied via inelastic neutron scattering in S = 1 chain material Ni(C₂H₈N₂)₂NO₂(ClO₄) (NENP), confirming the theoretical predictions. For higher S the experiments are scarce; the Haldane phase was reported to be found in the S = 2AF chain material MnCl₃(2, 2' – bipyridine) on the basis of the magnetization measurements [113]: under application of an external magnetic field, the magnetization remained zero in a finite field range, indicating presence of a gapped phase. We postpone to Sect. 1.6 the discussion of interesting physics which arises if one succeeds to close the gap by the magnetic field, and concentrate here on the properties of the Haldane phase itself.

Anisotropic S = 1 Haldane Chain

An interesting phase diagram emerges if one considers a S = 1 chain with anisotropies as described by the Hamiltonian

$$\mathcal{H} = \sum_{n} (S_n^x S_{n+1}^x + S_n^y S_{n+1}^y) + J^z S_n^z S_{n+1}^z + D(S_n^z)^2$$
(1.43)

The effects of exchange anisotropy J_z and single-ion anisotropy D are very different, and the system exhibits a rich phase diagram [47,106] shown in Fig. 1.8. To visualize the characteristic features of different phases, it is sometimes convenient to resort to the language of "solid-on-solid" models of surface phase transitions [106]. One identifies $|\pm\rangle$ spin-1 states with $\Delta h = \pm 1$ steps of the interface (domain walls), and treats those domain walls as particles with an internal degree of freedom – "spin" $\pm \frac{1}{2}$. Then one can interpret the Néel phase as a "solid flat," or "AF spin-ordered solid" one, i.e., a phase where there is a long-range correlation of particle positions, and their "spins" exhibit a long-range AF order. The gapped Haldane phase corresponds to the


Fig. 1.8. Phase diagram of the S = 1 Heisenberg chain with exchange anisotropy J_z and single-ion anisotropy D

"AF spin-ordered fluid" phase, characterized by the AF order in "spin" but with no order in the position of particles. The AF order disappears along the transition to the gapless XY1 phase which is a "spin-disordered fluid". Another gapless phase, the XY2 phase, can be described as a "spin-disordered solid" with the restored order in the particle positions. The so-called large-D phase large-D phase, which is achieved at sufficiently large values of the single-ion anisotropy, can be characterized as a gas of bound pairs of particles with opposite "spin". Those pairs unbind when D is decreased, and this transition is of the first order if it is to the ferromagnetic or to the Néel phase, of the Kosterlitz-Thouless (KT) type on the boundary to the XY1 phase, and Gaussian along the boundary to the Haldane phase.

The phase diagram of the anisotropic S = 1 chain was studied numerically [114, 115]. For purely exchange anisotropy (D = 0) the Haldane phase was found to exist in the interval from $J_z \approx 0 J_z \approx 1.2$, while for purely single-ion anisotropy $(J_z = 1)$ it persists from $D \approx -0.2$ to $D \approx 1$.

The role of anisotropy was also investigated for a $S = \frac{1}{2}$ chain with alternating ferro- and antiferromagnetic exchange, and a rich phase diagram was found [105]. In the limit of strong ferromagnetic bonds this system may be viewed as another physical model of the S = 1 Haldane chain, with the ferro exchange playing the role of the Hund coupling.

The phase diagram in the (D, J_z) space was analyzed by Schulz [47] for general S in the bosonization approach, which is able to capture the topology of the phase diagram. According to his results, the diagram of Fig. 1.8 should be generic for integer S, while for half-integer S the Haldane and large-D phases disappear, being replaced by the XY1 phase. Numerical studies [116] revealed that for S = 2 the XY1 phase creeps in between the Haldane phase and large-D one, squeezing the Haldane phase to a narrow region near the boundary to the Néel phase.

1.3.2 Integer vs Half-Odd-Integer S

The emergence of an energy gap in spite of rotational invariance comes as a surprise, especially because the classical Heisenberg chain, as well as the only exactly solvable quantum model of a Heisenberg spin chain, namely the $S = \frac{1}{2}$ one, are gapless. Classical intuition expects that a state arbitrarily close in energy to the ground state can be created by infinitesimally changing the angles between neighboring spins. For a quantum system whose ground state is a global singlet (the total spin $S_{tot} = 0$), however, this operation may just reproduce the initial state and thus fail to demonstrate the existence of gapless excitations.

Nonlinear σ -Model Description

Haldane's prediction, which created a surge of interest to one-dimensional magnets, was based on a large-S mapping to the continuum field theory, the so-called nonlinear sigma model (NLSM) (see e.g. [117]) which we will briefly review (for details, see the chapter by Cabra and Pujol).

Consider a spin-S antiferromagnetic Heisenberg chain described by the Hamiltonian

$$\mathcal{H} = J \sum \boldsymbol{S}_{j} \cdot \boldsymbol{S}_{j+1} - \boldsymbol{H} \cdot \sum_{j} \boldsymbol{S}_{j}, \qquad (1.44)$$

where we have included the external magnetic field H for the sake of generality. In the quasiclassical NLSM description one starts with introducing the set of coherent states

$$|\boldsymbol{n}\rangle = e^{iS^z\varphi}e^{iS^y\theta}|S^z = S\rangle,\tag{1.45}$$

where \boldsymbol{n} is the unit vector parameterizing the state and having the meaning of the spin direction. The partition function $Z = \text{Tr}(e^{-\beta \mathcal{H}})$, where $\beta = 1/T$ is the inverse temperature, can be rewritten as a coherent state path integral $Z = \int \mathcal{D}\boldsymbol{n}e^{-\mathcal{A}_E/\hbar}$, where $\mathcal{A}_E = \int_0^{\beta\hbar} d\tau L_E$ is the Euclidean action and $\tau = it$ is the imaginary time.

Breaking the spin variable \boldsymbol{n} into the smooth and staggered parts, $\boldsymbol{n}_j = \boldsymbol{m}_j + (-1)^j \boldsymbol{l}_j$, one can pass from discrete variables to the continuum fields \boldsymbol{m} , \boldsymbol{l} subject to the constraints $\boldsymbol{m}\boldsymbol{l} = 0$, $\boldsymbol{l}^2 + \boldsymbol{m}^2 = 1$. We assume that the magnetization for the low-energy states of the antiferromagnet is small, $|\boldsymbol{m}| \ll |\boldsymbol{l}|$, and therefore neglect higher than quadratic terms in \boldsymbol{m} . Then one can show that on the mean-field level \boldsymbol{m} is a slave variable, which can be excluded from the action,

$$\boldsymbol{m} = \frac{1}{4JS} \big\{ i\hbar (\boldsymbol{l} \times \partial_{\tau} \boldsymbol{l}) + \boldsymbol{H} - \boldsymbol{l} (\boldsymbol{H} \cdot \boldsymbol{l}) \big\}.$$
(1.46)

In weak fields and at low energies m^2 may be neglected in the constraint, so that l can be regarded as a unit vector and one arrives at the following effective Euclidean action depending on the unit vector l only:

$$\mathcal{A}_E = \mathcal{A}_B + \frac{\hbar}{2g} \int_0^{\beta\hbar c} dx_0 \int dx_1 \Big\{ (\partial_0 \boldsymbol{l} + \frac{i}{\hbar c} \, \boldsymbol{l} \times \boldsymbol{B})^2 + (\partial_1 \boldsymbol{l})^2 \Big\}, \qquad (1.47)$$

where $x_0 = c\tau$, $x_1 = x$, $c = \frac{2JSa}{\hbar}$, and g = 2/S. In absence of the magnetic field the model is Lorentz invariant (*c* plays the role of the limiting velocity) and is known as the O(3) NLSM with topological term. The so-called topological, or *Berry term* \mathcal{A}_B is given by

$$\mathcal{A}_B = i2\pi\hbar SQ, \qquad Q = \frac{1}{4\pi} \int d^2x \, \boldsymbol{l} \cdot (\partial_0 \boldsymbol{l} \times \partial_1 \boldsymbol{l}), \qquad (1.48)$$

The integer-valued quantity Q is the so-called *Pontryagin index* indicating how many times the vector l sweeps the unit sphere when x sweeps the two-dimensional space-time.

Without the topological term, the T = 0 partition function of the quantum AF spin-S chain is equivalent to that of a classical 2D ferromagnet at the effective temperature $T_{\text{eff}} = g$ in the continuum approximation. For integer spin S the topological term is ineffective since \mathcal{A}_B is always a multiple of $2\pi\hbar$, and the properties of the 1D quantum antiferromagnet can be taken over from the 2D classical ferromagnet. (This correspondence is in fact quite general, connecting the behavior of a *Lorentz invariant* quantum system in dimension d to that of its classical counterpart in dimension D = d + 1, and is often referred to as the quantum-classical correspondence).

At finite temperature the 2D classical ferromagnet is known [118, 119] to have a finite correlation length $\xi \propto e^{2\pi/T_{\rm eff}}$, which, in view of the Lorentz invariance, corresponds in the original spin chain to a finite *Haldane gap*

$$\Delta_{\rm Hald} \propto \hbar c / \xi = J S e^{-\pi S}$$

Thus, the T = 0 ground state of the integer-S isotropic Heisenberg onedimensional (D = 1 + 1) antiferromagnet is disordered, and the spectrum of elementary excitations has a gap. The degeneracy of the lowest excitations is threefold (in contrast to only double degeneracy obtained in spin wave approximation which is absent on the Néel state with broken symmetry). Spin correlations in real space are given by the so-called *Ornstein-Zernike correlation*

$$\langle \boldsymbol{l}(x)\boldsymbol{l}(0)\rangle \propto \frac{e^{-|x|/\xi}}{|x|^{(D-1)/2}}, \quad |x| \to \infty.$$
 (1.49)

For half-odd-integer spins, the contribution of any field configuration into the partition function carries a nontrivial phase factor $e^{-i2\pi SQ}$, which leads to the interference of configurations with different Q, and at the end to the absence of a gap in Heisenberg spin-S chains with half-odd-integer S. There is an argument due to Affleck [117] which connects this effect to the contribution of *merons* – objects with the topological charge $Q = \pm \frac{1}{2}$ which may be thought of as elementary entities constituting a Q = 1 solution known as the Belavin-Polyakov soliton [120].

Although in the NLSM formulation the presence of the topological term renders the half-odd integer spin chain theoretically more complicated than the integer-S one, the emergence of an energy gap in the latter in spite of rotational invariance calls for a simple physical explanation. It is instructive to see where the intuition goes wrong; this can be seen from the statement known as the Lieb-Schultz-Mattis theorem [26] for spins $\frac{1}{2}$, generalized later by Affleck and Lieb [121] to arbitrary half-odd-integer S and by Oshikawa et al. to finite magnetization [122]:

Generalized Lieb-Schultz-Mattis Theorem

Assume that (i) we have a spin-S chain with short-range exchange interaction, (ii) the Hamiltonian \mathcal{H} is invariant with respect to a translation by l lattice constants and (iii) \mathcal{H} is invariant with respect to arbitrary rotation around the z axis, so that the ground state has a definite $S_{\text{tot}}^z = LM$, where L is the number of spins in the chain.

Then, if l(S - M) is a half-odd-integer, there system is either gapless in the thermodynamic limit $L \to \infty$, or the ground state is degenerate, with spontaneously broken translational symmetry.

The proof runs as follows: let $|\psi_0\rangle$ be the ground state with certain magnetization M per spin. Consider the unitary *twist operator*

$$\widehat{U} = \exp\{i\frac{2\pi}{L}\sum_{j=1}^{L}jS_{j}^{z}\}$$

and construct a new state $|\psi_1\rangle = \hat{U}|\psi_0\rangle$. Assume for definiteness that

$$\mathcal{H} = \sum_{nm} \left\{ \frac{1}{2} J_{nm} (S_n^+ S_{n+m}^- + S_n^- S_{n+m}^+) + J_{nm}^z S_n^z S_{n+m}^z \right\};$$

this exact form is not essential, the same course of derivation can be performed assuming presence of any powers $(S_n^+ S_{n+m}^-)^k$. Operator S^z remains invariant under the unitary transformation, and $U^{\dagger}S_n^+U = e^{i2\pi n/L}S_n^+$, so that the energy difference between $|\psi_0\rangle$ and $|\psi_1\rangle$ is

$$\Delta E = \sum_{nm} J_{nm} e_{nm} (\cos \frac{2\pi m}{L} - 1), \qquad e_{nm} \equiv \langle \psi_0 | S_n^+ S_{n+m}^- | \psi_0 \rangle.$$

Denoting $\sum_{n=1}^{L} J_{nm} e_{nm} = L f_m$ and taking the thermodynamic limit $L \to \infty$, one obtains

$$\Delta E = E_1 - E_0 \propto \frac{1}{L} \sum_m m^2 f_m,$$

and, if the last sum is finite (which is true for J_{nm} being a reasonably fast decaying function of the distance m), we come to the conclusion that the energy E_1 of the state $|\psi_1\rangle$ tends to the ground state energy E_0 in the thermodynamic limit.

Now consider the overlap of $|\psi_0\rangle$ and $|\psi_1\rangle$: if they are orthogonal, one can be sure that E_1 gives a variational upper bound of the energy of the true eigenstate, otherwise no statement can be made.

Assume that the original translational symmetry of the Hamiltonian is not broken, i.e. that $T_l |\psi_0\rangle = |\psi_0\rangle$, where T_l is the operator of translation by l lattice sites, $T_l S_n T_l^{-1} = S_{n+l}$. Then the overlap

$$z_1 = \langle \psi_0 | \psi_1 \rangle = \langle \psi_0 | T_l U T_l^{-1} | \psi_0 \rangle.$$

The transformed twist operator can be rewritten as

$$T_{l}UT_{l}^{-1} = \exp\{i\frac{2\pi}{L}\sum_{j=1}^{L}jS_{j+l}^{z}\} = \exp\{i\frac{2\pi}{L}\sum_{j=1}^{L}(j-l)S_{j}^{z} + i2\pi\sum_{k=1}^{l}S_{k}^{z}\},\$$

where we have used periodic boundary conditions $S_{L+n}^z = S_n^z$. It is easy to see that $e^{i2\pi S_n^z} |\psi\rangle = e^{i2\pi S} |\psi\rangle$, since $|\psi\rangle$ contains only spin-S states, and $e^{i2\pi S^z}$ yields ± 1 depending on whether S is integer or half-integer. Thus, the equation for the overlap takes the form

$$z_1 = e^{i2\pi l(S-M)} z_1. \tag{1.50}$$

From that equation it is clear that l(S-M) = integer is a necessary condition for the overlap z_1 to be nonzero. Thus, for l(S-M) = half-odd-integer the system is either gapless, or our assumption that $T_l|\psi_0\rangle = |\psi_0\rangle$ is wrong.

The spin-S Heisenberg chain in its ground state corresponds to l = 1 and M = 0. From the above theorem it follows that, if a spontaneous breaking of the translational symmetry is excluded, a spin-S Heisenberg chain can only be gapped if S is integer. We will come back to this result later in Sect. 1.6 since it establishes also a connection to the phenomenon known as *magnetization plateau*; actually, the integer spin chain ground state with the Haldane gap is the simplest example of a magnetization plateau at M = 0.

1.3.3 The AKLT Model and Valence Bond Solid States

Although the large-S NLSM description allows one to get some basic understanding for the S = 1 chain, chains with low integer S exhibit several important features which go beyond the large-S limit. These deficiencies are to some extent filled by the additional insight obtained from the so-called *valence bond solid* (VBS) models. The prototype of these models was proposed by Afleck, Kennedy, Lieb, and Tasaki [123] and is thus known as the *AKLT model*. In the following we introduce this model and use it as a starting point

to discuss the matrix product representation and an approximate treatment of excitations in the Haldane chain.

Let us introduce the *projector* operator $P_{12}^{J=2}$ which projects the states of two S = 1 spins S_1 , S_2 onto the subspace with the total spin J = 2. Consider the Hamiltonian defined in terms of this projector:

$$\mathcal{H} = \frac{1}{12} \sum_{i} \{ P_{i,i+1}^{(J=2)} - 8 \} = \sum_{i} \mathbf{S}_{i} \mathbf{S}_{i+1} + \frac{1}{3} (\mathbf{S}_{i} \mathbf{S}_{i+1})^{2}.$$
(1.51)

Obviously, the minimum energy is obtained for a state with the property that the total spin of any two neighboring spins is never equal to 2. Such a state can be constructed by regarding every S = 1 as a composite object consisting of two symmetrized $S = \frac{1}{2}$ spins, and linking each $S = \frac{1}{2}$ spin to its neighbor from the nearest site with a singlet bond, see Fig. 1.9a. Remarkably, uniform VBS states can be constructed in the same way for any integer S (Fig. 1.9b), while for half-integer S only dimerized VBS states are possible. For periodic boundary conditions the ground state is unique and is a global singlet, while for open boundary conditions there are two free $\frac{1}{2}$ spins at the open ends of the chain, so that the ground state is fourfold degenerate and consists of a singlet and of the so-called Kennedy triplet [124].



Fig. 1.9. Valence bond solid (VBS) wave functions: (a) the ground state (1.52) of the S = 1 AKLT model (1.51); (b) S = 2 VBS state

The AKLT model (1.51), which can be obviously generalized for higher S, serves as a good example visualizing the nature of the Haldane phase.

The S = 1 VBS state, taken as a variational trial wave function, yields for the Haldane chain the ground state energy per spin $E = -\frac{4}{3}$, rather close to the numerically obtained value $E \simeq -1.40$ [102].

Though the construction looks simple, it seems to be rather a nontrivial task to write down the VBS wave function in terms of the original spin states. There exists, however, a simple and elegant representation of VBS wavefunctions in the language of *matrix product states* [125, 126]. The AKLT wave function can be presented in the following form:

$$|\Psi\rangle = \operatorname{Tr}(g_1 g_2 \cdots g_N), \qquad g_n^{AKLT} = \frac{1}{\sqrt{3}} \begin{pmatrix} -|0\rangle_n - \sqrt{2}|-\rangle_n \\ \sqrt{2}|+\rangle_n & |0\rangle_n \end{pmatrix}, \quad (1.52)$$

where $|\mu\rangle_n$ denotes the state of the spin S = 1 at site n with $S^z = \mu$.

Indeed, it is easy to show that a product of any two matrices g_1g_2 does not contain states with the total spin J = 2, which is exactly the property of the AKLT wave function. The trace corresponds to periodic boundary conditions, and the four matrix elements of $\Omega = g_1 g_2 \cdots g_N$ are nothing but the four degenerate ground states of the open chain. A similar representation exists for higher-*S* VBS states [127].

The matrix product (MP) formulation is remarkable since it allows to write complicated states in a factorized (product) form. Technically, averages over VBS states can be easily calculated using the transfer matrix technique [127], e.g., for any operator \hat{L}_{12} involving two neighboring spins one has

$$\langle \Psi | \hat{L}_{12} | \Psi \rangle = Tr(G^{N-2}M_{12}),$$

$$G = g_i^* \otimes g_i, \quad M_{12} = (g_1g_2)^* \otimes \hat{L}_{12}(g_1g_2), \quad (1.53)$$

where \otimes denotes the direct (tensor) product of matrices.

The correlation function of the AKLT model for an infinite chain is explicitly given by

$$\langle S_n^{\alpha} S_{n'}^{\beta} \rangle = (-1)^{|n-n'|} (4/3) \ e^{-|n-n'| \ln 3} \delta_{\alpha\beta}; \tag{1.54}$$

for finite chains the free spins at the edges give an additional contribution which also decays exponentially when moving away from the boundary [128]. All correlations decay purely exponentially, which is a peculiarity of the AKLT model connected to the fact that it is a special *disorder point* where the so-called dimensional reduction of the generic D = 2 Ornstein-Zernike behavior (1.49) takes place [129]. The correlation length of the AKLT model $\xi = 1/\ln 3$ is very short in comparison with $\xi \simeq 6.0$ in the Haldane chain, [102]. This means, that despite the qualitative similarity to the ground state of the S = 1 Haldane chain, quantitatively the AKLT state is rather far from it. However, one may say that S = 1 Haldane chain and the AKLT model are in the same phase, i.e., in any reasonable phase space the points corresponding to those two models can be connected by a line which does not cross any phase boundary. Respectively, those two models can be said to belong to the same *universality class* in the sense that corresponding quantum phase transitions caused by changing some parameter in the general phase space occur at the same phase boundary and thus have the same universal behavior.

The MP representation makes it easy to see the presence of the string order in the VBS wave function. Since the elementary matrix g_i can be rewritten through the Pauli matrices σ^{μ} as

$$g_i^{AKLT} = 1/\sqrt{3}(\sigma^+|-\rangle_i + \sigma^-|+\rangle_i - \sigma^0|0\rangle_i), \qquad (1.55)$$

it is clear that, since $(\sigma^{\pm})^2 = 0$ and $\sigma^+ \sigma^0 = -\sigma^+$, the ground state contains only such spin states where the $|+\rangle$ must be followed by a $|-\rangle$, with an arbitrary number of $|0\rangle$ in between. The "diluted AF order" is thus perfect in the AKLT model. The AKLT state is rotationally invariant, and states $|0\rangle$, $|+\rangle$ and $|-\rangle$ appear with the equal probability of 1/3. Nonzero contribution to the correlator (1.42) comes only from states with no $|0\rangle$ at sites n and n', so that the value $\mathcal{O}_1^{AKLT} = 4/9$, which is the maximal value for a rotational invariant state, to be compared with $\mathcal{O}_1^{\text{Hald}} \simeq 0.37$ for the Haldane chain [104].

The hidden order, together with the fourfold degeneracy of the ground state for open chain, is a characteristic feature of the Haldane phase for S = 1 chains. This provides an elegant way of detecting the Haldane state [130]: doping a S = 1 Haldane chain with Cu^{2+} ions having spin $\frac{1}{2}$, one breaks it effectively into finite pieces, and effectively free $S = \frac{1}{2}$ spins are created at the edges adjacent to the impurity site. The resulting three spins $\frac{1}{2}$ are bound together by a weak host-impurity interaction, forming a loose cluster practically decoupled from the bulk of the chain. In applied magnetic field, resonant transitions between the cluster levels should be visible inside the Haldane gap. Such a response was successfully observed in the ESR experiment on Cu-doped NENP [130], confirming that the system is in the Haldane phase.

Excitations in the AKLT Chain

The lowest excitation above the singlet ground state of the Haldane chain is known to be a massive triplet with the total spin equal to 1. Creating such an excitation may be visualized as replacing one of the singlet links in the AKLT state by a triplet one. The resulting trial wave function for a triplet excitation with $S^z = \mu$ at site *n* can be written down as follows:

$$|\mu, n\rangle = \text{Tr}\{g_1^{AKLT} g_2^{AKLT} \dots g_{n-1}^{AKLT} (g_n^{1\mu}) g_{n+1}^{AKLT} \dots g_N^{AKLT}\}, \qquad (1.56)$$

where $g^{(1\mu)}$ is in the most general case defined as

$$g^{(1\mu)} = a\sigma^{\mu} \cdot g^{AKLT} + bg^{AKLT} \cdot \sigma^{\mu}, \qquad (1.57)$$

the ratio a/b being a free parameter. States $|\mu, n\rangle$ with different n are generally not orthogonal. However, one may achieve such an orthogonality by setting a/b = 3 [131].

Those states are in fact solitons in the string order [109–112]. One can straightforwardly check that in the soliton state $|r, n\rangle$ the string order correlators $\mathcal{O}_1^{r'}(l, l')$ with $r' \neq r$ change sign when n gets inside the (l, l') interval, while $\mathcal{O}_1^{r}(l, l')$ remains insensitive to the presence of the soliton. The variational dispersion relation for such a soliton takes an especially simple form for the AKLT model [132]:

$$\varepsilon(k) = \frac{10}{27}(5+3\cos k).$$
 (1.58)

The one-particle gap $\Delta = \varepsilon(k = \pi)$ is at $k = \pi$, and the overall structure of excitation spectrum is qualitatively very similar to that of the Haldane chain. Numerical analysis [103, 112] confirms that the above picture of excitations, constructed for the AKLT model, remains qualitatively correct in case of the S = 1 Haldane chain as well, also in anisotropic case [133].

The difference between the ground states of the Haldane chain and of the AKLT model may be visualized as follows: the Haldane chain contains a finite number of bound pairs of solitons with opposite spin, which reduce the hidden order and renormalize the excitation energy [134].

1.3.4 Spin Chains with Alternating and Frustrated Exchange

If the exchange integral is allowed to alternate along the chain, i.e., $J_n = J[1 + (-1)^n \delta]$, the NLSM analysis shows [135] that the topological term (1.48) gets multiplied by $(1 - \delta)$. The theory is gapless if $2\pi S(1 - \delta) = \pi \mod(2\pi)$, which yields 2S critical points if $\delta \in [-1; 1]$. The same conclusion is supported by the VBS approach which allows exactly 2S + 1 different dimerized VBS states for a given S, so that there are 2S transitions between them. Numerically, such transitions were observed in chains with S up to 2 [136].

Recently, a dimerized S = 1 VBS state was detected in the ESR experiment on Zn-doped NTENP [137]. The idea of the experiment was similar to that of detecting the Haldane state: due to the dimerized nature of the ground state, effective free S = 1 spins emerge on doping at the edges adjacent to the impurities, and the corresponding resonance response can be measured.

If one adds a small frustrating next-nearest-neighbor interaction j, the 2S critical points can be expected to continue as critical lines in the (j, δ) plane. In the strong frustration region, however, little is known, except for the cases $S = \frac{1}{2}$ and S = 1.

In the $S = \frac{1}{2}$ case there is a single critical line $\delta_c = 0$ extending up to the point $j \simeq 0.24$, and continuing till $j = \infty$ as a first-order line [65]. For S = 1there are two symmetrical lines $\delta = \pm \delta_c(j)$, with $\delta_c(0) \simeq 0.25$ [136], which, according to the numerical results [138, 139], extend up to about $j \simeq 0.2$ as second-order transition lines, continue afterwards as first-order ones and cross the symmetry line $\delta = 0$ at a finite $j \simeq 0.75$. The symmetry line (i.e., a frustrated chain without alternation) was studied in [131, 140] and the point $j_c \simeq 0.75$ was identified as that of the first-order "connectivity transition" from the Haldane phase to the so-called "double Haldane" phase. The string order (1.42) disappears discontinuously at $j > j_c$ [140], signaling a breakdown of the Haldane phase (Fig. 1.10b).

The "double Haldane" phase at $j > j_c$ can be visualized (see Fig. 1.10a) as a VBS state consisting of two interconnected AKLT chains [131]; the corresponding order parameter can be written as

$$\mathcal{O}_{2}^{\alpha}(n,n') = \left\langle -S_{n-1}^{\alpha}S_{n}^{\alpha} e^{i\pi\sum_{l=n+1}^{n'-1}S_{l}^{\alpha}} S_{n'}^{\alpha}S_{n'+1}^{\alpha} \right\rangle, \qquad \alpha = x, y, z, \quad (1.59)$$

and turns out to emerge discontinuously at $j > j_c$ (Fig. 1.10b). It is, however, not clear at present how the "double Haldane" phase is connected to the



Fig. 1.10. (a) visual interpretation of the "double-Haldane" phase; (b) behavior of string order parameters (1.42) and (1.59) on the frustration j [131]

dimerized phase: the string order (1.59) was found to survive in the dimerized phase as well [141].

1.3.5 Frustrated Chains with Anisotropy: Quantum Chiral Phases

In recent few years, the problem of possible nontrivial ordering in frustrated quantum spin chains with easy-plane anisotropy has attracted considerable attention [142–146]. The simplest model of this type is described by the Hamiltonian:

$$\mathcal{H} = J \sum_{n} \{ (\boldsymbol{S}_{n} \boldsymbol{S}_{n+1})_{\Delta} + j (\boldsymbol{S}_{n} \boldsymbol{S}_{n+2})_{\Delta} \}, \qquad (1.60)$$

where $(\mathbf{S}_1\mathbf{S}_2)_{\Delta} \equiv S_1^x S_2^x + S_1^y S_2^y + \Delta S_1^z S_2^z$, and $0 < \Delta < 1$ is the anisotropy parameter.

In the classical ground state of (1.60) spins always lie in the easy plane (xy), i.e. in terms of angular variables θ , φ for the classical spins $(S_n^x + iS_n^y = S \sin \theta_n e^{i\varphi_n}, S_n^z = \cos \theta_n)$ one has $\theta = \frac{\pi}{2}$. For $j < \frac{1}{4}$ the alignment of spins is antiferromagnetic, $\varphi_n = \varphi_0 + \pi n$, and for $j > \frac{1}{4}$ one obtains an incommensurate helical structure with $\varphi_n = \varphi_0 \pm (\pi - \lambda_0)n$, where $\lambda_0 = \arccos(1/4j)$, and the \pm signs above correspond to the two possible chiralities of the helix.

The classical *isotropic* $(\Delta = 1)$ system has for $j > \frac{1}{4}$ three massless modes with wave vectors q = 0, $q = \pm \delta$, where $\delta \equiv \pi - \lambda_0$ is the pitch of the helix. The effective field theory for the isotropic case is the so-called SO(3) nonlinear sigma model, with the order parameter described by the local rotation matrix [148, 149].

Quantum fluctuations make the long-range helical order impossible in one dimension, since it would imply a spontaneous breaking of the continuous inplane symmetry; in contrast to that, the existence of the finite vector chirality

$$\boldsymbol{\kappa}_n = \langle (\boldsymbol{S}_n \times \boldsymbol{S}_{n+1}) \rangle \tag{1.61}$$

is not prohibited by the Coleman theorem, as first noticed by Villain [151]. Positive (negative) chirality means, that spins on average prefer to rotate to the left (right), respectively, thus the discrete symmetry between left and right is spontaneously broken in the chiral phase. Nersesyan *et al.* [142] predicted the existence of a gapless *chiral phase* for $S = \frac{1}{2}$ in the $j \gg 1$ limit, using the bosonization technique combined with a subsequent mean-field-type decoupling procedure. Except having the chiral order, this phase is characterized by the power-law decaying incommensurate in-plane spin correlations of the form $\langle S_0^+ S_n^- \rangle \propto n^{-\eta} e^{iQn}$, where Q is very close to π in the limit $j \gg 1$, and $\eta = \frac{1}{4}$ for $S = \frac{1}{2}$ [142].

Early attempts [143, 145] to find this *chiral gapless* phase in numerical calculations for $S = \frac{1}{2}$ were unsuccessful. At the same time, to much of surprise, DMRG studies for frustrated S = 1 chain [145, 146] have shown the presence of *two* different types of chiral phases, *gapped* and *gapless*.

The model (1.60) was studied analytically in the large-S limit and for j close to the classical Lifshitz point $\frac{1}{4}$ by mapping it to a planar helimagnet [147, 152]. This mapping is based on the fact that in presence of anisotropy the modes with $q = \pm \delta$ acquire a finite mass and can be integrated out. It was shown that the existence of two types of chiral phases is not specific for S = 1, but is a generic large-S feature for integer S [147]. The predicted large-S phase diagram for integer S is shown in Fig. 1.11. Later large-S study [152] has shown that the chiral gapped phase should be absent for half-integer S, due to the effect of the topological term.

In subsequent works, chiral phases were numerically found for $S = \frac{1}{2}$, [150, 153] as well as for $S = \frac{3}{2}$ and S = 2 [150]; the resulting phase diagrams



Fig. 1.11. Predicted phase diagram of frustrated anisotropic chains with integer S in the large-S approximation, according to [147]

are shown in Fig. 1.12 and one can see that there is a qualitative agreement with the predictions of the large-S theory. The predicted dependence of the critical exponent η on j in the vicinity of the transition into a chiral phase, $\eta \propto \frac{1}{S\sqrt{j-1/4}} \rightarrow \frac{1}{4}$ at $j \rightarrow j_c$, also agrees qualitatively with the numerical results of [150]. However, the large-S theory is unable to describe the transition into the dimerized phase for half-integer S.



Fig. 1.12. Phase diagrams of frustrated anisotropic chains with $S = \frac{1}{2}$, 1, $\frac{3}{2}$ and 2, obtained by means of DMRG [150]

Another theoretical approach using bosonization [154] suggests that the phase diagram for integer and half-integer S should be very similar, with the only difference that the Haldane phase gets replaced by the dimerized phase in the case of half-integer S. This is in contradiction with the recent numerical results [150] indicating that the chiral gapped phase is absent for half-integer S. On the other hand, the bosonization prediction of the asymptotic value of the critical exponent, $\eta \to 1/(8S)$ at $j \to \infty$, agrees well with the numerical data.

There are indications [155] that chiral order may have been found experimentally in the 1D molecular magnet $Gd(hfac)_3NITiPr$.

1.4 $S = \frac{1}{2}$ Heisenberg Ladders

Spin ladders consist of two or more coupled spin chains and thus represent an intermediate position between one- and two-dimensional systems. The prototype of a spin ladder is shown in Fig. 1.13a and consists of two spin chains (legs) with an additional exchange coupling between spins on equivalent positions on the upper and lower leg (i.e. on rungs). The interest in spin ladders started with the observation that this ladder with standard geometry and antiferromagnetic couplings is a spin liquid with a singlet ground state and a Haldane type energy gap even for S = 1/2 [156]. More generally, spin ladders with an arbitrary number of antiferromagnetically coupled chains and arbitrary spin value S extend the class of spin liquids: For half-odd-integer spin and an odd number of legs they are gapless, whereas they exhibit a Haldane type energy gap otherwise (for a review of the early phase of spin ladder research see [11] and for a review of experiments and materials see [157]). Spin ladders are realized in a number of compounds and interest in these materials was in particular stimulated by the hope to find a new class of high temperature superconductors. However, so far only two SrCuO spin ladder materials were found which become superconducting under high pressure: T_c is about 10 K for $Sr_{0.4}Ca_{13.6}Cu_{24}O_{41}$ at 3 GPa pressure [158]. Nevertheless, theoretical interest continued to be strong since generalized spin ladder models cover a wide range of interesting phenomena in quantum spin systems and on the other hand allow to study in a reduced geometry interacting plaquettes of quantum spins identical to the CuO_2 plaquettes which are the basic building blocks of HTSC's. In this section we will concentrate on reviewing the properties of spin ladder models which connect seemingly disjunct quantum spin models.



Fig. 1.13. (a) generic spin ladder with only "leg" and "rung" exchange interactions J_L , J_R ; (b) zigzag spin ladder

1.4.1 Quantum Phases of Two-Leg S = 1/2 Ladders

The prototype of quantum spin ladders has the geometry shown in Fig. 1.13a and is defined by the Hamiltonian

$$\mathcal{H} = \sum_{n} \sum_{\alpha=1,2} J_{\mathrm{L}} \boldsymbol{S}_{n,\alpha} \cdot \boldsymbol{S}_{n+1,\alpha} + \sum_{n} J_{\mathrm{R}} \boldsymbol{S}_{n,1} \cdot \boldsymbol{S}_{n,2}$$
(1.62)

with exchange energies $J_{\rm L}$ along the legs and $J_{\rm R}$ on rungs. The 'standard' ladder results for equal antiferromagnetic exchange $J_{\rm L} = J_{\rm R} = J > 0$. Whereas the corresponding classical system has an ordered ground state of the Néel type the quantum system is a spin liquid with short range spin correlations, $\xi \approx 3.2$ (in units of the spacing between rungs) and an energy gap $\Delta \approx 0.5 J_{\rm R}$ [159,160] at wave vector π . Regarding the similarity to the Haldane chain indicated by these properties it was therefore tempting to speculate that the ladder gap is nothing but the Haldane gap of a microscopically somewhat more complicated system. In order to discuss this speculation we consider the system of (1.62) with varying ratio $J_{\rm R}/J_{\rm L}$. In the strong coupling limit with $J_{\rm R}/J_{\rm L}$ positive and large, the ladder reduces to a system of noninteracting dimers with the dimer excitation gap $\Delta_{\text{dimer}} = J_{\text{R}}$. With increasing $J_{\rm L}$ the gap decreases to become $\Delta \approx 0.4 J_{\rm R}$ in the weak coupling limit [161,162]. On the other hand, for large negative values, the formation of S = 1 units on rungs is favored and the system approaches an antiferromagnetic S = 1 chain (with effective exchange $\frac{1}{2}J_{\rm L}$). However, these two simple and apparently similar limiting cases are separated by the origin, $J_{\rm R} = 0$, corresponding to the gapless case of two independent S = 1/2 chains. The relation between ladder gap and Haldane gap therefore does not become clear by this simple procedure (see the early discussion by Hida [163]).

Before we approach this point in more detail, we shortly consider the ladder Hamiltonian (1.62) for the alternative case of ferromagnetically interacting legs, $J_{\rm L} < 0$: The classical ground state then is the state of two chains with long range ferromagnetic order, oriented antiparallel to each other. One would speculate that this ferromagnetic counterpart of the standard ladder is less susceptible to quantum fluctuations since without rung interactions the ground state for S = 1/2 is identical to the classical ground state. This is, however, not the case: An arbitrarily small amount of (antiferromagnetic) rung exchange leads to the opening up of a gap as shown by analytical [164–166] and numerical [167] methods. The situation is somewhat more involved (and interesting) when the exchange interactions are anisotropic: up to some finite rung coupling the classical ground state survives for an anisotropy of the Ising-type in the leg interactions and a spin liquid ground state of the Luttinger liquid type appears for leg anisotropy of the XY type [165, 166].

The relation between Haldane and ladder gap can be clarified when the somewhat generalized model for a S = 1/2 ladder shown in Fig. 1.13b, with the Hamiltonian

$$\mathcal{H} = \sum_{n} \sum_{\alpha=1,2} J_L \boldsymbol{S}_{n,\alpha} \cdot \boldsymbol{S}_{n+1,\alpha} + \sum_{n} \left(J_1 \, \boldsymbol{S}_{n,1} \cdot \boldsymbol{S}_{n,2} + J_2 \, \boldsymbol{S}_{n,2} \cdot \boldsymbol{S}_{n+1,1} \right)$$
(1.63)

is studied. This model is mostly known under the name of zigzag ladder, i.e. two Heisenberg chains with zigzag interactions, but it can be viewed alternatively as a chain with alternating exchange J_1 , J_2 and NNN interactions J_L . If either J_1 or J_2 vanishes the Hamiltonian reduces to the ladder geometry with two legs and rungs. For $J_1 = J_2$, the model reduces to the Heisenberg chain with NNN interactions already discussed in Sect. 1.2, including the quantum phase transition from the Heisenberg chain universality class to the (twofold degenerate and gapped) dimer crystal ground state at $J_1 = J_2 = \alpha_c^{-1} J_L$ (with $\alpha_c \simeq 0.2411$) and the Majumdar-Ghosh point $J_1 = J_2 = 2J_L$ with two degenerate ground states, see Sect. 1.2.6 above. Upon including alternation, $J_1 \neq J_2$, the Majumdar-Ghosh point extends into two Shastry-Sutherland lines [168], $J_2 = \frac{1}{2}$ for $J_1 > \frac{1}{2}$ and $J_1 = \frac{1}{2}$ for $J_2 > \frac{1}{2}$: If the exchange coupling along the chain alternates between J_1 on even bonds and $J_2 < J_1$ on odd bonds, $|0_I\rangle$ continues to be the ground state for $J_2 = \frac{1}{2}$ as long as $J_2 > -1$.

It is instructive to study this more general model introduced by White [169], for several reasons: The ground state phase diagram for various combinations of the variables J_1 , J_2 , J_L allows to discuss the relations between a number of seemingly different models by continuous deformation of the interaction parameters [169–171] and it serves as an instructive example for quantum phase transitions depending on the parameters in interaction space. Moreover it allows to make contact to real quasi 1D materials by showing the position in this diagram in rough correspondence to their interaction parameters.

In the following we present and discuss three ground state phase diagrams, in order to cover (partly overlapping) the full phase space in the variables J_1, J_2, J_L . Evidently the phase diagrams are symmetric under exchange of J_1 and J_2 and we will discuss only one of the two possible cases.

(a) Figure 1.14a shows the phase diagram J_2 vs J_1 , assuming a finite value of $J_{\rm L} > 0$ as energy unit. It has been established by various methods that the only phase transition lines occur at $J_2 = -2J_1/(2+J_1)$ (transition to the ferromagnetically ordered ground state) and along the line $J_1 = J_2 > -4$. This line is a line of first order quantum phase transitions for $0 < J_1 = J_2 < \alpha_c^{-1}$ and of second order quantum phase transitions for $J_1 = J_2 > \alpha_c^{-1}$ (in the following we use finite value of $J_{\rm L} > 0$ as energy unit and restrict to the $J_1 > J_2$ half of the plane).

The origin $J_1 = J_2 = 0$ corresponds to the gapless case of *two* noninteracting Heisenberg chains, whereas on the line $J_1 = J_2 > 0$ one has *one* S = 1/2 Heisenberg chain with NNN interaction. This line separates two distinct gapped regimes, each containing the limit of noninteracting dimers $J_1 \to \infty$ resp. $J_2 \to \infty$, the standard ladder, an effective S = 1 chain and the Shastry-Sutherland (SS) line.

The concept of string order can be extended to ladders [172, 173] introducing two complementary string order parameters in the $J_1 - J_2$ phase diagram:

$$\mathcal{O}_{\text{lad},1}^{\alpha}(n,m) = \left\langle -\left(S_{n,1}^{\alpha} + S_{n,2}^{\alpha}\right) e^{i\pi\sum_{j=n}^{m-1}\left(S_{j,1}^{\alpha} + S_{j,2}^{\alpha}\right)} \left(S_{m,1}^{\alpha} + S_{m,2}^{\alpha}\right) \right\rangle, \quad (1.64)$$



Fig. 1.14. Phase diagrams of the $S = \frac{1}{2}$ zigzag ladder: (a) $J_L = 1$, (b) $J_1 = 1$, (c) $J_1 = -1$. Solid (dashed) lines correspond to the second (first) order transitions, respectively

$$\mathcal{O}_{\mathrm{lad},2}^{\alpha}(n,m) = \left\langle -\left(S_{n,1}^{\alpha} + S_{n+1,2}^{\alpha}\right) e^{i\pi \sum_{j=n}^{m-1} \left(S_{j,1}^{\alpha} + S_{j+1,2}^{\alpha}\right)} \left(S_{m,1}^{\alpha} + S_{m+1,2}^{\alpha}\right) \right\rangle.$$
(1.65)

For $J_1 > J_2$ (phase D2) singlets are found preferably on the rungs and the remaining antiferromagnetic leg exchange then leads to a tendency towards triplets, i.e. S = 1 units on diagonals. This implies a vanishing value for $\mathcal{O}_{\mathrm{lad},1}$ whereas a finite string order parameter $\mathcal{O}_{\mathrm{lad},2}$ develops. This type of string order characterizes the standard ladder $(J_1 = 1, J_2 = 0)$ and becomes identical with the S = 1 chain string order parameter for $J_2 \to -\infty$. The complementary situation is true for $J_1 < J_2$: rungs and diagonals as well as $\mathcal{O}_{\mathrm{lad},1}$ and $\mathcal{O}_{\mathrm{lad},2}$ exchange their roles. In the field theoretic representation of the generalized ladder [13,174,175] $\mathcal{O}_{\mathrm{lad},1}$ and $\mathcal{O}_{\mathrm{lad},2}$ correspond to Ising order resp. disorder parameters. Both order parameters become zero on the line $J_1 = J_2$ for $J_1 = J_2 > \alpha_c^{-1}$ (gapless line) whereas they change discontinuously following the discontinuous change in ground state when the line $J_1 = J_2$ for $J_1 = J_2 < \alpha_c^{-1}$ (line with two degenerate ground states) is crossed.

Thus it is possible to deform various gapped models, noninteracting dimers, the standard ladder and the S = 1 Haldane chain, continuously into each other without closing the gap if one stays on the same side of the line $J_1 = J_2$. Then the ladder gap evolves into the dimer gap when the rung coupling increases to infinity and the dimer gap evolves into the Haldane gap when two dimers on neighboring rungs interact ferromagnetically via J_2 , forming S = 1 units on diagonals. However, when the standard ladder is deformed into a S = 1 chain by changing rung dimers from antiferromagnetic to strongly ferromagnetic, one moves to a different symmetry class since the line $J_1 = J_2$ is crossed.

For ferromagnetic couplings $J_1, J_2 < 0$ there is a regime of disorder due to competing interactions before ferromagnetic order sets in. This applies in particular to the limit $-4 < J_1 = J_2 < 0$, a ferromagnetic chain with AF NNN exchange. It is usually taken for granted that the corresponding ground state of this frustrated chain is in an incommensurate phase and gapless; however, a recent interesting speculation [176] suggests the presence of a tiny but finite gap on some part of this line.

(b) In Fig. 1.14b the phase diagram in the variables J_2 vs J_L is presented, assuming a finite value of $J_1 > 0$ as energy unit. This choice of variables displays most clearly the neighborhood of the dimer point (the origin in this presentation) and the situation when ferromagnetic coupling is considered on the legs and on one type of inter-leg connections. The dividing line between the two dimer/Haldane phases D1 and D2 appears now as the line $J_2 = 1$ with the end of the gapless phase at $J_L = \alpha_c$ and the Majumdar-Ghosh point at $J_L = \frac{1}{2}$. The gap on this line starts exponentially small from zero at the Kosterlitz Thouless transition at $J_L = \alpha_c$, goes through a maximum at $J_L \approx 0.6$ and drops to zero exponentially for $J_L \to \infty$ (two decoupled chains) [149, 177].

The Shastry-Sutherland (SS) lines $J_L = \frac{1}{2}J_2$ (in D2) resp. $J_L = \frac{1}{2}$ in D1 are to be considered as disorder lines where spin-spin correlations in real space become incommensurate [178, 179]. The precise properties in the incommensurate regime beyond these lines have not been fully investigated up to now. The SS line extends into the range of ferromagnetic couplings and (in D2) ends at $J_L = \frac{1}{2}J_2 = -1$. This point lies on the boundary of the ferromagnetic phase, $J_2 = -2J_L/(1 + 2J_L)$. This boundary is obtained from the instability of the ferromagnetic state against spin wave formation. There are indications that ground states on this line are highly degenerate: states with energies identical to the ferromagnetic ground state are explicitly known for $J_1 = J_L = -1$ (end of the SS line, dimers on J_1 bonds), for $J_1 = J_L = -\frac{3}{2}$ (a matrix product ground state, see Sect. 1.4.2) and for a family of states which exhibit double chiral order as studied in ref. [180].

As mentioned before, the ladder is gapless on the line $J_2 = 1, J_L < 0$ (antiferromagnetic Heisenberg chain with ferromagnetic NNN exchange), but an infinitesimal alternation, $J_2 \neq 1$ drives it into the gapless phase, smoothly connected to the Haldane/dimer phase. At strongly negative values of J_1 the phase diagram of Fig. 1.14b shows the second order phase transition from the ferromagnetic to the antiferromagnetic S = 1 chain at $J_L = -\frac{1}{2}$.

(c) In Fig. 1.14c the phase diagram in the same variables J_2 vs J_L is shown, but assuming a finite *ferromagnetic* value of $|J_1| = -J_1 > 0$ as energy unit. This choice of variables allows to discuss the situation for two ferromagnetic couplings. The origin is identified as the limit of noninteracting spins 1 and the neighborhood of the origin covers both the ferro- as the antiferromagnetic S = 1 chain, depending on the direction in parameter space.

1.4.2 Matrix Product Representation for the Two Leg S = 1/2 Ladder

The matrix product representation introduced for the S = 1 chain above can be extended to ladders and is found to be a powerful approach to describe spin ladder ground states in the regime covered by the J_1 - J_2 phase space of the model of (1.63). It formulates possible singlet ground states as a product of matrices g_n referring to a single rung n, $|..\rangle = \prod_n g_n$. Matrices g_n as used in Sect. 1.3.3 are generalized to include the possibility of singlets on a rung and read [170]:

$$g_n(u) = u\widehat{1} |s\rangle_n + v(\frac{1}{\sqrt{2}}\sigma^-|t_-\rangle_n - \frac{1}{\sqrt{2}}\sigma^+|t_+\rangle_n + \sigma^z |t_0\rangle_n)$$

$$= \begin{pmatrix} u|s\rangle + v|t_0\rangle & -\sqrt{2}v|t_+\rangle\\ \sqrt{2}v|t_-\rangle & u|s\rangle - v|t_0\rangle \end{pmatrix}.$$
 (1.66)

(Note that the triplet part of (1.66) is equivalent to (1.55) up to a unitary transformation; here we keep the original nonation of [170].) We now show that the ground states of the Majumdar-Ghosh chain can be written in the form of a matrix product. This is trivially true for $|0\rangle_{II}$ which is obtained for u = 1, v = 0. It is also true for the state $|0\rangle_{I}$ if it is formulated in terms of the complementary spin pairs [2,3], [4,5]... used in $|0_{II}\rangle$: We start from the representation of a singlet as in (1.34, 1.35) and write

$$|0\rangle_{I} = \frac{1}{2^{N/2}} \sum_{\{\dots,s,s',t,\dots\}} \cdots \chi_{2p-1}(s) \ \epsilon^{s,s'} \ \chi_{2p}(s')$$
$$\times \ \chi_{2p+1}(t) \ \epsilon^{t,t'} \ \chi_{2p+2}(t') \ \chi_{2p+3}(r) \ \epsilon^{r,r'} \ \chi_{2p+4}(r') \cdots = \operatorname{Tr}\Big(\prod_{p} g_{p}\Big)$$

after defining the matrix with state valued elements

$$g_p(s,t) := \frac{1}{\sqrt{2}} \sum_{s'} \chi_{2p}(s) \ \chi_{2p+1}(s') \ \epsilon^{s',t}$$

to replace the singlet, (1.34) as new unit. The explicit form for g is

$$\frac{1}{\sqrt{2}} \begin{pmatrix} |\uparrow,\downarrow\rangle & -|\uparrow,\uparrow\rangle \\ |\downarrow,\downarrow\rangle & -|\downarrow,\uparrow\rangle \end{pmatrix}$$

which is identical to (1.66) with $u = v = 1/\sqrt{2}$.

1.4.3 Matrix Product States: General Formulation

The above construction of the matrix product ansatz for $S = \frac{1}{2}$ ladders can be generalized for arbitrary 1D spin systems [181]. Let $\{|\gamma S\mu\rangle\}$ be the complete set of the spin states of the elementary cell of a given 1D spin system, classified according to the total spin *S*, its *z*-projection μ and an (arbitrary) additional quantum number γ . Define the object *g* as follows:

$$g^{(jm)} = \sum_{\lambda q, S\mu} c_{\gamma} \langle jm | \lambda q, S\mu \rangle \, \widehat{T}^{\lambda q} | \gamma S\mu \rangle \,, \tag{1.67}$$

where $\langle jm|\lambda q, S\mu \rangle$ are the standard Clebsch-Gordan coefficients, c_{γ} are free *c*-number parameters, and $\hat{T}^{\lambda q}$ are irreducible tensor operators acting in some auxiliary space, which transform under rotations according to the \mathcal{D}^{λ} representation. Then it is clear that *g* transforms according to \mathcal{D}^{j} and thus can be assigned "hyperspin" quantum numbers jm. Then, building on those elementary objects g_i (where *i* denotes the *i*-th unit cell) one can construct wave functions with certain total spin almost in the same way as from usual spin states. For instance, for a quantum 1D ferrimagnet with the excess spin *j* per unit cell the state with the total spin and its *z*-projection both equal to Njwould have the form

$$|\Psi^{Nj,Nj}\rangle = \operatorname{Tr}_{\mathcal{M}}(\Omega_N), \quad \Omega_N = g_1^{(jj)} \cdot g_2^{(jj)} \cdots g_N^{(jj)}, \qquad (1.68)$$

where the trace sign denotes an appropriate trace taken over the auxiliary space. The choice of the auxiliary space \mathcal{M} determines the specific matrix representation of the operators $T^{\lambda q}$; the space \mathcal{M} can be always chosen in a form of a suitable decomposition into multiplets $\mathcal{M} = \sum_{\alpha J} \oplus \mathcal{M}^{\alpha J}$, and then the structure of the matrix representation is dictated by the Wigner-Eckart theorem:

$$\langle \alpha JM | T^{\lambda q} | \alpha' J'M' \rangle = \widetilde{T}_{\lambda, \alpha J, \alpha' J'} \langle JM | \lambda q, J'M' \rangle.$$
(1.69)

The reduced matrix elements $\widetilde{T}_{\lambda,\alpha J,\alpha' J'}$ and the coefficients c_{γ} are free parameters.

Matrix product states (MPS) are particularly remarkable because the matrices g_1g_2 , $g_1g_2g_3$, etc. all have the same structure (1.67) if they are constructed from the "highest weight" components $g^{(j,m=j)}$. This self-similarity is actually an indication of the deep connection of singlet MPS and the densitymatrix renormalization group technique, as first pointed out by Ostlund and Römmer [182] and developed later in works of Sierra *et al.* [183–185].

A Few Examples

In the simplest case of a two-dimensional $\mathcal{M} = \{|J = \frac{1}{2}, M\rangle\}$, the allowed values of λ are 0 and 1, and T^{1q} are just proportional to the usual Pauli matrices σ^q , and T^{00} is proportional to the unit matrix. If one wants the wavefunction to be a global singlet, the simplest way to achieve that is to have the construction (1.68) with j = 0. Then, for the case of S = 1 chain with one spin in a unit cell, one obtains exactly the formula (1.55), with no free parameters.

Higher-S AKLT-type VBS states can be also easily represented in the matrix product form. In this case one has to choose $\mathcal{M} = \{|S/2, M\rangle\}$, then the only possible value of λ is S, and, taking into account that $\langle 00|Sq, S\mu\rangle = \delta_{q,-\mu}(-1)^{S-\mu}$, we obtain

$$g_S = \sum_{\mu} (-1)^{S-\mu} T^{S,-\mu} |S,\mu\rangle.$$

For a generic quantum ferrimagnet, i.e., a chain of alternating spins 1 and $\frac{1}{2}$, coupled by antiferromagnetic nearest-neighbor exchange, the elementary unit contains now two spins. The ground state has the total spin $\frac{1}{2}$ per unit cell, then one would want to construct the elementary matrix $g^{1/2,1/2}$. If \mathcal{M} is still two-dimensional, the elementary matrix has according to (1.67) the following form:

$$g = \begin{pmatrix} (u-v)|\uparrow\rangle - |\frac{1}{2}\rangle & \sqrt{3}|\frac{3}{2}\rangle \\ -2v|\downarrow\rangle - |-\frac{1}{2}\rangle & (u+v)|\uparrow\rangle + |\frac{1}{2}\rangle \end{pmatrix},$$
(1.70)

where $|\uparrow\rangle$, $|\downarrow\rangle$ and $|\pm\frac{1}{2}\rangle$, $|\pm\frac{3}{2}\rangle$ are the cell states with the total spin $\lambda = \frac{1}{2}$ and $\lambda = \frac{3}{2}$, respectively.

1.4.4 Excitations in Two-Leg S=1/2 Ladders

The excitation spectrum in this simplest ladder type spin liquid is similar to that of a Haldane chain: The lowest excitation is a triplet band with minimum energy at $q = \pi$ and a continuum at q = 0. Since the ground state is a disordered singlet, a spin wave approach (which would result in a gapless spectrum) is inappropriate. In different regimes of the space of coupling constants, different methods have been developed to deal approximately with the low-lying excitations:

Weak Coupling Regime

In the *weak coupling regime*, close to two independent chains, the bosonization approach can be applied to decide whether the excitation is gapless or gapped. The standard situation is that the coupling between legs is relevant and a gap develops for arbitrarily small coupling. Some examples are: antiferromagnetic interactions in the standard rung geometry [177] (the gap is linear in $J_{\rm R}$, the numerical result is $\Delta \approx 0.4 J_{\rm R}$ [162]), antiferromagnetic interactions in the zigzag geometry [186], and antiferromagnetic interactions for isotropic ferromagnetic legs [165]. The gapless (Luttinger liquid) regime of the decoupled chains can survive, e.g. for ferromagnetic legs with XY-type anisotropy and antiferromagnetic coupling [166].

Strong Coupling Regime

In the strong coupling regime, close to the dimer limit the lowest elementary excitation develops from the excited triplet state of a dimer localized on one of the rungs which starts propagating due to the residual interactions. For the Hamiltonian of (1.63) the dispersion to first order is (we choose $J_1 \gg J_2$ to be the strong dimer interaction)

$$\omega(q) = J_1 + \left(J_L - \frac{1}{2}J_2\right)\cos q + J_1\left(\frac{3}{4}(\alpha_L - \frac{1}{2}\alpha_2)^2 + -\frac{1}{4}\alpha_2^2(1 + \cos q) - \frac{1}{4}(\alpha_L - \frac{1}{2}\alpha_2)^2\cos 2q \dots\right) \quad (1.71)$$

with $\alpha_L = J_L/J_1$ and $\alpha_2 = J_2/J_1$. The excitation gap is at either q = 0 (for $J_2 > 2J_L$ in the lowest order, alternating AF chain type spectrum) or $q = \pi$ ($J_2 < 2J_L$, ladder type spectrum). For a finite regime in the space of coupling constants an expansion in the dimer-dimer couplings leads to converging expressions for the low-energy frequencies. Expansions have now been carried out up to 14^{th} order by the methods of cluster expansion [68, 187, 188] and are convergent even close to the isotropic point.

We note two curiosities: In a small but finite transition regime, the minimum of the dispersion curve changes continuously from q = 0 to $q = \pi$ [187,189]; on the Shastry-Sutherland line, $\alpha_L = \alpha_2/2$ the energy of the mode at $q = \pi$ is known exactly, $\omega(q = \pi) = J_1$.

For nearly Heisenberg chains with NNN interaction and small alternation dimer series expansions have been used extensively to investigate further details of the spectra in e.g. $CuGeO_3$ [68]. Bound states for the standard spin ladder have been calculated to high order [190] and used to describe optically observed two-magnon states in $(La,Ca)_{14}Cu_{24}O_{41}$ [191].

The strong coupling approach has also been applied to describe interacting dimer materials such as KCuCl₃, TlCuCl₃ [192,193] with 3D interactions and $(C_4H_{12}N_2)Cu_2Cl_6$ (= PHCC) [194] with 2D interactions. These interactions are quantitatively important but not strong enough to close the spin gap and to drive the system into the 3D ordered state. The dimer expansions are much more demanding than in 1D, but nevertheless were done successfully up to 6th order [195, 196].

Bond Boson Operator Approach

This approach makes use of the representation of spin operators in terms of the so-called *bond bosons* [197]. On each ladder rung, one may introduce four bosonic operators s, t_a ($a \in (x, y, z)$) which correspond to creation of the singlet state $|s\rangle$ and three triplet states $|t_a\rangle$ given by

$$|s\rangle = \frac{1}{\sqrt{2}} (|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle), \quad |t_z\rangle = \frac{1}{\sqrt{2}} (|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle), \quad (1.72)$$
$$|t_x\rangle = -\frac{1}{\sqrt{2}} (|\uparrow\uparrow\rangle - |\downarrow\downarrow\rangle), \quad |t_y\rangle = \frac{i}{\sqrt{2}} (|\uparrow\uparrow\rangle + |\downarrow\downarrow\rangle),$$

Then the rung spin- $\frac{1}{2}$ operators $S_{1,2}$ can be expressed through the bond bosons as

$$\boldsymbol{S}_{1,2} = \pm \frac{1}{2} (s^{\dagger} \boldsymbol{t} + \boldsymbol{t}^{\dagger} s) - \frac{1}{2} i (\boldsymbol{t}^{\dagger} \times \boldsymbol{t}).$$
(1.73)

One may check that the above representation satisfies all necessary commutation relations, if the following *local* constraint is assumed to hold:

$$s^{\dagger}s + \boldsymbol{t}^{\dagger} \cdot \boldsymbol{t} = 1, \qquad (1.74)$$

which implies that the bond bosons are 'hardcore' (no two bosons are allowed to occupy one bond), and, moreover, exactly one boson must be present at each bond/rung. The constraint is easy to handle formally (e.g. in the path integral formulation), but practically one can do that only at the meanfield level [198], replacing the local constraint by a global one, i.e., (1.74) is assumed to be true only on average, which introduces rather uncontrollable approximations.

In a slightly different version of the bond boson approach [199], the vacuum state is introduced as corresponding to the state with fully condensed s bosons. Then for spin operators one obtains the formulae of the form (1.73) with s replaced by 1, and instead of the constraint (1.74) one has just a usual hardcore constraint $\mathbf{t}^{\dagger} \cdot \mathbf{t} = 0, 1$. This version is most useful in the limit of weakly coupled dimers (e.g., $J_1 \gg J_2, J_L$). Passing to the momentum representation, one obtains on the quadratic level the effective Hamiltonian of the form

$$\mathcal{H}_{eff} = \sum_{ka} A_k t_{k,a}^{\dagger} t_{k,a} + \frac{1}{2} B_k (t_{k,a}^{\dagger} t_{k,a}^{\dagger} + \text{h.c.}), \qquad (1.75)$$

where the amplitudes A_k , B_k are given by the expressions

$$B_k = (J_L - J_2/2)\cos(k), \qquad A_k = J_1 + B(k).$$
 (1.76)

Thus, neglecting the boson interaction, one obtains for the excitation energy

$$\omega(k) = \sqrt{J_1^2 + 2J_1\left(J_L - \frac{1}{2}J_2\right)\cos k}, \qquad (1.77)$$

which coincides with the corresponding RPA expression. Upon comparison to the full systematic series of the perturbation theory, one can see that (1.77) contains only the leading contributions at each cosine term $\cos(nk)$ of the complete series and misses the remaining terms starting in the second order [187].

The Hamiltonian (1.75) does not take into account any interaction between the bosons. One may argue that the most important contribution to the interaction comes from the hardcore constraint, which is effectively equivalent to the infinite on-site repulsion U.

The effect of the local hardcore constraint can be handled using the socalled *Brueckner approximation* as proposed by Kotov *et al.* [199]. In this approach, one neglects the contribution of anomalous Green's functions and obtains in the limit $U \to \infty$ the vertex function $\Gamma_{aa',ss'} = \Gamma(k,\omega)(\delta_{as}\delta_{a's'} + \delta_{as'}\delta_{a's})$, where k and $\hbar\omega$ are respectively the total momentum and energy of the incoming particles, with

$$\frac{1}{\Gamma(k,\omega)} = -\frac{1}{N} \sum_{q} \frac{Z_q Z_{k-q} u_q^2 u_{k-q}^2}{\omega - \Omega_q - \Omega_{k-q}}.$$
(1.78)

The corresponding normal self-energy $\Sigma(k,\omega)$ is

$$\Sigma(k,\omega) = (4/N) \sum_{q} Z_q v_q^2 \Gamma(k+q,\omega-\Omega_q)$$
(1.79)

Here \varOmega_k is the renormalized spectrum, which is found as a pole of the normal Green function

$$G(k,\omega) = \frac{\omega + A_k + \Sigma(-k,-\omega)}{(\omega - \Sigma_-)^2 - (A_k + \Sigma_+)^2 + B_k^2},$$
(1.80)

where $\Sigma_{\pm} \equiv \frac{1}{2} \{ \Sigma(k, \omega) \pm \Sigma(-k, -\omega) \}$. The quasiparticle contribution to the above Green function is given by

$$G(k,\omega) = \frac{Z_k u_k^2}{\omega - \Omega_k + i\varepsilon} - \frac{Z_k v_k^2}{\omega + \Omega_k - i\varepsilon}$$
(1.81)

which defines the renormalization factors Z_k , the Bogoliubov coefficients u_k , v_k and the spectrum Ω_k as follows [131]:

$$\Omega_{k} = \Sigma_{-} + E_{k}, \quad E_{k} = \{(A_{k} + \Sigma_{+})^{2} - B_{k}^{2}\}^{1/2}, \\
u_{k}^{2} = \frac{1}{2}\{1 + (A_{k} + \Sigma_{+})/E_{k}\}, \quad v_{k}^{2} = u_{k}^{2} - 1, \\
\frac{1}{Z_{k}} = 1 - \frac{\partial \Sigma_{-}}{\partial \omega} - \frac{(A_{k} + \Sigma_{+})}{E_{k}}\frac{\partial \Sigma_{+}}{\partial \omega}$$
(1.82)

where Σ_{\pm} and their derivatives are understood to be taken at $\omega = \Omega_k$. The system of equations (1.78), (1.79), (1.82) has to be solved self-consistently with respect to Z and Σ . This approach is valid as long as the boson density $\rho = \frac{3}{N} \sum_q Z_q v_q^2$ remains small, ensuring that the contribution of anomalous Green's functions is irrelevant [199].

It should be remarked that the original expressions of Kotov *et al.* [199] can be obtained from (1.82) as a particular case, assuming that $\Sigma(k,\omega)$ is almost linear in ω in the frequency interval $(-\Omega_k, \Omega_k)$; however, this latter assumption fails if one is far away from the phase transition, i.e. if the resulting frequency ω is not small comparing to J_1 .

The above way of handling the hardcore constraint is quite general and can be used in other problems as well, e.g., one can apply it to improve the results of using the variational soliton-type ansatz (1.56), (1.57) for the S = 1 Haldane chain [131].

Bound Domain Wall Approach

The low-lying excited states in spin ladders in the dimer phase can be discussed in a domain wall representation qualitatively rather similar to the antiferromagnetic Ising chain in Sect. 2.3. In the limit of a twofold degenerate ground state (i.e. on the line $J_1 = J_2 = J < \alpha_c^{-1} J_L$), excitations can be discussed in terms of pairs of domain walls, mediating between these two states [168].

Moving away from this line into the regime $J_1 \neq J_2$ where bond strengths alternate, a pair of domain walls feels a potential energy linear in the distance between them since the two dimer configurations now have different energies. As a consequence, all domain walls become bound with well defined dispersion $\omega(q)$. The frequency is lowest for the state originating from the simplest pair of domain walls, obtained by exciting one dimer leading to a triplet state. Thus one makes connection with the strong coupling limit and establishes that the free domain wall continuum upon binding develops into the sharp triplet excitation ('magnon') of the Haldane type. For a more quantitative description of the transition between bound and unbound limits, several variational formulations have been developed [189, 200, 201]. Of particular interest is the limit of $J_L \gg J_1$, i.e. weakly coupled gapless chains which can be studied by bosonization techniques [186]. The zigzag structure is responsible for a "twist" interaction which induces incommensurabilities in the spin correlations.

A particular simple example for a system with unbound domain walls is the Majumdar-Ghosh state $(J_1 = J_2 = 2J_L = J \text{ in (1.63)})$; a domain wall here means a transition from dimers on even bonds to dimers on odd bonds or vice versa and implies the existence of a free spin 1/2, justifying the name spinons for these excitations. For each free spin 1/2 the binding energy of half a dimer bond is lost, producing an energy gap J/2 which is lowered to a minimum value of J/4 at q = 0. For a chain with periodic boundary conditions the excitation spectrum consists of pairs of these spinons which, owing to isotropy, bind into 4 degenerate states, a triplet and a singlet. Because of the degeneracy of the two ground states these spinons can move independently (completely analogous to the domain walls of the Ising chain with small transverse interactions of Sect. 2.3), their energies therefore simply add and lead to an excitation continuum. For a finite range of wave vectors centered around $q = \pi$ bound states with lower energies exist [168, 200]. The excited state with lowest energy, however, remains the triplet/singlet at q = 0.

Moving away from the Majumdar-Ghosh point on the line with two degenerate ground states towards the quantum phase transition at $J_{\rm L} = J \alpha_c$, the energy of the spinons diminishes until they become gapless at the phase transition. Similar in spirit to the approach from the antiferromagnetic Ising phase, this is another way to approach the gapless excitation spectrum of the Heisenberg chain [202]. Since it preserves isotropy in spin space at each stage, it nicely demonstrates the fourfold degeneracy of the spinon spectrum with one triplet and one singlet, originating from the two independent spins 1/2.

1.4.5 Multileg Ladders

A natural generalization of the two-leg AF ladder is a general n-leg $S = \frac{1}{2}$ ladder model with all antiferromagnetic rung and leg couplings. Except being an interesting theoretical concept representing a system "in between" one and two dimensions, this model is realized in strontium copper oxides of the $Sr_{n-1}Cu_{n+1}O_{2n}$ family [11]. It turns out that the analogy between the regular two-leg $S = \frac{1}{2}$ ladder and the S = 1 Haldane chain can be pursued further, and n-leg ladders with odd n are gapless, while ladders with even n exhibit a nonzero spectral gap Δ [203,204]. One may think of this effect as cancellation of the topological terms coming from single $S = \frac{1}{2}$ chains [174, 204–206]. The problem can be mapped to the nonlinear sigma model [206] with the topological angle $\theta = \pi n$ and coupling constant $g \propto n^{-1}$, so that there is a similarity between the n-leg $S = \frac{1}{2}$ ladder and a single chain with S = n/2. The gap $\Delta \propto e^{-2\pi/g}$ vanishes exponentially in the limit $n \to \infty$, recovering the proper two-dimensional behavior.

Instructive numerical results are available for systems of up to 6 coupled chains: improving earlier DMRG studies [159], calculations for standard *n*-leg ladders using loop cluster algorithms [161,162] clearly show the decrease of the gap for *n* even (from 0.502 *J* for n = 2 to 0.160 *J* for n = 4 and 0.055 *J* for n = 6). Further detailed results by this method were obtained for correlation lengths and susceptibilities [162,207].

1.5 Modified Spin Chains and Ladders

Until now, we have considered only models with purely Heisenberg (bilinear) spin exchange. One should remember, however, that the Heisenberg Hamiltonian is only an approximation, and generally for S > 1/2 one has also "non-Heisenberg" terms such as $(\mathbf{S}_l \cdot \mathbf{S}_{l'})^m, m = 2, \ldots, 2S$ whose strength depends on the Hund's rule coupling. For $S = \frac{1}{2}$, exchange terms involving four or more spins emerge in higher orders of the perturbation theory in the Hubbard model. Those non-Heisenberg terms are interesting since they lead to a rather rich behavior, and even small admixture of such interactions may drive the system in the vicinity of a phase transition.

1.5.1 $S = \frac{1}{2}$ Ladders with Four-Spin Interaction

In case of a two-leg spin- $\frac{1}{2}$ ladder the general form of the isotropic translationally invariant spin ladder Hamiltonian with exchange interaction only between spins on plaquettes formed by neighboring rungs reads as

$$\mathcal{H} = \sum_{i} J_{R} \mathbf{S}_{1,i} \cdot \mathbf{S}_{2,i} + J_{L} \mathbf{S}_{1,i} \cdot \mathbf{S}_{1,i+1} + J'_{L} \mathbf{S}_{2,i} \cdot \mathbf{S}_{2,i+1}$$
(1.83)
+ $J_{D} \mathbf{S}_{1,i} \cdot \mathbf{S}_{2,i+1} + J'_{D} \mathbf{S}_{2,i} \cdot \mathbf{S}_{1,i+1} + V_{LL} (\mathbf{S}_{1,i} \cdot \mathbf{S}_{1,i+1}) (\mathbf{S}_{2,i} \cdot \mathbf{S}_{2,i+1})$
+ $V_{DD} (\mathbf{S}_{1,i} \cdot \mathbf{S}_{2,i+1}) (\mathbf{S}_{2,i} \cdot \mathbf{S}_{1,i+1}) + V_{RR} (\mathbf{S}_{1,i} \cdot \mathbf{S}_{2,i}) (\mathbf{S}_{1,i+1} \cdot \mathbf{S}_{2,i+1}),$

where the indices 1 and 2 distinguish lower and upper legs, and i labels rungs. The model is schematically represented in Fig. 1.15.



Fig. 1.15. A generalized ladder model with four-spin interactions

There is an obvious symmetry with respect to interchanging S_1 and S_2 on every other rung and simultaneously interchanging J_L , V_{LL} with J_D , V_{DD} . Less obvious is a symmetry corresponding to the so-called *spin-chirality dual* transformation [208]. This transformation introduces on every rung a pair of new spin- $\frac{1}{2}$ operators σ , τ , which are connected to the 'old' operators $S_{1,2}$ through

$$\boldsymbol{S}_{1,2} = \frac{1}{2}(\boldsymbol{\sigma} + \boldsymbol{\tau}) \pm (\boldsymbol{\sigma} \times \boldsymbol{\tau}). \tag{1.84}$$

Applying this transformation to the generalized ladder (1.83) generally yields new terms containing mixed products of three neighboring spins; however, in case of a symmetric ladder with $J_{L,D} = J'_{L,D}$ those terms vanish and one obtains the model of the same form (1.83) with new parameters

$$\begin{aligned} \widetilde{J}_{L} &= J_{L}/2 + J_{D}/2 + V_{LL}/8 - V_{DD}/8 \\ \widetilde{J}_{D} &= J_{L}/2 + J_{D}/2 - V_{LL}/8 + V_{DD}/8 \\ \widetilde{J}_{R} &= J_{R}, \qquad \widetilde{V}_{RR} = V_{RR} \\ \widetilde{V}_{LL} &= 2J_{L} - 2J_{D} + V_{LL}/2 + V_{DD}/2 \\ \widetilde{V}_{DD} &= -2J_{L} + 2J_{D} + V_{LL}/2 + V_{DD}/2 \end{aligned}$$
(1.85)

It is an interesting fact that all models having the product of singlet dimers on the rungs as their exact ground state are self-dual with respect to the above transformation, because the necessary condition for having the rung-dimer ground state is [209]

$$J_L - J_D = \frac{1}{4} (V_{LL} - V_{DD}).$$
(1.86)

It is worthwhile to remark that there are several families of generalized $S = \frac{1}{2}$ ladder models which allow an exact solution. First Bethe-ansatz solvable ladder models were those including three-spin terms explicitly violating the time reversal and parity symmetries (see the review [210] and references therein). Known solvable models with four-spin interaction include those constructed from the composite spin representation of the S = 1 chain [211], models solvable by the matrix product technique [209], and some special models amenable to the Bethe ansatz solution [212–214]. Among the models solvable by the matrix product technique, there exist families which connect smoothly the dimer and AKLT limits [215]. This proves that these limiting cases are in the same phase.

There are several physical mechanisms which may lead to the appearance of the four-spin interaction terms in (1.83). The most important mechanism is the so-called ring (four-spin) exchange. In the standard derivation based on the Hubbard model at half-filling, in the limit of small ratio of hopping tand on-site Coulomb repulsion U, the magnitude of standard (two-spin) Heisenberg exchange is $J \propto t^2/U$. Terms of the fourth order in t/U yield, except bilinear exchange interactions beyond the nearest neighbors, also exchange terms containing a product of four or more spin operators [216–218]. Those higher-order terms were routinely neglected up to recent times, when it was realized that they can be important for a correct description of many physical systems. Four-spin terms of the V_{LL} type can arise due to the spin-lattice interaction [219], but most naturally they appear in the so-called spin-orbital models, where orbital degeneracy is for some reason not lifted [220].

Ring Exchange

Ring exchange was introduced first to describe the magnetic properties of solid ³He [221]. Recently it was suggested that ring exchange is non-negligible in some strongly correlated electron systems like spin ladders [222, 223] and cuprates [224,225]. The analysis of the low-lying excitation spectrum of the pd-model shows that the Hamiltonian describing CuO₂ planes should contain a finite value of ring exchange [224, 225]. The search for ring exchange in cuprates was additionally motivated by inelastic neutron scattering experiments [226] and NMR experiments [227–229] on Sr₁₄Cu₂₄O₄₁ and Ca₈La₆Cu₂₄O₄₁. These materials contain spin ladders built of Cu atoms. The attempts to fit the experimental data with standard exchange terms yielded an unnaturally large ratio of $J_L/J_R \approx 2$ which is expected neither from the geometrical structure of the ladder nor from electronic structure calculations [230]. It can be shown that inclusion of other types of interactions, e.g., additional diagonal exchange, does not help to solve this discrepancy [223].

The ring exchange interaction corresponds to a special structure of the four-spin terms in (1.83), namely $V_{LL} = V_{RR} = -V_{DD} = 2J_{\text{ring}}$. Except adding the four-spin terms, ring exchange renormalizes the "bare" values of the bilinear exchange constants as well: $J_{L,L'} \rightarrow J_{L,L'} + \frac{1}{2}J_{\text{ring}}, J_{D,D'} \rightarrow J_{D,D'} + \frac{1}{2}J_{\text{ring}}, J_R \rightarrow J_R + J_{\text{ring}}$. Thus, an interesting and physically motivated special case of (1.83) is that of a regular ladder with rung exchange J_1 , leg exchange coupling J_2 , and with added ring exchange term, i.e., $J_R = J_1 + J_{\text{ring}}, J_L = J'_L = J_2 + \frac{1}{2}J_{\text{ring}}, J_D = J'_D = \frac{1}{2}J_{\text{ring}}, V_{LL} = V_{RR} = -V_{DD} = 2J_{\text{ring}}$.

It turns out that the line $J_{\rm ring} = J_2$ belongs to the general family of models (1.86) with two remarkable properties [209]: (i) on this line the product of singlets on the rungs is the ground state for $J_{\rm ring} < J_1/4$ and (ii) a propagating triplet is an exact excitation which softens at $J_{\rm ring} = J_1/4$ [222]. Thus, on this line there is an exactly known phase transition point and one knows also the exact excitation responsible for the transition. The transition at $J_{\rm ring} = J_2 = J_1/4$ is from the rung-singlet phase (dominant J_1) to the phase with a checkerboard-type long range dimer order along the ladder legs (see Fig. 1.18). In the $(J_{\rm ring}, J_1)$ plane, there is a transition boundary separating the rung singlet and dimerized phase [222, 231], and arguments based on bosonization suggest that in the limit $J_{\rm ring}, J_1 \to 0$ this boundary is a straight line $J_{\text{ring}} = \text{const} \cdot J_1$. In the vicinity of this line, even a small value of $J_{\rm ring}$ can strongly decrease the gap. For higher values of $J_{\rm ring}$, according to recent numerical studies [208, 232], additional phases appear in the phase diagram (see Fig. 1.16): one phase is characterized by the long-range scalar chiral order defined as mixed product of three spins on two neighboring ladder rungs, and another phase has dominating short-range correlations of vector chirality (1.61). Actually, under the dual transformation (1.84) staggered magnetization maps onto vector chirality, and checkerboard-type dimerizations is connected with the scalar chirality, so that the two additional phases may be viewed as duals of the Haldane and dimerized phase.



Fig. 1.16. Phase diagram of the $S = \frac{1}{2}$ ladder with equal rung and leg exchange $J_L = J_R = J$ and ring exchange $J_{\text{ring}} = K$ (from [232], LRO stands for long range order)

It is now believed [223] that inclusion of ring exchange is necessary for a consistent description of the excitation spectrum in the spin ladder material $La_6Ca_8Cu_{24}O_{41}$. This substance turns out to be close to the transition line to the dimerized phase, and therefore has an unusually small gap. Since the measured value of the energy gap sets the scale for the determination of the exchange parameters, this implies that actual values of these parameters are considerably higher compared to an analysis neglecting ring exchange. This solves the long-standing puzzle of apparently different exchange strength on the Cu-O-Cu bonds in ladders and 2D cuprates. Stimulated by infrared absorption results [233] and neutron scattering results on zone boundary magnons in pure La_2CuO_4 [234], ring exchange is now also believed to be relevant in 2D cuprates with large exchange energy. In the following we shortly discuss this related question:

In 2D magnetic materials with CuO_2 -planes the basic plaquette is the same as in the ladder material discussed above. The signature of cyclic exchange in the 2D Heisenberg model which is usually assumed for materials with CuO_2 -planes is a nonzero difference in the energies of two elementary excitations at the boundary of the Brillouin zone,

$$\Delta = \omega(q_x = \pi, q_y = 0) - \omega(q_x = \frac{\pi}{2}, q_y = \frac{\pi}{2}).$$

For the 2D Heisenberg antiferromagnet with its LRO, elementary excitations are described to lowest order in the Holstein-Primakoff (HP) spin wave approximation. In this approximation $\Delta \propto J_{\rm ring}$ results, i.e. Δ vanishes for the Heisenberg model with only bilinear exchange. Higher order corrections to the HP result as calculated in [235, 236] lead to $\Delta \approx -1.4 \cdot 10^{-2} J$. This theoretical prediction is in agreement with the experimental result in copper deuteroformate tetradeuterate (CFTD), another 2D Heisenberg magnet, but differs from the value $\Delta \approx +3 \cdot 10^{-2} J$ found from neutron scattering experiments in pure La₂CuO₄. In this latter material, diagonal, i.e. NNN interactions would have to be ferromagnetic to account for the discrepancy and can therefore be excluded, but a finite amount of ring exchange, $J_{\rm ring} \sim 0.1 J$, is in agreement with observations.

CFTD and La₂CuO₄ appear to differ in nothing but their energy scale $(J \approx 1400K \text{ for La}_2\text{CuO}_4 \text{ and } J \approx 70K \text{ for CFTD})$ and experimental results would be contradictory when bilinear and biquadratic exchange scale with the same factor. This is, however, not the case: In terms of the basic Hubbard model with hopping amplitude t and on-site Coulomb energy U one has $J \propto |t|^2/U$ and $J_{\text{ring}} \propto |t|^4/U^3$. Thus, the relative strength of the ring exchange $J_{\text{ring}}/J \propto J/U$ is material-dependent. In two materials with the same ions and therefore identical single-ion Coulomb energies, any differences result from different hopping rates. Thus in materials with high energy scale J such as La₂CuO₄ the relative importance of cyclic exchange is enhanced and it is therefore observable whereas cyclic exchange goes unnoticed in materials with low energy scale such as CFTD.

Spin-Orbital Models

Modified ladder models (1.83) arise also in one-dimensional systems with coupled spin and orbital degrees of freedom which can be described by a twoband orbitally degenerate Hubbard model at quarter filling (Fig. 17). In this case orbital degrees of freedom may be viewed as pseudospin- $\frac{1}{2}$ variables: one of the ladder legs can be interpreted as carrying the real spins $S_{1,i} \equiv S_i$ and the other one corresponds to the pseudospins $S_{2,i} \equiv \tau_i$. The corresponding effective Hamiltonian for the two-band Hubbard model was first derived by Kugel and Khomskii [220]. In addition to the spin exchange J_S and effective orbital exchange J_{τ} , its characteristic feature is the presence of strong spinorbital interaction terms of the form $(S_i \cdot S_{i+1})(\tau_i \cdot \tau_{i+1})$, which is equivalent to the four-spin interaction of the V_{LL} type in (1.83).



Fig. 1.17. Pseudospin variables au describe two degenerate orbital states of the magnetic ion

Generally, the above Hamiltonian has an SU(2) symmetry in the spin sector, but only U(1) or lower symmetry in the orbital sector. Under certain simplifying assumptions (neglecting Hund's rule coupling, nearest neighbor hopping between the same type of orbitals only, and only one Coulomb on-site repulsion constant) one obtains a Hamiltonian of the form

$$\mathcal{H} = \sum_{i} J_{S}(\boldsymbol{S}_{i} \cdot \boldsymbol{S}_{i+1}) + J_{\tau}(\boldsymbol{\tau}_{i} \cdot \boldsymbol{\tau}_{i+1}) + K(\boldsymbol{S}_{i} \cdot \boldsymbol{S}_{i+1})(\boldsymbol{\tau}_{i} \cdot \boldsymbol{\tau}_{i+1}) \quad (1.87)$$

with $J_S = J_{\tau} = J$ and $K = \frac{1}{4}J$, which possesses hidden SU(4) symmetry [212,237]. At this special point, the model is Bethe ansatz solvable [238] and gapless. This high symmetry can be broken in several ways depending on the microscopic details of the interaction, e.g., finite Hund's rule coupling and existence of more than one Coulomb repulsion constant makes the three parameters J_S , J_{τ} and K independent, reducing the symmetry to SU(2) × SU(2), and further breaking to SU(2) × U(1) is achieved through local crystal fields which can induce considerable anisotropy in the orbital sector.

The phase diagram of the model (1.87) is extensively studied analytically [239-241] as well as numerically [240, 242, 243]. The SU(4) point lies on the boundary of a critical phase which occupies a finite region of the phase diagram. Moving off the SU(4) point towards larger J_S , J_{τ} , one runs into the spontaneously dimerized phase with a finite gap and twofold degenerate ground state. The weak coupling region $J_S = J_{\tau} \gg |K|$ of the dimerized phase is a realization of the so-called non-Haldane spin liquid [219] where magnons become incoherent excitations since they are unstable against the decay into soliton-antisoliton pairs. At the special point $J_S = J_\tau = \frac{3}{4}K$ the exact ground state [244] is a checkerboard-type singlet dimer product shown in Fig. 1.18a, which provides a visual interpretation of the dimerized phase for K > 0. Solitons can be understood as domain walls connecting two degenerate ground states, see Fig. 1.18b, and magnons may be viewed as soliton-antisoliton bound states, in a close analogy to the situation at the Majumdar-Ghosh point for the frustrated spin- $\frac{1}{2}$ chain [168]. Numerical and variational studies [245] show that solitons remain the dominating low-energy excitations in the finite region around the point $J_S = J_\tau = \frac{3}{4}K$, but as one moves from it towards the SU(4) point, magnon branch separates from the soliton continuum and magnons quickly become the lowest excitations.



Fig. 1.18. Schematic representation of the spin-orbital model: (a) checkerboardtype dimerized ground state of (1.87) at $J_S = J_{\tau} = \frac{3}{4}K$; (b) a soliton connecting two equivalent dimerized states

For weak negative K one also expects a spontaneously dimerized phase [219], but now instead of a checkerboard dimer order one has spin and orbital singlets placed on the same links. A representative exactly solvable point inside this phase is $J_S = J_\tau = J = -\frac{1}{4}K$, K < 0, which turns out to be equivalent to the 16-state Potts model. At this point, the model has a

large gap of about 0.78J and its ground state can be shown to be twofold degenerate [214].

1.5.2 S = 1 Bilinear-Biquadratic Chain

The isotropic Heisenberg spin-1 AF chain is a generic example of a system in the Haldane phase. However, the most general isotropic exchange interaction for spin S = 1 includes biquadratic terms as well, which naturally leads to the model described by the following Hamiltonian:

$$\mathcal{H} = \sum_{n} \cos \theta \left(\boldsymbol{S}_{n} \cdot \boldsymbol{S}_{n+1} \right) + \sin \theta \left(\boldsymbol{S}_{n} \cdot \boldsymbol{S}_{n+1} \right)^{2}.$$
(1.88)

The AKLT model considered in Sect. 1.3 is a particular case of the above Hamiltonian with $\tan \theta = \frac{1}{3}$. There are indications [246] that strong biquadratic exchange is present in the quasi-one-dimensional compound LiVGe₂O₆. The points $\theta = \pi$ and $\theta = 0$ correspond to the Heisenberg ferro- and antiferromagnet, respectively. The bilinear-biquadratic chain (1.88) has been studied rather extensively, and a number of analytical and numerical results for several particular cases are available (Fig. 19). It is firmly established that the Haldane phase with a finite spectral gap occupies the interval $-\pi/4 < \theta < \pi/4$, and the ferromagnetic state is stable for $\pi/2 < \theta < 5\pi/4$, while $\theta = 5\pi/4$ is an SU(3) symmetric point with highly degenerate ground state [247].



Fig. 1.19. Phase diagram of the S = 1 bilinear-biquadratic chain (1.88)

An exact solution is available [238] for the Uimin-Lai-Sutherland (ULS) point $\theta = \pi/4$ which has SU(3) symmetry. The ULS point was shown [248] to mark the Berezinskii-Kosterlitz-Thouless (BKT) transition from the massive Haldane phase into a massless phase occupying the interval $\pi/4 < \theta < \pi/2$ between the Haldane and ferromagnetic phase; this is supported by numerical studies [249].

The properties of the remaining region between the Haldane and ferromagnetic phase are more controversial. The other Haldane phase boundary $\theta = -\pi/4$ corresponds to the exactly solvable Takhtajan-Babujian model [250]; the transition at $\theta = -\pi/4$ is of the Ising type and the ground state at $\theta < -\pi/4$ is spontaneously dimerized with a finite gap to the lowest excitations [249, 251–256]. The dimerized phase extends at least up to and over the point $\theta = -\pi/2$ which has a twofold degenerate ground state and finite gap [257–259].

Chubukov [260] used the Holstein-Primakoff-type bosonic representation of spin-1 operators [261] based on the quadrupolar ordered "spin nematic" reference state with $\langle \mathbf{S} \rangle = 0$, $\langle S_{x,y}^2 \rangle = 1$, $\langle S_z^2 = 0 \rangle$, and suggested, on the basis of the renormalization group arguments, that the region with $\theta \in [5\pi/4, \theta_c]$, where $\frac{5\pi}{4}\theta_c < \frac{3\pi}{2}$, is a disordered nematic phase. Early numerical studies [262] have apparently ruled out this possibility, forming a common belief [263,264] that the dimerized phase extends all the way up to the ferromagnetic phase, i.e., that it exists in the entire interval $5\pi/4 < \theta < 7\pi/4$. However, recent numerical results [265,266] indicate that the dimerized phase ends at certain $\theta_c > 5\pi/4$, casting doubt on the conclusions reached nearly a decade ago.

Using special coherent states for S = 1,

$$|\boldsymbol{u},\boldsymbol{v}\rangle = \sum_{j} (u_j + iv_j)|t_j\rangle, \qquad |\pm\rangle = \mp \frac{1}{\sqrt{2}} (|t_x\rangle \pm i|t_y\rangle), \quad |0\rangle = |t_z\rangle, \quad (1.89)$$

subject to the normalization condition $u^2 + v^2 = 1$ and gauge-fixing constraint $u \cdot v = 0$, one can show [267] that for θ slightly above $\frac{5\pi}{4}$ the effective lowenergy physics of the problem can be described by the nonlinear sigma model of the form (1.47). The topological term is absent and the coupling constant is given by

$$g = (1 - \operatorname{ctg} \theta)^{1/2} \ll 1 \tag{1.90}$$

(note that in this case smallness of g is not connected to the large-S approximation). By the analogy with the Haldane phase, this mapping suggests that for $\theta > 5\pi/4$ the system is in a disordered state with a short-range nematic order and exponentially small gap $\Delta \propto e^{-\pi/g}$. The antiferromagnetism unit vector \boldsymbol{l} gets replaced by the unit director \boldsymbol{u} and the opposite vectors \boldsymbol{u} and $-\boldsymbol{u}$ correspond to the same physical state, which makes the model live in the RP^2 space instead of O(3). The main difference from the usual O(3) NLSM is that the RP^2 space is doubly connected, which supports the existence of disclinations – excitations with a nontrivial π_1 topological charge. However, the characteristic action of a disclination is of the order of $\sin \theta$ and thus the low-energy physics on the characteristic scale of Δ should not be affected by the disclinations.

1.5.3 Mixed Spin Chains: Ferrimagnet

In the last decade there has been much interest in 'mixed' 1d models involving spins of different magnitude S. The simplest system of this type is actually of a fundamental importance since it represents the generic model of a quantum ferrimagnet described by the Hamiltonian

$$\mathcal{H} = \sum_{n} (\boldsymbol{S}_{n} \boldsymbol{\tau}_{n} + \boldsymbol{\tau}_{n} \boldsymbol{S}_{n+1})$$
(1.91)

where S_n and τ_n are respectively spin-1 and spin- $\frac{1}{2}$ operators at the *n*-th elementary magnetic cell (with S^z eigenstates denoted in the following as (+, 0, -) and (\uparrow, \downarrow) , respectively). An experimental realization of such a system is the molecular magnet NiCu(pba)(D₂O)₃ · D₂O [268].

According to the Lieb-Mattis theorem [269], the ground state of the system has the total spin $S_{\text{tot}} = L/2$, where L is the number of unit cells. There are two types of magnons [270, 271]: a gapless "acoustical" branch with $S^z = L/2 - 1$, and a gapped "optical" branch with $S^z = L/2 + 1$. The energy of the "acoustical" branch rises with field, and in strong fields those excitations can be neglected, while the "optical" magnon gap closes at the critical field.

A good quantitative description of the ferrimagnetic chain can be achieved with the help of the variational matrix product states (MPS) approach [34, 181]. The MP approach is especially well suited to this problem since the fluctuations are extremely short-ranged, with the correlation radius smaller than one unit cell length [181,270,271]. The ground state properties, including correlation functions, are within a few percent accuracy described by the MPS $|\Psi_0\rangle = \text{Tr}(g_1g_2\cdots g_L)$, where the elementary matrix has the form (1.70) and the variational parameters u, v are determined from the energy minimization. The variational energy per unit cell is $E_{\text{var}} = -1.449$, to be compared with the numerical value $E_{\text{g.s.}} \simeq 1.454$ [139,181]. According to (1.67), the above matrix has the "hyperspin" quantum numbers $(\frac{1}{2}, \frac{1}{2})$, which in turn ensures that the variational state $|\Psi_0\rangle$ has correct $S_{\text{tot}} = S_{\text{tot}}^z = L/2$.

The MPS approach works also very well for the excited states [34]. The dispersion of optical magnons can be reproduced within a few percent by using the MPS ansatz $|n\rangle = \text{Tr}(g_1g_2\cdots g_{n-1}\tilde{g}_ng_{n+1}\cdots g_L)$ with one of the ground state matrices g_n replaced by the matrix

$$\widetilde{g}_n = \frac{f-1}{\sqrt{2}} g_n \,\sigma^{+1} - \frac{f+1}{\sqrt{2}} \,\sigma^{+1} \,g_n + \widetilde{w} \,\sigma^{+1} \,\psi_{\frac{1}{2},\frac{1}{2}} \,, \tag{1.92}$$

which carries the "hyperspin" $(\frac{3}{2}, \frac{3}{2})$ and contains two free parameters f, w. Generally the states $|n\rangle$ are orthogonal to Ψ_0 , but are not orthogonal to each other. Since the states with a certain momentum $|k\rangle = \sum_n e^{ikn} |n\rangle$ obviously depend only on \tilde{w} , one parameter in (1.92) is redundant and can be fixed by requiring that one-magnon states $\{|n\rangle\}$ become mutually orthogonal [34]. The resulting variational dispersion for the optical magnon is in excellent agreement with the exact diagonalization data [34]; the variational value for the optical magnon gap is $\Delta_{\text{var}} \simeq 1.754 J$, to be compared with the numerically exact value $\Delta_{\text{opt}} = 1.759 J$ [139,270].

Several other mixed-spin systems were studied, particularly mixed-spin ladders which may exhibit either ferrimagnetic or singlet ground states depending on the ladder type [272, 273].

1.6 Gapped 1D Systems in High Magnetic Field

The presence of an external magnetic field brings in a number of new features. In gapped low-dimensional spin systems, the gap will be closed by a sufficiently strong external magnetic field $H = H_c$, and a finite magnetization will appear above H_c [274]. For a system with high (at least axial) symmetry the high-field phase at $H > H_c$ is critical [275–277] and the low-energy response is dominated by a two-particle continuum [278–280]. When the field is further increased, the system may stay in this critical phase up to the saturation field H_s , above which the system is in a saturated ferromagnetic state. Under certain conditions, however, the excitations in this high-field phase may again acquire a gap, making the magnetization per spin m "locked" in some field range; this phenomenon is known as a *magnetization plateau* and has been receiving much attention from both theoretical and experimental side [122, 203, 281-291]. Other singularities of the m(H) dependence, the so-called magnetization cusps [292, 293], may arise in frustrated systems. In anisotropic systems with no axial symmetry the high-field phase has longrange order and the response is of the quasi-particle type [275, 276].

1.6.1 The Critical Phase and Gapped (Plateau) Phase

In a one-dimensional spin chain with the spin S, a necessary condition for the existence of a plateau is given by the generalized Lieb-Schulz-Mattis theorem [122] discussed in Sect. 1.3.2 as the requirement that lS(1-M) is an integer number, where l is the number of spins in the magnetic unit cell, and M = m/S is the magnetization per spin in units of saturation. This condition ensures that the system is allowed to have a spectral gap at finite magnetization, so that one needs to increase the magnetic field by a finite value to overcome the gap and make the magnetization grow. It yields the allowed values of M at which plateaux may exist, but it does not guarantee their existence. For a mixed spin system with ions having different spins S_i the quantity lS in the above condition would be replaced by the sum of spin values over the unit cell $\sum_{i} S_{i}$. The number l may differ from that dictated by the Hamiltonian in case of a spontaneous translational symmetry breaking. A trivial plateau at M = 0 is obviously possible for any integer-S spin chain, which is just another way to say that the ground state has a finite gap to magnetic excitations.

As an intuitively clear example of a magnetization plateau one can consider the $S = \frac{3}{2}$ chain with large easy-plane single-ion anisotropy described by the Hamiltonian

$$\mathcal{H} = \sum_{l} J \boldsymbol{S}_{l} \cdot \boldsymbol{S}_{l+1} + D(S_{l}^{z})^{2} - H S_{l}^{z}.$$
(1.93)

If $D \gg J$, the spins are effectively suppressed to have $S^z = \pm 1/2$, and with increasing field to $H \sim J$ one gets first to the polarized m = 1/2 (M = 1/3)



Fig. 1.20. VBS states visualizing (a) M = 1/3 plateau in the large- $D S = \frac{3}{2}$ chain (1.93); (b) M = 1/2 plateau in the bond-alternated S = 1 chain

state (see Fig. 1.20a), and the magnetization remains locked at m = 1/2 up to a much larger field $H \sim D$, where it gets finally switched to m = 3/2 [122].

An experimentally more relevant example is a S = 1 chain with alternating bond strength, where l = 2 and a nontrivial plateau at $M = \frac{1}{2}$ is allowed. In the strong alternation regime (weakly coupled S = 1 dimers) this plateau can be easily visualized as the state with all dimers excited to S = 1, $S^z = +1$ (see Fig. 1.20b). The M = 1/2 plateau was experimentally observed in magnetization measurements up to 70 T in NTENP [294].

Very distinct magnetization plateaux at $M = \frac{1}{4}$ and $M = \frac{3}{4}$ were observed in NH₄CuCl₃ [295], a material which contains weakly coupled $S = \frac{1}{2}$ dimers. The nature of those plateaux is, however, most probably connected to three-dimensional interactions in combination with an additional structural transition which produces three different dimer types [296].

Plateaux and Critical Phase in an Alternated $S = \frac{1}{2}$ Zigzag Chain

Another simple example illustrating the occurrence of a plateau and the physics of a high-field critical phase is a strongly alternating $S = \frac{1}{2}$ zigzag chain, which can be also viewed as a ladder in the regime of weakly coupled dimers, as shown in Fig. 1.21. For a single dimer in the field, the energy of the $S^z = +1$ triplet state $|t_+\rangle$ becomes lower than that of the singlet $|s\rangle$ at H = J. If the dimers were completely decoupled, then there would be just one critical field $H_c^{(0)} = J$ and the magnetization M would jump from zero to one at $H = H_c^{(0)}$. A finite weak interdimer coupling will split the point $H = H_c$ into a small but finite field region $[H_c, H_s]$. Assuming that the coupling is small and thus H_c and H_s are close to J, one can neglect for each dimer all states except the two lowest ones, $|s\rangle$ and $|t_+\rangle$ [289,290]. The Hilbert space is reduced to two states per dimer, and one may introduce pseudospin- $\frac{1}{2}$ variables, identifying $|s\rangle$ with $|\widetilde{\downarrow}\rangle$ and $|t_+\rangle$ with $|\widetilde{\uparrow}\rangle$. The effective spin- $\frac{1}{2}$ Hamiltonian in the reduced Hilbert space takes the form

$$\mathcal{H} = \sum_{n} \widetilde{J}_{xy} (\widetilde{S}_{n}^{x} \widetilde{S}_{n+1}^{x} + \widetilde{S}_{n}^{y} \widetilde{S}_{n+1}^{y}) + \widetilde{J}_{z} \widetilde{S}_{n}^{z} \widetilde{S}_{n+1}^{z} - \widetilde{h} \widetilde{S}_{n}^{z}, \qquad (1.94)$$

where the effective coupling constants are given by

$$\widetilde{J}_{xy} = \alpha - \beta/2, \quad \widetilde{J}_z = \alpha/2 + \beta/4, \quad \widetilde{h} = H - J - \alpha/2 - \beta/4.$$
 (1.95)
At $\tilde{h} = 0$, depending on the value of the parameter $\varepsilon = \tilde{J}_z/|\tilde{J}_{xy}|$, the effective spin- $\frac{1}{2}$ chain can be in three different phases: the Néel ordered, gapped phase for $\varepsilon > 1$, gapless XY phase for $-1 < \varepsilon < 1$, and ferromagnetic phase for $\varepsilon < -1$. Boundaries between the phases are lines $\beta = 6\alpha$ and $\beta = 2\alpha/3$, as shown in Fig. 1.21.



Fig. 1.21. (a) alternating zigzag chain in the strong coupling limit $\alpha, \beta \ll J$; (b) its phase diagram in the high-field regime $\tilde{h} \simeq 0$ (see (1.95)); (c) the magnetization behavior in the XY and Néel phases

It is easy to understand what the magnetization curve looks like in different phases. In the XY phase the magnetization per spin of the effective chain $\tilde{m}(\tilde{h})$ reaches its saturation value $\frac{1}{2}$ at $\tilde{h} = \pm h_c$, where $h_c = |\tilde{J}_{xy}| + \tilde{J}_z$. Point $\tilde{h} = -h_c$ can be identified with the first critical field $H = H_c$, and $\tilde{h} = +h_c$ corresponds to the saturation field H_s . The symmetry $\tilde{h} \mapsto -\tilde{h}$ corresponds to the symmetry against the middle point $H = (H_c + H_s)/2$. This symmetry is only valid in the first order in the couplings α, β and is a consequence of our reduction of the Hilbert space. The magnetization $M = \tilde{m} + \frac{1}{2}$ of the original chain has only trivial plateaux at M = 0 and M = 1, as shown in Fig. 1.21c.

Near the first critical field H_c the magnetization behaves as $(H - H_c)^{1/2}$. This behavior is easy to understand for the purely XY point $\tilde{J}_z = 0$. At this point the model can be mapped to free fermions with the dispersion $E(k) = \tilde{J}_{xy} \cos k - \tilde{h}$ which is quadratic at its bottom. The magnetization Mis connected to the Fermi momentum k_F via $M = 1 - k_F/\pi$, which yields the square root behavior. Further, if the fermions are interacting, this interaction can be neglected in the immediate vicinity of H_c where the particle density is low, so that the square root behavior is universal in one dimension (it can be violated only at special points where the fermion dispersion is not quadratic, or in presence of anisotropy which breaks the axial symmetry).

In the Néel phase there is a finite gap Δ , and \tilde{m} stays zero up to $\tilde{h} = \Delta$, so that in the language of the original chain there is a nontrivial plateau at $M = \frac{1}{2}$ whose width is 2Δ (Fig. 1.21c).

A Few Other Examples

A similar mapping to an effective $S = \frac{1}{2}$ chain can be sometimes achieved for systems with no obvious small parameter. An instructive example is the AKLT chain (1.51) in strong magnetic field H [297,298]. The zero-field gap of the AKLT model is known to be $\Delta \simeq 0.70$ [111], and we are interested in the high-field regime $H > H_c \equiv \Delta$ where the gap closes. One may use the matrix product soliton ansatz (1.56), (1.57) to describe the triplet excitation with $\mu = +1$. States $|\mu, n\rangle$ with different n can be orthogonalized by putting in (1.57) a/b = 3 [131]. Further, one may introduce effective spin- $\frac{1}{2}$ states $|\alpha_n\rangle = |\uparrow\rangle, |\downarrow\rangle$ at each site, making the identification

$$|\alpha_1 \alpha_2 \cdots \alpha_L\rangle = \operatorname{Tr}(g_1 g_2 \cdots g_L), \qquad (1.96)$$

where the matrix g_n is either the ground state matrix (1.55) if $|\alpha_n\rangle = |\uparrow\rangle$, or the matrix (1.57) corresponding to the lowest $S^z = +1$ triplet if $|\alpha_n\rangle = |\downarrow\rangle$, respectively. Then the desired mapping is achieved by restricting the Hilbert space to the states of the above form (1.96). The resulting effective $S = \frac{1}{2}$ chain is described by the Hamiltonian

$$\mathcal{H}_{S=1/2} = \sum_{n} \widetilde{J}_{xy} \left(\widetilde{S}_{n}^{x} \widetilde{S}_{n+1}^{x} + \widetilde{S}_{n}^{y} \widetilde{S}_{n+1}^{y} \right) - \widetilde{h} \widetilde{S}_{n}^{z} + \sum_{n,m} V_{m} \widetilde{S}_{n}^{z} \widetilde{S}_{n+m}^{z}, \quad (1.97)$$

where $\tilde{J}_{xy} = \frac{10}{9}$, $\tilde{h} \simeq (H - 1.796)$, and the interaction constants V_m are exponentially decaying with m and always very small, $V_1 = -0.017$, $V_2 = -0.047$, $V_3 = 0.013$, $V_4 = -0.0046$, etc. [297, 298] Thus, if one neglects the small interaction V_m , then in the vicinity of H_c the AKLT chain is effectively described by the XY model, i.e. by noninteracting hardcore bosons.

The critical phase appears also in a ferrimagnet (1.91): in an applied field the ferromagnetic magnon branch acquires a gap which increases with the field, while the optical branch goes down and its gap closes at $H = \Delta_{\text{opt}} \simeq$ 1.76 J. A mapping to a $S = \frac{1}{2}$ chain can be performed can be performed [34] in a way very similar to the one described above for the AKLT model, using the MP ansatz with the elementary matrices (1.70) and (1.92). Restricting all effective interactions to nearest neighbors only, one obtains the effective Hamiltonian of the form (1.94), where $\tilde{J}_{xy} \simeq 0.52$, $\tilde{J}_z \simeq 0.12$, $h_e \simeq (H-2.44)$ are determined by the numerical values of the optimal variational parameters in the matrices (1.70) and (1.92) [34]. Similarly to (1.97), the complete effective Hamiltonian contains exchange interactions exponentially decaying with distance, but this decay is very rapid, e.g., the next-nearest neigbor exchange constants $\tilde{J}_{xy}^{(2)} \simeq 0.04$, $\tilde{J}_{z}^{(2)} \simeq 0.02$, so that one may safely use the reduced nearest-neighbor Hamiltonian.

For both the ladder and the ferrimagnet, in the critical phase the temperature dependence of the low-temperature part of the specific heat C exhibits a rather peculiar behavior [34, 299, 300]. With the increase of the field H, a single well-pronounced low-T peak pops up when H is in the middle between H_c and H_s . When H is shifted towards H_c or H_s , the peak becomes flat and develops a shoulder with another weakly pronounced peak at very low temperature. This phenomenon can be fully explained within the effective $S = \frac{1}{2}$ chain model [34] and results from unequal bandwidth of particle-type and hole-type excitations in the effective spin- $\frac{1}{2}$ chain [301]: In zero field the contributions into the specific heat from particles and holes are equal; with increasing field, the hole bandwidth grows up, while the particle bandwidth decreases, and the average band energies do not coincide. This leads to the presence of two peaks in C(T): holes yield a strong, round peak moving towards higher temperatures with increasing the field, and the other peak (due to the particles) is weak, sharp, and moves to zero when \tilde{h} tends to $\pm h_c$.

1.6.2 Magnetization Cusp Singularities

Cusp singularities were first discovered in integrable models of spin chains [302], but later were found to be a generic feature of frustrated spin systems where the dispersion of elementary excitations has a minimum at an incommensurate value of the wave vector [292,293]. The physics of this phenomenon can be most easily understood on the example of a frustrated $S = \frac{1}{2}$ chain described by the isotropic version of (1.60) with $\Delta = 1$ and $j > \frac{1}{4}$. Assume we are above the saturation field, so that the ground state is fully polarized. The magnon dispersion

$$\varepsilon(k) = H - 1 - j + \cos k + j \cos(2k)$$

has a minimum at $k = k_0 = \pi \pm \arccos(1/4j)$. The gap at $k = k_0$ closes if the field H is reduced below the saturation value $H_s = 1 + 2j + 1/(8j)$. If one treats magnons as hardcore bosons, they are in one dimension equivalent to fermions, and in the vicinity of H_s , when the density of those fermions is low, they can be treated as free particles. If $H_{\text{cusp}} < H < H_s$, where $H_{\text{cusp}} = 2$ corresponds to the point where $\varepsilon(k = \pi) = 0$, there are *two* Fermi seas (four Fermi points), and if H is reduced below H_{cusp} they join into a single Fermi sea. It is easy to show that the magnetization m behaves as

$$m(H) - m(H_{\rm cusp}) \propto \begin{cases} (H - H_{\rm cusp})^{1/2} &, & H > H_{\rm cusp} \\ H - H_{\rm cusp} &, & H < H_{\rm cusp} \end{cases},$$

so that there is indeed a cusp at $H = H_{\text{cusp}}$, see Fig. 1.22.



Fig. 1.22. Schematic explanation of cusp singularities: two Fermi seas join at $H = H_{\text{cusp}}$ (left) leading to a cusp in the magnetization curve (*right*)

1.6.3 Response Functions in the High-Field Phase

The description of the critical phase in terms of an effective $S = \frac{1}{2}$ chain is equivalent to neglecting certain high-energy degrees of freedom, e.g., two of the three rung triplet states in case of the strongly coupled spin ladder. Those neglected states, however, form excitation branches which contribute to the response functions at higher energies, and this contribution is generally easier to see experimentally than the highly dispersed low-energy continuum of the particle-hole ("spinon") excitations coming from the effective $S = \frac{1}{2}$ chain. In case of an axially anisotropic system, the continuum will collapse into a delta-function, and weights of low- and high-energy branches will be approximately equal. Those high-energy branches were found to exhibit interesting behavior in electron spin resonance (ESR) and inelastic neutron scattering (INS) experiments in two quasi-one-dimensional materials, Ni(C₂H₈N₂)₂Ni(CN)₄ (known as NENC) [303] and Ni(C₅H₁₄N₂)₂N₃(PF₆) (abbreviated NDMAP) [304].

As mentioned before, the physics of the high-field phase depends strongly on whether the field is applied along a symmetry axis or not.

Response in an Axially Symmetric Model

Let us consider the main features of the response in the critical phase of the axially symmetric system using the example of the strongly coupled ladder addressed in the previous subsection. In order to include the neglected $|t_-\rangle$ and $|t_0\rangle$ states, it is convenient to use the hardcore boson language. One may argue [298,305] that the most important part of interaction between the bosons is incorporated in the hardcore constraint. Neglecting all interactions except the constraint, one arrives at the simplified effective model of the type

$$\mathcal{H}_{\text{eff}} = \sum_{n\mu} \varepsilon_{\mu} b_{n,\mu}^{\dagger} b_{n,\mu} + t(b_{n,\mu}^{\dagger} b_{n+1,\mu} + \text{h.c.}), \qquad (1.98)$$

where $\mu = 0, \pm 1$ numbers three boson species (triplet components with $S^z = \mu$), $t = \alpha - \beta/2$ is the hopping amplitude which is equal for all species, and $\varepsilon_{\mu} = J - \mu H$.

The ground state at $H > H_c$ contains a "condensate" (Fermi sea) of b_{+1} bosons. Thus, at low temperatures for calculating the response it suffices to take into account only processes involving states with at most one b_0 or b_{-1} particle: (A) creation/annihilation of a low-energy b_{+1} boson; (B) creation/annihilation of one high-energy $(b_{-1} \text{ or } b_0)$ particle, and (C) transformation of a b_{+1} particle into b_0 one.

The processes of the type (A) can be considered completely within the model of an effective $S = \frac{1}{2}$ chain, for which analytical results are available [306–308]. For example, the transversal dynamical susceptibility $\chi^{xx}(q,\omega) = \chi^{yy}(q,\omega)$ for q close to the antiferromagnetic wave number π is given by the expression

$$\chi^{xx}(\pi + k, \omega) = A_x(H) \frac{\sin(\frac{\pi\eta}{2})\Gamma^2(1 - \frac{\eta}{2})u^{1-\eta}}{(2\pi T)^{2-\eta}} \\ \times \frac{\Gamma\left(\frac{\eta}{4} - i\frac{\omega - vk}{4\pi T}\right)\Gamma\left(\frac{\eta}{4} - i\frac{\omega + vk}{4\pi T}\right)}{\Gamma\left(1 - \frac{\eta}{4} - i\frac{\omega - vk}{4\pi T}\right)\Gamma\left(1 - \frac{\eta}{4} - i\frac{\omega + vk}{4\pi T}\right)} .$$
(1.99)

Here $A_x(H)$ is the non-universal amplitude which is known numerically [309], v is the Fermi velocity, and $\eta = 1 - \frac{1}{\pi} \arccos(\tilde{J}_z/\tilde{J}_{xy})$ (neglecting interaction between $b_{\pm 1}$ bosons corresponds to $\tilde{J}_z = 0$). This contribution describes a lowenergy "spinon" continuum, and the response function has an edge singularity at its lower boundary. A similar expression is available for the longitudinal susceptibility [306]; for the longitudinal DSF of the XY chain in case of zero temperature a closed exact expression is available as well [49], and for $T \neq 0$ the exact longitudinal DSF can be calculated numerically [56]. Applying the well-known relation $S^{\alpha\alpha}(q,\omega) = \frac{1}{\pi} \frac{1}{1-e^{-\omega/T}} \operatorname{Im}\chi^{\alpha\alpha}(q,\omega)$, one obtains in this way the contribution $I^A(q,\omega)$ of the (A) processes to the dynamic structure factor. The processes of (B) and (C) types, which correspond to excitations with higher energies, cannot be analyzed in the language of the $S = \frac{1}{2}$ chain.

Consider first the zero temperature case for (B)-type processes. The model (1.98) with just one high-energy particle present is equivalent to the problem of a single mobile impurity in the hardcore boson system. The hopping amplitudes for the impurity and for particles are equal, and in this case the model can be solved exactly [310]. Creation of the impurity leads to the orthogonality catastrophe [311] and to the corresponding edge-type singularity in the response.

In absence of the impurity, the eigenstates of the hardcore boson Hamiltonian (1.98) can be represented in the form of a Slater determinant constructed of the free plane waves $\psi_i(x) = \frac{1}{\sqrt{L}}e^{ik_ix}$ (*L* is the system length), with an additional antisymmetric sign factor attached to the determinant, which ensures symmetry of the wave function under permutations of k_i (this construction points to the equivalence between fermions and hardcore bosons which is a peculiarity of dimension one).

Let us assume for definiteness that the total number of b_{+1} particles in the ground state N is even. The allowed values of momenta k_i are then given by

$$k_i = \pi + (2\pi/L)I_i, \quad i = 1, \dots, N$$
 (1.100)

where the numbers I_i should be all different and half-integer. The ground state $|g.s.\rangle$ is given by the Fermi sea configuration with the momenta filling the $[k_F, 2\pi - k_F]$ interval, the Fermi momentum being defined as

$$k_F = \pi (1 - N/L). \tag{1.101}$$

The energy of is $E = \sum_{i=1}^{N} (\varepsilon_{i+1} + 2t \cos k_i)$, and the total momentum $P = \sum_i k_i$ of the ground state is zero (mod 2π).

Since the hopping amplitudes for "particles" and "impurities" are equal, it is easy to realize that the above picture of the distribution of wave vectors remains true when some of the particles are replaced by the impurities: they form a single "large" Fermi sea.

The excited configuration $|(\mu, \lambda)_{k'_1...k'_N}\rangle$ with a single impurity boson b_{μ} having the momentum λ can be also exactly represented in the determinantal form [310] with determinants containing wave functions $\varphi_i(x)$ which become asymptotically equivalent to the free scattering states $\frac{1}{\sqrt{L}}e^{i(k'_ix+\delta_i)}$ in the thermodynamic limit; for noninteracting hardcore particles the phase shifts $\delta_i = -\pi/2$. The total momentum of the excited state is $P' = \sum_{i=1}^N k'_i + \lambda$, and its energy is given by $E' = \sum_{i=1}^N (\varepsilon_{+1} + 2t \cos k'_i) + \varepsilon_{\mu} + 2t \cos \lambda$. Here the allowed wave vectors k'_i and λ are determined by the same formula (1.100), but since the total number of particles has changed by one, the numbers I_i are now integer.

The matrix element $\langle (\mu, \lambda)_{k'_1...k'_N} | b^{\dagger}_{\mu}(q) | \text{g.s.} \rangle$, which determines the contribution to the response from the (B)-type processes, is nonzero only if the selection rules $\lambda = q$, P' = P + q are satisfied [298], and is proportional to the determinant $M_{fi} = \text{det}\{\langle \varphi_i | \psi_j \rangle\}$ of the overlap matrix. Due to the orthogonality catastrophe (OC), the overlap determinant is generally algebraically vanishing in the thermodynamic limit, $|M_{fi}|^2 \propto L^{-\beta}$. The response is, however, nonzero and even singular because there is a macroscopic number of "shake-up" configurations with nearly the same energy.

The OC exponent β can be calculated using the results of boundary conformal field theory (BCFT) [312]. For this purpose it is necessary to calculate the energy difference ΔE_f between the ground state and the excited state $|f\rangle$, including the 1/L corrections. Then in case of *open boundary conditions* the OC exponent β , according to BCFT, can be obtained as

$$\beta = \frac{2L\widetilde{\Delta E_f}}{\pi v_F} \equiv \frac{2\widetilde{\Delta E_f}}{\Delta E_{\min}}.$$
(1.102)

Here $v_F = 2t \sin k_F$ is the Fermi velocity, so that $\Delta E_{\min} = \pi v_F/L$ is the lowest possible excitation energy, and ΔE_f is the O(1/L) part of ΔE_f (i.e., with the bulk contribution subtracted). In this last form this formula should be also valid for the *periodic boundary conditions*, then ΔE_{\min} should be replaced by $2\pi v_F/L$. For noninteracting hardcore bosons one obtains $\beta = \frac{1}{2}$. It is worthwhile to note that this value for the OC exponent coincides with the one obtained earlier for the regime of weak coupling [313] by means of the bosonization technique.

The value of the OC exponent is connected to another exponent $\alpha = 1 - \beta$ which determines the character of the singularity in the response,

$$S^B(q,\omega) \propto \frac{1}{(\omega - \omega_\mu(q))^{\alpha}},$$
 (1.103)

where $\omega_{\mu}(q)$ is the minimum energy difference between the ground state and the excited configuration. For example, at $q = \pi$, where the strongest response is expected, the lowest energy excited configuration is symmetric about $k = \pi$ and is given by $\lambda = \pi$, $k'_j = \pi \pm \frac{2\pi}{L}j$, $j = 1, \ldots, N/2$, so that

$$\omega_{\mu}(q=\pi) = \varepsilon_{\mu} + 2t \cos k_F = (1-\mu)H.$$
(1.104)

Note that the quantity $\omega_{\mu}(\pi)$, which determines the position of the peak in the response, and in an inelastic neutron scattering experiment would be interpreted as the energy of the corresponding mode with $S^z = \mu$, has a counter-intuitive dependence on the magnetic field: one would rather expect that it behaves as $-\mu H$. The resulting picture of modes which should be seen e.g. in the INS experiment is schematically shown in Fig. 1.23.



Fig. 1.23. The schematic dependence of "resonance" lines (peaks in the dynamic structure factor at $q = \pi$, shown as solid lines) on the magnetic field in an axially symmetric system. The dashed areas represent continua. The processes responsible for the transitions are indicated near the corresponding lines, e.g. $v \to -1$ denotes the (B)-type process of creating one boson with $S^z = -1$ from the vacuum, etc.

As q moves further from π , λ must follow q, and in order to satisfy the selection rules one has to create an additional particle-hole pair to compensate the unwanted change of momentum. Away from $q = \pi$ this configuration does not necessarily have the lowest energy, and there are other configurations with generally large number of umklapp-type of particle-hole pairs, whose energy may be lower, but, as discussed in [305], their contribution to the response can be neglected because the corresponding OC exponent is larger than 1 for this type of configurations.

At finite temperature $T \neq 0$ the singularity gets damped. The contribution of *B*-type processes to the dynamical susceptibility $\chi(q, \omega)$ is proportional to the following integral:

$$\chi(q,\omega) \propto \int_0^\infty dt e^{i \Omega t} \Bigl(\frac{\pi T}{\sinh \pi T t}\Bigr)^\beta \,,$$

where $\Omega \equiv \omega - \omega_{\mu}(q)$ is the deviation from the edge. Then for the dynamical structure factor $S(q, \omega)$ one obtains

$$S^{B}(q,\omega) \propto \frac{\cos(\pi\beta/2)}{1 - e^{-\omega/T}} \sinh\left(\frac{\Omega}{2T}\right) T^{\beta-1} \left| \Gamma\left(\frac{\beta}{2} + i\frac{\Omega}{2\pi T}\right) \right|^{2}.$$
 (1.105)

From (1.105) one recovers the edge singularity behavior (1.103) at T = 0.

For $H > H_c$ there will be also a contribution from C-type transitions corresponding to the transformation of b_{+1} bosons into b_0 ones. Those processes do not change the total number of particles and thus do not disturb the allowed values of the wave vector, so that there is no OC in this case. The problem of calculating the response is equivalent to that for the 1D Fermi gas, with the only difference that we have to take into account the additional change in energy $\varepsilon_0 - \varepsilon_{+1}$ which takes place in the transition. The well-known formula for the susceptibility of a Fermi gas yields the contribution of C-type processes into the response:

$$S^{C}(q,\omega) = \frac{1}{1 - e^{-\omega/T}} \frac{\pi - k_F}{2\pi^2}$$

$$\times \int dk \left[n_{+1}(k) - n_0(k+q) \right] \delta \left(\omega - \varepsilon_0(k+q) + \varepsilon_{+1}(k) \right),$$
(1.106)

where $\varepsilon_{\mu}(k) = \varepsilon_{\mu} + 2t \cos k$, and $n_{\mu} = (e^{\varepsilon_{\mu}/T} + 1)^{-1}$ is the Fermi distribution function. This contribution contains a square-root singularity, whose edge is located at

$$\omega = \varepsilon_0 - \varepsilon_{+1} + 2t\sqrt{2(1 - \cos q)} \tag{1.107}$$

and which survives even for a finite temperature.

Role of Weak 3D Coupling in the Axially Symmetric Case: Bose-Einstein Condensation of Magnons

In the axially symmetric case, the high-field phase is gapless and thus is extremely sensitive to even a small 3D interaction. If one views the process of formation of the high-field phase as an accumulation of hardcore bosonic particles (magnons) in the ground state, then the most important effect is that in a 3D system those bosons can undergo the Bose-Einstein Condensation (BEC) transition. In one dimension there is no difference between hardcore bosons and fermions, and instead of BEC one obtains, as we have seen, a Fermi sea.

In 3D coupled system, increasing the field beyond H_c leads to the formation of the Bose-Einstein condensate of magnons. The U(1) symmetry gets spontaneously broken, and the condensate wave function picks a certain phase which is physically equivalent to the transverse (with respect to the field) staggered magnetization.

The idea of field-induced BEC was discussed theoretically several times [275, 278, 280], but only recently such a transition was observed [314] in TlCuCl₃, which can be viewed as a system of weakly coupled $S = \frac{1}{2}$ dimers. The observed behavior of magnon density (longitudinal magnetization) n as a function of temperature T was in a qualitative agreement with the predictions of the BEC theory: with increasing T from zero to the critical temperature T_c the magnetization decreases, and then starts to increase, so that the minimum of n occurs at $T = T_c$. There was, however, some discrepancy between the predicted and observed field dependence of the critical temperature: according to the BEC theory, $T_c \propto (H - H_c)^{\phi}$ with $\phi = 2/3$, while the experiment yields rather $\phi \approx 1/2$ [314, 315]. The reason for this discrepancy seems to be clarified in the recent work [316]: since in TlCuCl₃ experiments the critical temperature T_c becomes comparable with the magnon gap Δ , one has to take into account the "relativistic" nature of the magnon dispersion $\varepsilon(q) = \sqrt{\Delta^2 + v^2 k^2}$, which modifies the theoretical $T_c(H)$ curves and brings them in a good agreement with the experiment. The BEC exponent $\phi = 2/3$ is recovered only in a very narrow interval of fields close to H_c [317].

Due to the spontaneous symmetry breaking the elementary excitations in the ordered (BEC) phase become of a quasiparticle type, i.e., edge-type singularities characteristic for the purely 1D axially symmetric system (with unbroken symmetry) are replaced by delta functions. The response in the 3D-ordered (BEC) phase of TlCuCl₃ was measured in INS experiments of Rüegg et al. [318,319] and was successfully described within the bond-boson mean-field theory [320]. The observed field dependence of gaps resembles the 1D picture of Fig. 1.23, with a characteristic change of slope at $H = H_c$ where the long-range 3D order appears.

To understand the main features of the dynamics in the 3D ordered highfield phase of a weakly coupled dimer system, it is instructive to consider an effective dimer field theory which is in fact a continuum version of the very successful bond boson calculation of [320]. The theory can be constructed using dimer coherent states [321]

$$|\mathbf{A}, \mathbf{B}\rangle = (1 - A^2 - B^2)^{1/2} |s\rangle + \sum_j (A_j + iB_j) |t_j\rangle,$$
 (1.108)

where the singlet state $|s\rangle$ and three triplet states $|t_j\rangle$, j = (x, y, z) are given by (1.72), and A, B are real vectors which are in a simple manner connected with the magnetization $M = \langle S_1 + S_2 \rangle$, sublattice magnetization $L = \langle S_1 - S_2 \rangle$, and vector chirality $\kappa = (S_1 \times S_2)$ of the spin dimer:

$$M = 2(A \times B)$$
, $L = 2A\sqrt{1 - A^2 - B^2}$, $\kappa = 2B\sqrt{1 - A^2 - B^2}$.
(1.109)

We will assume that we are not too far above the critical field, so that the magnitude of the triplet components is small, $A, B \ll 1$. Assuming further that all exchange interactions are isotropic, one gets the following effective Lagrangian density in the continuum limit:

$$\mathcal{L} = \hbar (\boldsymbol{A} \cdot \partial_t \boldsymbol{B} - \boldsymbol{B} \cdot \partial_t \boldsymbol{A}) - \frac{1}{2} \beta a^2 (\nabla \boldsymbol{A})^2 - (m \boldsymbol{A}^2 + \widetilde{m} \boldsymbol{B}^2) + 2\boldsymbol{H} \cdot (\boldsymbol{A} \times \boldsymbol{B}) - \lambda_0 (\boldsymbol{A}^2)^2 - \lambda_1 (\boldsymbol{A}^2 \boldsymbol{B}^2) - \lambda_2 (\boldsymbol{A} \cdot \boldsymbol{B})^2.$$
(1.110)

Here a plays the role of the lattice constant, $(\nabla \mathbf{A})^2 \equiv (\partial_k \mathbf{A})(\partial_k \mathbf{A})$, and the energy constants β , m, \tilde{m} , $\lambda_{0,1,2}$ depend on the details of interaction between the dimers. For example, in case of purely bilinear exchange only between neighboring dimers of the type shown in Fig. 1.15, they are given by

$$\alpha = J_L + J'_L + J_D + J'_D, \quad \beta = |J_L + J'_L + J_D + J'_D|$$

$$\widetilde{m} = J, \quad m = \widetilde{m} - \beta Z/2, \qquad (1.111)$$

$$\lambda_0 = \beta Z, \quad \lambda_1 = (\alpha + \beta) Z/2, \quad \lambda_2 = -\alpha Z/2$$

The spatial derivatives of \boldsymbol{B} are omitted in (1.110) because they appear only in terms which are of the fourth order in $\boldsymbol{A}, \boldsymbol{B}$. Generally, we can assume that spatial derivatives are small (small wave vectors), but we shall not assume that the time derivatives (frequencies) are small since we are going to describe high-frequency modes as well.

The vector \boldsymbol{B} can be integrated out, and under the assumption $A \ll 1$ it can be expressed through \boldsymbol{A} as follows:

$$\boldsymbol{B} = \widehat{Q}\boldsymbol{F}, \quad \boldsymbol{F} = -\hbar\partial_t \boldsymbol{A} + (\boldsymbol{H} \times \boldsymbol{A})$$
$$Q_{ij} = (1/\widetilde{m})\,\delta_{ij} - (\lambda_2/\widetilde{m}^2)A_iA_j. \tag{1.112}$$

After substituting this expression back into (1.110) one obtains the effective Lagrangian depending on A only:

$$\mathcal{L} = \frac{\hbar^2}{\widetilde{m}} \{ (\partial_t \mathbf{A})^2 - v^2 (\nabla \mathbf{A})^2 \} - \frac{2\hbar}{\widetilde{m}} (\mathbf{H} \times \mathbf{A}) \cdot \partial_t \mathbf{A} - U_2 - U_4,$$
(1.113)

where v is the magnon velocity, $v^2 = \frac{1}{2}(\beta \tilde{m} a^2/\hbar^2)$, and the quadratic and quartic parts of the potential are given by

$$U_{2}(\boldsymbol{A}) = m\boldsymbol{A}^{2} - \frac{1}{\widetilde{m}}(\boldsymbol{H} \times \boldsymbol{A})^{2}, \qquad (1.114)$$
$$U_{4}(\boldsymbol{A}, \partial_{t}\boldsymbol{A}) = \lambda_{0}(\boldsymbol{A}^{2})^{2} + \frac{\lambda_{1}}{\widetilde{m}^{2}}\boldsymbol{A}^{2}\boldsymbol{F}^{2} + \frac{\lambda_{2}}{\widetilde{m}^{2}}(\boldsymbol{A} \cdot \boldsymbol{F})^{2}$$

Note that the cubic in A term in (1.112) must be kept since it contributes to the U_4 potential.

Now it is easy to calculate the excitation spectrum in the whole range of the applied field \boldsymbol{H} which we assume do be directed along the z axis. At zero field, there is a triplet of magnons with the gap $\Delta = \sqrt{m\tilde{m}}$, which gets trivially split by fields below the critical field $H_c = \Delta$, so that there are three distinct modes with the energies $E_{\mu} = \Delta + \mu H$, $\mu = S^z = 0, \pm 1$. For $H > H_c$ the potential energy minimum is achieved at a finite $\boldsymbol{A} = \boldsymbol{A}_0$,

$$A_0^2 = \frac{(H^2 - \Delta^2)\widetilde{m}}{2(\lambda \widetilde{m}^2 + \lambda_1 H^2)}$$

All orientations of A_0 in the plane perpendicular to H are degenerate. This U(1) symmetry is spontaneously broken, so that A_0 chooses a certain direction, let us say $A_0 \parallel x$. Then above H_c the Bose-condensed ground state is to leading order a product of single-dimer wavefunctions of the type (1.108), which mix *three* states: a singlet $|s\rangle$ and two triplets $|\uparrow\uparrow\rangle$, $|\downarrow\downarrow\rangle$. From this, it is clear that this BEC transition cannot be correctly described within an approach based on the reduced Hilbert space with only *two* states $|s\rangle$, $|\uparrow\uparrow\rangle$ per dimer.

The spectrum at $H > H_c$ can be obtained in a straightforward way. One of the modes always remains gapless (the Goldstone boson), while the two other modes have finite gaps given by

$$\Delta_z^2 = (1 - \gamma_1)^{-1} \left\{ \Delta^2 + 2\gamma_0 \widetilde{m}^2 + \gamma_1 H^2 \right\}$$

$$\Delta_{xy}^2 = \left[(1 - \gamma_1 - \gamma_2)(1 - \gamma_1) \right]^{-1} \left\{ 2(H^2 - \Delta^2) + 4H^2(1 - 2\gamma_1)^2, \right\}$$
(1.115)

where the coefficients $\gamma_{\nu} \equiv \lambda_{\nu}(H^2 - \Delta^2)/[2(\lambda_0 \tilde{m}^2 + \lambda_1 H^2)]$. In the limit of a simplified interaction with $\lambda_{1,2} = 0$ the gaps do not depend on the interaction parameters and acquire the compact form $\Delta_z = H$, $\Delta_{xy} = \sqrt{6H^2 - \Delta^2}$, which compares rather well with the INS data [318, 319] on TlCuCl₃. It is worthwhile to note a certain similarity in the field dependence of the spectra in 3D and 1D case: the quasiparticle modes in the 3D case behave roughly in the same way as the edges of continua in the 1D case.

Response in an Anisotropic System

Typically, quasi-one-dimensional materials are not completely isotropic. For example, up to our knowledge there is no experimental realization of the isotropic S = 1 Haldane chain, and in real materials like NENP or NDMAP the single-ion anisotropy leads to splitting of the Haldane triplet into three distinctive components. When the axial symmetry is *explicitly* broken, the system behavior changes drastically: the high-field phase is no more critical and acquires a long-range order even in the purely 1D case.

We will illustrate the general features of the behavior of a gapped anisotropic 1D system in magnetic field by using the example of the strongly alternated anisotropic $S = \frac{1}{2}$ chain described by the Hamiltonian

$$\mathcal{H} = \sum_{n\alpha} J_{\alpha} S_{2n-1}^{\alpha} S_{2n}^{\alpha} + \sum_{n} \{ J'(\boldsymbol{S}_{2n} \cdot \boldsymbol{S}_{2n+1}) - \boldsymbol{H} \cdot \boldsymbol{S}_{n} \}, \qquad J' \ll J.$$
(1.116)

Since this system consists of weakly coupled anisotropic dimers, one may again use a mapping to the dimer field theory as considered above for 3D coupling. One again obtains a Lagrangian of the form similar to (1.110), but the quadratic part of the potential energy gets distorted by the anisotropy: instead of $(\mathbf{mA}^2 + \tilde{\mathbf{m}B}^2)$ one now has $\sum_j \{m_j A_j^2 + \tilde{m}_j B_j^2\}$. For the alternated chain (1.116) the Lagrangian parameters are given by $m_i = \tilde{m}_i - J'$, $\tilde{m}_i = \frac{1}{4} \sum_{jn} |\epsilon_{ijn}| (J_j + J_n), \lambda_0 = J', \lambda_1 = 2J', \lambda_2 = -J', \beta = J'$. Due to this "distortion", the effective Lagrangian obtained after integrating out \mathbf{B} takes a somewhat more complicated form

$$\mathcal{L} = \frac{\hbar^2}{\widetilde{m}_i} \left\{ (\partial_t A_i)^2 - v_i^2 (\partial_x A_i)^2 \right\} - 2 \frac{\hbar}{\widetilde{m}_i} (\boldsymbol{H} \times \boldsymbol{A})_i \partial_t A_i - U_2 - U_4, \quad (1.117)$$

where $v_i^2 = \frac{1}{2} J' \widetilde{m}_i a^2 / \hbar^2$, and

$$U_{2}(\boldsymbol{A}) = m_{i}A_{i}^{2} - \frac{1}{\widetilde{m}_{i}}(\boldsymbol{H} \times \boldsymbol{A})_{i}^{2},$$

$$U_{4}(\boldsymbol{A}, \frac{\partial \boldsymbol{A}}{\partial t}) = \lambda(\boldsymbol{A}^{2})^{2} + \lambda_{1}\boldsymbol{A}^{2}\frac{1}{\widetilde{m}_{i}^{2}}F_{i}^{2} + \lambda_{2}\frac{A_{i}A_{j}}{\widetilde{m}_{i}\widetilde{m}_{j}}F_{i}F_{j}, \qquad (1.118)$$

with \boldsymbol{F} defined in (1.112).

Having in mind that the alternated $S = \frac{1}{2}$ chain, the Haldane chain, and $S = \frac{1}{2}$ ladder belong to the same universality class, one may now conjecture that in the form (1.117-1.118) the above theory can be also applied to a variety of other anisotropic gapped 1D systems, with the velocities v_i and interaction constants m_i , \tilde{m}_i , λ_i treated as phenomenological parameters.

Several phenomenological field-theoretical description of the strong-field regime in the anisotropic case were proposed in the early 90s [275, 276, 322]. One can show that the Lagrangian (1.117) contains theories of Affleck [275]

and Mitra and Halperin [322] as particular cases: after restricting the interaction to the simplified form with $\lambda_{1,2} = 0$ and assuming isotropic velocities $v_i = v$, Affleck's Lagrangian corresponds to the isotropic **B**-stiffness $\tilde{m}_i = \tilde{m}$, while another choice $\tilde{m}_i = m_i$ yields the theory of Mitra and Halperin.

For illustration, let us assume that $H \parallel \hat{z}$. Then the quadratic part of the potential takes the form

$$U_2 = (m_x - \frac{H^2}{\tilde{m}_y})A_x^2 + (m_y - \frac{H^2}{\tilde{m}_x})A_y^2 + m_z A_z^2, \qquad (1.119)$$

and the critical field is obviously $H_c = \min\{(m_x \tilde{m}_y)^{1/2}, (m_y \tilde{m}_x)^{1/2}\}$. At zero field the three triplet gaps are given by $\Delta_i = (m_i \tilde{m}_i)^{1/2}$. Below H_c the energy gap for the mode polarized along the field stays constant $E_z = \Delta_z$, while the gaps for the other two modes are given by

$$(E_{xy}^{\pm})^{2} = \frac{1}{2}(\Delta_{x}^{2} + \Delta_{y}^{2}) + H^{2}$$

$$\pm \left[(\Delta_{x}^{2} - \Delta_{y}^{2})^{2} + H^{2}(m_{x} + m_{y})(\tilde{m}_{x} + \tilde{m}_{y}) \right]^{1/2}.$$
(1.120)

Below H_c the mode energies do not depend on the interaction constants λ_i , while the behavior of gaps at $H > H_c$ is sensitive to the details of the interaction potential.

It is easy to see that in the special case $m_i = \tilde{m}_i$, the above expression transforms into

$$E_{xy}^{\pm} = \frac{1}{2} (\Delta_x + \Delta_y) \pm \left[\frac{1}{4} (\Delta_x - \Delta_y)^2 + H^2 \right]^{1/2}, \qquad (1.121)$$



Fig. 1.24. Measured field dependence of the gap energies in NDMAP at T = 30 mK and H applied along the crystallographic a axis (open symbols). Dashed and dashdot lines are predictions of the theoretical models proposed in [275] and [276], respectively. The solid lines are the best fit to the data using the alternative model (1.117). (From [304])

which exactly coincides with the formulas obtained in the approach of Tsvelik [276], as well as with the perturbative formulas of [323, 324] and with the results of modified bosonic theory of Mitra and Halperin [322] who postulated a bosonic Lagrangian to match Tsvelik's results for the field dependence of the gaps below H_c .

The present approach was applied to the description of the INS [304] and ESR [325] experiments on the S = 1 Haldane material NDMAP and yielded a very good agreement with the experimental data, see Fig. 1.24. It turns out that for a satisfactory quantitative description the inclusion of $\lambda_{1,2}$ is important, as well as having unequal stiffness constants $m_i \neq \tilde{m}_i$.

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2 Quantum Magnetism in Two Dimensions: From Semi-classical Néel Order to Magnetic Disorder

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Abstract. It is known from the Mermin-Wagner theorem that magnetic long-range order can exist in two dimensions only at zero temperature, but even then it can still be destroyed e.g. by quantum fluctuations or geometric frustration. In this context, we review ground-state features of the s = 1/2 Heisenberg antiferromagnet on two-dimensional lattices. In order to discuss the interplay of lattice topology and quantum fluctuations we focus on the 11 two-dimensional uniform Archimedean lattices which include e.g. the square, triangular and kagomé lattice. The ground state of the spin-1/2 Heisenberg antiferromagnet is likely to be semi-classically ordered in most cases. However, the interplay of geometric frustration and quantum fluctuations gives rise to a quantum paramagnetic ground state without semi-classical long-range order on two lattices which are precisely those among the 11 uniform Archimedean with a highly degenerate ground state in the classical limit $s \to \infty$. The first one is the famous kagomé lattice where many low-lying singlet excitations are known to arise in the spin gap. The second lattice, called star lattice, is a new example for a quantum paramagnet and has a clear gap to all excitations.

Modification of certain bonds leads to quantum phase transitions which are also discussed briefly. Furthermore, we discuss the magnetization process of the Heisenberg antiferromagnet on the 11 Archimedean lattices, focusing on anomalies like plateaus and a magnetization jump just below the saturation field. As an illustration we discuss the two-dimensional Shastry-Sutherland model which is used to describe $SrCu_2(BO_3)_2$.

2.1 Introduction

The subject of quantum spin-half antiferromagnetism in two-dimensional (2D) systems has attracted a great deal of interest in recent times in connection with the magnetic properties of layered cuprate high-temperature superconductors [1–3] and with the recent progress in synthesizing novel quasi-2D magnetic materials exhibiting a spin-gap behavior like CaV_4O_9 [4] or $SrCu_2(BO_3)_2$ [5]. Another striking feature is the plateau structure in the magnetization process of frustrated quasi-two-dimensional magnetic materials like $SrCu_2(BO_3)_2$ [5] or Cs_2CuBr_4 [6] (for more details concerning the experiments see chapter by P. Lemmens and P. Millet in this book). However, low-dimensional quantum spin systems are of interest in their own right as examples of strongly interacting quantum many-body systems. Although we know from the Mermin-Wagner theorem [7] that thermal fluctuations are strong enough to destroy magnetic long-range order (LRO) for Heisenberg spin systems in one and two dimensions at any finite temperature, the role of quantum fluctuations is less understood. For the magnetic ordering in the ground state (GS) the transition from one to two dimensions seems to be crucial. It is well known that the GS of the one-dimensional Heisenberg quantum antiferromagnet does not possess Néel LRO (see chapter by H.-J. Mikeska and A.K. Kolezhuk in this book). On the other hand as a result of intensive work in the late eighties it is now well-established that the GS of the Heisenberg antiferromagnet on the square lattice exhibits semi-classical Néel LRO (see for example the reviews [1,2]). However, Anderson's and Fazekas' investigations [8,9] of the triangular lattice led to the conjecture that quantum fluctuations plus frustration may be sufficient to destroy the Néel-like LRO in two dimensions.

Besides frustration, there is another mechanism favoring the "melting" of Néel ordering in the ground states of unfrustrated Heisenberg antiferromagnets, namely the competition of non-equivalent nearest-neighbor (NN) bonds leading to the formation of local singlets of two (or even four) coupled spins. By contrast to frustration, which yields competition in quantum as well as in classical systems, this type of competition is present only in quantum systems.

Several notations for the quantum phases without semi-classical Néel order are used in the literature, where one often finds the terms 'quantum disorder' or 'quantum spin liquid'. However, these quantum phases may exhibit quite different complex properties. We shall prefer the notation 'quantum paramagnet' (see, e.g. [10]) to stress their common feature, namely the absence of magnetic order at T = 0.

A more specific classification of GS phases of 2D quantum magnets has been proposed recently by Lhuillier, Sindzingre, Fouet and Misguich [11–15]. Besides the semi-classical Néel like LRO, these authors also characterize three quantum GS phases, namely the so-called valence bond crystal, the type I spin liquid and the type II spin liquid (for more details see [11–15] and also Sect. 2.4.4).

We note that quantum paramagnetic phases may be observed also in three-dimensional strongly frustrated quantum magnets like the Heisenberg antiferromagnet on the pyrochlore lattice [16] although the tendency to order is more pronounced in three than in two dimensions.

In this review we focus on the GS of the 2D isotropic Heisenberg antiferromagnet (HAFM)

$$H = \sum_{\langle i,j \rangle} J_{ij} \mathbf{S}_i \mathbf{S}_j = \sum_{\langle i,j \rangle} J_{ij} \left(S_i^x S_j^x + S_i^y S_j^y + S_i^z S_j^z \right)$$
(2.1)

and consider the extreme quantum case of spin quantum number 1/2. Of course, there is a long history of investigations of this model. Nevertheless, much interesting new physics has been discovered in recent years. The 2D systems are of particular interest because the competition between quantum fluctuations and interactions seems to be well balanced, and fine tuning of this competition may lead to zero-temperature transitions between semiclassical and quantum phases (see chapter by S. Sachdev in this book and also Sect. 2.5).

The calculation of the GS of the spin half HAFM is challenging. Besides the conventional methods like spin-wave theory and general quantummany body techniques like the coupled cluster method also new numerical methods like quantum Monte Carlo and exact diagonalization are powerful instruments. However, only a few of them (e.g. exact diagonalization or the coupled cluster method) are universally applicable, whereas some methods suffer from the sign problem in frustrated systems. More details regarding analytical and numerical methods can be found in chapters by N.B. Ivanov and D. Sen; D.C. Cabra and P. Pujol; N. Laflorencie and D. Poilblanc; D.J.J. Farnell and R.F. Bishop. The majority of the results presented in this chapter were obtained by exact diagonalization using the program package spinpack [17].

Quantum magnetism in 2D systems is a very broad field. To be specific and different from other existing reviews we focus our discussion on the ground state properties of the spin half HAFM on the 11 uniform Archimedean lattices (tilings). These lattices are the prototypes of 2D arrangements of spins and vary in their geometrical and topological properties. Hence they present an ideal possibility for a systematic study of the interplay of lattice geometry and magnetic interactions in 2D quantum spin systems. Many of the lattices considered find their realization in nature either in a pure or in a modified form. Furthermore, almost all lattices can be transformed into each other by bond or site depletion/addition. One now has the opportunity to study GS transitions caused by modifying the strength of some bonds [18].

With regard to other aspects of 2D quantum magnetism like e.g. finite temperature properties we recommend among others [1, 2, 12, 13, 19–22].

The plan of this review is as follows. In Sect. 2.2 we describe the main geometrical features of the 11 uniform Archimedean lattices and discuss their mutual relationships. In Sect. 2.3 we discuss several criteria for semi-classical Néel like order in quantum antiferromagnets with a particular focus on the information that can be extracted from exact diagonalization of finite lattices. The subsequent Sect. 2.4 is devoted to the analysis of the magnetic ground-state ordering of the spin-half HAFM on the Archimedean lattices, where we consider separately bipartite (Sect. 2.4.1) and frustrated (Sects. 2.4.2 and 2.4.3) lattices. The findings for all these lattices are compared and sum-

marized in Sect. 2.4.4. Readers uninterested in the detailed discussion of the particular lattices are referred to this Sect. 2.4.4. In Sect. 2.5 we consider briefly quantum phase transitions occurring in the 2D HAFM due to the interplay of competition in the interactions and strong quantum fluctuations. In the final Sect. 2.6 we discuss the magnetization process of the spin-half HAFM on the Archimedean lattices using the square (Sect. 2.6.1), triangular (Sect. 2.6.2) and kagomé lattice (Sect. 2.6.3) as main examples. We further discuss exact eigenstates that appear for the kagomé and star lattices in Sect. 2.6.4 and the relation between the Shastry-Sutherland model and $SrCu_2(BO_3)_2$ in Sect. 2.6.5.

2.2 Archimedean Lattices

2.2.1 Characteristics and Geometry

In 2D magnetism we are faced with a large number of different lattices with differing coordination numbers and topologies and therefore we cannot expect a general statement concerning zero-temperature semi-classical Néel-like LRO in 2D quantum spin systems. Nevertheless, we can try to find some systematics concerning the main geometric features relevant for the magnetic ordering in antiferromagnets.

The 11 uniform Archimedean tilings (lattices) shown in Fig. 2.1 represent the prototypes of 2D arrangements of regular polygons. The first investigations of 2D regular tilings go back to Johannes Kepler (*Harmonice Mundi*, 1619). 2D (spin) lattices are obtained from the tilings by putting sites (spins) on each vertex connecting neighboring polygons. The HAFM for these lattices is obtained by assuming antiferromagnetic exchange bonds J = 1 on each edge of the polygons.

The Archimedean lattices vary in coordination number z (from 3 to 6) and in topology (frustrated and nonfrustrated; equivalent nearest-neighbor (NN) bonds and non-equivalent NN bonds). Therefore a systematic study of the influence of lattice geometry on magnetic ordering may be made.

Among them we have three 2D lattices built by a periodic arrangement of **identical** regular polygons, namely the square lattice (T2), the triangular lattice (T1) and the honeycomb lattice (T3). Other uniform tilings are obtained by combining different regular polygons such as hexagons and triangles or hexagons, squares and triangles with the restriction that all lattice sites are equivalent and all polygons have identical edge length. Under these geometric restrictions precisely 11 uniform Archimedean tilings are possible, where one tiling exists in two enantiomorphic forms (left and right handed). Only two of them, namely the square lattice (T2), and the triangular lattice (T1) are primitive lattices having only one site per geometric unit cell; all other ones have at least two sites per unit cell. More information can be found, for example, in [23].



Fig. 2.1. The 11 Archimedean tilings T1...T11. The mathematical description $n_1.n_2.n_3...n_r$ by numbers n_i separated by dots corresponds to the number of vertices of the polygons arranged around a vertex. The tilings T1, T2, T3, T8 are well-known as triangular (T1), square (T2), honeycomb (T3) and kagomé (T8) lattices. For the other lattices no standardized names are available. For T4, T5, T6, T10 and T11 we employ the names maple-leaf (T4), trellis (T5), SrCuBO or Shastry-Sutherland (T6), SHD (i.e. square-hexagonal-dodecagonal, T10) and CaVO (T11) lattice previously used in papers dealing with magnetic properties of these lattices (see also Sect. 2.4). We shall denote the tilings T7 and T9 by the names bounce (T7) and star (T9) lattice, proposed in [24]

In this section we will illustrate the Archimedean tilings and discuss their main geometric properties. As mentioned above, they represent the prototypes of 2D tilings, from which a large variety of 2D lattices can be derived. As a result we obtain bipartite, i.e. non-frustrated (only even polygons, tilings T2, T3, T10, T11) as well as non-bipartite, i.e. frustrated spin lattices (tilings with odd polygons (triangles), i.e. T1, T4, T5, T6, T7, T8, T9). Furthermore, we can differentiate between lattices with only equivalent NN bonds (T1, T2, T3, T8) and lattices with non-equivalent NN bonds (T4, T5, T6, T7, T9, T10, T11).

The degree of geometric frustration and the coordination number are important quantities that strongly influence the magnetic properties. In order to give a more precise characteristics of the frustration, we use an idea proposed by Kobe and coworkers [25] and consider the GS energy of the classical HAFM (i.e. the spins **S** are ordinary classical vectors of length s = 1/2). Non-frustrated lattices (T2, T3, T10, T11) have minimal energy per bond $E_0^{\text{class}}/\text{bond} = -1s^2$. Geometric frustration leads to unsatisfied bonds yielding an increase of classical GS energy. This increase of energy can be used as a measure of frustration. The tilings with maximal frustration are the triangular lattice (T1) and the kagomé lattice (T8) having $E_0^{\text{class}}/\text{bond} = -s^2/2$. The combination of strong frustration and low coordination number z favors strong quantum fluctuations. In Fig. 2.2 we show the location of the lattices in a parameter space spanned by the coordination number z and the frustration. The suppression of classical Néel-like LRO is most likely in the upper left corner in Fig. 2.2, whereas in the opposite region Néel ordered systems are expected.



Fig. 2.2. Location of the Archimedean tilings in parameter space spanned by frustration (classical GS energy per bond divided by s^2 , see text) and coordination number z

2.2.2 Relationships Between the Lattices

As mentioned above, we interpret the edges of the polygons as exchange bonds which connect the spins sitting on the vertices. In real magnetic systems often we are faced with the situation that bonds may vary in strength for instance due to lattice distortions. Hence it is interesting to consider also bonds varying in strength. In particular, a given lattice may interpolate into another different lattice as selected bonds are forced to reach the limit J' = 0. The relationships between the Archimedean lattices based on removing bonds are summarized in Fig. 2.3.⁴ A continuous change of those bonds from J' = 1to J' = 0 is therefore accompanied by a transition or a crossover between the ground states of the related lattices. We illustrate some of these relationships between lattices in Figs. 2.4 and 2.5.



Fig. 2.3. Relationships (arrows) between the tilings (represented by numbers). A related tiling is obtained from an initial tiling by removing certain edges (bonds) and a subsequent appropriate distortion



Fig. 2.4. Relationships between triangular (T1, left), square (T2, middle) and honeycomb lattice (T3, right), see text for details

⁴ It is also possible to find transformations between lattices by removing sites (site depletion). That is not considered here.



Fig. 2.5. Relationships between the honeycomb (left), maple-leaf (middle) and bounce lattice (right), see text for details

Figure 2.4 shows the relationships between triangular, square and honeycomb lattices. The square lattice is obtained from the triangular lattice by omitting the dotted bond J''. The geometric distortion of the square lattice obtained in this manner is irrelevant for the HAFM because the interaction matrix J_{ij} of the distorted lattice is identical to the regular lattice.⁵ The honeycomb lattice is then obtained from the square lattice by omitting the dashed bonds J' (the model with variable J' is known as the J - J' model on the square lattice and shows interesting quantum phase transitions [21, 26, 27]).

In Fig. 2.5 the relationships between the honeycomb, the maple-leaf and the bounce lattice are shown. Starting from the maple-leaf lattice one obtains the bounce lattice by omitting the dotted bonds J_D . Further removing the dashed-dotted bond in the bounce lattice one obtains the star lattice (T9, not shown in Fig. 2.5). On the other hand the honeycomb lattice is obtained from the maple-leaf lattice by removing the dashed bonds J_T . Again the distortion of the lattices is irrelevant for the HAFM.

2.3 Criteria for Néel Like Order

2.3.1 Order Parameter

The definition of the magnetic order parameter is usually related to the classical ground state (GS). Thus supposing that in the classical GS a spin at site *i* is directed along the unit vector \mathbf{e}_i , we choose the spin orientation \mathbf{e}_i as local *z*-direction $\mathbf{e}_i^{z'} = \mathbf{e}_i$, which may in general vary from site to site. In order to break the rotational symmetry we add a field to the Hamiltonian (2.1)

⁵ Of course, the distorted lattices obtained by removing bonds may also be transformed to the regular (non-distorted) lattice by an appropriate shift of the sites.

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$$H' = H - h \sum_{i} \mathbf{e}_{i}^{z'} \mathbf{S}_{i}.$$
 (2.2)

We define the order-parameter operator as

$$\hat{m}^{z} = \frac{1}{N} \sum_{i} S_{i}^{z'} = \frac{1}{N} \sum_{i} \mathbf{e}_{i}^{z'} \mathbf{S}_{i}.$$
(2.3)

Then the order parameter for a GS spontaneously breaking the rotational symmetry of H is defined as

$$m^{z} = \lim_{h \to 0} \lim_{N \to \infty} \langle \hat{m}^{z} \rangle, \qquad (2.4)$$

where $\langle \hat{O} \rangle$ means the expectation value of the operator \hat{O} in the GS. This definition of the order parameter corresponds, e.g., to the order parameter used in spin-wave theory (SWT). However, symmetry breaking is introduced in this case by the Holstein-Primakoff transformation, which starts from a symmetry broken classical GS. In order to be more specific let us consider a classical spin system having a **planar** magnetic GS ordering. We choose the z-x plane of a fixed global coordinate system to describe the order. Then the relation between the spin \mathbf{S}'_i in the local coordinate system and the spin \mathbf{S}_i in the global coordinate system is given by

$$\mathbf{S}'_{i} = \hat{U}(\phi_{i})\mathbf{S}_{i} = (\cos(\phi_{i})S_{i}^{x} - \sin(\phi_{i})S_{i}^{z}, S_{i}^{y}, \sin(\phi_{i})S_{i}^{x} + \cos(\phi_{i})S_{i}^{z}),$$
(2.5)

where ϕ_i is the angle between the local $\mathbf{e}_i^{z'}$ and the global z axis \mathbf{e}^z . The last component in (2.5) enters the order-parameter operator \hat{m}^z in (2.3).

The definition of the order-parameter operator (2.3) yields the well-known order parameter of the ferromagnet ($\phi_i = 0$) $m^z = \lim_{h \to 0; N \to \infty} \frac{1}{N} \langle \sum_i S_i^{z'} \rangle = \lim_{h \to 0; N \to \infty} \frac{1}{N} \langle \sum_i S_i^{z} \rangle$ (magnetization) as well as the order parameter for the conventional two-sublattice Néel antiferromagnet ($\phi_{i \in A} = 0, \phi_{i \in B} = \pi$) $m_s^z = \lim_{h \to 0; N \to \infty} \frac{1}{N} \langle \sum S_i^{z'} \rangle = \lim_{h \to 0; N \to \infty} \frac{1}{N} \langle \sum \epsilon_i S_i^z \rangle$ (staggered magnetization), where the staggered factor ϵ_i is $\epsilon_i = +1$ ($\epsilon_i = -1$) for sites belonging to sublattice A(B). The staggered magnetization can be expressed by the sublattice magnetizations $S_A^z = \sum_{i \in A} S_i^z$ and $S_B^z = \sum_{i \in B} S_i^z$, we have $m_s^z = \frac{1}{N} \langle S_A^z - S_B^z \rangle_{h \to 0, N \to \infty}$. The general definition (2.4) is also applicable for non-collinear (canted) spin structures appearing on frustrated lattices. For example, the classical GS of the HAFM on the triangular lattice consists of three sublattices A, B, C with an angle of 120° between the sublattice spins, i.e. we have $\phi_{i \in A} = 0, \phi_{i \in B} = 2\pi/3$ and $\phi_{i \in C} = 4\pi/3$. Consequently we find $m^z = \frac{1}{N} \langle S_A^z + \sqrt{3} S_B^z / 2 - S_B^z / 2 - \sqrt{3} S_C^z / 2 - S_C^z / 2 \rangle_{h \to 0, N \to \infty}$. The extension to arbitrary non-collinear spin structures is straightforward.

The situation is changed for the HAFM on finite lattices considered in numerical studies because the GS of a finite system cannot possess the spontaneous symmetry breaking used for the infinite lattice ((2.2)-(2.4)). Therefore

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the square of the order-parameter operator $(\hat{m}^z)^2$ has to be used. Furthermore, we have to take into account the fact that the GS of finite antiferromagnetic systems with even number of sites N is a rotationally invariant singlet state.⁶ Then the magnetic correlations are equally distributed over all three components $\langle S_i^x S_j^x \rangle = \langle S_i^y S_j^y \rangle = \langle S_i^z S_j^z \rangle$. Thus, taking into account this symmetry, one defines the relevant order parameter for finite systems as

$$\bar{m} = \sqrt{\left\langle \left(\frac{1}{N}\sum_{i}\mathbf{S}_{i}^{\prime}\right)^{2}\right\rangle} = \sqrt{3\langle (\hat{m}^{z})^{2}\rangle}.$$
(2.6)

One may write this order parameter as

$$\bar{m} = \sqrt{\left\langle \left(\frac{1}{N}\sum_{i}\epsilon_{i}\mathbf{S}_{i}\right)^{2}\right\rangle} = \sqrt{\frac{1}{N^{2}}\sum_{i,j}\epsilon_{i}\epsilon_{j}\left\langle \mathbf{S}_{i}\mathbf{S}_{j}\right\rangle}$$

$$= \sqrt{\frac{3}{N^{2}}\left\langle (S_{A}^{z})^{2} + (S_{B}^{z})^{2} - 2S_{A}^{z}S_{B}^{z}\right\rangle}$$
(2.7)

for bipartite antiferromagnets and

$$\bar{m} = \sqrt{\frac{3}{N^2}} \langle (S_A^z)^2 + (S_B^z)^2 + (S_C^z)^2 - S_A^z S_B^z - S_A^z S_C^z - S_B^z S_C^z \rangle$$
(2.8)

for the triangular lattice. Note that $\langle S_i^x S_j^z \rangle = 0$ was used in the last equation. Obviously the analysis of magnetic order is then based on the spin pair correlation function $\langle \mathbf{S}_i \mathbf{S}_j \rangle$. We notice that an alternative definition to (2.4) of the order parameter for infinite systems uses the asymptotic large-distance behavior of the spin pair correlation function.

The order parameter \bar{m} is widely used for finite lattices such as square or triangular lattices. Finite-size extrapolations of \bar{m} yield good agreement with m^z defined in (2.4) calculated e.g. by spin-wave theory or the coupled cluster method (see e.g. [31–33]).

However, the definition of the order parameter given above is to some extent problematic for the following reasons: (i) The definition is biased because it supposes the same type of ordering in the quantum system as in the classical system. Investigations of spin systems with spiral order demonstrate, that the characteristic angles ϕ_i entering (2.5) may be different in the classical and quantum case [27]. (ii) There are systems with a huge non-trivial degeneracy of the classical GS (e.g. the HAFM on T8 (kagomé) and on T9 (star), see Sect. 2.4.3). The question now arises: which of the large number of degenerate classical ground states should be used? (iii) A significant problem is also posed if the classical GS is not known.

⁶ Note that although there is a rigorous proof for the singlet character of the GS of finite systems only for the HAFM on bipartite lattices [28, 29], much numerical evidence suggests that the same statement is true for nonbipartite frustrated antiferromagnetic Heisenberg systems (see e.g. [30]).

We therefore use a universal definition of the order parameter, given by

$$m^{+} = \sqrt{\frac{(M^{+})^{2}}{N^{2}}} = \left(\frac{1}{N^{2}} \sum_{i,j}^{N} |\langle \mathbf{S}_{i} \mathbf{S}_{j} \rangle|\right)^{1/2}, \qquad (2.9)$$

which avoids the problems listed above. For bipartite systems this definition is identical to the staggered magnetization \bar{m} defined in (2.7). For spin systems with noncollinear GSs both definitions \bar{m} and m^+ are not identical, although there is a relation between them. For instance, we have in the classical limit $(m_{\rm class}^+)^2 = \frac{4}{3}(\bar{m}_{\rm class})^2$ for the HAFM on the triangular lattice. Note that this relation remains valid also for the singlet GS of the quantum spin-1/2 HAFM on the triangular lattice.

Finally, we mention that the universality of the definition (2.9) of the order parameter may lead to a certain loss of distinction between different types of ordering and for the detection of the type of order an additional inspection of the spin-spin correlation function is necessary.

2.3.2 Mechanism of Symmetry Breaking – The Pisa Tower of Quasi-degenerate Joint States (QDJS)

As pointed out already by P.W. Anderson [34] the spontaneous symmetry breaking in semi-classically Néel ordered antiferromagnets at the thermodynamic limit is revealed in the spectrum of a finite system. This idea has been picked-up in several papers [31,35–45] dealing with two-dimensional quantum antiferromagnetism.

In the limit $N \to \infty$ a whole set of non-rotationally invariant excited states collapses onto the true GS (e.g. the semi-classical two-sublattice Néel state for the HAFM on a bipartite lattice). Therefore a large amount of information on possible Néel like LRO is contained in the spectrum of HAFM on finite lattices. There are extensive systematic studies for HAFM on the square, triangular and kagomé lattice [31,36–39,42] and some recent reviews by Lhuillier, Sindzingre, Fouet and Misguich [11–15]. We follow the lines of their studies and illustrate some main features using the HAFM on the square lattice as an example.

For a system with two-sublattice Néel LRO in the GS the low-energy part of the spectrum up to $S \approx \sqrt{N}$ is roughly described by the dynamics of a quantum top, i.e. the effective low-energy Hamiltonian reads

$$H_{\text{eff}} \simeq E_0 + \frac{\mathbf{S}^2}{2N\chi_0} + H_{\text{magnons}} \rightarrow E_{\text{min}}(S) \simeq E_0 + \frac{S(S+1)}{2N\chi_0} + E_{\text{magnons}}$$
(2.10)

with E_0 as the GS energy, χ_0 as uniform susceptibility and \mathbf{S}^2 as the square of total spin, cf. Fig. 2.6. The inverse 'moment of inertia' $\frac{1}{N\chi_0}$ vanishes in



Fig. 2.6. HAFM on the square lattice. a: Low-energy spectrum for N = 32 (the inset shows the **k** points in the Brillouin zone). b: Energy of the QDJS versus S(S + 1) for various system sizes N

the thermodynamic limit (see Fig. 2.7b) and therefore the so-called quasidegenerate joint states (QDJS) described by (2.10) collapse to the symmetry broken Néel state in the thermodynamic limit. In case of more complex Néel order e.g. with three sublattices as for the HAFM on the triangular lattice the basic features of the low-energy Hamiltonian as given in (2.10)are maintained but the moment of inertia then contains both in-plane and out-of-plane susceptibilities. Also the number of the QDJS for a given total spin S depends on the number of sublattices in the Néel state. There is only one QDJS in each sector of S for the two-sublattice HAFM, but e.g. $N_S = \min(2S+1, N/2 - S + 1)$ QDJS for a three-sublattice Néel state such as in the triangular lattice [37]. Furthermore, the translational symmetry of the QDJS depends on the relation between the translational symmetry of the Néel state and of the lattice. For instance, the size of the magnetic unit cell for the Néel ordered square-lattice (triangular-lattice) HAFM is twice (three times) as large as the geometric unit cell. Consequently, the QDJS belong to k-vectors $\mathbf{Q}_1 = (0,0)$ and $\mathbf{Q}_2 = (\pi,\pi)$ ($\mathbf{Q}_1 = (0,0)$ and $\mathbf{Q}_2^+ = (+4\pi/3, 0), \ \mathbf{Q}_2^- = (-4\pi/3, 0))$ of lattice translational symmetry with \mathbf{Q}_2 (\mathbf{Q}_2^+ and \mathbf{Q}_2^-) mapping on the center of the magnetic Brillouin zone. However, only $\mathbf{Q} = (0,0)$ appears for the QDJS for the honeycomb lattice which has two atoms in the geometric unit cell as well as in the magnetic unit cell.
Indeed, a linear relation between the lowest eigenvalues $E_{\min}(S)$ and S(S+1) and a similar relation for the family of magnon excitations has been observed for the HAFM on the 2D square lattice [35], the honeycomb lattice [42] as well as for the triangular-lattice HAFM [37]. We show in Fig. 2.6a the low-energy spectrum for the HAFM on the square lattice with N = 32 sites in more detail.

According to (2.10) the collapsing QDJS follow in the energy-S(S + 1)diagram for small $S \leq \sqrt{N}$ in good approximation a straight line with increasing inclination (with decreasing slope), see Fig. 2.6b, and are sometimes called the Pisa tower of states. The strong deviation from this linear relation has been used as one argument for the absence of semi-classical LRO for the HAFM on the kagomé lattice [38]. A similar argumentation has been used in [40] for the $J_1 - J_2$ square-lattice HAFM and in [41] for the HAFM on the fractal Sierpiński gasket.

Well separated above the family of QDJS a second family of levels exists describing the magnon excitations typical for a HAFM with Néel ordering. This family represents the 'softest magnons', i.e. magnons of energy $E_{\min}^{M} = c |\mathbf{k}_{\min}|$ with c as lowest spin-wave velocity and $|\mathbf{k}_{\min}| \sim \frac{1}{L}$ ($L = \sqrt{N}$) as smallest finite wave vector (related to the wave vector \mathbf{Q} of the corresponding QDJS) allowed by the periodic boundary conditions of the finite lattice. The energy of these magnons also collapses, however, with

$$E_{\min}^{M} \simeq \frac{c}{\sqrt{N}} \tag{2.11}$$

much slower than $E_{\min}(S) \propto \frac{1}{N}$ from (2.10). This scaling behavior of the QDJS and of the softest magnons is shown in Fig. 2.7a, where the logarithmic scale in this figure makes it obvious that the slope of the $E_0(S = 1) - E_0$ -curve belonging to the QDJS is about twice as large as the slope of the $E_1(S = 1) - E_0$ -curve belonging to the softest magnons.

Finally we want to emphasize two special aspects of the spectrum of semiclassically Néel ordered HAFMs. The first one is the so-called spin gap, i.e. the gap $\Delta = E_0(S = 1) - E_0(S = 0)$ between the first triplet excitation and the singlet GS. According to (2.10) and (2.11) this gap vanishes in the thermodynamic limit for a Néel ordered GS. Note that (2.11) is related to the existence of gapless Goldstone modes. However, a non-vanishing spin gap for $N \to \infty$ is an indication for a quantum paramagnetic GS. The second aspect is that the singlet GS is unique and the lowest singlet excitation above the GS is well separated from it in a finite system (see Fig. 2.6a). The first triplet excitation above the rotationally invariant singlet GS generally is the lowest excitation at all. Therefore the existence of low-lying singlets deep within the spin gap also can be understood as an indication for a non-Néel ordered GS.

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Fig. 2.7. HAFM on the square lattice. a: Finite-size scaling of lowest excitations (double logarithmic scale). b: Finite-size scaling of inverse moment of inertia

2.3.3 Finite-Size Scaling

Effective continuum field-theory studies as well as spin-wave theory and quantum Monte Carlo (QMC) calculations have led to detailed predictions for the low-energy physics and the finite-size scaling of Néel ordered quantum antiferromagnets in two dimensions [35,44–47], which we will use below in (2.12), (2.13) and (2.14). As already discussed in the last section the inverse 'moment of inertia' is obtained from the QDJS and so the spin gap scales in leading order with 1/N, see (2.10). However, for finite-lattice sizes accessible in exact diagonalization the asymptotic behavior is often not reached and boundary effects are important. Hence, the extrapolation to $N \to \infty$ possesses some uncertainty. According to [35, 44–47] the GS energy per site $e_0 = \frac{E_0}{N}$ for a semi-classical Néel state scales as

$$e_0(L) = A_0 + \frac{A_3}{L^3} + \mathcal{O}(L^{-4})$$
(2.12)

where $L = N^{1/2}$ is the linear size of the lattice, $A_0 = e_0(\infty)$ and A_3 is proportional to the spin-wave velocity c. For the order parameter m^+ we use

$$m^+(L) = B_0 + \frac{B_1}{L} + \mathcal{O}(L^{-2})$$
 (2.13)

where $B_0 = m^+(\infty)$. For the spin gap Δ we apply

$$\Delta(L) = G_0 + \frac{G_2}{L^2} + \mathcal{O}(L^{-3})$$
(2.14)

where $G_0 = \Delta(\infty)$. In case that there are many appropriate finite lattices with $N \leq 36$ the large number of data points leads to reliable extrapolation to the thermodynamic limit [32,33]. On the other hand, for systems with only a few appropriate finite lattices the extrapolation is much stronger influenced by the boundary effects and the extrapolated results exhibit a larger uncertainty. Furthermore, only the leading terms in (2.12), (2.13), (2.14) can be used in case of a small number of data points. Particular problems may arise for the spin gap: (i) Boundary effects are present in both E_0 and E_1 leading to a larger error in $E_1 - E_0$. (ii) As discussed in the last section the first triplet excitation belongs to the QDJS with a definite symmetry. However, it may appear that this symmetry is not present in a certain finite lattice, i.e. the calculated first excitation belongs to another symmetry and consequently it has higher energy leading to an overestimation of the gap. Therefore the extrapolation of the gap will not be a main focus of our discussion of the ordering of the HAFM on the Archimedean tilings in Sect. 2.4.

We use only the leading terms even in case that the number of data points would allow a scaling including next-to-leading term in order to have the same systematics for all the 11 Archimedean tilings in the comparative discussion given below. By way of illustration we compare the results obtained by both variants of extrapolation for the square lattice (for a comparison with data available in literature, see Sect. 2.4.1):

- GS energy per site: $E_0/N = -0.6701$ (leading term only); $E_0/N = -0.6685$ (next-to-leading term included);
- singlet-triplet gap: $\Delta = 0.0605$ (leading term only); $\Delta = 0.0247$ (next-to-leading term included);
- order parameter: $m^+ = 0.3173$ (leading term only); $m^+ = 0.3235$ (next-to-leading term included).

For the extrapolation altogether 12 finite square lattices from N = 18 to N = 40 are used, the next-to-leading terms changes the energy by less than 0.2% and the order parameter by less than 2%. The values for the extrapolated gap can be understood as a measure of the accuracy of the extrapolation, since we know that the excitations about a Néel ordered GS become gapless for $N \to \infty$.

Finally, we mention that the finite-size scaling for systems with a critical GS or with a GS having only short-range spin pair correlations $\langle \mathbf{S}_i \mathbf{S}_j \rangle$ can be different from the above given relations. The concrete relations may depend on details of GS correlations. Nevertheless one aspect shall be noted: due to the absence of long-range correlations in $\langle \mathbf{S}_i \mathbf{S}_j \rangle$ the finite-size effects should be weaker for the GS energy. As a simple example we can consider a HAFM with a valence-bond GS as realized for the Shastry-Sutherland model for stronger frustration (see Sect. 2.5). The GS energy per site is completely independent of N in this case.

2.4 Magnetic Ground-State Ordering for the Spin Half HAFM on the Archimedean Lattices

In this section we present and discuss results obtained by exact diagonalization for the 11 Archimedean lattices. For some of these lattices such studies have not been performed so far or the presented results go beyond the system sizes published until now. If available we also discuss results obtained by other methods to get a reliable picture on the magnetic ordering. From our results we conclude that three categories of ground state ordering appear: Collinear two-sublattice Néel long-range order (LRO), non-collinear (multi-sublattice Néel or spiral) LRO and quantum paramagnetic ground states without LRO in the spin pair correlation $\langle \mathbf{S}_i \mathbf{S}_j \rangle$.

At first we consider in Sects. 2.4.1, 2.4.2 and 2.4.3 each lattice separately and present results for the GS energy, the spin gap and the order parameter as well as the spectrum. In a second step we summarize and compare in Sect. 2.4.4 the magnetic ordering on the various Archimedean lattices. We refer the reader who is not interested in the detailed discussion of the individual lattices to Sect. 2.4.4.

2.4.1 Semi-classical Néel Ordering on Bipartite Lattices

The classical GS for bipartite lattices is the perfect Néel state having a GS energy per bond $E_0^{\text{class}}/\text{bond} = -s^2 = -0.25$ and an order parameter $m_{\text{class}}^+ = s = 0.5$. However, this classical order is very sensitive to fluctuations. Indeed the 1D HAFM does not exhibit Néel LRO. For the 2D HAFM we know from the Mermin-Wagner theorem [7] that at arbitrarily small finite temperatures T the thermal fluctuations are strong enough to destroy the Néel LRO. However, it was for a long time an open question whether also quantum fluctuations are able to destroy Néel LRO in 2D at absolute zero. Each 2D lattice needs its individual consideration because the strength of quantum fluctuations can vary from lattice to lattice. Stronger quantum fluctuations appear in lattices with low coordination number z and in lattices with non-equivalent NN bonds. Although this non-equivalency of NN bonds is irrelevant for classical bipartite HAFM it leads to a competition between the bonds in quantum models. This quantum competition favors local singlet formation weakening that way Néel ordering (see, e.g. [27, 48] and Sect. 2.5).

The Square Lattice (T2)

Starting from P.W. Anderson's pioneering work [34], the spin half HAFM on the square lattice has been studied over many decades. There are some excellent reviews [1–3] which can be used to get more detailed information on this work. Although till now there is no rigorous proof for the existence of Néel LRO⁷ after intensive studies over many decades it became clear in the late eighties that there is no doubt of semi-classical Néel LRO at absolute zero. The quantum fluctuations lead to a substantial renormalization of the order parameter (sublattice magnetization), which amounts to about 60% of the classical value. Experimentally there are some layered antiferromagnetic inorganic materials like the parent compound La₂CuO₄ for high- T_c super-conductors or Sr₂CuO₂Cl₂ [2, 19, 53] but also organic compounds [54] which are well described by the (quasi-)2D HAFM on the square lattice.

The spin half HAFM on the square lattice can serve as the canonical example for a quantum HAFM on a 2D bipartite lattice. Already about ten years ago Schulz and coworkers [55] published large-scale exact diagonalization studies for the GS of systems up to N = 36. Recently Betts and coworkers [33] have presented a systematic study of a complete set of all finite square lattices up to N = 32. In particular, one finds in [33] a guideline how to find systematically the so-called defining edge vector in finite lattices for arbitrary dimension and type of lattice. We use this scheme to generate the finite lattices discussed below. We have recalculated and extended Schulz' and Betts' results for systems up to N = 40 sites including the results for the low-lying excitations. We have presented some of our results already in Sects. 2.3.2 and 2.3.3.

The classical GS breaks the translational symmetry of the lattice. The magnetic unit cell is twice as large as the geometric one. On a finite bipartite lattice the quantum GS is a rotationally invariant singlet state (Lieb-Mattis theorem [29]). As can be seen from Fig. 2.6a there is one QDJS in each sector S (cf. Sect. 2.3.2) and the translational symmetry of the QDJS alternates between $\mathbf{Q}_0 = (0,0)$ and $\mathbf{Q}_1 = (\pi,\pi)$. Note that \mathbf{Q}_0 and \mathbf{Q}_1 are different in the geometric but they coincide in the magnetic Brillouin zone. The energies of the QDJS are well described by $E_0 + S(S+1)/2N\chi_0$, see (2.10). The family of one-magnon states is well separated from the QDJS. Their energies follow the dispersion relation obtained by spin-wave theory, see Fig. 2.8. The lowest singlet excitation is significantly above the first triplet excitation.

The GS energy, the first excitation and the square of the order parameter for the largest lattices with N = 38 and N = 40 and for comparison for N = 36 are given in Table 2.1. Furthermore, we present for the first time all spin-spin correlations for N = 40 in Table 2.2. It is obvious that the decay of the spin-spin correlations is weak.

Altogether 12 finite square lattices from N = 18 to N = 40 are used for the extrapolation of the GS energy and the order parameter to infinite N(see Figs. 2.20a and 2.21a). We compare our results with some corresponding data obtained by other means:

 $^{^7}$ We mention that the existence of Néel LRO was proven for the HAFM with $S \geq 1~[49,50]$ and for the spin half anisotropic XXZ antiferromagnet [51,52] on the square lattice.



Fig. 2.8. Energy of the one-magnon excitations $\Delta_k = E_0(\mathbf{k}, S = 1) - E_0(S = 0)$ versus \mathbf{k} (dispersion relation). The solid and dashed lines show results of the linear (LSWT) and higher-order spin-wave theory (SWT1) [56], the points are exact data for finite lattices of size N = 16, 18, 32 und 36. The inset in the right part of the figure shows the path through the Brillouin zone

Table 2.1. Ground-state energy E_0 (singlet), singlet-triplet gap (spin gap) $\Delta = E_0(S=1) - E_0$ and square of the order parameter $m^2 \equiv (m^+)^2$ of the HAFM on finite square lattices with N = 38, N = 40 and N = 36 sites (the results for N = 36 are in agreement with those of [55]). (l_{11}, l_{12}) and (l_{21}, l_{22}) are the components of the two edge vectors defining the finite 2D lattice

N	l_{11}	l_{12}	l_{21}	l_{22}	E_0	Δ	m^2
36	6	0	0	6	-24.4393974	0.287538	0.20983715
38	1	7	-5	3	-25.7607925	0.272791	0.20751801
40	2	6	-6	2	-27.0948503	0.261623	0.20361937

Table 2.2. Ground-state spin-spin correlations $\langle S_0^z S_R^z \rangle = \frac{1}{3} \langle \mathbf{S}_0 \mathbf{S}_R \rangle$ for all lattice vectors **R** of the HAFM on a square lattice with N = 40 sites. Note that due to the reduced symmetry of the finite lattice we have slightly different values for lattice vectors $\mathbf{R} = (3, 1)$ and (3, -1) as well as for $\mathbf{R} = (1, 2)$ and (1, -2)

\mathbf{R}	(0, 0)	(0, 1)	(1, 1)	(0, 2)	(1, -2)	(1, 2)
$\langle S_0^z S_R^z \rangle$	0.250000	-0.112895	0.069066	0.061711	-0.059679	-0.059055
R	(2, 2)	(0, 3)	(3, 1)	(3, -1)	(3, -2)	(4, -2)
$\langle S_0^z S_R^z \rangle$	0.053826	-0.055663	0.054700	0.052344	-0.052074	0.050275

- GS energy per bond: our result: E_0 /bond = -0.3350; high-order SWT [56]: E_0 /bond = $-s^2 0.157948s 0.006237 + 0.0000108/s = -0.335233;$ QMC [47]: E_0 /bond = -0.334719; coupled cluster method (CCM) [57]: E_0 /bond = -0.3349; series expansion [58]: E_0 /bond = -0.3347; previous exact diagonalization up to N = 32 [33]: E_0 /bond = -0.33404;
- order parameter (sublattice magnetization): our result: $m^+ = 0.3173 \sim 0.635 \ m^+_{class}$; high-order SWT [56]: $m^+ = s 0.1966019 + 0.0000866(25)/s^2 = 0.3037$; QMC [47]: $m^+ = 0.3070$; CCM [57]: $m^+ = 0.31$; series expansion [58]: $m^+ = 0.307$; previous exact diagonalization up to N = 32 [33]: $m^+ = 0.30676$

(for the extrapolation of the gap, see Sect. 2.3.3). A more detailed collection of results for the sublattice magnetization and the GS energy obtained by different methods can be found in [33,57].

The existence of Néel LRO for the square lattice does not automatically imply the conclusion, that all other bipartite lattices are also Néel longrange ordered. Stronger quantum fluctuations can appear in lattices with coordination number z < 4 and in lattices with non-equivalent NN bonds.

The Honeycomb Lattice (T3)

For this lattice the geometric and the magnetic (Néel state) unit cell are identical and include two sites. All NN bonds are equivalent but the coordination number z = 3 is less than in the square lattice giving rise to stronger quantum fluctuations. Nevertheless there is a lot of evidence obtained by several methods [27,42,59–62] and also from the data presented below, that the GS is a semi-classical Néel state.

The low-energy spectrum is shown in Fig. 2.9. The QDJS are well separated from the other states and follow (2.10). Due to the coincidence of geometric and magnetic unit cell they all have translational symmetry vector $\mathbf{Q} = (0, 0)$. There are no low-lying singlets within the spin gap. The similarity between the spectra of the square and the honeycomb lattice is obvious.

The largest lattice considered has N = 38 sites and is defined by the edge vectors (3, 2); (-2, 5) and has GS energy E_0 /bond = -0.366768, spin gap $\Delta = 0.213953$ and square of the order parameter $(m^+)^2 = 0.184396$.

For the finite-size extrapolation of the GS energy (Fig. 2.20a), the spin gap and the order parameter (Fig. 2.21a) we have used 14 finite lattices from N = 6 up to N = 38. The extrapolation according to formulae (2.12), (2.13), (2.14) leads to the following results:

- GS energy per bond: E_0 /bond = -0.3632(for comparison: QMC [59]: E_0 /bond = -0.3630; 2nd order SWT [60]: E_0 /bond = $-s^2 - 0.209842s - 0.0110084 = -0.365929$; series expansion [61]: E_0 /bond = -0.3629; CCM [27]: E_0 /bond = -0.3631);
- spin gap: $\Delta = 0.0504$ (for comparison: CCM [27]: $\Delta = 0.02$);



Fig. 2.9. Low-energy spectrum for the HAFM on the honeycomb lattice with N = 32 sites (the inset shows the **k** points in the Brillouin zone)

• order parameter: $m^+ = 0.2788 \sim 0.558 \ m_{class}^+$ (for comparison: QMC [59]: $m^+ = 0.235$; 2nd order SWT [60]: $m^+ = 0.2418$; series expansion [61]: $m^+ = 0.266$; CCM [27]: $m^+ = 0.28$).

Obviously due to the lower coordination number the magnetization is approximately 10% smaller than for the square lattice, but the existence of Néel LRO is not in question.

The CaVO (T11) and the SHD (T10) Lattices

Both these lattices have non-equivalent NN bonds and low coordination number z = 3 leading to strong quantum fluctuations. The lattice T11 has attracted much attention since 1995 when in susceptibility measurements on CaV₄O₉ [4] for the first time a rotationally invariant quantum paramagnetic GS with a finite spin gap of $\Delta \approx 110$ K was discovered experimentally for a quasi-2D antiferromagnetic spin half system. The underlying lattice of CaV₄O₉ is a 1/5 site-depleted square lattice which can be transformed by an appropriate distortion to the Archimedean lattice T11 (see Fig. 2.10). We use therefore the name 'CaVO' to denote this lattice. The experimental findings stimulated a series of theoretical studies for the spin half HAFM on the CaVO lattice [48, 63–77]. The geometric unit cell of the CaVO lattice contains four sites. However, the translational symmetry of the lattice and of the classical Néel GS do not fit to each other and consequently the magnetic unit cell must



Fig. 2.10. Arrangement of the V^{4+} atoms (points) in the V-O layers of CaV₄O₉ (left) and the corresponding Archimedean tiling T11 (right)



Fig. 2.11. Low-energy spectrum for HAFM on the CaVO (T11) and on the SHD (T10) lattice (the insets show the **k** points in the Brillouin zone). a: CaVO with N = 32. b: SHD with N = 36.

be chosen as twice as large as the geometric one. This makes the symmetry of the QDJS similar to that of the square lattice (see Figs. 2.11a and 2.6a).

The Archimedean lattice T10 is built by regular squares, hexagons and dodecagons (SHD) and is therefore more complex than the CaVO lattice. As far as we know till now no antiferromagnetic material was synthesized having the lattice structure of tiling 10. The low coordination number, the quantum competition of non-equivalent NN bonds and the complex structure of the lattice have stimulated the search for a possible non-Néel ordered GS for this lattice [43, 78]. The geometric unit cell of the SHD lattice contains twelve sites. The translational symmetry of the lattice and of the classical Néel GS fit to each other leading to identical magnetic and geometric unit cell. Hence the symmetry of the QDJS is similar to that of the honeycomb lattice (see Figs. 2.11b and 2.9). The similarities between the spectra of the square and the CaVO lattice as well as the SHD and the honeycomb lattice are obvious: The QDJS are well separated from the other states and follow (2.10). There are no low-lying singlets within the spin gap.

Since the magnetic unit cell contains 8 sites, the largest CaVO lattice we consider has N = 32 sites and is defined by the edge vectors (2, -2); (2, 2). It has GS energy per bond E_0 /bond = -0.372903, spin gap $\Delta = 0.281788$ and square of the order parameter $(m^+)^2 = 0.178018$. The two non-equivalent NN correlations functions for N = 32 are $\langle \mathbf{S}_i \mathbf{S}_j \rangle_J = -0.311103$ for J bonds belonging to squares and $\langle \mathbf{S}_i \mathbf{S}_j \rangle_{J'} = -0.403803$ for J' dimer bonds (cf. Fig. 2.10).

The largest SHD lattice considered has N = 36 sites and is defined by the edge vectors (2, 1); (-1, 1). It has GS energy per bond E_0 /bond = -0.373118, spin gap $\Delta = 0.270929$ and square of the order parameter $(m^+)^2 = 0.163243$. The three non-equivalent NN correlations functions for N = 36 are $\langle \mathbf{S}_i \mathbf{S}_j \rangle_{SH} = -0.414324$ for NN bonds belonging to squares and hexagons, $\langle \mathbf{S}_i \mathbf{S}_j \rangle_{SD} = -0.395046$ for NN bonds belonging to squares and dodecagons and $\langle \mathbf{S}_i \mathbf{S}_j \rangle_{HD} = -0.309984$ for NN bonds belonging to hexagons and dodecagons.

The finite-size extrapolation for the CaVO and even more for the SHD lattice suffers from the restriction to a small number of unit cells in the accessible finite lattices. Hence the extrapolation is particularly uncertain and should be taken with extra care. For the finite-size extrapolation of the GS energy (Fig. 2.20a), and the order parameter (Fig. 2.21a) according to formulae (2.12), (2.13), (2.14) we use finite lattices of N = 16, 24, 32 (CaVO) and N = 12, 24, 36 (SHD). The extrapolation leads to the following results for the CaVO lattice:

- GS energy per bond: E_0 /bond = -0.3689(for comparison: linear SWT [64]: E_0 /bond = -0.3584);
- spin gap: $\Delta = 0.1149$ (for comparison: QMC [48]: $\Delta \sim 0$);
- order parameter: $m^+ = 0.2303 \sim 0.461 \ m_{class}^+$ (for comparison: linear SWT [64]: $m^+ = 0.212$; QMC [48]: $m^+ = 0.178$)

and for the SHD lattice:

- GS energy per bond: E_0 /bond = -0.3713(for comparison: variational (Huse-Elser) [78]: E_0 /bond = -0.3605; variational (resonating valence bond (RVB)) [43]: E_0 /bond = -0.3688);
- spin gap: $\Delta = 0.1435;$

• order parameter: $m^+ = 0.2126 \sim 0.425 \ m^+_{class}$ (for comparison: variational (RVB) [43]: $m^+ = 0.2546$).

Due to the competition between the bonds the order parameters for the CaVO and the SHD lattice are smaller than for the honeycomb lattice. Nevertheless we find convincing evidence that the GS is semi-classically Néel ordered. This conclusion is well supported by other methods [43, 48, 64, 66, 67, 72, 78]. However, the quantum competition between non-equivalent bonds leads to a tendency to form local singlets either on neighboring bonds or along polygons. In connection with the observed rotationally invariant quantum paramagnetic GS with a finite spin gap in CaV₄O₉ for the CaVO lattice a J - J'-HAFM with different strengths of NN bonds J and J' is considered, where J is the NN bond belonging to a square and J' is the NN bond not belonging to a dimer (Fig. 2.10). Within this model a quantum paramagnetic rotationally invariant singlet phase with gapped excitations is obtained. We will discuss this J - J'- HAFM and its quantum phase transition in more detail in Sect. 2.5.

2.4.2 Semi-classical LRO on Frustrated Lattices

The classical GS for non-bipartite frustrated lattices may be collinear (weak frustration) or non-collinear (strong frustration) and depends on the special features of the lattice. The frustration may enhance the effect of quantum fluctuations so that the magnetic order may be stronger weakened than for the bipartite lattices. Thus the frustrated HAFM on 2D lattices is an interesting candidate for a magnetic system with a quantum paramagnetic GS.

The Triangular Lattice (T1)

The triangular lattice is strongly frustrated but has largest coordination number z = 6 (see Fig. 2.2). Already in the 70ties Anderson and Fazekas [8,9] considered the spin half HAFM on the triangular lattice. They argued that the GS for the 2D triangular lattice might be similar to that for the 1D HAFM and proposed a spin-liquid like rotationally invariant resonating valence bond GS instead of a semi-classical Néel state. Starting in the late eighties several authors found, however, more and more evidence for a Néel ordered GS (see e.g. [31, 36, 37, 79–86]).

The classical GS is a three-sublattice Néel state with an angle of 120° between the spins of different sublattices (Fig. 2.12a). It breaks the translational symmetry of the lattice. The energy per bond is $E_0^{\text{class}}/\text{bond} = -s^2/2 = -0.125$ and the order parameter is $m_{\text{class}}^+ = \frac{1}{2}\sqrt{2/3} = 0.40825$.

The magnetic unit cell is three times as large as the geometric one and thus the QDJS belong to vectors $\mathbf{Q}_1 = (0,0)$, $\mathbf{Q}_2^+ = (+4\pi/3,0)$ and $\mathbf{Q}_2^- = (-4\pi/3,0)$. Low-lying states have been tabulated in [37], however for



Fig. 2.12. The HAFM on the triangular lattice (T1). a: Classical GS. b: Lowenergy spectrum for N = 36 (the inset shows the **k** points in the Brillouin zone)

N = 36 only in the sector $\mathbf{Q}_1 = (0, 0)$. Fig. 2.12b shows our results for the low-lying states on the N = 36 lattice. Apparently, the QDJS are well separated from the other states and follow (2.10) for $S \leq 4$. The lowest singlet excitation energy is above the first triplet excitation. A special feature of the E(S) behavior of the QDJS is a deviation from the linearity starting in the vicinity of S = N/6. This comes from the Ising part of the Hamiltonian and is connected with distinguished Ising states having two spins up and one spin down per triangle [87,88] and results in a plateau in the magnetization versus external magnetic field curve (for a more detailed discussion, see Sect. 2.6). However, this peculiarity emerging around S(S + 1) = 42 in Fig. 2.12b is relevant only if $\sqrt{N} \leq N/6$, i.e. for small N.

The largest lattice considered has N = 36 sites and is defined by the edge vectors (6,0); (0,6). It has GS energy per bond E_0 /bond = -0.186791, spin gap $\Delta = 0.344211$ and square of the order parameter $(m^+)^2 = 0.124802$ (cf. [37]). A detailed discussion of the spectra can be found in [31,36,37].

For the finite-size extrapolation of the GS energy (Fig. 2.20b), the spin gap and the order parameter (Fig. 2.21b) we use only even finite lattices of size N = 24, 30, 36. The extrapolation according to formulae (2.12), (2.13), (2.14) leads to the following results:

• GS energy per bond: E_0 /bond = -0.1842 (for comparison: RVB [8]: E_0 /bond = -0.154; SWT [81]: E_0 /bond = -0.1823; former exact diagonalization [37]: E_0 /bond = -0.1815, Green's function Monte Carlo [84]: E_0 /bond = -0.1819; CCM [86]: E_0 /bond = -0.1835);

- spin gap: $\Delta = 0.1293;$
- order parameter: $m^+ = 0.1577 \sim 0.386 \ m_{class}^+$ (for comparison: sublattice magnetization $m^{sl} = \langle S_i^z \rangle$ in SWT [81]: $m^{sl} = 0.2387 = 0.4474 \ m_{class}^{sl}$; Green's function Monte Carlo [84]: $m^{sl} = 0.205 = 0.41 \ m_{class}^{sl}$; CCM [86]: $m^{sl} = 0.2107 = 0.4214 \ m_{class}^{sl}$).

Obviously, the extrapolated gap is quite large, whereas the order parameter is smaller than that obtained by other means. This suggests stronger finite-size effects than for bipartite lattices. Nevertheless, the existence of semi-classical Néel LRO is not in question.

The Maple-Leaf (T4) and the Bounce (T7) Lattices

The maple-leaf lattice [89] is obtained from the triangular lattice by a 1/7 depletion of sites. Its geometric unit cell contains six sites and the underlying Bravais lattice is a triangular one (cf. Fig. 2.13). It is also strongly frustrated but has lower coordination number (z = 5) than the triangular lattice. Furthermore, it has three non-equivalent NN bonds (solid, dashed and dotted lines in Fig. 2.5). Thus the quantum fluctuations might be more important



Fig. 2.13. The classical GS of the HAFM on the maple-leaf lattice (T4). The geometric unit cell is shown with dashed lines. The magnetic unit cell contains three geometric unit cells labeled by A,B,C

and the HAFM on the maple-leaf lattice was considered as a candidate for a quantum paramagnet [90].

The bounce lattice is related to the maple-leaf lattice. It can be obtained from the maple-leaf lattice by bond depletion as described in Sect. 2.2.2 (see Fig. 2.5). It has also a geometric unit cell with 6 sites, an underlying triangular Bravais lattice and contains two non-equivalent NN bonds. The coordination number z = 4 is lower than for the maple-leaf lattice but it is less frustrated, since the omitted bond was a frustrating one. As far as we know no antiferromagnetic material has, as yet, been synthesized with the lattice structure of tilings 4 or 7.

The classical GS of the maple-leaf lattice is a six-sublattice Néel state shown in Fig. 2.13 with $E_0^{\text{class}}/\text{bond} = -s^2(\sqrt{3}+1)/5 = -0.13660$ and $m_{\text{class}}^+ = 0.39434$. The less frustrated bounce lattice has also a six-sublattice Néel GS with a 120° structure on each triangle and a collinear up-down structure on each hexagon leading to $E_0^{\text{class}}/\text{bond} = -2s^2/3 = -0.16667$ and $m_{\text{class}}^+ = 1/\sqrt{6} = 0.40825$. Both classical GSs break the translational symmetry of the lattice, the corresponding magnetic unit cell is three times as large as the geometric one and contains 18 sites. Therefore the applicability of finite-size calculations is particularly limited.

The low-lying spectra for both lattices with N = 36 sites are shown in Fig. 2.14. The lowest states in each sector of S are QDJS belonging to appropriate symmetries $\mathbf{Q}_1 = (0,0)$ and $\mathbf{Q}_2^+ = (+4\pi/3,0)$, $\mathbf{Q}_2^- = (-4\pi/3,0)$. They follow (2.10). The lowest singlet excitation energy is above the first triplet excitation.

The largest finite lattices considered have N = 36 sites and are defined by the edge vectors (3, 0); (1, 2) for both tilings. Note that these finite lattices do not have the full symmetry of the corresponding infinite lattices. The N = 36maple-leaf lattice has GS energy per bond $E_0/\text{bond} = -0.215589$, spin gap $\Delta = 0.400009$ and square of the order parameter $(m^+)^2 = 0.106101$. A picture of this lattice and a table of the correlation functions are given in [90]. The non-equivalent NN bonds lead to different NN correlations: $\langle \mathbf{S}_i \mathbf{S}_j \rangle_T =$ -0.1777 (belonging to a dashed line in Fig. 2.5, middle), $\langle \mathbf{S}_i \mathbf{S}_j \rangle_H = -0.3656$ (belonging to a solid line in Fig. 2.5, middle) and $\langle \mathbf{S}_i \mathbf{S}_j \rangle_D = 0.0086$ (belonging to a dotted line in Fig. 2.5, middle).⁸ It appears that the correlation functions of the quantum system fit quite well to the classical GS.

The N = 36 bounce lattice has GS energy per bond E_0 /bond = -0.286540, spin gap $\Delta = 0.445138$ and square of the order parameter $(m^+)^2 = 0.119073$. The non-equivalent NN bonds lead to different NN correlations: $\langle \mathbf{S}_i \mathbf{S}_j \rangle_T = -0.1723$ (belonging to a dashed line in Fig. 2.5 right) and $\langle \mathbf{S}_i \mathbf{S}_j \rangle_H = -0.4008$ (belonging to a solid line in Fig. 2.5 right). The

⁸ Note that these values and the corresponding values for the bounce lattice are averaged values, since the N = 36 lattices do not have all lattice symmetries of the infinite lattice. As a result one has to average over three different values for a certain correlation function.



Fig. 2.14. Low-energy spectrum for HAFM on the maple-leaf (T4) and on the bounce (T7) lattice (the insets show the **k** points in the Brillouin zone). a: maple-leaf lattice with N = 36. b: bounce lattice with N = 36

correlation function of the omitted bond (see Fig. 2.5) is $\langle \mathbf{S}_i \mathbf{S}_j \rangle_D = 0.1116$. It is obvious that the NN correlations $\langle \mathbf{S}_i \mathbf{S}_j \rangle_D$ and $\langle \mathbf{S}_i \mathbf{S}_j \rangle_H$ are enhanced by omitting the frustrating bond, whereas $\langle \mathbf{S}_i \mathbf{S}_j \rangle_T$ remains almost the same.

We use finite maple-leaf lattices and bounce lattices of size N = 18 and 36 for the finite-size extrapolation of the GS energy (Fig. 2.20b), the spin gap and the order parameter (Fig. 2.21b). By using formula (2.12), (2.13) and (2.14) we obtain for the maple-leaf lattice:

- GS energy per bond: E_0 /bond = -0.2171(for comparison: SWT [90]: E_0 /bond = -0.20486; variational [90]: E_0 / bond = -0.1988);
- spin gap: $\Delta = 0.2548;$
- order parameter: $m^+ = 0.0860 \sim 0.218 \ m_{class}^+$ (for comparison: sublattice magnetization $m^{sl} = \langle S_i^z \rangle$ in SWT [90]: $m^{sl} = 0.154 = 0.308 \ m_{class}^{sl}$).

An extrapolation of the gap based on a variational approach was presented in [90] and leads to $\Delta = 0.0180$.

The corresponding extrapolation for the bounce lattice yields:

- GS energy per bond: E_0 /bond = -0.2837;
- spin gap: $\Delta = 0.2926;$
- order parameter: $m^+ = 0.1095 \sim 0.268 \ m_{class}^+$.

Obviously, the extrapolated order parameters are small but finite. The fact that the order parameter for the bounce lattice is larger than for the mapleleaf lattice seems to be related to the lower frustration. Taking into consideration results of the spin-wave theory and the variational approach presented for the maple-leaf lattice in [90] we conclude that the semi-classical six-sublattice Néel LRO survives for both lattices. However, this statement needs confirmation by further studies.

The Trellis Lattice (T5)

The trellis lattice is to some extent exceptional since its structure corresponds to a system of coupled ladders or alternatively of coupled zigzag chains. Its geometric unit cell contains 2 sites (cf. Fig. 2.15a). It has the same coordination number z = 5 as the maple-leaf lattice but its frustration is slightly smaller (cf. Fig. 2.2). Furthermore, it has three non-equivalent NN bonds, labeled by J_1 , J_2 and J_3 in Fig. 2.15a.

The HAFM on the trellis lattice is related to the magnetism of $SrCu_2O_3$, CaV_2O_5 and MgV_2O_5 [91, 92]. However, the J_1 , J_2 and J_3 bonds are not of equal strength in these materials (for instance in $SrCu_2O_3$ the zigzag J_1



Fig. 2.15. The trellis lattice (T5). a: Illustration of the lattice with basis vectors \mathbf{b}_1 and \mathbf{b}_2 , geometric unit cell (dashed) and the non-equivalent NN bonds J_1 , J_2 and J_3 . In the classical GS the spins form a spiral along the zigzag chains (J_1 , J_2 bonds) whereas the spins along a J_3 bond are antiparallel. b: Low-energy spectrum for N = 28 (the inset shows the **k** points in the Brillouin zone)

coupling is weak leading to a quasi-1D ladder structure). The classical GS is a Néel state for $J_2 < J_1/4$, and is an incommensurate spiral state along the zigzag chains (x-direction) for $J_2 > J_1/4$, where the angles between neighboring bonds are $\alpha_2 = 2 \arccos(J_1/4J_2)$ (J_2 bond), $\alpha_1 = \pi + \alpha_2/2$ (J_1 bond) and $\alpha_3 = \pi$ (J_3 bond). This leads to a classical GS with pitch angles $\alpha_1 = \pi + \arccos(1/4) = 1.41957\pi$; $\alpha_2 = 2 \arccos(1/4) = 0.83914\pi$, GS energy per bond E_0^{class} /bond = $-0.65s^2 = -0.1625$ and $m_{\text{class}}^+ = 0.39894$ for the perfect lattice ($J_1 = J_2 = J_3$).

The incommensurability of the classical GS creates additional difficulties applying exact diagonalization for finite lattices since the classical pitch angles α_1 and α_2 may be in conflict with periodic boundary conditions. In order to minimize this boundary effect we consider only finite lattices of N = 20, 28 and 36 sites having pitch angles $\alpha_2^{(N)}$ deviating by not more than 6% from the true values α_2 . The N = 36 lattice is defined by the edge vectors (9,0); (-1,2) and has a pitch angle $\alpha_2^{(36)} = 1.059\alpha_2$. Its GS energy per bond is E_0 /bond = -0.247578, spin gap $\Delta = 0.605227$ and square of the order parameter $(m^+)^2 = 0.109897$. The three non-equivalent NN correlations functions for N = 36 are $\langle \mathbf{S}_i \mathbf{S}_j \rangle_{J_1} = -0.098835, \langle \mathbf{S}_i \mathbf{S}_j \rangle_{J_2} = -0.283938$ and $\langle \mathbf{S}_i \mathbf{S}_j \rangle_{J_3} = -0.472341$ (cf. Fig. 2.15a).

In Fig. 2.15b the QDJS are shown. Although the boundary conditions are not perfect it can be seen that the QDJS are separated from the other states and follow approximately (2.10). The lowest singlet excitation is above the first triplet excitation. The translational symmetry of the QDJS is more complex than in the other lattices. It is connected with the **q** vector of the spiral state. We find $Q_y = 0$, π and $Q_x = 6\pi (N/2 - S)/7 \mod 2\pi$ for N = 28and $Q_x = 8\pi (N/2 - S)/9 \mod 2\pi$ for N = 36.

For the finite-size extrapolation of the GS energy (Fig. 2.20b), the spin gap and the order parameter (Fig. 2.21b) we use finite lattices of size N = 20, 28, 36. The extrapolation according to formulae (2.12), (2.13), (2.14) leads to the following results:

- GS energy per bond: E_0 /bond = -0.2471;
- spin gap: $\Delta = 0.49;$
- order parameter: $m^+ = 0.0885 \sim 0.222 \ m_{class}^+$.

Although our data do not allow a secure conclusion the results are in favor of a spiral long-range ordered phase. This conclusion is in agreement with the findings in [91] based on a Schwinger boson technique and linear spin-wave theory.

The SrCuBO Lattice (T6)

The SrCuBO lattice is weakly frustrated, has four sites in the geometric unit cell and two non-equivalent NN bonds J_1 and J_2 (see Fig. 2.16a, top). It can be transformed by an appropriate distortion to a square lattice with one



Fig. 2.16. The SrCuBO lattice (T6). a: Comparison of the SrCuBO lattice (above) and the Shastry-Sutherland model (below). The unit cell is illustrated by the dashed square. b: Low-energy spectrum for N = 32 (the inset shows the **k** points in the Brillouin zone)

diagonal bond in each second square (see Fig. 2.16a, bottom). This frustrated square lattice is known as Shastry-Sutherland model [93,94] introduced in the 80ties as a 2D spin half HAFM with an exactly known quantum GS. Indeed for large frustrating J_2 the GS is a so-called orthogonal dimer product state with dimer singlets on each J_2 bond. Although the Shastry-Sutherland model initially was understood as a 'toy model' it has attracted much renewed attention as it provides a representation of the magnetic properties of the recently discovered 2D spin gap system $SrCu_2(BO_3)_2$ [5,95]. The experimental findings stimulated a series of theoretical studies for the spin half HAFM on the SrCuBO lattice with varying bonds J_1 , J_2 , see [96–104] and the recent review [105]. We will discuss the GS phase diagram in the $J_1 - J_2$ plane below in Sect. 2.5. In this section we consider $J_1 = J_2$, only. In this case the classical GS is the two-sublattice Néel state with energy per bond $E_0^{\text{class}}/\text{bond} = -0.6s^2 = -0.15$ and with order parameter $m_{\text{class}}^+ = 0.5$. The geometric unit cell of the SrCuBO lattice contains four sites and the translational symmetry of the lattice and of the classical Néel GS fit to each other. The spectrum of the SrCuBO lattice (Fig. 2.16b) is therefore comparable with that of the honeycomb lattice (Fig. 2.9). The QDJS are well separated from the other states and follow (2.10). The lowest singlet excitation is above the first triplet.

The largest lattice considered with N = 36 sites is defined by the edge vectors (3,0); (0,3) and has GS energy per bond E_0 /bond = -0.233410, spin gap $\Delta = 0.319735$ and square of the order parameter $(m^+)^2 = 0.169048$, that is 80% of the order parameter of the corresponding square lattice. The two nonequivalent NN correlations functions for N = 36 are $\langle \mathbf{S}_i \mathbf{S}_j \rangle_{J_1} = -0.332886$ (almost the same as for the square lattice) and $\langle \mathbf{S}_i \mathbf{S}_j \rangle_{J_2} = 0.164493$.

For the finite-size extrapolation of the GS energy (Fig. 2.20b), the spin gap and the order parameter (Fig. 2.21b) we use finite lattices of size N = 20, 32 and 36. The extrapolation according to formulae (2.12), (2.13), (2.14) leads to the following results:

- GS energy per bond: E_0 /bond = -0.2310(for comparison: series expansion [96]: E_0 /bond = -0.231; Schwinger boson mean field [94]: E_0 /bond = -0.231; CCM [106]: E_0 /bond = 0.2311);
- spin gap: $\Delta = 0.0927;$
- order parameter: $m^+ = 0.2280 \sim 0.456 \ m_{class}^+$ (for comparison: series expansion [96]: $m^+ = 0.200$; Schwinger boson mean field [94]: $m^+ = 0.203$; CCM [106]: $m^+ = 0.211$).

Due to frustration the order parameter is only about 70% of that of the square lattice but it is the largest one of all frustrated lattices. There is no doubt of semi-classical GS Néel order for this lattice. This conclusion is in agreement with several other studies like series expansion [96, 97, 99] and bosonic representations [94, 104]. However, the Néel LRO is destroyed by further increasing the frustrating bond J_2 (see Sect. 2.5).

2.4.3 Absence of Semi-classical LRO on Frustrated Lattices – The Kagomé (T8) and the Star (T9) Lattices

Among the non-bipartite frustrated lattices the kagomé⁹ and the star lattice play an exceptional role. The kagomé lattice is strongest frustrated (as strong as the triangular lattice) and has low coordination number z = 4, cf. Fig. 2.17. It can be obtained by a 1/4 site depletion or alternatively by a 1/3 bond depletion (with an appropriate subsequent distortion) of the triangular lattice. Whereas the triangles in the kagomé lattice are corner sharing, they are separated by a dimer in the star lattice. Its degree of frustration is less than for the kagomé lattice but it has an even lower coordination number z = 3 and two non-equivalent NN bonds J_D and J_T , cf. Fig. 2.18. As indicated in Fig. 2.3, the star lattice can be obtained by a 2/5 bond depletion of the maple-leaf (T4) or alternatively by a 1/4 bond depletion of the bounce lattice (T7) with an appropriate subsequent distortion. Both the kagomé and the star lattices are characterized by strong quantum fluctuations.

After realizing in the early nineties that the quantum GS of the HAFM on the triangular lattice is Néel ordered the HAFM on the kagomé lattice came

⁹ The name kagomé stems from the Japanese language and means a bamboo-basket woven pattern [107].



Fig. 2.17. Two variants of the GS of the classical HAFM on the kagomé lattice (T8): the $\sqrt{3} \times \sqrt{3}$ state (left) and the q=0 state (right). The dotted ellipses show further degrees of freedom of the highly degenerate classical GS. The Wigner-Seitz geometrical unit cell contains three sites (A1, A2, A3). The light and gray triangles symbolize different chiralities



Fig. 2.18. Two variants of the GS of the classical HAFM on the star lattice (T9): the $\sqrt{3} \times \sqrt{3}$ state (left) and the q=0 state (right). The dotted ellipses show further degrees of freedom of the highly degenerate classical GS. The Wigner-Seitz geometrical unit cell consists of six sites located on a hexagon (sites A1,..., A6). The light and gray triangles symbolize different chiralities

into the focus of interest as a hot candidate for a 2D quantum spin system with an exotic non-Néel ordered GS [36, 38, 39, 86, 108–120]. Indeed, most of the recent investigations are in favor of a quantum paramagnetic GS, although its nature is far from being well understood. A possible physical realization of the kagomé HAFM is SrCrGa oxide, which is, however, a layered kagomé HAFM with spin 3/2 [121,122]. A novel spin-1/2 kagomé like HAFM has been found recently in volborthite $Cu_3V_2O_7(OH)_2 \cdot 2H_2O$ [123]. By contrast, the spin half HAFM on the star lattice has not been considered in the literature so far, nor is a physical realization currently known. However, we mention that a projection of the three-dimensional non-frustrated magnetic compound green dioptase $Cu_6Si_6O_{18} \cdot 6H_2O$ has the shape of the star lattice [124].

The geometric unit cell of the kagomé (star) lattice contains three (six) sites and the underlying Bravais lattice is a triangular one (cf. Figs. 2.17 and 2.18). The classical GS for the kagomé lattice was studied in [125–127]. In analogy to the triangular lattice the angle between neighboring spins is 120° . Its energy per bond is $E_0^{\text{class}}/\text{bond} = -s^2/2 = -0.125$. However, in contrast to the triangular lattice there is a non-trivial infinite degeneracy of the classical GS typical for a classical HAFM with corner-sharing triangles.

In the classical GS of the star lattice the two non-equivalent NN bonds carry different NN spin correlations: the angle between neighboring spins on dimer bonds J_D is 180°, whereas the angle on triangular bonds J_T is 120°. Its energy per bond is $E_0^{\text{class}}/\text{bond} = -2s^2/3 = -0.1667$. Although the star lattice is not built by corner-sharing triangles, the classical GS for this lattice also exhibits a non-trivial infinite degeneracy very similar to that of the kagomé lattice.

Two particular variants of the classical GS characterized by a certain wave vector are shown in Figs. 2.17 and 2.18. The states on the left side of Figs. 2.17 and 2.18 exhibit the same symmetry as the classical GS for the triangular lattice having a magnetic unit cell three times as large as the geometric unit cell (so-called $\sqrt{3} \times \sqrt{3}$ state). The states on the right side of Figs. 2.17 and 2.18 have the same translational symmetry as the lattice (socalled q = 0 state) and therefore the magnetic and the geometric unit cell are identical. Both states are highly degenerate as indicated by the dotted elliptic lines at the top of spins.

Let us consider the order parameter (2.9) for the classical GS. If we take the perfect ordered planar $\sqrt{3} \times \sqrt{3}$ and q=0 state, then we get for both lattices $m^+_{\text{class},\sqrt{3} \times \sqrt{3}} = m^+_{\text{class},q=0} = \frac{1}{2}\sqrt{2/3} = 0.40825$. However, one has to take into account the high degeneracy of the GS. In order to average over these degenerate states we performed numerical calculations of the ground states for classical systems of up to N = 432 sites. The numerical results lead to the conclusion that for large N we have $m^+_{\text{class,averaged}} = 0.25$ for both lattices. This corresponds to a GS phase with decoupled spins for larger spin-spin separations.

In the quantum case the largest kagomé lattice considered has N = 36sites and is defined by the edge vectors (4, 2); (2, 4). It has GS energy per bond E_0 /bond = -0.219188, spin gap $\Delta = 0.164190$ and square of the order parameter $(m^+)^2 = 0.076630$. We mention that the result for E_0 /bond was already given in [39, 111].



Fig. 2.19. Low-energy spectrum for HAFM on the kagomé (T8) and on the star (T9) lattice (the insets show the **k** points in the Brillouin zone). a: kagomé with N = 36. b: star with N = 30

The largest star lattice considered has N = 36 sites and is defined by the edge vectors (2,0); (1,3). It has GS energy per bond E_0 /bond = -0.310348, spin gap $\Delta = 0.480343$ and square of the order parameter $(m^+)^2 = 0.082299$. Note that the value of the spin gap is particularly large. The only N = 36 lattice having a larger spin gap is the trellis lattice for which, however, the large spin-gap is most likely a finite-size artifact due to the incommensurate structure of the states. The two non-equivalent NN correlation functions for N = 36 are $\langle \mathbf{S}_i \mathbf{S}_j \rangle_{J_T} = -0.170339$ (that is weaker than for the kagomé and the triangular lattice) and $\langle \mathbf{S}_i \mathbf{S}_j \rangle_{J_D} = -0.590367$.¹⁰ We mention that the NN correlation $\langle \mathbf{S}_i \mathbf{S}_j \rangle_{J_D}$ is the strongest correlation we found in all lattices, thus indicating a strong tendency to form local singlets on the J_D bonds.

The spectra of both lattices are shown Fig. 2.19. For both spectra it is obvious that the lowest states $E_{\min}(S)$ are not well described by (2.10). In particular, the lowest states belonging to S = 0 and S = 1 deviate significantly from a straight line. We do not see well separated QDJS as well as spin-wave excitations. Furthermore, the symmetries of the lowest states in each sector of S cannot be attributed to the classical $\sqrt{3} \times \sqrt{3}$ or q=0 ground states in general. The kagomé lattice is an exceptional case in that a large number of non-magnetic singlets fill the singlet-triplet gap. For instance for

¹⁰ Note that these values are averaged values, since the N = 36 star lattice does not have all lattice symmetries of the infinite lattice. As a result one has to average over two different values.

N = 27 there are 153 [39] and for N = 36 on finds 210 [11] non-magnetic excitations within the spin gap and in the thermodynamic limit possibly a gapless singlet continuum. This unusual number of low-lying singlets is attributed to the non-trivial huge degeneracy of the classical GS. By contrast, the star lattice does not show low-lying singlets. This can be attributed to the special property of the quantum GS to form local singlets on the J_D bonds which somehow makes the singlet GS of the star lattice exceptional. As a consequence, the quantum GS of the star lattice has lowest energy per bond among all frustrated lattices and is well separated from the excitations. Especially the first singlet excitation has comparably high energy.

We mention that a detailed discussion of the spectrum for the kagomé lattice was given in [38,39].

For the finite-size extrapolation of the GS energy (Fig. 2.20b), the gap and the order parameter (Fig. 2.21b) we use finite lattices of size N = 12, 18, 24, 30 and 36 (kagomé) and of N = 18, 24, 30 and 36 (star). The extrapolation leads to the following results for the kagomé lattice:

- GS energy per bond: E_0 /bond = -0.2172 (for comparison: SWT [109]: E_0 /bond = -0.2353; former exact diagonalization (N = 9, ..., 21) [108]: E_0 /bond = -0.217; CCM [86]: E_0 /bond = -0.2126; Green's function decoupling [115, 120]: E_0 /bond = -0.215);
- spin gap: $\Delta = 0.0397;$
- order parameter: $m^+ = 0.000 \sim 0.0 \ m_{\text{class}}^+$.



Fig. 2.20. Finite-size extrapolations of GS energy per bond E_0 /bond, a: bipartite lattices, b: frustrated lattices



Fig. 2.21. Finite-size extrapolations of m^+ , a: bipartite lattices, b: frustrated lattices

In fact, the extrapolation gives the unphysical value $m^+ = -0.0146 < 0$ (cf. Fig. 2.21b). We interpret this as vanishing order parameter.

For the star lattice we obtain:

- GS energy per bond: E_0 /bond = -0.3093;
- spin gap: $\Delta = 0.3809;$
- order parameter: $m^+ = 0.0385 \sim 0.094...0.150 m^+_{class}$ (the first value corresponds to $m^+_{class} = 0.40825$ of the perfect ordered planar $\sqrt{3} \times \sqrt{3}$ and q=0 classical GS, see Fig. 2.18, whereas the second value corresponds to $m^+_{class,averaged} = 0.25$ obtained by averaging over all degenerate classical ground states).

The extrapolated spin gap for the kagomé lattice is small but finite and corresponds to the values reported in the literature (see e.g. [12]), but we should remark that the existence of a spin gap at all is not a fully secure statement.

For both lattices the exact diagonalization data yield indications for a quantum paramagnetic GS. For the kagomé lattice this statement is known from detailed studies by C. Lhuillier, H.-U. Everts and coworkers as well as other groups published over the last 10 years. However, the star lattice represents a new example for a quantum HAFM on a uniform 2D lattice without semi-classical GS ordering. We emphasize that the quantum paramagnetic GS for the star lattice is different in nature to the quantum GS for the kagomé lattice. The quantum GS for the star lattice is characterized by an

extremely strong NN correlation on the dimer bonds (more than 60% larger than the NN correlation of the honeycomb lattice having the same coordination number z = 3) and a weak NN correlation on the triangular bonds (only about 30% of the NN dimer correlation and significantly weaker than the triangular NN correlation of the kagomé and the triangular lattices). The singlet-triplet spin gap is particularly large (about ten times larger than that for the kagomé lattice). Although the classical GS exhibits a huge non-trivial degeneracy, remarkably one does not find low-lying singlets within this large spin gap, rather the first singlet excitation is well above the lowest triplet state. The low-lying spectrum as a whole resembles the spectrum of weakly coupled dimers [18]. All these features support the conclusion that the quantum GS of the HAFM on the star lattice is dominated by local singlet pairing and represents a so-called valence-bond crystal state (see also Sect. 2.4.4).

2.4.4 Summary and Comparison

Based on extensive exact diagonalization studies and on available results in the literature we discuss the GS ordering of the spin half HAFM on the 11 uniform Archimedean tilings in two dimensions. Of course we are not able to clarify all aspects of the GS properties of these quantum many-body systems. Nevertheless the comparative discussion of the 11 lattices leads to conclusions on the influence of lattice structure on GS magnetic ordering in two dimensions and this way on the existence or absence of semi-classical LRO in these systems. The HAFM has been already studied intensively in the literature for some of these lattices (square (T2), triangular (T1), honeycomb (T3), kagomé (T8), SrCuBO (T6), CaVO (T11)) and the physical picture seems to be more or less clear for those lattices. For some other lattices (SHD (T10), maple-leaf (T4) and trellis (T5)) only a few results are available in the literature so far and the conclusions on the GS ordering are less reliable. The HAFM on the star lattice (T9) as well as on the bounce lattice (T7) has not been studied till now.

Let us summarize the results of the preceding sections: The GS of the spin half HAFM on the bipartite (i.e. non-frustrated) lattices is semi-classically Néel ordered. The reduction of the order parameter by quantum fluctuations depends on the coordination number and on the competition of nonequivalent NN bonds (cf. Table 2.3). The low-energy spectra exhibit some typical features for magnetic systems with semi-classical order, namely well separated quasi-degenerate joint states (QDJS) with symmetries belonging to the classical GS ordering. Another indication for semi-classical ordering is the disappearance of the spin gap in the thermodynamic limit. We find, at least for the lattices with not too large unit cells, indications for a vanishing spin gap. However, the finite-size extrapolation of the spin gap is less reliable than for the magnetization (see Sect. 2.3.3) and therefore we do not consider the spin gap as a main criterion for the existence of semi-classical LRO. The comparison of the finite-size behavior of the GS energy shown in **Table 2.3.** Comparison of the ground-state energy per bond E_0 /bond and the order parameter m^+ (2.9) of the spin half HAFM obtained by finite-size extrapolation (see text). In order to see the effect of quantum fluctuations, we present m^+ scaled by its corresponding classical value m^+_{class} . Furthermore, we show the coordination number z and indicate, whether all NN bonds are equivalent or not by EQ and NEQ, respectively. For the star lattice (last row) the first value corresponds to m^+_{class} of the perfect ordered planar $\sqrt{3} \times \sqrt{3}$ and q=0 classical GS, see Fig. 2.18, whereas the second value corresponds to m^+_{class} averaged over all degenerate classical ground states

tiling	z	NN bonds	$ E_0 $ bond	$m^+/m_{\rm class}^+$
bipartite				
square $(T2)$	4	\mathbf{EQ}	-0.3350	0.635
honeycomb $(T3)$	3	\mathbf{EQ}	-0.3632	0.558
CaVO $(T11)$	3	NEQ	-0.3689	0.461
SHD (T10)	3	NEQ	-0.3713	0.425
frustrated				
SrCuBO (T6)	5	NEQ	-0.2310	0.456
triangular (T1)	6	\mathbf{EQ}	-0.1842	0.386
bounce $(T7)$	4	NEQ	-0.2837	0.286
trellis (T5)	5	NEQ	-0.2471	0.222
maple-leaf $(T4)$	5	NEQ	-0.2171	0.218
kagomé (T8)	4	\mathbf{EQ}	-0.2172	0.000
star $(T9)$	3	NEQ	-0.3093	$0.094 \ldots 0.150$

Fig. 2.20 shows that the extrapolation coefficient A_3 (cf. (2.12)) for the bipartite lattices is largest in agreement with long-ranged spin-spin correlations. We mention that a suppression of semi-classical LRO in bipartite lattices can appear in systems with NN bonds of different strength this way increasing the competition of non-equivalent NN bonds (see Sect. 2.5).

The situation for the frustrated lattices is more complex. Some of the criteria for semi-classical LRO might be weaker pronounced. For the HAFM on the kagomé and on the star lattice we find evidence for a quantum paramagnetic GS whereas for the other frustrated lattices there are indications for semi-classical LRO. Although the order parameter m^+ is additionally weakened by the interplay of quantum fluctuations and frustration the extrapolated values of m^+ remain finite (between 22% and 45% of the classical values) for the tilings T1,T4,T5,T6,T7. It vanishes however for the kagomé lattice and is at least very small for the star lattice (see Table 2.3, Fig. 21). Except for the kagomé and the star lattices the low-energy spectra exhibit some typical features for magnetic systems with semi-classical order, namely well separated QDJS with symmetries belonging to the classical GS ordering. The comparison of the finite-size behavior of the GS energy (Fig. 2.20) shows the smallest extrapolation coefficient A_3 for the kagomé and the star lattice being in agreement with short-range spin-spin correlations. Although the extrapolation coefficient A_3 is very small for the trellis lattice, too, we interpret this as a particular finite-size effect due to the incommensurate structure of the classical GS.

We conclude that the interplay of lattice structure and quantum fluctuations may lead to a non-classical quantum paramagnetic singlet GS for frustrated lattices with low coordination number and strong frustration, i.e. for the kagomé and the star lattice (see Fig. 2.22). Although extensive studies have been performed for the kagomé lattice [36, 38, 39, 86, 108–120], the spin half HAFM on the star lattice is considered in this article for the first time. By contrast with all the other lattices, the kagomé and the star lattice show a huge non-trivial degeneracy of the classical GS due to strong frustration.



Fig. 2.22. Border line between semi-classical magnetic order and quantum magnetic disorder in a parameter space spanned by frustration (classical GS energy per bond divided by s^2 , see Sect. 2.2.1) and coordination number z. The numbers indicate the location of the Archimedean tilings in this parameter space

Although there is no semi-classical GS order for both lattices, the nature of both quantum ground states is basically different in the quantum case. We argue that the origin for this difference lies in the existence of non-equivalent NN bonds in the star lattice whereas all NN bonds in the kagomé lattice are equivalent. That leads also to significant differences in the low-lying spectrum of both lattices. The kagomé lattice has probably a finite spin gap, but within this spin gap a large number (increasing exponentially with system size) of low-lying singlets appear [38,39,113] which seem to be a remnant of the nontrivial classical GS degeneracy. However, the HAFM on the star lattice has a particularly large spin gap but also a well pronounced singlet-singlet gap (even larger than the spin gap) which is in accord with a GS dominated by local singlet pairing on non-equivalent NN J_D bonds assisted by frustration. As a consequence, the huge classical GS degeneracy has no remnant in the spectrum of the quantum model. We mention that the checkerboard (planar pyrochlore) lattice is another example, where the non-trivial classical GS degeneracy does not lead to a continuum of low-lying singlets and the ground-state is most likely a valence bond crystal (see e.g. [128, 129]). Furthermore, examples are known that many low-lying non-magnetic excitations within the spin gap may appear although the classical GS is not non-trivially degenerate [41, 130].

For all the other bipartite and frustrated lattices the quantum fluctuations seem to be not strong enough to destroy the classical order. However, we should again emphasize that our conclusions about semi-classical LRO possesses some uncertainty, in particular for the trellis, maple-leaf and bounce lattices.

The above presented study provides some criteria for the appearance of novel quantum ground states in 2D spin systems. Although only for a few of the lattices under consideration direct realizations in real materials have been found till now, in several cases slightly modified models, e.g. models with NN couplings of non-equal strength or with inclusion of next-nearest neighbor couplings, are appropriate for the description of real magnetic substances.

At the end of this paragraph we will classify the magnetic ordering on the 11 Archimedean tilings using the four basic types of low-energy physics in 2D isotropic quantum antiferromagnets proposed and described recently by Lhuillier, Sindzingre, Fouet and Misguich [11–15]. The first type of GS phases is the semi-classical LRO (collinear or noncollinear). Most of the lattices belong this class, namely all bipartite lattices (T2,T3,T10,T11) but also the frustrated tilings (T1,T4,T5,T6,T7). The GS of the HAFM on these lattices breaks the SU(2) symmetry. The low-lying excitations are gapless Goldstone modes (magnons). As discussed above, the order parameter is reduced by quantum fluctuations. The three other types of GS phases, namely the so-called valence bond crystal, type I spin liquid and type II spin liquid are purely quantum.

The so-called valence bond crystal is a phase characterized by the formation of local singlets with high binding energy built by an even number of spins (most likely by two or four spins) connected by NN bonds (singlet 'valence bonds'). The correlation between the singlets is weak leading to a fast exponential decay of the spin pair correlation to zero. The GS is a rotationally invariant singlet of the total spin without SU(2) symmetry breaking. However, breaking of translational symmetry of the lattice is possible but not necessary. The valence bond crystal possesses long-range singlet-singlet (dimerdimer or plaquette-plaquette) correlations. All excitations above the GS are gapped leading to an exponential (i.e. thermally activated) low-temperature behavior of the specific heat c and of the susceptibility χ . A candidate for such a GS phase is the HAFM on the star lattice. For this lattice the possible dimer-dimer LRO would fit to the lattice geometry. Another candidate is the $J_1 - J_2$ model on the square lattice, widely discussed in the literature (see [85, 131] and references therein), where the valence bond crystal phase would break the translational symmetry of the lattice. The type I spin liquid has some similarity to the valence bond crystal. It has also a rotationally invariant singlet GS without SU(2) symmetry breaking, it has a fast exponential decay of the spin pair correlation to zero and gapped excitations leading to thermally activated low-temperature behavior of c and χ . However, the GS does not possess singlet-singlet long-ranged correlations but is likely to be characterized by short ranged resonating valence bonds. There is no good candidate for this phase among the Archimedean tilings. But this phase might be realized in the $J_1 - J_2$ model on the honeycomb lattice [42]. The type II spin liquid has also a rotationally invariant singlet GS, a fast exponential decay of the spin pair correlation to zero and no long-ranged singlet-singlet correlations. The spin gap Δ to the first triplet excitation is finite giving rise to a thermally activated low-temperature behavior of the susceptibility χ . However, there is a gapless continuum of singlets which could be described by a family of short-ranged valence bond states [116] the number of which is exponentially growing with size N. This gapless continuum implies that the system has a zero-temperature residual entropy and that the low-temperature specific heat is not thermally activated. The best candidate for this type of spin liquid is the spin half HAFM on the kagomé lattice.

2.5 Quantum Phase Transitions in 2D HAFM – The CaVO J - J' Model and the Shastry-Sutherland Model

Phase transitions have been a subject of great interest to physicists over many decades. Besides thermal phase transitions, the so-called quantum phase transitions (or zero-temperature transitions) have started to attract a lot of attention (see chapter by S. Sachdev in this book). For zero-temperature orderdisorder transitions we basically need the interplay between the interparticle interactions and quantum fluctuations. Canonical models to discuss quantum phase transitions are quantum spin models. As discussed above the HAFM on most of the 2D lattices possesses semi-classical LRO in the GS, but the interplay of quantum fluctuations and strong competition between bonds may suppress this order. The competition may appear either as frustration or by non-equivalent NN bonds or a combination of both. Indeed, the strength of this competition may serve as the control parameter of a zero-temperature order-disorder transition. It can be tuned by changing the relative magnitude of non-equivalent NN bonds or by introducing next-nearest neighbor bonds. The Archimedean tilings therefore represent a wide playground for the investigation of zero-temperature transitions.

A generic model of a frustrated HAFM widely discussed in the literature (see, e.g., [55,85,131-138]) is the spin-half $J_1 - J_2$ model on the square lattice, where the frustrating J_2 bonds plus quantum fluctuations are believed to

lead to a second-order transition from a Néel-ordered state to a quantum paramagnetic state at about $J_2 \approx 0.38 J_1$. The properties of the latter state are still far from being understood. One favored quantum phase for $J_2 \sim 0.5 J_1$ is a valence bond crystal. However, there are examples where frustration leads to a first-order transition in quantum spin systems in contrast to a second-order transition in the corresponding classical model (see, e.g., [27,74,99,139–141]).

The competition between non-equivalent NN bonds melts the semiclassical Néel order by formation of local singlets. By contrast to frustration, which yields competition in quantum as well as in classical systems, the local singlet formation is a pure quantum effect. Both mechanisms may of course be mixed as, for instance, in CaV_4O_9 or in $SrCu_2(BO_3)_2$ (see, e.g., [64, 68, 99]).

Let us first discuss a mean-field like approach to describe the continuous quantum phase transition driven by local singlet formation. To that end we study the HAFM on the CaVO lattice having two non-equivalent NN bonds J and J', see Fig. 2.10. The uncorrelated mean-field state for Néel LRO is the two-sublattice Néel state $|\phi_{MF_1}\rangle = |\uparrow\rangle|\downarrow\rangle|\uparrow\rangle|\downarrow\rangle|\downarrow\rangle|\downarrow\rangle|\downarrow\rangle|\downarrow\rangle\dots$ and for the dimerized singlet state it is the rotationally invariant product state of local singlets of the two spins belonging to a J' bond $|\phi_{MF_2}\rangle = \prod_{\{i,j\}_{J'}} \{|\uparrow_i\rangle|\downarrow_j\rangle - |\downarrow_i\rangle|\uparrow_j\rangle\}/\sqrt{2}$, where i is a site in the sublattice A and j a site in sublattice B. In order to describe the transition between both states, we consider an uncorrelated product state interpolating between $|\phi_{MF_1}\rangle$ and $|\phi_{MF_2}\rangle$ of the form [27,142]

$$|\Psi_{MF}(t)\rangle = \prod_{\{i,j\}_{J'}} \frac{1}{\sqrt{1+t^2}} \left\{ |\uparrow_i\rangle|\downarrow_j\rangle - t|\downarrow_i\rangle|\uparrow_j\rangle \right\}.$$
 (2.15)

We have $|\Psi_{MF}(t=0)\rangle = |\phi_{MF_1}\rangle$ and $|\Psi_{MF}(t=1)\rangle = |\phi_{MF_2}\rangle$. The minimal value of the energy is given by

$$\frac{E_{MF}}{N} = \frac{\langle \Psi_{MF} | H | \Psi_{MF} \rangle}{N} = \begin{cases} -\frac{3J'}{8} - \frac{1}{16J} (2J - J')^2 & J' \le 2J \\ -\frac{3J'}{8} & J' > 2J. \end{cases}$$
(2.16)

Furthermore, it is found that the sublattice magnetization m^z has the following form

$$m^{z} = \langle \Psi_{MF} | S_{i \in A}^{z} | \Psi_{MF} \rangle = \begin{cases} \frac{1}{4J} \sqrt{(2J - J')(2J + J')} & J' \leq 2J \\ 0 & J' > 2J. \end{cases}$$
(2.17)

Note that m^z vanishes at a critical point $J'_c = 2J$, and that the critical index is the mean-field index 1/2 (see Fig. 2.23b). Using the relation between the variational parameter t and the sublattice magnetization m^z we find the relation $E_{MF}/N = -\frac{1}{8}J' - \frac{1}{4}J'\sqrt{1 - 4(m^z)^2} - J(m^z)^2$ showing the typical behavior of a second-order transition, see Fig. 2.23a. We can expand E_{MF} up to the fourth order in m^z near the critical point and find a



Fig. 2.23. Mean field results for the J-J'-HAFM on the CaVO lattice. a: Energy versus order parameter. b: Order parameter versus J'

Landau-type expression, given by $E_{MF}/N = -\frac{3}{8}J' + \frac{1}{2}(J'-2J)(m^z)^2 + \frac{1}{2}J'(m^z)^4$.

Although this mean-field like description gives some qualitative insight into the physics of the quantum phase transition for the CaVO lattice more elaborated investigations [48, 64, 66, 70] show that the quantum phase transition to a rotationally invariant gapped dimerized GS phase takes place at $J'/J \approx 1.7$ and to the plaquette singlet GS phase at $J'/J \approx 0.9$. The critical exponents of quantum phase transitions driven by the competition of nonequivalent NN bonds in 2D quantum HAFMs are not the mean field exponents but those of the three-dimensional classical Heisenberg model [70, 143].

Another interesting example for quantum phase transitions in spin systems appears in the Shastry-Sutherland model, i.e. the $J_1 - J_2$ HAFM on the SrCuBO lattice (T6). We will use in this section the Shastry-Sutherland representation (frustrated square lattice, see Fig. 2.16a, lower part). The classical GS of this model has two phases: The collinear Néel phase for $J_2 \leq J_1$ and a spiral phase for $J_2 > J_1$ (cf. Fig. 1 in [94]). The transition between the two classical phases is of second order.

For $J_2 \leq J_1$ the physics of the quantum model is similar to that of the classical model, i.e., we have semi-classical Néel order (see Sect. 2.4.2). However, the quantum model exhibits new features for stronger frustration $J_2 > J_1$. Firstly, one finds that the collinear Néel phase in the quantum model can survive into the region where classically it is already unstable [94, 99, 102, 104]. This effect is known as order from disorder [144, 145] and is widely observed in quantum spin systems (see, e.g. [27, 55, 110]).



Fig. 2.24. Spin-spin correlation and square of sublattice magnetization (order parameter) scaled by their values for the square lattice $(J_2 = 0)$ for the Shastry-Sutherland model (N = 32)

Secondly, one knows already from Shastry and Sutherland [93] that for large enough J_2 the quantum GS is a rotationally invariant product state of local pair singlets $|\phi\rangle = \prod_{\{i,j\}_{J_2}} [|\uparrow_i\rangle| \downarrow\rangle_j - |\downarrow_i\rangle|\uparrow\rangle_j]/\sqrt{2}$ (so-called orthogonal dimer state), where *i* and *j* correspond to those sites which cover the J_2 bonds. This orthogonal dimer phase sets in at around $J_2^c \approx (1.45 \dots 1.48) J_1$ [95–97,99,102,104]. The nature of the transition to the dimer phase is still a matter of discussion, although there are arguments that the transition is probably of first order [94,99]. In the region $1.2J_1 \leq J_2 \leq 1.45J_1$ the main challenging question is whether the system has an intermediate phase. Candidates are quantum spiral phases or more favorable a plaquette RVB like phase. Despite numerous investigations, a definite picture concerning the existence and nature of an intermediate phase has not yet emerged.

We illustrate such behavior discussed above by finite-lattice results (N = 32) for the spin-spin correlation along the NN J_1 bond, along the diagonal J_2 bond and for the largest separation R = 4 available in the finite N = 32 Shastry-Sutherland lattice as well as for the square of sublattice magnetization \bar{m}^2 (cf. (2.6)) shown in Fig. 2.24. We have scaled the correlation functions and the sublattice magnetization by their corresponding values for the square lattice ($J_2 = 0$) for better comparison. The small changes in the correlation functions and the sublattice magnetization are in agreement with the survival of the collinear Néel ordering up to about $J_2 \sim 1.2J_1$. Beyond $J_2 \sim 1.2J_1$ the correlation functions change drastically up to $J_2 = 1.4785J_1$, where for N = 32 the rotationally invariant orthogonal dimer state becomes the GS. At this point the correlation functions and the sublattice magnetize magnetize magnetize magnetize magnetize magnetize the sublattice magnetize for the sublattice magnetize ($J_2 = 0$) for better comparison.

tization jump to their values of the orthogonal dimer state. The behavior in the region $1.2J_1 \leq J_2 < 1.4785J_1$ preceding the transition to the orthogonal dimer state seems to be in accordance with the existence of an intermediate phase.

2.6 Magnetization Process

In this final section we discuss the effect of a uniform external magnetic field on the models discussed so far. Once a small but finite magnetization is created by the external field, spins can no longer align completely antiparallel in the classical ground state even for a bipartite lattice. Since this is similar to the effect of geometric frustration, one can regard the magnetic field as introducing or enhancing frustration. One may therefore expect that a strong external field can induce further interesting quantum effects. In particular, we will discuss the quantum phenomena which are sketched in Fig. 2.25:

(a) Plateaux have a fixed magnetization m in a region of the applied magnetic field h. Note that a plateau with magnetization m = 0 corresponds to a spin gap at zero magnetic field h = 0. On a plateau, the magnetization m typically assumes a (simple) rational

On a plateau, the magnetization m typically assumes a (simple) rational fraction of its saturation value.

(b) Also some examples of jumps associated with a special degeneracy in the spectrum will be discussed in Sect. 2.6.4.

Specifically we consider the Heisenberg antiferromagnet (2.1) in a uniform external magnetic field h



Fig. 2.25. Schematic magnetization curve illustrating some plateaux (a) and a jump below saturation (b)

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$$H = \sum_{\langle i,j \rangle} J_{ij} \mathbf{S}_i \mathbf{S}_j - h \sum_i S_i^z \,. \tag{2.18}$$

In the following we will focus on the zero-temperature magnetization process of the Heisenberg antiferromagnet (2.18) on the 11 Archimedean and some related lattices. Some further aspects of two-dimensional s = 1/2 antiferromagnets in an external field have been summarized e.g. in [12,146].

In the present context it will sometimes be useful to allow for general length s of the local spin. One can also introduce an XXZ anisotropy as a prefactor Δ_I multiplying the z-z interaction term in (2.1). Note that a magnetic field $h \neq 0$ already breaks the symmetry from SU(2) down to U(1)such that in contrast to the case h = 0, there is no reason for the Heisenberg point $\Delta_I = 1$ to be special. We will nevertheless concentrate mainly on s = 1/2 and $\Delta_I = 1$.

An important observable is the magnetization

$$m = \frac{1}{s N} \sum_{i} S_i^z \tag{2.19}$$

which we normalize to saturation value m = 1 (recall that N is the total number of spins in the system). The magnetization (2.19) is a conserved quantity for the Hamiltonian (2.18): [H, m] = 0. One can therefore replace the operator (2.19) by its expectation value and by slight abuse of notation we will use the same symbol for both. The conservation of m is also technically useful for computing the magnetization curve since one can relate energies with a field E(h) to the energies $E(S^z, h = 0)$ for fixed total S^z at h = 0

$$E(h) = E(S^{z}, h = 0) - h S^{z}.$$
(2.20)

This implies that the GS energies in the sectors S^z and $S^z + 1$ cross at the magnetic field

$$h = E(S^{z} + 1, h = 0) - E(S^{z}, h = 0)$$
(2.21)

i.e. at this value of h the magnetization increases by 1/s N. The ground states with a given total spin S typically carry the maximal possible S^z and hence $S = S^z$ holds for them. In such a situation, $E(S^z, h = 0) = E(S)$ of the preceding sections and (2.21) implies that the h(m) curve is obtained by (discrete) differentiation of the E(S) curve at h = 0 with respect to $S \sim m$. In particular, if there is a regime with a quadratic dependence of E on S like in (2.10), the magnetization curve m(h) becomes linear in this regime.

If E(S) has a downward cusp, one obtains two different fields h_1 and h_2 when approaching the associated value of m from below and above, respectively, and one finds a plateau in m(h). In one dimension, the appearance of plateaux is governed by the following quantization condition on the magnetization m [147] (see also [146] for a more detailed discussion)

$$sV\left(1-m\right)\in\mathbb{Z}\,.\tag{2.22}$$

Here V is the number of spins in a translational unit cell of the **ground** state (i.e. the lowest state for a given m) which can be larger than (namely an integer multiple of) the unit cell of the Hamiltonian if translational symmetry is spontaneously broken.

In two dimensions, there is no proof yet that the condition (2.22) is a necessary one. Nevertheless, the condition (2.22) should apply to those cases where plateau states are ordered (e.g. valence bond crystals) and it is therefore at least a useful guide also in two dimensions.

Figure 2.26 shows results for magnetization curves of all 11 Archimedean lattices. With one exception, all these curves have been computed for finite lattices with N = 36 sites. Since for s = 1/2 only the discrete values $S^z = 0$, 1, ..., N/2 are allowed for a given N, one finds step-like curves on a finite lattice. The task is then to determine which parts of these curves will become smooth in the thermodynamic limit $N \to \infty$ or where anomalies like plateaux or jumps remain in this limit.

Clearly, the behavior in a magnetic field is even richer than the h = 0properties and we will therefore not aim at a complete analysis. Before we proceed with a discussion of some selected aspects, we would like to add some remarks on two tilings that we will not discuss further. Firstly, on the trellis lattice (T5) the ground states in a magnetic field carry incommensurate momenta. They correspond in the x-direction to the twist angle α_2 discussed in Sect. 2.4.2 (although for m < 1 the GS momenta in a quantum system are in general different from the classical twist angle). Since irrational momenta are not realized for any finite lattice, one obtains additional finite-size effects. However, we have checked that these effects are sufficiently small for the N =36 lattice which we have used to render the result in Fig. 2.26 qualitatively representative.

Secondly, the ground state on the CaVO lattice (T11) has a unit cell with 8 spins (see Sect. 2.4.1). Since this does not fit on a lattice with N = 36 sites, one observes large finite-size artifacts in this case. In fact, the CaVO lattice is the only one among the 11 Archimedean lattices where no good magnetization curve can be obtained for N = 36. For completeness, we nevertheless show this result as the dotted curve in Fig. 2.26, but we also show a curve for N = 32 (full line) which should be considered as representative.

In the following three sections we discuss the tilings T2 (square), T1 (triangular) and T8 (kagomé) in more detail.

2.6.1 Square Lattice

Let us start with a brief discussion of the magnetization process of the square lattice which is well understood and probably representative for the non-frustrated Archimedean tilings. Figure 2.27 shows the magnetization curve of the s = 1/2 square-lattice Heisenberg antiferromagnet obtained by different



Fig. 2.26. Magnetization curves of the s = 1/2 Heisenberg antiferromagnet with J = 1 on all 11 Archimedean tilings. Results are for N = 36 sites except for the tiling T11 (CaVO) where the full curve shows a result for N = 32 which should be more representative than N = 36 (shown as the dashed curve). For further details compare the text


Fig. 2.27. Magnetization curve of the s = 1/2 Heisenberg antiferromagnet on the square lattice. The thin solid line is for N = 40 sites, the full bold line is an extrapolation to the thermodynamic limit. A second-order spin-wave result [151] (bold dashed line) and QMC results (diamonds) are also shown

approaches. Firstly, the thin full line shows the result obtained by exact diagonalization for a finite lattice with N = 40 sites [148] (see also [88, 149, 150] for earlier exact diagonalization studies). The full bold line denotes an extrapolation of the exact diagonalization data to the thermodynamic limit which is obtained by connecting the midpoints of the finite-size steps at the largest available system size. One observes a smooth magnetization curve with no peculiar features (in particular no plateaux) for |m| < 1. Note that close to saturation the extrapolated curve includes data at large system sizes, which are not shown explicitly in Fig. 2.27 (the curve is based exclusively on finite lattices with at least 8×8 sites for $m \ge 0.84375$). The high-field part of the magnetization curve is therefore particularly well controlled by exact diagonalization.

The magnetization curve of a classical Heisenberg antiferromagnet would be just a straight line for all fields up to saturation. Hence, the curvature of the magnetization curve Fig. 2.27 is due to quantum effects. These quantum effects can also be studied by spin-wave theory; a second-order spin-wave result [151] is shown by the bold dashed line in Fig. 2.27.

Finally, the magnetization process of the square lattice can also be studied by quantum Monte Carlo (QMC) since this lattice is not frustrated, We have generated some values of m(h) on a 64 × 64 lattice (typically at T = J/50which we have lowered to T = J/200 upon approaching saturation) using the ALPS stochastic-series-expansion QMC application [152, 153]. These results are shown by the diamonds in Fig. 2.27 (statistical errors are much smaller than the size of the symbols) and agree with available stochasticseries-expansion QMC results [154].

The quantitative differences of the results of all three approaches are small, i.e., each approach yields a good description of the s = 1/2 HAFM on the square lattice. As the spin-wave approach [151] is based on a Néel state, we may therefore conclude that Néel order prevails in the transverse components for |m| < 1 (see also [150] for a discussion from the point of view of exact diagonalization).

The same picture is probably also valid for the other bipartite nonfrustrated tilings, namely T3 (honeycomb), T10 (SHD) and T11 (CaVO). All these lattices are believed to be Néel ordered at h = 0 (see Sect. 2.4.4). Upon application of a magnetic field, the Néel vector first turns perpendicular to the field and then the sublattice magnetizations are smoothly tilted towards the field direction until full polarization is reached. At least the numerical results for the magnetization curves shown in Fig. 2.26 for the lattices T3 (honeycomb – see also [88] for further details and numerical data), T10 (SHD) and T11 (CaVO) are consistent with a smooth magnetization curve.

From an experimental point of view, one needs a sufficiently small exchange constant J to render the saturation field accessible in a laboratory. Successful synthesis and measurement of the magnetization process of suitable s = 1/2 square lattice antiferromagnets have been reported in [54].

2.6.2 Triangular Lattice

The s = 1/2 XXZ model on the triangular lattice is among the first models whose magnetization process was studied by exact diagonalization [155]. These early studies already found a plateau with m = 1/3, at least for Isinglike anisotropies $\Delta_I > 1$. Due to the restriction to at most 21 sites, it was first not completely clear whether the plateau persists in the isotropic regime $\Delta_I \approx 1$. The magnetization process of the Heisenberg antiferromagnet $(\Delta_I = 1)$ was analyzed further using spin-wave theory [156]. This study provided evidence that the m = 1/3 plateau exists also at $\Delta_I = 1$ and estimates for its boundaries were obtained.

Figure 2.28 shows the magnetization curves obtained by exact diagonalization for the s = 1/2 Heisenberg antiferromagnet on finite lattices with N = 36and 39 sites (thin lines). There are small quantitative differences of the N =36 curve with exact diagonalization results presented previously [37, 88, 146] whose origin is discussed in [148]. Both curves in Fig. 2.28 exhibit a clear plateau at m = 1/3 in an otherwise smooth magnetization curve. The spin-wave results for the magnetic fields at the lower $h_1 = 3 (s - 0.084) J = 1.248 J$ and the upper boundaries $h_2 = 3 (s + 0.215) J = 2.145 J$ of the m = 1/3plateau [156] are smaller by about 0.13J (lower boundary) and 0.01J (upper boundary) than the exact diagonalization results presented here for N = 39and s = 1/2.



Fig. 2.28. Magnetization curve of the s = 1/2 Heisenberg antiferromagnet on the triangular lattice. The thin dashed and solid lines are for N = 36 and 39 sites, respectively. The bold line is an extrapolation to the thermodynamic limit

The full bold line in Fig. 2.28 denotes an extrapolation of the exact diagonalization data to the thermodynamic limit which is obtained by connecting the midpoints of the finite-size steps at the largest available system size (except for the boundaries of the m = 1/3 plateau where corners were used). Close to saturation this includes again bigger system sizes than those explicitly shown in Fig. 2.28.

The state of the m = 1/3 plateau can be easily understood in the Ising limit $\Delta_I \gg 1$ [87, 88]. Quantum fluctuations are completely suppressed in the limit $\Delta_I \to \infty$ and the m = 1/3 state is a classical state where all spins on two of the three sublattices of the triangular lattice point up and all spins on the third sublattice point down. This state corresponds to an ordered collinear spin configuration. It is threefold degenerate and breaks the translational symmetry. One can then use perturbation theory in $1/\Delta_I$ to study the m = 1/3 plateau of the XXZ model [88]. However, the current best estimate of the point $\Delta_{I,c}$ where the m = 1/3 plateau disappears is obtained from a numerical computation of the overlap of the Ising states and the m = 1/3 wave function of the full XXZ model with s = 1/2: $\Delta_{I,c} = 0.76 \pm 0.03$ [148]. This means that the m = 1/3 plateau states of the Ising antiferromagnet and the Heisenberg antiferromagnet on the triangular lattice are qualitatively the same.

In the absence of a magnetic field, order persists in the Heisenberg antiferromagnet on the triangular lattice despite the geometric frustration (see Sect. 2.4.2). We have now seen that the magnetic field enhances the frustration sufficiently in the Heisenberg antiferromagnet on the triangular lattice to open a spin gap and thus a plateau at m = 1/3.

Among the other magnetization curves shown in Fig. 2.26, the one of the bounce lattice (T7) looks most similar to the one of the triangular lattice. Indeed, also the tiling T7 consists of triangles and one may expect that also here an up-up-down spin structure on each triangle gives rise to an m = 1/3 plateau. However, the covering of the complete lattice with up-up-down triangles is not unique for the bounce lattice, indicating at least some differences in the magnetization process of the triangular and bounce lattices.

2.6.3 Kagomé Lattice

Among the Archimedean lattices, the kagomé (T8) and star (T9) lattices are characterized by the combination of strong frustration and low coordination number. As discussed in Sects. 2.4.3 and 2.4.4, we believe that they give rise to a quantum paramagnetic ground state at h = 0. The N = 36 magnetization curves in Fig. 2.26 indicate that these two lattices are presumably also those with the most complicated and rich magnetization processes among all 11 Archimedean lattices. Here we summarize the current understanding of the magnetization process of the s = 1/2 kagomé lattice and leave a detailed investigation of the star lattice to the future.

Figure 2.29 shows complete magnetization curves for the kagomé lattice with N = 27 and 36 sites as well as the high-field part of N = 45 and 54 curves [148,157,158]. Firstly, there should be a plateau at m = 0 associated to



Fig. 2.29. Magnetization curves of the s = 1/2 Heisenberg antiferromagnet on the kagomé lattice with N = 27, 36 (complete), 45 and 54 (partial). The inset shows a magnified version of the region around m = 7/9

the small spin gap above the quantum paramagnetic ground state. However, this is difficult to recognize in Fig. 2.29.

A plateau at m = 1/3 may be better recognized in Fig. 2.29. In fact, the presence of this plateau at m = 1/3 in the s = 1/2 Heisenberg antiferromagnet on the kagomé lattice has been established previously by considering also system sizes different from those shown in Fig. 2.29 [157, 159]. The state of this plateau is, however, quite non-trivial. For the classical Heisenberg antiferromagnet at m = 1/3, thermal fluctuations select collinear states, but due to the huge degeneracy of these states, there appears to be no real order on the classical level at m = 1/3 [160] (see also [161]). For s = 1/2, it is useful to consider the XXZ model. In the Ising limit $\Delta_I \to \infty$ one can then first establish [162] a relation to a quantum dimer model on the honeycomb lattice which was argued [163,164] to give rise to a valence bond crystal ground state with a $\sqrt{3} \times \sqrt{3}$ order. Fig. 2.30 shows a qualitative picture of this state. In the present context the circles indicate resonances between the two different Néel states on the surrounding hexagon. The next step is to compute the overlap of the m = 1/3 wave function of the XXZ model with that of the quantum dimer model as a function of Δ_I and one finds no evidence for a phase transition for $\Delta_I \geq 1$ [162]. This implies that also the m = 1/3state of the s = 1/2 Heisenberg antiferromagnet on the kagomé lattice is an ordered state with features similar to the valence bond crystal. There are many low-lying non-magnetic excitations above the lowest m = 1/3 state which can be considered as a remnant of the classical degeneracy. However, the valence-bond-crystal-type order implies just three degenerate m = 1/3ground states related by translational symmetry (see Fig. 2.30 for illustration) and a gap to all excitations above this three-fold degenerate ground state. Note that for the s = 1/2 Heisenberg antiferromagnet on the kagomé lattice this non-magnetic gap in the m = 1/3 sector turns out to be quite small (estimates are of the order of J/25 [162]).



Fig. 2.30. Part of the kagomé lattice with a $\sqrt{3} \times \sqrt{3}$ superstructure indicated by the circles in certain hexagons. Arrows indicate spins which are aligned with the magnetic field

There may be a further plateau at m = 5/9 in Fig. 2.29 although it is difficult to draw unambiguous conclusions from the available numerical data in this region of magnetization values.

Finally, one can see a pronounced jump of height $\delta m = 2/9$ just below saturation and a plateau at m = 7/9 in the magnetization curve of the s = 1/2 kagomé lattice. Both features will be discussed in more detail in the next section.

2.6.4 Independent Magnons and Macroscopic Magnetization Jumps

For the s = 1/2 Heisenberg model on a given two-dimensional lattice it is a very rare event that one can write down the ground state exactly. One such exceptional case is the dimerized ground state arising in the two-dimensional Shastry-Sutherland model [93] (see Sect. 2.5). It is therefore remarkable that in the high-field region of some popular frustrated lattices such as the kagomé lattice one can construct a macroscopic number of exact ground states. We will discuss some aspects of the construction in more detail in this section, focusing in particular on the kagomé lattice. Note that similar constructions can be given for other lattices [158, 165] and finite clusters [166] (for other points of view we also refer to [158, 165]). We also wish to remark that the construction of exact eigenstates to be described below works for models where no non-trivial conservation laws are known. However, it is restricted to the transition to saturation, since, as will become clear in the following, it relies on the knowledge of a reference state (namely the ferromagnetically polarized state $|\uparrow \dots \uparrow\rangle$ which is a trivial eigenstate of the Hamiltonian) and an analytic determination of the one-magnon excitations above it.

Now let us be more specific and, as the first step, consider very high magnetic fields such that the ground state is the ferromagnetically polarized state. In highly frustrated spin models, the lowest branch $\omega_0(\mathbf{k})$ of the one-magnon excitations above the ferromagnetically polarized state often has some flat directions (i.e. does not depend on some of the components k_i) or is completely flat (i.e. independent of \mathbf{k}). In the latter case, one finds a special type of jump just below the saturation field as well as indications for a plateau below the jump [158, 165].

The explicit computation of the one-magnon spectrum above the ferromagnetically polarized state depends on the model. For example, the kagomé lattice has a unit cell containing three sites and the spectrum is obtained by diagonalization of a 3×3 matrix. For the spin-s XXZ model one then finds the three magnon branches $\omega_i(\mathbf{k})$ (i = 0, 1, 2) which are shown in Fig. 2.31. Remarkably, the lowest branch $\omega_0(\mathbf{k}) = h - (2 + 4\Delta_I) Js$ is completely flat, i.e. independent of \mathbf{k} . This property is a fingerprint of the strong frustration caused by the triangles in the kagomé lattice. In fact, the lowest magnon branch relative to the ferromagnetically polarized state is also completely flat for



Fig. 2.31. The three branches $\omega_i(\mathbf{k})$ of one-magnon excitations above the ferromagnetic background for the kagomé lattice along the path in the Brillouin zone shown in the inset. Note that $\omega_0(\mathbf{k})$ is completely independent of \mathbf{k}

some other popular highly frustrated lattices including the pyrochlore lattice and its two-dimensional projection, namely the checkerboard lattice [158].

The one-magnon excitations can be localized in the real-space directions corresponding to a flat direction in **k**-space by using an inverse Fourier transformation. If the dispersion is completely flat, one can construct a magnon excitation that is localized in a finite volume. For the kagomé lattice, these local magnon excitations are located on the hexagons marked by circles in Fig. 2.30. Apart from normalization, this state is given by

$$|1\rangle \sim \sum_{x} (-1)^{x} S_{x}^{-} |\uparrow \dots \uparrow\rangle$$
(2.23)

where the sum runs over the 6 corners of the hexagon. Localization can be verified since each spin next to the hexagon is coupled to two spins in the hexagon such that contributions of flipped spins propagating onto the exterior site add with different signs and thus cancel. Therefore, a localized magnon is an exact eigenstate of the XXZ Hamiltonian on the kagomé lattice.

Now one can create further localized magnon excitations. As long as the local magnons are sufficiently well separated in space, they do not interact and consequently the many-magnon state is still an exact eigenstate. The non-trivial step is to verify that these non-interacting localized magnon excitations are not only eigenstates but in fact ground states in their respective magnetization subspaces. This result is probably true for general s, general coupling geometries with $J_{i,j} \geq 0$ and XXZ anisotropy $\Delta_I \geq 0$. In [158] the ground state property was verified numerically for some cases and it has been shown rigorously for certain subsets of the parameters, namely for $s = 1/2, \Delta_I \ge 0$ and all coupling constants $J_{i,j}$ equal [166] or for general s and $J_{i,j} \ge 0$, but isotropic interaction $\Delta_I = 1$ [167].

If the localization region is finite, a macroscopic fraction of the spins in the system can be flipped using local magnon excitations. Since the energies of the individual excitations add without interaction terms, one obtains a finite interval of the magnetization m where the ground state energy E(m)becomes a linear function. Due to the relation (2.21), this linear behavior leads to a finite jump in the magnetization curve m(h) at the saturation field h_{sat} .

Inspection of Fig. 2.30 shows that at most N/9 local magnons fit on a finite kagomé lattice. Therefore, a jump of height $\delta m = 1/(9 s)$ is predicted for the kagomé lattice. For the s = 1/2 Heisenberg antiferromagnet on the kagomé lattice one indeed observes numerically a jump of height $\delta m = 2/9$ which is independent of the system size if boundary conditions are chosen appropriately (see Fig. 2.29). Note that the height of the jump is in general proportional to 1/s and vanishes in the classical limit $s \to \infty$. Therefore, the macroscopic jump caused by independent local magnons is a true macroscopic quantum effect.

The maximal number of local excitations is obtained for their closest possible packing. The circles in Fig. 2.30 indicate this state for the kagomé lattice. This clearly is an ordered (crystalline) state. According to general arguments [98, 168], one expects a gap above such a crystalline state and consequently a plateau in the magnetization curve at the foot of the jump. This conclusion is supported by the numerical magnetization curve of the s = 1/2 Heisenberg antiferromagnet on the kagomé lattice, Fig. 2.29, which exhibits a clear plateau at m = 7/9 with a width around 0.07J [169].

The excitation energy of a local magnon is exactly zero at the saturation field $h_{\rm sat}$. Hence, all independent magnon states are exactly degenerate at $h = h_{\rm sat}$. The number of these states grows exponentially with N. This can be seen by considering the subset of states where magnons sit only on the positions of the crystalline state. Since the number of such positions is proportional to N and each position can be empty or occupied by a magnon, one finds an exponentially growing lower bound on the number of independent magnon states (this lower bound is $2^{N/9}$ for the kagomé lattice). In other words, the local magnon excitations give rise to a finite zero-temperature entropy at $h = h_{\rm sat}$ for a quantum spin system !

The star lattice (T9) is the other Archimedean tiling which supports local magnon excitations. In this case, the magnons are localized around dodecagons as shown by the circles in the inset of Fig. 2.32. The adjacent triangles again ensure localization via destructive interference of hopping processes out of a dodecagon. One can read off from the inset of Fig. 2.32 that a finite star lattice can in general accomodate at most N/18 local magnons. This implies a jump of height $\delta m = 1/(18 s)$ below saturation with a plateau at m = 1 - 1/(18 s) corresponding to the crystalline pattern of local magnon



Fig. 2.32. High-field part of the magnetization curves of the s = 1/2 Heisenberg antiferromagnet on the star lattice (T9) with N = 54 (dashed line) and N = 72 sites (solid line). The inset indicates the closest packing of local magnon excitations

excitations sketched in the inset of Fig. 2.32. The main panel of Fig. 2.32 shows that a jump of the expected height $\delta m = 1/9$ is indeed present in the magnetization curves of the s = 1/2 model on lattices with N = 54 and 72. Note that the N = 36 lattice whose magnetization curve is shown in Fig. 2.26 is not generic, but an exception from the point of view of local magnons. Due to its small linear extent, it has more and shorter cycles wrapping around the boundary than present in the infinite system, namely of length eight while the dodecadons yield cycles with length twelve. This N = 36 lattice then supports not only two but three local magnons and therefore the jump is higher than in the generic situation. Note further that a plateau is expected below this jump, i.e. at m = 8/9 for s = 1/2. However, the N = 54 and 72 curves in Fig. 2.32 do not allow an unambiguous confirmation of the presence of such a plateau.

The checkerboard and a square-kagomé lattice are further two-dimensional lattices supporting local magnon excitations [158, 169]. On the checkerboard lattice, a magnon is localized around a square. This leads to a jump of size $\delta m = 1/(8 s)$, as one can verify numerically for s = 1/2 [165].

We would like to mention in passing that there are instabilities towards lattice deformations. However, it can be argued that the most favorable instability is one which preserves the local magnon excitations as exact eigenstates and the associated degeneracy [169].

A related but different situation arises in two dimensions if the minima of the one-magnon excitations form a one-dimensional manifold. One example is the two-dimensional Shastry-Sutherland lattice [170] whose magnetization process will be discussed in the next section, another one is the frustrated square lattice mentioned in Sect. 2.5 at $J_2 = J_1/2$ [171, 172]. In this case, magnon excitations can be constructed [146, 158] that are localized in some directions, but not all. The frustrated square lattice can accommodate L/2 local magnon excitations [146, 158] if the linear extent of the lattice is L, leading to a finite-size jump $\delta m = L/(2 N s)$. A finite-size jump of height $\delta m = L/N$ is indeed observed in exact diagonalization studies of the s = 1/2 frustrated square lattice at $J_2 = J_1/2$ [146, 172]. However, due to the incomplete localization, the height of the jump vanishes in the thermodynamic limit, i.e. the transition to saturation remains continuous in such a case. Although the magnetization curve should be exceptionally steep just below saturation, the precise asymptotic form has been discussed controversially [150, 158, 172]. A recent diagrammatic analysis of the condensation problem into the one-magnon dispersion yields a square-root dependence with a logarithmic correction for the frustrated square lattice at $J_2 = J_1/2$ [173].

2.6.5 Shastry-Sutherland Model Versus SrCu₂(BO₃)₂

For the purpose of high-field magnetization experiments one does not only need materials which realize a given lattice structure, but in addition J must be small in order to be able to achieve full or at least a macroscopic polarization of the sample in (pulsed) magnetization experiments. $\text{SrCu}_2(\text{BO}_3)_2$ is an s = 1/2 material whose lattice structure corresponds to the tiling T6 and where the exchange constants are sufficiently small to close the spin gap by an external magnetic field and study the material at finite magnetizations in a laboratory. The magnetization process of $\text{SrCu}_2(\text{BO}_3)_2$ has attracted considerable attention because plateaux are observed in the magnetization curve¹¹ at m = 1/8, 1/4 and 1/3 [5,174–176] (see Fig. 2.33).

By contrast, the tiling T6 at $J = J_1 = J_2$ has a smooth magnetization curve (see Fig. 2.26), hence we need to consider the Shastry-Sutherland model with $J_1 \neq J_2$. The theoretical analysis of the magnetization process of the two-dimensional Shastry-Sutherland model [93] has been summarized in [105] – here we discuss only some selected aspects.

For $J_2 \rightarrow 0$, the Shastry-Sutherland model reduces to the square lattice antiferromagnet which is Néel ordered in the transverse components for all magnetic fields (see Sect. 2.6.1). As discussed in Sect. 2.5, this Néel phase extends beyond $J_2 = J_1$ for m = 0. For $m \rightarrow 1$, Néel order in the transverse components is stable for $J_2 \leq J_1$ [170]. We have performed a finite-size analysis of the widths of the m = 1/8, 1/4, 1/3 and 1/2 steps and found no indications for plateaux in the thermodynamic limit for $J_2 = J_1$. These considerations indicate the absence of quantum phase transitions between

¹¹ Magnetization experiments are controlled by a material-dependent and anisotropic g-factor. The s = 1/2 spins in SrCu₂(BO₃)₂ are localized on Cu²⁺-ions, hence $g \approx 2$ – see e.g. [174] for more details.



Fig. 2.33. Magnetization curves of a SrCu₂(BO₃)₂ single crystal scaled by g = 2.28 for $h \parallel c$ and g = 2.05 for $h \perp c$ [174]. Also shown are magnetization curves of the s = 1/2 Heisenberg antiferromagnet on N = 32 and 36 Shastry-Sutherland lattices for $J_2 = 57$ T, $J_1 = 0.6 J_2$

 $J_2 = 0$ and $J_2 = J_1$ for any value of the field h such that Néel order persists for the tiling T6 with $J_2 = J_1$ at all magnetic fields.

This is one indication that $SrCu_2(BO_3)_2$ should be described by $J_2 > J_1$ since several plateaux are observed in its magnetization curve Fig. 2.33, namely at m = 0, 1/8, 1/4 and 1/3 [5,174–176]. In this regime, one can perform perturbation expansions around the limit of decoupled dimers $J_1 = 0$ and indeed perturbation theory plays a central role in the theoretical approaches [98, 170, 177–179]. Plateaux at m = 0, 1/2, 1/3 and 1/4 then arise in zeroth, first, second and fourth order perturbation theory in J_1 , as has been clearly pointed out in [179].

For a direct comparison with the Shastry-Sutherland model, we adopt the estimates $J_1 \approx 0.6 J_2$ and $J_2 \approx 70 - 75$ K obtained by analyzing inelastic neutron scattering data [180–182] or the specific heat in a magnetic field [176]. The magnetization curves for the Shastry-Sutherland model shown in Fig. 2.33 were computed by choosing first $J_1 = 0.6 J_2$ and then setting the overall scale to $J_2 = 57$ T (≈ 77 K with g = 2). The m = 1/8 and 1/4 plateaux (present only for N = 32 in Fig. 2.33), the m = 1/3 plateau (present only for N = 36 in Fig. 2.33) and the m = 0 plateau agree roughly with the experimental results [174]. We have also performed computations for the value $J_1 = 0.68 J_2$ proposed in [174] and have found less good agreement.

Only the region with $m \leq 1/3$ has so far been accessed with magnetization experiments on SrCu₂(BO₃)₂. Hence, the magnetization curves for the N = 32 and N = 36 Shastry-Sutherland lattices are also restricted to



Fig. 2.34. Magnetization curve of the s = 1/2 Heisenberg antiferromagnet on the Shastry-Sutherland lattice for $J_2 = 1$, $J_1 = 0.6$. The dashed and solid lines are for N = 32 and N = 36 sites, respectively. The diamonds denote the exact value of the saturation field at m = 1 [170] and a series expansion result for the gap to S = 1 excitations at m = 0 [180], respectively

 $m \leq 1/3$ in Fig. 2.33. Fig. 2.34 shows the corresponding complete magnetization curves for $J_2 = 0.6 J_1$. Note that we have chosen an N = 32 lattice which is compatible with the structure of the m = 1/8 plateau in $\mathrm{SrCu}_2(\mathrm{BO}_3)_2$ as determined by NMR [175]. For both finite lattices, the saturation field agrees well with the analytical result [170] shown by one of the diamonds in Fig. 2.34. Also the boundary of the m = 0 plateau is in good agreement with the spin gap (i.e. the gap to S = 1 excitations) computed by expansion around the dimer limit $J_1 = 0$ [180] (compare the second diamond in Fig. 2.34).

As on other lattices, it is more difficult to draw unambiguous conclusions from finite-size data for intermediate values of m. One complication which the Shastry-Sutherland model shares with the trellis lattice are incommensurate ground states arising from the spiral phase for $J_1 < J_2$ in the classical model (see Sect. 2.5). A more general aspect is that given magnetizations are realized only for a limited number of sizes N. For example, lattices with N = 32 and 36 share only m = 1/2 in addition to m = 0 and 1. Even for m = 1/2finite-size effects are still important in Fig. 2.34 although the presence of a plateau at m = 1/2 is well established in the Shastry-Sutherland model (see above and [105]). m = 1/8 is realized only for N = 16 apart from N = 32. From these two lattice sizes one may estimate a plateau width $\leq J_2/10$ for the Shastry-Sutherland model at $J_1 = 0.6 J_2$, but the evidence in favor of a plateau at m = 1/8 is not very strong yet although its structure has already been analyzed in detail [175, 183].

2.6.6 Summary of Plateaux and Related Topics

Let us summarize the findings of this section. Firstly, in Sect. 2.6.1 we have discussed the square lattice which we believe to be representative for the non-frustrated bipartite tilings T2 (square), T6 (honeycomb), T10 (SHD) and T11 (CaVO). In these cases, Néel order probably persists in the transverse components for all magnetic fields up to saturation, leading to a smooth magnetization curve. The frustrated tilings T4 (maple leaf), T5 (trellis) and T6 (SrCuBO) may behave similarly. At least their N = 36 magnetization curves shown in Fig. 2.26 appear smooth and provide no evidence for any plateaux or jumps.

Also the triangular (T1) and bounce lattices (T7) are magnetically ordered at h = 0. However, in these two cases a plateau appears at m = 1/3 in the magnetization curves (see Sect. 2.6.2). In both cases, the appearance of a plateau at m = 1/3 may be attributed to the fact that these lattices are built from triangles. Nevertheless, the structure of the m = 1/3 state on the bounce lattice may be different from the one of the triangular lattice which corresponds to a long-range ordered collinear up-up-down spin configuration.

The tilings T8 (kagomé) and T9 (star) have the most interesting magnetization curves. According to Sect. 2.4.3, at h = 0 the kagomé lattice is expected to have a small spin gap whereas the star lattice has a large one. This gives rise to a narrow and pronounced plateau at m = 0, respectively. Comparison of results for the s = 1/2 Heisenberg antiferromagnet on the kagomé lattice with different sizes N (see Sect. 2.6.3) shows that the magnetization curve has a plateau at m = 1/3. Evidence has been provided recently [162] that the state of this m = 1/3 plateau on the kagomé lattice has a structure of the valence-bond-crystal type. The N = 36 magnetization curve of the star lattice shown in Fig. 2.26 indicates an m = 1/3 plateau, too. Since the lattice T9 also consists of triangles, it is plausible that this m = 1/3 plateau persists in the limit $N \to \infty$. Further plateaux are suspected on both lattices, including one at m = 5/9 on the kagomé lattice (see Sect. 2.6.3) and a similar one at m = 7/9 on the star lattice (compare Fig. 2.32) even if the currently available numerical data do not allow definite conclusions.

Close to saturation, exact eigenstates can be constructed for the strongly frustrated tilings T8 (kagomé) and T9 (star) – see Sect. 2.6.4. For general s they give rise to a jump below saturation of height $\delta m = 1/(9s)$ (T8) and $\delta m = 1/(18s)$ (T9). Furthermore, a plateau is expected directly below this jump and such a plateau is indeed observed in the s = 1/2 Heisenberg antiferromagnet at m = 7/9 for the kagomé lattice (T8) and possibly at m = 8/9 for the star lattice (T9).

Although there are still open issues concerning the magnetization process on the 11 Archimedean lattices, it is already clear that even richer behavior is found if one allows different exchange constants on non-equivalent bonds or adds further couplings. Examples include the following:

- The two-dimensional Shastry-Sutherland model has been discussed in Sect. 2.6.5. Here plateaux with m = 0, 1/4, 1/3 and 1/2 have been found and a further one is expected at m = 1/8. In contrast, its ancestor, the tiling T6 has a smooth magnetization curve with no particular features (see Fig. 2.26).
- A similar situation arises in the CaVO lattice (T11) if one allows for two different exchange constants J and J' as shown in Fig. 2.10. It is clear at least in the limit $J' \rightarrow 0$ that plateaux can then arise for m = 0 and 1/2 [77]. Further plateaux with m = 1/4, 3/4 and 1/8 arise in some parameter regions if one adds a second-neighbor interaction J_2 [77,98].
- Not only the zero-field properties of the frustrated square lattice have attracted considerable attention (see Sect. 2.5), but also its magnetization process has been studied intensively [146, 149, 150, 171–173, 184–187]. In this model, a collinear up-up-up-down state arises at half the saturation field [184, 185]. For s = 1/2, this state is found to be stabilized in the region $0.5 \leq J_2/J_1 \leq 0.66$ where it gives rise to a plateau with m = 1/2 [146, 172, 184, 185]. A further plateau at m = 1/3 is predicted by a Chern-Simons theory [187] and might also be observable in exact diagonalization studies although the latter do not allow definite conclusions about the presence or absence of an m = 1/3 plateau yet [146].
- Another variant of the square lattice is the checkerboard lattice, a planar projection of the pyrochlore lattice. The s = 1/2 checkerboard lattice has a pronounced spin gap at h = 0 [128, 129, 188], i.e. a plateau at m = 0. In the limit of decoupled four-spin units [129] another plateau arises at m = 1/2. Numerical data for N = 32, 40 [165] and 36 sites support the presence of an m = 1/2 plateau also in the checkerboard model where all coupling constants are equal. The construction of Sect. 2.6.4 predicts another plateau at m = 3/4 in the s = 1/2 checkerboard model although here the numerical evidence [165] is less clear.
- One can also add multi-spin interactions. On the triangular lattice, inclusion of four-spin cyclic exchange terms in the s = 1/2 model gives rise to an additional plateau at m = 1/2 [12,130,189,190]. This m = 1/2 plateau is already present in the classical model where one also finds an m = 1/3 plateau for a suitable choice of parameters [191]. The latter differs from the m = 1/3 plateau of Sect. 2.6.2 which arises only in the quantum Heisenberg antiferromagnet on the triangular lattice and is absent in the classical limit.

All the aforementioned plateaux for $m \neq 0$ give rise to ordered ground states (at least in those cases where the state of the plateau is sufficiently well understood). The unit cell of the ground state then has a volume V such that the magnetization m satisfies the quantization condition (2.22). Hence, also in two dimensions this quantization condition seems to hold generically.

The transitions to saturation in 9 of the 11 Archimedean lattices appear to be continuous quantum phase transition (see Fig. 2.26). Generically, the dispersion of the one-magnon excitations above the ferromagnetically polarized state should be quadratic close to their minima. An analysis of the associated condensation problem then predicts the following universal asymptotic behavior of the magnetization curve close to the saturation field $h_{\rm sat}$ [192–194]

$$1 - m \sim \left(\frac{h_{\text{sat}} - h}{J}\right) \ln \left(\frac{b J}{h_{\text{sat}} - h}\right)$$
(2.24)

where b is a non-universal constant. The logarithmic correction in (2.24) is characteristic for two dimensions and arises because of a logarithmic singularity in the density of states [192]. The functional form (2.24) has been verified by a first-order spin-wave approximation for the square lattice [151] and numerically for the s = 1/2 Heisenberg antiferromagnet on the square, honeycomb and triangular lattices [88, 146]. We note that a behavior of the form (2.24) is expected to be valid at generic continuous transitions at plateau boundaries in two dimensions [194] (at least in those cases where the fundamental excitations are magnons).

Deviations from (2.24) are expected if the one-magnon dispersion is not quadratic close to the minimum which in general requires fine-tuning of parameters. Nevertheless, completely flat bands arise in two Archimedean lattices, namley the kagomé and star lattices (T8 and T9). In these cases, we find a macroscopic jump in the magnetization curve just below saturation as we have discussed in Sect. 2.6.4

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3 Molecular Magnetism

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Abstract. Magnetic molecules are fascinating new substances with a wide variety of applications in physics, magneto-chemistry, biology, biomedicine and material sciences as well as in quantum computing. Each of the identical molecular units can contain as few as two and up to several dozens of paramagnetic ions whose interaction is rather well described by the Heisenberg model with isotropic nearest neighbor interaction sometimes augmented by anisotropy terms. Studying such finite spin systems focuses on qualitatively new physics caused by the finite size of the system. Among the findings discussed in this chapter are extensions of the theorems of Lieb, Schultz, and Mattis, the introduction of rotational bands as well as the discovery of giant magnetization jumps.

3.1 Introduction

The synthesis of molecular magnets has undergone rapid progress in recent years [1–6]. Each of the identical molecular units can contain as few as two and up to several dozens of paramagnetic ions ("spins"). One of the largest paramagnetic molecules synthesized to date, the polyoxometalate {Mo₇₂Fe₃₀} [7] contains 30 iron ions of spin s = 5/2. Although these materials appear as macroscopic samples, i. e. crystals or powders, the intermolecular magnetic interactions are utterly negligible as compared to the intramolecular interactions. Therefore, measurements of their magnetic properties reflect mainly ensemble properties of single molecules.

Their magnetic features promise a variety of applications in physics, magneto-chemistry, biology, biomedicine and material sciences [1,3,4] as well as in quantum computing [8–10]. The most promising progress so far is being made in the field of spin crossover substances using effects like "Light Induced Excited Spin State Trapping (LIESST)" [11].

It appears that in the majority of these molecules the localized singleparticle magnetic moments couple antiferromagnetically and the spectrum is rather well described by the Heisenberg model with isotropic nearest neighbor interaction sometimes augmented by anisotropy terms [12–16]. Thus, the interest in the Heisenberg model, which is known already for a long time [17], but used mostly for infinite one-, two-, and three-dimensional systems, was renewed by the successful synthesis of magnetic molecules. Studying such spin arrays focuses on qualitatively new physics caused by the finite size of the system.

Several problems can be solved with classical spin dynamics, which turns out to provide accurate quantitative results for static properties, such as magnetic susceptibility, down to thermal energies of the order of the exchange coupling. However, classical spin dynamics will not be the subject of this chapter, it is covered in many publications on Monte-Carlo and thermostated spin dynamics. One overview article which discusses classical spin models in the context of spin glasses is given by [18].

Theoretical inorganic chemistry itself provides several methods to understand and describe molecular magnetism, see for instance [19]. In this chapter we would like to focus on those subjects which are of general interest in the context of this book.

3.2 Substances

From the viewpoint of theoretical magnetism it is not so important which chemical structures magnetic molecules actually have. Nevertheless, it is very interesting to note that they appear in almost all branches of chemistry. There are inorganic magnetic molecules like polyoxometalates, metal-organic molecules, and purely organic magnetic molecules in which radicals carry the magnetic moments. It is also fascinating that such molecules can be synthesized in a huge variety of structures extending from rather unsymmetric structures to highly symmetric rings.

One of the first magnetic molecules to be synthesized was Mn-12-acetate [20] $(Mn_{12}) - [Mn_{12}O_{12}(CH_3COO)_{16}(H_2O)_4] -$ which by now serves as the "drosophila" of molecular magnetism, see e. g. [1, 4, 21–23]. As shown in Fig. 3.1 the molecules contains four Mn(IV) ions (s = 3/2) and eight Mn(III) ions (s = 2) which are magnetically coupled to give an S = 10 ground state. The molecules possesses a magnetic anisotropy, which determines the observed relaxation of the magnetization and quantum tunneling at low temperatures [21, 24].

Although the investigation of magnetic molecules in general – and of Mn-12-acetate in particular – has made great advances over the past two decades, it is still a challenge to deduce the underlying microscopic Hamiltonian, even if the Hamiltonian is of Heisenberg type. Mn-12-acetate is known for about 20 years now and investigated like no other magnetic molecule, but only recently its model parameters could be estimated with satisfying accuracy [25, 26].

Another very well investigated class of molecules is given by spin rings among which iron rings ("ferric wheels") are most popular [27–34]. Iron-6 rings for instance can host alkali ions such as lithium or sodium which allows to modify the parameters of the spin Hamiltonian within some range [16,35]. Another realization of rings is possible using chromium ions as paramagnetic



Fig. 3.1. Structure of Mn-12-acetate: On the top the Mn ions are depicted by large spheres, on the bottom the dominant couplings are given. With friendly permission by G. Chaboussant



Fig. 3.2. Structure of a chromium-4 and a chromium-8 ring. The Cr ions are depicted by large spheres

centers. Figure 3.2 shows the structure of two rings, one with four chromium ions the other one with eight chromium ions.

A new route to molecular magnetism is based on so-called Keplerate structures which allow the synthesis of truly giant and highly symmetric spin arrays. The molecule $\{Mo_{72}Fe_{30}\}$ [7,36] containing 30 iron ions of spin s = 5/2may be regarded as the archetype of such structures. Figure 3.3 shows on the l.h.s. the inner skeleton of this molecule – Fe and O-Mo-O bridges – as well as the classical ground state [37] depicted by arrows on the r.h.s. [36].



Fig. 3.3. Structure of $\{Mo_{72}Fe_{30}\}$, a giant Keplerate molecule where 30 iron ions are placed at the vertices of an icosidodecahedron. L.h.s.: sketch of the chemical structure, r.h.s. magnetic structure showing the iron ions (spheres), the nearest neighbor interactions (edges) as well as the spin directions in the classical ground state. The dashed triangle on the l.h.s. corresponds to the respective triangle on the r.h.s.. With friendly permission by Paul Kögerler [36]



Fig. 3.4. Square lattice of $\{Mo_{72}Fe_{30}\}$ -molecules: Each molecule is connected with its four nearest neighbors by an antiferromagnetic coupling. With friendly permission by Paul Kögerler [38,39]

One of the obvious advantages of magnetic molecules is that the magnetic centers of different molecules are well separated by the ligands of the molecules. Therefore, the intermolecular interactions are utterly negligible and magnetic molecules can be considered as being independent. Nevertheless, it is desirable to build up nanostructured materials consisting of magnetic molecules in a controlled way. Figure 3.4 gives an example of a planar structure consisting of layers of $\{Mo_{72}Fe_{30}\}$ [38,39] which has been synthesized recently together with a linear structure consisting of chains of $\{Mo_{72}Fe_{30}\}$ [40]. These

systems show new combinations of physical properties that stem from both molecular and bulk effects.

Many more structures than those sketched above can be synthesized nowadays and with the increasing success of coordination chemistry more are yet to come. The final hope of course is that magnetic structures can be designed according to the desired magnetic properties. But this goal is not close at all, it requires further understanding of the interplay of magneto-chemistry and magnetic phenomena. One of the tools used to clarify such questions is density functional theory or other *ab initio* methods [41–46].

3.3 Experimental Work

3.3.1 Experimental Methods

The properties of magnetic molecules are investigated with a variety of wellknown techniques. Standard investigations include the determination of atomic positions of crystallized substances by means of x-ray spectroscopy as well as the determination of the zero-field (weak-field) magnetic susceptibility. The latter method is often used to obtain a first estimate of the involved exchange parameters analyzing the high temperature behavior of the susceptibility, see e. g. [47].

If the exchange parameters turn out to be of the order of some (ten) Kelvin or less it is illuminative to perform high-field low-temperature magnetization measurements for instance with pulsed fields that can extend up to 60 Tesla, see e. g. [36,48]. The measured low-temperature magnetization curve exhibits a step structure which reveals additional and often more accurate information about the exchange parameters and the Landé g-factor. Alternatively torque magnetometry [16, 33] and micro squid techniques [49, 50] are used. These methods are as well applied in slowly-varying fields where magnetic molecules may behave as single-molecule magnets and show a noticeable hysteresis.

Resonance techniques are exploited in order to determine the gap structure of the energy spectrum of a magnetic molecule subject to a magnetic field. Electron Spin/Parametric Resonance (ESR/EPR) [12] measures in the GHz region whereas Nuclear Magnetic Resonance (NMR) measures in the MHz region and therefore the latter one is often used to determine level crossings [22, 23, 29, 30, 51]. Sometimes ESR is replaced by μ SR, a method where a muon is implanted in the sample in order the have a rather local probe [52, 53].

Neutron scattering [54] can also be considered as a resonance technique. It also probes the gap structure of the energy spectrum at a given field [16,55,56].

Not so often applied are Mössbauer [57] and photoemmission spectroscopy [58, 59] as well as calorimetric methods [60], since the latter suffers from phonon contributions.

3.3.2 Phenomena of Current Interest

Besides the desire to understand magnetism on a molecular scale and the structure and origin of the appropriate spin Hamiltonian some investigated phenomena are of very general interest for a broad community.

From the point of view of further applications the exciting fact, that some magnetic molecules possess a large ground state spin, e. g. S = 10 for Mn_{12} and Fe₈ as well as a sufficient anisotropy barrier, attracts very much attention. Such molecules show a pronounced hysteresis and are thus called single-molecule magnets (SMM) [1]. Nevertheless, an initial magnetization is not stable in molecular magnets but can tunnel through the anisotropy barrier which in this context is called quantum tunneling of the magnetic moment [21, 24, 61–64]. The tunnel rates especially for the low-lying magnetic levels, with e. g. $M = \pm 10$, are very small, but can be enhanced if the system is first excited to higher-lying levels with smaller magnetic moment. Depending on the mechanism used these processes are termed thermally assisted tunneling [65] or phonon assisted tunneling [66].

Another very promising effect is the so called "Light Induced Excited Spin State Trapping" (LIESST) [11] which can be observed in spin crossover substances. Such substances are characterized by a low-spin ground state and a higher-lying high-spin state which is meta stable. It is then possible to switch between these states by means of light irradiation, temperature, or pressure. The meachanism can be understood as follows. Spin-crossover molecules can be modeled with a one-dimensional effective oscillator Hamiltonian where the oscillator potential parametrically depends on total spin, for instance of the six-fold coordinated iron in the center of the molecule. It is then possible to switch the molecules from the low-spin ground state via an intermediate state to a high-spin state by irradiation with laser light of an appropriate frequency. The magnetic transition is accompanied by a structural transition, i. e. the molecule changes its size etc. and consequently some of its properties like color. This process is reversible, i. e. with a different frequency the molecule can be switched back to the original low-spin state. Due to the very different density of states of the two spin configurations the switching can also be performed thermally or even by applying pressure.

A rather young field is the investigation of low-dimensional structures built from interacting magnetic molecules. These structures may grow "naturally" while the substance crystallizes [67, 68] or "on purpose" driven by chemical reactions which interlink neighboring molecules [38–40]. The additional interaction may lead to additional features of the substance for instance to an ordering of the total spins of the molecules at low temperatures. The disappearance of such an order between moments of magnetic molecules expresses itself as a phase transition and is visible for instance in the specific heat and susceptibility data [68].

3.4 Theoretical Techniques and Results

3.4.1 Hamiltonian

It appears that in the majority of these molecules the interaction between the localized single-particle magnetic moments can be rather well described by the Heisenberg model with isotropic (nearest neighbor) interaction and an additional anisotropy term [12–16]. Dipolar interactions are usually of minor importance. It is also found that antiferromagnetic interactions are favored in most molecules leading to nontrivial ground states.

Heisenberg Hamiltonian

For many magnetic molecules the total Hamilton operator can be written as

$$H_{\sim} = H_{\rm Heisenberg} + H_{\rm anisotropy} + H_{\rm Zeeman}$$
(3.1)

$$H_{\sim}_{\text{Heisenberg}} = -\sum_{u,v} J_{uv} \underline{s}(u) \cdot \underline{s}(v)$$
(3.2)

$$\mathcal{H}_{\text{anisotropy}} = -\sum_{u=1}^{N} d_u (\boldsymbol{e}(u) \cdot \boldsymbol{s}(u))^2$$
(3.3)

$$H_{\text{Zeeman}} = g\mu_B \boldsymbol{B} \cdot \boldsymbol{S} . \tag{3.4}$$

The Heisenberg Hamilton operator¹ in the form given in (3.2) is isotropic, J_{uv} is a symmetric matrix containing the exchange parameters between spins at sites u and v. The exchange parameters are usually given in units of energy, and $J_{uv} < 0$ corresponds to antiferromagnetic, $J_{uv} > 0$ to ferromagnetic coupling². The sum in (3.2) runs over all possible tuples (u, v). The vector operators $\mathbf{s}(u)$ are the single-particle spin operators.

The anisotropy terms (3.3) usually simplify to a large extend, for instance for spin rings, where the site-dependent directions e(u) are all equal, e. g. $e(u) = e_z$ and the strength as well is the same for all sites $d_u = d$.

The third part (Zeeman term) in the full Hamiltonian describes the interaction with the external magnetic field. Without singe-site and g-value anisotropy the direction of the field can be assumed to be along the z-axis which simplifies the Hamiltonian very much.

Although the Hamiltonian looks rather simple, the eigenvalue problem is very often not solvable due to the huge dimension of the Hilbert space or because the number of exchange constants is too big to allow an accurate determination from experimental data. Therefore, one falls back to effective single-spin Hamiltonians for molecules with non-zero ground state spin and a large enough gap to higher-lying multiplets.

¹ Operators are denoted by a tilde.

 $^{^{2}}$ One has to be careful with this definition since it varies from author to author

Single-Spin Hamiltonian

For molecules like Mn_{12} and Fe_8 which possess a high ground state spin and well separated higher lying levels the following single-spin Hamiltonian

$$H_{\widetilde{\omega}} = -D_2 S_z^2 - D_4 S_z^4 + H' \tag{3.5}$$

$$H' = g\mu_B B_x S_x \tag{3.6}$$

is appropriate, see e. g. [23]. The first two terms of the Hamilton operator $\underset{\sim}{H}$ represent the anisotropy whereas $\underset{\sim}{H'}$ is the Zeeman term for a magnetic field along the *x*-axis. The total spin is fixed, i. e. S = 10 for Mn₁₂ and Fe₈, thus the dimension of the Hilbert space is dim $(\mathcal{H}) = 2S + 1$.

The effective Hamiltonian (3.5) is sufficient to describe the low-lying spectrum and phenomena like magnetization tunneling. Since $\underline{H'}$ does not commute with the z-component of the total spin \underline{S}_z , every eigenstate $|M\rangle$ of \underline{S}_z , i. e. the states with good magnetic quantum number M, is not stationary but will tunnel through the barrier and after half the period be transformed into $|-M\rangle$.

3.4.2 Evaluating the Spectrum

The ultimate goal is to evaluate the complete eigenvalue spectrum of the full Hamilton operator (3.1) as well as all eigenvectors, compare also the chapter by Laflorencie and Poilblanc. Since the total dimension of the Hilbert space is usually very large, e. g. $\dim(\mathcal{H}) = (2s+1)^N$ for a system of N spins of equal spin quantum number s, a straightforward diagonalization of the full Hamilton matrix is not feasible. Nevertheless, very often the Hamilton matrix can be decomposed into a block structure because of spin symmetries or space symmetries. Accordingly the Hilbert space can be decomposed into mutually orthogonal subspaces. Then for a practical evaluation only the size of the largest matrix to be diagonalized is of importance (relevant dimension).

Product Basis

The starting point for any diagonalization is the product basis $|\mathbf{m}\rangle = |m_1, \ldots, m_u, \ldots, m_N\rangle$ of the single-particle eigenstates of all $s_z(u)$

$$\underbrace{s}_{z}(u) \mid m_{1}, \dots, m_{u}, \dots, m_{N} \rangle = m_{u} \mid m_{1}, \dots, m_{u}, \dots, m_{N} \rangle .$$
(3.7)

These states are sometimes called Ising states. They span the full Hilbert space and are used to construct symmetry-related basis states.

Symmetries of the Problem

Since the isotropic Heisenberg Hamiltonian includes only a scalar product between spins, this operator is rotationally invariant in spin space, i. e. it commutes with S and thus also with S_z

$$\begin{bmatrix} H_{\text{Heisenberg}}, \mathbf{S}^2 \end{bmatrix} = 0 \quad , \qquad \begin{bmatrix} H_{\text{Heisenberg}}, \mathbf{S}_z \end{bmatrix} = 0 \quad . \tag{3.8}$$

In a case where anisotropy is negligible a well-adapted basis is thus given by the simultaneous eigenstates $|S, M, \alpha\rangle$ of \mathfrak{S}^2 and \mathfrak{S}_z , where α enumerates those states belonging to the same S and M [69,70]. Since the applied magnetic field can be assumed to point into z-direction for vanishing anisotropy the Zeeman term automatically also commutes with $\mathcal{H}_{\text{Heisenberg}}, \mathfrak{S}^2$, and \mathfrak{S}_z . Since M is a good quantum number the Zeeman term does not need to be included in the diagonalization but can be added later.

Besides spin symmetries many molecules possess spatial symmetries. One example is given by spin rings which have a translational symmetry. In general the symmetries depend on the point group of the molecule; for the evaluation of the eigenvalue spectrum its irreducible representations have to be used [13, 16,69]. Thus, in a case with anisotropy one looses spin rotational symmetries but one can still use space symmetries. Without anisotropy one even gains a further reduction of the relevant dimension.

Dimension of the Problem

The following section illuminates the relevant dimensions assuming certain symmetries³.

If no symmetry is present the total dimension is just

dim
$$(\mathcal{H}) = \prod_{u=1}^{N} (2s(u) + 1)$$
 (3.9)

for a spin array of N spins with various spin quantum numbers. In many cases the spin quantum numbers are equal resulting in a dimension of the total Hilbert space of $\dim(\mathcal{H}) = (2s+1)^N$.

If the Hamiltonian commutes with S_z then M is a good quantum number and the Hilbert space \mathcal{H} can be divided into mutually orthogonal subspaces $\mathcal{H}(M)$

$$\mathcal{H} = \bigoplus_{M=-S_{\max}}^{+S_{\max}} \mathcal{H}(M) , \quad S_{\max} = \sum_{u=1}^{N} s(u) .$$
 (3.10)

³ Work done with Klaus Bärwinkel and Heinz-Jürgen Schmidt, Universität Osnabrück, Germany.

For given values of M, N and of all s(u) the dimension dim $(\mathcal{H}(M))$ can be determined as the number of product states (3.7), which constitute a basis in $\mathcal{H}(M)$, with $\sum_{u} m_{u} = M$. The solution of this combinatorial problem can be given in closed form [70]

$$\dim\left(\mathcal{H}(M)\right) = \frac{1}{(S_{\max} - M)!} \left[\left(\frac{d}{dz}\right)^{S_{\max} - M} \prod_{x=1}^{N} \frac{1 - z^{2s(x) + 1}}{1 - z} \right]_{z=0} (3.11)$$

For equal single-spin quantum numbers $s(1) = \cdots = s(N) = s$, and thus a maximum total spin quantum number of $S_{\max} = Ns$, (3.11) simplifies to

dim
$$(\mathcal{H}(M)) = f(N, 2s + 1, S_{\max} - M)$$
 with (3.12)

$$f(N, \mu, \nu) = \sum_{n=0}^{\lfloor \nu/\mu \rfloor} (-1)^n \binom{N}{n} \binom{N-1+\nu-n\mu}{N-1}.$$

In both formulae (3.11) and (3.12), M may be replaced by |M| since the dimension of $\mathcal{H}(M)$ equals those of $\mathcal{H}(-M)$. $\lfloor \nu/\mu \rfloor$ in the sum symbolizes the greatest integer less or equal to ν/μ . Equation (3.12) is known as a result of de Moivre [71].

If the Hamiltonian commutes with \underline{S}^2 and all individual spins are identical the dimensions of the orthogonal eigenspaces $\mathcal{H}(S, M)$ can also be determined. The simultaneous eigenspaces $\mathcal{H}(S, M)$ of \underline{S}^2 and \underline{S}_z are spanned by eigenvectors of \underline{H} . The one-dimensional subspace $\mathcal{H}(M = S_{\max}) = \mathcal{H}(S_{\max}, S_{\max})$, especially, is spanned by $|\Omega\rangle$, a state called magnon vacuum. The total ladder operators (spin rising and lowering operators) are

$$\underline{S}^{\pm} = \underline{S}_x \pm i \,\underline{S}_y \,\,. \tag{3.13}$$

For S > M, \underline{S}^- maps any normalized $\underline{\mathcal{H}}$ -eigenstate $\in \mathcal{H}(S, M + 1)$ onto an $\underline{\mathcal{H}}$ -eigenstate $\in \mathcal{H}(S, M)$ with norm $\sqrt{S(S+1) - M(M+1)}$.

For $0 \leq M < S_{\max}$, $\mathcal{H}(M)$ can be decomposed into orthogonal subspaces

$$\mathcal{H}(M) = \mathcal{H}(M, M) \oplus \underline{S}^{-} \mathcal{H}(M+1)$$
(3.14)

with

$$S_{\sim}^{-}\mathcal{H}(M+1) = \bigoplus_{S \ge M+1} \mathcal{H}(S,M) .$$
(3.15)

In consequence, the diagonalization of H in \mathcal{H} has now been traced back to diagonalization in the subspaces $\mathcal{H}(S,S)$, the dimension of which are for $S < S_{\max}$

$$\dim \left(\mathcal{H}(S,S)\right) = \dim \left(\mathcal{H}(M=S)\right) - \dim \left(\mathcal{H}(M=S+1)\right)$$
(3.16)

and can be calculated according to (3.12).

As an example for space symmetries I would like to discuss the translational symmetry found in spin rings. The discussed formalism can as well be applied to other symmetry operations which can be mapped onto a translation. Any such translation is represented by the cyclic shift operator T or a multiple repetition. T_{i} is defined by its action on the product basis (3.7)

$$T_{\sim} | m_1, \dots, m_{N-1}, m_N \rangle = | m_N, m_1, \dots, m_{N-1} \rangle .$$

$$(3.17)$$

The eigenvalues of \underline{T} are the N-th roots of unity

$$z_k = \exp\left\{-i\frac{2\pi k}{N}\right\}$$
, $k = 0, \dots, N-1$, $p_k = 2\pi k/N$, (3.18)

where k will be called translational (or shift) quantum number and p_k momentum quantum number or crystal momentum. The shift operator \underline{T} commutes not only with the Hamiltonian but also with total spin. Any $\mathcal{H}(S, M)$ can therefore be decomposed into simultaneous eigenspaces $\mathcal{H}(S, M, k)$ of \underline{S}^2 , \underline{S}_z and \underline{T} .

In the following we demonstrate how an eigenbasis of both S_z and T_{\sim} can be constructed, this basis spans the orthogonal Hilbert spaces $\mathcal{H}(M, k)$. How total spin can be included by means of an irreducible tensor operator approach is described in Refs. [13, 16, 69].

A special decomposition of \mathcal{H} into orthogonal subspaces can be achieved by starting with the product basis and considering the equivalence relation

$$|\psi\rangle \cong |\phi\rangle \Leftrightarrow |\psi\rangle = \underline{T}^n |\phi\rangle, \ n \in \{1, 2, \dots, N\}$$
(3.19)

for any pair of states belonging to the product basis. The equivalence relation then induces a complete decomposition of the basis into disjoint subsets, i. e. the equivalence classes. A "cycle" is defined as the linear span of such an equivalence class of basis vectors. The obviously orthogonal decomposition of \mathcal{H} into cycles is compatible with the decomposition of \mathcal{H} into the various $\mathcal{H}(M)$. Evidently, the dimension of a cycle can never exceed N. Cycles are called "proper cycles" if their dimension equals N, they are termed "epicycles" else. One of the N primary basis states of a proper cycle may arbitrarily be denoted as

$$|\psi_1\rangle = |m_1, \dots, m_N\rangle \tag{3.20}$$

and the remaining ones may be enumerated as

$$|\psi_{n+1}\rangle = \sum_{n=1}^{n} |\psi_1\rangle, \ n = 1, 2, \dots, N-1.$$
 (3.21)

The cycle under consideration is likewise spanned by the states

$$|\chi_k\rangle = \frac{1}{\sqrt{N}} \sum_{\nu=0}^{N-1} \left(e^{i\frac{2\pi k}{N}} T \right)^{\nu} |\psi_1\rangle$$
(3.22)

which are eigenstates of \underline{T} with the respective shift quantum number k. Consequently, every k occurs once in a proper cycle. An epicycle of dimension D is spanned by D eigenstates of \underline{T} with each of the translational quantum numbers $k = 0, N/D, \ldots, (D-1)N/D$ occurring exactly once.

As a rule of thumb one can say that the dimension of each $\mathcal{H}(M, k)$ is approximately $\dim(\mathcal{H}(M, k)) \approx \dim(\mathcal{H}(M))/N$. An exact evaluation of the relevant dimensions for spin rings can be obtained from [70].

Exact Diagonalization

If the relevant dimension is small enough the respective Hamilton matrices can be diagonalized, either analytically [70, 72, 73] or numerically, see e. g. [13, 69, 74–79], and the chapter by Laflorencie and Poilblanc.

Again, how such a project is carried out, will be explained with the help of an example, a simple spin ring with N = 6 and s = 1/2. The total dimension is dim $(\mathcal{H}) = (2s+1)^N = 64$. The Hamilton operator (3.2) simplifies to

$$\underset{\sim}{H}_{\text{Heisenberg}} = -2J \sum_{u=1}^{N} \underline{s}(u) \cdot \underline{s}(u+1) , \quad N+1 \equiv 1 .$$
 (3.23)

We start with the magnon vacuum $|\Omega\rangle = |+++++\rangle$ which spans the Hilbert space $\mathcal{H}(M)$ with M = Ns = 3. "±" are shorthand notations for $m = \pm 1/2$. The dimension of the subspace dim $(\mathcal{H}(M = Ns))$ is one and the energy eigenvalue is $E_{\Omega} = -2JNs^2 = -3J$. $|\Omega\rangle$ is an eigenstate of the shift operator with k = 0. Since S is also a good quantum number in this example $|\Omega\rangle$ has to be an eigenstate of S^2 , too, the quantum number is S = Ns.

The next subspace $\mathcal{H}(M)$ with M = Ns - 1 = 2 is spanned by $|-++++\rangle$ and the five other vectors which are obtained by repetitive application of \mathcal{T} . This subspace obviously has the dimension N, and the cycle spanned by $\mathcal{T}^n |-++++\rangle$, $n = 0, \ldots, N-1$ is a proper one. Therefore, each k quantum number arises once. The respective eigenstates of \mathcal{T} can be constructed according to (3.22) as

$$|M = 2, k\rangle = \frac{1}{\sqrt{N}} \sum_{\nu=0}^{N-1} \left(e^{i\frac{2\pi k}{N}} T \right)^{\nu} |-++++\rangle .$$
 (3.24)

All subspaces $\mathcal{H}(M, k)$ have dimension one. Since $S^{-} | \Omega \rangle$ is a state belonging to $\mathcal{H}(M = Ns - 1)$ with the same k-quantum number as $| \Omega \rangle$ it is clear that

 $|M = 2, k = 0\rangle$ is a already an eigenstate of S^2 with S = Ns. The other $|M = 2, k \neq 0\rangle$ must have S = Ns - 1.

The next subspace $\mathcal{H}(M)$ with M = Ns - 2 = 1 is spanned by three basic vectors, i. e. $|--++++\rangle$, $|-+-+++\rangle$, $|-++-++\rangle$ and the repetitive applications of T onto them. The first two result in proper cycles, the third vector $|-++-++\rangle$ results in an epicycle of dimension three, thus for the epicycle we find only k quantum numbers k = 0, 2, 4. The energy eigenvalues found in the subspace $\mathcal{H}(M = Ns - 1)$ ("above") must reappear here which again allows to address an S quantum number to these eigenvalues. The dimension of the subspace $\mathcal{H}(M = 1)$ is 15, the dimensions of the subspaces $\mathcal{H}(M, k)$ are 3 (k = 0), 2 (k = 1), 3 (k = 2), 2 (k = 3), 3 (k = 4), and 2 (k = 5).

The last subspace which has to be considered belongs to M = 0 and is spanned by $|--+++\rangle$, $|-+++\rangle$, $|-+-++\rangle$, $|-++-+\rangle$ and repetitive applications of T. Its dimension is 20. Here $|-+-++\rangle$ leads to an epicycle of dimension two.

The Hamilton matrices in subspaces with M < 0 need not to be diagonalized due to the S_z -symmetry, i. e. eigenstates with negative M can be obtained by transforming all individual $m_u \to -m_u$. Summing up the dimensions of all $\mathcal{H}(M)$ then yields $1 + 6 + 15 + 20 + 15 + 6 + 1 = 64 \sqrt{.}$

Figure 3.5 shows the resulting energy spectrum both as a function of total spin S as well as a function of translational quantum number k.



Fig. 3.5. Energy eigenvalues as a function of total spin quantum number S (l.h.s.) and k (r.h.s.)

Projection and Lanczos Method

Complex hermitian matrices can be completely diagonalized numerically up to a size of about 10,000 by 10,000 which corresponds to about 1.5 Gigabyte of necessary RAM. Nevertheless, for larger systems one can still use numerical methods to evaluate low-lying energy levels and the respective eigenstates with high accuracy. A simple method is the projection method [77] which rests on the multiple application of the Hamiltonian on some random trial state.

To be more specific let's approximate the ground state of a spin system. We start with a random trial state $|\phi_0\rangle$ and apply an operator which "cools" the system. This operator is given by the time evolution operator with imaginary time steps

$$|\tilde{\phi}_1\rangle = \exp\left\{-\varepsilon \underline{H}\right\} |\phi_0\rangle . \qquad (3.25)$$

Expanding $\,|\,\phi_0\,\rangle$ into eigenstates $\,|\,\nu\,\rangle$ of the Hamilton operator elucidates how the method works

$$|\tilde{\phi}_{1}\rangle = \sum_{\nu=0} \exp\left\{-\varepsilon E_{\nu}\right\} |\nu\rangle \langle\nu|\phi_{0}\rangle$$
(3.26)

$$= \exp\left\{-\varepsilon E_0\right\} \sum_{\nu=0} \exp\left\{-\varepsilon (E_{\nu} - E_0)\right\} |\nu\rangle \langle \nu | \phi_0\rangle . \quad (3.27)$$

Relative to the ground state component all other components are exponentially suppressed. For practical purposes equation (3.26) is linearized and recursively used

$$|\tilde{\phi}_{i+1}\rangle = \left(1 - \varepsilon H\right) |\phi_i\rangle , \quad |\phi_{i+1}\rangle = \frac{|\dot{\phi}_{i+1}\rangle}{\sqrt{\langle \tilde{\phi}_{i+1} | \tilde{\phi}_{i+1}\rangle}} .$$
(3.28)

 ε has to be small enough in order to allow the linearization of the exponential. It is no problem to evaluate several higher-lying states by demanding that they have to be orthogonal to the previous ones. Restricting the calculation to orthogonal eigenspaces yields low-lying states in these eigenspaces which allows to evaluate even more energy levels. The resulting states obey the properties of the Ritz variational principle, i. e. they lie above the ground state and below the highest one.

Another method to partially diagonalize a huge matrix was proposed by Cornelius Lanczos in 1950 [80,81]. Also this method uses a (random) initial vector. It then generates an orthonormal system in such a way that the representation of the operator of interest is tridiagonal. Every iteration produces a new tridiagonal matrix which is by one row and one column bigger than the previous one. With growing size of the matrix its eigenvalues converge against the true ones until, in the case of finite dimensional Hilbert spaces, the eigenvalues reach their true values. The key point is that the extremal eigenvalues converge rather quickly compared to the other ones [82]. Thus it might be that after 300 Lanczos steps the ground state energy is already approximated to 10 figures although the dimension of the underlying Hilbert space is 10^8 .

A simple Lanczos algorithm looks like the following. One starts with an arbitrary vector $|\psi_0\rangle$, which has to have an overlap with the (unknown)
ground state. The next orthogonal vector is constructed by application of $\underset{\sim}{H}$ and projecting out the original vector $|\psi_0\rangle$

$$|\psi_{1}'\rangle = (1 - |\psi_{0}\rangle\langle\psi_{0}|) \underbrace{H}_{\approx} |\psi_{0}\rangle = \underbrace{H}_{\approx} |\psi_{0}\rangle - \langle\psi_{0}|\underbrace{H}_{\approx} |\psi_{0}\rangle |\psi_{0}\rangle, \quad (3.29)$$

which yields the normalized vector

$$|\psi_1\rangle = \frac{|\psi_1'\rangle}{\sqrt{\langle\psi_1'|\psi_1'\rangle}} . \tag{3.30}$$

Similarly all further basis vectors are generated

$$|\psi_{k+1}'\rangle = (1 - |\psi_k\rangle\langle\psi_k| - |\psi_{k-1}\rangle\langle\psi_{k-1}|) \underset{\sim}{H} |\psi_k\rangle$$

$$= \underset{\sim}{H} |\psi_k\rangle - \langle\psi_k|H|\psi_k\rangle|\psi_k\rangle - \langle\psi_{k-1}|\underset{\sim}{H} |\psi_k\rangle|\psi_{k-1}\rangle$$

$$(3.31)$$

and

$$|\psi_{k+1}\rangle = \frac{|\psi'_{k+1}\rangle}{\sqrt{\langle\psi'_{k+1}|\psi'_{k+1}\rangle}}$$
 (3.32)

The new Lanczos vector is by construction orthogonal to the two previous ones. Without proof we repeat that it is then also orthogonal to all other previous Lanczos vectors. This constitutes the tridiagonal form of the resulting Hamilton matrix

$$T_{i,j} = \langle \psi_i | \underset{\sim}{H} | \psi_j \rangle \quad \text{with} \quad T_{i,j} = 0 \quad \text{if} \quad |i-j| > 1 .$$
(3.33)

The Lanczos matrix T can be diagonalized at any step. Usually one iterates the method until a certain convergence criterion is fulfilled.

The eigenvectors of $\underset{\sim}{H}$ can be approximated using the eigenvectors $\mid \phi_{\mu} \, \rangle$ of T

$$|\chi_{\mu}\rangle \approx \sum_{i=0}^{n} \langle \psi_{i} | \phi_{\mu} \rangle | \psi_{i} \rangle , \qquad (3.34)$$

where μ labels the desired energy eigenvalue, e. g. the ground state energy. *n* denotes the number of iterations.

The simple Lanczos algorithm has some problems due to limited accuracy. One problem is that eigenvalues may collapse. Such problems can be solved with more refined formulations of the method [81].

DMRG

The DMRG technique [83] has become one of the standard numerical methods for quantum lattice calculations in recent years [84,85]. Its basic idea is the reduction of Hilbert space while focusing on the accuracy of a target state. For this purpose the system is divided into subunits – blocks – which are represented by reduced sets of basis states. The dimension m of the truncated block Hilbert space is a major input parameter of the method and to a large extent determines its accuracy.

DMRG is best suited for chain-like structures. Many accurate results have been achieved by applying DMRG to various (quasi-)one-dimensional systems [78, 86, 87]. The best results were found for the limit of infinite chains with open boundary conditions. It is commonly accepted that DMRG reaches maximum accuracy when it is applied to systems with a small number of interactions between the blocks, e. g. systems with only nearest-neighbor interaction [84].

It is not a priori clear how good results for finite systems like magnetic molecules are⁴. Such systems are usually not chain-like, so in order to carry out DMRG calculations a mapping onto a one-dimensional structure has to be performed [84]. Since the spin array consists of a countable number of spins, any arbitrary numbering is already a mapping onto a one-dimensional structure. However, even if the original system had only nearest-neighbor exchange, the new one-dimensional system has many long-range interactions depending on the way the spins are enumerated. Therefore, a numbering which minimizes long range interactions is preferable. Figure 3.6 shows the graph of interactions for the molecule $\{Mo_{72}Fe_{30}\}$ which we want to consider as an example in the following [88].



Fig. 3.6. One-dimensional projection of the icosidodecahedron: the lines represent interactions

For finite systems a block algorithm including sweeps, which is similar to the setup in White's original article [83], has turned out to be most efficient. Two blocks are connected via two single spin sites, these four parts form the superblock see Fig. 3.7.



Fig. 3.7. Block setup for DMRG "sweep" algorithm: The whole system of N spins constitutes the superblock. The spins $\{1, 2, ..., p\}$ belong to the left block, the other spins $\{p + 1, ..., N\}$ to the right block

⁴ Work done with Matthias Exler, Universität Osnabrück, Germany.

For illustrative purposes we use a simple Heisenberg Hamiltonian, compare (3.2). The Hamiltonian is invariant under rotations in spin space. Therefore, the total magnetic quantum number M is a good quantum number and we can perform our calculation in each orthogonal subspace $\mathcal{H}(M)$ separately.

Since it is difficult to predict the accuracy of a DMRG calculation, it is applied to an exactly diagonalizable system first. The most realistic test system for the use of DMRG for $\{Mo_{72}Fe_{30}\}$ is the icosidodecahedron with spins s = 1/2. This fictitious molecule, which possibly may be synthesized with vanadium ions instead of iron ions, has the same structure as $\{Mo_{72}Fe_{30}\}$, but the smaller spin quantum number reduces the dimension of the Hilbert space significantly. Therefore a numerically exact determination of low-lying levels using a Lanczos method is possible [89]. These results are used to analyze the principle feasibility and the accuracy of the method.

The DMRG calculations were implemented using the enumeration of the spin sites as shown in Figs. 3.6 and 3.8. This enumeration minimizes the average interaction length between two sites.



Fig. 3.8. Two-dimensional projection of the icosidodecahedron, the site numbers are those used in our DMRG algorithm

In Fig. 3.9 the DMRG results (crosses) are compared to the energy eigenvalues (circles) determined numerically with a Lanczos method [88, 89]. Very good agreement of both sequences, with a maximal relative error of less than 1% is found. Although the high accuracy of one-dimensional calculations (often with a relative error of the order of 10^{-6}) is not achieved, the result demonstrates that DMRG is applicable to finite 2D spin systems. Unfortunately, increasing *m* yields only a weak convergence of the relative error, which is defined relative to the width of the spectrum

$$\epsilon(m) = \frac{E_{\rm DMRG}(m) - E_0}{|E_0^{\rm AF} - E_0^{\rm F}|} .$$
(3.35)

The dependence for a quasi two-dimensional structure like the icosidodecahedron is approximately proportional to 1/m (see Fig. 3.10). Unfortunately,



Fig. 3.9. Minimal energy eigenvalues of the s = 1/2 icosidodecahedron. The DMRG result with m = 60 is depicted by crosses, the Lanczos values by circles. The rotational band is discussed in subsection 3.4.3



Fig. 3.10. Dependence of the approximate ground state energy on the DMRG parameter m. E_0 is the true ground state energy in the case s = 1/2 and the extrapolated one for s = 5/2

such weak convergence is characteristic for two-dimensional systems in contrast to one-dimensional chain structures, where the relative error of the approximate energy was reported to decay exponentially with m [83]. Nevertheless, the extrapolated ground state energy for s = 1/2 deviates only by $\epsilon = 0.7$ % from the ground state energy determined with a Lanczos algorithm.

The major result of the presented investigation is that the DMRG approach delivers acceptable results for finite systems like magnetic molecules. Nevertheless, the accuracy known from one-dimensional systems is not reached.

Spin-Coherent States

Spin-coherent states [90] provide another means to either treat a spin system exactly and investigate for instance its dynamics [91] or to use spin coherent states in order to approximate the low-lying part of the spectrum. They are also used in connection with path integral methods. In the following the basic ideas and formulae will be presented.

The obvious advantage of spin-coherent states is that they provide a bridge between classical spin dynamics and quantum spin dynamics. Spin coherent states are very intuitive since they parameterize a quantum state by the expectation value of the spin operator, e. g. by the two angles which represent the spin direction.

Spin coherent states $|z\rangle$ are defined as

$$|z\rangle = \frac{1}{(1+|z|^2)^s} \sum_{p=0}^{2s} \sqrt{\binom{2s}{p}} z^p |s,m=s-p\rangle , \quad z \in \mathbb{C} . \quad (3.36)$$

In this definition spin-coherent states are characterized by the spin length s and a complex value z. The states (3.36) are normalized but not orthogonal

$$\langle z \, | \, z \, \rangle = 1 \, , \quad \langle y \, | \, z \, \rangle = \frac{(1 + y^* z)^{2s}}{(1 + |y|^2)^s (1 + |z|^2)^s} \, .$$

Spin-coherent states provide a basis in single-spin Hilbert space, but they form an overcomplete set of states. Their completeness relation reads

$$1_{\sim} = \frac{2s+1}{\pi} \int d^2 z \frac{|z\rangle\langle z|}{(1+|z|^2)^2} , \quad d^2 z = d\text{Re}(z) \, d\text{Im}(z) .$$
(3.37)

The intuitive picture of spin-coherent states becomes obvious if one transforms the complex number z into angles on a Riemann sphere

$$z = \tan(\theta/2)e^{i\phi}$$
, $0 \le \theta < \pi$, $0 \le \phi < 2\pi$. (3.38)

Thus, spin-coherent states may equally well be represented by two polar angles θ and ϕ . Then the expectation value of the spin operator s is simply

$$\langle \theta, \phi | \underset{\sim}{\boldsymbol{s}} | \theta, \phi \rangle = s \begin{pmatrix} \sin(\theta) \cos(\phi) \\ \sin(\theta) \sin(\phi) \\ \cos(\theta) \end{pmatrix} .$$
(3.39)

Using (3.38) the definition of the states $|\theta, \phi\rangle$ which is equivalent to (3.36) is then given by

$$|\theta,\phi\rangle = \sum_{p=0}^{2s} \sqrt{\binom{2s}{p}} \left[\cos(\theta/2)\right]^{(2s-p)} \left[e^{i\phi}\sin(\theta/2)\right]^p |s,m=s-p \,(3.40)$$

and the completeness relation simplifies to

$$\mathbb{1}_{\sim} = \frac{2s+1}{4\pi} \int \mathrm{d}\Omega \ |\,\theta,\phi\,\rangle\langle\,\theta,\phi\,| \ . \tag{3.41}$$

Product states of spin-coherent states span the many-spin Hilbert space. A classical ground state can easily be translated into a many-body spin-coherent state. One may hope that this state together with other product states can provide a useful set of linearly independent states in order to approximate low-lying states of systems which are too big to handle otherwise. But it is too early to judge the quality of such approximations.

3.4.3 Properties of Spectra

33excitation spectrum!spinningIn the following chapter I am discussing some properties of the spectra of magnetic molecules with isotropic and antiferromagnetic interaction.

Non-bipartite Spin Rings

With the advent of magnetic molecules it appears to be possible to synthesize spin rings with an odd number of spins. Although related to infinite spin rings and chains such systems have not been considered mainly since it does not really matter whether an infinite ring has an odd or an even number of spins. In addition the sign rule of Marshall and Peierls [92] and the famous theorems of Lieb, Schultz, and Mattis [93, 94] provided valuable tools for the understanding of even rings which have the property to be bipartite and are thus non-frustrated. These theorems explain the degeneracy of the ground states in subspaces $\mathcal{H}(M)$ as well as their shift quantum number k or equivalently crystal momentum quantum number $p_k = 2\pi k/N$.

Nowadays exact diagonalization methods allow to evaluate eigenvalues and eigenvectors of H for small even and odd spin rings of various numbers N of spin sites and spin quantum numbers s where the interaction is given by antiferromagnetic nearest neighbor exchange [74–76,95–97]. Although Marshall-Peierls sign rule and the theorems of Lieb, Schultz, and Mattis do not apply to non-bipartite rings, i. e. frustrated rings with odd N, it turns out that such rings nevertheless show astonishing regularities⁵. Unifying the picture for even and odd N, we find for the ground state without exception [96,97]:

1. The ground state belongs to the subspace $\mathcal{H}(S)$ with the smallest possible total spin quantum number S; this is either S = 0 for $N \cdot s$ integer, then the total magnetic quantum number M is also zero, or S = 1/2 for $N \cdot s$ half integer, then $M = \pm 1/2$.

⁵ Work done with Klaus Bärwinkel and Heinz-Jürgen Schmidt, Universität Osnabrück, Germany.

Table 3.1. Properties of ground and first excited state of AF Heisenberg rings for various N and s: ground state energy E_0 , gap ΔE , degeneracy deg, total spin S and shift quantum number k

s		N									
	2	3	4	5	6	7	8	9	10		
$\frac{1}{2}$	1.5	0.5	1	0.747	0.934	0.816	0.913	0.844	0.903	$E_0/(NJ)$	
	1	4	1	4	1	4	1	4	1	deg	
	0	1/2	0	1/2	0	1/2	0	1/2	0	S	
	1	1, 2	0	1, 4	3	2, 5	0	2, 7	5	k	
$\frac{1}{2}$	4.0	3.0	2.0	2.236	1.369	2.098	1.045	1.722	0.846	$\Delta E/ J $	
	3	4	3	2	3	8	3	8	3	deg	
	1	3/2	1	1/2	1	3/2	1	3/2	1	S	
	0	0	2	0	0	1, 6	4	3, 6	0	k	
1	4	2	3	2.612	2.872	2.735	2.834	2.773	2.819	$E_0/(NJ)$	
	1	1	1	1	1	1	1	1	1	deg	
	0	0	0	0	0	0	0	0	0	S	
	0	0	0	0	0	0	0	0	0	k	
1	4.0	2.0	2.0	1.929	1.441	1.714	1.187	1.540	1.050	$\Delta E/ J $	
	3	9	3	6	3	6	3	6	3	deg	
	1	1	1	1	1	1	1	1	1	S	
	1	0, 1, 2	2	2, 3	3	3, 4	4	4, 5	5	k	

2. If $N \cdot s$ is integer, then the ground state is non-degenerate.

- 3. If $N \cdot s$ is half integer, then the ground state is fourfold degenerate.
- 4. If s is integer or $N \cdot s$ even, then the shift quantum number is k = 0.
- 5. If s is half integer and $N \cdot s$ odd, then the shift quantum number turns out to be k = N/2.
- 6. If $N \cdot s$ is half integer, then $k = \lfloor (N+1)/4 \rfloor$ and $k = N \lfloor (N+1)/4 \rfloor$ is found. $\lfloor (N+1)/4 \rfloor$ symbolizes the greatest integer less or equal to (N+1)/4.

In the case of s = 1/2 one knows the k-quantum numbers for all N via the Bethe ansatz [76,95], and for spin s = 1 and even N the k quantum numbers are consistent with [75].

It appears that for the properties of the first excited state such rules do not hold in general, but only for "high enough" N > 5 [97]. Then, as can be anticipated from Tables 3.1 and 3.2, we can conjecture that

- if N is even, then the first excited state has S = 1 and is threefold degenerate, and
- if N is odd and the single particle spin is half-integer, then the first excited state has S = 3/2 and is eightfold degenerate, whereas
- if N is odd and the single particle spin is integer, then the first excited state has S = 1 and is sixfold degenerate.

Considering relative ground states in subspaces $\mathcal{H}(M)$ one also finds – for even as well as for odd N – that the shift quantum numbers k show a

Table 3.2. Properties of ground and first excited state of AF Heisenberg rings for various N and s (continuation): ground state energy E_0 , gap ΔE , degeneracy deg, total spin S and shift quantum number k. $\dagger - O$. Waldmann, private communication. $\dagger \dagger -$ projection method [77]

s	N										
	2	3	4	5	6	7	8	9	10		
$\frac{3}{2}$	7.5	3.5	6	4.973	5.798	5.338	5.732	5.477	$5.704^{\dagger\dagger}$	$E_0/(NJ)$	
	1	4	1	4	1	4	1	4	1	deg	
	0	1/2	0	1/2	0	1/2	0	1/2	0	S	
	1	1, 2	0	1, 4	3	2, 5	0	2, 7	5	k	
$\frac{3}{2}$	4.0	3.0	2.0	2.629	1.411	2.171	1.117	1.838	$0.938^{\dagger\dagger}$	$\Delta E/ J $	
	3	16	3	8	3	8	3	8	3	deg	
	1	3/2	1	3/2	1	3/2	1	3/2	1	S	
	0	0, 1, 2	2	2, 3	0	1, 6	4	3, 6	0	k	
2	12	6	10	8.456	9.722	9.045	9.630	$9.263^{\dagger\dagger}$	$9.590^{\dagger\dagger}$	$E_0/(NJ)$	
	1	1	1	1	1	1	1	1	1	deg	
	0	0	0	0	0	0	0	0	0	S	
	0	0	0	0	0	0	0	0	0	k	
2	4.0	2.0	2.0	1.922	1.394	1.652	1.091	$1.431^{\dagger\dagger}$	$0.906^{\dagger\dagger}$	$\Delta E/ J $	
	3	9	3	6	3	6	3	6	3	deg	
	1	1	1	1	1	1	1	1	1	S	
	1	0, 1, 2	2	2, 3	3	3, 4	4	4, 5	5	k	
$\frac{5}{2}$	17.5	8.5	15	12.434	14.645	13.451	14.528^{\dagger}	$13.848^{\dagger\dagger}$	$14.475^{\dagger\dagger}$	$E_0/(NJ)$	
	1	4	1	4	1	4	1	4	1	deg	
	0	1/2	0	1/2	0	1/2	0	1/2	0	S	
	1	1, 2	0	1,4	3	2, 5	0	2, 7	5	k	

strikingly simple regularity for $N \neq 3$

$$k \equiv \pm (Ns - M) \lceil \frac{N}{2} \rceil \mod N , \qquad (3.42)$$

where $\lceil N/2 \rceil$ denotes the smallest integer greater than or equal to N/2 [98]. For N = 3 and $3s - 2 \ge |M| \ge 1$ one finds besides the ordinary k-quantum numbers given by (3.42) extraordinary k-quantum numbers, which supplement the ordinary ones to the complete set $\{k\} = \{0, 1, 2\}$.

For even N the k values form an alternating sequence 0, N/2, 0, N/2, ...on descending from the magnon vacum with M = Ns as known from the sign-rule of Marshall and Peierls [92]. For odd N it happens that the ordinary k-numbers are repeated on descending from $M \leq Ns-1$ to M-1 iff N divides [2(Ns - M) + 1].

Using the k-rule one can as well derive a rule for the relative ground state energies and for the respective S quantum numbers:

• For the relative ground state energies one finds that if the k-number is different in adjacent subspaces, $E_{\min}(S) < E_{\min}(S+1)$ holds. If the k-number is the same, the energies could as well be the same.

- Therefore, if N (even or odd) does not devide $(2(Ns-M)+1)\lceil N/2 \rceil$, then any relative ground state in $\mathcal{H}(M)$ has the total spin quantum number S = |M|.
- This is always true for the absolute ground state which therefore has S = 0 for Ns integer and S = 1/2 for Ns half integer.

The k-rule (3.42) is founded in a mathematically rigorous way for N even [92–94], N = 3, M = Ns, M = Ns - 1, and M = Ns - 2 [98]. An asymptotic proof for large enough N can be provided for systems with an asymptotically finite excitation gap, i. e. systems with integer spin s for which the Haldane conjecture applies [99,100]. In all other cases numerical evidence was collected and the k-rule as a conjecture still remains a challenge [98].

Rotational Bands

For many spin systems with constant isotropic antiferromagnetic nearest neighbor Heisenberg exchange the minimal energies $E_{min}(S)$ form a rotational band, i. e. depend approximately quadratically on the total spin quantum number S [48, 101, 102]

$$E_{min}(S) \approx E_a - J \frac{D(N,s)}{N} S(S+1)$$
 (3.43)

The occurrence of a rotational band has been noted on several occasions for an even number of spins defining a ring structure, e. g. see [102]. The minimal energies have been described as "following the Landé interval rule" [28–30, 32]. However, we⁶ find that the same property also occurs for rings with an odd number of spins as well as for the various polytope configurations we have investigated, in particular for quantum spins positioned on the vertices of a tetrahedron, cube, octahedron, icosahedron, triangular prism, and an axially truncated icosahedron. Rotational modes have also been found in the context of finite square and triangular lattices of spin-1/2 Heisenberg antiferromagnets [103,104], compare also the chapter of Richter, Schulenburg, and Honecker.

There are several systems, like spin dimers, trimers, squares, tetrahedra, and octahedra which possess a strict rotational band since their Hamiltonian can be simplified by quadrature. As an example the Heisenberg square, i. e., a ring with N = 4 is presented. Because the Hamilton operator (3.23) can be rewritten as

$$H_{\sim}^{2} = -J \left(\sum_{n=1}^{\infty} 2^{n} - \sum_{n=1}^{\infty} 2^{n} - \sum_{n=1}^{\infty} 2^{n} \right) , \qquad (3.44)$$

$$\mathbf{S}_{213} = \mathbf{s}(1) + \mathbf{s}(3) , \ \mathbf{S}_{24} = \mathbf{s}(2) + \mathbf{s}(4) , \qquad (3.45)$$

with all spin operators \underline{S}^2 , \underline{S}^2_{13} and \underline{S}^2_{24} commuting with each other and with \underline{H} , one can directly obtain the complete set of eigenenergies, and these are $\frac{1}{6}$ Work done together with Marshall Luban, Ames Lab, Iowa, USA.

characterized by the quantum numbers S, S_{13} and S_{24} . In particular, the lowest energy for a given total spin quantum number S occurs for the choice $S_{13} = S_{24} = 2s$

$$E_{min}(S) = -J \left[S \left(S+1 \right) - 2 \cdot 2s \left(2s+1 \right) \right] = E_0 - J S \left(S+1 \right) , \quad (3.46)$$

where $E_0 = 4s(2s + 1)J$ is the exact ground state energy. The various energies $E_{min}(S)$ form a rigorous parabolic rotational band of excitation energies. Therefore, these energies coincide with a parabolic fit (crosses connected by the dashed line on the l.h.s. of Fig. 3.11) passing through the antiferromagnetic ground state energy and the highest energy level, i. e., the ground state energy of the corresponding ferromagnetically coupled system.



Fig. 3.11. Energy spectra of antiferromagnetically coupled Heisenberg spin rings (horizontal dashes). The crosses connected by the dashed line represent the fit to the rotational band according to (3.46), which matches both the lowest and the highest energies exactly. On the l.h.s the dashed line reproduces the exact rotational band, whereas on the r.h.s. it only approximates it, but to high accuracy. The solid line on the r.h.s. corresponds to the approximation (3.47)

It turns out that an accurate formula for the coefficient D(N, s) of (3.46) can be developed using the sublattice structure of the spin array [101]. As an example we repeat the basic ideas for Heisenberg rings with an even number of spin sites [32]. Such rings are bipartite and can be decomposed into two sublattices, labeled A and B, with every second spin belonging to the same sublattice. The classical ground state (Néel state) is given by an alternating sequence of opposite spin directions. On each sublattice the spins are mutually parallel. Therefore, a quantum trial state, where the individual spins on each sublattice are coupled to their maximum values, $S_A = S_B =$ Ns/2, could be expected to provide a reasonable approximation to the true ground state, especially if s assumes large values. For rings with even N the approximation to the respective minimal energies for each value of the total spins $\Sigma = \Sigma_A + \Sigma_B$ is then given by [32]

$$E_{min}^{\text{approx}}(S) = -\frac{4J}{N} \left[S(S+1) - 2\frac{Ns}{2} \left(\frac{Ns}{2} + 1\right) \right] .$$
(3.47)

This approximation exactly reproduces the energy of the highest energy eigenvalue, i. e., the ground state energy of the corresponding ferromagnetically coupled system (S = Ns). For all smaller S the approximate minimal energy $E_{min}^{\text{approx}}(S)$ is bounded from below by the true one (Rayleigh-Ritz variational principle). The solid curve displays this behavior for the example of N = 6, s = 3/2 in Fig. 3.11 (r.h.s.). The coefficient "4" in (3.47) is the classical value, i. e. for each fixed even N the coefficient D(N, s) approaches 4 with increasing s [101].

The approximate spectrum, (3.47), is similar to that of two spins, S_A and S_B , each of spin quantum number Ns/2, that are coupled by an effective interaction of strength 4J/N. Therefore, one can equally well say, that the approximate rotational band considered in (3.47) is associated with an effective Hamilton operator

$$H^{\text{approx}}_{\sim} = -\frac{4J}{N} \left[\mathbf{S}^2 - \mathbf{S}^2_A - \mathbf{S}^2_B \right] , \qquad (3.48)$$

where the two sublattice spins, S_A, S_B , assume their maximal value $S_A = S_B = Ns/2$. Hamiltonian (3.48) is also known as Hamiltonian of the Lieb-Mattis model which describes a system where each spin of one sublattice interacts with every spin of the other sublattice with eaqual strength [94,105].

It is worth noting that this Hamiltonian reproduces more than the lowest levels in each subspace $\mathcal{H}(S)$. At least for bipartite systems also a second band is accurately reproduced as well as the gap to the quasi-continuum above, compare Figure 3.12 and [102]. This property is very useful since the approximate Hamiltonian allows the computation of several observables without diagonalizing the full Hamiltonian.

It is of course of utmost importance whether the band structure given by the approximate Hamiltonian (3.48) persists in the case of frustrated molecules. It seems that at least the minimal energies still form a rotational band which is understandable at least for larger spin quantum numbers s taking into account that the parabolic dependence of the minimal energies on Smainly reflects the classical limit for a wide class of spin systems [106].

The following example demonstrates that even in the case of the highly frustrated molecule {Mo₇₂Fe₃₀} the minimal energies arrange as a "rotational band"⁷. In the case of {Mo₇₂Fe₃₀} the spin system is decomposable into three sub-lattices with sub-lattice spin quantum numbers S_A , S_B , and S_C [48,101]. The corresponding approximate Hamilton operator reads

$$H_{\alpha \text{approx}} = -J \frac{D}{N} \left[\mathbf{S}^2 - \gamma \left(\mathbf{S}^2_A + \mathbf{S}^2_B + \mathbf{S}^2_C \right) \right] , \qquad (3.49)$$

where $\underset{\sim}{S}$ is the total spin operator and the others are sub-lattice spin operators. D and γ are allowed to deviate from their respective classical values, D = 6 and $\gamma = 1$, in order to correct for finite s.

⁷ Work done with Matthias Exler, Universität Osnabrück, Germany.



Fig. 3.12. The low-lying levels of a spin ring, N = 6 and s = 5/2 in this example, can be grouped into the lowest (Landé) band, the first excited (Excitation) band and the quasi-continuum (QC). For the spin levels of the L- and E-band k is given in brackets followed by the energy. Arrows indicate strong transitions from the L-band. Associated numbers give the total oscillator strength f_0 for these transitions. With friendly permission by Oliver Waldmann [102]



Fig. 3.13. DMRG eigenvalues and lowest rotational band of the s = 5/2 icosidodecahedron; m = 60 was used except for the lowest and first exited level which were calculated with m = 120

We use the DMRG method to approximate the lowest energy eigenvalues of the full Hamiltonian and compare them to those predicted by the rotational band hypothesis (3.49). Figure 3.13 shows the results and a fit to the lowest rotational band. Assuming the same dependence on m as in the s = 1/2 case, the relative error of the DMRG data should also be less than 1%. The agreement between the DMRG energy levels and the predicted quadratic dependence is very good. Nevertheless, it remains an open question whether higher lying bands are present in such a highly frustrated compound.

Magnetization Jumps

Although the spectra of many magnetic molecules possess a rotational band of minimal energies $E_{min}(S)$ and although in the classical limit, where the single-spin quantum number s goes to infinity, the function $E_{min}(S)$ is even an exact parabola if the system has co-planar ground states [106], we⁸ find that for certain coupling topologies, including the cuboctahedron and the icosidodecahedron (see Fig. 3.14), that this rule is violated for high total spins [89, 107]. More precisely, for the icosidodecahedron the last four points of the graph of E_{min} versus S, i. e. the points with $S = S_{max}$ to $S = S_{max}-3$, lie on a straight line

$$E_{min}(S) = 60Js^2 - 6Js(30s - S) . (3.50)$$

An analogous statement holds for the last three points of the corresponding graph for the cuboctahedron. These findings are based on numerical calculations of the minimal energies for several s both for the icosidodecahedron as well as for the cuboctahedron. For both and other systems a rigorous proof of the high spin anomaly can be given [89, 108].



Fig. 3.14. Structure of the icosidodecahedron (l.h.s.) and the cuboctahedron (r.h.s.)

The idea of the proof can be summarized as follows: A necessary condition for the anomaly is certainly that the minimal energy in the one-magnon space is degenerate. Therefore, localized one-magnon states can be constructed which are also of minimal energy. When placing a second localized magnon on the spin array there will be a chance that it does not interact with the

⁸ Work done with Heinz-Jürgen Schmidt, Universität Osnabrück, Andreas Honecker, Universität Braunschweig, Johannes Richter and Jörg Schulenburg, Universität Magdeburg, Germany.

first one if a large enough separation can be achieved. This new two-magnon state is likely the state of minimal energy in the two-magnon Hilbert space because for antiferromagnetic interaction two-magnon bound states do not exist. This procedure can be continued until no further independent magnon can be placed on the spin array. In a sense the system behaves as if it consists of non-interacting bosons which, up to a limiting number, can condense into a single-particle ground state. In more mathematical terms: In order to prove the high-spin anomaly one first shows an inequality which says that all points $(S, E_{min}(S))$ lie above or on the line connecting the last two points. For specific systems as those mentioned above what remains to be done is to construct particular states which exactly assume the values of E_{min} corresponding to the points lying on the bounding line, then these states are automatically states of minimal energy.

The observed anomaly – linear instead of parabolic dependence – results in a corresponding jump of the magnetization curve \mathcal{M} versus B, see Fig. 3.15. In contrast, for systems which obey the Landé interval rule the magnetization curve at very low temperatures is a staircase with equal steps up to the highest magnetization. The anomaly could indeed be observed in magnetization measurements of the Keplerate molecules {Mo₇₂Fe₃₀}. Unfortunately, the magnetization measurements [36, 48] performed so far suffer from too high temperatures which smear out the anomaly.



Fig. 3.15. Icosidodecahedron: L.h.s. – minimal energy levels $E_{min}(S)$ as a function of total spin S. R.h.s. – magnetization curve at T = 0 [89]

Nevertheless, it may be possible to observe truly giant magnetization jumps in certain two-dimensional spin systems which possess a suitable coupling (e. g. Kagomé) [107]. In such systems the magnetization jump can be of the same order as the number of spins, i. e. the jump remains finite – or in other words is macroscopic – in the thermodynamic limit $N \to \infty$. Thus, this effect is a true macroscopic quantum effect, see also the chapter by Richter, Schulenburg, and Honecker.

Bounds

Rigorous results on spin systems have sharpened our understanding of magnetic phenomena. In addition such results can serve as a basis or source of inspiration for the development of approximate models. For example, the inequalities of Lieb and Berezin [109,110] relating spectral properties of quantum systems to those of their classical counterparts provide a foundation for classical or semi-classical treatments of spin systems.

Exact bounds of the spectra belong to this class of rigorous results. They may be used as guidance or benchmark for approximate, e. g. variational methods. In earlier works linear bounds [89] as well as parabolic bounds [111] could be derived for magnetic spectra.

In this section I would like to present two examples of exact bounds for antiferromagnetic Heisenberg systems which explicitly use the topological structure of the spin array⁹.

The first concept which leads to upper bounds of the minimal energies $E_{min}(S)$ rests on "*n*-cyclicity", which for n = 2 is related to bi-partiteness [112]. The key point of the new concept of *n*-cyclicity is that the oriented graph of interactions between the spins of the molecule or spin array can be mapped onto the oriented cyclic graph with *n* vertices.

It is only in certain cases that different cyclicities n and n' mean an essential distinctness. This is because for $n \ge 4$ any *n*-cyclic system is also (n-2)-cyclic since three successive vertices and the corresponding edges can be mapped in a forward-backward-forward way, compare the l.h.s. of Fig. 3.16, which shows a homomorphism of a pentagon onto a triangle, as an example. Each 2m-ring and hence any 2m-cyclic system is *n*-cyclic for any positive integers m, n, since it is 2-cyclic and a 2-cycle can be homomorphically embedded into any *n*-cycle.



Fig. 3.16. The pentagon is 5-cyclic and also 3-cyclic (l.h.s.) whereas the tetrahedron is not 3-cyclic (r.h.s.), because if the four vertices of the tetrahedron are attached to the numbers 1, 2, 3 one number must repeat and occurs at adjacent vertices, which does not happen in the 3-cycle

⁹ Work done with Klaus Bärwinkel and Heinz-Jürgen Schmidt, Universität Osnabrück, Germany.

Hence it makes only sense to distinguish between even-cyclic systems, which will be called 2-cyclic, and (2n+1)-cyclic system with maximal integer n. If a spin system is 2-cyclic in our sense it will be bi-partite in the sense of Refs. [93, 94], but that notion of bi-partiteness is more general than ours since it also comprises cases with different coupling constants.

We consider some more examples which illustrate the definition of cyclicity. A triangular plane lattice with suitable periodic boundary conditions is 3-cyclic, a square lattice or cubic lattice is 2-cyclic. The kagomé lattice is 3-cyclic but not 2-cyclic. 3-cyclicity is equivalent to 3-colorability [37]. Hence the octahedron, the dodecahedron, the cuboctahedron, and the icosidodecahedron are 3-cyclic, cf. [37], but the tetrahedron is not, see r.h.s. of Fig. 3.16.

The construction of an upper bound is then realized by constructing an appropriate trial state which of course according to the Ritz variational principle must lead to an upper bound in the energy. In order to find a good low-lying state in a subspace $E_{min}(M)$ the total spin lowering operator S^- is applied (Ns - M)-times to the magnon vacuum state $|\Omega\rangle$. Of course this state is still an eigenstate of the Heisenberg Hamiltonian with the same energy as the magnon vacuum, but using the knowledge about the topological structure the components $|\mathbf{m}\rangle$ can be brought together with appropriate phases which reflect the *n*-cyclicity of the spin array. This transformation is done with the help of a unitary "Bloch" operator U_{ℓ} generalizing ideas of [113]. This operator produces a shift in the *k*-quantum number. For spin rings it is defined by the following action on the product states

$$U_{\mathcal{U}\ell} | \boldsymbol{m} \rangle = \prod_{j=1}^{N} \exp\left\{ i \frac{2\pi j (s-m_j)\ell}{n} \right\} | \boldsymbol{m} \rangle .$$
 (3.51)

The trial state is chosen to be

$$|\varphi\rangle = C_M U_\ell \left(\sum_{n=1}^{\infty} \right)^{(Ns-M)} |\Omega\rangle , \qquad (3.52)$$

where C_M is a normalization constant,

$$C_M^2 = \frac{(Ns+M)!}{(2Ns)! (Ns-M)!} .$$
(3.53)

For the Heisenberg Hamiltonian

$$H_{\sim} = -J \sum_{u,v} \, \underline{s}(u) \cdot \underline{s}(v) \,, \qquad (3.54)$$

where the sum runs over all (unsorted) interacting pairs (u, v) of spins at sites u and v, one obtains as the expectation value

$$\langle \varphi | \underset{\sim}{H} | \varphi \rangle = -J\gamma s^2 \cos \alpha_\ell \qquad (3.55)$$
$$-J(1 - \cos \alpha_\ell) \frac{\gamma}{N} \left(Ns^2 - \frac{2s \left((Ns)^2 - M^2 \right)}{2Ns - 1} \right) .$$

 γ denotes the number of interacting spin pairs, i. e. in a spin ring of length N it would be 2N. The best bound for $E_{\min}(M)$ is obtained if $\cos \alpha_{\ell}$ is as low as possible, i. e. $\ell = \frac{n}{2}$ and $\cos \alpha_{\ell} = -1$ for even n and $\ell = \frac{n \pm 1}{2}$ for odd n. Therefore, the final result for upper bounds is

$$E_{\min}(M) \le -Jc\gamma s^2 - J(1-c)\frac{\gamma}{N} \left(Ns^2 - \frac{2s\left((Ns)^2 - M^2\right)}{2Ns - 1}\right) , (3.56)$$

where c = -1 in the case of even *n* and $c = -\cos \frac{\pi}{n}$ for odd *n*.

The Figs. 3.17 and 3.18 display examples for upper bounds. For the spin ring with N = 6 presented in Fig. 3.17 which could be regarded as 2- or 3-cyclic one realizes that 2-cyclicity always leads to the best approximation.



Fig. 3.17. Upper and lower bounds of $E_{\min}(S)$ for Heisenberg spin rings with N = 6 and s = 1/2 (l.h.s.) as well as s = 5/2 (r.h.s.). The solid curves display the bounds for the minimal energies considering 2-cyclicity and 2-homogeneity



Fig. 3.18. Upper and lower bounds of $E_{\min}(S)$ for the triangular spin lattice with N = 12 and s = 1/2 (l.h.s.) as well as s = 1 (r.h.s.). The solid curves display the bounds for the minimal energies considering 3-cyclicity and 3-homogeneity

The second concept which rests on homogeneity of the interaction matrix leads to lower bounds of the minimal energies $E_{min}(S)$ [112]. For this purpose the Heisenberg Hamiltonian can be written in the general form¹⁰

¹⁰ Please be aware of the missing "-" sign.

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$$H_{\sim} = \sum_{\mu\nu} J_{\mu\nu} \underbrace{s}_{\mu\nu} \cdot \underbrace{s}_{\nu} \cdot \underbrace{s}_{\nu} . \qquad (3.57)$$

The matrix \mathbb{J} of exchange parameters $J_{\mu\nu}$ is assumed to be symmetric and having constant row sums j.

Next we turn to the suitable definition of *n*-homogeneity. Let the set of spin sites $\{1, \ldots, N\}$ be divided into *n* disjoint subsets of equal size *m*, $\{1, \ldots, N\} = \bigcup_{\nu=1}^{n} \mathcal{A}_{\nu}$, such that the coupling constants within each \mathcal{A}_{ν} are ≤ 0 , but ≥ 0 between \mathcal{A}_{ν} and \mathcal{A}_{μ} for $\nu \neq \mu$. Moreover, the partial row sums are assumed to be constant:

$$\sum_{b \in \mathcal{A}_{\mu}} J_{ab} = \begin{cases} j^{\text{in if } a} \in \mathcal{A}_{\mu} \\ j^{\text{ex if } a} \notin \mathcal{A}_{\mu} \end{cases} .$$
(3.58)

A spin system satisfying these assumptions will be called *n*-homogeneous, compare [92–94]). Note that this notion is incommensurable to "*n*-cyclicity" defined previously. However, certain rings, the triangular lattice, the kagomé lattice, and the icosidodecahedron satisfy both definitions. A necessary condition for nearest neighbor Heisenberg systems to be *n*-homogeneous is that the number of nearest neighbors, which is assumed to be constant, is divisible by (n-1). Actually, spin rings of even N are 2-homogeneous, rings of odd N are 3-homogeneous if N is divisible by 3.

We recall that $\mathbf{1} = \frac{1}{\sqrt{N}}(1, 1, \dots, 1)$ is an eigenvector of \mathbb{J} with eigenvalue j. Due to the *n*-homogeneity there are, after a suitable permutation of the spin sites, further eigenvectors of the form

$$u^{(k)} = (m:1, m:\rho^k, m:\rho^{2k}, \dots, m:\rho^{(n-1)k}), \ k = 1, \dots, n-1 \ , \quad (3.59)$$

where (m : x, ...) denotes the *m*-fold repetition of the entry *x*, and $\rho \equiv e^{2\pi i/n}$. The corresponding eigenvalues are $j_k = j^{\text{in}} + j^{\text{ex}} \sum_{p=1}^{n-1} \rho^{pk} = j^{\text{in}} - j^{\text{ex}}$, hence they coalesce into one (n-1)-fold degenerate eigenvalue. By applying the theorem of Geršgorin (c.f. [114], 7.2) this eigenvalue is shown to be the smallest one j_{\min} .

Next we construct a coupling matrix \widetilde{J} with the same eigenspaces as J but only three different eigenvalues. The three eigenvalues of \widetilde{J} can be chosen as

$$\tilde{j} = j, \ \tilde{j}_{\min} = j_{\min}, \ \tilde{j}_2 = j_2 \ .$$
 (3.60)

 j_2 is the remaining smallest eigenvalue of \mathbb{J}' (\mathbb{J} restricted to the subspace orthogonal to 1) after eliminating (n-1)-times j_{\min} from the set of eigenvalues. Thus it can happen that $j_2 = j_{\min}$ if j_{\min} is more than (n-1)-fold degenerate.

Let us write $S_{\mathcal{A}} \equiv \sum_{a \in \mathcal{A}} S_a$ for any subset $\mathcal{A} \subset \{1, \ldots, N\}$. After choosing \widetilde{H} to be a Heisenberg Hamiltonian of the form (3.57) with coupling matrix \widetilde{J} one finally finds that

$$\begin{aligned} H &\geq \widetilde{H} = -\alpha \left(\sum_{\nu} S_{\mathcal{A}_{\nu}}^{2} \right) + (\alpha + \beta) N s(s+1) + \gamma \left(S_{\nu}^{2} - \sum_{\nu} S_{\mathcal{A}_{\nu}}^{2} \right) \\ &\geq \frac{j - j_{\min}}{N} S(S+1) + N j_{\min} s(s+1) + (N-n)(j_{2} - j_{\min}) s . \end{aligned} \tag{3.61}$$

Since $j_2 - j_{\min} \ge 0$ the bound (3.61) is the better, the smaller *n* is. This is in contrast to the upper bound considered in the last section, which is improved for large odd *n*.

The Figs. 3.17 and 3.18 display examples for lower bounds. One realizes that the lower bounds are not as good as the upper ones.

3.5 Dynamics

In this section I would like to outline two branches – tunneling and relaxation – where the dynamics of magnetic molecules is investigated. The section is kept rather introductory since the field is rapidly evolving and it is too early to draw a final picture on all the details of the involved processes.

3.5.1 Tunneling

Tunneling dynamics has been one of the corner stones in molecular magnetism since its very early days, see e. g. [21,24,61–63].

The subject can roughly be divided into two parts, one deals with tunneling processes of the magnetization in molecules possessing a high ground state spin and an anisotropy barrier, the second deals with the remaining tunneling processes, e. g. in molecules which have an S = 0 ground state.

As already mentioned in Sect. 3.4.1 some molecules like Mn_{12} and Fe_8 possess a high ground state spin. Since the higher lying levels are well separated from the low-lying S = 10 levels a single-spin Hamiltonian (3.5), which includes an anisotropy term, is appropriate. Figure 3.19 sketches the energy landscape for an anisotropy term which is quadratic in S_z . If the Hamiltonian includes terms like a magnetic field in x-direction that do not commute with S_z resonant tunneling is observed between states $|S, M\rangle$ and $|S, -M\rangle$. This behavior is depicted on the l.h.s. of Fig. 3.19 for the transition between M = -10 and M = 10. If an additional magnetic field is applied in z-direction the quadratic barrier acquires an additional linear Zeeman term and is changed like depicted on the r.h.s. of Fig. 3.19. Now tunneling is possible between states of different |M|, see e. g. [115].

It is rather simple to model the tunneling process in the model Hilbert space of S = 10. i. e. a space with dimension 2S + 1 = 21. Nevertheless, in a real substance the tunneling process is accompanied and modified by other influences. The first major factor is temperature which may enhance the process, this leads to thermally assisted tunneling [65]. Each such substance hosts phonons which modify the tunneling process, too, resulting in



Fig. 3.19. Sketch of the tunneling barrier for a high spin molecule with S = 10, l.h.s. without magnetic field, r.h.s. with magnetic field, compare (3.5). The arrows indicate a possible resonant tunneling process

phonon assisted tunneling [66,116–118]. Then local dipolar fields and nuclear hyperfine fields may strongly affect the relaxation in the tunneling regime [6]. In addition there may be topological quenching due to the symmetry of the material [119–121]. And last but not least describing such complicated molecules not in effective single-spin models but in many-spin models is still in an unsatisfactory state, compare [122].

Another kind of tunneling is considered for Heisenberg spin rings with uniaxial single-ion anisotropy. Classically the ground state of even rings like Na:Fe₆ and Cs:Fe₈ is given by a sequence of spin up and down like in Fig. 3.20. It now turns out that such a Néel-like state, which is formulated in terms of spin-coherent states (3.40), contributes dominantly to the true ground state as well as to the first excited state if the anisotropy is large enough [91]. Thus it is found that the ground state $|E_0\rangle$ and the first excited state $|E_1\rangle$ can be approximated as

$$|E_{0}\rangle \approx \frac{1}{\sqrt{2}} (|N\acute{e}l,1\rangle \pm |N\acute{e}l,2\rangle)$$

$$|E_{1}\rangle \approx \frac{1}{\sqrt{2}} (|N\acute{e}l,1\rangle \mp |N\acute{e}l,2\rangle) ,$$
(3.62)

where the upper sign is appropriate for rings where the number of spins N is a multiple of 4, e. g. N = 8, and the lower sign is for all other even N.



Fig. 3.20. Sketch of the tunneling process between Néel-like states on a spin ring. Without loss of generality the state on the l.h.s. will be denoted by $|Néel, 1\rangle$ and the state on the r.h.s. will be denoted by $|Néel, 2\rangle$

Therefore, the tunneling frequency is approximately given by the gap between ground and first excited state. Experimentally, such a tunnel process is hard to observe, especially since ESR is sensitive only to the total spin. What would be needed is a local probe like NMR. This could be accomplished by replacing one of the iron ions by another isotope.

The tunneling process was further analyzed for various values of the uniaxial single-ion anisotropy [123]. Since in such a case the cyclic shift symmetry persists, k is still a good quantum number. Therefore, mixing of states is only allowed between states with the same k quantum number. This leads to the conclusion that the low-temperature tunneling phenomena can be understood as the tunneling of the spin vector between different rotational modes with $\Delta S = 2$, compare Fig. 3.21 and the subsection on rotational bands on page 177.



Fig. 3.21. Energy spectrum of spin rings with N = 6 and vanishing anisotropy at two magnetic fields drawn as a function of the magnetic quantum number M. The dashed curves represent the lowest-lying parabolas $E_{min}(M)$ discussed in Sect. 3.4.3. A white or black circle indicates that a state belongs to k = 0 or k = N/2, compare Fig. 3.12. States belonging to one spin multiplet are located on straight lines like that plotted in panel (a) for the S = 4 multiplet. With friendly permissions by Oliver Waldmann [123]

3.5.2 Relaxation Dynamics

In a time-dependent magnetic field the magnetization tries to follow the field. Looking at this process from a microscopic point of view, one realizes that, if the Hamiltonian would commute with the Zeeman term, no transitions would occur, and the magnetization would not change a tiny bit. There are basically two sources which permit transitions: non-commuting parts in the spin Hamiltonian and interactions with the surrounding. In the latter case the interaction with phonons seems to be most important.

Since a complete diagonalization of the full Hamiltonian including noncommuting terms as well as interactions like the spin-phonon interaction is practically impossible, both phenomena are modeled with the help of rate equations.

If we start with a Hamiltonian H_0 , which may be the Heisenberg Hamiltonian and add a non-commuting term H' like anisotropy the eigenstates of the full Hamiltonian are superpositions of those of H_0 . This expresses itself in avoided level crossings where the spectrum of H_0 would show level crossings, compare Fig. 3.22. Transitions between eigenstates of H_0 , which may have good M quantum numbers, can then effectively be modeled with the formula by Landau, Zener, and Stückelberg [124–128]

$$p = 1 - \exp\left\{-\frac{\pi\Delta^2}{2\hbar g\mu_B |M_1 - M_2| \frac{d}{dt}B}\right\} .$$
 (3.63)

 \varDelta denotes the energy gap at the avoided level crossing.



Fig. 3.22. Schematic energy spectrum in the vicinity of an avoided level crossing. The formula by Landau, Zener, and Stückelberg (3.63) approximates the probability p for the tunneling process from M_1 to M_2

The effect of phonons is taken into account by means of two principles: detailed balance, which models the desire of the system to reach thermal equilibrium and energy conservation, which takes into account that the energy released or absorbed by the spin system must be absorbed or released by the phonon system and finally exchanged with the thermostat. The interesting effects arise since the number of phonons is very limited at temperatures in the Kelvin-range or below, thus they may easily be used up after a short time (phonon bottleneck) and have to be provided by the thermostat around which needs a characteristic relaxation time. Since this all happens in a timedependent magnetic field, the Zeeman splittings change all the time and phonons of different frequency are involved at each time step. In addition the temperature of the spin system changes during the process because the equilibration with the thermostat (liq. Helium) is not instantaneous. More accurately the process is not in equilibrium at all, especially for multi-level spin systems. Only for two-level systems the time-dependent occupation can be translated into an apparent temperature. In essence the retarded dynamics

leads to distinct hysteresis loops which have the shape of a butterfly [129–131]. For more detailed information on dissipative two-level systems the interested reader is referred to [131,132].

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4 Spin Wave Analysis of Heisenberg Magnets in Restricted Geometries

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Abstract. In the last decade it has been proven that the standard spin-wave theory was able to provide accurate zero-temperature results for a number of low-dimensional Heisenberg spin systems. In this chapter we introduce the main ingredients of the spin-wave technique using as a working model the two-leg mixed-spin ferrimagnetic ladder and the Dyson–Maleev boson formalism up to second order in the spin-wave interaction. In the remainder, we survey typical applications in low-space dimensionality as well as some recent modifications of the theory admitting a quantitative analysis in magnetically disordered phases. The presented spin-wave results are compared with available numerical estimates.

4.1 Introduction

The spin-wave theory is probably one of the most powerful tools ever used in the theory of magnetism. Originally proposed by Bloch [1,2] and Holstein and Primakoff [3] as a theory of the ferromagnetic state, it was later extended for the antiferromagnetic Néel state by Anderson [4], Kubo [5], and Oguchi [6]. Dyson's profound analysis of spin-wave interactions [7,8] demonstrated that spin waves may be used to obtain asymptotic expansions for the thermodynamic functions of the Heisenberg ferromagnet at low temperatures. Dyson's method was generalized by Harris et al. [9] to calculate in a systematic way spin-spin correlations, spin-wave damping, and various thermodynamic properties of antiferromagnetic insulators.

It should be noticed that the basis of the spin-wave theory (SWT) for antiferromagnets is much less established than for ferromagnets. The Dyson– Maleev transformation [10] gives a correspondence between any operator defined on the Hilbert space of the spin system and an operator on the boson Hilbert space. Evaluating the required averages for the Bose system, we necessarily make two approximations. First, we expand these quantities, by using a perturbation formalism in which the unperturbed Hamiltonian is quadratic in boson operators and the perturbation is the remaining quartic interaction. Second, we neglect the projection operator in the averages, which takes into

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account the so-called kinematic interactions by canceling the boson states with more than 2S bosons per lattice site, S being the spin quantum number of the lattice spin. In the ferromagnetic case, Dyson has argued that these approximations would lead to results which are asymptotically correct at low temperatures (T) to all orders in T. In the antiferromagnetic case, the situation is less settled due to the zero-point motion, i.e. quantum spin fluctuations in the Néel state. In principle, one may suspect that there are errors in the perturbation theory even at zero T. The same problem appears in the Holstein–Primakoff formalism [3]. We refer the interested reader to the original papers cited above as well as to the monographs [11-13] for details concerning this problem. In principle, the spin-wave approach is less effective for low-dimensional quantum spin systems, as quantum spin fluctuations typically increase in reduced space dimensions (D) and for small spin quantum numbers S. Moreover, since at finite T thermal fluctuations completely destroy the magnetic long-range order in 1D and 2D Heisenberg models with isotropic short-range interactions [14], in such cases the conventional SWT completely fails.

In view of the mentioned drawbacks of SWT, it seems surprising that for the last decade the standard spin-wave approach has been found to give very accurate description of the zero-temperature physics of a number of low-dimensional spin models, the best example being the $S = \frac{1}{2}$ Heisenberg antiferromagnet on a square lattice [15]. Probably, another good example is the mixed-spin Heisenberg chain describing a large class of recently synthesized quasi-1D molecular magnets [16] (cf. Chap. 4). The following analysis reveals some common features of these examples, the most important being the weakness (in a sense) of spin-wave interactions. Fortunately, in low-space dimensions many numerical techniques – such as the quantum Monte Carlo method (QMC), the exact numerical diagonalization (ED), and the densitymatrix renormalization group method (DMRG) – are more effective, so that the discussed drawbacks of the spin-wave analysis may be partially reduced by a direct combination with numerical methods.

A goal of the present review is to summarize typical applications and some recent developments of the spin-wave approach related to low-dimensional quantum spin systems. The spin-wave technique is presented in the following section, using the mixed-spin Heisenberg ladder as a working model and the Dyson–Maleev boson formalism. Due to the asymptotic character of spinwave series, the calculation up to second order in the spin-wave interaction is a reasonable approximation for most of the applications at zero T. As far as at this level perturbative corrections can easily be calculated in the framework of the Rayleigh–Schrödinger theory, we will not consider in detail perturbation techniques based on magnon Green's functions [9, 17]. Typical applications of the spin-wave formalism in low-dimensional spin systems are presented in Sects. 3 and 4. In particular, Sect. 3 involves an analysis of the parameters of the quantum ferrimagnetic phase in mixed-spin quasi-1D models, such as the (s_1, s_2) Heisenberg chain. The SWT results are compared with available DMRG and ED numerical estimates. Section 4 collects basic SWT results concerning 2D Heisenberg antiferromagnets. Some recent modifications of the SWT – admitting a quantitative analysis in magnetically disordered phases – are presented in Sect. 5. Section 6 contains concluding remarks.

4.2 Dyson–Maleev Formalism

In this section we describe the formal apparatus of the SWT. We choose as a working model the mixed-spin Heisenberg ladder (Fig. 4.1) defined by the Hamiltonian

$$\mathcal{H} = \sum_{n=1}^{N} \left[\boldsymbol{s}_n \cdot \boldsymbol{\sigma}_{n+1} + \boldsymbol{\sigma}_n \cdot \boldsymbol{s}_{n+1} \right] + J_{\perp} \sum_{n=1}^{N} \boldsymbol{s}_n \cdot \boldsymbol{\sigma}_n , \qquad (4.1)$$

where the index $n \ (= 1, \dots, N)$ labels the rungs of the ladder, and N is an even integer. The ladder is composed of two types of spins (s_n, σ_n) characterized by the spin quantum numbers s_1 and $s_2 \ (s_1 > s_2)$: $s_n^2 = \hbar^2 s_1(s_1+1)$ and $\sigma_n^2 = \hbar^2 s_2(s_2+1)$. In the following text we use the notation $r_s \equiv s_1/s_2 > 1$, and set $\hbar = 1$ and $a_0 = 1$, a_0 being the lattice spacing along the ladder.



Fig. 4.1. Mixed-spin Heisenberg ladder composed of two types of site spins. The arrows show one of the classical ground states for $J_{\perp} > 0$, defined by the orientation of the ferromagnetic moment $M = \sum_{n=1}^{N} (s_n + \sigma_n)$. The intrachain coupling J = 1

It is worth noticing that the model (4.1) is not purely academic. For instance, recently published experimental work on bimetallic quasi-1*D* molecular magnets (cf. Chap. 4) implies that the magnetic properties of these mixedspin compounds are basically described by the Heisenberg spin model with antiferromagnetically coupled nearest-neighbor localized spins. The ladder structure in Fig. 4.1 reproduces, in particular, arrangements of the Mn ($s_1 = \frac{5}{2}$) and the Cu ($s_2 = \frac{1}{2}$) magnetic atoms along the *a* axis in the compounds MnCu(pbaOH)(H₂O)₃ (pbaOH = 2-hydroxy-1,3-propylenebisoxamato) [18].

4.2.1 Classical Reference State

The first step in constructing a spin-wave expansion is to find the lowestenergy classical spin configurations of the related classical model. As a rule, this is a straightforward task, apart from some magnetic models with competing interactions which may exhibit complicated non-collinear spin states (see, e.g. [19]). Another serious problem at this stage may be related to a macroscopic degeneracy of the classical ground state, a typical example being the Heisenberg model on a kagomé lattice (cf. Chap. 3) which exhibits a magnetically disordered ground state. Further analysis of the problem involves quantum fluctuations and the so-called *order-from-disorder phenomenon* [20, 21].

Turning to our model (4.1), it is easy to see that the required reference state for $J_{\perp} > 0$ is a ferrimagnetic spin configuration where the s_n spins are oriented in a given direction, and the σ_n spins point in the opposite direction (see Fig. 4.1). The state is degenerate under arbitrary rotations (as a whole) in the spin space. One may pick up a reference state by introducing a small staggered field, say, for the s_n spins. We can actually get more information even in the quantum case, by using the Lieb–Mattis theorem for bipartite lattices [22]. First, the theorem predicts that the quantum ground state belongs to a subspace with the total-spin quantum number $(S_1 - S_2)N$, i.e. for $J_{\perp} > 0$ the system has a ferrimagnetic ground state characterized by the ferromagnetic moment per site $M_0 = (s_1 - s_2)/2$. Second, the theorem states that the energies of the ground states $E(S_T)$ characterized by the total-spin quantum numbers $S_T \geq N(s_1 - s_2)$ are arranged as follows

$$E(S_T + 1) > E(S_T).$$
 (4.2)

Notice that the classical and quantum ferrimagnetic ground states have one and the same magnetization M_0 , but otherwise they are different because the classical ground state is not an eigenstate of the quantum model (4.1). The quantum ferrimagnetic state is $[2N(s_1 - s_2) + 1]$ -fold degenerate, since the z component of the total spin – being a good quantum number – takes the values $-N(s_1-s_2), -N(s_1-s_2)+1, \cdots, N(s_1-s_2)$. This quantum magnetic phase may also be characterized by the following sublattice magnetizations

$$\boldsymbol{m}_{A} = \frac{1}{N} \sum_{n=1}^{N} \langle \boldsymbol{s}_{n} \rangle \quad \boldsymbol{m}_{B} = \frac{1}{N} \sum_{n=1}^{N} \langle \boldsymbol{\sigma}_{n} \rangle , \qquad (4.3)$$

where the symbol $\langle \cdots \rangle$ means a quantum-mechanical average over the ground state. We shall later see that quantum spin fluctuations reduce the classical sublattice magnetizations s_1 and s_2 , but the magnetic long-range order is preserved, i.e. $\mathbf{m}_A, \mathbf{m}_B \neq 0$.

In the region $J_{\perp} < 0$ the situation is different, i.e. the lowest-energy spin configuration is the Néel antiferromagnetic state based on the composite rung spins $s_1 + s_2$. Now the Lieb–Mattis theorem predicts that the exact quantum ground state is a spin-singlet state, i.e. $S_T = 0$ and $M_0 = 0$. Therefore, it may be generally expected a magnetically disordered phase, t.e. $m_A, m_B = 0$, as the isotropic Heisenberg model (4.1) is defined on a bipartite 1D lattice (see, e.g. [23]). In terms of the SWT this would mean that the classical antiferromagnetic state is swept out by quantum fluctuations, so that the concept of the spin-wave expansion does not work at all.

4.2.2 Boson Hamiltonian

Now we describe the second step in constructing the spin-wave expansion, t.e. the transformation of (4.1) to a boson Hamiltonian. The most popular boson representation of spin operators has been suggested by Holstein and Primakoff [3]. Other useful representations have been devised by Schwinger [24], Maleev [10], Villain [25], and Goldhirsch [26,27].

We start by defining the Holstein–Primakoff representation for the spins s_n (n = 1, ..., N):

$$s_n^+ = \sqrt{2s_1} \sqrt{1 - \frac{a_n^\dagger a_n}{2s_1}} a_n, \ s_n^- = \sqrt{2s_1} a_n^\dagger \sqrt{1 - \frac{a_n^\dagger a_n}{2s_1}}, \ s_n^z = s_1 - a_n^\dagger a_n , (4.4)$$

where $s_n^{\pm} = s_n^x \pm s_n^y$ and s_1 is the spin quantum number. a_n and a_n^{\dagger} are annihilation and creation boson operators satisfying the commutation relations

$$[a_n, a_m^{\dagger}] = \delta_{nm}, \quad [a_n, a_m] = [a_n^{\dagger}, a_m^{\dagger}] = 0.$$
(4.5)

Using the last equations, it is easy to show that the operators defined by (4.4) satisfy the commutation relations for spin operators

$$[s_n^+, s_n^-] = 2s_n^z, \qquad [s_n^z, s_n^\pm] = \pm s_n^\pm, \tag{4.6}$$

and the equation $s_n^2 = s_1(s_1+1)$. The operators a_n and a_n^{\dagger} act in the infinitedimensional boson Hilbert space spanned by the orthonormal basis states

$$|n_1, n_2, \dots, n_N) = \frac{(a_1^{\dagger})^{n_1} (a_2^{\dagger})^{n_2} \cdots (a_N^{\dagger})^{n_N}}{\sqrt{n_1! n_2! \dots n_N!}} |0\rangle, \qquad (4.7)$$

where $n_i \ (=0, 1, ..., \infty)$ is the occupation number of site *i*. The reference vacuum state $|0\rangle$ is defined by the relations $a_i|0\rangle = 0$ (for $\forall i$).

It is possible to rationalize the square roots in (4.4) by the Maleev similarity transformation

$$a_n \longmapsto \left(1 - \frac{a_n^{\dagger} a_n}{2s_1}\right)^{1/2} a_n, \qquad a_n^{\dagger} \longmapsto a_n^{\dagger} \left(1 - \frac{a_n^{\dagger} a_n}{2s_1}\right)^{-1/2}.$$
(4.8)

This transformation is not unitary, but preserves the number operator $a_n^{\dagger}a_n$ as well as the commutation relations (4.5) within the physically relevant Hilbert space $(n_i \leq 2s_1 \text{ for } \forall i)$. Applying the last transformation to (4.4), we get the Dyson–Maleev boson representation

$$s_n^+ = \sqrt{2s_1} \left(1 - a_n^\dagger a_n / 2s_1 \right) a_n \,, \qquad s_n^- = \sqrt{2s_1} a_n^\dagger \,, \qquad s_n^z = s_1 - a_n^\dagger a_n \,. \tag{4.9}$$

Note that the operators s_n^{\pm} in this representation are not Hermitian conjugate in the boson space (4.7) so that in the general case they will generate non-Hermitian Hamiltonians. Treatment of such Hamiltonians requires some care, but it seems that – at least up to second order in the spin-wave interaction – this does not cause serious problems. More problematic is the relation between physical and unphysical states. The latter appear in the exact Holstein–Primakoff representation as well, as any actual calculation requires truncation of the asymptotic square-root series. Dyson's method [7] eliminates the unphysical boson states by a projection operator giving zero on these states. In practice, however, we are enforced to eliminate this operator. As already mentioned, this is the basic approximation of SWT. As a whole, the Dyson–Maleev formalism has many advantages if one needs to go beyond the linear spin-wave theory (LSWT) within a perturbation scheme. This is because the interactions between spin waves are better handled so that the unphysical singularities caused by the long-wavelength spin waves cancel out.

To continue, we write a representation similar to (4.9) for the spins σ_n , by using a new set of boson fields $(b_n, n = 1, \ldots, N)$:

$$\sigma_n^+ = \sqrt{2s_2} \, b_n^\dagger \left(1 - b_n^\dagger b_n / 2s_2 \right), \qquad \sigma_n^- = \sqrt{2s_2} \, b_n \,, \qquad \sigma_n^z = -s_2 + b_n^\dagger b_n \,.$$
(4.10)

 b_n and b_n^{\dagger} satisfy the same commutation relations (4.5), and are supposed to commute with the set of *a* bosons. Here the reference state is chosen in the opposite direction, in accord with the classical spin configuration in Fig. 4.1.

Using (4.9) and (4.10), we can find the boson image of any function of spin operators. In particular, we are interested in the boson representation of the spin Hamiltonian (4.1), which we denote by \mathcal{H}_B . For the purposes of SWT, it is instructive to express \mathcal{H}_B in terms of the Fourier transforms a_k and b_k of the boson operators a_n and b_n , by using the unitary Fourier transformations

$$a_n = \frac{1}{\sqrt{N}} \sum_k e^{ikn} a_k , \qquad b_n = \frac{1}{\sqrt{N}} \sum_k e^{-ikn} b_k , \qquad (4.11)$$

and the identity

$$\frac{1}{N}\sum_{n=1}^{N} \mathrm{e}^{\mathrm{i}(k-k')n} = \delta_{kk'}.$$

It may be verified that this transformation is canonical, by showing that the new operators a_k and b_k obey a set of commutation relations identical to (4.5). The wave vectors k in the last expressions are defined in the first Brillouin zone:

$$k = \frac{2\pi}{N}l$$
, $l = -\frac{N}{2} + 1, -\frac{N}{2} + 2, \dots, \frac{N}{2}$,.

Notice that the rung spins (s_n, σ_n) in Fig. 4.1 compose the *n*-th magnetic (and lattice) elementary cell: this may be easily observed by interchanging the site spins of every (say) even rung in Fig. 4.1.

We leave the Fourier transformation of \mathcal{H}_B as an exercise, and directly present the result in terms of the new operators a_k and b_k :

$$\mathcal{H}_B = -2\gamma_0 r_s S^2 + \mathcal{H}_0 + V'_{DM} \,, \qquad (4.12)$$

where

$$\mathcal{H}_0 = 2S \sum_k \left[\gamma_0 \left(a_k^{\dagger} a_k + r_s b_k^{\dagger} b_k \right) + \sqrt{r_s} \gamma_k \left(a_k^{\dagger} b_k^{\dagger} + a_k b_k \right) \right], \qquad (4.13)$$

$$V_{DM}^{'} = -\frac{1}{N} \sum_{1-4} \delta_{12}^{34} \left(2\gamma_{1-4}a_{3}^{\dagger}a_{2}b_{1}^{\dagger}b_{4} + \sqrt{r_{s}}\gamma_{1+2-4}a_{3}^{\dagger}b_{2}^{\dagger}b_{1}^{\dagger}b_{4} + \frac{1}{\sqrt{r_{s}}}\gamma_{4}a_{3}^{\dagger}a_{2}a_{1}b_{4} \right).$$

$$(4.14)$$

Here $\gamma_k = J_{\perp}/2 + \cos k$ ($\gamma_0 = J_{\perp}/2 + 1$), $\delta_{12}^{34} \equiv \Delta(k_1 + k_2 - k_3 - k_4)$ is the *Kronecker function*, and we have introduced the abbreviations $(k_1, k_2, k_3, k_4) \equiv (1, 2, 3, 4)$ for the wave vectors.

In a standard spin-wave expansion, $1/s_1$ and $1/s_2$ are treated as small parameters, whereas the parameter r_s may be considered as a fixed number of order unity. In such a perturbation scheme, it is convenient to set $1/S \equiv 1/s_2$ and use 1/S as a small parameter. Thus, the first term in (4.12) – the classical ground-state energy – is proportional to S^2 , the LSWT Hamiltonian \mathcal{H}_0 is multiplied by S, and the spin-wave interaction term V'_{DM} has the order $\mathcal{O}(1)$. We shall follow a perturbation scheme where the diagonal terms of V'_{DM} , i.e. terms proportional to the occupation-number operators $a_k^{\dagger}a_k$ and $b_k^{\dagger}b_k$, are treated together with \mathcal{H}_0 as a zeroth-order Hamiltonian, whereas the rest of V'_{DM} is taken as a perturbation [9]. This is a more generic approach because for some reasons the spin-wave interactions may be weak even in the extreme quantum systems with 1/S = 2.

4.2.3 Quasiparticle Representation

In the next step, we diagonalize the quadratic Hamiltonian \mathcal{H}_0 , by using the Bogoliubov canonical transformation to quasiparticle boson operators (α_k and β_k) [3]:

$$a_k = u_k(\alpha_k - x_k\beta_k^{\dagger}), \quad b_k = u_k(\beta_k - x_k\alpha_k^{\dagger}), \quad u_k^2(1 - x_k^2) = 1.$$
 (4.15)

It is a simple exercise to find the transformation parameters u_k and x_k from the condition which eliminates the off-diagonal terms $\alpha_k \beta_k$ appearing in \mathcal{H}_0 after the transformation (4.15). The result reads

$$u_k = \sqrt{\frac{1+\varepsilon_k}{2\varepsilon_k}}, \quad x_k = \frac{\eta_k}{1+\varepsilon_k},$$
(4.16)

where

$$\varepsilon_k = \sqrt{1 - \eta_k^2}, \quad \eta_k = \frac{2\sqrt{r_s}}{r_s + 1} \frac{\gamma_k}{\gamma_0}. \tag{4.17}$$

In some applications, the quadratic Hamiltonian \mathcal{H}_0 may include additional ferromagnetic bilinear terms (such as $a_k^{\dagger}b_k$) so that the actual diagonalization is more involved due to the increased number of parameters (4.16). Some diagonalization techniques for systems with large number of boson operators are presented in [11, 28].

A quasiparticle representation of the quartic terms (4.14) requires more technical work. As mentioned above, it is instructive to pick up the quadratic diagonal terms in V'_{DM} and to treat them together with \mathcal{H}_0 as a zeroth-order approximation. A simple way to do this is based on the presentation of V'_{DM} as a sum of normal-ordered products of boson quasiparticle operators. Apart from a constant, the resulting expression for V'_{DM} contains diagonal and off-diagonal quadratic operator terms, and normal-ordered quartic operator terms. We leave as an exercise this simple but somewhat cumbersome procedure and give the final result for \mathcal{H}_B expressed in terms of the quasiparticle boson operators α_k and β_k :

$$\mathcal{H}_B = E_0 + \mathcal{H}_D + \lambda V, \quad V = V_2 + V_{DM}, \quad \lambda \equiv 1.$$
 (4.18)

Here E_0 is the ground-state energy of the ferrimagnetic state calculated up to the order $\mathcal{O}(1)$ in the standard 1/S expansion:

$$\frac{E_0}{N} = -2\gamma_0 r_s S^2 - \gamma_0 (1+r_s) \left(1 - \frac{1}{N} \sum_k \varepsilon_k\right) S + e_1 + \mathcal{O}\left(\frac{1}{S}\right) , \quad (4.19)$$

where $e_1 = -2(c_1^2 + c_2^2) - J_{\perp}(c_1^2 + c_3^2) - (2c_2 + J_{\perp}c_3)c_1(r_s + 1)r_s^{-1/2}$ and

$$c_1 = -\frac{1}{2} + \frac{1}{2N} \sum_k \frac{1}{\varepsilon_k}, \quad c_2 = -\frac{1}{2N} \sum_k \cos k \frac{\eta_k}{\varepsilon_k}, \quad c_3 = -\frac{1}{2N} \sum_k \frac{\eta_k}{\varepsilon_k}.$$
(4.20)

 \mathcal{H}_D is the quadratic Hamiltonian resulting from \mathcal{H}_0 and the diagonal terms picked up from (4.14):

$$\mathcal{H}_D = 2S \sum_k \left[\omega_k^{(\alpha)} \alpha_k^{\dagger} \alpha_k + \omega_k^{(\beta)} \beta_k^{\dagger} \beta_k \right] \,, \tag{4.21}$$

where up to $\mathcal{O}(1/S)$ the dressed dispersions read

$$\omega_k^{(\alpha,\beta)} = \gamma_0 \left(\frac{r_s + 1}{2} \varepsilon_k \mp \frac{r_s - 1}{2} \right) + \frac{g_k^{\pm}}{2S} + \mathcal{O}\left(\frac{1}{S^2} \right)$$
(4.22)

where $g_k^{\pm} = (g_k \eta_k - d_0) \varepsilon_k^{-1/2} / 2 \pm (r_s - 1) (2c_2 + c_3 J_{\perp}) r_s^{-1/2} / 2$, $g_k = 2c_1 (r_s + c_3 J_{\perp}) r_s^{-1/2} / 2$ $1)\gamma_k r_s^{-1/2} + 4c_2 \cos k + 2c_3 J_{\perp}, d_0 = 4c_1\gamma_0 + (r_s + 1)(2c_2 + J_{\perp}c_3)r_s^{-1/2}.$ The functions $\omega_k^{(\alpha,\beta)}$ without $\mathcal{O}(1/S)$ corrections will be referred to as

bare dispersions.

Finally, the quasiparticle interaction V includes two different terms, i.e. the two-boson interaction

$$V_2 = \sum_k \left[V_k^+ \alpha_k^\dagger \beta_k^\dagger + V_k^- \alpha_k \beta_k \right]$$
(4.23)

defined by the vertex functions

$$V_k^{\pm} = \frac{d_0 \eta_k - g_k}{2\varepsilon_k} \mp \frac{r_s - 1}{\sqrt{r_s}} c_1 \gamma_k , \qquad (4.24)$$

and the quartic Dyson–Maleev interaction

$$V_{DM} = -\frac{J}{2N} \sum_{1-4} \delta_{12}^{34} \Big[V_{12;34}^{(1)} \alpha_1^{\dagger} \alpha_2^{\dagger} \alpha_3 \alpha_4 + 2V_{12;34}^{(2)} \alpha_1^{\dagger} \beta_2 \alpha_3 \alpha_4 + 2V_{12;34}^{(3)} \alpha_1^{\dagger} \alpha_2^{\dagger} \beta_3^{\dagger} \alpha_4 + 4V_{12;34}^{(4)} \alpha_1^{\dagger} \alpha_3 \beta_4^{\dagger} \beta_2 + 2V_{12;34}^{(5)} \beta_4^{\dagger} \alpha_3 \beta_2 \beta_1 + 2V_{12;34}^{(6)} \beta_4^{\dagger} \beta_3^{\dagger} \alpha_2^{\dagger} \beta_1 + V_{12;34}^{(7)} \alpha_1^{\dagger} \alpha_2^{\dagger} \beta_3^{\dagger} \beta_4^{\dagger} + V_{12;34}^{(8)} \beta_1 \beta_2 \alpha_3 \alpha_4 + V_{12;34}^{(9)} \beta_4^{\dagger} \beta_3^{\dagger} \beta_2 \beta_1 \Big], \qquad (4.25)$$

defined by the vertex functions $V_{12:34}^{(i)}$, $i = 1, \ldots, 9$. We have adopted the symmetric form of vertex functions used in [17]. The explicit form of $V_{12:34}^{(i)}$ depends on the concrete model. For the ladder model (4.1), the vertex functions may be obtained from those of the Heisenberg ferrimagnetic chain [29], using the formal substitution $\cos k \mapsto \cos k + J_{\perp}/2$.

In the following we shall treat the spin-wave interaction V as a small perturbation to the diagonal Hamiltonian $E_0 + H_D$. To restore the standard 1/S series, one should (i) use bare dispersion functions, and (ii) resume the series in powers of 1/S.

4.3 Spin Wave Analysis of Quasi-1D Ferrimagnets

In this section we analyze the magnon spectrum and basic parameters of the quantum ferrimagnetic phase of the model (4.1), by using the developed spin-wave formalism and the Rayleigh–Schrödinger perturbation theory up to second order in λ . The SWT results are compared with available DMRG and ED numerical estimates.

4.3.1 Linear Spin Wave Approximations

In a standard linear spin-wave approximation we consider only the first two terms in (4.12), and discard V'_{DM} as a next-order term in 1/S. This corresponds to the first two terms in the expression for the ground-state energy (4.19), and to the first term in the expression for the quasiparticle dispersions (4.22). As a matter of fact, by using the normal-ordering procedure, we have already got even the next-order terms of the expansions in 1/S for these quantities.

Magnon Excitation Spectrum

The quadratic Hamiltonian \mathcal{H}_D defines two branches of spin-wave excitations $(\alpha \text{ and } \beta \text{ magnons})$ described by the dispersion functions $\omega_k^{(\alpha,\beta)}$ in the first Brillouin zone $-\pi \leq k \leq \pi$ (see Fig. 4.2). The excited states $\alpha_k^{\dagger}|0\rangle$ $(\beta_k^{\dagger}|0\rangle)$ belong to the subspace characterized by the quantum number $S_T^z = S_T - 1$ $(S_T^z = S_T + 1)$, where $S_T = (s_1 - s_2)N$. In the long wavelength limit $k \ll 1$, the energies of α magnons $E_k^{(\alpha)}$ have the Landau–Lifshitz form

$$E_k^{(\alpha)} \equiv 2S\omega_k^{(\alpha)} = \frac{\varrho_s}{M_0}k^2 + \mathcal{O}(k^4), \qquad (4.26)$$

where ρ_s is the spin stiffness constant [30]. This form of the Goldstone modes is typical for Heisenberg ferromagnets, and reflects the fact that the order parameter, i.e. the ferromagnetic moment, is itself a constant of the motion.



Fig. 4.2. Magnon excitation spectrum of the mixed-spin ladder $(s_1, s_2) = (1, \frac{1}{2})$ for interchain couplings $J_{\perp} = 0.1$ and $J_{\perp} = 1$. The dashed lines display the energy of β magnons $E_k^{(\beta)}$ related to the Hamiltonian \mathcal{H}_D . The solid lines show the magnon spectra as obtained from the second-order approximation in V. The energy of α magnons related to (4.22) is not displayed, as it closely follows the respective solid lines. The symbols indicate ED numerical results. The figure is taken from [31]

The spin stiffness constant ρ_s as well as M_0 play a basic role in the lowtemperature thermodynamics [32]. The parameter ρ_s may be obtained from the Landau–Lifshitz relation and (4.22):
4 Spin Wave Analysis of Heisenberg Magnets in Restricted Geometries 205

$$\frac{\varrho_s}{2s_1s_2} = 1 - \frac{1}{S} \left(c_1 \frac{r_s + 1}{r_s} + \frac{c_2}{\sqrt{r_s}} \right) + \mathcal{O}\left(\frac{1}{S^2}\right). \tag{4.27}$$

The function $E_k^{(\alpha)}$ exhibits an additional minimum at the zone boundary, so that in the vicinity of π it reads

$$E_k^{(\alpha)} = \Delta_{\pi}^{(\alpha)} + \text{const}(\pi - k)^2$$
 (4.28)

Here $\Delta_{\pi}^{(\alpha)}$ is the excitation gap at the zone boundary. In the limit $J_{\perp} \to 0$, the excitation gap $\Delta_{\pi}^{(\alpha)} (\propto J_{\perp})$ goes to zero. For ferromagnetic couplings $J_{\perp} < 0$, the $k = \pi$ mode becomes unstable and produces global instability of the ferrimagnetic phase.

The function $E_k^{(\beta)} \equiv 2S\omega_k^{(\beta)}$ may be characterized by the spectral gaps $\Delta_0^{(\beta)}$ (at k = 0) and $\Delta_{\pi}^{(\beta)}$ (at $k = \pi$). The expression for $\Delta_0^{(\beta)}$ reads

$$\Delta_0^{(\beta)} = 2\gamma_0(s_1 - s_2) \left(1 - \frac{2c_2 + c_3 J_\perp}{2S\gamma_0\sqrt{r_s}} \right) + \mathcal{O}\left(\frac{1}{S}\right) \,. \tag{4.29}$$

For the $(s_1, s_2) = (1, \frac{1}{2})$ chain $(J_{\perp} = 0)$, the last equations give the results $\rho_s/2s_1s_2 = 0.761$ and $\Delta_0^{(\beta)} = 1.676$, to be compared with the results $\rho_s/2s_1s_2 = 1$ and $\Delta_0^{(\beta)} = 1$ obtained in a standard linear approximation using the Hamiltonian \mathcal{H}_0 [33, 34]. A comparison with the numerical QMC result $\Delta_0^{(\beta)} = 1.759$ [35] clearly demonstrates the importance of the 1/S corrections to the dispersion functions (4.22) in the extreme quantum limit.

Summarizing, it may be stated that the linear approximation – based on the quadratic Hamiltonian \mathcal{H}_D – gives a good qualitative description of the magnon excitation spectrum of the model (4.1). The same conclusion is valid for the ground-state energy: The expression (4.19) has been found to produce an excellent fit to the numerical ED results in a large interval up to $J_{\perp} = 10$ [31].

Sublattice Magnetizations

The on-site magnetizations $m_A = \langle s_n^z \rangle$ and $m_B = -\langle \sigma_n^z \rangle$ are parameters of the quantum ferrimagnetic phase which keep information for the long-range spin correlations. The simple LSWT results $m_A = s_1 - c_1$ and $m_B = s_2 - c_1$ show that quantum spin fluctuations reduce the classical on-site magnetizations already at the level of non-interacting spin waves. \mathcal{H}_0 produces the same results. The ratio

$$\frac{s_2 - m_B}{s_2} = \frac{c_1}{S} \tag{4.30}$$

may be used as a measure of the zero-point motion in the quantum ground state. Thus, there appears to be a well-defined semiclassical limit $S \to \infty$

where \mathcal{H}_0 is a sufficiently accurate approximation, provided $c_1/S \ll 1$. In this connection, it seems surprising that the spin-wave series for the $S = \frac{1}{2}$ square-lattice Heisenberg antiferromagnet produces the excellent result $m_A =$ 0.3069(2) [36] – the recent stochastic-series QMC estimate is 0.3070(3) [37] – in spite of the fact that in this case the parameter $c_1/S \approx 0.393$ is not small. Even more illuminating is the $(1, \frac{1}{2})$ ferrimagnetic chain: In spite of the large parameter $c_1/S \approx 0.610$, the second-order SWT gives the precise result $m_A = 0.79388$ [38] (DMRG estimate is $m_A = 0.79248$ [39]). It is difficult to explain the accuracy of SWT in terms of the standard 1/S series. However, as will be shown below, the quasiparticle interaction V produces numerically small corrections to the principal zeroth-order approximation.

In the mixed-spin model (4.1) there appears an important first-order correction to the sublattice magnetizations which is connected to the quadratic interaction V_2 . Let us go beyond the linear approximation and calculate the $\mathcal{O}(\lambda)$ correction to m_A . The on-site magnetization m_B may be obtained from the exact relation $m_A = s_1 - s_2 + m_B$ resulting from the conservation law for the ferromagnetic moment. The expression of m_A in terms of quasiparticle operators reads

$$m_A = s_1 - c_1 - \frac{1}{2N} \sum_k \left[\frac{1}{\varepsilon_k} \langle \alpha_k^{\dagger} \alpha_k + \beta_k^{\dagger} \beta_k \rangle - \frac{\eta_k}{\varepsilon_k} \langle \alpha_k^{\dagger} \alpha_k^{\dagger} + \beta_k^{\dagger} \beta_k^{\dagger} \rangle \right]. \quad (4.31)$$

Now we make use of the standard perturbation formula

$$\langle \hat{O} \rangle^{(1)} = \sum_{n \neq 0} \frac{\langle 0|V|n \rangle \langle n|\hat{O}|0 \rangle}{E_0 - E_n} + \sum_{n \neq 0} \frac{\langle n|V|0 \rangle \langle 0|\hat{O}|n \rangle}{E_0 - E_n}$$
(4.32)

giving the first-order correction in V of $\langle \hat{O} \rangle$. Here \hat{O} is an arbitrary operator and $\langle \cdots \rangle$ means a quantum-mechanical average over the exact ground state. The formula is also valid in the case of non-Hermitian perturbations V. In our case, \hat{O} is a quadratic operator, so that the sum in (4.32) is restricted to the two-boson eigenstates $|n_k\rangle = \alpha_k^{\dagger}\beta_k^{\dagger}|0\rangle$ of \mathcal{H}_D , k being a wave vector from the first Brillouin zone. The energies of these states are $E_k - E_0 = 2S(\omega_k^{(\alpha)} + \omega_k^{(\beta)})$. Finally, using the matrix elements

$$\langle 0|V_2|n_k\rangle = V_k^{(-)}, \quad \langle n_k|V_2|0\rangle = V_k^{(+)}, \quad (4.33)$$

we get the following result for m_A calculated up to first order in V:

$$m_A = s_1 - c_1 - \frac{1}{4SN} \sum_k \frac{\eta_k}{\varepsilon_k} \frac{V_k^{(+)} + V_k^{(-)}}{\omega_k^{(\alpha)} + \omega_k^{(\beta)}} + \mathcal{O}(\lambda^2) \,. \tag{4.34}$$

To find the standard 1/S correction to m_A , we have to use in (4.34) the bare dispersion functions.

Figure 4.3 shows the results for m_A , as obtained from (4.34) by using the bare and dressed dispersion functions (4.22). It is seen that the expansion in



Fig. 4.3. On-site magnetization (sublattice \mathcal{A}) of the $(1, \frac{1}{2})$ ladder as a function of the interchain coupling J_{\perp} . The dashed and dashed-dotted lines display the series results up to first order in 1/S (bare dispersions) and V (dressed dispersions). The solid line shows the series result up to second order in V. The Lanczos ED results for ladders with N = 12 rungs are denoted by open circles. The figure is taken from [31]

1/S gives a small (unexpected) decrease of m_A in the vicinity of $J_{\perp} = 0$, whereas the expansion in V produces a correct qualitative result in this limit. The indicated problem of the standard 1/S series probably results from enhanced fluctuations of the individual chain magnetizations about the common quantization axis. Indeed, at the special point $J_{\perp} = 0$ the classical ground state acquires an additional degeneracy under independent rotations of the chain ferromagnetic moments. Thus, the quartic diagonal interaction – included in \mathcal{H}_D – seems to stabilize the common quantization axis connected to the global ferromagnetic moment. We have an example where the expansion in powers of V gives better results.

Antiferromagnetic Chain

It is instructive to consider the antiferromagnetic chain as a special case $(s_1 = s_2, J_{\perp} = 0)$ of the mixed-spin model (4.1). After some algebra, from (4.19) and (4.22) we find the following simplified expressions for the ground-state energy (per site)

$$e_0 = -S^2 \left[1 + \frac{1}{2S} \left(1 - \frac{2}{\pi} \right) \right]^2 + \mathcal{O} \left(\frac{1}{S} \right)$$

$$(4.35)$$

and the magnon spectrum

$$\omega_k^{(\alpha,\beta)} \equiv \frac{E_k}{2S} = \left[1 + \frac{1}{2S}\left(1 - \frac{2}{\pi}\right)\right] |\sin k| + \mathcal{O}\left(\frac{1}{S^2}\right) \tag{4.36}$$

of the antiferromagnetic chain. For $S = \frac{1}{2}$, the standard LSWT gives the result $e_0 = -0.4317$ which is close to Hulthen's exact result $-\ln 2 + 1/4 \approx$ -0.443147 [40]. It is an illuminating agreement, as the theory might have been expected to fail for magnetically disordered states. Notice, however, that the next-order approximation, i.e. $e_0 = -0.4647$, does not improve the SWT result. This indicates a poor convergence of the 1/S expansion. We can also check the series for $S = \frac{3}{2}$, by using the numerical result $e_0 = -2.82833(1)$ [41] based on DMRG estimates for finite systems and the finite-size corrections to the energy, as derived from the Wess-Zumino-Witten theory [42]. The first two terms in the series (4.35) for $S = \frac{3}{2}$ give the result $e_0 = -2.79507$. In this case, an inclusion of the next-order term in (4.35) produces the precise SWT result $e_0 = -2.82808$. Thus, already for $S = \frac{3}{2}$ the spin-wave series shows a good convergence.

Turning to the magnon spectrum (4.36), we find that for $S = \frac{1}{2}$ SWT qualitatively reproduces Des Cloizeaux and Pearson's exact result for the one-magnon triplet excitation spectrum $E_k = \frac{\pi}{2} |\sin k|$ [43]. It is interesting that the 1/S correction in (4.36) improves the standard LSWT result for the spin-wave velocity (c = 1) to the value c = 1.3634: the exact result is $c = \pi/2 \approx 1.5708$. The magnon spectrum (4.36) is doubly degenerate and has the relativistic form $E_k = c|k|$ ($c|\pi - k|$) near the point k = 0 ($k = \pi$), to be compared with the rigorous result where the spin-wave states, being eigenstates of spin 1, are triply degenerate. Long-wavelength spin waves correspond to states where all regions are locally in a Néel ground state but the direction of the sublattice magnetization makes long-wavelength rotations.

Using (4.20) and (4.30), we find the following expression for the on-site magnetization in the antiferromagnetic chain

$$m = S - c_1 = S + \frac{1}{2} - \frac{1}{2N} \sum_k \frac{1}{|\sin k|} = -\infty.$$
(4.37)

We see that in 1D the quantum correction is divergent at small wave vectors already in the leading LSWT approximation, no matter how large is S. This indicates that the Néel state is destabilized by quantum fluctuations, so that the concept of spin-wave expansion fails.

Finally, it is instructive to calculate the long-wavelength behavior of the correlation function $\langle \mathbf{s}_n \cdot \boldsymbol{\sigma}_{n+x} \rangle$. Using the Dyson–Maleev representation and (4.15), one finds $\langle \mathbf{s}_n \cdot \boldsymbol{\sigma}_{n+x} \rangle = -S^2 + 2S \langle a_n b_{n+x} \rangle + \cdots$ where $\langle a_n b_{n+x} \rangle = -(1/2N) \sum_k (\cos k/|sink|) \exp(ikx)$. Thus, in the limit $x \gg 1$ one obtains

$$\langle \boldsymbol{s}_n \cdot \boldsymbol{\sigma}_{n+x} \rangle = -S^2 \left[1 - \frac{1}{\pi S} \ln x + \mathcal{O}\left(\frac{1}{S^2}\right) \right].$$
 (4.38)

This indicates that in the semiclassical limit $S \to \infty$ the antiferromagnetic chain is ordered at exponentially large scales $\xi \simeq a_0 \exp(\pi S)$ [44]. Here we have restored the lattice spacing a_0 .

4.3.2 Spin Wave Interactions

We have already discussed some effects of the quasiparticle interaction V, by calculating the first-order correction to the sublattice magnetizations m_A and m_B . Notice that $\mathcal{O}(\lambda)$ corrections to the ground-state energy (4.19) as well as to the dispersion functions (4.22) do not appear. Indeed, it is easy to see that the corresponding matrix elements $\langle 0|V|0\rangle$ and $\langle n_k|V|n_k\rangle$ ($|n_k\rangle = \alpha_k^{\dagger}|0\rangle$, or $\beta_k^{\dagger}|0\rangle$) vanish as a result of the normal ordering of V. It will be shown below that the $\mathcal{O}(\lambda^2)$ corrections lead to further improvement of the spin-wave results. To that end, we consider two examples, i.e. the ground-state energy E_0 and the dispersion function $\omega_k^{(\alpha)}$. The reader is referred to the original literature for similar calculations concerning the parameters m_A , ϱ_s [31], and $\Delta_0^{(\beta)}$ [29].

The calculations may be performed within the standard perturbation formula

$$E_i^{(2)} = \sum_{j \neq i} \frac{\langle i|V|j\rangle\langle j|V|i\rangle}{E_i - E_j}$$
(4.39)

giving the second-order correction in V to the eigenvalue E_i of the eigenstate $|i\rangle$ of a non-perturbed Hamiltonian. In our case, the zeroth-order Hamiltonian is $E_0 + \mathcal{H}_D$, and the perturbation V is given by (4.12). The sum in (4.39) runs over the eigenstates of \mathcal{H}_D .

Second-Order Corrections to E_0

We consider corrections to the vacuum state $|i\rangle \equiv |0\rangle$, so that the energy $E_i \equiv E_0$ is given by (4.19). There are two types of $\mathcal{O}(\lambda^2)$ corrections to E_0 which are connected with the interactions V_2 and V_{DM} .

First, we proceed with the quadratic interaction V_2 . It is easily seen that only the states $|j\rangle \equiv |n_k\rangle = \alpha_k^{\dagger} \beta_k^{\dagger} |0\rangle$ produce non-zero matrix elements in (4.39). The dominator for these two-boson states reads $E_0 - E_k = -2S(\omega_k^{(\alpha)} + \omega_k^{(\beta)})$, where $\omega_k^{(\alpha,\beta)}$ are defined by (4.22). Using the above results and (4.33), we get the following correction to the ground-state energy (4.19) coming from V_2 :

$$E_0^{(2)'} = -\frac{1}{2S} \sum_k \frac{V_k^{(+)} V_k^{(-)}}{\omega_k^{(\alpha)} + \omega_k^{(\beta)}}.$$
(4.40)

Next, we consider the Dyson–Maleev interaction V_{DM} . Looking at the explicit expression of V_{DM} (4.25), we find that only the term with the vertex function $V_{12;34}^{(7)}$ ($V_{12;34}^{(8)}$) does not annihilate the vacuum state $|0\rangle$ ($\langle 0|$). Thus, the sum in (4.39) runs over the four-boson eigenstates $|1234\rangle = (2!2!)^{-1/2} \alpha_{k_1}^{\dagger} \alpha_{k_2}^{\dagger} \beta_{k_4}^{\dagger} |0\rangle$. The related matrix elements read

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$$\langle 1234|V_{DM}|0\rangle = -\frac{1}{N}V_{12;34}^{(7)}\delta_{12}^{34}, \quad \langle 0|V_{DM}|1234\rangle = -\frac{1}{N}V_{43;12}^{(8)}\delta_{12}^{34}.$$

Using these expressions, we find the following correction to the ground-state energy resulting from V_{DM} :

$$E_0^{(2)''} = -\frac{1}{2S} \frac{1}{N^2} \sum_{1-4} \delta_{12}^{34} \frac{V_{43;12}^{(8)} V_{12;34}^{(7)}}{\omega_1^{(\alpha)} + \omega_2^{(\alpha)} + \omega_3^{(\beta)} + \omega_4^{(\beta)}}.$$
 (4.41)

Notice that the second-order correction to E_0 in powers of 1/S is the sum of $E_0^{(2)'}$ and $E_0^{(2)''}$ but calculated with the bare dispersion functions.

Second-Order Corrections to $\omega_k^{(\alpha)}$

Now we are interested in perturbations to the one-magnon states $|i\rangle \equiv |k\rangle = \alpha_k^{\dagger}|0\rangle$. The calculations may be performed by following the method already used for E_0 . Since we are treating an excited eigenstate, there appear new types of corrections connected to the vertex functions $V_{12;34}^{(2)}$ and $V_{12;34}^{(3)}$. These terms may be predicted, e.g. by drawing the diagrams shown in Fig. 4.4. Notice that the graphical representation of the vertex functions in Fig. 4.4 is connected to the quasiparticle operator forms of V_2 (4.23) and V_{DM} (4.25). The interested reader is referred to the original literature (see, e.g. [9,17,45]) where this diagram technique is explained in detail. We leave these simple calculations as an exercise, and directly present the expression for the second-order corrections to $\omega_k^{(\alpha)}$:



Fig. 4.4. Second-order self-energy diagrams giving the corrections to the dispersion function $\omega_k^{(\alpha)}$. Solid and dashed lines represent, respectively, the bare propagators for α and β magnons. The figure is taken from [29]

$$\begin{split} \delta\omega_{k}^{(\alpha)} &= -\frac{1}{(2S)^{2}} \left[\frac{V_{k}^{(+)}V_{k}^{(-)}}{\omega_{k}^{(\alpha)} + \omega_{k}^{(\beta)}} - \frac{2}{N} \sum_{p} \frac{V_{p}^{(+)}V_{kp;pk}^{(2)} + V_{p}^{(-)}V_{kp;pk}^{(3)}}{\omega_{p}^{(\alpha)} + \omega_{p}^{(\beta)}} \right. \\ &+ \frac{2}{N^{2}} \sum_{2-4} \delta_{k2}^{34} \left(\frac{V_{43;2k}^{(8)}V_{k2;34}^{(7)}}{\omega_{k}^{(\alpha)} + \omega_{2}^{(\alpha)} + \omega_{3}^{(\beta)} + \omega_{4}^{(\beta)}} + \frac{V_{43;2k}^{(3)}V_{k2;34}^{(2)}}{-\omega_{k}^{(\alpha)} + \omega_{2}^{(\beta)} + \omega_{3}^{(\alpha)} + \omega_{4}^{(\alpha)}} \right) \right].$$

$$(4.42)$$

It is interesting to note that the vertex functions $V_k^{(-)}$, $V_{kp;pk}^{(2)}$, $V_{kp;pk}^{(3)}$, $V_{43;2k}^{(3)}$, and $V_{43;2k}^{(3)}$ vanish at the zone center $k = 0^{-1}$, so that the gapless structure of $\omega_k^{(\alpha)}$ is preserved separately by each of the second-order corrections in (4.42). Thus, we have an example demonstrating some of the good features of the Dyson–Maleev formalism.

4.3.3 Comparison with Numerical Results

We have already presented in Figs. 2 and 3 second-order SWT results for the dispersion functions $\omega_k^{(\alpha,\beta)}$ and the on-site magnetization m_A of the $(1,\frac{1}{2})$ ladder. The comparison shows that the SWT dispersion functions closely follow the ED data in the whole Brillouin zone. For instance, the SWT result for the gap $\Delta_0^{(\beta)}$ at $J_{\perp} = 0.1$ differs by less than 0.5% from the ED estimate. Turning to m_A , we find a precision higher than 0.3% in the whole interval $0 \leq J_{\perp} \leq 3$. These are illuminating results, as in the considered system the perturbation parameter 1/S = 2 is large. To understand these results, let us consider, e.g. the λ series for the spectral gap $\Delta_0^{(\beta)}$ of the $(1, \frac{1}{2})$ chain [29]:

$$\frac{\Delta_0^{(\beta)}}{2(s_1 - s_2)} = 1.6756\lambda^0 + 0.1095\lambda^2 - 0.0107\lambda^3 + \mathcal{O}(\lambda^4)$$

Although 1/S = 2, we see that the quasiparticle interaction V introduces numerically small corrections to the zeroth-order approximation \mathcal{H}_D .

Finally, in Table 4.1 we have collected SWT results for different ferrimagnetic chains. It is interesting to note that even in the extreme quantum cases $(1, \frac{1}{2})$ and $(\frac{3}{2}, 1)$, deviations from the DMRG estimates are less than 0.03% for the energy and 0.2% for the on-site magnetization. Moreover, it is seen that the increase of $r_s = s_1/s_2$ – keeping $s_2 = \frac{1}{2}$ fixed – leads to a rapid improvement of the 1/S series. The above results suggest that the Heisenberg ferrimagnetic chains and ladders are examples of low-dimensional quantum spin systems where the spin-wave approach is an effective theoretical tool.

¹ Analytical properties of the vertex functions have been studied in [46].

Table 4.1. Spin-wave results for the parameters $e_0 = E_0/N$, m_A , and $\Delta_0 = \Delta_0^{(\beta)}/2(s_1 - s_2)$ of different (s_1, s_2) Heisenberg chains calculated, respectively, up to the orders 1/S, $1/S^2$, and $1/S^3$. The SWT results are compared with available DMRG estimates which are, respectively, denoted by \bar{e}_0 , \bar{m}_A [39], and $\bar{\Delta}_0$ [47]

(s_1,s_2)	e ₀	$\bar{\mathrm{e}}_{\mathrm{0}}$	m_A	\bar{m}_A	Δ_0	$\bar{\Delta}_0$
$ \begin{array}{c} \left(1, \frac{1}{2}\right) \\ \left(\frac{3}{2}, 1\right) \\ \left(\frac{3}{2}, \frac{1}{2}\right) \\ \left(2, \frac{1}{2}\right) \end{array} $	-1.45432 -3.86321 -1.96699 -2.47414	-1.45408 -3.86192 -1.96727	$\begin{array}{c} 0.79388 \\ 1.14617 \\ 1.35666 \\ 1.88984 \end{array}$	$\begin{array}{c} 0.79248 \\ 1.14427 \\ 1.35742 \end{array}$	$1.7744 \\ 1.6381 \\ 1.4217 \\ 1.2938$	$1.76 \\ 1.63 \\ 1.42 \\ 1.29$

4.4 Applications to 2D Heisenberg Antiferromagnets

In this section we survey recent applications of the spin-wave approach to 2D Heisenberg spin systems, the emphasis being on the ground-state parameters of the square- and triangular-lattice Heisenberg antiferromagnets. We shall skip most of the technical details, as the discussed spin-wave formalism actually does not depend on the space dimension. As already mentioned, for the last decade SWT has been found to produce surprisingly accurate results for the ground-state parameters of the square-lattice Heisenberg antiferromagnet even in the extreme quantum limit $S = \frac{1}{2}$. Below we collect these results and compare them with recent QMC numerical estimates. As to the triangular antiferromagnet, it seems to be a rare example of magnetically frustrated spin system where the spin-wave expansion is effective. In this case, we also give some technical details concerning the spin-wave expansion, as it includes some new issues resulting from the coplanar arrangement of classical spins.

4.4.1 Square-Lattice Antiferromagnet

The square-lattice $S = \frac{1}{2}$ Heisenberg antiferromagnet – being a simple and rather general model to describe the undoped copper-oxide materials – has received a great deal of interest for the last decade. Now it is widely accepted that the ground state of the model is characterized by antiferromagnetic long-rage order. Thus, the role of quantum spin fluctuations is restricted to reduction of the sublattice magnetization from its classical value $\frac{1}{2}$ by about 39%. ² In a seminal work by Chakravarty, Halperin, and Nelson [48] – using the renormalization-group approach to study the quantum non-linear σ model in 2 + 1 space-time dimension – it has been shown that in the socalled renormalized classical regime $k_BT \ll \rho_s$ the thermodynamic properties

² Compare with the reduction of about 42% of the classical on-site magnetization $\frac{1}{2}$ in the $(1, \frac{1}{2})$ ferrimagnetic chain (see Table 1).

of the 2D quantum Heisenberg antiferromagnet are dominated by magnon excitations, so that the leading and next-to-leading corrections in $k_B T/\rho_s$ are fully controlled by three physical parameters, i.e the spin stiffness constant ρ_s ,³ the spin-wave velocity c, and the on-site magnetization m, calculated at T = 0 (see also [49]). Moreover, it has been argued that the discussed universal thermodynamic properties appear for arbitrary $k_B T/\rho_s$, provided that $0 < \rho_s \ll J$ and $k_B T \ll J$, J being the nearest-neighbor exchange constant [50].

The quantities ρ_s , and c appear as input parameters in the quantum non-linear σ model defined by the Lagrangian density

$$\mathcal{L} = \frac{\rho_s}{2c^2} \left(\frac{\partial \boldsymbol{n}}{\partial t}\right)^2 - \frac{\rho_s}{2} \left[\left(\frac{\partial \boldsymbol{n}}{\partial x}\right)^2 + \left(\frac{\partial \boldsymbol{n}}{\partial y}\right)^2 \right], \qquad (4.43)$$

where the vector staggered field $\boldsymbol{n} = \boldsymbol{n}(t, x, y)$ satisfies the non-linear constraint $n^2 = 1$. This model may be introduced using arguments based on general grounds: As long as the continuous O(3) symmetry is spontaneously broken, the symmetry of the problem requires that the interaction of the Goldstone modes, i.e. spin waves, of the system in the long-wavelength limit be described by this model regardless of the details of the macroscopic Hamiltonian and the value of the spin. For the square-lattice antiferromagnet, close to $\mathbf{k} = (0,0)$ and (π,π) the magnon spectrum takes the relativistic forms $E_{\mathbf{k}} = c|\mathbf{k}|$ and $|\mathbf{\pi} - \mathbf{k}|$, c being the spin-wave velocity. If we expand \mathbf{n} as $(1, \epsilon_1, \epsilon_2)$, where the ϵ_i are small compared to unity, then the equations of motion following from (4.43) show that there are two modes both of which have the dispersion $E_{\mathbf{k}} = c|\mathbf{k}|$, as expected. If we expand the Lagrangian to higher orders in ϵ_i , we find that there are interactions between the spin waves whose strength is proportional to c/ρ_s , which is of order 1/S. We thus see that all the parameters appearing in (4.43) can be determined by SWT. Compared to the standard 1/S expansion, the hydrodynamic approach is more generic in two points, i.e. (i) it is applicable to magnetically disordered phases, and (ii) it may lead to non-perturbative results which are beyond the reach of SWT (see, e.g. [51-53]).

Ground-state parameters of the $S = \frac{1}{2}$ square-lattice Heisenberg antiferromagnet have been studied in great detail using a variety of techniques, including SWT, QMC, and series expansions [15]. An early QMC study by Reger and Young [54] indicated that the SWT gives a good quantitative description of the ground state. Series expansions around the Ising limit performed by Singh [55,56] found the results $\rho_s \approx 0.18J$ and $c \approx 1.7J$, both in good agreement with the first-order SWT [6]. Later on, higher-order calculations demonstrated that the second-order corrections in 1/S to the parameters

³ This quantity, measuring the response of the system to an infinitesimal twist of the spins around an axis perpendicular to the direction of the broken symmetry, should not be confused with the spin stiffness constant of the ferromagnetic state ρ_s connected to the Landau–Lifshitz relation (4.26).

 ρ_s , c and m are small – even in the extreme quantum limit $S = \frac{1}{2}$ – and improve the SWT results. For instance – using both the Dyson–Maleev and Holstein–Primakoff formalisms up to second order in 1/S – Hamer et al. calculated the ground-state energy E_0/N and the sublattice magnetization m [36]. Both formalisms were shown to give identical results closely approximating previous series estimates [57]. Different scientific groups have presented consistent second-order SWT results for the spin-wave velocity c [58–60], the uniform transverse susceptibility χ_{\perp} [59,61] and the spin stiffness constant ρ_s^4 [59, 61]. In Table 4.2 we have collected some of these results, demonstrating an excellent agreement with recent high-precision numerical estimates [154] obtained by using the stochastic series expansion QMC method for $L \times L$ lattices with L up to 16.

Table 4.2. Second-order SWT results for the ground-state energy per site $e_0 = E_0/N$ [36], the on-site magnetization m [36, 59], the spin-wave velocity c [59, 60], the uniform transverse susceptibility χ_{\perp} [59, 61], and the spin stiffness constant ρ_s [59, 61] of the $S = \frac{1}{2}$ square-lattice Heisenberg antiferromagnet. The SWT results are compared to recent stochastic series expansion QMC estimates for $L \times L$ lattices with L up to 16 [154]. The series results for e_0 , m and χ_{\perp} are taken from [57], and those for ρ_s and c – from [61]. The figures in parentheses show the errors in the last significant figure. $\hbar = a_0 = J = 1$

Quantity	SWT	QMC	Series
$ \begin{array}{c} -e_0 \\ m \\ c \\ \chi_{\perp} \\ \rho_c \end{array} $	$\begin{array}{c} 0.669494(4)\\ 0.3069(1)\\ 1.66802(3)\\ 0.06291(1)\\ 0.180978 \end{array}$	$\begin{array}{c} 0.669437(5)\\ 0.3070(3)\\ 1.673(7)\\ 0.0625(9)\\ 0.175(2) \end{array}$	$\begin{array}{c} 0.6693(1)\\ 0.307(1)\\ 1.655(12)\\ 0.0659(10)\\ 0.182(5) \end{array}$

The accuracy of SWT may be understood in terms of the spin-wave interaction V. Indeed, let us consider the 1/S series for m [36]

$$m = S - 0.1966019 + \frac{0.003464}{(2S)^2} + \mathcal{O}\left(\frac{1}{S^3}\right).$$
(4.44)

For $S = \frac{1}{2}$, the related series in powers of λ simply reads $m = 0.3033981\lambda^0 + 0.003464\lambda^2 + \mathcal{O}(\lambda^3)$, so that the spin-wave interaction V introduces numerically small corrections to the leading approximation. The same conclusion is valid for the other parameters.

⁴ The reported third-order SWT result for this parameter is 0.1750(1) [61].

4.4.2 Triangular-Lattice Antiferromagnet

The Heisenberg antiferromagnet on a triangular lattice with nearest-neighbor exchange interactions is a typical example of a strongly frustrated spin model.⁵

After a long period of intensive studies – see, e.g. [64] and references therein – it is now widely accepted that the classical coplanar ground state survives quantum fluctuations. This state may be represented by the ansatz

$$\frac{\boldsymbol{s_r}}{S} = \hat{\boldsymbol{z}}\cos(\boldsymbol{q}_M \cdot \boldsymbol{r}) + \hat{\boldsymbol{x}}\sin(\boldsymbol{q}_M \cdot \boldsymbol{r}), \qquad (4.45)$$

where $\boldsymbol{q}_M = (\frac{4\pi}{3}, 0)$ is the wave vector of the magnetic pattern, $\hat{\boldsymbol{x}} \perp \hat{\boldsymbol{z}}$ are unit coordinate vectors in the spin space, and \boldsymbol{r} runs on the lattice sites. As usual, the lattice spacing a_0 is set to unity. The classical spins lay in the (x, z) plane, and point in three different directions so that the angle $\frac{2\pi}{3}$ is settled between any pair of spins in the elementary triangle $(\boldsymbol{s}_a, \boldsymbol{s}_b, \boldsymbol{s}_c)$.

In performing the 1/S expansion about non-collinear reference states such as (4.45), one faces some novelties which will be discussed in the remainder of this section. One of them concerns the number of boson fields needed to keep track of the whole magnon spectrum. This is an important practical issue, as higher-order spin-wave expansions involving more than two boson fields are, as a rule, technically intractable. In the general case, this number should be equal to the number of spins in the magnetic elementary cell, so that for the magnetic structure (4.45) we would need three boson fields. However, in several special cases we can transform the non-collinear magnetic structures into a ferromagnetic configuration by applying a uniform twist on the coordinate frame. These special systems have the property that their magnon spectrum has no gaps at the boundaries of the reduced magnetic Brillouin zone connected to the magnetic pattern. The triangular-lattice antiferromagnet fulfills this rule, so that we may describe the system by a single boson field, as in the ferromagnetic case. In the remainder of this section we shall follow this approach [65].

To that end, let us rotate the spin coordinate frame about the y axis by the angle $\theta_{rr'} = q_M \cdot (r - r')$ for any pair of neighboring spins $(s_r, s_{r'})$, in accord to the reference state (4.45). In the local reference frame, the Heisenberg Hamiltonian acquires the form

$$\mathcal{H} = \sum_{(\boldsymbol{r},\boldsymbol{r}')} \left[\cos \theta_{\boldsymbol{r}\boldsymbol{r}'} \left(s_{\boldsymbol{r}}^{x} s_{\boldsymbol{r}'}^{x} + s_{\boldsymbol{r}}^{z} s_{\boldsymbol{r}'}^{z} \right) + \sin \theta_{\boldsymbol{r}\boldsymbol{r}'} \left(s_{\boldsymbol{r}}^{z} s_{\boldsymbol{r}'}^{x} - s_{\boldsymbol{r}}^{x} s_{\boldsymbol{r}'}^{z} \right) + s_{\boldsymbol{r}}^{y} s_{\boldsymbol{r}'}^{y} \right], \quad (4.46)$$

where the sum runs over all pairs of nearest-neighbor sites of the triangular lattice.

 $^{^{5}}$ For a recent review on frustrated quantum magnets, see [63].

Next, using the Holstein–Primakoff transformation $(4.4)^6$ and the procedures described in Sect. 2, we find the following boson representation for (4.46)

$$\mathcal{H}_B = -\frac{3}{2}S^2N + 3S\sum_{k} \left[A_{\boldsymbol{k}} a_{\boldsymbol{k}}^{\dagger} a_{\boldsymbol{k}} + \frac{B_{\boldsymbol{k}}}{2} \left(a_{\boldsymbol{k}}^{\dagger} a_{-\boldsymbol{k}}^{\dagger} + a_{\boldsymbol{k}} a_{-\boldsymbol{k}} \right) \right] + V, \quad (4.47)$$

 $A_{\mathbf{k}} = 1 + \nu_{\mathbf{k}}/2$, $B_{\mathbf{k}} = -3\nu_{\mathbf{k}}/2$, and $\nu_{\mathbf{k}} = \frac{1}{3}[\cos k_x + 2\cos(k_x/2)\cos(\sqrt{3}k_y/2)]$. Here and in the remainder of this section, \mathbf{k} takes N values from the first Brillouin zone of the triangular lattice.

Up to quartic anharmonic terms, the expansion of the square root in (4.46) produces the following spin-wave interaction $V = V_3 + V_4$, where

$$V_3 = i\sqrt{\frac{S}{2}} \frac{3}{2\sqrt{N}} \sum_{1-3} (\kappa_1 + \kappa_2) (a_1^{\dagger} a_2^{\dagger} a_3 - a_3^{\dagger} a_2 a_1), \qquad (4.48)$$

$$V_4 = -\frac{3}{16N} \sum_{1-4} \left[\Gamma_{12;34}^{(1)} a_1^{\dagger} a_2^{\dagger} a_3 a_4 + \Gamma_{123}^{(2)} (a_1^{\dagger} a_2^{\dagger} a_3^{\dagger} a_4 + a_4^{\dagger} a_3 a_2 a_1) \right] , (4.49)$$

 $\kappa_{\mathbf{k}} = \frac{1}{3} [\sin k_x - 2\sin(k_x/2)\cos(\sqrt{3}k_y/2)], \Gamma_{12;34}^{(1)} = 4\nu_{1-3} + 4\nu_{2-3} + \nu_1 + \nu_2 + \nu_3 + \nu_4, \text{ and } \Gamma_{123}^{(2)} = -2(\nu_1 + \nu_2 + \nu_3).$ For simplicity, in the last expressions we have omitted the Kronecker δ function, and have used the abbreviations for the wave vectors introduced in Sect. 4.2.2.

A novelty here is the triple boson interaction $V_3 = \mathcal{O}(S^{1/2})$, which is typical for systems exhibiting non-collinear magnetic patterns. We shall see below that such kind of interactions complicate the calculation of higher-order 1/S corrections.

Linear Spin Wave Approximation

In a standard LSWT, we discard V and diagonalize the quadratic part of (4.47) by the Bogoliubov transformation $a_{\mathbf{k}} = u_{\mathbf{k}}(\alpha_{\mathbf{k}} - x_{\mathbf{k}}\alpha^{\dagger}_{-\mathbf{k}})$. The parameters $u_{\mathbf{k}}$ and $x_{\mathbf{k}}$ are defined by (4.16) and (4.17), but in this case $\eta_{\mathbf{k}} = -3\nu_{\mathbf{k}}/(2 + \nu_{\mathbf{k}})$. The diagonalization yields the free-quasiparticle Hamiltonian $\mathcal{H}_0 = 3S \sum_{\mathbf{k}} \omega_{\mathbf{k}} \alpha^{\dagger}_{\mathbf{k}} \alpha_{\mathbf{k}}$, where the dispersion function

$$E_{\boldsymbol{k}} \equiv 3S\omega_{\boldsymbol{k}} = 3S\sqrt{(1-\nu_{\boldsymbol{k}})(1+2\nu_{\boldsymbol{k}})} \tag{4.50}$$

gives the magnon energies in a LSWT approximation, to be compared with the magnon spectrum resulting from the approach using three boson fields [66]. It is easy to check that the dispersion function (4.50) exhibits three zero modes, as it should be since the Hamiltonian symmetry O(3) is completely

 $^{^6}$ The choice of the transformation is a matter of convenience, as the final results – at least to second order in 1/S – are independent of the boson representation.

broken by the magnetic pattern (4.45). Two of these modes are at the ordering wave vectors $\mathbf{k} = \pm \mathbf{q}_M$, whereas the third zero mode at $\mathbf{k} = 0$ describes soft fluctuations of the total magnetization. Expanding about the zero modes, we find the following expressions for the spin-wave velocities [67]

$$c_{0\perp} \equiv c_{\pm q_M} = \left(\frac{3}{2}\right)^{3/2} S, \quad c_{0\parallel} \equiv c_{k=0} = \frac{3\sqrt{3}}{2} S.$$
 (4.51)

Let us now calculate the on-site magnetization $m = \langle s_{\boldsymbol{r}}^z \rangle = S - \langle a_{\boldsymbol{r}}^{\dagger} a_{\boldsymbol{r}} \rangle$. Using the Bogoliubov transformation, we find for the density of particles $\langle a_{\boldsymbol{k}}^{\dagger} a_{\boldsymbol{k}} \rangle = -1/2 + 1/(2\varepsilon_{\boldsymbol{k}})$, so that the LSWT result for *m* reads [66]

$$m = S + \frac{1}{2} - \frac{1}{2N} \sum_{k} \frac{1}{\sqrt{1 - \eta_{k}^{2}}} = S - 0.2613.$$
(4.52)

For $S = \frac{1}{2}$, the LSWT result is m = 0.2387. Since the reported leading 1/S correction to m is small and positive⁷, there is a clear disagreement with the recent QMC estimate m = 0.20(6) [69].

Spin Wave Interactions

Here we consider as an example the calculation of 1/S corrections to the magnon spectrum (4.50). There are two different types of corrections related to the spin-wave interactions V_3 and V_4 in (4.48). Turning to V_4 , notice that we have already learned (Sect. 4.2.3) that the required correction may be obtained by expressing V_4 as a sum of normal products of quasiparticle operators: the diagonal quadratic terms give the required 1/S correction to the spectrum. However, in several cases we are not interested in the quasiparticle representation of V_4 . Then, it is possible to follow another way by decoupling the quartic operator products in V_4 . Actually, this procedure takes into account the so-called one-loop diagrams, and may be performed within a formal substitution of the operator products, such as $a_1^{\dagger}a_2^{\dagger}a_3a_4$, by the following sum over all the non-zero pair boson correlators

$$a_1^{\dagger}a_2^{\dagger}a_3a_4 \longmapsto \sum_{\text{pair}} \left[\langle a_1^{\dagger}a_2^{\dagger} \rangle a_3a_4 + a_1^{\dagger}a_2^{\dagger} \langle a_3a_4 \rangle - \langle a_1^{\dagger}a_2^{\dagger} \rangle \langle a_3a_4 \rangle \right] .$$
(4.53)

As suggested by the quadratic form in (4.47), there are two types of boson correlators, i.e. $\langle a_1^{\dagger}a_2 \rangle$ and $\langle a_1a_2 \rangle = \langle a_1^{\dagger}a_2^{\dagger} \rangle$, contributing in (4.53). The constant terms in (4.53) give first-order corrections to the ground state energy, whereas the quadratic operator products renormalize the coefficients A_k and B_k in (4.47). Thus, the interaction V_4 renormalizes the bare dispersion function ω_k to

⁷ We are aware of two such calculations reporting, however, somewhat different corrections, i.e. 0.0055/S [68] and 0.00135/S [65].

$$\bar{\omega}_{\boldsymbol{k}} = \sqrt{\bar{A}_{\boldsymbol{k}}^2 - \bar{B}_{\boldsymbol{k}}^2}, \qquad (4.54)$$

where the new coefficients \bar{A}_{k} and \bar{B}_{k} can be expressed in the form⁸

$$\bar{A}_{k} = A_{k} \left(1 + \frac{a_{1}}{2S} \right) + \frac{a_{2}}{2S}, \quad \bar{B}_{k} = B_{k} \left(1 + \frac{b_{1}}{2S} \right) + \frac{b_{2}}{2S}.$$

An analysis of (4.54) indicates that the renormalized spectrum still preserves the zero mode at k = 0, but at the same time acquires non-physical gaps at $k = \pm q_M$. The reason for such kind of behavior of the SWT is connected with the fact that we have omitted the 1/S corrections resulting from V_3 . Indeed, the spin-wave interaction V_3 has the order $\mathcal{O}(S^{1/2})$, so that a simple power counting indicates that 1/S corrections to ω_k appear in the second-order of the perturbation theory in V_3 . We shall skip the details of this calculation, as it may be performed entirely in the framework of the method presented in Sect. 4.2. Namely, one should express V_3 in terms of quasiparticle operators, and then apply the general perturbation formula (4.32) for the interaction V_3 , by using the dressed dispersions (4.54). As a matter of fact, as we are interested in corrections up to 1/S, we can use the bare dispersion function (4.50). The final result of this calculation shows that the 1/S correction resulting from V_3 exactly vanishes the gap (produced by V_4), so that the structure of magnon spectrum (4.50) – containing three zero modes - is preserved in the leading first-order approximation [70]. Based on therenormalized dispersion, the following expressions for the spin-wave velocities (4.51) have been reported [65]:

$$c_{\parallel} = c_{0\parallel} \left(1 - \frac{0.115}{2S} \right), \quad c_{\perp} = c_{0\perp} \left(1 + \frac{0.083}{2S} \right).$$

Notice that the 1/S corrections diminish the ratio c_{\parallel}/c_{\perp} from the LSWT result 1.41 to the value 1.16. These expressions indicate that the leading corrections to the magnon spectrum are numerically small even in the case $S = \frac{1}{2}$. Good convergence has been found also for the 1/S series of the magnetic susceptibilities χ_{\perp} and χ_{\parallel} [71, 72] which appear as parameters of the magnetic susceptibility tensor [73]

$$\chi_{\alpha\beta} = \chi_{\perp} \delta_{\alpha\beta} + (\chi_{\parallel} - \chi_{\perp}) y_{\alpha}) y_{\beta} \,.$$

Here \hat{y} is a unit vector directed perpendicular to the basal (x, z) plane of the planar magnetic structure.

Summarizing, the available SWT results point towards a good convergence of the perturbative spin-wave series in the triangular-lattice Heisenberg antiferromagnet. This is remarkable, as the spin-wave expansion might have been expected to fail for strongly frustrated magnetic systems.

⁸ For brevity, here we omit the expressions for the constants a_1 , a_2 , b_1 , and b_2 [65].

4.5 Modified Spin Wave Theories

Here we consider some modifications of the standard spin-wave theory allowing for an analysis of magnetically disordered phases. These may appear either as a result of quantum fluctuations – a classical example being the spin-S Heisenberg antiferromagnetic chain discussed in Sect. 4.3.1 – or due to thermal fluctuations, as in 1D and 2D Heisenberg magnets with short-range isotropic interactions [14]. For the antiferromagnetic chain, we have indicated that the failure of SWT arises already in the linear spin-wave approximation as a divergency in the boson-occupation numbers $n_i = \langle a_i^{\dagger} a_i \rangle = \infty$ implying $\langle s_i^z \rangle = -\infty$. Infinite number of spin waves also appears at T > 0, when the T = 0 magnetic phases of low-dimensional Heisenberg systems do not survive thermal fluctuations. Actually, the occupation numbers n_i should not exceed 2S – as dictated by the spin algebra – and the magnetization should be zero, as required by the symmetry of the phases. In the remainder of this section we discuss modifications of the SWT based on *ad hoc* constraints imposing fixed number of bosons.

The first generalized spin-wave theory of this kind has been formulated by Takahashi to study the low-T thermodynamics of 1D and 2D Heisenberg ferromagnets [74,75]. Takahashi's idea was to supplement the standard SWT of Heisenberg ferromagnets with the constraint imposing zero ferromagnetic moment at T > 0:

$$M = \sum_{n=1}^{N} \langle s_n^z \rangle = SN - \sum_{\boldsymbol{k}} \langle a_{\boldsymbol{k}}^{\dagger} a_{\boldsymbol{k}} \rangle = 0.$$
(4.55)

Depending on the context, in the remainder of this section $\langle A \rangle$ means the expectation value of the operator A at T = 0 or T > 0. Quite surprisingly, it was found an excellent agreement with the Bethe-ansatz low-temperature expansions of the free energy and magnetic susceptibility for the $S = \frac{1}{2}$ Heisenberg ferromagnetic chain. Similar extensions of SWT have been suggested for Heisenberg antiferromagnets both at T = 0 [76,77] and at T > 0 [78,79], by using the same constraint equation (4.55) but for the sublattice magnetization. Below we discuss some applications of the modified SWT to low-dimensional Heisenberg antiferromagnets both at T = 0 and at finite temperatures.

4.5.1 Square-Lattice Antiferromagnet at Finite T

Using the Dyson–Maleev transformations (4.9) and (4.10), the boson Hamiltonian \mathcal{H}'_B of the square-lattice antiferromagnet reads

$$\mathcal{H}_{B}^{'} = -\frac{N}{2}JzS^{2} + \sum_{\boldsymbol{k}} \left[A_{\boldsymbol{k}}(a_{\boldsymbol{k}}^{\dagger}a_{\boldsymbol{k}} + b_{\boldsymbol{k}}^{\dagger}b_{\boldsymbol{k}}) + B_{\boldsymbol{k}}(a_{\boldsymbol{k}}^{\dagger}b_{\boldsymbol{k}}^{\dagger} + a_{\boldsymbol{k}}b_{\boldsymbol{k}}) \right] + V_{DM}^{'}, \quad (4.56)$$

whereas the constraint equation for the total sublattice magnetization takes the form

$$\sum_{\boldsymbol{k}} \langle a_{\boldsymbol{k}}^{\dagger} a_{\boldsymbol{k}} + b_{\boldsymbol{k}}^{\dagger} b_{\boldsymbol{k}} \rangle = SN \,. \tag{4.57}$$

The wave vector \mathbf{k} runs in the small (magnetic) Brillouin zone $|k_x \pm k_y| \leq \pi$ containing N/2 points. $A_{\mathbf{k}} = JSz$, $B_{\mathbf{k}} = JSz\gamma_{\mathbf{k}}$, $\gamma_{\mathbf{k}} = \frac{1}{2}(\cos k_x + \cos k_y)$, and z = 4 is the lattice coordination number.

In essence, the constraint equation (4.57) introduces an effective cut-off for unphysical states [80]. To see this, let us consider the $S = \frac{1}{2}$ system. According to (4.57), the average number of, say, the α magnons is N/4, whereas the total number of one-magnon states in the magnetic Brillouin zone is N/2. Thus, after introducing the constraint (4.57), the effective number of allowed states in the boson Hilbert space is

$$\left[\frac{(N/2)!}{(N/4)!(N/4)!}\right]^2 \sim \frac{4}{\pi} \frac{2^N}{N} \,,$$

so that with logarithmic accuracy the correct dimension 2^N is restored.

To implement the constraint equation in the theory, we introduce, as usual, a chemical potential μ for the boson fields, i.e. instead of \mathcal{H}'_B we consider the Hamiltonian

$$\mathcal{H}_B = \mathcal{H}_B' - \mu \sum_{\boldsymbol{k}} (a_{\boldsymbol{k}}^{\dagger} a_{\boldsymbol{k}} + b_{\boldsymbol{k}}^{\dagger} b_{\boldsymbol{k}}), \qquad (4.58)$$

where μ is fixed by the constraint equation (4.57). Notice that the introduction of a chemical potential simply renormalizes the coefficient $A_{\mathbf{k}} \rightarrow A_{\mathbf{k}} - \mu$ so that we can apply the formalism from Sect. 4.2 without any changes.

Using the Bogoliubov transformation (4.15) with the parameter $\eta_{\mathbf{k}} = JzS\gamma_{\mathbf{k}}/(JzS-\mu)$, one finds the following quasiparticle representation of \mathcal{H}_B (see, e.g. [17])

$$\mathcal{H}_B = E_0 + \mathcal{H}_D + V_{DM} \,, \tag{4.59}$$

where E_0 is the ground-state energy calculated up to first-order of the perturbation theory in 1/S:

$$E_0 = -\frac{N}{2}zJS^2 \left(1 + \frac{r}{2S}\right)^2, \quad r = 1 - \frac{2}{N}\sum_{k}\sqrt{1 - \eta_k^2}.$$
 (4.60)

As we know from Sect. 4.2.3, the free-quasiparticle Hamiltonian

$$\mathcal{H}_D = \sum_{\boldsymbol{k}} E_{\boldsymbol{k}} (\alpha_{\boldsymbol{k}}^{\dagger} \alpha_{\boldsymbol{k}} + \beta_{\boldsymbol{k}}^{\dagger} \beta_{\boldsymbol{k}})$$
(4.61)

includes the diagonal quadratic terms resulting from V'_{DM} , so that the magnon energies E_k are calculated up to first-order corrections in 1/S:

$$E_{\mathbf{k}} = JzS\left(1 + \frac{r}{2S}\right)\sqrt{1 - \eta_{\mathbf{k}}^2}.$$
(4.62)

Here the factor r/2S is Oguchi's correction to the magnon spectrum [6].

We want to treat the spin-wave interaction up to first order in the 1/S perturbation theory, so that the Dyson–Maleev interaction V_{DM} will be discarded. It is important to notice that here the off-diagonal quadratic interaction V_2 does not appear, as dictated by the sublattice interchange symmetry. This means that the lowest-order corrections to the sublattice magnetization m have the order $\mathcal{O}(S^{-2})$, see the series (4.44), so that the constraint equation (4.57) calculated in a LSWT approximation can be safety used at this level.

Turning to the magnon spectrum (4.62), we see that the chemical potential introduces a spectral gap Δ so that close to the zone center the excitation spectrum acquires the relativistic form

$$E_{k} = \sqrt{\Delta^{2} + c^{2}k^{2}}, \quad c = \frac{JzS}{\sqrt{2}} \left(1 + \frac{r}{2S}\right), \quad (4.63)$$

where $\Delta = 2c(-\mu/JzS)^{1/2}$ and c is the spin-wave velocity calculated up to first order in 1/S. Using the standard expression for free bosons $n_{\mathbf{k}} = \langle \alpha_{\mathbf{k}}^{\dagger} \alpha_{\mathbf{k}} \rangle = \langle \beta_{\mathbf{k}}^{\dagger} \beta_{\mathbf{k}} \rangle = [\exp(-E_{\mathbf{k}}/k_BT) - 1]^{-1}$, the constraint equation (4.57) takes the form

$$S + \frac{1}{2} = \frac{1}{N} \sum_{k} \frac{1}{\sqrt{1 - \eta_{k}^{2}}} \operatorname{coth} \frac{E_{k}}{k_{B}T}.$$
 (4.64)

At low T, the main contributions in the last sum come from small wave vectors so that, using (4.63), the gap equation (4.64) yields

$$\Delta = \frac{c}{\xi} = 2T \operatorname{arcsinh}\left[\frac{1}{2} \exp\left(-\frac{2\pi\rho_s}{k_B T}\right)\right].$$
(4.65)

Here ρ_s is the T = 0 spin stiffness constant calculated up to first order in 1/S, and ξ is the spin correlation length. This result exactly reproduces the saddle-point equation in the 1/N expansion of the O(N) nonlinear σ model in 2 + 1 space-time dimensions (see, e.g. [81]). It is well known that (4.65) describes three different regimes, i.e. (i) the renormalized classical, (ii) the quantum critical, and (iii) the quantum disordered regimes [53].

As an example, we consider the renormalized classical regime defined by the condition $k_B T \ll \rho_s$. In this case, the last equation yields the following result for the correlation length

$$\xi \sim \frac{c}{T} \exp\left(\frac{2\pi\rho_s}{k_B T}\right) \,. \tag{4.66}$$

This coincides with the one-loop approximation of the 2 + 1 nonlinear σ model [48]. As is well known, at a two-loop level the *T* dependence in the pre-exponential factor disappears, whereas the exponent argument does not change.

Finally, let us calculate the leading temperature correction to the internal energy $U = \langle \mathcal{H}_B \rangle$. The expression for U reads

$$U = E_0 + \sum_{\boldsymbol{k}} E_{\boldsymbol{k}} \left(\coth \frac{E_{\boldsymbol{k}}}{k_B T} - 1 \right) \,. \tag{4.67}$$

Using (4.63), after some algebra one finds the following result:

$$U = E_0 + \frac{2\zeta(3)N}{\pi c^2} T^3.$$
(4.68)

Here $\zeta(x)$ is the Riemann zeta function. The above temperature correction describes the contribution from two zero modes, i.e. $\mathbf{k} = (0,0)$ and $\mathbf{k} = (\pi,\pi)$, and reproduces the expected universal behavior known from the 2+1 nonlinear σ model and the chiral perturbation theory [49,82].

4.5.2 Applications to Finite-Size Systems

The modified SWT can also be applied to finite-size systems [76, 77]. This opens an opportunity directly to compare SWT results with finite-size numerical data. As is known, the standard SWT is not applicable to finite systems due to divergences related to the Goldstone zero modes. Actually, the divergency comes from the Bogoliubov transformation (4.15) which is not defined for these modes.

Turning to the example from Sect. 4.5.1, notice that in the infinite system the chemical potential μ goes to zero as $T \to 0$. At T = 0 the constraint equation takes the form

$$S + \frac{1}{2} - \frac{2}{N\sqrt{1 - \eta_0^2}} - \frac{1}{N} \sum_{\boldsymbol{k} \neq 0} \frac{1}{\sqrt{1 - \eta_{\boldsymbol{k}}^2}} = 0.$$
 (4.69)

Here we have selected the contribution from the two zero modes at $\mathbf{k} = (0, 0)$ having $S^z = \pm 1$.

According to (4.69), on a finite lattice the parameter $\eta_0 = JzS/(JzS - \mu)$ is less than unity, so that the divergences associated with the zero modes disappear. The constraint (4.69) takes into account the fact that in finite systems there are no spontaneously broken continuous symmetries.

To find the staggered magnetization m appearing in the thermodynamic limit of the 2D system, we calculate the antiferromagnetic structure factor $S(\pi, \pi)$ for large N:

$$m^{2}(N) = \frac{2}{N}S(\pi,\pi) = \frac{4}{(1-\eta_{0}^{2})N^{2}} + \frac{1}{N^{2}}\sum_{\boldsymbol{k}\neq0}\frac{1+\eta_{\boldsymbol{k}}^{2}}{1-\eta_{\boldsymbol{k}}^{2}},$$
(4.70)

where we have again selected the contribution from the zero modes.

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In the large-N limit, the last sum transforms into an integral which is $\propto \ln N$, so that the main contribution comes from the first term in (4.70). Thus, we find the relation

$$m^2 = \lim_{N \to \infty} \frac{4}{(1 - \eta_0^2)N^2} \,. \tag{4.71}$$

Equation (4.69) induces a gap in the magnon spectrum which is defined by $\Delta = c\sqrt{2(1-\eta_0^2)}$. Using (4.71) and the notations from Sect. 4.5.1, we find the following result for the magnon excitation gap in the large-N limit

$$\Delta = \frac{c^2}{\rho_s L^2} \,. \tag{4.72}$$

 $L = N^{1/2}$ is the linear size in a square geometry. The last expression reproduces the result for Δ obtained by other methods [49,83,84].

Finally, let us return to the Heisenberg antiferromagnetic chain discussed in Sect. 4.3.1, this time using the modified SWT [79]. We have seen that in 1Dthe expression for the staggered magnetization (4.37) contained an infrared divergency indicating that the magnetic order is destabilized by quantum fluctuations. Using the concept of the modified theory, we can resolve the problem by replacing (4.37) with the constraint equation

$$S + \frac{1}{2} = \frac{1}{N} \sum_{k} \frac{1}{\sqrt{1 - \eta_0^2 \cos^2 k}} = \frac{K(\eta_0)}{\pi}, \qquad (4.73)$$

where $K(\eta_0)$ is the complete elliptic integral of the first kind.

Since $K(\eta_0) \ge \pi/2$, the gap equation (4.73) has a solution for arbitrary S. However, the constraint introduces an excitation gap, so that the discussed theory makes sense only for integer S. To find the gap, we may use for small $(1 - \eta_0^2)^{1/2}$ the asymptotic result $K(\eta_0) = \ln 4(1 - \eta_0^2)^{-1/2}$, so that the excitation gap reads

$$\Delta \sim c \, \exp\left(-\pi S\right). \tag{4.74}$$

Here c is the spin-wave velocity of the antiferromagnetic chain (4.36). The obtained gap has the asymptotic form $\Delta \sim S \exp(-\pi S)$, to be compared with Haldane's result $\Delta \sim S^2 \exp(-\pi S)$ obtained from the σ -model mapping [85,86]. It is remarkable that the simple modified SWT is capble to reproduce the asymptotic expression for the Haldane gap.

4.6 Concluding Remarks

We have surveyed the spin-wave technique and its typical applications to Heisenberg magnetic systems in restricted geometries. In most of the cases the SWT results were compared with the available numerical estimates. As a result, the systematic large-S technique has been found to give very accurate description of the zero-temperature parameters and magnon excitation spectra of a number of low-dimensional quantum spin models, such as the Heisenberg antiferromagnet on square and triangular lattices and various quasi-one-dimensional mixed-spin Heisenberg systems exhibiting ferrimagnetic ground states. Presented analysis of the asymptotic series up to second order in the parameter 1/S implies that in these systems the spin-wave interaction introduces numerically small corrections to the principal approximation, even in the extreme quantum limit $S = \frac{1}{2}$. Thus, indicated effectiveness of the spin-wave technique – as applied to magnetic systems with small spin quantum numbers and in restricted geometries – may be attributed to the observed weakness of spin-wave interactions.

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5 Simulations of Pure and Doped Low-Dimensional Spin-1/2 Gapped Systems

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Abstract. Low dimensional spin- $\frac{1}{2}$ systems with antiferromagnetic interactions display very innovative features, driven by strong quantum fluctuations. In particular, geometrical effects or competing magnetic interactions can give rise to the formation of a spin gap between the singlet ground state and the first excited triplet state. In this chapter, we focus on the numerical investigation of such systems by Exact Diagonalisation methods and some extensions of it including a simultaneous mean-field treatment of some perturbative couplings. After a presentation of the Lanczos algorithm and a description of the space group symmetries, we give a short review on some pure low-dimensionnal frustrated spin gapped systems. In particular, we outline the role of the magnetic frustration in the formation of disordered phase. A large part is also devoted to frustrated Spin-Peierls systems for which the role of interchain couplings as well as impurity doping effects has been studied numerically.

5.1 Introduction

Many systems of Condensed Matter consist of fermions (electrons) moving on a lattice and experiencing strong repulsive interactions [1]. In such cases, the traditional perturbative methods to treat the electronic correlations often break down. In a pioneering work, Bonner and Fisher [2] revealed the exact diagonalisation (ED) method as a powerful tool to study the properties of one dimensional (1D) spin chains. Later on, it was extended to investigate two dimensional (2D) localised spin systems [3]. This work initiated a more extensive use of the method to investigate a wide variety of different systems such as strongly correlated lattice electrons (Hubbard-like models), mesoscopic systems [4], electron-phonon models, etc....

The success of this method first comes from the rapidly growing power of supercomputers which are being equipped with faster and faster processors and larger and larger memories, disk space and storage facilities. In addition, ED are clearly *unbiased* methods as we shall discuss later on in the course of this Chapter. The systematic errors can be, in most cases, easily estimated and, hence, this method is a very controlled one. Of course, it has its limitations (which we shall also discuss later) but, clearly, the efficiency of this technique will steadily increase in the future as the power of supercomputers booms up. The following Chapter will be dedicated mostly to the numerical technique based on the Lanczos algorithm. However, we shall focus on a specific area of strongly correlated models, namely low-dimensional spin- $\frac{1}{2}$ gapped systems, to illustrate various technical aspects of the method and to discuss the physics of these topics. References to related specialised work dealing in more details with the physical aspects will also be given.

Low dimensional spin- $\frac{1}{2}$ systems with antiferromagnetic (AF) interactions display very innovative features, driven by strong quantum fluctuations. In particular, geometrical effects or competing magnetic interactions can give rise to the formation of a spin gap between the singlet ground state and the first excited triplet state. In this chapter, we focus on the numerical investigation of such systems by Exact Diagonalisation (ED) methods and some extensions of it including a simultaneous mean-field (MF) treatment of some perturbative couplings.

This chapter is organised as follows: in Sect. 2 a description of the Lanczos algorithm is given with special emphasis on the practical use of space group symmetries. A very short review on the well-known planar frustrated Heisenberg model and some linear chain Heisenberg models is given in Sect. 3. In particular, we outline the role of the magnetic frustration in the formation of a disordered phase. We also introduce a MF treatment of interchain couplings. Section 4 is devoted to more recent studies of impurity doping and to the derivation of effective models describing interaction between dopants.

5.2 Lanczos Algorithm

5.2.1 Algorithm

The exact diagonalisation method is based on the Lanczos algorithm [5] which we shall describe here. This algorithm is particularly suited to handle sparse matrices and there is, in fact, a wide variety of lattice models belonging to this class as we shall see later on [6].

Let us consider some lattice model corresponding to some Hamiltonian H with its symmetry group $\mathcal{G} = \{g\}$, namely [H, g] = 0. Let us also assume, for the moment, that irreducible representations of the symmetry group can be constructed. They consist of complete subsets of states $\mathcal{A}_l = \{|\alpha\rangle\}$ which are globally invariant under the application of the Hamiltonian H (we postpone to the next part of this section the explicit construction of these states). Clearly, H can be diagonalized in each of the subsets \mathcal{A}_l independently. It can be written as a tridiagonal matrix in a new orthonormal basis set $\{|\Phi_m\rangle\}$ defined as [7]

$$H|\Phi_{1}\rangle = e_{1}|\Phi_{1}\rangle + b_{2}|\Phi_{2}\rangle,$$

$$\vdots$$

$$H|\Phi_{n}\rangle = e_{n}|\Phi_{n}\rangle + b_{n+1}|\Phi_{n+1}\rangle + b_{n}|\Phi_{n-1}\rangle,$$
(5.1)

where the various coefficients and new basis states can be calculated recursively. The proof is as follows: let us suppose that the procedure has been applied until the step n, i.e. an orthonormal set of states $|\Phi_1\rangle, ..., |\Phi_n\rangle$ has been constructed. Assuming the knowledge of $e_1, ..., e_{n-1}, b_2, ..., b_n, |\Phi_{n-1}\rangle$ and $|\Phi_n\rangle$, one can then determine

$$e_n = \langle \Phi_n | H | \Phi_n \rangle. \tag{5.2}$$

Hence, the new state defined by

$$|\phi_{n+1}\rangle = H|\Phi_n\rangle - e_n|\Phi_n\rangle - b_n|\Phi_{n-1}\rangle, \qquad (5.3)$$

is clearly orthogonal to $|\Phi_n\rangle$. Moreover, $\langle \Phi_{n-1}|\phi_{n+1}\rangle = \langle \Phi_{n-1}|H|\Phi_n\rangle - b_n$ which is also vanishing as can be seen by substituting the expression for $H|\Phi_{n-1}\rangle$. More generally, $|\phi_{n+1}\rangle$ is, in fact, orthogonal to all the previous states $|\Phi_p\rangle$, $p \leq n$ as can be shown recursively. Indeed, let us assume that, for p < n,

$$\forall i, \quad p \le i \le n \quad \langle \Phi_i | \phi_{n+1} \rangle = 0, \tag{5.4}$$

then $\langle \Phi_{p-1} | \phi_{n+1} \rangle = \langle \Phi_{p-1} | H | \Phi_n \rangle$ where $\langle \Phi_{p-1} | \Phi_n \rangle = \langle \Phi_{p-1} | \Phi_{n-1} \rangle = 0$ has been used. Substituting the expression given by (5.2) for $H | \Phi_{p-1} \rangle$ leads to the expected result

$$\langle \Phi_{p-1} | \phi_{n+1} \rangle = 0. \tag{5.5}$$

The (positive) number b_{n+1} is simply defined as a normalisation factor,

$$b_{n+1}^2 = \langle \phi_{n+1} | \phi_{n+1} \rangle, \tag{5.6}$$

i.e. $|\Phi_{n+1}\rangle = \frac{1}{b_{n+1}} |\phi_{n+1}\rangle$.

In principal, a zero vector will be generated after iterating the Hamiltonian a number of times corresponding to the size \mathcal{N}_l of the Hilbert space. However, the number of iterations necessary to obtain the lowest eigenvalues and eigenvectors is much smaller. Typically, for $\mathcal{N}_l \sim 10^6$, the ground state can be obtained with an accuracy better than 10^{-8} , by truncating the procedure after only $N_{it} \sim 100$ iterations and by diagonalizing the resulting tri-diagonal matrix by using a standard library subroutine. However, for a given size \mathcal{N}_l of the Hilbert space, the necessary number N_{it} of iterations might vary by a factor of 2 or 3 depending on the model Hamiltonian. In practice, the convergence is faster for models for which high energy configurations have been integrated out (e.g. t–J models in contrast to Hubbard models). Note however that, in some cases (models with strong finite range interaction), the energy vs N_{it} curve can exhibit steps before convergence to the true ground state is achieved. Once space group symmetries have been implemented, the best choice for the initial state $|\Phi_1\rangle$ is a purely random vector. The ground state is also easily obtained as a function of the states

 $|\Phi_n\rangle$. However, to express it in terms of the initial basis $\{|\alpha\rangle\}$ (as it is often useful) it becomes necessary to store temporarily the intermediate vectors $|\Phi_n\rangle$. This step is usually the most demanding in terms of disk space and/or mass storage. However, note that, at runtime, only three Lanczos vectors of size \mathcal{N}_l need to be assigned in memory.

<u>Full diagonalisation</u>: In some special cases (spectrum statistics [8], thermodynamics [9], etc ...) the complete spectrum (or a least the lower part of it) is needed. This can also be performed by the Lanczos algorithm. In this case, usage of a more sophisticated standard library package (e.g. EA15 of Harwell) is preferable. Indeed, more involved tests become then necessary to eliminate the unphysical "ghost" levels appearing (always above the ground state energy) after the diagonalisation of the tridiagonal matrix. However, the input for the library subroutine consists only on the set of states $|\Phi_n\rangle$ which have to be calculated separately (see below).

5.2.2 Space Group Symmetries

Usually, some efforts have to be carried out in order to take full advantage of the symmetries of the Hamiltonian. Let us consider a model defined on a *D*-dimensional lattice describing interacting fermions as e.g. the simple Heisenberg (spin) model,

$$H_J = \sum_{\mathbf{x}, \mathbf{y}} J_{\mathbf{x}\mathbf{y}} \, \mathbf{S}_{\mathbf{x}} \cdot \mathbf{S}_{\mathbf{y}},\tag{5.7}$$

where interaction (in this case the exchange coupling) is not necessarily restricted to nearest neighbor (NN) sites but, nevertheless, is supposed to exhibit translation and point group symmetry. In other words, denoting the point group [10] by $\mathcal{G}_P = \{g_P\}$, we assume, e.g. in the case of (5.7),

$$J_{\mathbf{x}\mathbf{y}} = J(\mathbf{x} - \mathbf{y})$$

and
$$\forall g_P \in \mathcal{G}_P, \quad J(g_P(\mathbf{r})) = J(\mathbf{r}).$$
 (5.8)

Clearly, such properties are easily generalised to generic spin or fermionic Hamiltonians. In addition, we shall also assume, as in (5.7), spin rotational invariance (the total spin S is a good quantum number) or, at least, invariance of the Hamiltonian under a spin rotation around some quantisation axis. Translation symmetry can be preserved on finite systems provided the geometry is that of a D-dimensional torus with periodic or twisted boundary conditions (BC). On the torus geometry, the full space group reads,

$$\mathcal{G} = \mathcal{G}_P \otimes \mathcal{T},\tag{5.9}$$

where we denote by $\mathcal{T} = \{t_p\}, p = 1, ..., N$, the translation group. It is clear that H_J is invariant under any $g \in \mathcal{G}$.



Fig. 5.1. Schematic representation of of different lattices. (a) the $\sqrt{32} \times \sqrt{32}$ square lattice with NN and second NN couplings J_1 and J_2 . (b) the dimerized ring with dimerization δ . (c) The frustrated ring with NN and second NN couplings J_1 and J_2

5.2.3 Construction of the Hilbert Space

The motivation to take into account the Hamiltonian symmetries is two-fold. First, the Hamiltonian can be block diagonalized, each block corresponding to an irreducible representation of the symmetry group. Practically, the sizes of the various blocks \mathcal{N}_l are much smaller than the size of the full Hilbert space (see e.g. Table 5.1), hence, minimising the numerical effort to diagonalize the Hamiltonian matrix. Secondly, each irreducible representation of the symmetry group is characterised by quantum numbers (such as the momentum \mathbf{k}) which can be connected directly to physical properties.

Table 5.1. Symmetry groups and sizes of the reduced Hilbert spaces for various spin- $\frac{1}{2}$ AF Heisenberg models for one of the most symmetric irreducible representation (typical reduced size is written as the total size in the $S^z = 0$ sector divided by the number of symmetries). The 1D models are described in Sect. 5.3.2. T_N and I_2 stand for the translation group \mathcal{T} (see text) and the spin inversion symmetry $S_{\mathbf{x}}^Z \to -S_{\mathbf{x}}^Z$, respectively

Model	Lattice Size	Symmetry group	Typical reduced Hilbert space size
2D Isotropic	6×6	$T_{36} \otimes C_{4v} \otimes I_2$	9 075 135 300/576
$1D \ J_1 - J_2$	32×1	$T_{32} \otimes C_2 \otimes I_2$	601 080 390/128
1D $J_1 - J_2 - \delta$	32×1	$T_{16} \otimes I_2$	601 080 390/32

We shall focus here on the Heisenberg model (5.7) but t–J and Hubbard models can also be studied. In order to minimise memory occupation, configurations can be stored in binary form. One, first, assumes an arbitrary labelling of the lattice sites from 1 to L and denotes a configuration in real space by

$$|c\rangle = |s_1, ..., s_i, ..., s_L\rangle$$
 (5.10)

For the Heisenberg model, the information s_i on each lattice site \mathbf{x}_i can be stored on a single bit, e.g. \uparrow and \downarrow correspond to 1 and 0 respectively. A

spin configuration with up to 64 spins can then be represented by a single 64-bit word. As an example, a N = 4 site configuration Heisenberg model such as $|\uparrow,\downarrow,\uparrow,\downarrow\rangle$ is coded as $0_{64}...0_51_40_31_20_1$ (where the subscripts indicate the place of the bits) i.e. the integer "10". An integer $N(|c\rangle)$ can then be uniquely associated to each configuration $|c\rangle$ and formally written as,

$$N(|c\rangle) = \sum_{1}^{N} 2^{i-1} \sigma_i , \qquad (5.11)$$

for spin-1/2 models ($\sigma_i = 0$, or 1).

At this point, it becomes useful to consider space group symmetry. Each irreducible representation can be characterised by a momentum

$$\mathbf{K} = \sum_{\mu} n_{\mu} \mathbf{K}_{\mu} \,, \tag{5.12}$$

where \mathbf{K}_{μ} are the reciprocal lattice vectors (e.g., in 2D, $\mathbf{K}_{\mu} = \frac{2\pi}{N} \mathbf{T}_{\mu} \wedge \mathbf{e}_{z}$) and n_{μ} are integers. For each value of \mathbf{K} , one then considers $\mathcal{G}_{\mathbf{K}}^{P}$, the so-called little group of \mathbf{K} ($\mathcal{G}_{\mathbf{K}}^{P} \subset \mathcal{G}_{P}$), containing group elements g_{P} such that

$$g_P(\mathbf{K}) = \mathbf{K} \,. \tag{5.13}$$

The relevant subgroup of \mathcal{G} to be considered is then

$$\mathcal{G}_{\mathbf{K}} = \mathcal{G}_{\mathbf{K}}^P \otimes \mathcal{T} \,. \tag{5.14}$$

For a given symmetry sector $l = (\mathbf{K}, \tau_{\mathbf{K}})$ ($\tau_{\mathbf{K}}$ is one of the irreducible representations of $\mathcal{G}_{\mathbf{K}}^{P}$) a "symmetric" state $|\alpha\rangle$ can then be constructed from a single configuration $|c\rangle$ as a linear combination which reads, up to a normalisation factor,

$$|\alpha\rangle \equiv |\alpha\rangle \{|c\rangle\} = \sum_{g_P \in \mathcal{G}_{\mathbf{K}}^P, t \in \mathcal{T}} e(\tau_{\mathbf{K}}, g_P) \exp\left(i\mathbf{K} \cdot \mathbf{T}_t\right) \, (g_P \, t)(|c\rangle), \qquad (5.15)$$

where $e(\tau_{\mathbf{K}}, g_P)$ are the characters (tabulated) of the representation $\tau_{\mathbf{K}}$ and \mathbf{T}_t are the translation vectors associated to the translations t. Since the procedure to construct the symmetric state is well defined, it is clear that one needs to keep only a single one of the related configurations $g_P t(|c\rangle)$, this state being called "representative" of the symmetric state $|\alpha\rangle$ and denoted by $|r\rangle = R(|c\rangle)$. This naturally implies,

$$\forall g \in \mathcal{G}_{\mathbf{K}}, \ R(g(|c\rangle)) = |r\rangle.$$
(5.16)

In other words, one retains only the configurations $|c\rangle$ which can not be related to each other by any space symmetry $g \in \mathcal{G}_{\mathbf{K}}$. The set of all the representatives (labelled from 1 to \mathcal{N}_l) defines then unambiguously the Hilbert space $\mathcal{A}_l = \{|\alpha\rangle\}$. Typically, the size of this symmetric subspace is reduced by a factor of $\operatorname{card}(\mathcal{G}_{\mathbf{K}})$ compared to the size of the original basis $|c\rangle$. Note that, in some cases, there might exist some configurations $|c\rangle$ (to be eliminated) which do not give rise to any representative. This occurs when there is a subset $\mathcal{G}' \subset \mathcal{G}_{\mathbf{K}}$ such that

$$\forall g \in \mathcal{G}', \qquad g(|c\rangle) = |c\rangle$$

and (5.17)
$$\sum_{g \in \mathcal{G}'} e(\tau_{\mathbf{K}}, g_P(g)) \exp\left(i\mathbf{K} \cdot \mathbf{T}(g)\right) = 0.$$

The choice of the representative among the set of equivalent states (i.e. states related by some symmetry operations of $\mathcal{G}_{\mathbf{K}}$) is, in principle, arbitrary. However, as we shall see in the following, it is convenient to define it as the state $|c\rangle$ of a given class giving rise to the *smallest* integer $N(|c\rangle)$ i.e.

$$N(|r\rangle) = \min_{g \in \mathcal{G}_{\mathbf{K}}} \{ N(g(|c\rangle)) \}$$
(5.18)

For simplicity, we shall here extend our coding procedure to more general Hubbard-like models where one can construct the configurations by a tensorial product of the up and down spin parts

$$|c\rangle = |c(\uparrow)\rangle \otimes |c(\downarrow)\rangle.$$
(5.19)

 $N(|c\rangle)$ contains now 2N bits and is of the form

$$N(|c\rangle) = N'(|c(\uparrow)\rangle) \times 2^N + N'(|c(\downarrow)\rangle), \qquad (5.20)$$

where $N'(|c(\sigma)\rangle)$ corresponds to the binary coding $(N'(|c(\sigma)\rangle) < 2^N))$ of the σ -spin part of the configuration, by using (5.11) where 1 (resp. 0) refers now to the presence (resp. absence) of a spin $\sigma(=\uparrow \text{or}\downarrow)$ at a given site *i*. Although this coding is more costly in term of memory space (2N bits per configuration instead of N), it is more general and applies both to Hubbard-like models (where \uparrow and \downarrow spins can leave on the same site) and to Heisenberg models. The minimisation of $N(g(|c\rangle))$ over $g \in \mathcal{G}_{\mathbf{K}}$ can be done in two steps. First, one generates all possible configurations for the up spins (this usually involves a limited number of states) and only representatives $|r(\uparrow)\rangle$ are kept. At this stage, one needs to keep track of the subsets of symmetries $\mathcal{E}_{\mathbf{K}}[r(\uparrow)\rangle]$ of $\mathcal{G}_{\mathbf{K}}$ leaving these representatives invariant. In a second step, one constructs the full set of configurations as a tensorial product of the form $|r(\uparrow)\rangle \otimes |c(\downarrow)\rangle$. The remaining symmetries of $\mathcal{E}_{\mathbf{K}}[r(\uparrow)\rangle]$ are then applied to the spin \downarrow part and one only retains the configurations $|c(\downarrow)\rangle$ such that,

$$\forall g' \in \mathcal{E}_{\mathbf{K}}[r(\uparrow)\rangle] \ N'(|c(\downarrow)\rangle) \le N'[g'(|c(\downarrow)\rangle)].$$
(5.21)

The Hilbert space is then formally defined by all the symmetric states $|\alpha\rangle\{|r\rangle\}$ (see (5.15)). However, one only needs to store the binary codes $N(|r\rangle)$ as well

as the normalisations of the related symmetric states. The normalisation of (5.15) requires some caution. In some cases, there might exist more than a single (i.e the identity operator \mathbb{I}) group element g' of $\mathcal{E}_{\mathbf{K}}[|r(\uparrow)\rangle]$ which keeps $|r\rangle$ invariant. Then,

$$\mathcal{F}_{\mathbf{K}}[|r\rangle] = \{g' \in \mathcal{E}_{\mathbf{K}}(|r(\uparrow)\rangle); N'[g'(|c(\downarrow)\rangle)] = N'(|c(\downarrow)\rangle)\}$$
(5.22)

defines the subgroup of such symmetry operations. The sum over the group elements in (5.15) should, in fact, be restricted to the elements of the coset $\mathcal{G}_{\mathbf{K}}/\mathcal{F}_{\mathbf{K}}[|r\rangle]$ and the appropriate normalisation factor is then given by

$$n(|r\rangle) = \left[\frac{\operatorname{card}\{\mathcal{F}_{\mathbf{K}}[|r\rangle]\}}{\operatorname{card}\{\mathcal{G}_{\mathbf{K}}\}}\right]^{1/2}.$$
(5.23)

The two integers $N(|r\rangle)$ and $n(|r\rangle)$ corresponding to the binary code of a representative and to the normalisation of the related symmetric state, respectively, can be combined and stored in a single 64-bit computer word.

In the case of the generic Heisenberg model, the previous two-step procedure can, in most cases, also be done using the N bits coding from (5.11) but it is more involved. We only indicate here the spirit of the method. More technical details for spin-1/2 models can be found in Refs. [11] and [12]. The decomposition between up and down spins is replaced here by an (appropriate) partition of the lattice sites into two subsets A and B. The computer word (integer) (5.11) coding each configuration contains then two parts, each part corresponding to one subset of the lattice sites, $|c(A)\rangle$ and $|c(B)\rangle$. The partition is chosen in a way such that the group $\mathcal{G}_{\mathbf{K}}$ can be decomposed as

$$\mathcal{G}_{\mathbf{K}} = \mathcal{G}_{\mathbf{K}}^{S} + \mathcal{S}(\mathcal{G}_{\mathbf{K}}^{S}), \qquad (5.24)$$

where S is a group symmetry which fulfils $S^2 = \mathbb{I}$, the subgroup $\mathcal{G}_{\mathbf{K}}^S$ leaves the two subsets of lattice sites *globally* invariant and $\mathcal{S}(\mathcal{G}_{\mathbf{K}}^S)$ is the coset of S relative to $\mathcal{G}_{\mathbf{K}}^S$. The decomposition (5.24) is not always unique. For 2D clusters such as the $\sqrt{32} \times \sqrt{32}$ lattice of Fig. 5.1, a convenient choice for Sis a reflection symmetry. Note that, in the case of 1D rings, (5.24) is only possible if the number of sites is of the form N = 4p + 2 or for very special values of the momentum K (like K = 0 or $K = \pi$). Then, the previous procedure can be extended to this case by writing the configurations as

$$|c\rangle = |c(A)\rangle \otimes |c(B)\rangle \tag{5.25}$$

and by (i) applying the subgroup $\mathcal{G}^{S}_{\mathbf{K}}$ on $|c(A)\rangle$ to generate its corresponding representative $|r(A)\rangle$ and then (ii) applying all the symmetries of $\mathcal{E}_{\mathbf{K}}[|r(A)\rangle]$ on the part $|c(B)\rangle$. The action of the remaining symmetry \mathcal{S} is considered at last; if the lattice sites are labelled in such a way that

$$\mathbf{x}_{i+N/2} = \mathcal{S}(\mathbf{x}_i), \qquad (5.26)$$

for $i \leq N/2$, the application of S can be implemented as a simple permutation of the two sub-words of $N(|c\rangle)$.

5.2.4 Construction of the Hamiltonian Matrix

We turn now to the implementation of the basic operation $|\Phi_n\rangle \rightarrow H|\Phi_n\rangle$ appearing in (5.2) which is always specific to the model Hamiltonian H and constitutes the central part of the Lanczos code. Since the states $|\Phi_n\rangle$ are expressed in terms of the symmetric states $|\alpha\rangle$ of (5.15) the Hamiltonian matrix has to be computed in this basis. For this purpose, it is only necessary to apply H on the set of representatives $|r_{\gamma}\rangle$ (labelled from 1 to \mathcal{N}_l). In general, each configuration $|r\rangle$ leads to a small number β_{max} (at most equal to the number of terms in H) of generated states,

$$H|r_{\gamma}\rangle \propto \sum_{\beta=1}^{\beta_{max}} (-1)^{\theta_{\gamma,\beta}} |c_{\gamma,\beta}\rangle ,$$
 (5.27)

where different signs $(-1)^{\theta_{\gamma,\beta}}$ might arise (in the case of fermion models) from fermionic commutation relations. The matrix is then very sparse. Note that the full Hamiltonian can always be split in a small number of separate terms so that the amplitude of the matrix elements in (5.27) is just a constant and hence does not need to be stored.

At this point, it becomes necessary to determine the representatives (in binary form) of the various generated states on the right hand side of (5.27) by applying to them all the symmetries of $\mathcal{G}_{\mathbf{K}}$. To achieve this, the choice of (5.20) for the binary coding of the configurations is very convenient. It is, indeed, a simple way to take advantage of the natural decomposition of the generated states,

$$|c_{\gamma,\beta}\rangle = |c_{\gamma,\beta}(\uparrow)\rangle \otimes |c_{\gamma,\beta}(\downarrow)\rangle .$$
(5.28)

Although, we restrict ourselves here, for sake of simplicity, to the general coding of the Hubbard-like models, the following procedure can be straightforwardly applied using the more restrictive Heisenberg form (5.25) for the configurations $|c_{\gamma,\beta}\rangle$. The calculation of the representative

$$|r_{\gamma,\beta}\rangle_f = R\{|c_{\gamma,\beta}(\uparrow)\rangle \otimes |c_{\gamma,\beta}(\downarrow)\rangle\}$$
(5.29)

can be done in two steps. First, one applies all the symmetries of $\mathcal{G}_{\mathbf{K}}$ to $|c_{\gamma,\beta}(\uparrow)\rangle$. Since this procedure has to be repeated a large number of times, the function

$$R: |c(\uparrow)\rangle \longmapsto |r(\uparrow)\rangle. \tag{5.30}$$

can be, in fact, tabulated, prior to the actual calculation of the matrix elements. This is made possible since the number of possible states $|c(\uparrow)\rangle$ remains, in general quite modest. This procedure enormously speeds up the calculation of the representatives and justifies the choice of (5.20). Note that one also needs, in this preliminary calculation, to store, for each configuration $|c(\uparrow)\rangle$, the corresponding ensembles,

$$\mathcal{R}_{\mathbf{K}}[(|c(\uparrow)\rangle] = \{g \in \mathcal{G}_{\mathbf{K}}; g(|c(\uparrow)\rangle) = |r(\uparrow)\rangle\}.$$
(5.31)

This also requires limited storage since, in most cases, $\mathcal{R}_{\mathbf{K}}[|c(\uparrow)\rangle]$ contains just a single element. In a second step, it is sufficient to apply only the remaining symmetries of $\mathcal{R}_{\mathbf{K}}[|c_{\gamma,\beta}(\uparrow)\rangle]$ to the $|c_{\gamma,\beta}(\downarrow)\rangle$ part. A standard hashing table [13] is then used to find the positions of the representatives in the list. The connectivity matrix connecting the labels of the initial set of representatives to the labels of the new set of generated states is then stored on disk.

Note that the phases related to commutations of fermion operators and/or to the characters of the symmetry operations involved in the transformation of the generated states to their representatives have also to be kept. These phases have the general form,

$$\lambda_{\gamma,\beta} = (-1)^{\theta_{\gamma,\beta}} e(\tau_{\mathbf{K}}, g_P(g^*_{\gamma,\beta})) \exp\left(i\mathbf{K} \cdot \mathbf{T}(g^*_{\gamma,\beta})\right) , \qquad (5.32)$$

where $g^*_{\gamma,\beta}$ is a group element of $\mathcal{R}_{\mathbf{K}}[|c_{\gamma,\beta}(\uparrow)\rangle]$ such that

$$|r_{\gamma,\beta}\rangle_f = g^*_{\gamma,\beta}(|c_{\gamma,\beta}\rangle) , \qquad (5.33)$$

which depends on γ and β . It is easy to show that, if there exists more than a single group element which fulfils (5.33) then, all of them lead to the same phase factor (5.32).

Since the number of possible different phases given by (5.32) is quite small, it is possible to store the $\lambda_{\gamma,\beta}$ (in some convenient integer form) together with $N\{|r_{\gamma,\beta}\rangle_f\}$ on a small number of computer bits. For an Hilbert space of 10^8 (hundred millions) representatives with typically an average of ~ 50 images per state, the occupation of the disk is of the order of 5000 Mw i.e. 40 Gb. This can even be reduced by a factor of two by using each computer word to store the informations corresponding to two images instead of a single one. Once it has been generated, the Hamiltonian matrix is cut into several pieces (typically of the order of 10 to 100 Mw) and the various parts are successively read from the disk in order to calculate $H|\Phi_n\rangle$. The best performances are obtained when the calculation using the nth part of the matrix and the access to the disk to read the $(n+1)^{th}$ part are simultaneous. Note that the Lanczos algorithm as it has been described above is well adapted to be implemented on a vector supercomputer (e.g. on a NEC-5 × 5).

5.3 Examples of Translationally Invariant Spin Gapped Systems

Here, we first restrict ourselves to systems where the symmetry analysis decribed above can be used. Note however that explicit symmetry breaking may be present but, in general, the remaining symmetry group contains a large number of symmetries which can be exploited. Note also that since spontaneous symmetry breaking can occur only in the thermodynamic limit, it does not prevent for finite systems the previous symmetry analysis.

5.3.1 Application to the 2D $J_1 - J_2$ Model

The study of quantum phase transitions in 2D is of great interest. One of the standard example is the so-called frustrated Heisenberg model (see Fig. 5.1a). It is defined by (5.7) when only NN and second NN exchange couplings are retained,

$$J_{\mathbf{x}\mathbf{y}} = J_1 \text{ if } |\mathbf{x} - \mathbf{y}| = 1,$$

$$J_{\mathbf{x}\mathbf{y}} = J_2 \text{ if } |\mathbf{x} - \mathbf{y}| = \sqrt{2},$$

$$J_{\mathbf{x}\mathbf{y}} = 0 \text{ otherwise }.$$
(5.34)

Various analytical approaches have been applied to this problem such as spinwave calculations [14], large-N SU(N) theories [15], series expansions [16], and Schwinger boson mean field approaches [17]. However, most of these methods are somewhat biased since they assume the existence of some particular ground states. Pioneering unbiased ED studies [18–20] have strongly suggested the existence of a disordered magnetic phase for intermediate couplings J_2/J_1 . Here, we briefly discuss some more recent subsequent work [11,12,21] which attempted to obtain more accurate results by a finite size scaling analysis. Similar studies have also been performed for other S = 1/2 2D spin models like the triangular lattice [22], the Kagome lattice [23], the 1/5-depleted square lattice [24] or the (2D) pyrochlore lattice [25].



Fig. 5.2. Finite size results for $M_N^2(\pi, \pi)$ for different values of J_2 . The dashed lines are least squares fits to the data, using all available clusters. The full lines are fits using only N = 20, 32, 36 (Reprinted from [12])

A magnetic ordered phase with a spin modulation \mathbf{Q} can be characterised by an order parameter $M_N(\mathbf{Q})$ defined by

$$M_N^2(\mathbf{Q}) = \frac{1}{N(N+2)} \langle \Psi_0 | (\sum_{i=1}^N \exp\left(i \,\mathbf{x}_i \cdot \mathbf{Q}\right) \mathbf{S}_i)^2 | \Psi_0 \rangle , \qquad (5.35)$$

where $|\Psi_0\rangle$ is the ground state. It is important to notice that, in any finite system, the order parameter itself has a zero expectation value in the ground state due to spin SU(2) symmetry. In other words, the macroscopic magnetisation can slowly fluctuate so that in average it vanishes. It is therefore essential to consider, as in (5.35), the square of the order parameter which can be interpreted as a generalised susceptibility.

For weak frustration J_2/J_1 , Néel order with $\mathbf{Q} = (\pi, \pi)$ is expected, while for large ratio J_2/J_1 a collinear phase with $\mathbf{Q} = (\pi, 0)$ or $\mathbf{Q} = (0, \pi)$ consisting of successive alternating rows of parallel spins is a serious candidate. Indeed, in such a collinear phase, each sub-lattice has Néel order so it is clear that it is stabilised by J_2 . Note that the normalisation factor of the staggered magnetisations (5.35) is chosen so that the order parameter is independent of the size in a perfect classical Néel or collinear state. In such ordered phases where the continuous spin symmetry is spontaneously broken, field theory arguments [26] suggest a scaling of the form,

$$M_N(\mathbf{Q}) \simeq m_0(\mathbf{Q}) + \frac{C(\mathbf{Q})}{N^{1/2}}$$
 (5.36)



Fig. 5.3. Comparison of the finite size fits for the anti-ferromagnetic and collinear order parameters (left and right curves, respectively) with linear spin wave theory (Reprinted from [12])

Square clusters with N = 4p sites (so that $(\pi, 0)$ belongs to the reciprocal lattice) are considered (see Sec. II.B), i.e. N=16, 20, 32 and 36. As seen in Fig. 5.2 corresponding to $\mathbf{Q} = (\pi, \pi)$ the scaling law (5.36) is very well satisfied. Note that the 4×4 cluster shows systematic deviations. Similar results are also obtained for the collinear order parameter at larger J_2/J_1 ratios.

The extrapolated results are shown in Fig. 5.3. The most interesting feature is the existence of a narrow range of J_2/J_1 around 0.5 where none of the ordered states is stable. Various candidates for this disordered phase have been proposed such as the dimer phase [15] and investigated numerically [18, 20, 21]. More details on this topic can be found e.g. in the review article [11].

5.3.2 Application to Spin-Peierls Chains

a) Purely 1D Models

We now move to systems with anisotropic couplings in space (quasi 1D materials). Let us first consider purely 1D models in order to describe the physical origin of the spin-Peierls (SP) transition. One of the simplest 1D model which diplays such a behavior is the frustrated spin- $\frac{1}{2}$ ring (see Fig. 5.1c), also called $J_1 - J_2$ or zig-zag chain, described by the Hamiltonian

$$H_{\text{frust}} = \sum_{i=1}^{L} (J_1 \boldsymbol{S}_i . \boldsymbol{S}_{i+1} + J_2 \boldsymbol{S}_i . \boldsymbol{S}_{i+2}).$$
(5.37)

Its symmetry properties are given in Table 5.1 for L = 32 sites.

The low energy properties of such a model are very interesting because it is gapless as long as $\alpha = J_2/J_1$ remains smaller than a critical value $\alpha_c \simeq 0.2412$ ([29], Fig. 4). For $\alpha > \alpha_c$, a gap $\Delta_S(\alpha) \propto e^{-(\alpha - \alpha_c)^{-1}}$ develops and a spontaneous dimerization appears, characteristic of the SP transition. At $\alpha = 0.5$, the so-called Majumdar-Ghosh (MG) point [30], the 2-fold degenerate ground state is known exactly and consists in the product of spin singlets located either on odd or on even bonds. Beyond the MG point, the shortrange correlations become incommensurate. The triplet (S = 1) spectrum of the SP phase is a two-particle (so-called kink or soliton) continuum as evidenced by the scaling of the soliton-antisoliton binding energy to zero [31].

Adding an explicit dimerization , i.e. a rigid modulation δ of the NN coupling (see Fig. 5.1), drives immediately the ground state into a SP phase for any $\delta \neq 0$ even if $J_2 = 0$. In term of symmetries, C_2 and the translations of an odd number of lattice spacings are lost (see Fig. 5.1b and Table 5.1)). If $J_2 \neq 0$, the dimerized *and* frustrated model

$$H_{\rm dim} = \sum_{i=1}^{L} J_1 \left[(1 + (-1)^i \delta) \boldsymbol{S}_i \cdot \boldsymbol{S}_{i+1} + \alpha \boldsymbol{S}_i \cdot \boldsymbol{S}_{i+2} \right]$$
(5.38)



Fig. 5.4. Critical value α_c of the frustration vs the inverse square of the system size obtained using the method developped in [27]. (Reprinted from [28])

displays an enhancement of the dimerized gapped phase, indeed $\Delta_S(\alpha, \delta) - \Delta_S(\alpha, 0) \propto \delta^{2/3}$ [32].



Fig. 5.5. Phase diagram of the frustrated dimerized Heisenbeg AF spin- $\frac{1}{2}$ chain in the $\alpha - \delta$ plane. The dotted line is the Shastry-Sutherland line $(2\alpha + \delta = 1)$ [33]. On its left side, the phase is SP gapped and commensurate whereas on the right side, the correlations are incommensurate

These properties are summarized in the phase diagram shown in Fig. 5.5. Note that the static modulation δ leads to soliton-antisoliton boundstates as shown in Fig. 5.6.

b) Chain Mean Field Theory for Coupled Spin Chains

Physically, the previous models are often inadequate to describe the properties of several compounds like $CuGeO_3$ [34] or LiV_2O_5 [35] which are


Fig. 5.6. Lowest lying triplet(•), singlet (•) and quintuplet (\otimes) excitations vs. the wave vector k for the dimerized frustrated chain (5.38) with $J_2 = 0.5$, $\delta = 0.05$, L = 28. Results for L = 32, k = 0 are shown to the left. ED results reprinted from [36]

excellent realizations of weakly interacting frustrated spin- $\frac{1}{2}$ chains. Let us first consider a set of frustrated spin chains which are coupled by a weak AF exchange J_{\perp} . This 2D model is governed by the following Hamiltonian

$$H_{2D}(\alpha, J_{\perp}) = \sum_{i=1}^{L} \sum_{a=1}^{M} [\mathbf{S}_{i,a} \cdot \mathbf{S}_{i+1,a} + \alpha \mathbf{S}_{i,a} \cdot \mathbf{S}_{i+2,a} + J_{\perp} \mathbf{S}_{i,a} \cdot \mathbf{S}_{i,a+1}].$$
(5.39)

where *i* is the lattice index along the chains of lenght *L* and *a* labels the *M* chains (*L* and *M* are chosen to be even and periodic boundary conditions are assumed in both directions). Obviously it should be possible to study exactly this spin model on the square lattice but, as we have seen above, it is hard to perform ED with system larger than 36 spins. Here, we take advantage of the fact that $J_{\perp} \ll 1$ to perform a MF treatment of the transverse coupling. Following Schulz [37], the chain mean-field (CMF) version of (5.39) is given by

$$H_{2D}^{\mathrm{MF}}(\alpha, J_{\perp}) = \sum_{i=1}^{L} \sum_{a=1}^{M} [\boldsymbol{S}_{i,a}.\boldsymbol{S}_{i+1,a} + \alpha \boldsymbol{S}_{i,a}.\boldsymbol{S}_{i+2,a} + h_{i,a}S_{i,a}^{z} - J_{\perp}\langle S_{i,a}^{z}\rangle\langle S_{i,a+1}^{z}\rangle], \qquad (5.40)$$

with

$$h_{i,a} = J_{\perp}(\langle S_{i,a+1}^z \rangle + \langle S_{i,a-1}^z \rangle), \qquad (5.41)$$

the local magnetic field to be computed self-consistently. In the absence of dopant (see Sect. 4), we expect an homogeneous AF phase characterized by a self-consistent staggered magnetization $\langle S_{i,a}^z \rangle = (-1)^{i+a}m$. Therefore the coupled chains problem is reduced to a single chain in a staggered magnetic field $h_i = \pm 2(-1)^i J_{\perp}m$.

$$H_{\text{single}}(\alpha, J_{\perp}) = \sum_{i=1}^{L} [\boldsymbol{S}_i \cdot \boldsymbol{S}_{i+1} + \alpha \boldsymbol{S}_i \cdot \boldsymbol{S}_{i+2} + 2m J_{\perp} (-1)^i S_i^z] + \text{constant},$$
(5.42)

and the symmetry group of such a model is $T_{L/2}$. In the absence of frustration $(\alpha = 0)$, it was shown that $m \sim \sqrt{J_{\perp}}$ [37]. By solving the self-consistency condition using ED of finite chains the transition line $J_{\perp} = J_{\perp}^{c}(\alpha)$ (see Fig. 5.7) separating the dimerised SP phase (m = 0) and the AF ordered phase (for which $m \neq 0$) has been obtained in agreement with field theoretic approaches [38]. Finite size effects are small in the gapped regime and especially at the MG point. Note also that numerical data suggest that the AF order sets up at arbitrary small coupling when $\alpha < \alpha_c$ with a clear finite size scaling $J_{\perp}^{c}(L) \propto 1/L$ at small α .



Fig. 5.7. Phase diagram of the coupled frustrated chains as a function of the frustration α and the inter-chain magnetic coupling J_{\perp} . The points, calculated for 2 different chain sizes (12 and 16 sites), separate a dimerized phase (SP) from a Néel ordered phase (AF). The closed diamond shows the order-disorder critical point at $\alpha_c \simeq 0.2412$. The dashed line represents the expected behavior in the thermodynamic limit. In the inset, the staggered magnetization $m(J_{\perp})$ has been calculated for a L = 12 sites chain along the MG line (dot-dashed line). Along the $\alpha = 0$ line, different symbols show the critical J_{\perp} for L = 8, 12, 16, 18 from top to bottom and we have checked its scaling to 0 according to a 1/L law

c) Convergence Issues of the Numerical CMF

The numerical procedure consists of successive Lanczos-diagonalizations of a frustrated (α) spin-1/2 chain. At each step the AF order parameter m is calculated and reinjected at the following step as the "new field". Starting the numerical procedure with an arbitrary value of $m(0) \neq 0$, the chain of size L is first diagonalized, m(1) is extracted and then used for the next iteration. Eventually the procedure converges to the fixed point m^* . A very interesting feature is that the convergence to m^* as function of the number of MF iterations p (see Fig. 5.8) is exponential.

$$m(p) - m^* \propto \exp(-p/\xi_\tau) \quad \text{for } p >> \xi_\tau, \tag{5.43}$$

with $\xi_{\tau}(J_{\perp})$ a typical convergence time scale.

In order to study convergence at large p we have considered here small systems (12 sites) at the MG point where the finite size effects are very small. We have checked this convergence issue by studing the speed V(p) = |m(p+1) - m(p)| which is exponentially vanishing

$$V(p) \propto \exp(-t)$$
 for $t = \frac{p}{\xi_{\tau}} >> 1$, (5.44)



Fig. 5.8. Universal behavior of the convergence speed of the MF iterative procedure plotted versus the renormalized iteration index $t = \frac{p}{\xi_{\tau}}$. Results are shown for a system of spins interacting via (5.42) with L = 12 and $\alpha = 0.5$. From top to bottom $J_{\perp} = 0.075, 0.13, 0.1, 0.11, 0.108, 0.106$. An initial value m(0) = 1/2 is used for all simulations. Convergences to $m^* = 0$ if the phase is SP (solid lines) or $m^* \neq 0$ (long-dashed lines) if the pase is AF are obtained. The inset shows the behavior of the typical convergence time scale ξ_{τ} as a function of the distance to the critical point $\delta J_{\perp} = J_{\perp} - J_{\perp}^c$. The curves are power law fits (see text)

as we can see in Fig. 5.8. It is very important to note that the convergence of the MF procedure is universal in the sense that the choice of the starting value m(0) is not crucial.

The time scale ξ_{τ} is J_{\perp} -dependant and diverges as a power law $\xi_{\tau} \sim |\delta J|^{-\mu}$ when approaching the critical line where $|\delta J| = |J_{\perp} - J_{\perp}^c|$. Our datas suggest $\mu \simeq 1.06$ if $J_{\perp}^c > J_{\perp}$ and $\mu \simeq 0.95$ if $J_{\perp} > J_{\perp}^c$ for L = 12 (see inset of Fig. 5.8).

5.4 Lanczos Algorithm for Non-uniform Systems: Application to Doped SP Chains

Doping a SP system with non-magnetic impurities leads to very surprising new features. For example in $Cu_{1-x}M_xGeO_3$ (M=Zn or Mg), the discovery of coexistence between dimerization and AF long range order at small impurity concentration has motivated extented experimental [39] and theoretical [36, 40–44] investigations. In the following we report numerical studies of models for doped coupled spin chains. For sake of completness we also include a four-spin coupling which originates from cyclic exchange [45, 46].

5.4.1 Doped Coupled Frustrated Spin- $\frac{1}{2}$ Chain with Four-Spin Exchange

As for the transverse coupling J_{\perp} in (5.40,5.41), we also apply the MF treatment to the added 4-spin coupling $J_4(\mathbf{S}_{i,a} \cdot \mathbf{S}_{i+1,a})(\mathbf{S}_{i+1,a+1} \cdot \mathbf{S}_{i,a+1})$. This leads to a self-consistent modulation of the NN couplings

$$H_{\text{eff}}(\alpha, J_{\perp}, J_4) = \sum_{i,a} [(1 + \delta J_{i,a}) \boldsymbol{S}_{i,a} \cdot \boldsymbol{S}_{i+1,a} + \alpha \boldsymbol{S}_{i,a} \cdot \boldsymbol{S}_{i+2,a} + h_{i,a} S_{i,a}^z] + \text{constant}, \qquad (5.45)$$

with $h_{i,a}$ given by (5.41) and

$$\delta J_{i,a} = J_4\{\langle \boldsymbol{S}_{i,a+1} \cdot \boldsymbol{S}_{i+1,a+1} \rangle + \langle \boldsymbol{S}_{i,a-1} \cdot \boldsymbol{S}_{i+1,a-1} \rangle\}.$$
(5.46)

Such a modulation produced by the J_4 term stabilizes the SP phase and raises the transition line in Fig. 5.7 (for more details, see [47]). Another interesting feature of this model is its direct link with the magneto-elastic model considered in [43] where the elastic coupling K plays a role very similar to that of $1/J_4$. In the following the parameters α , J_{\perp} and J_4 are set in order to constrain the system to be in a SP state in the absence of dopants.

A dopant is described as an inert site i.e. all couplings to this site will be set to zero. Contrary to what we have seen previously, the use of the translation invariance is now forbidden by the presence of a single defects or by randomly located defects (see Fig. 5.9). The maximal size accessible with a



Fig. 5.9. Schematic picture of the coupled chains model with nearest neighbor, next-nearest neighbor, inter-chain and 4-spin couplings J_1 , $J_2 = \alpha J_1$, J_{\perp} , and J_4 . Full (resp. open) circles stand for spin- $\frac{1}{2}$ sites (resp. non-magnetic dopants)

Lanczos procedure is then reduced because of this lack of symmetry and also because of the repeated iterative MF procedure. Indeed, the problem can not be reduced to a single chain model and hence the M non-equivalent chains have to be diagonalized independently. Following the method used in [44], the MF equations are solved self-consistently on finite $L \times M$ clusters. Therefore, in the doped case, the time scale of the MF convergence ξ_{τ} for the single chain problem (5.42) is typically multiplied by M.

5.4.2 Confinement

Replacing a single spin- $\frac{1}{2}$ in a *spontaneously* dimerized (isolated) spin chain by a non magnetic dopant (described as an inert site) liberates a free spin $\frac{1}{2}$, named a soliton, which does not bind to the dopant [36]. The soliton can be depicted as a single unpaired spin (domain) separating two dimer configurations [36]. The physical picture is completely different when a *static* bond dimerisation exists and produces an attractive potential between the soliton and the dopant [36, 40] and consequently leads, under doping, to the formation of local magnetic moments [36, 42] as well as a rapid suppression of the spin gap [41]. However, a coupling to a purely one-dimensional (1D) adiabatic lattice [43] does not produce confinement in contrast to more realistic models including an elastic inter-chain coupling (to mimic 2D or 3D lattices) [43, 44].

Here, we re-examine the confinement problem in the context of the previous model including interchain magnetic coupling.

a) Different Kinds of Dimer Orders

Let us return to model (5.45). For $J_4 = 0$, the MF treatment of the transverse magnetic coupling J_{\perp} does not break the degeneracy of the ground state: each chain displays a 2-fold degenerate ground state (dimers can stand either on even or odd bonds) independently from the other ones. The situation changes radically when $J_4 \neq 0$ because the degeneracy is reduced to 2. Indeed, each



Fig. 5.10. Energy difference $\Delta_{\text{Stg-Clm}}$ between ground states with staggered and collumnar dimer orders plotted versus J_4 for model (5.45) (without dopant) at $\alpha = 0.5$, $J_{\perp} = 0.1$ and L = 12

(a)
$$J_4 = 0$$

(b) $J_4 < 0$
(c) $J_4 > 0$

Fig. 5.11. Schematic picture of the soliton confinement mechanism induced by the coupling J_4 of the model (5.45). The non magnetic dopant is represented by an open circle and the large black bonds stand for stronger dimer bonds. The black arrow represents the soliton, released by the impurity, which is deconfined if $J_4 = 0$ (a) whereas it is linked to the dopant if $J_4 \neq 0$. We can see that this binding is imposed by the bulk dimerization which is columnar if $J_4 < 0$ (b) or staggered if $J_4 > 0$ (c)

chain displays the same dimerized pattern if $J_4 < 0$ (columnar dimer order) whereas the dimer order is staggered in the transverse direction if $J_4 > 0$, as we can see in Fig. 5.10. Consequently, the soliton remains deconfined [48] when $J_4 = 0$ as we can observe in Figs. 5.11,5.12. On the other hand, if $J_4 \neq 0$ the bulk dimerization constrains the soliton to lie in the vicinity of the impurity (see Fig. 5.11).

b) Enhancement of the Magnetization near a Dopant

Under doping, the system becoming inhomogeneous, we define a local mean staggered magnetization

$$\mathcal{M}_{i,a}^{\text{stag}} = \frac{1}{4} (-1)^{i+a} (2\langle S_{i,a}^z \rangle - \langle S_{i+1,a}^z \rangle - \langle S_{i-1,a}^z \rangle)$$
(5.47)



Fig. 5.12. Local magnetization $\mathcal{M}_{i,a}^{\text{stag}}$ for $L \times M = 16 \times 8$ coupled chains with one dopant D (shown by arrow) located at a = 1, i = 16 in the dimerised phase ($\alpha = 0.5, J_{\perp} = 0.1$). Circles correspond to $J_4 = 0$ (shown up to the third neighbor chain of the doped one) and squares (crosses) to $J_4 = 0.01$ ($J_4 = 0.08$). The coupling J_2 across the dopant has been set to 0 for convenience (Reprinted from [47])

which has been calculated for a single dopant in a system of size $L \times M = 16 \times$ 8. It is plotted for different values of the four-spin coupling in Fig. 5.12 where the confinement mechanism can clearly be observed. Note that the inter-chain coupling induces a "polarization cloud" with strong AF correlations in the neighbor chains of the doped one.

c) Confinement Length

In order to measure the strength of confinement, a confinement length can be defined as

$$\xi_{\parallel} = \frac{\sum_{i} i |S_{i}^{z}|}{\sum_{i} |S_{i}^{z}|}.$$
(5.48)

In the absence of confinement, the solitonic cloud is located at the center of the doped chain: $\xi_{\parallel} = L/2$. Otherwise, ξ_{\parallel} converges to a finite value when $L \to \infty$. In Fig. 5.13, the confinement lenght is plotted versus J_4 for 2 different system sizes at $\alpha = 0.5$ and $J_{\perp} = 0.1$. The finite size effects decrease for increasing J_4 . Note that $\xi_{\parallel}(J_4) \neq \xi_{\parallel}(-J_4)$ and a power law [40] with different exponents η is expected when $J_4 \to 0$. A fit gives $\eta \sim 0.33$ if $J_4 < 0$ and $\eta \sim 0.50$ for $J_4 > 0$ (Fig. 5.13). This asymmetry can be understood from opposite renormalisations of J_1 for different signs of J_4 . Indeed, if $J_4 < 0$ then $\delta J_{i,a} > 0$ and the nearest neighbor MF exchange becomes larger than the bare one. Opposite effects are induced by $J_4 > 0$.



Fig. 5.13. ED data of the soliton average position vs J_4 calculated for $\alpha = 0.5$ and $J_{\perp} = 0.1$. Different symbols are used for $L \times M = 12 \times 6$ and 16×8 clusters. The long-dashed line is a power-law fit (see text). Inset shows the magnetization profile in the doped (a = 1) chain at $J_4 = 0.08$, ie $\xi_{\parallel} \simeq 2.5$ (Reprinted from [47])

5.4.3 Effective Interaction

We now turn to the investigation of the effective interaction between dopants. A system of coupled chains with two dopants is considered here (see Fig. 5.9). Each impurity releases an effective spin $\frac{1}{2}$, localized at a distance $\sim \xi_{\parallel}$ from it due to the confining potentiel set by J_4 . We define an effective pairwise interaction J^{eff} as the energy difference of the S = 1 and the S = 0 ground states. When $J^{\text{eff}} = E(S = 1) - E(S = 0)$ is positive (negative) the spin interaction is AF (ferromagnetic). Let us first consider the case of two dopants in the same chain. (i) When the two vacancies are on the same sub-lattice the moments experience a very small ferromagnetic $J^{\text{eff}} < 0$ as seen in Fig. 5.14 with $\Delta a = 0$ so that the two effective spins $\frac{1}{2}$ are almost free. (ii) When the two vacancies, Δi is odd and the effective coupling is AF with a magnitude close to the singlet-triplet gap. Fig. 5.14 with $\Delta a = 0$ shows that the decay of J^{eff} with distance is in fact very slow for such a configuration. Physically, this result shows that a soliton and an anti-soliton on the same chain and different sublattices tend to recombine.

The behavior of the pairwise interaction of two dopants located on *dif*ferent chains ($\Delta a = 1, 2, 3, 4$) is shown on Fig. 5.14 for $\Delta a = 1, 2, 3, 4$ for $J_4 > 0$. When dopants are on opposite sub-lattices the effective interaction is antiferromagnetic. At small dopant separation $J^{\text{eff}}(\Delta i)$ increases with the dopant separation as the overlap between the two AF clouds increases until $\Delta i \sim 2\xi$. For larger separation, $J^{\text{eff}}(\Delta i)$ decays rapidly. Note that the released spin- $\frac{1}{2}$ solitons bind on the opposite right and left sides of the dopants as imposed by the the bulk dimerisation [49]. If dopants are on the same



Fig. 5.14. Magnitude of the effective magnetic coupling between two impurities located either on the same chain (a) or on different ones (b-c-d) vs the dopant separation Δi in a system of size $L \times M = 16 \times 8$ with $\alpha = 0.5$, $J_{\perp} = 0.1$, and $J_4 = 0.08$. Closed (resp. open) symbols correspond to AF (F) interactions

sub-lattice, solitons are located on the same side of the dopants [50] and the effective exchange $J^{\text{eff}}(\Delta i)$ is ferromagnetic and decays rapidly to become negligible when $\Delta i > 2\xi$. The key feature here is the fact that the effective pairwise interaction is *not* frustrating (because of its sign alternation with distance) although frustration is present in the microscopic underlying model. AF ordering is then expected (at T = 0) as seen for a related system of coupled Spin-Peierls chains [44].

5.5 Conclusion

The coexistence between AF order and SP dimer order under doping SP materials with non magnetic impurities [39] is one of the most surprising phenomenon in the field of quantum magnetism. Starting with the non frustating interaction between two solitonic clouds calculated above, we can construct an effective model of long range interacting spins $\frac{1}{2}$, randomly diluted on a square lattice. We have implemented a Quantum Monte Carlo (QMC) algorithm using the Stochastic Series Expansion (SSE) method [51] in order to study long distance interacting spin- $\frac{1}{2}$ models. The mechanism of AF ordering has been studied at very low temperature and dopant concentration with very large system sizes, up to 96×96 [52].

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6 Field-Theoretical Methods in Quantum Magnetism

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Abstract. We present a review of different field theory techniques that have proved very useful in the study of quantum magnets in low dimensions. We first review the application of the spin-wave analysis and non-linear σ -model techniques in one and two dimensional quantum antiferromagnets. We discuss in particular the emergence of Haldane's conjecture for spin chains and ladders within this formalism. We also present a brief discussion on the non-linear σ -model description for the two-dimensional antiferromagnet in the square lattice. In a second part we review the method of abelian bosonization and its application to the study of the XXZ spin 1/2 chain and its generalizations, such as the dimerized chain. Non-abelian bosonization is used to describe both SU(2) symmetric chains with arbitrary spin S and 2 leg ladders, rederiving Haldane's conjecture within this formalism. The inclusion of charge degrees of freedom leading to a Hubbard or a t - J model is also discussed. Finally, we apply the abelian bosonization approach to the study of N-leg ladders in amagnetic field, which leads to a further extension of Haldane's conjecture.

6.1 Introduction

Field theory techniques have proven in the last decades to be a powerful tool in the understanding of quantum magnetism. One of its main interests lie in the relatively simple and universal description it can provide in studying condensed matter system, and in particular all the exotic behaviors that can be found in low dimensional strongly correlated systems. For example, phenomena like fractional excitations or spin-charge separation, which are going to be presented in this chapter, are some of the topics that find a very natural description in the field theory context. This approach has indeed allowed to understand the experimental data reflecting the presence of such unusual behaviors. Another important and more pragmatic issue is the fact that, once a field theory is built for describing a particular model, physical quantities such as correlation functions, the magnetic susceptibility or the specific heat can in general be easily computed. Moreover, the effect of microscopic modifications of the system, as well as the specificity of low dimensional systems, can also be simply understood through this approach.

The field theory approach have to be considered as a fundamental tool within the different techniques that are currently used to study condensed matter systems. It is complementary and in close connection to other techniques such as integrable models and numerical methods. As we show in this chapter, the Bethe Ansatz solution of the XXZ spin chain provides information about the system that is then used to construct the precise field theory model that describes its large scale behavior. The knowledge of the field theory allows to compute, for example, the behavior at large distances of correlation function in a much simpler way than with integrable model techniques. Finite size scaling analysis is another subject in which field theory have proven to be very useful, providing a natural link with numerical techniques also commonly used in condensed matter physics.

This chapter provides a review of field theory techniques that are used in the study of quantum magnets in low dimensions. In the first part we provide an overview of the spin-wave analysis in one and two dimensional quantum antiferromagnets. We then concentrate on the derivation of the non-linear σ -model that describes the low energy dynamics of spin S chains within the large S approach. The behavior of this model with and without a topological term, giving rise to Haldane's conjecture, is discussed. The results obtained in the context of spin-wave analysis and the non-linear σ -model are then generalized to the case of spin ladders. A brief discussion on the applications of this description for the two-dimensional antiferromagnet in the square lattice closes the first part of this chapter.

In the second part we review the method of abelian bosonization and apply it to the study of the XXZ spin 1/2 chain in the presence of a magnetic field, which leads to the Luttinger liquid picture. We discuss how the microscopic data of the lattice model are related to the field theory parameters. We analyze in detail the computation of thermodynamic quantities and correlation functions within the bosonization method as well as certain modifications of the XXZ chain. The particularities and non-abelian bosonization description of the SU(2) Heisenberg point are also discussed. We then briefly treat the generalization of those results to the Hubbard and t-Jmodels to illustrate the inclusion of charge degrees of freedom. Non-abelian bosonization is also applied to the study of the two leg S = 1/2 SU(2) symmetric spin ladder and to higher spin one-dimensional chains where it is used to rederive Haldane's conjecture. Finally, we apply the abelian bosonization approach to the study of N-leg ladders, which leads to a further extension of Haldane's conjecture.

The general overview presented here aims not only at providing a description of the usual tools used in field theory for condensed matter physics, but also to show to the reader how this approach is in almost symbiotic connection with the other areas described in this book. We also mention in this chapter many topics that to date are still open questions with the hope that future progresses in field theory will help to elucidate those issues.

6.2 Path Integral for Spin Systems

Let us assume that we have an arbitrary number of spins $\{\mathbf{S}_n\}$ labeled by the generic site index *n* without, for the moment, making any further supposition about the geometry and dimensionality of such an array of spins. These operators satisfy an SU(2) algebra on each site

$$[S_n^x, S_n^y] = iS_n^z \quad \text{and} \quad \mathbf{S}_n^2 = S(S+1), \tag{6.1}$$

We assume also that the system has a Hamiltonian $H({\mathbf{S}_i})$ that we do neither need to specify for the moment. The idea is to define a path integral for such a system as proposed by Haldane (see for example [1], [2]). To avoid making heavier the notation, let us assume that we have first a single spin. Following [2], in the 2S + 1 dimensional Hilbert space, we define the states:

$$|\mathbf{n}\rangle = e^{i\theta(\hat{z}\times\mathbf{n})\cdot\mathbf{S}} |S,S\rangle \tag{6.2}$$

where **n** is a unit vector forming an angle θ with the quantization axis (z) and $|S, S\rangle$ is the highest weight state. A straightforward calculation shows that:

$$\langle \mathbf{n} | \mathbf{S} | \mathbf{n} \rangle = S \mathbf{n}. \tag{6.3}$$

One can also show that the internal product of two such states gives:

$$\langle \mathbf{n}_1 | \mathbf{n}_2 \rangle = e^{iS\Phi(\mathbf{n}_1, \mathbf{n}_2, \hat{z})} \left(\frac{1 + \mathbf{n}_1 \cdot \mathbf{n}_2}{2}\right)^S \tag{6.4}$$

where $\Phi(\mathbf{n}_1, \mathbf{n}_2, \hat{z})$ is the solid angle viewed from the origin formed by the triangle with vertices in \mathbf{n}_1 , \mathbf{n}_2 and \hat{z} . Note that $\Phi(\mathbf{n}_1, \mathbf{n}_2, \hat{z})$, as a solid angle, is defined modulo 4π . This ambiguity has however no importance in (6.4) because of the periodicity of the exponential. With this over-complete basis, one can also write the identity operator in the Hilbert space:

$$I = \int \left(\frac{2S+1}{4\pi}\right) d^3n \ \delta(\mathbf{n}^2 - 1) |\mathbf{n}\rangle \langle \mathbf{n}|$$
(6.5)

which can be obtained by using the properties of the rotation matrices

$$D_{M,M'}^{S}(\mathbf{n}) = \langle S, M | e^{i\theta(\hat{z} \times \mathbf{n}) \cdot \mathbf{S}} | S, M' \rangle$$

in the spins S representation :

$$\frac{2S+1}{4\pi} \int d^3 n \,\,\delta(\mathbf{n}^2-1) {D^S}^*_{M,M'}(\mathbf{n}) D^S_{N,N'}(\mathbf{n}) = \delta_{M,N} \delta_{N',M'}.$$

Imagine now that we want to compute the partition function

$$Z = Tr\{e^{-\beta H}\}$$

seen as the evolution of the system in imaginary time with periodic boundary conditions. We can decompose the evolution in imaginary time into N infinitesimal steps of length δt , with $N \to \infty$, $N\delta t = \beta$. using then the Trotter formula:

$$Z = \lim_{N \to \infty} \left(e^{-\delta tH} \right)^N$$

an inserting an identity at each intermediary step, we obtain:

$$Z = \lim_{N \to \infty} \left(\frac{2S+1}{4\pi} \right)^N \left(\prod_{a=1}^N \int d^3 n_a \ \delta(\mathbf{n}_a^2 - 1) \langle \mathbf{n}(t_a) | e^{-\delta t H} | \mathbf{n}(t_{a+1}) \rangle \right).$$

If now, as in the standard path integral construction, we only keep in each infinitesimal step the first order in δt :

$$\langle \mathbf{n}(t_a)|e^{-\delta tH}|\mathbf{n}(t_{a+1})\rangle = \left[\langle \mathbf{n}(t_a)|\mathbf{n}(t_{a+1})\rangle + \delta t\langle \mathbf{n}(t_a)|H|\mathbf{n}(t_a)\rangle + O(\delta t^2)\right]$$

and we formally define the path integral measure:

$$\int \mathcal{D}\mathbf{n} = \lim_{N \to \infty} \left(\frac{2S+1}{4\pi}\right)^N \left(\prod_{a=1}^N \int d^3 n_a \,\,\delta(\mathbf{n}_a^2 - 1)\right)$$

using (6.4) we can write the partition function as:

$$Z = \int \mathcal{D}\mathbf{n} \ e^{-S[\mathbf{n}]} \tag{6.6}$$

with

$$S[\mathbf{n}] = -iS\sum_{a} \Phi(\mathbf{n}(t_{a}), \mathbf{n}(t_{a+1}), \hat{z}) - S\sum_{a} \log\left(\frac{1 + \mathbf{n}(t_{a}) \cdot \mathbf{n}(t_{a+1})}{2}\right) + \delta t \sum_{a} \langle \mathbf{n}(t_{a}) | H | \mathbf{n}(t_{a}) \rangle.$$
(6.7)

Since in the computation of the partition function we used periodic boundary conditions, namely: $\mathbf{n}(0) = \mathbf{n}(\beta)$, and if we suppose that the path described by $\mathbf{n}(t)$ is smooth ³, we see that $\sum_{a} \Phi(\mathbf{n}(t_{a}), \mathbf{n}(t_{a+1}), \hat{z})$ describes the solid angle, or the area in the unit sphere bounded by the curve $\mathbf{n}(t)$, $\mathcal{A}\{\mathbf{n}(t)\}$. As before, the independence of the partition function of the choice of the quantization axis or the ambiguity in the definition of the solid angle is a consequence of the 4π invariance of the phase factor. In (6.7), the second term is of order $(\delta t)^2$, the imaginary time continuum limit of this action is then :

$$S[\mathbf{n}] = -iS\mathcal{A}\{\mathbf{n}(t)\} + \int_0^\beta dt \langle \mathbf{n}(t) | H | \mathbf{n}(t) \rangle.$$
(6.8)

³ This assumption is actually delicate, as in the standard Feynmann path integral, see [2] and references therein, but we ignore such technical details here.

Equation (6.8) is the main result of the path integral description for magnetic systems which we can now apply to spins chains, ladders and two-dimensional antiferromagnets.

6.3 Effective Action for Antiferromagnetic Spins Chains

Let us assume now that we have a collection of spins $\mathbf{S}_i^2 = S(S+1)$ forming a one dimensional array (chain) with the Hamiltonian:

$$H = J \sum_{k} \mathbf{S}_{n} \cdot \mathbf{S}_{n+1} \tag{6.9}$$

with J > 0. (6.9) is just the one-dimensional Heisenberg antiferromagnet. The action (6.8) takes then the explicit form:

$$S[\{\mathbf{n}_n\}] = -iS\sum_n \mathcal{A}\{\mathbf{n}_n(t)\} + JS^2 \int_0^\beta dt \sum_n \mathbf{n}_n(t) \cdot \mathbf{n}_{n+1}(t).$$
(6.10)

In order to take the continuum limit in the spatial direction, we need to identify the low energy, large scale degrees of freedom that can be considered as slowly varying fields in the action. We can, for this, make use of the known results from spin wave theory from which we know that low energy modes are found at zero and π momenta (see also below for the generalization to the case of ladders). We can then write the ansatz

$$\mathbf{n}_n = (-1)^n \sqrt{1 - a^2 \mathbf{l}_n^2} \,\mathbf{m}_n + a \mathbf{l}_n \tag{6.11}$$

with *a* the lattice spacing and $\mathbf{m}_n^2 = 1$. This result, which is valid for large *S*, is just telling us that the large scales behavior of the system is governed by fields representing a staggered and a quasi-homogeneous variation of the magnetization. The latter field, playing the rôle of angular momentum for **n** is chosen to have dimension of density and is responsible of a net magnetization which is supposed to be small. To order a^2 , the relation $\mathbf{n}_n^2 = 1$ is equivalent to $\mathbf{m}_n \cdot \mathbf{l}_n = 0$. We can now introduce this form for the field \mathbf{n}_n in (6.10) and keep only the lowest order in *a* to take the continuum limit. For the area term, by noticing that $\mathcal{A}\{-\mathbf{n}(t)\} = -\mathcal{A}\{\mathbf{n}(t)\}$, we can group terms two by two and write the sum as:

$$\sum_{i} \mathcal{A}\{\mathbf{n}_{2i}(t)\} + \mathcal{A}\{\mathbf{n}_{2i-1}(t)\} =$$
$$\sum_{i} \mathcal{A}\{\sqrt{1 - a^2 \mathbf{l}_{2i}^2} \mathbf{m}_{2i}(t) + a \mathbf{l}_{2i}(t)\}$$
$$-\mathcal{A}\{\sqrt{1 - a^2 \mathbf{l}_{2i-1}^2} \mathbf{m}_{2i-1}(t) - a \mathbf{l}_{2i-1}(t)\}.$$

We can now use the expression relating the difference in the areas produced by curves $\mathbf{n}(t)$ and $\mathbf{n}(t) + \delta \mathbf{n}(t)$:

$$\mathcal{A}\{\mathbf{n}(t) + \delta \mathbf{n}(t)\} = \mathcal{A}\{\mathbf{n}(t)\} + \int_0^\beta dt \ \delta \mathbf{n}(t). \left(\partial_t(\mathbf{n}(t)) \times \mathbf{n}(t)\right)$$

and we obtain

$$\sum_{i} \mathcal{A}\{\mathbf{n}_{2i}(t)\} + \mathcal{A}\{\mathbf{n}_{2i-1}(t)\} = -a \int_{0}^{\beta} dt \sum_{i} \left(\frac{\Delta}{a}(\mathbf{m}_{2i}(t)) + 2\mathbf{l}_{2i}(t)\right) \cdot (\mathbf{m}_{2i}(t) \times \partial_{t}(\mathbf{m}_{2i}(t))) + O(a^{2})$$

where we have used that $\mathbf{m}_{2i-1}(t) = \mathbf{m}_{2i}(t) - \Delta(\mathbf{m}_{2i}(t)) + O(a^2).$

In the same spirit, and omitting constant terms, the second term in (6.10) can be written as:

$$\frac{JS^2}{2} \int_0^\beta dt \sum_i \left[\left(\mathbf{n}_{2i}(t) + \mathbf{n}_{2i+1}(t) \right)^2 + \left(\mathbf{n}_{2i+1}(t) + \mathbf{n}_{2i+2}(t) \right)^2 \right]$$

and the lowest order in a gives:

$$\frac{JS^2a^2}{2}\int_0^\beta dt\sum_i\left\{\left[-\frac{\Delta}{a}\mathbf{m}_{2i}(t)+2\mathbf{l}_{2i}\right]^2+\left[\frac{\Delta}{a}\mathbf{m}_{2i+1}(t)+2\mathbf{l}_{2i+1}\right]^2\right\},\,$$

which, still to lowest order in a can also be written as:

$$JS^2a^2\int_0^\beta dt\sum_i \left[\left(\frac{\Delta}{a}\mathbf{m}_{2i}(t)\right)^2 + 4\mathbf{l}_{2i}^2\right].$$

We can now collect all the pieces together and take the continuum limit by replacing $\frac{\Delta}{a} \to \partial_x$, $2a \sum_i \to \int dx$ (the factor of 2 arises from the doubling of the chain index). We also take the limit of zero temperature $T \to 0$. We obtain for the total action:

$$S[\mathbf{m}, \mathbf{l}] = \frac{JS^2a}{2} \int dx \, dt \, \left[(\partial_x (\mathbf{m}(x, t))^2 + 4\mathbf{l}(x, t)^2 \right] \\ + \frac{iS}{2} \int dx dt \left(\partial_x (\mathbf{m}(x, t)) + 2\mathbf{l}(x, t) \right) \cdot \left(\mathbf{m}(x, t) \times \partial_t (\mathbf{m}(x, t)) \right). \quad (6.13)$$

We immediately notice that this action is quadratic in the variable **l**. We can then integrate out this variable and obtain the final result:

$$S[\mathbf{m}] = \int dx dt \frac{1}{2g} \left(v(\partial_x \mathbf{m})^2 + \frac{1}{v} (\partial_t \mathbf{m})^2 + \frac{i\theta}{8\pi} \epsilon_{ij} \mathbf{m} \cdot (\partial_i \mathbf{m} \times \partial_j \mathbf{m}) \right)$$
(6.14)

with g = 2/S the coupling constant, v = 2aJS the spin wave velocity and the topological angle $\theta = 2\pi S$. If we want the action to be finite in an infinite system and at zero temperature, we have to impose that **m** tends to a fixed vector \mathbf{m}_0 at infinity in space and imaginary time. By making all the points at infinity equivalent, we are just saying that our space time is equivalent to a sphere S_2 . Since in each point of the space time **m** can also be viewed as an element of S_2 , the mapping $\mathbf{m}(x,t)$ corresponds to an embedding of the sphere into itself. Such embeddings are classified by what is called the second homotopy group of the sphere $\Pi_2(S_2) = \mathbb{Z}$ [3]. To each embedding corresponds an integer (element of \mathbb{Z}) given by the Pontryagin index:

$$\frac{1}{8\pi} \int dx dt \ \epsilon_{ij} \mathbf{m} \cdot (\partial_i \mathbf{m} \times \partial_j \mathbf{m}) \in \mathbb{Z}$$
(6.15)

where we immediately recognize in this expression the last term of the action (6.14). We can then conclude from this result that for integer S, the imaginary part of the action in (6.14) (which we will call the topological term) is always a multiple of 2π and plays no role at all, while for half integer spins, as we will see, the situation is completely different.

6.4 The Hamiltonian Approach

The result (6.14) can also be derived using a Hamiltonian approach; we follow here the derivation given in [4], [5]. Let us group our spin operators two by two and define the variables \mathbf{L} and \mathbf{M} through

$$\mathbf{S}_{2i} = a\mathbf{L}_i - S\mathbf{M}_i$$

$$\mathbf{S}_{2i+1} = a\mathbf{L}_i + S\mathbf{M}_i.$$
 (6.16)

These relations can be inverted:

$$\mathbf{L}_{i} = \frac{1}{2a} \left[\mathbf{S}_{2i+1} + \mathbf{S}_{2i} \right]$$
$$\mathbf{M}_{i} = \frac{1}{2S} \left[\mathbf{S}_{2i+1} - \mathbf{S}_{2i} \right]$$
(6.17)

and one can then easily show using (6.1) that these variables satisfy the constraints:

$$a^{2}\mathbf{L}_{i}^{2} + S^{2}\mathbf{M}_{i}^{2} = S(S+1) ; \mathbf{L}_{i} \cdot \mathbf{M}_{i} = 0$$
 (6.18)

and the algebra:

$$\begin{split} [L_i^a, L_j^b] &= \frac{i}{2a} \epsilon^{abc} \delta_{i,j} L_i^c ; \\ [L_i^a, L_j^b] &= \frac{i}{2a} \epsilon^{abc} \delta_{i,j} M_i^c ; \end{split}$$

$$[L_i^a, L_j^b] == \frac{ia}{2S^2} \epsilon^{abc} \delta_{i,j} L_i^c.$$
(6.19)

We can rewrite the Hamiltonian (6.9) as:

$$H = J \sum_{i} [\mathbf{S}_{2i} \cdot \mathbf{S}_{2i+1} + \mathbf{S}_{2i+1} \cdot \mathbf{S}_{2i+2}]$$

= $J \sum_{i} [-S^2 \mathbf{M}_i^2 + a^2 \mathbf{L}_i^2 + a^2 \mathbf{L}_i \cdot \mathbf{L}_{i+1} + a^2 \mathbf{M}_i - \mathbf{M}_{i+1}) + \frac{S^2}{2} (\mathbf{M}_i - \mathbf{M}_{i+1})^2 - S^2 \mathbf{M}_i^2]$ (6.20)

where the index of the first term of the last line has been shifted by one for convenience. To make contact with the result of the preceding section, we take the continuum limit by keeping in the Hamiltonian only the terms of order a^2 . We start by defining the variable x as:

$$\mathbf{M}_i \to \mathbf{M}(x) ; \ \mathbf{M}_{i\pm 1} \to \mathbf{M}(x) \pm 2a\partial_x(\mathbf{M}(x)) + O(a^2).$$

Using then the identification

$$2a\sum_{i} \rightarrow \int dx \ ; \ \frac{1}{2a}\delta_{i,j} \rightarrow \delta(x-y)$$

and the relation (6.18), we obtain the continuous Hamiltonian (we omit constant terms):

$$H = \frac{v}{2} \int dx \left[g \left(\mathbf{L} - \frac{\theta}{4\pi} \partial_x(\mathbf{M}) \right)^2 + \frac{1}{g} \left(\partial_x(\mathbf{M}) \right)^2 \right]$$
(6.21)

where g, v and θ have already been defined. The key point is to realize that for $S \to \infty$, the constraint and the algebra become:

$$\mathbf{M}^{2}(x) = 1 \; ; \; \mathbf{L}(x) \cdot \mathbf{M}(x) = 0$$
 (6.22)

$$[L^{a}(x), L^{b}(y)] = i\epsilon^{abc}\delta(x-y)L^{c}(x) ; \ [L^{a}(x), M^{b}(y)] = i\epsilon^{abc}\delta(x-y)M^{c}(x)$$

$$[M^{a}(x), M^{b}(y)] = 0 (6.23)$$

and in this limit we can view $\mathbf{L}(x)$ as the angular momentum density $\mathbf{M}(x) \times \dot{\mathbf{M}}(x)$ associated to the normalized field \mathbf{M} (note the similarity between this operator relation and the ansatz (6.11)). Upon the replacement $\mathbf{m} \to \mathbf{M}$ in (6.14), and using an appropriate parametrization for this normalized field (as, for example, the azimuthal angles in the sphere), one can easily show that (6.21) is the Hamiltonian associated to the Lagrangian of the action (6.14), which completes our alternative derivation of the non-linear sigma model description of Heisenberg antiferromagnetic chains in the large S limit.

6.5 The Non-linear Sigma Model and Haldane's Conjecture

Let us first consider the case of integer spins, where the topological term is absent. We then have the usual O(3) non linear sigma model (NLSM)⁴ for which we can write the partition function as:

$$Z = \int D\{\mathbf{m}\}\delta(\mathbf{m}^2 - 1)e^{-\int dxdt\frac{1}{2g}\left((\partial_x \mathbf{m})^2 + (\partial_t \mathbf{m})^2\right)}$$
(6.24)

and where we have set the sound velocity v to unity. At the classical level, the action in (6.24) is scale invariant. Since there is no apparent scale parameter in the model, one would be tempted to conclude that the correlation function of this model are algebraically decaying, a phenomenon typical of scale invariant systems. We will see however that fluctuations change dramatically this scenario [6]. To see this, we start by expressing the δ function in (6.24) in terms of a Lagrange multiplier:

$$Z = \int_{c-i\infty}^{c+i\infty} D\{\lambda(x)\} \int D\{\mathbf{m}(x)\} e^{-\int dx dt \frac{1}{2g} \left((\partial_x \mathbf{m})^2 + (\partial_t \mathbf{m})^2 + \lambda(\mathbf{m}^2 - 1)\right)}.$$
 (6.25)

In order to understand the qualitative behavior of (6.24), we are going to do an approximation which consists in replacing the integral in λ by the maximal value of the integrand:

$$Z \sim \int D\{\mathbf{m}(x)\} e^{-\int dx dt \frac{1}{2g} \left((\partial_x \mathbf{m})^2 + (\partial_t \mathbf{m})^2 + \lambda_m (\mathbf{m}^2 - 1) \right)}, \tag{6.26}$$

where the optimal value λ_m is assumed to be a constant. As we will see below, such an approximation is valid if we generalize our model to the O(N) nonlinear sigma model and consider the limit $N \gg 1$. The approximate partition function in (6.26) has the advantage of being Gaussian and then all physical quantities can be easily calculated.

To obtain λ_m , we integrate over $\mathbf{m}(x)$ in (6.25):

$$Z = \int_{c-i\infty}^{c+i\infty} D\{\lambda(x)\} e^{\frac{1}{2g} \left(\int \lambda(x,t) dx dt - \frac{N}{2} \log(\det\{-\triangle + \lambda(x,t)\})\right)}, \tag{6.27}$$

where \triangle is the two dimensional Laplace operator. It is now apparent that for $N \gg 1$ we can estimate this integral by a saddle-point approximation. The condition for maximizing the integrand is:

⁴ The historical origin of this name comes from the way the field was written in some choice of variables where the O(N) symmetry is realized non-linearly.

$$\frac{1}{2g} = \frac{N}{2} \frac{\delta}{\delta\lambda(x,t)} \log(\det\{-\triangle + \lambda(x,t)\})$$
(6.28)

And, again, under the assumption of λ_m being constant, we get:

$$1 = gNTr\{\frac{1}{-\Delta + \lambda_m}\}$$

= $gN \int \frac{d^2p}{4\pi^2} \frac{1}{p^2 + \lambda_m}$
= $\frac{gN}{4\pi} log(\Lambda^2/\lambda_m)$ (6.29)

where Λ is an ultraviolet momentum cut-off. From (6.29) we obtain the optimal value:

$$\lambda_m = \Lambda^2 e^{-\frac{4\pi}{gN}} \tag{6.30}$$

which indicates us that fluctuations have dynamically generated a mass term in our original action. Indeed, by using (6.26) one easily sees that correlation functions are now exponentially decaying with the distance.

The arguments we used to derive the result (6.30) are strictly speaking valid for $N \gg 1$. It is however well established by many techniques that this result is indeed qualitatively correct for $N \ge 3$. The non-linear sigma model is integrable even at the quantum level and an exact S matrix has being proposed [7]. An intuitive way to understand this result is by seeing (6.24) as the partition function of a classical magnet in the continuum. In such an interpretation, the coupling g plays the role of temperature. Another approach to understand this phenomenon is given by the renormalization group analysis. We refer the reader to [6], [8] for a detailed presentation of the renormalization group techniques and give here the main steps of the procedure. The idea is to decompose the field in slowly and fast fluctuating parts; we then integrate over the fast degrees of freedom to obtain an effective action with renormalized parameters. Following [6] we start by writing our field as:

$$\mathbf{m} = \sqrt{1 - \sum_{i} \sigma_i^2} \mathbf{m}_s + \sum_{i=1}^{N-1} \sigma_i \mathbf{e}_i, \qquad (6.31)$$

where $\mathbf{m}_s^2 = 1$ and the vectors \mathbf{e}_i form an orthonormal basis for the space orthogonal to \mathbf{m}_s , and the fields σ_i are the fast fluctuating degrees of freedom. Keeping only the quadratic terms in the fields σ_i , the action (6.14) becomes:

-

$$\int dx dt \frac{1}{2g} \left[\sum_{\mu} \sum_{i,j} (\partial_{\mu} \boldsymbol{\mathbf{m}}_{i}) \cdot \boldsymbol{\mathbf{e}}_{j} \sigma_{j} \right]^{2} \\ \sum_{\mu} \sum_{i,j} (\partial_{\mu} \boldsymbol{\mathbf{m}}_{s}) \cdot \boldsymbol{\mathbf{e}}_{i} (\partial_{\mu} \boldsymbol{\mathbf{m}}_{s}) \cdot \boldsymbol{\mathbf{e}}_{j} (\sigma_{i} \sigma_{j} - \sum_{k} \sigma_{k}^{2} \delta_{ij}) + \sum_{\mu} (\partial_{\mu} \boldsymbol{\mathbf{m}}_{s})^{2} \right]. \quad (6.32)$$

We can now integrate over the fast degrees of freedom in the momentum shell $\Lambda - \delta \Lambda , or in real space between scales <math>L$ and $L + \delta L$ to obtain an effective coupling for the slow part of the action $\sum_{\mu} (\partial_{\mu} \mathbf{m}_s)^2$. To lowest order in g, the contribution to each component $(\partial_{\mu} \mathbf{m}_s^i)^2$, of the slow part of the action is given by the one point function:

$$\langle \sigma_i \sigma_i - (N-1) \sum_k \sigma_k^2 \rangle$$

and after some algebra, we can do the integration to obtain the new coupling:

$$\frac{1}{g+\delta g} = \frac{1}{g} - (N-2)g^2 \int_{\Lambda-\delta\Lambda}^{\Lambda} \frac{d^2p}{(2\pi)^2 p^2}.$$
(6.33)

An important observation is that the term $\partial_{\mu}(\mathbf{e}_i) \cdot \mathbf{e}_j$ does not contribute to this result. The way to understand this is to notice that the action is invariant under rotations in the N-1 dimensional space $\alpha_i \to M_{ij}\alpha_j$ with $\alpha_i = \sigma_i, (\partial_{\mu}\mathbf{m}_s) \cdot \mathbf{e}_i$ and the term $\partial_{\mu}(\mathbf{e}_i) \cdot \mathbf{e}_j$ behaves under this transformation as a gauge field. Since $\sum_{\mu} (\partial_{\mu}\mathbf{m}_s)^2$ is rotationally invariant, the lowest order contribution we can have in the effective action arising from $\partial_{\mu}(\mathbf{e}_i) \cdot \mathbf{e}_j$ is the (gauge) invariant term $\sum_{\mu,\nu,i,j} (\partial_{\mu}(\mathbf{e}_i) \cdot \partial_{\nu}(\mathbf{e}_j) - \partial_{\mu}(\mathbf{e}_i) \cdot \partial_{\nu}(\mathbf{e}_j))^2$. This gauge invariant term give rise to non-logarithmic divergences which can be shown

to give no contribution in (6.33). From this result we obtain the β function:

$$\beta(g) = -\frac{dg}{dln(\Lambda)} = \frac{dg}{dln(L)} = \frac{N-2}{2\pi}g^2 + O(g^3).$$
(6.34)

That is, by going to large scales g flows to strong coupling indicating a regime of high temperature where the system is disordered and with a finite correlation length. Of course the original cut-off Λ and coupling g depend on the microscopic details of the theory, but if we imagine varying such parameters in our field theory in such a way to keep the dynamically generated scale constant, the constant λ satisfies the equation:

$$\frac{\partial \lambda}{\partial A} + \frac{\partial \lambda}{\partial g} \frac{\partial g}{\partial A} = 0 . \qquad (6.35)$$

Using (6.34) and assuming that $\lambda = \Lambda^2 f(g)$, we obtain:

$$\lambda = \Lambda^2 e^{-\frac{4\pi}{g(N-2)}} \tag{6.36}$$

which coincides with (6.30) for $N \to \infty$. Remember that our case of interest corresponds to N = 3.

The case of half-integer spins is very different. We have to remember that in this case the action (6.14) contain the topological term which contributes

as a destructive phase term in the computation of the partition function. The coupling constant still flows to the strong coupling regime, but the topological term is protected against renormalization because of its discrete nature. Recalling that the coupling constant g = 2/S is inversely proportional to the spin S, the flow to strong coupling can be interpreted as a large scale behavior of the system with smaller spins. Since the topological term remains present at large scale, one can conclude that the large scales behavior of half integer spin chains corresponds to the one of spin 1/2 chain, which is known to be gapless. Shankar and Read have given further support to this conclusion [9]. This drastic difference between integer and half integer antiferromagnetic spin chains is known as the Haldane conjecture [10].

6.6 Antiferromagnetic Spin Ladders

The techniques we have used so far to obtain the large scales behavior of Heisenberg antiferromagnetic chains can be generalized to other geometries. The closest example is given by the spin ladder systems. Imagine an array of spins forming a strip composed of N chains. Neighboring spins belonging to the same chains are supposed to have a coupling J, as before, while neighboring spins of adjacent chains have a coupling given by J'. We assume also that both couplings J and J' are positive. The spins on this ladder are labelled by the chain index n and the row index $a, 1 \le a \le N$. N is to be considered as fixed and finite while the number of spins along the chains diverge in the thermodynamic limit. The analysis presented in this section follows the lines of [5].

At the classical level, the lowest energy configurations are given by a Néel order, say, in the \hat{z} direction given by:

$$\mathbf{S}_{a,n} = (-1)^{a+n} S\hat{z}.$$
(6.37)

The equations of motion for a spin belonging to an intermediate row is given by:

$$\frac{d\mathbf{S}_{a,n}}{dt} = -\mathbf{S}_{a,n} \times \left[J\left(\mathbf{S}_{a,n-1} + \mathbf{S}_{a,n+1}\right) + J'\left(\mathbf{S}_{a-1,n} + \mathbf{S}_{a+1,n}\right) \right], \quad (6.38)$$

(for the spins belonging to the edges rows, the a - 1 or a + 1 terms are absent). We can now use our experience in spin wave analysis to identify the low energy excitations around this Néel state. We linearize (6.38) and write the ansatz:

$$S_{a,n}^x + iS_{a,n}^y = e^{i(wt+nq)} \left(A_a(q) + (-1)^{a+n+1} B_a(q) \right).$$
(6.39)

Note that the example of decoupled chains is a particular case of this model. With the ansatz proposed here one must of course recover the well-known results for the simple chain in the limit $J' \to 0$. The resulting eigenvalue problem shows that there are N-1 families of solutions, which for $J' \neq 0$ have a gapped spectrum (one can show however that such modes become gapless for the case of decoupled chains J' = 0). There is only one family of excitations with vanishing energy at q = 0 and $q = \pi$. These modes are the counterpart of the gapless modes of the single Heisenberg chain and give us a clue of the form of the slowly varying fields in a field theory approach.

The solution for small q is given by $B_a = B$ and $A_a \propto q \sum_{b}^{a} L_{ab}^{-1}$ with

$$L = \begin{pmatrix} 4J + J' & J' & 0 & .. \\ J' & 4J + 2J' & J' & .. \\ 0 & J' & 4J + 2J' & .. \\ .. & .. & .. & .. \end{pmatrix}.$$
(6.40)

We can now work out the path integral description of the low energy physics of the ladder system. We refer the reader to [5] for the Hamiltonian derivation of it and the subtle differences between the path integral and Hamiltonian results. By using our basis of states $|\mathbf{n}\rangle$ in (6.2), we write the action of the ladder as:

$$S[\{\mathbf{n}_{a,n}\}] = -iS\sum_{a,n} \mathcal{A}\{\mathbf{n}_{a,n}(t)\} + JS^2 \int_0^\beta dt \sum_{n,a} \mathbf{n}_{a,n}(t) \cdot \mathbf{n}_{a,n+1}(t) + J'S^2 \int_0^\beta dt \sum_n \sum_{a=1}^{N-1} \mathbf{n}_{a,n}(t) \cdot \mathbf{n}_{a+1,n}(t) (6.41)$$

and, inspired by the spin wave result, we propose as an ansatz the generalization of (6.11):

$$\mathbf{n}_{a,n} = (-1)^n \sqrt{1 - a^2 \alpha_a^2 \mathbf{l}(n)^2} \ \mathbf{m} + a \alpha_a \mathbf{l}(n)$$
(6.42)

where $\alpha_a = A_a/(\sum_a A_a)$. Note that excitations along the transverse direction of the ladder are all supposed to be of high energy. This is due to the fact that N is kept finite implying a finite difference in the energy levels of transverse excitations. Then, the effective low energy degrees of freedom are one-dimensional in nature. The procedure is then standard: we insert expression (6.42) into (6.41) and work out the continuum limit. The final result is again the effective action (6.14) with the parameters:

$$g = \frac{1}{S\sqrt{\sum_{a,b} L_{ab}^{-1}}}; \ v = \frac{SJa}{\sqrt{\sum_{a,b} L_{ab}^{-1}}}; \ \theta = 2\pi S \sum_{a=1}^{N} (-1)^{a}.$$
(6.43)

The important result here is the contribution to the topological term which is easy to understand by noticing that, with the assumption we made in (6.42), each chain will contribute to it with a term $2\pi S(-1)^a$. We then see that only for an odd number of coupled chains, and for half-integer spin will the topological term give rise to a gapless behavior of our system at zero temperature. To summarize, the Non-Linear Sigma model approach predicts that the large scale behavior of the system is governed by the product SN: if it is an integer, the system is expected to be gaped, while for half-integer values, the system is gapless.

6.7 Chains with Alternating Bonds

As another example of the applications of the NLSM technique, we can consider the study of spin chains with alternating couplings, or dimerization [4] [5] with the Hamiltonian:

$$J\sum_{i} \left[(1+\delta)\mathbf{S}_{2i}(t) \cdot \mathbf{S}_{2i+1}(t) + (1-\delta)\mathbf{S}_{2i+1}(t) \cdot \mathbf{S}_{2i+2}(t) \right].$$
(6.44)

We can use both the path integral or Hamiltonian approach to obtain the effective action in the continuum limit. Within this last approach, we use again the operators (6.17). It is a straightforward computation to show that the Hamiltonian is now:

$$H = J \sum_{i} \left[-(1+\delta)S^{2}\mathbf{M}_{i}^{2} + a^{2}(1+\delta)\mathbf{L}_{i}^{2} + a^{2}(1-\delta)\mathbf{L}_{i} \cdot \mathbf{L}_{i+1} + aS(1-\delta)\left(\mathbf{M}_{i-1} \cdot \mathbf{L}_{i} - \mathbf{L}_{i} \cdot \mathbf{M}_{i+1}\right) + \frac{S^{2}}{2}(1-\delta)(\mathbf{M}_{i} - \mathbf{M}_{i+1})^{2} - S^{2}(1-\delta)\mathbf{M}_{i}^{2} \right]$$
(6.45)

and completing squares and taking the continuum limit as before, we obtain (see [4]):

$$H = \frac{\tilde{v}}{2} \int dx \left[\tilde{g} \left(\mathbf{L} - \frac{\tilde{\theta}}{4\pi} \partial_x(\mathbf{M}) \right)^2 + \frac{1}{\tilde{g}} \left(\partial_x(\mathbf{M}) \right)^2 \right]$$
(6.46)

with now $\tilde{g} = 2/(S\sqrt{1-\delta^2})$, $\tilde{v} = 2aJS\sqrt{1-\delta^2}$ and $\tilde{\theta} = 2\pi S(1-\delta)$. Building the corresponding Lagrangian we observe that the resulting sigma model has now a topological term with a factor of $1-\delta$ in front. This result can be easily obtained also within the path integral approach. The topological term changes sign under a parity transformation, as well as time reversal and $\mathbf{m} \to -\mathbf{m}$. In the non-dimerized case (which is parity invariant) this fact has no importance since the factor in front of it is a multiple of π and an overall sign has no effect in the computation of the partition function. The situation is different in the presence of dimerization. Now the total action is not anymore invariant under such transformation. One important question is what is the large scales behavior of the NLSM in the presence of a topological term with coefficient different from $\pm \pi$. We refer the reader to [4] to a discussion about this delicate issue and just announce the commonly believed scenario: the NLSM is massless only for $\theta = \pm \pi$. A result in support of the idea that for a nontrivial θ we obtain a massive behavior is the fact that a spin 1/2 chain with dimerization has a gap in the spectrum, as we are going to see below in the context of bosonization. Based in this belief we can then conclude that by varying the parameter δ one should encounter 2S + 1 gapless points in a spin S chain. Such results can also be extended to the case of spin ladders where different kinds of dimerizations are conceivable [5], and where, again, one recover a NLSM with a non-integer factor for the topological term.

6.8 The Two-Dimensional Heisenberg Antiferromagnet

We start our discussion on two-dimensional antiferromagnets by considering spins S located at the vertices of a square lattice $\mathbf{S}_{i,j}$, where *i* and *j* label the position on the lattice for each spin. The Hamiltonian is:

$$H = J \sum_{i,j} \mathbf{S}_{i,j} \cdot (\mathbf{S}_{i+1,j} + \mathbf{S}_{i,j+1}).$$
(6.47)

We are going to consider again the $T \rightarrow 0$ limit. Within the path integral approach, the effective action is given by:

$$S[\{\mathbf{n}_{i,j}\}] = -iS \sum_{i,j} \mathcal{A}\{\mathbf{n}_{i,j}(t)\} + JS^2 \int dt \sum_{i,j} \mathbf{n}_{i,j}(t) \cdot (\mathbf{n}_{i+1,j}(t) + \mathbf{n}_{i,j+1}(t)).$$
(6.48)

We are going again to make use of the result of spin wave theory and assume that, for large S, the low-energy physics of the system can be described by the ansatz:

$$\mathbf{n}_{i,j} = (-1)^{i+j} \sqrt{1 - a^2 \mathbf{l}_{i,j}^2} \ \mathbf{m}_{i,j} + a \mathbf{l}_{i,j}.$$
(6.49)

Before obtaining explicitly the effective action arising from this ansatz, let us discuss first which kind of topological terms one can expect in the computation of the partition function. In order to have a finite value for the action, we assume again that the field configuration tends to the same constant field at spatial and imaginary time infinity. By associating all the points at infinity, the space-(imaginary)time manifold corresponds now to S^3 . On the other hand, the order parameter field is still en element of S^2 . The possibility of having configurations of the spin field with non-trivial winding is given by the homotopy group $\Pi_2(S^3) = 1$ which turns out to be trivial. One can then already see that the specifics in the physics of one-dimensional systems are not recovered in the square lattice. The situation can be more subtle for frustrating systems, like the Heisenberg antiferromagnet in the triangular lattice. In this case the classical Néel configuration is obtained by imposing that adjacent spins in each triangle form a planar configuration with a relative angle of $2\pi/3$. The orientation of such triad is characterized by an element of SO(3): we have to specify a vector orthogonal to the plane of the triad and the angle that forms on this plane the triad with respect to a reference configuration. Since $\Pi_2(SO(3)) = \mathbb{Z}$, we can expect in this case to have non-trivial contributions to the partition function from a topological origin. A microscopic derivation of the effective action has revealed indeed the possibility of such kind of non-trivial contributions [11], but its consequences in the large scale physics are much less easy to understand than in the onedimensional case.

Let us now resume our discussion about the antiferromagnet in the square lattice, where topology can still play a rôle. Any field configuration at a given time can be characterized by an integer corresponding to the Pontryagin index that we have discussed before:

$$\frac{1}{4\pi} \int dx dy \mathbf{m} \cdot (\partial_x \mathbf{m} \times \partial_y \mathbf{m}). \tag{6.50}$$

If the field $\mathbf{m}(x, y, t)$ varies smoothly with the time, this quantity keeps the same integer value all along the time. This quantity corresponds to the total charge of textural defects of the field configuration, called skyrmions [12]. The presence of such a term in the effective action would have dramatic consequences on the statistic of such skyrmions. Haldane [13] has shown however that the effective action of the square lattice antiferromagnet has no such topological terms. He considered however the possibility of singular configurations of the field allowing for tunneling processes that change this integer index. Such a kind of singularity, called a hedgehog, can play a rôle if the system is disordered and Haldane found a non-trivial S dependence of that term on the basis of a microscopic derivation of the effective action. We limit ourselves in this discussion to the case of non-singular configurations of the field and derive the effective action arising from the ansatz (6.49).

The part arising from the Hamiltonian can be treated in the same spirit as in the one-dimensional case and we have:

$$JS^{2} \int dt \sum_{i,j} \mathbf{n}_{i,j}(t) \cdot (\mathbf{n}_{i+1,j}(t) + \mathbf{n}_{i,j+1}(t)) =$$
$$\frac{JS^{2}}{2} \int dt \left[\sum_{i,j} \left(\mathbf{n}_{i,j}(t) + \mathbf{n}_{i+1,j}(t) \right)^{2} + \sum_{i,j} \left(\mathbf{n}_{i,j}(t) + \mathbf{n}_{i,j+1}(t) \right)^{2} \right]$$

which in the continuum limit gives:

$$\frac{JS^2}{2} \int dt \int dx \int dy \left((\partial_x \mathbf{m})^2 + (\partial_y \mathbf{m})^2 + 8\mathbf{l}^2 \right).$$

The outcome of the area term is a bit more subtle. We start by grouping the contribution of spins two by two along, say, the \hat{x} direction, as we did in the one-dimensional case and we get:

$$-a \int dt \sum_{i,j} \left((-1)^j \frac{\Delta_i}{a} (\mathbf{m}_{2i,j}(t)) + 2\mathbf{l}_{2i,j}(t) \right) \cdot (\mathbf{m}_{2i,j}(t) \times \partial_t (\mathbf{m}_{2i,j}(t))) + O(a^2)$$
(6.51)

where Δ_i stands for the difference (or lattice derivative) in the $i(\hat{x})$ direction. We know that the term

$$\int dt \sum_{i} \left(\frac{\Delta_{i}}{a} (\mathbf{m}_{2i,j}(t)) \right) \cdot (\mathbf{m}_{2i,j}(t) \times \partial_{t} (\mathbf{m}_{2i,j}(t)))$$

is going to give rise to the integer associated to the Pontryagin index in the x-t space-time slice. Since the field **m** is assumed to be slowly varying and non-singular, this integer must be the same for each row j. Then, because of the alternating sign in the sum in (6.51), this term cancels. In the continuous limit, the only contribution from the area term is then:

$$\frac{iS}{a} \int dt \int dx \int dy \mathbf{l}(x, y, t) \cdot (\mathbf{m}(x, y, t) \times \partial_t(\mathbf{m}(x, y, t))) dx$$

Collecting all the terms together and, again, integrating over the field **l** we obtain the final result for the action:

$$S = \frac{1}{2g} \int dx dy dt \left(v \left[(\partial_x \mathbf{m})^2 + (\partial_y \mathbf{m})^2 \right] + \frac{1}{v} (\partial_t \mathbf{m})^2 \right)$$
(6.52)

with $g = 2\sqrt{2}a/S$, $v = 2\sqrt{2}aJS$. To understand the behavior of this action, we start by noticing that the partition function is equivalent to that of a continuous magnet in three dimensions. Again g plays the rôle of a temperature and one expects the existence of some critical value below which the O(3) symmetry is broken.

To see this in more detail, we proceed as in the (1+1) dimensional case and consider the O(N) non-linear sigma model. The procedure is strictly the same, and we obtain again the saddle-point equation:

$$1 = gN \operatorname{tr} \left\{ \frac{1}{-\Delta + \lambda_m} \right\}$$
$$= gN \int \frac{d^3p}{(2\pi)^3} \frac{1}{p^2 + \lambda_m}$$
(6.53)

where now the integral over momenta is three-dimensional. As in the (1+1) case, this integral is divergent at high momenta and has to be regularized by a cut-off $\Lambda \sim \frac{1}{a}$. By a careful inspection of the integral (6.53), one can see that there is a real and strictly positive solution for λ_m for any value of g bigger than the critical value g_c obtained from:

$$1 = g_c N \int \frac{d^3 p}{(2\pi)^3} \frac{1}{p^2} = \frac{g_c N \Lambda}{2\pi^2}.$$
 (6.54)

For any $g > g_c$ the scenario is similar to the (1+1) dimensional case where the symmetry is unbroken and excitations acquire a gap.

If $g < g_c$, there is no real and positive solution of (6.53). This phase corresponds to the broken symmetry phase we mentioned above. This scenario is also supported by a one loop computation of the β function in $2 + \epsilon$ dimensions (and setting $\epsilon = 1$ here):

$$\beta(g)=-g+\frac{N-2}{2\pi}g^2+\dots$$

(recall that now g is a dimension-full constant). This result suggests that there is a critical value of g which is the only point in which the system is truly scale invariant. Below that value the system flows to the low temperature phase and above it it flows to the high temperature phase.

For very small g, and taking back N = 3 we can describe our field as a small deformation of an homogeneous vector, say, in the \hat{z} direction:

$$\mathbf{m}(x, y, t) = (\sqrt{1 - \alpha_1^2 - \alpha_2^2}, \alpha_1(x, y, t), \alpha_2(x, y, t))$$

and the remaining action

$$S \sim \frac{1}{2g} \sum_{a=1}^{2} \int dx dy dt \left(c \left[(\partial_x \alpha_a)^2 + (\partial_y \alpha_a)^2 \right] + \frac{1}{c} (\partial_t \alpha_a)^2 + \dots \right)$$
(6.55)

is simply the one of two massless Goldstone modes. We thus conclude that there must be a critical value of the coupling constant, proportional to a that separates the ordered from the disordered phase. This means that there must be a critical value of the spin magnitude S_c above which the system is ordered at zero temperature. Since we have by now numerical and experimental evidence that the spin 1/2 Heisenberg antiferromagnet has an ordered ground state, we then conclude that it is ordered for all values of S at T = 0.

6.9 Bosonization of 1D Systems

6.9.1 XXZ Chain in a Magnetic Field: Bosonization and Luttinger Liquid Description

We consider now a generalization of the one-dimensional SU(2) Hamiltonian (6.9) for S = 1/2, by including an anisotropy term in the z direction, which we parameterize by Δ , and an external magnetic field h applied along the z-axis. The resulting model is known as the XXZ chain which, being integrable, allows for a detailed analysis of the low energy theory using abelian

bosonization. This simple theory also serves as a starting point for the study of many different situations which can be described by its perturbations as the case of modulated chains or N leg ladders made up of XXZ chains. Given its importance, we present the bosonization analysis in detail.

The lattice Hamiltonian is given by

$$H_{XXZ}^{latt} = J \sum_{n} \left(\frac{1}{2} \left(S_n^+ S_{n+1}^- + S_n^- S_{n+1}^+ \right) + \Delta S_n^z S_{n+1}^z \right) - h \sum_{n} S_n^z \,. \quad (6.56)$$

where we consider J > 0. $S_n^{\pm} = S_n^x \pm i S_n^y$ are the spin raising and lowering operators where $S_n^{x,y,z}$ are the spin operators acting on site n and satyisfying the SU(2) algebra (6.1). In this section we restrict ourselves to S = 1/2.

This model has a U(1) invariance corresponding to rotations around the internal z axis for generic Δ . For $\Delta = 0$ we have the XY model which can be solved exactly using the Jordan-Wigner transformation and it is the starting point of the bosonization procedure that we describe below. The full SU(2) spin symmetry is recovered at $\Delta = 1$ and h = 0 where it is more convenient to apply non-abelian bosonization. This case will be discussed in Sect. 9.3.

We first summarize the outcome of the bosonization of the XXZ chain and then present its derivation in detail. For a complete bibliography see [4,14–17] and references therein.

The Hamiltonian (6.56) is exactly solvable by Bethe ansatz and it can be shown that its low-energy properties are described by a scalar boson with a Hamiltonian given by

$$H_{XXZ}^{cont} = \frac{v}{2} \int \mathrm{d}x \left(K \left(\partial_x \tilde{\phi}(x) \right)^2 + \frac{1}{K} \left(\partial_x \phi(x) \right)^2 \right) \tag{6.57}$$

where ϕ is the field dual to the scalar field ϕ and it is defined in terms of its canonical momentum as $\partial_x \tilde{\phi} = \Pi$. This notation is usually introduced in order to simplify the expressions of the spin operators in the continuum limit; see the Appendix for details on our conventions.

The Fermi velocity v and the so-called Luttinger parameter K depend on both the magnetic field and the anisotropy parameter Δ . These two parameters determine completely the low energy dynamics of the lattice model and they can be computed from the Bethe Ansatz solution. For zero magnetic field and $-1 < \Delta < 1$ they can be found in closed form:

$$K(\Delta) = \frac{\pi}{2(\pi - \theta)} \qquad v(\Delta) = \frac{\pi}{2} \frac{\sin \theta}{\theta}$$
(6.58)

where $\cos(\theta) = \Delta$ and we have set J = 1. Otherwise, one has to solve numerically a set of integro-differential equations (see [18]) which result is discussed below.

The Hamiltonian (6.57) corresponds to a conformal field theory with central charge c = 1 and the free boson is compactified at radius R, *i.e.* it satisfies $\phi = \phi + 2\pi R$, where R is related to the Luttinger parameter K as $R^2 = 1/(2\pi K)$. The importance of this restriction is discussed in the Appendix.

Let us consider first the XY case, *i.e.* $\Delta = 0$, and for convenience let us rotate by π the spins on every second site around the z axis in spin space, which simply amounts to an irrelevant change of the overall sign of the exchange term (in this case J and -J lead to equivalent models).

Then it is convenient to write the spin operators in terms of spinless fermions ψ_n , through the so-called Jordan-Wigner transformation:

$$S_n^z = \psi_n^\dagger \psi_n - 1/2 \tag{6.59}$$

$$S_n^+ = e^{-i\alpha_n} \psi_n^\dagger , \qquad \alpha_n = \pi \sum_{j=0}^{n-1} \left(\psi_j^\dagger \psi_j \right) . \tag{6.60}$$

It is easy to show that these operators satisfy the SU(2) algebra (6.1) provided S = 1/2 and the spinless fermions ψ_n are canonical, *i.e.* $\{\psi_n, \psi_{n'}^{\dagger}\} = \delta_{n,n'}$.

The Hamiltonian (6.56) can then be written as

$$H_{XY}^{latt} = J \sum_{n=1}^{N} \left(-\frac{1}{2} \left(\psi_n^{\dagger} \psi_{n+1} - \psi_n \psi_{n+1}^{\dagger} \right) - h \left(\psi_n^{\dagger} \psi_n - 1/2 \right) \right).$$
(6.61)

This problem can be readily solved by Fourier transforming

$$\tilde{\psi}_k = \frac{1}{\sqrt{N}} \sum_n \psi_n e^{-ikna} \tag{6.62}$$

where a is the lattice spacing and the momentum k is restricted to the first Brillouin zone, $k \in (-\pi/a, \pi/a]$.

$$H_{XY}^{latt} = -J\sum_{k}\cos(k)\tilde{\psi}_{k}^{\dagger}\tilde{\psi}_{k} - h\sum_{k}\tilde{\psi}_{k}^{\dagger}\tilde{\psi}_{k}.$$
(6.63)

where we see that we have one band of fermions with dispersion $e(k) = -J\cos(k)$ and chemical potential -h. The ground state is obtained by filling all single particle states which have energies e(k) < h as in Fig. 6.1

Normalizing the magnetization as $M = \frac{2}{N} \sum_{n} S_{n}^{z} = M(h)$, we see that the Fermi momentum is given by

$$k_F = \pm \frac{\pi}{2} (1+M). \tag{6.64}$$

Since we are interested in the low energy properties of the model, we keep only the modes close to the Fermi surface (here consisting of two points) by restricting the sum in (6.63) to $|k \pm k_F| \leq \Lambda$, with Λ an ultraviolet cutoff. This allows us to study the system at length scales larger than $1/\Lambda$.



Fig. 6.1. Dispersion

Writing the fermions as (x = na)

$$\frac{\psi(x)}{\sqrt{a}} \approx e^{ik_F x} \psi_L(x) + e^{-ik_F x} \psi_R(x) \tag{6.65}$$

where ψ_R and ψ_L vary slowly with x in a scale of order $a > 1/\Lambda$, and contain the Fourier modes around $\pm k_F$ respectively, we obtain

$$H_{XY}^{cont} = iv \int dx [\psi_R^{\dagger} \partial_x \psi_R - \psi_L^{\dagger} \partial_x \psi_L]$$
(6.66)

where the Fermi velocity $v = \partial e(k)/\partial k|_{k=k_F} = Ja \sin(k_F)$ which we set to 1 in what follows. This is the Dirac Hamiltonian in (1 + 1) dimensions. This means that the low energy theory for the XX case (*i.e.* $\Delta = 0$) corresponds to free fermions.

One can easily compute the fermion two point functions for right and left movers that are given by

$$\langle \psi_R(x,t)\psi_R^{\dagger}(0,0)\rangle = \frac{1}{2\pi a}\frac{1}{z}$$
 (6.67)

$$\langle \psi_L(x,t)\psi_L^{\dagger}(0,0)\rangle = \frac{1}{2\pi a} \frac{1}{\bar{z}}$$
 (6.68)

where z = t + ix, $\bar{z} = t - ix$.

From these correlators one can compute the one particle momentum distribution functions which show the characteristic Fermi liquid behaviour. We will see below that this behaviour is changed radically as soon as interactions are taken into account.

In order to treat the interactions that arise for $\Delta \neq 0$ it is more convenient to map the fermionic theory into an equivalent bosonic one, a procedure usually called bosonization. It can be shown [19] that the fermionic theory described by (6.66) can be equivalently reformulated in terms of bosonic variables with Hamiltonian

$$H = \frac{1}{2} \int dx [(\partial_x \phi)^2 + (\partial_x \tilde{\phi})^2], \qquad (6.69)$$

where ϕ is a scalar field and $\tilde{\phi}$ is defined in terms of the conjugate momentum $\Pi(x) = \partial_x \tilde{\phi}(x)$. Canonical commutation relations between ϕ and Π imply

$$[\phi(x), \tilde{\phi}(x')] = -\frac{i}{2} \operatorname{sign}(x - x')$$
(6.70)

while all other commutators are zero.

The key observation is that the fermion operators can be written in terms of the scalar field as

$$\psi_R(x) = \eta_R \frac{1}{\sqrt{2\pi a}} : e^{i\sqrt{4\pi}\phi_R(x)} : , \quad \psi_L(x) = \eta_L \frac{1}{\sqrt{2\pi a}} : e^{-i\sqrt{4\pi}\phi_L(x)} (6.71)$$

where $\eta_{R,L}$ are the so-called Klein factors which satisfy anticommutation relations $\{\eta_i, \eta_j\} = 2\delta_{ij}$. These Klein factors are operators which act on an auxiliary Hilbert space that can be chosen arbitrarily and this freedom is exploited to eliminate them from the effective theory (see below). The right and left components $\phi_{R,L}$ are defined in terms of the bosonic field and its dual as

$$\phi = \phi_R + \phi_L \qquad \tilde{\phi} = \phi_R - \phi_L . \tag{6.72}$$

The fields in the right hand side of (6.71) obey anticommutation rules, as can be easily verified using (6.70), and their two-point functions reproduce the free fermion results (6.68) (see the Appendix for details).

One can further show that the fermionic currents can be bosonized as

$$J_R = : \psi_R^{\dagger} \psi_R : (x) = -\frac{i}{\sqrt{\pi}} \partial_z \phi_R(x),$$

$$J_L = : \psi_L^{\dagger} \psi_L : (x) = \frac{i}{\sqrt{\pi}} \partial_{\bar{z}} \phi_L(x)$$
(6.73)

where Klein factors do not appear here since $\eta_i^{\dagger}\eta_i = 1$ for i = R, L. In the following we will not include the Klein factors explicitly to simplify the notation. This is only possible whenever one can simultaneously diagonalize all the Klein operators which appear in a given problem, which is trivially the case for a single chain: In this case we have only two different Klein operators η_R and η_L and henceforth the only non-trivial products that could appear in the interaction terms are $t_{RL} \equiv \eta_R \eta_L$ and $t_{LR} = -t_{RL}$. We can then choose a basis of the Hilbert space where Klein operators act which diagonalizes t_{RL} and t_{LR} simultaneously and then forget about them. We discuss this issue in more detail in the case of N-leg ladders in Sect. 9.9 where the situation is a bit more complicated. The interaction terms which arise when $\Delta \neq 0$ *i.e.*,

$$\delta H = \Delta \sum_{n=1}^{N} \left(S_n^z S_{n+1}^z \right) = \Delta \sum_{n=1}^{N} \left(\left(\psi_n^{\dagger} \psi_n - 1/2 \right) \left(\psi_{n+1}^{\dagger} \psi_{n+1} - 1/2 \right) \right) ,$$
(6.74)

can be rewritten using (6.59) and (6.65) as

$$\delta H = \Delta \int dx \left[\rho(x) + (-1)^x M(x) \right] \cdot \left[\rho(x+a) + (-1)^{x+a} M(x+a) \right], \quad (6.75)$$

where

$$\rho(x) =: \psi_R^{\dagger} \psi_R + \psi_L^{\dagger} \psi_L : \quad \text{and} \quad M(x) = \psi_L^{\dagger} \psi_R + \psi_R^{\dagger} \psi_L. \tag{6.76}$$

Expanding up to first order in a and eliminating oscillatory terms one obtains

$$\delta H = \Delta \int dx \left(4J_R J_L + J_R^2 + J_L^2 - \left((\psi_L^{\dagger} \psi_R)^2 + H.c. \right) \right) .$$
 (6.77)

The first three terms which are quadratic in the currents are marginal in the renormalization group sense and can be easily handled using bosonization. The last one is irrelevant for $\Delta < 1$ so we postpone its analysis to a later stage. Quadratic interactions between currents arise in the so-called Thirring model and hence are usually termed "Thirring-like" terms.

The current-current terms are bosonized using (6.71) and (6.73) as

$$\delta H = \frac{1}{\pi} \Delta \int dx \left(4\partial_x \phi_L \partial_x \phi_R - (\partial_x \phi_R)^2 - (\partial_x \phi_L)^2 \right) . \tag{6.78}$$

This term can be absorbed in (6.69) and the full bosonized XXZ Hamiltonian then reads

$$H = \frac{v}{2} \int dx \left[\frac{1}{K} (\partial_x \phi)^2 + K (\partial_x \tilde{\phi})^2 \right], \qquad (6.79)$$

where, up to first order in Δ , we have

$$K = 1 - \frac{2\Delta}{\pi} , \qquad (6.80)$$

which provide the first term in the expansion of (6.58) for small Δ . The situation with the effective velocity v is less straightforward, as discussed in [20]. In this case one has to take into account the renormalization of the Fermi velocity due to the Δ interaction on the lattice before taking the continuum limit. In this way one gets to first order in Δ

$$v = 1 + \frac{2\Delta}{\pi} . \tag{6.81}$$

One can improve these results by using the exact Bethe Ansatz solution from which one can extract the exact values of v and K, as given in (6.58). The idea is to compare the asymptotics of the XXZ chain correlation functions obtained via the Bethe Ansatz solution in a finite volume L with that of the free boson defined by (6.79). It should be stressed that the relation between k_F and M (6.64) is not modified by the interactions. One further shows in this way that the bosonic field has to be compactified with a radius R given in terms of the Luttinger parameter K as $R^2 = 1/(2\pi K)$. This means that ϕ and $\phi + 2\pi R$ are identified at each point, and this leads to strong restrictions to the possible perturbations which could appear (see the Appendix).

We can now study the effects of the interactions on the low energy behaviour.

To this end, let us first compute the two point functions of the fermions for $\Delta \neq 0$. Using (6.71) and (6.79) one can easily show that (6.67) modifies to

$$\langle \psi_R(x,t)\psi_R^{\dagger}(0,0)\rangle = \frac{1}{2\pi a} \frac{1}{z^{2d}\bar{z}^{2\bar{d}}}$$
 (6.82)

where d = (K+1/K+2)/8, $\bar{d} = (K+1/K-2)/8$ and $K(\Delta)$ is given in (6.58). A similar expression is obtained for the left-handed fermions. One can already observe the drastic change in the exponents caused by the interactions.

The most dramatic effect of the interactions is the disappearance of the quasiparticle peak in the Fourier transformed Green function, with the consequent disappearance of the finite jump in the momentum distribution function.

More precisely, the spectral function at zero temperature which is defined as

$$\rho(q,\omega) \equiv -\frac{1}{\pi} \text{Im} G^R(k_F + q,\omega) , \qquad (6.83)$$

where $G^{R}(k,\omega)$ is the Fourier transformed retarded two point function

$$G^{R}(x,t) \equiv -i\Theta(t) \left\langle \left\{ \psi_{R}(x,t), \psi_{R}^{\dagger}(0,0) \right\} \right\rangle , \qquad (6.84)$$

can be computed to give

$$\rho(q,\omega) = -2\sin(2\pi D)\Gamma(1-2d)\Gamma(1-2\bar{d})|w-q|^{2d-1}|w+q|^{2\bar{d}-1} . \quad (6.85)$$

where $D = d + \bar{d}$ is the scaling dimension of the interacting fermion.

From this last expression one can obtain the single particle density of states by integrating over the momentum, which leads to a power law behaviour

$$N(\omega) \approx |w|^{2D-1} \tag{6.86}$$

instead of the delta function peak characteristic of a Fermi liquid.
One can also compute the momentum distribution which gives

$$n(k) \approx n(k_F) + \text{const. sign}(k - k_F)|k - k_F| + \cdots$$
 (6.87)

instead of the Fermi liquid behaviour in which n(k) presents a finite jump at k_F , showing again the radical difference between the low energy theory of the XXZ chain and a Fermi liquid.

Another crucial difference between a Fermi liquid and our present theory is that in the former the exponents that control the space decay of correlations are universal (in the sense that they do not depend on the interactions) while they do depend on the interactions in the latter case.

All these features have motivated the name of Luttinger liquid to describe this kind of systems [10].

As a final step, the bosonized expressions for the spin operators are obtained using (6.59), (6.60), (6.65) and (6.71) leading to

$$S_x^z \approx \frac{1}{\sqrt{2\pi}} \frac{\partial \phi}{\partial x} + a : \cos(2k_F x + \sqrt{2\pi}\phi) : + \frac{\langle M \rangle}{2}, \qquad (6.88)$$

and

$$S_x^{\pm} \approx (-1)^x : \mathrm{e}^{\pm i\sqrt{2\pi}\tilde{\phi}}(b\cos(2k_F x + \sqrt{2\pi}\phi) + c) : \qquad (6.89)$$

where we have rescaled $K \to 2K$ in what follows, so that the free fermion point now corresponds to K = 2. The colons denote normal ordering with respect to the groundstate with magnetization $\langle M \rangle$, which leads to the constant term in (6.88). The prefactor 1/2 arises from our normalization of the magnetization to saturation values $\langle M \rangle = \pm 1$. The constants a, b and c are non-universal and can be computed numerically and in particular an exact expression for b has been proposed in [21] for h = 0.

As we mentioned above, the parameter K in (6.57) can be computed by solving a set of integral equations obtained in the Bethe ansatz solution [22]. The results obtained from them are summarized in the magnetic phase diagram for the XXZ-chain (Fig. 6.2). There are two gapped phases: A ferromagnetic one at sufficiently strong fields and an antiferromagnetic phase for $\Delta > 1$ at small fields. In between is the massless phase where the bosonized form (6.57) is valid [18].

The transition between the ferromagnetic commensurate phase and the massless incommensurate phase, which occurs on the line $h_{uc} = (1 + \Delta)J$, is an example of the Dzhaparidze-Nersesyan-Pokrovsky-Talapov, universality class [23, 24], *i.e.* for $\langle M \rangle \rightarrow 1$ the magnetization behaves as

$$\left(\langle M \rangle - M_c\right)^2 \sim h^2 - h_{uc}^2 \tag{6.90}$$

with here $M_c = 1$.

This transition, which is an example of a commensurate-incommensurate (C-IC) transition can be described in the bosonization language by noticing



Fig. 6.2. Magnetic phase diagram of the XXZ-chain (6.56).

that for magnetic fields above saturation, $(h > h_{uc})$, one has to consider an additional operator which becomes commensurate for $\langle M \rangle = 1$. The Hamiltonian is then given by

$$H = H_0 + \int dx \cos \sqrt{2\pi} \phi(x) + h_{\text{eff}} \int dx \partial_x \phi$$
 (6.91)

where the last term corresponds to the interaction with the magnetic field in the bosonized language $(h_{\text{eff}} \propto h)$ has no effect for $h > h_{uc}$ due to the presence of the gap [25]. The cos term which arises at $\langle M \rangle = 1$ is relevant and then responsible for the gap. By decreasing $h \rightarrow h_{uc}$ one can then drive the system into a massless regime and precisely at the transition point the Luttinger parameter takes the universal value K = 2 [23, 24].

The other transition line starts at $\Delta = 1$ and h/J = 0, *i.e.* at the SU(2) point (see Sect. 9.3 for the study of this case using non-abelian bosonization). The Luttinger parameter takes the value K = 1 at this point and hence one has to include in the analysis of the low-energy dynamics the operator of dimension 2K

$$\mathcal{O}(x) = \cos(\sqrt{8\pi\phi(x)}) , \qquad (6.92)$$

which is marginal at this point and becomes relevant for smaller K (bigger Δ). One can easily show how this operator arises by plugging the bosonized expression of S^z (6.88) in the Δ interaction term (6.74). This operator opens a gap in the spectrum via a Kosterlitz-Thouless transition [26] and from the Bethe Ansatz equations one readily obtains the characteristic stretched exponential decay for the gap for Δ slightly bigger than one:

$$\frac{h_c}{J} \sim 4\pi e^{-\frac{\pi^2}{2\sqrt{2(\Delta-1)}}} \qquad (\text{for } \Delta \text{ slightly bigger than 1}). (6.93)$$

6.9.2 Thermodynamics and Correlations

We are now ready to analyze the thermodynamic properties of the XXZ chain in the low energy limit. Spin-spin correlation functions can be computed using (6.88, 6.89) together with the Hamiltonian (6.57) as well as (6.186, 6.189, 6.195) in the Appendix, with g = 1/K. One obtains in this way the following expressions for the equal time correlators (we set m = 1 hereafter):

$$\langle S_{x_1}^z S_{x_2}^z \rangle \approx \frac{\langle M \rangle^2}{4} + \frac{K}{4\pi^2} \frac{1}{|x_1 - x_2|^2} + \frac{a^2}{2} \frac{\cos(2k_F(x_1 - x_2))}{|x_1 - x_2|^K}$$
(6.94)

$$\langle S_{x_1}^+ S_{x_2}^- \rangle \approx -\frac{b^2}{2} \frac{\cos((2k_F - \pi)(x_1 - x_2))}{|x_1 - x_2|^{K + \frac{1}{K}}} + (-1)^{(x_1 - x_2)} \frac{c^2}{|x_1 - x_2|^{\frac{1}{K}}} (6.95)$$

where both staggered and non-staggered contributions are obtained.

From (6.94) we observe that for $\Delta > 0$, *i.e.* in the AF region, K < 2 and hence the staggered contribution dominates, signaling the expected tendency towards antiferromagnetic ordering. For $\Delta > 2$ instead, since K > 2, it is the non-staggered term that dominates at long distances, as expected in the ferromagnetic side. However, as expected in one dimension, there is no true long range order since the correlators decay slowly with a power law, which is called quasi-long range order. More importantly, it should be stressed that the power law decay is given by the Luttinger parameter K which is nonuniversal and depends on the microscopic details, such as the anisotropy Δ the magnetic field, etc.

Using the above expressions one can compute different thermodynamic properties such as the magnetic static susceptibility [27] and transport properties such as the dynamical susceptibility and thermal conductivity. These computations can be extended to finite (small) temperature by performing a conformal transformation which maps the plane (z) into the cylinder (ζ) . This transformation compactifies the imaginary time direction via

$$z(\zeta) = \exp(2\pi\zeta/\beta) , \qquad (6.96)$$

where $\beta = 1/T$.

Following [27], let us compute the magnetic susceptibility, which is defined as

$$\chi \equiv \frac{\partial M}{\partial h} = \beta \; \frac{Tr[(\sum_{n} S_{n}^{z})^{2} e^{-\beta H}]}{Tr[e^{-\beta H}]} - \beta \frac{Tr\left[(\sum_{n} S_{n}^{z}) e^{-\beta H}\right]^{2}}{[Tr[e^{-\beta H}]]^{2}}.$$
 (6.97)

and hence

$$\chi = \beta \left(L \sum_{n} \left\langle S_n^z S_0^z \right\rangle - M^2 \right) \tag{6.98}$$

Using the bosonized expression for the spin operators and noticing that after the integration the oscillating terms are eliminated, we are led to compute

$$\beta L \sum_{n} \langle S_n^z S_0^z \rangle \to \beta \int_{-\infty}^{\infty} dx \, \langle S_x^z S_0^z \rangle = \frac{\beta}{2\pi} \int_{-\infty}^{\infty} dx \, \langle \partial_x \phi(x) \partial_x \phi(0) \rangle \,, \quad (6.99)$$

Using (6.187), (6.188) one can easily compute the needed zero temperature correlations (recovering the Fermi velocity)

$$\langle \partial_x \phi_{R,L}(x,\tau) \partial_x \phi_{R,L}(0,0) \rangle = -\frac{K}{4\pi (v\tau \pm ix)^2} . \tag{6.100}$$

We can extend this result to finite (but small) temperatures by means of the conformal transformation (6.96), which leads to the replacement

$$v\tau \pm ix \to (v\beta/\pi)\sin(\pi \frac{v\tau \pm ix}{v\beta})$$
 (6.101)

in (6.100).

We are thus led to evaluate

$$\beta \int_{-\infty}^{\infty} dx \left\langle S_x^z S_0^z \right\rangle = -\frac{K}{8v^2 \beta} \int_{-\infty}^{\infty} dx \left(\frac{1}{\sin^2(\pi \frac{v\tau + ix}{v\beta})} + \frac{1}{\sin^2(\pi \frac{v\tau - ix}{v\beta})} \right),\tag{6.102}$$

which can be easily done by using the following change of variables $u = \tan(\pi \tau/\beta); w = -i \tan(i\pi x/(v\beta)).$

We finally obtain

$$\chi = \frac{K}{\pi v}.\tag{6.103}$$

This result is valid for small temperatures and independent of T as it is expected from the scale invariance of the system.

By including the effects of the operator (6.92) which is irrelevant for $\Delta < 1$ and becomes marginal at the SU(2) point as we already discussed, one can compute the next to leading term in the low temperature behavior of the susceptibility. One can do this by computing the two point correlator of the current in (6.99) using perturbation theory to include the perturbation term. For $1/2 < \Delta < 1$ one obtains a correction term proportional to $T^{4(K-1)}$ and for $\Delta < 1/2$ it takes the universal form T^2 . In the SU(2) case, $\Delta = 1$, the perturbation is marginally irrelevant and the correction term to the low temperature susceptibility is then logarithmic, $\propto \ln^{-1}(T_0/T)$ with T_0 a given constant. This result has been shown to agree quite well with the exact Bethe Ansatz result [27].

Notice that the susceptibility diverges when we approach the ferromagnetic point $\Delta \to -1$ because both v and K^{-1} vanish in this limit (see (6.58)). In the massive regime, which occurs for $\Delta > 1$, one obtains the expected exponential decay for $T \to 0$.

6.9.3 SU(2) Point via Non-abelian Bosonization

For $\Delta = 1$ and h = 0, which corresponds to K = 1 (or $R = 1/\sqrt{2\pi}$) and $\langle M \rangle = 0$ (and hence $k_F = \pi/2$) one recovers the full SU(2) spin symmetry. This can be observed *e.g.* in (6.94, 6.95), since they coincide at this particular point:

$$\langle S_{x_1}^z S_{x_2}^z \rangle \approx -\frac{1}{4\pi^2} \frac{1}{|x_1 - x_2|^2} + (-1)^{(x_1 - x_2)} \frac{a^2}{|x_1 - x_2|}$$
(6.104)

$$\langle S_{x_1}^+ S_{x_2}^- \rangle \approx \frac{b^2}{|x_1 - x_2|} + (-1)^{(x_1 - x_2)} \frac{c^2}{|x_1 - x_2|}$$
(6.105)

For certain purposes it is more convenient to use non-abelian bosonization [28] and rewrite both the low energy Hamiltonian and the continuum expressions for the spin operators in this new language.

It can be shown that the scalar boson compactified at radius $R = 1/\sqrt{2\pi}$ is equivalent to the theory describing a SU(2) group valued (matrix) field g with dynamics given by the Wess-Zumino-Witten (WZW) action [28]

$$S[g]_{WZW} = \frac{k}{8\pi} \int d^2 x \operatorname{tr} \left(\partial_{\mu} g \partial^{\mu} g^{-1} \right) + \frac{k}{12\pi} \int d^3 y \epsilon_{ijk} \operatorname{tr} \left(g^{-1} \partial_i g g^{-1} \partial_j g g^{-1} \partial_k g \right).$$
(6.106)

where the trace is taken over the group indices and the so called level k equals 1 in the present case. This theory has been studied in [28] in the context of the non-Abelian bosonization of fermions and in [29] using conformal field theory techniques, where e.g. four point correlators were computed. See [30] for details.

The corresponding Hamiltonian can be written in the Sugawara form which is quadratic in the SU(2) currents

$$H_{WZW} = \frac{1}{k+2} \int dx \left(\mathbf{J}_R \cdot \mathbf{J}_R + \mathbf{J}_L \cdot \mathbf{J}_L \right)$$
(6.107)

where $\mathbf{J}_{R,L} = \operatorname{tr}(\sigma g^{-1}\partial_{z,\bar{z}}g)$ and the spin operators can be compactly written as

$$\mathbf{S}_x \approx (\mathbf{J}_R + \mathbf{J}_L) + \operatorname{const}(-1)^x \operatorname{tr}(\sigma g) \tag{6.108}$$

The two formulation are related as follows

$$g \propto \begin{pmatrix} : \exp(i\sqrt{2\pi}\phi) : : : \exp(-i\sqrt{2\pi}\tilde{\phi}) : \\ - : \exp(i\sqrt{2\pi}\tilde{\phi}) : : : \exp(-i\sqrt{2\pi}\phi) : \end{pmatrix}$$
(6.109)

and

$$J_{R,L}^{z} = \pm \partial_{z,\bar{z}}\phi$$

$$J_{L}^{+} = :\exp(-i\sqrt{8\pi}\phi_{L}:) , \quad J_{R}^{+} = :\exp(-i\sqrt{8\pi}\phi_{R}): . \quad (6.110)$$

The marginally irrelevant perturbation (6.92) can be written in this language as the product of left and right handed currents $\mathbf{J}_R \cdot \mathbf{J}_L$.

At this point this (more complicated) formulation may appear unnecessary except for the fact that the expressions exhibit the SU(2) invariance more naturally. However, the description of the S = 1/2 Heisenberg chain in terms of the level 1 WZW theory is crucial in the study of interacting systems, such as *e.g* the two leg Heisenberg ladder in the weak interchain coupling regime [31,32]. In this case one can exploit the powerful machinery of CFT in two dimensions to study the low energy dynamics of these systems. This particular example is discussed in Sect. 9.7.

6.9.4 Modifications of the XXZ Chain

Using the formalism just developed one can study any modification of the XXZ chain provided that perturbation theory can be safely applied. We discuss now the case in which the exchange couplings J in the Hamiltonian (6.56) have a spatial periodicity of two sites (usually called dimerization) as a sample case, but other perturbations like next-nearest-neighbors, terms breaking XY symmetry, etc. can be treated similarly.

The Hamiltonian is given by

$$H_{XXZ}^{latt} = \sum_{n} J_n \left(\frac{1}{2} \left(S_n^+ S_{n+1}^- + S_n^- S_{n+1}^+ \right) + \Delta S_n^z S_{n+1}^z \right) , \qquad (6.111)$$

where $J_n = J(1 + (-1)^n \delta)$. For $\Delta = 0$ we can map it into a model of free fermions using the Jordan-Wigner transformation (6.60)

$$H = \frac{J}{2} \sum_{n} \left((1 - \delta)(\psi_{2n}^{\dagger} \psi_{2n+1} + H.c.) + (1 + \delta)(\psi_{2n+1}^{\dagger} \psi_{2n+2} + H.c.) \right)$$
(6.112)

Defining on even and odd sites the fermions $\chi_n = \psi_{2n}$ and $\xi_n = \psi_{2n+1}$ and Fourier transforming, one obtains a two by two Hamiltonian which can be diagonalized to give

$$H = J \sum_{k} \left(E_{+} \ \psi_{-k}^{(+)\dagger} \psi_{k}^{(+)} + E_{-} \ \psi_{-k}^{(-)\dagger} \psi_{k}^{(-)} \right)$$
(6.113)

where $\psi_k^{(+)}$ and $\psi_k^{(-)}$ are defined in terms of the Fourier component of χ and ξ as

$$\psi_k^{(+)} = \frac{(1-\delta) + (1+\delta)e^{-ik}}{\sqrt{2}E_+} \chi_k + \frac{1}{\sqrt{2}} \xi_k ,$$

$$\psi_k^{(-)} = \frac{(1-\delta) + (1+\delta)e^{-ik}}{\sqrt{2}E_-} \chi_k + \frac{1}{\sqrt{2}} \xi_k .$$
(6.114)

We have then two bands of fermions with dispersions given by

$$E_{\pm} = \pm \sqrt{(1 + \delta^2 + (1 - \delta^2) \cos k)/2} \tag{6.115}$$

which shows that a half filling there is a gap δ in the spectrum (see Fig. 6.3). Notice that the momentum k here is twice the momentum we have used in (6.62), due to the distinction between even and odd sites we made in going to the new variables χ and ξ .



Fig. 6.3. Energy bands for the dimerized case.

The same model can be studied using bosonization now for arbitrary Δ but perturbatively in the dimerization δ . In this scheme one treats the term $J\delta\sum_{n}(-1)^{n}\left(\frac{1}{2}\left(S_{n}^{+}S_{n+1}^{-}+S_{n}^{-}S_{n+1}^{+}\right)+\Delta S_{n}^{z}S_{n+1}^{z}\right)$ as a perturbation which can be written using (6.88), (6.89). It is easy to show that a new term arises, which is of the form

$$O(x) = \cos\sqrt{2\pi}\phi \tag{6.116}$$

which is relevant and is responsible for the opening of a gap. This operator could have been predicted by symmetry arguments, since once the translation symmetry is broken in the lattice, as it happens in the dimerized case, it is no longer forbidden to appear. This can be seen as follows: translation by one lattice site $x \to x + 1$ implies that the chiral fermions in (6.65) transform as

$$\psi_R \to e^{ik_F}\psi_R, \quad \psi_L \to e^{-ik_F}\psi_L \tag{6.117}$$

and henceforth, for $k_F = \pi/2$, the bosonic field is transformed as $\phi \rightarrow \phi - \sqrt{\pi}/2$. Breaking of this symmetry then allows for a term like (6.116) to appear, which was otherwise forbidden. In the non Abelian SU(2) formulation, the parity breaking operator is simply given by trg.

The gap can be estimated by power counting to be of order $\approx \delta^{(1/(2-K/2))}$. Note that in the XY case (*i.e.* for $\Delta = 0$) K = 2 and we recover the free fermion result. In the next section we compute the RG equations for this effective theory.

6.9.5 RG Analysis of the Scalar Field Perturbed by Vertex Operators

We already used the renormalization group technique when treating the nonlinear sigma model. The case of the scalar bosonic field with a vertex operator is much simpler. Following [8], the action is given by:

$$S = \frac{1}{2} \int dx \ dt \left[\frac{1}{K} \left(\partial_x \phi \right)^2 + \lambda \cos(\beta \phi) \right] \ , \tag{6.118}$$

where $\beta = \sqrt{2\pi}$ corresponds to the dimerized case.

The scaling dimension of the operator $\cos(\beta\phi)$ is $\frac{K\beta^2}{4\pi}$ and the coupling λ has then the dimension $2 - \frac{K\beta^2}{4\pi}$ in order to have a dimensionless action.

Imagine now that, as before, we integrate over the fast degrees of freedom within the momenta shell $\Lambda - \delta \Lambda$ and Λ , or shifting from the scale L to $L + \delta L$. Since λ is a dimensional constant, it has to be rescaled accordingly. Simple dimensional analysis tells us that:

$$\frac{d\lambda}{dln(L)} = \left(2 - \frac{K\beta^2}{4\pi}\right)\lambda \tag{6.119}$$

Of course one may anticipate that fluctuations can change this naïve scaling relation but to lowest order in the coupling constant we can keep this equation for describing the behavior of the system under renormalization group transformations. This is not, however, the end of the story. Let us assume that $\left(2 - \frac{K\beta^2}{4\pi}\right)$ is small *i.e.* we are close to the point where λ is marginal. In the process of integration, we define an effective partition function which we can define through the formal notation:

$$Z = Z_{\text{eff}}(1 - \lambda \int dx \ dt \langle \cos(\beta \phi(x, t)) \rangle + \frac{\lambda^2}{2} \int dx_1 \ dx_2 \ dt_1 \ dt_2 \langle \cos(\beta \phi(x_1, t_1)) \cos(\beta \phi(x_2, t_2)) \rangle + \dots) \quad (6.120)$$

where the integration is taken over scales smaller than δL . The term in parenthesis can then be re-exponentiated and we can define our effective action in terms of the original one:

$$S_{eff} = S - \lambda \int dx \ dt \langle \cos(\beta \phi(x,t)) \rangle + \frac{\lambda^2}{2} \int dx_1 \ dx_2 \ dt_1 \ dt_2 \langle \cos(\beta \phi(x_1,t_1)) \cos(\beta \phi(x_2,t_2)) \rangle + \dots \quad (6.121)$$

We then see that in the process of integrating out the degrees of freedom at small scales, we can have the merging of two vertex operators separated by a distance smaller than δL . In this operator product, there is certainly the term $\cos(2\beta\phi)$ which is present and contributes to the renormalization group equations, but can also be neglected to first order. There is however another term in the product expansion of the vertex operators:

$$e^{i\beta\phi(x)}e^{-i\beta\phi(x+\delta x)} \to -\frac{\beta^2 \left(\partial_u \phi\right)^2}{|\delta x|^{\frac{K\beta^2}{2\pi}-2}} + \dots$$
(6.122)

So the constant K gets a correction:

$$\frac{1}{K_{eff}} = \frac{1}{K} + \lambda^2 \beta^2 \int_{L}^{L+\delta L} \frac{d^2 \delta x}{|\delta x|^{2-\epsilon}}$$
$$= \frac{1}{K} + \frac{\lambda^2 \beta^2}{\epsilon} 2\pi ((L+\delta L)^{\epsilon} - L^{\epsilon})$$
(6.123)

where $\epsilon = \frac{4-K\beta^2}{2\pi}$ is supposed to be small. From this result, and from (6.119) we can write the renormalization group equations to lowest order:

$$\frac{d\lambda}{dln(L)} = \left(2 - \frac{K\beta^2}{4\pi}\right)\lambda + \dots$$
$$\frac{dK}{dln(L)} = -\frac{K^2\beta^2}{4\pi}\lambda^2 + \dots$$
(6.124)

which are known as the Kosterlitz renormalization group equations. The flow diagram for these equations is well known [8]. The flow is depicted in Fig. 6.4. In the vicinity of $\lambda = 0$, the line $\lambda = \lambda_c(K) = \left(\frac{K\beta^2}{8\pi} - 1\right)$ separates the regions of initial conditions that flow to weak coupling and strong coupling respectively. If $K > \frac{8\pi}{\beta^2}$ and $\lambda < \lambda_c(K)$, the large scale behavior of the system corresponds to a massless scalar field theory, while elsewhere the system presents a massive behavior with a finite correlation length.

6.9.6 Charge Degrees of Freedom: Hubbard and t - J Models

The methods described in the previous sections can be extended to study systems including spin and charge degrees of freedom, provided they are Bethe ansatz solvable. Such is the case of the Hubbard model which is exactly solvable for arbitrary values of the on-site repulsion U, filling and magnetic field [33]. The exact solution can then be used to construct a low energy bosonized effective field theory [34–36] which can then be used to study perturbations of this model (see *e.g.* [14, 37]).

Here we present some aspects of the bosonization description of the Hubbard chain and its applications (see [15, 16, 37] and references therein).



Fig. 6.4. Renormalization Group flow

The Hubbard model describes electrons hopping on a lattice which interact repulsively via an on-site Coulomb energy U with the lattice Hamiltonian given by

$$H = -t \sum_{n,\alpha} (c_{n+1,\alpha}^{\dagger} c_{n,\alpha} + H.c.) + U \sum_{n} c_{n,\uparrow}^{\dagger} c_{n,\uparrow} c_{n,\downarrow}^{\dagger} c_{n,\downarrow}$$
$$+ \mu \sum_{n} (c_{n,\uparrow}^{\dagger} c_{n,\uparrow} + c_{n,\downarrow}^{\dagger} c_{n,\downarrow}) - \frac{h}{2} \sum_{n} (c_{n,\uparrow}^{\dagger} c_{n,\uparrow} - c_{n,\downarrow}^{\dagger} c_{n,\downarrow}) . \quad (6.125)$$

Here $c_{n,\alpha}^{\dagger}$ and $c_{n,\alpha}$ are electron creation and annihilation operators at site $n, \alpha = \uparrow, \downarrow$ the two spin orientations, h the external magnetic field and μ the chemical potential. As we already mentioned, this model has been exactly solved by Bethe Ansatz already in 1968 [33] but it took until 1990 for the correlation functions to be computed by combining Bethe Ansatz results with Conformal Field Theory (CFT) techniques [34].

Spin-charge separation is one of the important features of the Hubbard chain at zero magnetic field. Interestingly, it is no longer spin and charge degrees of freedom that are separated if an external magnetic field is switched on [34]. Nevertheless it has been shown that in the presence of a magnetic field, the spectrum of low energy excitations can be described by a semi-direct product of two CFT's with central charges c = 1 [34]. This in turn implies that the model is still in the universality class of the Tomonaga-Luttinger (TL) liquid and therefore allows for a bosonization treatment.

We proceed as before by setting U = 0 and writing the fermion operators as (now we have fermions with spin, and hence the number of equations is duplicated)

$$c_{n,\alpha} \to \psi_{\alpha}(x) \sim \mathrm{e}^{ik_{F,\alpha}x} \psi_{L,\alpha}(x) + \mathrm{e}^{-ik_{F,\alpha}x} \psi_{R,\alpha}(x) + \dots \quad (6.126)$$

$$= e^{ik_{F,\alpha}x} e^{-i\sqrt{4\pi\phi_{L,\alpha}(x)}} + e^{-ik_{F,\alpha}x} e^{i\sqrt{4\pi\phi_{R,\alpha}(x)}} + \dots , (6.127)$$

where $k_{F,\alpha}$ are the Fermi momenta for up and down spin electrons, which are related to the filling and the magnetization as

$$k_{+} = k_{F,\uparrow} + k_{F,\downarrow} = \pi n ; \qquad k_{-} = k_{F,\uparrow} - k_{F,\downarrow} = \pi \langle M \rangle , \qquad (6.128)$$

The fields $\phi_{R,L,\alpha}$ are the chiral components of two bosonic fields, which bosonize the spin up and down chiral fermion operators $\psi_{R,L,\alpha}$, as in (6.71). The dots stand for higher order terms which have to be computed in order to reproduce the correct asymptotics of correlations obtained from the Bethe Ansatz solution. They take into account the corrections arising from the curvature of the dispersion relation due to the Coulomb interaction. The effects of band curvature due to interactions are also present in the case of the XXZ chain. However, in that case the effects are, for most practical purposes, negligible, since they lead in general to additional terms in the bosonization formulae which are strongly irrelevant operators. In the present case, though, these terms can be important since in some cases they could be relevant and should then be taken into account. For non-zero Hubbard repulsion U and magnetic field h, the low energy effective Hamiltonian corresponding to (6.125) written in terms of the bosonic fields ϕ_{\uparrow} and ϕ_{\downarrow} has a complicated form, mixing up and down degrees of freedom [36].

The crucial step to obtain a simpler bosonized Hamiltonian is to consider the Hamiltonian of a generalized (two component) TL model and identify the excitations of the latter with the exact Bethe Ansatz ones for the model (6.125), providing in this way a *non-perturbative* bosonic representation of the low energy sector of the full Hamiltonian (6.125). This program has been carried out in [36] and reviewed in [37].

The fixed point (*i.e.* neglecting all irrelevant terms) bosonized Hamiltonian can be written as

$$\sum_{i=c,s} \frac{u_i}{2} \int dx \left[\left(\partial_x \phi_i \right)^2 + \left(\partial_x \theta_i \right)^2 \right] , \qquad (6.129)$$

where $\phi = \phi_R + \phi_L$ and $\theta = \phi_R - \phi_L$ and the new bosonic fields ϕ_c and ϕ_s are related to ϕ_{\uparrow} and ϕ_{\downarrow} through

$$\begin{pmatrix} \phi_c \\ \phi_s \end{pmatrix} = \frac{1}{\det Z} \begin{pmatrix} Z_{ss} & Z_{ss} - Z_{cs} \\ Z_{sc} & Z_{sc} - Z_{cc} \end{pmatrix} \begin{pmatrix} \phi_{\uparrow} \\ \phi_{\downarrow} \end{pmatrix} , \qquad (6.130)$$

In these expressions Z_{ij} , i, j = c, s, are the entries of the dressed charge matrix Z taken at the Fermi points

$$Z = \begin{pmatrix} Z_{cc} \ Z_{cs} \\ Z_{sc} \ Z_{ss} \end{pmatrix} . \tag{6.131}$$

These matrix elements are solutions of a set of coupled integral equations obtained from the Bethe Ansatz [34] and depend on the Hubbard coupling U, the chemical potential μ and the magnetic field h. These parameters play a similar role as that played by K in the case of the XXZ chain.

At zero magnetic field, the matrix Z reduces to

$$Z(h=0) = \begin{pmatrix} \xi & 0\\ \xi/2 & 1/\sqrt{2} \end{pmatrix} , \qquad (6.132)$$

with $\xi = \xi(\mu, U)$. In this case we recover the expressions for the charge and spin fields for zero magnetic field

$$\phi_c = \frac{1}{\xi} \left(\phi_{\uparrow} + \phi_{\downarrow} \right) , \quad \phi_s = \frac{1}{\sqrt{2}} \left(\phi_{\uparrow} - \phi_{\downarrow} \right) , \quad (6.133)$$

where the compactification radius of the spin field (*i.e.* the parameter which indicates the period of ϕ_s , $\phi_s = \phi_s + 2\pi R_s$, $R_s = 1/\sqrt{2\pi}$) is fixed by the SU(2) symmetry of the spin sector (it corresponds to the Luttinger parameter for the spin sector being $K_s = 1$). The radius for the charge field, on the other hand, depends on the chemical potential μ and the Coulomb coupling U.

One very important fact that we already mentioned is that for h = 0 the charge and spin degrees of freedom are completely decoupled, a phenomenon which is known as spin-charge separation. In particular, since the velocities for the two kinds of excitations are different, it is easy to verify that if one creates a particle (true electron) on the ground state, its constituents (spin and charge parts) will, after some time, be located in different points in space.

It should be noted that for $M \neq 0$, the fields arising in the diagonalized form of the bosonic Hamiltonian (6.129) are no longer the charge and spin fields.

For generic values of the parameters of the model (6.125), we can now write down for example the bosonized expressions for the charge density operator and for the z component of the spin operator

$$\rho(x) = \psi_{\uparrow}^{\dagger}\psi_{\uparrow}(x) + \psi_{\downarrow}^{\dagger}\psi_{\downarrow}(x)
= \frac{1}{\sqrt{\pi}}\partial_{x}\left(Z_{cc}\phi_{c} - Z_{cs}\phi_{s}\right) + a \sin[k_{+}x - \sqrt{\pi}\left(Z_{cc}\phi_{c} - Z_{cs}\phi_{s}\right)]
\times \cos[k_{-}x - \sqrt{\pi}\left((Z_{cc} - 2Z_{sc})\phi_{c} - (Z_{cs} - 2Z_{ss})\phi_{s}\right)]
+ b\sin(2k_{+}x - \sqrt{4\pi}(Z_{cc}\phi_{c} - Z_{cs}\phi_{s})), \quad (6.134)$$

$$2S^{z} = \psi_{\uparrow}^{\dagger}\psi_{\uparrow} - \psi_{\downarrow}^{\dagger}\psi_{\downarrow} = c \;\partial_{x}((Z_{cc} - 2Z_{sc})\phi_{c} - (Z_{cs} - 2Z_{ss})\phi_{s}) \\ + d \;\cos[k_{+}x - \sqrt{\pi}(Z_{cc}\phi_{c} - Z_{cs}\phi_{s})] \\ \times \sin[k_{-}x - \sqrt{\pi}((Z_{cc} - 2Z_{sc})\phi_{c} - (Z_{cs} - 2Z_{ss})\phi_{s})] \\ - e \sin[2k_{-}x - \sqrt{4\pi}((Z_{cc} - 2Z_{sc})\phi_{c} - (Z_{cs} - 2Z_{ss})\phi_{s})] \;(6.135)$$

where a, b, c, d, e are non-universal constants. Other operators can be constructed similarly and then correlations can be easily computed following similar lines as for the XXZ chain.

In the limit of large U, double occupancy will be forbidden and one can use perturbation theory in t/U to show that this Hamiltonian reduces to the so called t - J model in this limit which for zero magnetic field reads

$$H_{t-J} = -t \sum_{n,\alpha} (c_{n+1,\alpha}^{\dagger} c_{n,\alpha} + H.c.) + J \sum_{n} \mathbf{S}_{n} \cdot \mathbf{S}_{n+1}$$
(6.136)

where the operator \mathbf{S}_n represents the spin of the electron at site n,

$$\mathbf{S}_n = c_{n,\alpha}^{\dagger} \frac{\sigma_{\alpha\beta}}{2} c_{n,\beta} \tag{6.137}$$

with σ the Pauli matrices and the spin exchange constant is given by $J = t^2/U$.

In the case of zero field, $k_{F,\uparrow} = k_{F,\downarrow} = k_F$, and the expressions for the charge density and the S^z spin operators are simplified to

$$\rho(x) = \frac{\xi}{\sqrt{\pi}} \partial_x \phi_c + a \sin(2k_F x - \sqrt{\pi} \xi \phi_c) \times \cos(\sqrt{2\pi} \phi_s) + b \sin(4k_F x - \sqrt{4\pi} \xi \phi_c) , \qquad (6.138)$$

$$S^{z} = c \ \partial_{x}\phi_{s} + d \ \cos(2k_{F}x - \sqrt{\pi} \ \xi\phi_{c}) \times \cos(\sqrt{2\pi}\phi_{s}) + e\sin(\sqrt{8\pi}\phi_{s})$$
(6.139)

If one works at half-filling, which in this language means one electron per lattice site and hence $k_F = \pi/2$, there is an extra operator perturbing the charge sector which opens a charge gap even for arbitrarily small U. Then one can integrate out the charge degrees of freedom to recover the S = 1/2Heisenberg chain studied before, which describes the Mott insulating phase of the Hubbard model.

After freezing the massive charge degrees of freedom, the spin operator reads

$$S^{z} = c \ \partial_{x}(\phi_{s}) + const \ \times (-1)^{x} \cos(\sqrt{2\pi\phi_{s}}) , \qquad (6.140)$$

where $const \propto (\cos(\sqrt{\pi} \xi \phi_c))$ and we recover the expression in (6.88) for $k_F = \pi/2$.

The t - J model is not Bethe Ansatz solvable in general, but only at the specific point J = 2t where it becomes supersymmetric [38]. At this point one can follow a similar procedure as described above to construct the bosonized low energy theory from the Bethe Ansatz solution.

6.9.7 Two-Leg Heisenberg Ladder

We have seen by using the NLSM approach that one should expect a spin gap in the spectrum of the two-leg Heisenberg ladder. In the present section we study the same problem using a different technique which further supports this conclusion.

We apply the combination of non-abelian bosonization techniques and the powerful machinery of conformal field theories in two dimensions to the case of a two leg S = 1/2 Heisenberg antiferromagnetic spin ladder following [31,32]. This is one of the simplest examples where the combination of these techniques shows its power by allowing for a complete analysis of the low energy dynamics.

The Hamiltonian is defined as

$$H_{2-leg}^{latt} = J\left(\mathbf{S}_{n}^{1} \cdot \mathbf{S}_{n+1}^{1} + \mathbf{S}_{n}^{2} \cdot \mathbf{S}_{n+1}^{2}\right) + J'\mathbf{S}_{n}^{1} \cdot \mathbf{S}_{n}^{2},$$
(6.141)

where J, J' are the intrachain and interchain couplings respectively. We work in the weak interchain coupling limit $J' \ll J$, which allows us to apply the bosonization procedure described in Sect. 9.3 to each of the chains as if they were decoupled. We then treat the interchain couplings with the aid of (6.108) in perturbation theory.

The low energy limit Hamiltonian then takes the form

$$H_{2-leg}^{cont} = H_{WZW}^1 + H_{WZW}^2 + \lambda_1 \int dx \left(\left(\mathbf{J}_R^1 + \mathbf{J}_L^1 \right) \cdot \left(\mathbf{J}_R^2 + \mathbf{J}_L^2 \right) \right) + \lambda_2 \int dx \left(\operatorname{tr}(\sigma g^1) \cdot \operatorname{tr}(\sigma g^2) \right)$$
(6.142)

where $\lambda_{1,2} \propto J'/J$.

The key observation here is that the free theory (J' = 0) corresponds to two $SU(2)_1$ WZW factors and this CFT theory can be conformally embedded into

$$SU(2)_1 \otimes SU(2)_1 \supset SU(2)_2 \otimes Z_2 , \qquad (6.143)$$

where $SU(2)_2$ stands for the level 2 WZW theory and Z_2 corresponds to the Ising CFT.

This last equation does not indicate the complete equivalence of the theory on the r.h.s. with that on the l.h.s. What is true is that both theories have the same conformal central charge and all the primary fields of the theory on the l.h.s. are contained in the r.h.s theory. The idea is to try to map all the interaction terms into the new language, which in fact turns out to be possible in this case (though it is not generically true).

One can write the interaction terms in (6.142) using this embedding and the outcome is quite nice, since the two sectors are decoupled from each other, each of them with their respective mass terms.

There are two kinds of interaction terms in (6.142), the first being the current-current terms, which have the effect of renormalizing the effective Fermi velocity to first order apart from marginal terms. We then have the more relevant terms which are the product of the two WZW fields.

To study the effect of these relevant terms we use the conformal embedding mentioned above. The first observation is that the product of the WZW fields in the two $SU(2)_1$ sectors has scaling dimension 1 and should hence be writable in terms of dimension 1 operators in the Ising and $SU(2)_2$ WZW sectors. In this way, one obtains the following correspondence

$$\operatorname{tr}(\sigma g^1) \cdot \operatorname{tr}(\sigma g^2) = \operatorname{tr}(\Phi_{j=1}) - 3 \epsilon , \qquad (6.144)$$

which can be proved by comparing the operator product expansions of the operators on the left and right hand sides. In the above equation, the field $\Phi_{j=1}$ is the spin 1 field in the WZW theory $SU(2)_2$ and ϵ is the energy operator in the Ising sector, which can be described by one Majorana fermion.

We can then conclude that the Ising sector, being perturbed by the energy operator, has a mass m_1 proportional to J'/J.

This theory can be further simplified by noticing that the level 2 SU(2) WZW theory can be equivalently described as three Majorana fermions. In this new language, the corresponding interaction term, $tr(\Phi_{j=1})$ simply provides the mass m_2 for these Majorana fermions, which is different from m_1 and again proportional to J'/J. The ratio between the masses of the different Ising sectors has been fixed using Abelian bosonization in [32], showing that $m_1/m_2 = -3$.

The effective Hamiltonian can then be written as

$$H_{2-leg}^{eff} = -\frac{i}{2} \left(\zeta_R \partial_x \zeta_R - \zeta_L \partial_x \zeta_L \right) - im_1 \zeta_R \zeta_L + \sum_{a=1}^3 \left(-\frac{i}{2} \left(\xi_R^a \partial_x \xi_R^a - \xi_L^a \partial_x \xi_L^a \right) - im_2 \xi_R^a \xi_L^a \right) , \quad (6.145)$$

apart from marginal terms coming from the current-current interactions.

A similar result can be obtained using Abelian bosonization as in [32]. Different modifications of the two leg ladder considered here, as the inclusion of dimerization, extra diagonal couplings between the chains, etc. can be treated using the same formalism.

6.9.8 Higher Spin Chains: Non-abelian Bosonization

In the case of the Heisenberg antiferromagnet with higher values of the spin S one can still represent the spin variables in terms of fermions, which now carry an extra internal (color) index. The generalization of (6.137) for arbitrary S reads

$$\mathbf{S}_{n} = \sum_{i=1}^{25} \sum_{\alpha,\beta=\uparrow,\downarrow} c_{\alpha in}^{\dagger} \frac{\sigma_{\alpha\beta}}{2} c_{\beta in}, \qquad (6.146)$$

where $c_{\alpha in}$ are fermionic variables with α the spin index, i = 1, ..., 2S the extra color index and n the site index respectively. As before, $\sigma_{\alpha\beta}$ are the Pauli matrices.

~ ~

In this case, non-abelian bosonization is more suitable to deal with the low energy theory. This approach has been first introduced in [39] (see also [40]). Here we follow the path-integral approach presented in [41], which is more suitable for our purposes.

In order to correctly represent the spin S chain, the physical states $|phys\rangle$ must satisfy at each lattice site the constraints

$$\sum_{i} c^{\dagger}_{\alpha i n} c_{\alpha i n} |\text{phys}\rangle = 2S |\text{phys}\rangle$$
$$\sum_{i,j} c^{\dagger}_{\alpha i n} \tau^{a}_{ij} c_{\alpha j n} |\text{phys}\rangle = 0, \qquad (6.147)$$

where τ^a are the generators of the SU(2S) algebra. The first constraint imposes the condition that allows only one spin per site, whereas the second one states that the physical states must be color singlets.

The Heisenberg Hamiltonian (6.56) with spin operators satisfying (6.1) with $\Delta = 1$ and h = 0, can then be expressed as

$$H = -\frac{1}{2} \sum_{n} c^{\dagger}_{\alpha i n} c_{\alpha j n+1} c^{\dagger}_{\beta j n+1} c_{\beta i n} + \text{constant}$$
(6.148)

which has a local $SU(2S) \times U(1)$ local gauge invariance introduced by the parametrization (6.146). This quartic interaction can be rewritten by introducing an auxiliary field B as

$$H = \frac{1}{2} \sum_{n} (B_{n,n+1}^{ij} c_{\alpha i n}^{\dagger} c_{\alpha j n+1} + H.c. + \bar{B}_{n+1,n}^{ji} B_{n,n+1}^{ij}).$$
(6.149)

To obtain an effective low energy theory we perform a mean field approximation taking B as a constant $2S \times 2S$ matrix whereafter H can then be diagonalized. We then introduce the fluctuations around this mean solution, which we are able to integrate in a path-integral setup.

We write each color fermion as in (6.65)

$$\frac{\psi_{i\alpha}(x)}{\sqrt{a}} \approx e^{ik_F x} \psi_{L,i\alpha}(x) + e^{-ik_F x} \psi_{R,i\alpha}(x) , \qquad (6.150)$$

with $k_F = \pi/2$ and expand the auxiliary field around its mean field value, keeping the fluctuations to first order in the lattice spacing, since we are interested in the low energy dynamics

$$B_{xy} = B_0 e^{aV_{xy}} \simeq B_0 (1 + aV_{xy}), \tag{6.151}$$

We define the fields $A^1 \equiv \frac{1}{2}(V_{xy} - V_{xy}^{\dagger})$ and $R_{xy} \equiv \frac{1}{2}(V_{xy} + V_{xy}^{\dagger})$ in the algebra of U(2S) which are the fluctuation fields which we have to integrate to obtain the effective low energy partition function. When substituted back into the Hamiltonian, the expansion (6.151) leads to a quadratic integral in R_{xy} which can be performed to give

$$H = B_0 \left(-i\Psi_{R,i\alpha}^{\dagger} (\delta_{ij}\partial_x + A_{ij}^{\dagger})\Psi_{R,j\alpha} + i\Psi_{L,i\alpha}^{\dagger} (\delta_{ij}\partial_x + A_{ij}^{\dagger})\Psi_{L,j\alpha} \right) + \frac{1}{4} \left(\Psi_{L,i\alpha}^{\dagger}\Psi_{R,j\alpha} - \Psi_{R,i\alpha}^{\dagger}\Psi_{L,j\alpha} \right)^2, \qquad (6.152)$$

where the last term arises from the integration over the R field.

In order to implement the constraints (6.147) we first rewrite them in the continuum limit using (6.150). In terms of the continuum fermions the constraints read

$$\begin{split} \bar{\Psi}_{i\alpha}\gamma_0\Psi_{i\alpha}|\text{phys}\rangle &= 2S|\text{phys}\rangle ,\\ \bar{\Psi}_{i\alpha}\gamma_0\tau_{ij}\Psi_{j\alpha}|\text{phys}\rangle &= 0 ,\\ \bar{\Psi}_{i\alpha}\Psi_{j\alpha}|\text{phys}\rangle &= 0 \quad \text{for all } i,j , \end{split}$$
(6.153)

where $\Psi^{\dagger} = (\Psi_R, \Psi_L)$ and $\bar{\Psi} = \Psi^{\dagger} \gamma_0$.

The first two constraints are implemented by introducing a Lagrange multiplier A^0 in the Lie algebra of U(2S), which together with A^1 in (6.152) provide the two space-time components of a gauge field in U(2S). The third constraint is instead imposed with the use of the identity (see [41] for details)

$$\delta[\bar{\Psi}_{i\alpha}\Psi_{j\alpha}] = \lim_{\lambda_2 \to \infty} e^{-\lambda_2 \int d^2 x \ (\bar{\Psi}_{i\alpha}\Psi_{j\alpha})^2} \ . \tag{6.154}$$

After some algebra, the effective Lagrangian reads

$$L = \bar{\Psi}\gamma^{\mu}iD_{\mu}\Psi - \lambda_1(i\bar{\Psi}_i\gamma_5\Psi_j)^2 - \lambda_2(i\bar{\Psi}_i\Psi_j)^2, \qquad (6.155)$$

where the covariant derivative is defined as $D_{\mu} = \partial_{\mu} - ia_{\mu} + B_{\mu}$, and we have decomposed, for later convenience, the $U(2S) A_{\mu}$ field into a U(1) field a_{μ} and a SU(2S) field B_{μ} .

The Lagrangian can be further rewritten as

$$L = \bar{\Psi}_{i\alpha} \gamma^{\mu} i (\partial_{\mu} - i a_{\mu} \delta_{ij} \delta_{\alpha\beta} + B^{ij}_{\mu} \delta_{\alpha\beta}) \Psi_{j\beta} + 4 (\lambda_1 + \lambda_2) \mathbf{J}_R \cdot \mathbf{J}_L + (\lambda_1 + \lambda_2) j_R j_L - (\lambda_1 - \lambda_2) (\Psi^{\dagger}_{Ri\alpha} \Psi_{Lj\alpha} \Psi^{\dagger}_{Rj\beta} \Psi_{Li\beta} + H.c.),$$
(6.156)

where

$$\mathbf{J}_{R,L} = \Psi_{R,Li\alpha}^{\dagger} \frac{\sigma_{\alpha\beta}}{2} \Psi_{R,Li\beta}$$
$$j_{R,L} = i \Psi_{R,Li\alpha}^{\dagger} \Psi_{R,Li\alpha} \qquad (6.157)$$

are $SU(2)_{2S}$ and U(1) currents respectively.

For $\lambda_1 = \lambda_2 = 0$ we are left with the theory of 2S Dirac fermions coupled to gauge fields in U(1) and SU(2S). Since these gauge fields have no dynamics, they act as Lagrange multipliers and it can be shown that the resulting theory corresponds to the fermionic realization of the coset model [42]

$$\frac{U(2S)}{U(1) \otimes SU(2S)_2} \equiv SU(2)_{2S}$$
(6.158)

as was already observed in [41]. The third term can be absorbed by a redefinition of the U(1) gauge field a_{μ} .

The second and last terms in (6.156) can then be expressed as fields in the resulting WZW theory $SU(2)_{2S}$

$$\Delta \mathcal{L} = (\lambda_1 - \lambda_2) \left(\Phi_{\alpha\beta}^{(1/2)} \Phi_{\beta\alpha}^{(1/2)} + H.c. \right) + 4(\lambda_1 + \lambda_2) \mathbf{J}_R \cdot \mathbf{J}_L$$
(6.159)

where we have identified the spin 1/2 primary field of the $SU(2)_{2S}$ WZW theory, $\Phi^{(1/2)}$, in terms of its fermionic constituents

$$\Phi_{\alpha\beta}^{(1/2)} \equiv \Psi_{R,i\alpha}^{\dagger} \Psi_{L,i\beta} \tag{6.160}$$

which has conformal dimensions $d = \bar{d} = 3/(8(S+1))$. The first term in (6.159) corresponds then to the spin 1 affine primary $\Phi^{(1)}$ with conformal dimensions $d = \bar{d} = 1/(S+1)$, as can be seen after some simple algebra.

We can finally write

$$\Delta \mathcal{L} = -4 \ (\lambda_1 - \lambda_2) \ tr \ \Phi^{(1)} + 4(\lambda_1 + \lambda_2) \mathbf{J}_R \cdot \mathbf{J}_L \tag{6.161}$$

For $S = \frac{1}{2}$ we recover the effective model we derived in Sect. 9.3. In this case, the first term in (6.161) is proportional to the identity operator and the second is marginally irrelevant since $\lambda_1 + \lambda_2$ is positive and gives the well known logarithmic corrections to correlators.

For higher spins, we have to consider the interaction term (6.161) and we also have to include all other terms which are radiatively generated. We then need the operator product expansion (OPE) coefficients among the different components of $\Phi^{(1)}$ which have been computed in [43]. The OPE coefficients are non-vanishing if the so called "Fusion Rules" are non-vanishing. In the level k SU(2) WZW theory they are given by [44]

$$\Phi_{m,\bar{m}}^{(j)} \times \Phi_{m',\bar{m}'}^{(j')} = \sum_{n=|j-j'|}^{\min(j+j',k-j-j')} \Phi_{m+m',\bar{m}+\bar{m}'}^{(n)}$$
(6.162)

We can now make use of the following equivalence [43, 45]

$$SU(2)_k \equiv Z_k \otimes U(1) \tag{6.163}$$

We will exploit this equivalence to derive an effective low energy action for the spin S Heisenberg chain. Indeed, it was shown in [43] that the primary fields of the $SU(2)_k$ WZW theory are related to the primaries of the Z_k parafermion theory and the U(1) vertex operators. They are connected by the relation

$$\Phi_{m,\bar{m}}^{(j)}(z,\bar{z}) = \phi_{2m,2\bar{m}}^{(2j)}(z,\bar{z}) : e^{\frac{i}{\sqrt{2S}}(m\phi_R(z) + \bar{m}\phi_L(\bar{z}))} :, \tag{6.164}$$

where the Φ fields are the invariant fields of the $SU(2)_k$ WZW theory, the ϕ fields are the Z_k parafermion primaries and ϕ_R and ϕ_L are the holomorphic and antiholomorphic components of a compact massless free boson field. In the same way, the currents are related as

$$J_R^+(z) = (2S)^{1/2} \psi_1(z) : \exp\left(\frac{i}{\sqrt{2S}} \phi_R(z)\right) : ,$$

$$J_R^z(z) = (2S)^{1/2} \partial_z \phi_R(z)$$
(6.165)

where $J_R^{\pm} = J_R^x \pm i J_R^y$ and ψ_1 is the first parafermionic field. (A similar relation holds for the left-handed currents).

Using this equivalence we can express the relevant perturbation term (6.161) in the new language as

$$\Delta \mathcal{L} = -4(\lambda_1 - \lambda_2) \left(\phi_{0,0}^{(2)} + \phi_{2,-2}^{(2)} : e^{\frac{i}{\sqrt{2S}}(\phi_R(z) - \phi_L(\bar{z}))} : + \phi_{-2,2}^{(2)} : e^{-\frac{i}{\sqrt{2S}}(\phi_R(z) - \phi_L(\bar{z}))} : \right) + 4S(\lambda_1 + \lambda_2) \left(\psi_1 \bar{\psi}_1^{\dagger} : e^{\frac{i}{\sqrt{2S}}(\phi_R(z) - \phi_L(\bar{z}))} : + H.c. \right), \quad (6.166)$$

where we absorbed the derivative part of the U(1) field coming from (6.165) into a redefinition of the constant in front of the unperturbed Lagrangian. The first term corresponds to the first "thermal" field of the parafermion theory, $\phi_{0,0}^{(2)} = \epsilon_1$, with conformal dimensions $d = \bar{d} = 1/(1+S)$, while the second and third terms correspond to the p = 2 disorder operator in the PF theory, $\phi_{2,-2}^{(2)} = \mu_2$ and its adjoint $\phi_{-2,2}^{(2)} = \mu_2^{\dagger}$ with dimensions $d_2 = \bar{d}_2 = (S-1)/(2S(S+1))$. It is assumed that all the operators which are radiatively generated have to be included in the complete effective theory.

We use now the fact that the Z_{2S} PF theory perturbed by its first thermal operator ϵ_1 flows into a massive regime irrespectively of the sign of the coupling [46]. Assuming that, as for the Z_2 case, due to the sign of the coupling $\lambda_1 - \lambda_2$ in (6.166) the theory is driven into a low temperature ordered phase, we have that vacuum expectation values (v.e.v.'s) of disorder operators μ_j , vanish for $j \neq 2S \mod(2S)$ as well as v.e.v.'s of the parafermionic fields $\langle \psi_k \bar{\psi}_k^{\dagger} \rangle = 0$, for $2k \neq 2S \mod(2S)$. This will be important in the computation of spin-spin correlation functions below.

Since the parafermionic sector is massive, the effective theory for large scales can be obtained by integrating out these degrees of freedom. One can obtain then the most general effective action for the remaining U(1) field, by including all the vertex operators which are invariant under the symmetry $Z_{2S} \times \tilde{Z}_{2S}$ [43]

$$\phi_R \to \phi_R - \frac{\sqrt{2\pi}m}{\sqrt{S}} ; \ \phi_L \to \phi_L - \frac{\sqrt{2\pi}n}{\sqrt{S}}$$
 (6.167)

with $m, n \in \mathbb{Z}$, which is preserved after the integration of the massive parafermions.

One obtains in this way the effective action for the remaining U(1) theory

$$Z_{\text{eff}} = \int d\phi \, \exp\left(-\int K_S(\partial_\mu \phi)^2 + \alpha_S \int \cos(\sqrt{\frac{S}{2}}(\phi_R - \phi_L)) + \beta_S \int \cos(\sqrt{2S}(\phi_R - \phi_L)) + \cdots\right),$$
(6.168)

for S integer while α_S vanishes for S half integer. Here the dots indicate irrelevant fields corresponding to higher harmonics of the scalar field and K_S is an effective constant arising from the OPE of vertex and parafermionic operators in the process of integration of the massive degrees of freedom.

Using the generalization of (6.65) to the case with 2S colors together with (6.146), (6.157) and (6.160) we can write the continuum expression of the original spin operator $\mathbf{S}(x)$ as

$$\mathbf{S}(x) = \mathbf{J}_R + \mathbf{J}_L + \text{const} (-1)^x \text{tr}(\frac{\sigma}{2} (\Phi^{(1/2)} + \Phi^{(1/2)\dagger})) , \qquad (6.169)$$

which is the generalization of (6.108) for arbitrary spin S.

Let us study the behavior of the spin-spin correlation function at large scales, to see whether the system has a gap or not. In the new language of (6.169), these correlators have a staggered and a non-staggered part which correspond respectively to current-current correlators and correlators of the components of the fundamental field $\Phi^{(1/2)}$.

Let us focus on the staggered part of the $S^z S^z$ correlator: Since our original SU(2) WZW model is perturbed, correlation functions of the fundamental field will contain supplementary operators coming from the OPE of the product of $\Phi^{(1/2)}$ and the perturbing fields. With the help of the fusion rules (6.162) it is easy to see that, for example, the effective alternating

z-component of the spin operator containing the scalar field will be given by:

$$\sum_{k \le 2S, \ k \text{ odd}} a_k \ \mu_k : e^{\frac{ik}{2\sqrt{2S}}(\phi_R(z) - \phi_L(\bar{z}))} : +H.c., \tag{6.170}$$

where only odd k fields appear in the sum.

For S half-integer, the operator $\Phi^{(S)}$ is present in (6.170), and we can easily check that, (since μ_{2S} corresponds to the identity), this operator is simply given by

 $e^{\frac{iS}{\sqrt{2S}}(\phi_R - \phi_L)} + H.c.$

The other operators in the series contain parafermionic disorder operators whose correlators will decay exponentially to zero at large scales. Thus, considering only the Gaussian part of (6.168), we can show that the spin correlation functions at large scales behave like:

$$\langle S_z(x)S_z(y)\rangle \sim (-1)^{(x-y)}|x-y|^{-2SK_S} \langle S_+(x)S_-(y)\rangle \sim (-1)^{(x-y)}|x-y|^{-1/(2SK_S)}$$
(6.171)

The fact that the SU(2) symmetry is unbroken at all scales fixes then the value of K_S to be

$$K_S = 1/(2S) \tag{6.172}$$

For this value of K_S one can show that the perturbing operator with coupling β_S in (6.168) is marginally irrelevant (remember that $\alpha_S = 0$ in the half-integer case).

We conclude then that the large scale behavior of half-integer spin chains is given by the level 1 SU(2) WZW model with logarithmic corrections as for the spin 1/2 chain.

Let us consider now integer spins S. Since the series (6.170) for the effective spin operator contains only half-integer spins j (odd k's), all the operators in the series will contain non-trivial parafermionic operators. Then all the terms in the spin-spin correlation function will decay exponentially to zero with the distance indicating the presence of a gap in the excitation spectrum, thus confirming Haldane's conjecture.

6.9.9 N-Leg Ladders in a Magnetic Field: Gap for Non-zero Magnetization

Another interesting situation is the one of antiferromagnetic spin ladders which we have already studied in Sect. 7 using NLSM techniques in the SU(2)symmetric case. The Hamiltonian for coupled XXZ chains in the presence of a magnetic field is a generalization of that presented in (6.141) [18]

$$H_{N-ladder}^{latt} = \sum_{a=1}^{N} H_{XXZ}^{(a)} + J' \sum_{n,a=1}^{a=N} \mathbf{S}_{n}^{a} \cdot \mathbf{S}_{n}^{a+1} - h \sum_{i,n} S_{n}^{a,z}, \qquad (6.173)$$

where $H_{XXZ}^{(a)}$ is given by an expression like (6.56) for each chain labeled by a.

For J' = 0 one can map the low energy sector of each XXZ chain into a bosonic field theory as described in Sect. 9. One obtains in this way an effective description which consists in a collection of identical Hamiltonians like (6.57), with N bosonic fields, ϕ_a , describing the low energy dynamics of chain $a, a = 1, \dots, N$. The interchain exchanges give rise to perturbation terms which couple the fields of the different chains.

In the case in which many chains are considered, one has to introduce as many different Klein factors as the number of chains considered for both right and left components, η_R^a , η_L^a , a = 1, ..., N, to ensure the correct commutation relations between spin fields (see (6.71) for the case of a single chain). In the present case, the interactions contain generically products of four Klein factors of the form

$$t_{ijkl} \equiv \eta_i \eta_j \eta_k \eta_l \tag{6.174}$$

where the subindices here indicate the pair index (α, a) , with $\alpha = R, L$ and athe chain index. One can easily show using the Klein algebra, $\{\eta_i, \eta_j\} = 2\delta_{ij}$, that $t^2 = 1$ when all indices are different and then these operators have eigenvalues ± 1 . As discussed in [15], one could get rid of the t operators which appear in the interaction terms (and hence bosonize completely the problem) provided one can simultaneously diagonalize all the operators like (6.174) appearing in a given situation. This in turn can be done if all these operators are mutually commuting, which has to be studied for each case separately. In the present situation this can be easily shown by noticing that interchain interactions between a and b chains contain products of the form

$$\eta_R^a \eta_L^a \eta_R^b \eta_L^b \tag{6.175}$$

with $a \neq b$, a, b = 1, ..., N and using the algebra of the Klein factors one can show that they are all mutually commuting.

After a careful RG analysis, one can show that at most one degree of freedom, given by the combination of fields $\phi_D = \sum_a \phi_a$, remains massless. The large scale effective action for the ladder systems is then given again by a Hamiltonian (6.57) for ϕ_D and the perturbation term

$$H_{pert} = \lambda \int dx \, \cos(2Nk_F x + \sqrt{2\pi}\phi_D) \,, \qquad (6.176)$$

where $k_F = (1 + \langle M \rangle)\pi/2$ is related to the total magnetization $\langle M \rangle$.

The key point if to identify the values of the magnetization for which the perturbation operator (6.176) can play an important rôle. In fact, this operator is commensurate at values of the magnetization given by

$$N/2(1 - \langle M \rangle) \in \mathbb{Z}, \qquad (6.177)$$

otherwise the integral over x will make this term vanish due to the fast oscillations of the phase factor since the continuum fields are slowly varying.

If this operator turns out to be also relevant in the RG sense (this depends on the parameters of the effective Hamiltonian (6.57), the model will have a finite gap, implying a plateau in the magnetization curve.

Let us see how this condition can be obtained: in the weak-coupling limit along the rungs, $J' \ll J$, the bosonized low-energy effective Hamiltonian for the N-leg ladder reads

$$H_{N-ladder}^{cont} = \int \mathrm{d}x \left[\frac{1}{2} \sum_{a=1}^{N} \left(v_a K_a \left(\partial_x \tilde{\phi}_a(x) \right)^2 + \frac{v_a}{K_a} \left(\partial_x \phi_a(x) \right)^2 \right) + \lambda_1 \sum_{a,b} \left(\partial_x \phi_a(x) \right) \left(\partial_x \phi_b(x) \right) + \sum_{a,b} \left\{ \lambda_2 : \cos(2(k_F^a + k_F^b)x + \sqrt{2\pi}(\phi_a + \phi_b)) : (6.178) \right\}$$

$$+\lambda_3: \cos\left(2(k_F^a - k_F^b)x + \sqrt{2\pi}(\phi_a - \phi_b)\right): +\lambda_4: \cos\left(\sqrt{2\pi}(\tilde{\phi}_a - \tilde{\phi}_b)\right): \right\} \right],$$

where only the most relevant perturbation terms are kept. The four coupling constants λ_i essentially correspond to the coupling J' between the chains: $\lambda_i \sim J'/J$. In arriving to the Hamiltonian (6.178) we have discarded a constant term and absorbed a term linear in the derivatives of the free bosons into a redefinition of the applied magnetic field. For simplicity we have used here periodic boundary conditions (PBC's) along the transverse direction.

Note that the λ_2 and λ_3 perturbation terms contain an explicit dependence on the position (in the latter case this *x*-dependence disappears for symmetric configurations with equal k_F^i). Such operators survive in passing from the lattice to the continuum model, assuming that the fields vary slowly, only when they are commensurate. In particular, the λ_2 term appears in the continuum limit only if the oscillating factor $\exp(i2x(k_F^i + k_F^j))$ equals unity. If the configuration is symmetric, this in turn happens only for zero magnetization (apart from the trivial case of saturation).

Let us describe this in some detail for the case of the three leg ladder, N = 3. In this case we first diagonalize the Gaussian (derivative) part of the Hamiltonian by the following change of variables in the fields:

$$\psi_1 = \frac{1}{\sqrt{2}} (\phi_1 - \phi_3) , \psi_2 = \frac{1}{\sqrt{6}} (\phi_1 + \phi_3 - 2\phi_2) , \psi_D = \frac{1}{\sqrt{3}} (\phi_1 + \phi_2 + \phi_3) .$$
(6.179)

In terms of these fields the derivative part of the Hamiltonian can be written as: 300 D.C. Cabra and P. Pujol

$$\bar{H}_{\text{der.}} = \frac{vK}{2} \int dx \left[(1+a) \left(\partial_x \psi_D(x) \right)^2 + (1-b) \left(\left(\partial_x \psi_1(x) \right)^2 + \left(\partial_x \psi_2(x) \right)^2 \right) \right]$$
(6.180)

where a = J'K/J = 2b. We can now study the large-scale behaviour of the effective Hamiltonian (6.178) where we assume all k_F^i equal due to the symmetry of the chosen configuration of couplings. Let us first consider the case when the magnetization $\langle M \rangle$ is non-zero. In this case only the λ_3 and λ_4 terms are present. The one-loop RG equations are:

$$\frac{dK}{d\ln(L)} = -2K^2 \lambda_3^2 + 2\lambda_4^2$$
$$\frac{d\lambda_3}{d\ln(L)} = \left(2 - \frac{K}{(1-b)}\right)\lambda_3 - \pi\lambda_3^2$$
$$\frac{d\lambda_4}{d\ln(L)} = \left(2 - \frac{(1-b)}{K}\right)\lambda_4 - \pi\lambda_4^2.$$
(6.181)

It is important to notice that only the fields ψ_1 and ψ_2 enter in these RG equations, since the perturbing operators do not contain the field ψ_D . The behaviour of these RG equations depends on the value of K. The main point is that always one of the two λ perturbation terms will dominate and the corresponding cosine operator tends to order the associated fields. This gives a finite correlation length in correlation functions containing the fields ψ_1 and ψ_2 (or their duals). For example, for $\Delta \leq 1$ we have that K > 1 since $\langle M \rangle \neq 0$. Then, from (6.181) one can easily see that the dominant term will be the λ_4 one. This term orders the dual fields associated with ψ_1 and ψ_2 . Then, the correlation functions involving these last fields decay exponentially to zero. In either case, the field ψ_D remains massless. In the case of open boundary conditions the situation is similar and again it is the diagonal field the one which stays generically massless, in spite of the asymmetry of the Gaussian part of the action.

A more careful analysis of the original Hamiltonian shows that this diagonal field will be coupled to the massive ones only through very irrelevant operators giving rise to a renormalization of its Luttinger parameter K. However, due to the strong irrelevance of such coupling terms these corrections to K are expected to be small, implying that its large-scale effective value stays close to the zero-loop result.

At the values of the magnetization where this operator is commensurate, the field ψ_D can then undergo a K-T transition to a massive phase, indicating the presence of a plateau in the magnetization curve. An estimate of the value of J' at which this operator becomes relevant can be obtained from its scaling dimension. In the zero-loop approximation and for $\Delta = 1$ one then obtains $J'_c \approx 0.09J$ for the $\langle M \rangle = 1/3$ plateau at N = 3.

Appendix: The Scalar Boson in 2D, a c = 1 Conformal Field Theory

Primary Field Content and Correlators

The action for the scalar Euclidean boson is

$$S(\phi) = \frac{g}{2} \int d^2 x \left(\partial_\mu \phi\right)^2 \tag{6.182}$$

and in condensed matter applications g is related to the Luttinger parameter as g = 1/K and hence contains the information about the interactions.

This action is invariant under constant translations of the field

$$\phi(x) \to \phi'(x) = \phi(x) + \alpha , \qquad (6.183)$$

with the corresponding conserved current $J_{\mu}(x) = \partial_{\mu}\phi(x)$. There exists another (trivially) conserved current $\tilde{J}_{\mu}(x) = \epsilon_{\nu\mu}\partial_{\nu}\phi(x)$ (usually referred to as "topological" current).

The corresponding Hamiltonian reads

$$H = \frac{1}{2} \int dx \left(\frac{1}{g} \Pi(x)^2 + g (\partial_x \phi)^2 \right) , \qquad (6.184)$$

where the wave propagation velocity has been set to 1 and the canonical conjugate momentum $\Pi \equiv \delta \mathcal{L}/\delta \dot{\phi} = g \dot{\phi}$. The dual field $\tilde{\phi}$ which is usually defined for convenience, since it allows to write certain fields in a local way, is related to Π as $\partial_x \tilde{\phi} = \Pi$. One can eliminate g from (6.184) by making a canonical transformation

$$\phi' = \sqrt{g}\phi , \qquad \Pi' = \frac{1}{\sqrt{g}}\Pi . \qquad (6.185)$$

The propagator is then given by

$$\Delta(z,\bar{z};w,\bar{w}) \equiv \langle 0|\phi'(z,\bar{z})\phi'(w,\bar{w})|0\rangle = -\frac{1}{4\pi}\log m^2|z-w|^2 \qquad (6.186)$$

where $z = v\tau + ix$, $\bar{z} = v\tau - ix$ and m is a small mass which has been added as an infrared regulator. Ultraviolet divergences are naturally regulated in the problems we will be interested in by the lattice constant a. We will drop the primes in the scalar fields from now on, but the reader should keep in mind (6.185).

From this correlator one can read the chiral parts $(\phi(z, \bar{z}) = \phi_R(z) + \phi_L(\bar{z}))$

$$\langle \phi_R(z)\phi_R(w)\rangle = -\frac{1}{4\pi}\log m(z-w) \tag{6.187}$$

and

$$\langle \phi_L(\bar{z})\phi_L(\bar{w})\rangle = -\frac{1}{4\pi}\log m(\bar{z}-\bar{w}) . \qquad (6.188)$$

In terms of the chiral components the dual field reads $\tilde{\phi} = \phi_R(z) - \phi_L(\bar{z})$.

Taking derivatives from (6.187) one obtains

$$\langle \partial_z \phi_R(z) \partial_w \phi_R(w) \rangle = -\frac{1}{4\pi} \frac{1}{(m(z-w))^2} \tag{6.189}$$

and similarly for the anti-holomorphic components.

The energy-momentum tensor for the free massless boson is

$$T_{\mu\nu} =: \left(\partial_{\mu}\phi\partial_{\nu}\phi - \frac{1}{2}\eta_{\mu\nu}\partial_{\rho}\phi\partial_{\rho}\phi\right):, \qquad (6.190)$$

where the dots : : denote normal ordering defined by substracting the singular part of the product when the arguments coincide.

Its holomorphic component reads

$$T \equiv -2\pi T_{zz} = -2\pi : \partial \phi_R \partial \phi_R := \lim_{z \to w} \left(\partial \phi_R(z) \partial \phi_R(w) - \langle \partial \phi_R(z) \partial \phi_R(w) \rangle \right)$$
(6.191)

and similarly for the anti-holomorphic component $\overline{T}(\overline{z})$.

The two point correlators of the energy-momentum tensor components can be easily computed using (6.189) and (6.191) to give

$$\langle T(z) | T(w) \rangle = \frac{c/2}{(m(z-w))^4} , \qquad \langle \bar{T}(\bar{z}) | \bar{T}(\bar{w}) \rangle = \frac{c/2}{(m(\bar{z}-\bar{w}))^4}, \quad (6.192)$$

with the numerical constant c = 1. These expressions define the central charge of the model. Indeed, it can be shown that the modes of the energy momentum tensor components T and \overline{T} satisfy respective Virasoro algebras with central charge c = 1.

One usually defines vertex operators in terms of the chiral components of the field ϕ as

$$V_{\alpha,\bar{\alpha}}(z,\bar{z}) := \exp\left(i\alpha\phi_R(z) + i\bar{\alpha}\phi_L(\bar{z})\right) :=$$

$$: \exp\left(i\frac{(\alpha + \bar{\alpha})}{2}\phi(z,\bar{z}) + i\frac{(\alpha - \bar{\alpha})}{2}\tilde{\phi}(z,\bar{z})\right) :, \qquad (6.193)$$

for arbitrary real numbers α and $\bar{\alpha}$, where normal ordering is defined as usual and for the vertex operators we can write

$$:e^{i\alpha\phi}: \equiv e^{i\alpha\phi_{\text{creation}}} e^{i\alpha\phi_{\text{annihilation}}}$$
(6.194)

Their two-point functions are readily evaluated to give

$$\left\langle V_{\alpha,\bar{\alpha}}(z,\bar{z})V_{\alpha,\bar{\alpha}}^{\dagger}(w,\bar{w})\right\rangle = \frac{1}{(m(z-w))^{\frac{\alpha^2}{4\pi}}(m(\bar{z}-\bar{w}))^{\frac{\bar{\alpha}^2}{4\pi}}}$$
 (6.195)

Since this expression is infrared divergent, one has to renormalize the vertex operators in order to make their correlators IR finite

$$V_{\alpha,\alpha}(z,\bar{z})|_{\text{ren}} \equiv m^{\frac{\alpha^2}{4\pi}} : e^{(i\alpha\phi(z,\bar{z}))} :$$
 (6.196)

From (6.195) one can read off the conformal dimensions d and \bar{d} of $V_{\alpha,\bar{\alpha}}$

$$d = \frac{\alpha^2}{8\pi} \qquad \bar{d} = \frac{\bar{\alpha}^2}{8\pi} . \tag{6.197}$$

The scaling dimension Δ and the conformal spin S are defined as $D = d + \bar{d}$ and $S = d - \bar{d}$ respectively. Below we will see how the restriction on the conformal spin to be integer or half-integer restricts the possible values of α and $\bar{\alpha}$.

Multipoint correlators are also easily evaluated and the general result is (for simplicity we take $\alpha_i = \bar{\alpha}_i$)

$$\left\langle \prod_{i=1}^{N} V_{\alpha_i,\alpha_i}(z_i, \bar{z}_i) \right\rangle \bigg|_{ren} = \prod_{i < j} |z_i - z_j|^{\frac{\alpha_i \alpha_j}{2\pi}}, \quad \text{if} \quad \sum_{i=1}^{N} \alpha_i = 0, \quad (6.198)$$

and zero otherwise.

The neutrality condition $\sum_{i=1}^{N} \alpha_i = 0$ is necessary for the cancellation of the renormalization constants. Otherwise the result vanishes in the zero mass limit.

Compactified Free Boson

So far we have not imposed any condition on the bosonic variable ϕ . However, in many applications in condensed matter systems, like in the XXZ chain (see discussion below (6.81)), the bosonic variable is constrained to live on a circle of radius R (usually called "compactification radius"), *i.e.* ϕ and $\phi + 2\pi R$ are identified at each space-time point. This condition restricts the allowed values for the charges α to integer multiples of 1/R in order for the operators to be well defined. If one further imposes that the conformal spins have to be integers (to ensure single-valuedness of correlators) then the dual field $\tilde{\phi}$ is compactified with $\tilde{R} = \frac{1}{2\pi gR}$ and the allowed charges are restricted to the set

$$\{(\alpha,\bar{\alpha})\} = \{(n/R + 2\pi gmR, n/R - 2\pi gmR), n, m \in \mathbb{Z}\}$$
(6.199)

which correpond to fields with conformal dimensions

$$h_{n,m} = 2\pi g \left(\frac{n}{4\pi g R} + \frac{1}{2}mR\right)^2 , \quad \bar{h}_{n,m} = 2\pi g \left(\frac{n}{4\pi g R} - \frac{1}{2}mR\right)^2 .$$
(6.200)

Notice that the theory is dual under the transformation $R \leftrightarrow \frac{1}{2\pi gR}$, which amounts to the interchange of the so called electric and magnetic charges (respectively n and m in (6.200)).

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7 The Coupled Cluster Method Applied to Quantum Magnetism

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Abstract. The Coupled Cluster Method (CCM) is one of the most powerful and universally applied techniques of quantum many-body theory. In particular, it has been used extensively in order to investigate many types of lattice quantum spin system at zero temperature. The ground- and excited-state properties of these systems may now be determined routinely to great accuracy. In this Chapter we present an overview of the CCM formalism and we describe how the CCM is applied in detail. We illustrate the power and versatility of the method by presenting results for four different spin models. These are, namely, the XXZ model, a Heisenberg model with bonds of differing strengths on the square lattice, a model which interpolates between the Kagomé- and triangular-lattice antiferromagnets, and a frustrated ferrimagnetic spin system on the square lattice. We consider the ground-state properties of all of these systems and we present accurate results for the excitation energies of the spin-half square-lattice XXZ model. We utilise an "extended" SUB2 approximation scheme, and we demonstrate how this approximation may be solved exactly by using Fourier transform methods or, alternatively, by determining and solving the SUB2-m problem. We also present the results of "localised" approximation schemes called the LSUBm or SUBm-m schemes. We note that we must utilise computational techniques in order to solve these localised approximation schemes to "high order." We show that we are able to determine the positions of quantum phase transitions with much accuracy, and we demonstrate that we are able to determine their quantum criticality by using the CCM in conjunction with the coherent anomaly method (CAM). Also, we illustrate that the CCM may be used in order to determine the "nodal surfaces" of lattice quantum spin systems. Finally, we show how connections to cumulant series expansions may be made by determining the perturbation series of a spin-half triangular-lattice antiferromagnet using the CCM at various levels of LSUBm approximation.

7.1 Introduction

Key experimental observations in fields such as superfluidity, superconductivity, nuclear structure, quantum chemistry, quantum magnetism and strongly correlated electronic systems have often implied that the strong quantum correlations inherent in these systems should be fully included, at least conceptually, in any theoretical calculations that aim fully to describe their basic properties. Until fairly recently a common problem in many of these fields has been that the "conceptual school" of quantum many-body theory (QMBT) has been rather divorced from the "quantitative school" of fully microscopic QMBT. In this context the conceptual school typically simplifies the original, fundamental theoretical model to a more tractable one. This is done either by replacing the original Hamiltonian with a simpler or effective one that still implies or includes the most important of the observed features, and/or by postulating that these key features can be captured via an (approximate) wave function with specific inbuilt correlations. The BCS state for superconductors is a typical example of the latter. By contrast, the quantitative school attempts to solve the original quantum many-body problem as accurately as possible. Nowadays the boundaries between the two schools are becoming increasingly blurred for several important reasons. Thus, on one hand, many of the most interesting problems, such as high- temperature superconductivity, are so difficult that neither school can present convincing solutions. On the other hand, the techniques now available in the field of *ab initio* QMBT have become increasingly refined over the last 15 years or so, and have also become more accessible to a wider group of researchers who can utilise the experience and expertise built up in other fields of application.

Ab initio techniques of microscopic QMBT are, at their best, designed to include the important effects of quantum correlations in an unbiased and systematic manner. In particular, over the last decade or so, some of the QMBT tools that have proven to be versatile in describing very accurately a wide range of both finite and extended systems of interest in physics and chemistry, and which are defined in continuous space, have begun to be applied to quantum lattice systems. They are now beginning to provide unified treatments of such systems, which can not only compete, for example, with other much more computationally intensive stochastic simulations, but can also provide an almost unique means to study in a systematic and unbiased manner the physically interesting (zero-temperature) quantum phase transitions that many such quantum lattice systems display in abundant variety. At the same time the conceptual school of QMBT can often provide a good starting point for the quantitative school, as we shall see in more detail below, in the form of "model" reference states that become the starting or zeroth-order approximations on top of which further many-body correlations can be systematically included within well-defined hierarchical approximation schemes.

Foremost among the most versatile techniques in the modern arsenal of QMBT are those such as quantum Monte Carlo (QMC) methods [1–4] the correlated basis function (CBF) method [5–15] and the coupled cluster method (CCM) [16–24], on the last of which we concentrate in this Chapter. The latter two methods are undoubtedly the most powerful and most universally applicable of all fully microscopic techniques presently available for *ab initio* calculations in QMBT. Each of the above methods has its own particular strengths and weaknesses, as we discuss in more detail below. Before

doing so, however, we first give a short overview of the CBF method since, for reasons discussed more fully below, we shall concentrate our main attention hereafter on the CCM.

The most common, and perhaps the simplest, of the variational methods in QMBT are based on trial wave functions of the (Bijl-Dingle-)Jastrow form [25]. Early calculations of this sort relied on various cluster expansions of the ensuing approximate matrix elements [25–27]. It was realised later that these variational approaches may also be formulated diagrammatically [28]. This feature has been of considerable help in the construction of such powerful approximations as the Percus-Yevick and hypernetted chain (HNC) summations and their variants, which have their origins in the classical theory of liquids and which have been adapted for both bosonic and fermionic systems [7, 29]. The review article by Clark [8] gives a good overview of the variational theory sketched above as applied to extended nuclear matter. The interested reader is also referred to [30].

Two basic flaws mar the above variational approaches. Firstly, the particular partial summations of the graphs considered by such approximations as the HNC approach destroy one of the most attractive features of variational techniques, namely that they yield upper bounds to the exact ground-state energy. Secondly, even a complete summation of graphs (or a variational Monte Carlo evaluation of the corresponding expectation values) for a given trial wave function (of Jastrow type, for example) gives only the exact variational result and not the true ground state. This latter deficiency may be remedied by the inclusion of more general state-dependent correlations and higher-order correlation functions of the Feenberg type. Alternatively, and more generally, one may extend the Jastrow wave function to a complete set of correlated basis functions, which is the CBF approach.

The CBF method was introduced some 45 years ago by Feenberg and his collaborators [5–7], and was later developed largely by Clark and his collaborators [8–11]. Introductory surveys of the method are given in [12–15]. We simply note here that the CBF method has as its central ingredient the direct incorporation of the most important interparticle correlations into the approximate wave functions on which the microscopic description is based. At its simplest level the method only involves a single configuration, and hence reduces to ordinary variational theory. This further reduces to Jastrow theory if the simplest reasonable choice of correlation operator is made in terms of the usual symmetric product over all pairs in the system of state-independent two-body correlation functions.

Since we shall be dealing extensively with applications of the CCM in this Chapter, we postpone a comparable introduction of it until Sec. 1.2, and before doing so we return to a review of the relative merits and weaknesses of the QMC, CBF and CCM approaches to QMBT. We first note that QMC techniques are severely restricted in the choice of problems to which they can readily be applied by the infamous "sign problem" [31, 32], which arises whenever we have a lack of prior knowledge of the nodal surface of the manybody wave function under discussion. For spin systems on a regular lattice it is often related to the occurrence of (strong) frustration. Conversely, it can only readily be circumvented when we have such prior knowledge via, for example, the Marshall-Peierls sign rule [33], or some such analogous relation. Nevertheless, QMC numerical results for spin-lattice systems often provide the benchmark for other methods for the cases in which the technique can be applied, especially for lattices in two or more spatial dimensions. We note in passing that for the special case of one spatial dimension (i.e., chains) the methods of choice usually include exact solutions when available [34–37], the density matrix renormalisation group (DMRG) method [38], and techniques from quantum field theory [39].

By contrast, the CBF method is not limited in the range of systems to which it can be applied by the presence of strong (geometric or dynamic) frustration. However, its applications up till now have been restricted in practice to a very limited number of spin-lattice systems (namely, the transverse Ising model [40–43] and the XY model [44]. We note that part of the reason for this limited usage of the method for problems in quantum magnetism lies in the fact that, in practical calculations, it is often difficult to include correlations beyond the two-body level in the Jastrow-Feenberg trial states. Such higher-order correlations are often important for very accurate calculations.

By further contrast, the CCM is limited neither by the presence of frustration in the system nor to the inclusion of only two-body correlations. As we shall see later, the inclusion of many-body correlations between spins up to about the 8-body level or so is nowadays quite routine. It is important to note that the Goldstone linked-cluster theorem is explicitly obeyed by the CCM at any level of approximate implementation, and hence results may always be determined directly from the outset in the infinite-lattice limit, $N \to \infty$ (where N is the number of spins in the system). This is in sharp contrast to the QMC results that are always obtained for finite-sized lattices, from which the results for the infinite lattice need to be extrapolated using finitesize scaling arguments. Furthermore, the very important Hellmann-Feynman theorem is also obeyed by the CCM at all levels of approximation. On the other hand, we note that in order to retain all of these useful and important features, it turns out to be necessary to relax the condition that the corresponding bra- and ket-states are manifestly Hermitian conjugates of one another. At a given level of truncation, this Hermiticity property may be only approximately obeyed, although it is certainly restored in the exact limit. As we shall see, a consequence is that we lose the property in the CCM that the results for the ground-state energy form an upper bound to the true results. In practice this lack of manifest Hermiticity poses few actual problems. Indeed, it can often be used as an internal quality check on the accuracy of the method. Finally, we note that the CCM lends itself extremely well for applications on the lattice to the use of computer-algebraic techniques both to derive and to solve the fundamental sets of coupled nonlinear

equations that lie at its heart in practical implementations, via well-defined hierarchies of approximations.

In the rest of this Chapter we will focus attention only on spin-lattice applications of the CCM, for reasons already cited. Nevertheless, we believe that the CBF method still has a worthy future in this field. We hope that others will still develop it further, since it certainly shares many highly desirable features with the CCM. Before concentrating in the rest of this Chapter solely on the CCM, we take a final opportunity to list some of the more important of these features below:

- Both methods are extremely versatile, and they have been extensively tested. There is by now a large amount of experience in using them.
- An impressively wide range of applications to systems of physical interest has been made of one and/or the other method. These include finite nuclei; nuclear matter; quantum field theory (including systems of anharmonic oscillators, ϕ^4 field theory, and pion-nucleon field theory); atoms and molecules of interest in quantum chemistry; the electron gas; quantum hydrodynamics; and the liquids helium (including bulk ³He and ⁴He and their mixtures, and films).
- Both methods are capable of very high accuracy at attainable levels of implementation. In most applications the CBF and/or CCM results are either the best or among the best from all available microscopic techniques. They are now often at the point of being fully competitive with the large-scale QMC simulations in the cases where the latter can be performed.
- Neither method is restricted in principle to particular forms of the Hamiltonian. Both are easily capable of handling very complicated interactions.
- Both the CBF method and the CCM are intrinsically nonperturbative in nature. Some correlations are retained to infinite order, even at the lowest levels of implementation. The CCM, in particular, can often be used to derive (or reconstruct) perturbation theory (PT) series, by a suitable choice of truncation hierarchy for the subsets of terms retained in the multiconfigurational expansions of the ground- or excited-state correlation operators, as described more fully below. In such cases, the CCM provides a natural analytic continuation of the PT series, which in practice is usually found to be valid far outside the radius of convergence of the PT series, and also to be quantitatively superior to such alternative schemes as (generalised) Padé resummations.
- Although nonperturbative in principle, the CCM can be easily related to the Goldstone diagram expansions of time-independent perturbation theory. This feature facilitates comparisons with other methods.
- Similarly, at the optimised Jastrow level implemented via the HNC approximation, the CBF method has been shown [45] to be equivalent to two-body localised parquet theory, and hence to a sum of planar Feynman diagrams of time-dependent perturbation theory.

- In both methods one may work from the outset in the bulk thermodynamic limit, $N \to \infty$, thereby avoiding problems connected with finite-size effects. This is always done in the CCM, although, for technical reasons, it is not always practicable in the CBF case.
- Both methods have the virtue of great flexibility. One may choose "uncorrelated" or "model" ground-state reference states, for example, in many ways. In particular, this presents an opportunity for the "conceptual school" of many-body theory to provide a good starting-point for the "quantitative school." Similarly, many different approximation hierarchies for the correlation operators of the CBF and CCM schemes can be envisaged, and there is again room for external experience or physical intuition to be utilised in their choice.
- Both methods are capable of handling phase transitions. Even when the "uncorrelated" or "model" reference state is a poor choice, both the CBF and CCM schemes have been shown in particular cases to be able to predict phase changes. In the case of the CCM we discuss this in more detail below.
- Both methods, but particularly the CCM, often have the practical capability of implementation to high orders of approximation. The CCM has especially been shown to be very amenable to the use of computer algebra to derive the high-order basic coupled sets of nonlinear equations that underpin it. This feature is particularly marked for lattice systems, and it is a key reason why the CCM is now proving to be fully competitive with large-scale QMC stochastic simulations at a fraction of the computing cost, in those cases where the latter can be performed.

For further details of the CBF method and some of its applications to various quantum lattice systems, the interested reader is referred to the overview in [46], where comparisons are also made with the CCM.

Henceforth we confine our attention to the CCM, whose applications over the last ten or so years to quantum magnetic systems at zero temperature [47–68] have proven to be extremely successful. In particular, the use of computer-algebraic implementations of the CCM for quantum systems of large or infinite numbers of particles has largely been pioneered with respect to these spin-lattice problems. We note too in this context that there have been subsequent applications of these highly accurate computational CCM techniques to other types of lattice quantum systems, such as U(1) and SU(N) lattice gauge field theory [69–71], and the latticised O(N) nonlinear sigma model of relevance to chiral meson field theory [72].

In the remainder of this Chapter we firstly give a brief description of the CCM formalism. We then describe four specific applications of the method to various spin-lattice systems at zero temperature. The first application is to the unfrustrated spin-half XXZ model (or anisotropic Heisenberg model) on the linear chain and on the bipartite square lattice. This simple model serves both to illustrate how the method may be applied in practice and
to indicate the quality of the results attainable at practical levels of implementation. By contrast to this simple model, frustrated systems generally are both more difficult to deal with and have richer phase diagrams, which contain phases of novel forms of order. Three such strongly frustrated systems are then considered. The first of these, the so-called J-J' model, is a spin-half Heisenberg model on the square lattice with two different, competing nearest-neighbour couplings with different bond strengths arranged in a regular zigzag pattern. For the case where the bond strengths have different signs the square plaquettes are thus dynamically frustrated, whereas when the bond strengths have the same sign the model exhibits competition (between magnetic order and dimerisation) without frustration. The third model exhibits geometric frustration, and is again a spin-half Heisenberg model that interpolates smoothly between a triangular lattice and a Kagomé lattice. The last model considered is another model that includes the possibility of dynamical frustration, in which we have both nearest-neighbour and next- nearest-neighbour Heisenberg interactions with unequal strengths. Furthermore, the model is taken to represent a spin-half/spin-one ferrimagnet in which one sublattice of the bipartite square lattice is populated entirely with spin-one spins, while the other sublattice is populated entirely with spin-half spins. The Chapter is concluded with a discussion of the implications of these illustrative results for further work, and with some ideas for future extensions and applications of the CCM.

7.2 The CCM Formalism

A brief description of the normal coupled cluster method (NCCM) formalism is now provided, although the interested reader is referred to [16–24, 47–68]. for further details. The exact ket and bra ground-state energy eigenvectors, $|\Psi\rangle$ and $\langle \tilde{\Psi}|$, of a general many-body system described by a Hamiltonian H,

$$|H|\Psi\rangle = E_g|\Psi\rangle; \quad \langle \Psi|H = E_g\langle \Psi|, \qquad (7.1)$$

are parametrised within the single-reference CCM as follows:

$$|\Psi\rangle = e^{S}|\Phi\rangle \quad ; \quad S = \sum_{I \neq 0} S_{I}C_{I}^{+} ,$$

$$\langle \tilde{\Psi}| = \langle \Phi|\tilde{S}e^{-S} \quad ; \quad \tilde{S} = 1 + \sum_{I \neq 0} \tilde{S}_{I}C_{I}^{-} . \tag{7.2}$$

The single model or reference state $|\Phi\rangle$ is required to have the property of being a cyclic vector with respect to two well-defined Abelian subalgebras of *multi-configurational* creation operators $\{C_I^+\}$ and their Hermitian-adjoint destruction counterparts $\{C_I^- \equiv (C_I^+)^{\dagger}\}$. Thus, $|\Phi\rangle$ plays the role of a vacuum state with respect to a suitable set of (mutually commuting) many-body creation operators $\{C_I^+\}$. Note that $C_I^- |\Phi\rangle = 0$, $\forall I \neq 0$, and that $C_0^- \equiv 1$, the identity operator. These operators are furthermore complete in the manybody Hilbert (or Fock) space. Also, the correlation operator S is decomposed entirely in terms of these creation operators $\{C_I^+\}$, which, when acting on the model state $(\{C_I^+|\Phi\rangle\})$, create excitations from it. We note that although the manifest Hermiticity, $(\langle \tilde{\Psi}|^{\dagger} = |\Psi\rangle / \langle \Psi|\Psi\rangle)$, is lost, the normalisation conditions $\langle \tilde{\Psi}|\Psi\rangle = \langle \Phi|\Psi\rangle = \langle \Phi|\Phi\rangle \equiv 1$ are explicitly imposed. The correlation coefficients $\{S_I, \tilde{S}_I\}$ are regarded as being independent variables, and the full set $\{S_I, \tilde{S}_I\}$ thus provides a complete description of the ground state. For instance, an arbitrary operator A will have a ground-state expectation value given as,

$$\bar{A} \equiv \langle \tilde{\Psi} | A | \Psi \rangle = \langle \Phi | \tilde{S} e^{-S} A e^{S} | \Phi \rangle = \bar{A} \left(\{ \mathcal{S}_{I}, \tilde{\mathcal{S}}_{I} \} \right) .$$
(7.3)

We note that the exponentiated form of the ground-state CCM parametrisation of (7.2) ensures the correct counting of the *independent* and excited correlated many-body clusters with respect to $|\Phi\rangle$ which are present in the exact ground state $|\Psi\rangle$. It also ensures the exact incorporation of the Goldstone linked-cluster theorem, which itself guarantees the size-extensivity of all relevant extensive physical quantities. We also note that any operator in a similarity transform may be written as

$$\tilde{A} \equiv e^{-S} A e^{S} = A + [A, S] + \frac{1}{2!} [[A, S], S] + \cdots$$
 (7.4)

The determination of the correlation coefficients $\{S_I, \tilde{S}_I\}$ is achieved by taking appropriate projections onto the ground-state Schrödinger equations of (7.1). Equivalently, they may be determined variationally by requiring the ground-state energy expectation functional $\bar{H}(\{S_I, \tilde{S}_I\})$, defined as in (7.3), to be stationary with respect to variations in each of the (independent) variables of the full set. We thereby easily derive the following coupled set of equations,

$$\delta \bar{H} / \delta \tilde{\mathcal{S}}_I = 0 \Rightarrow \langle \Phi | C_I^- e^{-S} H e^S | \Phi \rangle = 0, \ \forall \ I \neq 0 \ ; \tag{7.5}$$

$$\delta \bar{H}/\delta \mathcal{S}_I = 0 \Rightarrow \langle \Phi | \tilde{S} e^{-S} [H, C_I^+] e^S | \Phi \rangle = 0, \ \forall \ I \neq 0 .$$
(7.6)

Equation (7.5) also shows that the ground-state energy at the stationary point has the simple form

$$E_g = E_g(\{\mathcal{S}_I\}) = \langle \Phi | \mathrm{e}^{-S} H \mathrm{e}^{S} | \Phi \rangle \quad . \tag{7.7}$$

It is important to realize that this (bi-)variational formulation does *not* lead to an upper bound for E_g when the summations for S and \tilde{S} in (7.2) are truncated, due to the lack of exact Hermiticity when such approximations are made. However, one can prove that the important Hellmann-Feynman theorem *is* preserved in all such approximations. We note that (7.5) represents a coupled set of non-linear multinomial equations for the *c*-number correlation coefficients $\{S_I\}$. The nested commutator expansion of the similarity-transformed Hamiltonian

$$\tilde{H} \equiv e^{-S} H e^{S} = H + [H, S] + \frac{1}{2!} [[H, S], S] + \cdots$$
(7.8)

and the fact that all of the individual components of S in the sum in (7.2) commute with one another, together imply that each element of S in (7.2) is linked directly to the Hamiltonian in each of the terms in (7.8). Thus, each of the coupled equations (7.5) is of *linked cluster* type. Furthermore, each of these equations is of finite length when expanded, since the otherwise infinite series of (7.8) will always terminate at a finite order, provided only (as is usually the case) that each term in the second-quantised form of the Hamiltonian, H, contains a finite number of single-body destruction operators, defined with respect to the reference (vacuum) state $|\Phi\rangle$. Hence, the CCM parametrisation naturally leads to a workable scheme which can be efficiently implemented computationally. It is important to note that at the heart of the CCM lies a similarity transformation, in contrast with the unitary transformation in a standard variational formulation in which the bra state $\langle \tilde{\Psi} |$ is simply taken as the explicit Hermitian conjugate of $|\Psi\rangle$.

In the case of spin-lattice problems of the type considered here, the operators C_I^+ become products of spin-raising operators s_k^+ over a set of sites $\{k\}$, with respect to a model state $|\Phi\rangle$ in which all spins points "downward" in some suitably chosen local spin axes. The CCM formalism is exact in the limit of inclusion of all possible such multi-spin cluster correlations for S and \tilde{S} , although in any real application this is usually impossible to achieve. It is therefore necessary to utilise various approximation schemes within S and \tilde{S} . The three most commonly employed schemes previously utilised have been: (1) the SUB*n* scheme, in which all correlations involving only *n* or fewer spins are retained, but no further restriction is made concerning their spatial separation on the lattice; (2) the SUB*n*-*m* sub-approximation, in which all SUB*n* correlations spanning a range of no more than *m* adjacent lattice sites are retained; and (3) the localised LSUB*m* scheme, in which all multi-spin correlations over all distinct locales on the lattice defined by *m* or fewer contiguous sites are retained.

An excited-state wave function, $|\Psi_e\rangle$, is determined by linearly applying an excitation operator X^e to the ket-state wave function of (7.2), such that

$$|\Psi_e\rangle = X^e \ e^S |\Phi\rangle \quad . \tag{7.9}$$

This equation may now be used to determine the low-lying excitation energies, such that the Schrödinger equation, $H|\Psi_e\rangle = E_e|\Psi_e\rangle$, may be combined with its ground-state counterpart of (7.1) to give the result,

$$\epsilon_e X^e |\Phi\rangle = e^{-S} [H, X^e] e^S |\Phi\rangle \quad , \tag{7.10}$$

where $\epsilon_e \equiv E_e - E_g$ is the excitation energy. By analogy with the ground-state formalism, the excited-state correlation operator is written as,

$$X^e = \sum_{I \neq 0} \mathcal{X}_I^e C_I^+ \quad , \tag{7.11}$$

where the set $\{C_I^+\}$ of multi-spin creation operators may differ from those used in the ground-state parametrisation in (7.2) if the excited state has different quantum numbers than the ground state. We note that (7.11) implies the overlap relation $\langle \Phi | \Psi_e \rangle = 0$. By applying $\langle \Phi | C_I^-$ to (7.10) we find that,

$$\epsilon_e \mathcal{X}_I^e = \langle \Phi | C_I^- e^{-S} [H, X^e] e^S | \Phi \rangle \quad , \forall \ I \neq 0 \quad ,$$
(7.12)

which is a generalised set of eigenvalue equations with eigenvalues ϵ_e and corresponding eigenvectors \mathcal{X}_I^e , for each of the excited states which satisfy $\langle \Phi | \Psi_e \rangle = 0$.

We note that lower orders of approximation may be determined analytically and an example of applying the LSUB2 and SUB2 approximations to the spin-half linear chain XXZ model is given later in order to show clearly how this is performed. However, it rapidly becomes clear that analytical determination of the CCM equations for higher orders of approximation is impractical. We therefore employ computer algebraic techniques in order efficiently to determine and solve the CCM ket- and bra-state equations. A full exposition of this topic is beyond the scope of this chapter, although we note that the problem essentially becomes one of pattern matching in order to determine the CCM ground-state ket equations. The bra-state equations may be determined easily thereafter and the ket- and bra-state equations are readily solved using standard techniques for the solution of coupled polynomial equations (e.g., the Newton-Raphson method). The excited-state eigenvalue equations may be also determined in an analogous manner, and, although this is not strictly necessary, we restrict the level of approximation to the same for the excited state as for the ground state in calculations presented here. A full exposition of the details in applying the CCM to high orders of approximation is given for the ground state in [54, 59, 67] and for excited states in [62].

Note that the results of SUB*m*-*m* and LSUB*m* approximation schemes may be extrapolated to the exact limit, $m \to \infty$, using various "heuristic' approaches. How to do this is not discussed here, although the interested reader is referred to [59,67] for more details.

7.3 The XXZ Model

We wish to apply the CCM to the spin-half XXZ model on the linear chain and the square lattice in order to illustrate how one applies the CCM to a practical problem and also to demonstrate the accuracy and power of the method. We note that these systems are unfrustrated and, in global spin coordinates, the XXZ Hamiltonian is specified as follows,

$$H = \sum_{\langle i,j \rangle} [s_i^x s_j^x + s_i^y s_j^y + \Delta s_i^z s_j^z] \quad , \tag{7.13}$$

where the sum on $\langle i, j \rangle$ counts all nearest-neighbour pairs once. The Néel state is the ground state in the trivial Ising limit $\Delta \to \infty$, and a phase transition occurs at (or near to) $\Delta = 1$. Indeed, the ground state demonstrates Néellike order in the z-direction for $\Delta > 1$ and a similar x-y planar phase for $-1 < \Delta < 1$. The system is ferromagnetic for $\Delta < -1$.

7.3.1 The CCM Applied to the XXZ Model Using a z-Aligned Néel Model State

We turn now to the choice of $|\Phi\rangle$ and the operators $\{C_I^+\}$ for the case of spin-half quantum antiferromagnets on bipartite lattices, in regimes where the corresponding classical limit is described by a Néel-like order in which all spins on each sublattice are separately aligned in some global spin axes. It is then convenient to introduce a different local quantisation axis and different spin coordinates on each sublattice, by a suitable rotation in spin space, so that the corresponding reference state becomes a fully aligned ("ferromagnetic") state, with all spins pointing along, say, the negative z-axis in the corresponding local axes. Such rotations are canonical tranformations that leave the spin commutation relations unchanged. In the same local axes, the configuration indices $I \to \{k_1, k_2, \dots, k_M\}$, a set of site indices, such that $C_I^+ \to s_{k_1}^+ s_{k_2}^+ \cdot \cdots s_{k_M}^+$, where $s_k^{\pm} \equiv s_k^x \pm i s_k^y$ are the usual spin-raising and spin-lowering operators at site k.

For the Hamiltonian of (7.13) we first choose the z-aligned Néel state as our reference state (which is the exact ground state for $\Delta \to \infty$, and is expected to be a good starting point for all $\Delta > 1$, down to the expected phase transition at $\Delta = 1$). We then perform a rotation of the up-pointing spins by 180° about the y-axis, such that $s^x \to -s^x$, $s^y \to s^y$, $s^z \to -s^z$ on this sublattice. The Hamiltonian of (7.13) may thus be written in these local coordinates as,

$$H = -\frac{1}{2} \sum_{\langle i,j \rangle} [s_i^+ s_j^+ + s_i^- s_j^- + 2\Delta s_i^z s_j^z] \quad .$$
 (7.14)

The results presented below are based on the SUB2 approximation scheme and the localised LSUB*m* scheme, in which we include all multispin correlations over all possible distinct locales (or "lattice animals") on the lattice defined by *m* or fewer contiguous sites. We include all *fundamental configurations*, $I \to \{k_1, k_2, \dots, k_n\}$, with $n \leq m$, which are distinct under the point and space group symmetries of both the lattice and the Hamiltonian. The numbers, N_F and N_{F_e} , of such fundamental configurations for the ground and excited states, respectively, may be further restricted by the use of additional conservation laws. For example, the Hamiltonian of (7.13) commutes with the total uniform magnetisation, $s_T^z = \sum_k s_k^z$, where the sum on k runs over all lattice sites. The ground state is known to lie in the $s_T^z = 0$ subspace, and hence we exclude configurations with an odd number of spins or with unequal numbers of spins on the two equivalent sublattices. Similarly for the excited states, since we are only interested in the lowest-lying excitation, we restrict the choice of configurations to those with $s_T^z = \pm 1$.

7.3.2 The LSUB2 Approximation for the Spin-Half, Linear-Chain XXZ Model

We start the LSUB2 calculation by specifying the commutation relations $[s_l^{\pm}, s_k^z] = \mp s_k^{\pm} \delta_{l,k}$ and $[s_l^{+}, s_k^{-}] = 2s_k^z \delta_{l,k}$. We again note that the similarity transform may be expanded as a series of nested commutators in (7.4). We write the LSUB2 ket-state operator in the following simple form for the spin-half linear chain model,

$$S = b_1 \sum_{i}^{N} s_i^+ s_{i+1}^+ \quad , \tag{7.15}$$

where i runs over all sites on the linear chain and b_1 is the sole ket-state correlation coefficient. In this approximation we may therefore determine similarity transformed versions of the spin operators, given by

$$\tilde{s}_{l}^{+} = s_{l}^{+}
\tilde{s}_{l}^{z} = s_{l}^{z} + b_{1}(s_{l}^{+}s_{l+1}^{+} + s_{l-1}^{+}s_{l}^{+})
\tilde{s}_{l}^{-} = s_{l}^{-} - 2b_{1}(s_{l}^{z}s_{l+1}^{+} + s_{l-1}^{+}s_{l}^{z}) - 2b_{1}^{2}s_{l-1}^{+}s_{l}^{+}s_{l+1}^{+}$$
(7.16)

We note that the otherwise infinite-series of operators in the expansion of the similarity transform terminates to finite order. We also note that $(s_l^+)^2 |\Phi\rangle = 0$ for any lattice site (which is true only for spin-half systems), and this is implicitly assumed in the last of (7.16). Clearly we may also write the similarity transformed version of the Hamiltonian as

$$\tilde{H} = -\frac{1}{2} \sum_{\langle i,j \rangle} [\tilde{s}_i^+ \tilde{s}_j^+ + \tilde{s}_i^- \tilde{s}_j^- + 2\Delta \tilde{s}_i^z \tilde{s}_j^z] \quad .$$
(7.17)

We may now substitute the expressions for the spin operators in (7.16) into the above expression. The ground-state energy is given by

$$\frac{E_g}{N} = -\frac{1}{4} \{ \Delta + 2b_1 \} \quad . \tag{7.18}$$

We note that our expression for the ground-state energy is size-extensive (i.e., it scales linearly with N), as required by the Goldstone theorem which is obeyed by the NCCM. Furthermore, this expression terminates to finite order, as for the similarity transformed versions of spin operators. Finally, we note that *any* other non-trivial choice for S will always yield this expression for the ground-state energy. The task is now to find b_1 and we note that if we could include all possible spin correlations in S then we would obtain an exact result for the ground-state energy. However, this is found to be impossible to achieve for most cases in practice, and we make an approximation (such as the LSUB2 approximation presented here). The LSUB2 ket-state equation is given by

$$3b_1^2 + 2\Delta b_1 - 1 = 0 \quad , \tag{7.19}$$

which therefore implies that the LSUB2 ground-state energy may be written explicitly in terms of Δ as,

$$\frac{E_g}{N} = -\frac{\Delta}{12} - \frac{1}{6}\sqrt{\Delta^2 + 3} \quad . \tag{7.20}$$

We note that this expression gives the correct result in the Ising limit $\Delta \to \infty$. We again note that the bra state does not manifestly have to be the Hermitian conjugate of the ket state, and we note that the bra-state correlation operator for the LSUB2 approximation is given by,

$$\tilde{S} = 1 + \tilde{b}_1 \sum_{j=1}^{N} s_j^- s_{j+1}^- \quad , \tag{7.21}$$

where the index j runs over all sites on the linear chain and \tilde{b}_1 is the sole bra-state correlation coefficient in the LSUB2 approximation. In order to determine the bra-state equation, we now explicitly determine $\bar{H}/(\{S_I, \tilde{S}_I\})$,

$$\bar{H} = -\frac{N}{4}(\Delta + 2b_1) + N\tilde{b}_1\left(-\frac{1}{2} + \Delta b_1 + \frac{3}{2}b_1^2\right) \quad , \tag{7.22}$$

such that LSUB2 bra-state equation is given from $\partial \bar{H} \partial b_1 = 0$ as

$$-\frac{1}{2} + \Delta \tilde{b}_1 + 3b_1 \tilde{b}_1 = 0 \quad , \tag{7.23}$$

which gives $\tilde{b}_1 = \frac{1}{2}(\Delta^2 + 3)^{-1/2}$. Finally, we note that once the values for the bra- and ket-state correlation coefficients have been determined (at a given level of approximation) then we may also obtain the values for expectation values, such as the sublattice magnetisation given by

$$M \equiv -\frac{2}{N} \langle \tilde{\Psi} | \sum_{i}^{N} s_{i}^{z} | \Psi \rangle = -\frac{2}{N} \langle \Phi | \tilde{S} e^{-S} (\sum_{i}^{N} s_{i}^{z}) e^{S} | \Phi \rangle \quad .$$
(7.24)

The sublattice magnetisation is written here in terms of the "rotated" spin coordinates. We note that this is given by

$$M_{\rm LSUB2} = 1 - 4b_1 \tilde{b}_1 \quad ,$$

= $\frac{1}{3} \left[1 + \frac{2\Delta}{\sqrt{\Delta^2 + 3}} \right]$ (7.25)

for the LSUB2 approximation.

7.3.3 The SUB2 Approximation for the Spin-Half, Linear-Chain XXZ Model

The SUB2 approximation allows us to include all possible two-spin correlations in our wave function. We note that the SUB2 ket-state operator is given by

$$S = 1/2 \sum_{i}^{N} \sum_{r} b_{r} s_{i}^{+} s_{i+r}^{+} \quad , \qquad (7.26)$$

and that the index i runs over all sites on the linear chain. Furthermore, the index r runs over all lattice vectors which connect one sublattice to the other and b_r is its corresponding SUB2 ket-state correlation coefficient for this vector. We again determine a similarity transformed version of the spin operators and we are able to determine the SUB2 equations, given by

$$\sum_{\rho} \left\{ (1 + 2\Delta b_1 + 2b_1^2) \delta_{\rho,r} - 2(\Delta + 2b_1) b_r + \sum_s b_{r+s+\rho} b_s \right\} = 0 \quad , \qquad (7.27)$$

where ρ runs over all (1D) nearest-neighbour lattice vectors. Equation (7.27) may now be solved by employing a sublattice Fourier transform, given by

$$\Gamma(q) = \sum_{r} e^{\mathbf{i}rq} b_r \quad , \tag{7.28}$$

where r again is a lattice vector (i.e., an odd integer number in 1D) which connects the different sublattices. This expression has an inverse given by

$$b_r = \int_0^\pi \frac{dq}{\pi} \cos(rq) \Gamma(q) \quad . \tag{7.29}$$

The SUB2 equations (7.27) and (7.28) therefore lead to an expression for $\Gamma(q)$ given by

$$\Gamma(q) = \frac{K}{\cos(q)} [1 \pm \sqrt{1 - k^2 \cos^2(q)}] \quad , \tag{7.30}$$

where $K = \Delta + 2b_1$ and $k^2 = (1 + 2\Delta b_1 + 2b_1^2)/K^2$. (Note that we choose the negative solution in (7.30) in order to reproduce results in the trivial limit

 $\Delta \to \infty$.) These equations now yield a self-consistency requirement on the variable b_1 and they may be solved iteratively at a given value of Δ . Indeed, we know that all correlation coefficients must tend to zero (namely, for SUB2: $b_r \to 0, \forall r$) as $\Delta \to \infty$ and we *track* this solution for large Δ by reducing Δ in small successive steps. We find that the discriminant in (7.30) becomes negative at a *critical points*, $\Delta_c \approx 0.3728$. Furthermore, the behaviour of b_r changes from exponential to algebraic decay with respect to r at Δ_c . These are strong indications that the CCM critical point is detecting the known quantum phase transition in the system at $\Delta = 1$. Furthermore, the SUB2 approximation for the ground state may be used in conjunction with a SUB1 approximation for the excited state operator X^e in (7.11) in order to determine the excitation spectrum. We note that the excitation spectrum becomes soft at the critical point, Δ_c . This is further evidence for a phase transition and the interested reader is referred to [48] for more details.

We may also solve the SUB2-*m* equations directly using computational techniques. Indeed, we study the limit points of these approximations by using solution-tracking software (PITCON), which allows one to solve coupled non-linear equations. We again track our solution from the limit $\Delta \to \infty$ down to and beyond the limit point and Fig. 7.1 shows our results. In particular, we note that we have two distinct branches, although only the upper branch is a "physical" solution. We again note that the CCM does not necessarily always provide an upper bound on the ground-state energy – although this is often the case for the physical solution! By tracking from a point at which we are sure of, the solution we guarantee that our solution is valid, and this approach is also used for LSUB*m* approximations.

We find that the two branches collapse onto the same line, namely, that of the full SUB2 solution, as we increase the level of SUB2-*m* approximation with respect to *m*. Indeed, we may plot the positions of the SUB2-*m* limit points against $1/m^2$ and we note that these data points are found to be both highly linear and they tend to the critical value, Δ_c , for the full SUB2 equations in the limit $m \to \infty$. Again, we note that the LSUB*m* and SUB*m*-*m* approximations also show similar branches (namely, one "physical" and one "unphysical" branch) which appear to converge as one increases the magnitude of the truncation index, *m*. This is a strong indication that our LSUB*m* and SUB*m*-*m* critical points are also reflections of phase transitions in the real system and that our extrapolated LSUB*m* and SUB*m*-*m* results should tend to the exact solution.

7.3.4 CCM Results for the Spin-Half Square-Lattice XXZ Model Using a z-Aligned Model State

We shall now illustrate the power and accuracy of the CCM by presenting results in Figs. 7.2–7.4 respectively for the energy per spin (E_g/N) and the sublattice magnetisation (M) for the ground state, and the energy gap (ϵ_e)



Fig. 7.1. CCM SUB2-*m* and full SUB2 results for the ground-state energy of the spin-half linear-chain XXZ model compared to exact results of the Bethe Ansatz [34–37]. The CCM model state is the z-aligned Néel state. SUB2-*m* limit points converge to the SUB2 limit at $\Delta_c = 0.3728$, at which point the solution to the full SUB2 equations becomes complex, as $m \to \infty$, and these are reflections of the infinite-order phase transition at $\Delta = 1$ in the 'true' system. Note that the upper branch of the SUB2-*m* results are physical and the lower branch is unphysical

of the lowest-lying excited state for the spin-half $X\!X\!Z$ model on the square lattice.

We find that for all LSUB*m* approximations with m > 2 the physical branch of ground-state solutions (i.e., the one which becomes exact in the $\Delta \to \infty$ limit) terminates at a critical value Δ_c , such that for $\Delta < \Delta_c$ no real solution exists. These LSUB*m* "critical points" are analogous to the SUB2-*m* limit points of the previous subsection, and they are again taken to be a signal of the phase transition at (or near to) $\Delta = 1$. (Note that the "unphysical" LSUB*m* branches, as seen for the SUB2-*m* approximations above, are not plotted here in order to present a clear illustration of our results, although they certainly exist.)

The SUB2 and LSUB*m* results using the *z*-aligned state as model state are compared in Table 7.1 for the isotropic ($\Delta = 1$) case with results from linear spin-wave theory (LSWT) [74], series expansion techniques [75], and quantum Monte Carlo (QMC) simulations [76]. Figures 7.2 and 7.4 show the corresponding results for the ground-state energy and lowest-lying excitation energy ϵ_e as functions of Δ . Our results for the ground- and excited-state properties of the *XXZ* systems are seen to be in excellent agreement with those results of the best of other approaches. We also note that values for the spin stiffness of the Heisenberg model (see [77] and later on in this Chapter for further details) are also found to be very accurate. Furthermore, calculations have been carried out for this model using an extended version of the CCM in which the bra-state correlation operator \tilde{S} is also written in an exponentiated form, analogous to that of the ket-state operator. The interested reader is referred to [61] for more details.

7.3.5 CCM Results for the Spin-Half Square-Lattice XXZ Model Using a Planar Model State

There is never a unique choice of model state $|\Phi\rangle$. Indeed, our choice should be guided by any physical insight available to us concerning the system or, more specifically, that particular phase of it which is under consideration. In the absence of any other insight into the quantum many-body system, we may sometimes be guided by the behaviour of the corresponding classical system. The XXZ model under consideration provides just such an illustrative example. Thus, for $\Delta > 1$ the classical Hamiltonian of (7.13) on the 2D square lattice (and, indeed, on any bipartite lattice) is minimized by a perfectly antiferromagnetically Néel-ordered state in the z-direction, and we have already utilised this information in the preceding subsections. However, the classical ground-state energy is minimized by a Néel-ordered state with spins pointing



Fig. 7.2. CCM LSUB*m* results using the *z*-aligned and planar Néel model states for the ground-state energy of the spin-half square-lattice *XXZ* model compared to quantum Monte Carlo results of [73]. Results for the LSUB6 approximation using both model states end at their respective *critical* points



Fig. 7.3. CCM LSUB*m* results using the *z*-aligned and planar Néel model states for the sublattice magnetisation of the spin-half square-lattice XXZ model. Results for the LSUB4 and LSUB6 approximation using both model states end at their respective *critical* points



Fig. 7.4. CCM LSUB*m* results using the *z*-aligned Néel state as model state for the lowest-lying excitation energies of the spin-half square-lattice *XXZ* model

Table 7.1. CCM results [59, 62] for the isotropic ($\Delta = 1$) spin-half square-lattice Heisenberg antiferromagnet compared to results of other methods. The numbers of fundamental configurations in the ground-state and excited-state CCM wave functions for the z-aligned Néel model state are given by N_f^z and $N_{f_e}^z$, respectively, and the number of fundamental configurations in the ground-state CCM wave function for the planar Néel model state is given by N_f^p . Results for the critical points of the z-aligned Néel model state are indicated by Δ_c^z and results for the critical points of the planar Néel model state are indicated by Δ_c^p . (Note that results for the ground-state expectation values for both model states are identical for the isotropic Heisenberg model at $\Delta = 1$)

Method	E_g/N	M	ϵ_e	N_f^z	$N_{f_e}^z$	N_f^p	Δ_c^z	Δ_c^p
LSUB2	-0.64833	0.841	1.407	1	1	1	_	_
SUB2	-0.65083	0.827	1.178	-	-	_	0.799	1.204
LSUB4	-0.66366	0.765	0.852	7	6	10	0.577	1.648
LSUB6	-0.66700	0.727	0.610	75	91	131	0.763	1.286
LSUB8	-0.66817	0.705	0.473	1273	2011	2793	0.843	_
Extrapolated CCM	-0.6697	0.62	0.00	-	-	-	1.03	-
LSWT [74]	-0.658	0.606	0.0	-	-	-	1.0	-
Series Expansions [75]	-0.6693(1)	0.614(2)	-	-	-	-	-	-
QMC [76]	-0.669437(5)	0.6140(6)	-	-	-	_	-	-

along any direction in the xy plane, say along the x-axis for $-1 < \Delta < 1$. Thus, in order to provide CCM results in the region $-1 < \Delta < 1$, we now take this state to be our model state and we shall refer to it as the "planar" model state.

In order to produce another "ferromagnetic" model state for the planar model state in the local frames, we rotate the axes of the left-pointing spins (i.e., those pointing in the negative x-direction) in the planar state by 90° about the y-axis, and the axes of the corresponding right-pointing spins by -90° about the y-axis. (Note that the positive z-axis is defined here to point upwards and the positive x-axis is defined to point rightwards.) Thus, the transformations of the local axes are described by

$$s^x \to s^z$$
 , $s^y \to s^y$, $s^z \to -s^x$ (7.31)

for the left-pointing spins, and by

$$s^x \to -s^z$$
, $s^y \to s^y$, $s^z \to s^x$ (7.32)

for the right-pointing spins. The transformed Hamiltonian of (7.13) may now be written in these local axes as

$$H = -\frac{1}{4} \sum_{\langle i,j \rangle} \left[(\Delta + 1)(s_i^+ s_j^+ + s_i^- s_j^-) + (\Delta - 1)(s_i^+ s_j^- + s_i^- s_j^+) + 4s_i^z s_j^z \right] \quad ,$$
(7.33)

In this case we track the CCM solution for the planar model state from the point $\Delta = -1$. We note that all of the CCM correlation coefficients are zero at $\Delta = -1$ because the model state is an exact ground eigenstate of the Hamiltonian of (7.33) at this point. The results for the ground-state energy using the planar model state are plotted in Fig. 7.2, and the corresponding results for the sublattice magnetisation (M, again defined with respect to the rotated local spin axes) are shown in Fig. 7.3. Furthermore, we note that the Hamiltonian for the planar model state of (7.33) is identical to the Hamiltonian for the z-aligned model state of (7.17) at $\Delta = 1$. Indeed, we obtain identical results for the ground-state expectation values at $\Delta = 1$, and this is an excellent test of the validity of our results.

7.3.6 Quantum Criticality of the Antiferromagnetic Phase Transition for the Spin-Half Square-Lattice XXZ Model

We wish to investigate the quantum criticality of the phase at (or near to) $\Delta = 1$ for the case of the square lattice. The critical index for the singular (non-analytic) term in E_g/N near an LSUB*m* critical point $\Delta_c(m)$ can first be obtained, for example, by direct examination of the anisotropy susceptibility, $\chi_a \equiv -\partial^2 (E_g/N)/\partial \Delta^2$. For m > 2 we find,

$$\chi_a^m(\Delta) \longrightarrow \bar{\chi}_a^m |\Delta - \Delta_c(m)|^{-\alpha_0} ; \quad \Delta \to \Delta_c(m) .$$
 (7.34)

Direct calculation for the LSUB*m* approximations using both the *z*-aligned and planar Néel model states shows that for m > 2 we have $\alpha_0 \approx 1.500 \pm 0.005$. However, the prefactors $\bar{\chi}_a^m$ in (7.34) are themselves strongly dependent on the truncation index *m*. We may now use a variant of the so-called coherent anomaly method (CAM) of Suzuki [78] to extract further information. Thus, we attempt to fit $\bar{\chi}_a^m$ with the coherent anomaly form,

$$\bar{\chi}_a^m \longrightarrow K |\Delta_c(\infty) - \Delta_c(m)|^{\nu} \quad ; \quad \Delta \to \Delta_c(\infty) \quad , \tag{7.35}$$

where K is a constant. Thus, as explained by Suzuki [78], one may intuit or prove that the exact $\chi_a(\Delta)$ has the critical form,

$$\chi_a(\Delta) \longrightarrow \kappa |\Delta - \Delta_c(\infty)|^{-\alpha_0 + \nu} ; \quad \Delta \to \Delta_c(\infty) \equiv \Delta_c , \qquad (7.36)$$

where κ is a constant.

A CAM analysis along these lines of the LSUB*m* results based on the *z*-aligned Néel state gives $\nu \approx 1.25$ using the $\Delta_A^z(4)$ and $\Delta_A^z(6)$ data, and $\nu \approx 0.97$ using the $\Delta_A^z(6)$ and $\Delta_A^z(8)$ data. We thus obtain a singular term in E_g/N near Δ_A^z with a critical exponent $2 - \alpha_0 + \nu \approx 1.50 - 1.75$. This may be compared with the corresponding value of 3/2 for both the mean-field-like CCM SUB2 approximation (in which all 2-spin-flip correlation terms are retained, however far apart on the lattice) and linear spin-wave theory (LSWT). A similar treatment for the planar model state yields a critical exponent of $2 - \alpha_0 + \nu \approx 1.77$, which is in good agreement with the result for the *z*-aligned Néel model state.



Fig. 7.5. Results for the Ising-expansion coefficients, plotted as a function of the lattice distance R, corresponding to two-body excitations with respect to the model state for the spin-half, square lattice Heisenberg model at $\Delta = 1$ obtained via the LSUB*m* approximation scheme (with $m = \{4, 6, 8\}$) and the SUB2 approximation. (Figure taken from [32])

7.3.7 CCM Prediction of the Nodal Surface of the Spin-Half Square-Lattice Heisenberg Model

We consider an expansion of the ground-state wave function in a complete Ising basis $\{|I\rangle\}$ (in terms of the *local* coordinates after rotation). This may be again written as, $|\Psi\rangle = \sum_{I} \Psi_{I} |I\rangle$, where the sums over I goes over all 2^{N} Ising states, and we find that this expression naturally leads from (7.2) (also see [32,60]) to an exact mapping of the CCM correlation coefficients $\{S_{I}\}$ to the Ising-expansion coefficients $\{\Psi_{I}\}$, which is given by

$$\Psi_I = \langle \Phi | C_I^- e^S | \Phi \rangle \equiv \langle \Phi | s_{i_1}^- s_{i_2}^- \cdots s_{i_l}^- e^S | \Phi \rangle .$$
(7.37)

It is possible to match the terms in the exponential to the 'target' configuration of C_I^- in (7.37), and so obtain a numerical value for the $\{\Psi_I\}$ coefficients once the CCM ket-state equations have been derived and solved for a given value of the anisotropy. Note that we may plot the Ising-expansion coefficients as a function of the lattice distance R, corresponding to two-body excitations with respect to the model state, and results are shown in Fig. 7.5.

We observe that all of the coefficients are found to be positive, and this shows that the exact Marshall-Peierls sign rule is being obeyed for our *ab initio* calculation. We note that no such condition is imposed in our CCM treatment of this model. Indeed, it is also the case that all other four- or higher-body terms have corresponding Ising expansion terms which are positive. We also note that the Ising expansion coefficients appear to converge rapidly with increasing levels of approximation, and that a strength of the CCM is that it may be applied to even very strongly frustrated systems where no analogues of the Marshall-Peierls sign rule are usually known.

We note that it might be possible to use the CCM in order to simulate accurately the nodal surface of quantum problem and this information might be fed into a fixed-node QMC calculation in order to simulate very accurately the properties of this system. Indeed, general rules might be inferred from the CCM data and, if so, an exact solution, to within QMC statistical limits, might be determined. The interested reader is referred to [31, 32, 60] for more information.

7.4 The J-J' Model: A Square-Lattice Model with Competing Nearest-Neighbour Bonds

We now wish to show that the CCM can treat frustrated systems as easily as unfrustrated systems, and we begin by noting that the J-J' model is a spinhalf Heisenberg model on a square lattice with two kinds of nearest-neighbour bonds J and J', as shown in Fig. 7.6,

$$H = J \sum_{\langle ij \rangle_1} \mathbf{s}_i \cdot \mathbf{s}_j + J' \sum_{\langle ij \rangle_2} \mathbf{s}_i \cdot \mathbf{s}_j.$$
(7.38)

The sums over $\langle ij \rangle_1$, and $\langle ij \rangle_2$ represent sums over the nearest-neighbour bonds shown in Fig. 7.6 by dashed and solid lines respectively. Each square-



Fig. 7.6. Illustration of the J-J' model of (7.38), with two kinds of regularly distributed nearest-neighbour exchange bonds, J (dashed lines) and J' (solid lines) and its classical spiral state ($\Phi > 0$) shown for the ferromagnetic case (J < 0, J' > |J|/3). (Figure taken from [77])

lattice plaquette consists of three J bonds and one J' bond. A model with such a zigzag pattern has been treated by various methods [64,77,79–82]. For the cases in which J and J' have *different* signs (i.e., one bond is ferromagnetic while the other is antiferromagnetic) the plaquettes are frustrated, whereas competition without frustration is realized for antiferromagnetic bonds (J' > 0 and J > 0).

Using this model we discuss the influence of quantum fluctuations on the ground-state phase diagram and in particular on the nature of the zerotemperature phase transitions from phases with collinear magnetic order at small frustration to phases with noncollinear spiral order at large frustration. The role of quantum fluctuations is examined by comparing ferromagneticspiral and antiferromagnetic-spiral transitions within the same model. Whereas for the classical version of the J-J' model both situations can be mapped onto each other, the quantum model behaves differently in the two cases and this is because of the different nature of the collinear state. The quantum Néel state on two-dimensional lattices exhibits strong quantum fluctuations. For example, as we saw in the previous section, the sublattice magnetisation of the Heisenberg antiferromagnet (HAF) on the square lattice is only about 60% of its classical value. By contrast, the ferromagnetic state is the same for the quantum and the classical model and there are no quantum fluctuations in this state.

The classical ground state of this J-J' model is collinear (i.e., ferromagnetic or antiferromagnetic depending on the sign of J) for the unfrustrated cases. For |J'| > |J|/3 (and J and J' having different signs) the frustration is large enough in order to force the ground state to be a noncollinear state of spiral nature with a characteristic pitch angle $\Phi = \pm |\Phi_{cl}|$ given by

$$|\Phi_{\rm cl}| = \begin{cases} 0 & |J'| < \frac{|J|}{3} \\ \arccos\left(\frac{1}{2}\sqrt{1+\frac{1}{|J'|}}\right) & |J'| \ge \frac{|J|}{3} \end{cases}$$
(7.39)

Figure 7.6 shows the classical spiral state for the ferromagnetic case (J < 0, J' > |J|/3) where the spin orientations at A and B lattice sites as numbered on the figure are defined the angle $\theta_n = n\Phi_{\rm cl}$. For the antiferromagnetic case (J > 0, J' < -J/3) all of the spins on one sublattice are reversed. We note that $\Phi = 0$ corresponds to the collinear state. The classical transition between the collinear and the noncollinear state is of second order and takes place at the critical point $J'_c = -J/3$. Figure 7.7 gives an illustration of the complete classical ground-state phase diagram.

We choose the spiral state with the characteristic angle Φ (illustrated in Fig. 7.6) as our CCM model state. Further details concerning the treatment of the J-J' model via the CCM are given in [64, 77, 81]. We calculate the ground state and the low-lying excitations of the Hamiltonian of (7.38). We use the CCM for high orders of approximation up to LSUB8 which contains 4986 fundamental configurations for the Néel model state with $\Phi = 0$ and 42160 fundamental configurations for a helical model state with $\Phi \neq 0$. (We



Fig. 7.7. Classical ground-state phase diagram for the J-J' model on the square lattice with competing nearest-neighbour bonds, indicating the collinear Néel and ferromagnetic phases and the noncollinear spiral phases for various values of J and J'. (Figure taken from [77])

note that such a large number of configurations as the latter case may be considered only by using parallel processing techniques, although this is not performed here. The interested reader is referred to [83] for more details of a parallelised implementation of the CCM.) By way of comparison we also exactly diagonalise finite sized lattices of up to N = 32 spins with periodic boundary conditions. We extrapolate to the infinite-lattice limit using standard finite-size scaling laws.

For sufficiently strong antiferromagnetic J' bonds the J-J' model is characterised by a tendency to singlet pairing of the two spins coupled by a J'bond, and hence the long-range magnetic (collinear or noncollinear) order is destroyed. We observe clear indications of a second-order phase transition to a quantum paramagnetic dimerised phase at a certain critical value of $J' = J'_s$. However the only case examined in detail here is the antiferromagnetic case (J = +1). Evidence of a phase transition to a dimension phase is indicated by the sublattice magnetisation (see Fig. 7.8). The results of the CCM and exact diagonalisations (ED) agree well with each other and with the result $J_s' \approx 2.56$ from cumulant series expansions [79], whereas by contrast renormalised spin wave theories (RSWT) clearly overestimate the order. We also note that another indication of a dimerised phase is the appearance of a gap Δ between the singlet ground state and the first triplet excitation. The gap appears to open in the range $2.5 \lesssim J'_s \lesssim 3.0$ for both the ED and CCM calculations (see Fig. 7.8). This result is in good agreement with the corresponding estimates for the critical point using the magnetisation.

The phase transition to the dimerised phase is also indicated by the spin stiffness ρ_s . The ground-state stiffness is a variable which indicates the distance of the ground state from criticality, and the breakdown of the Néel long-range order is thus accompanied by ρ_s going to zero. The spin stiffness



Fig. 7.8. Indications of a phase transition to the dimerised phase for the J-J' model on the square lattice with competing nearest-neighbour bonds, with J = +1. Left graph: sublattice magnetisation versus J' using the CCM, exact diagonalisation, and spin wave theories. Right graph: spin gap versus J' using the CCM compared to results of exact diagonalisation. (Figure taken from [77])

measures the amount of energy used in introducing a twist θ to the direction of spin between every pair of neighbouring rows, such that

$$\rho_s = \left. \frac{d^2}{d\theta^2} \frac{E_0(\theta)}{N} \right|_{\theta=0},\tag{7.40}$$

and this quantity may be calculated directly using the CCM.

We note that the magnetic order parameters may only tell us whether certain types of long-range order are present, whereas the spin stiffness has the advantage of being unbiased with respect to the nature of the ordering. The spin stiffness constitutes, together with the spin wave velocity, the fundamental parameter that determines the low-energy dynamics of magnetic systems [84]. The CCM LSUBn results are given in Fig. 7.9. We calculate the stiffness using two different directions of in-plane rows, i.e., rows parallel to the J' bonds and rows perpendicular to the J' bonds. We note that, although the results of the stiffness for the two directions are different in general (see Fig. 7.9), the phase transition points (i.e., the values of J' where ρ_s becomes zero) agree well with each other for the various LSUBn approximations although the extrapolated CCM results are expected to be even more accurate. Our calculations predict that $J'_s \approx 2.8$ which is again in good agreement with the results of the other methods. We note that this phase transition to the dimensional O(3)universality class as indicated by the value of the correlation length critical exponent [82].

We now consider the frustrated region of the J-J' model for J and J' with *different* signs. We note that classically there is a second-order phase transition from collinear order to noncollinear order at J' = -J/3 for both



Fig. 7.9. The spin stiffness versus J' for two different rows within the plane of the lattice are good indicators of a phase transition to the dimerised phase for the J-J' model on the square lattice with competing nearest-neighbour bonds, with J = +1. Left graph: rows along the x direction (parallel to the J' bonds). Right graph: rows along the y direction (perpendicular to the J' bonds). (Figure taken from [77])



Fig. 7.10. Pitch angle Φ versus |J'| for the quantum and the classical case of the J-J' model on the square lattice with competing nearest-neighbour bonds. Although Φ is classically the same for the ferromagnetic case (J = -1, J' > 0) and for the antiferromagnetic case (J = +1, J' < 0) we note that the quantum pitch angle is different for both cases. The curves to the left of the classical (dashed) curve belong to J = -1 and those to the right of it belong to J = +1. (Figure taken from [77])

antiferromagnetic and ferromagnetic nearest-neighbour J-bonds. By contrast, the behaviour of the quantum model for the two cases is different concerning the phase transition. In particular, we find that the critical point is shifted to $J' \approx -1.35$ (see Fig. 7.10) for the antiferromagnetic case (J = +1), although no such shift is observed for the ferromagnetic case (J = -1).

The exact diagonalisation (ED) data of the structure factor $S(\mathbf{k})$ (see Fig. 7.11) also agree with these findings. For J = +1 the collinear Néel order



Fig. 7.11. Ground-state structure factor $S(\mathbf{k}) \propto \sum_{i,j \in A} e^{i(\mathbf{R}_j - \mathbf{R}_i) \cdot \mathbf{k}} \langle \mathbf{s}_i \cdot \mathbf{s}_j \rangle$ (note that the indices *i* and *j* run over one sublattice) for a 8×4 lattice of the *J*–*J'* model on the square lattice with competing nearest-neighbour bonds for the quantum and the classical case for various spiral vectors \mathbf{k} for antiferromagnetic J = +1 (left graph) and ferromagnetic J = -1 (right graph). (Figure taken from [77])

 $[\mathbf{k} = (0,0)]$ becomes unstable in comparison to the noncollinear spiral order $[\mathbf{k} = (\pi/4, 0)]$ in the classical model for $J' \lesssim -0.36$. We note that this occurs only for $J' \leq -0.95$ in the quantum case. The situation for the ferromagnetic case (J = -1) is again different, and the results of the structure factor show that the transition from $\mathbf{k} = (0,0)$ (collinear ferromagnetic order) to $\mathbf{k} = (\pi/4, 0)$ (spiral order) takes place at nearly the same value of $J' \approx 0.36$ for both the classical and the quantum cases. We may also use the difference between the amount of the on-site magnetic moment $\langle s_i \rangle$ and its classical value $\langle s_i \rangle_{\rm cl} = 1/2$ as a measure of quantum fluctuations. We compare the strength of quantum fluctuations near the collinear-noncollinear transitions for both the antiferromagnetic and the ferromagnetic cases. Although the quantum fluctuations are particularly strong for J = +1 near the antiferromagneticspiral transition (leading to an on-site magnetic moment less then 20% of its classical value [64], there are virtually no quantum fluctuations at the ferromagnetic-spiral transition for J = -1 because the on-site magnetic moment takes its classical value 1/2 up to $J' \approx 0.36$ (cf. [81]). Hence the shift of the critical J'_c in the antiferromagnetic case can clearly be attributed to the strong quantum fluctuations.

We may summarise by saying that our findings are generally consistent with the statement that quantum fluctuations (which we have in the antiferromagnetic case only) prefer a collinear ordering. We note that the quantum collinear state can survive for the quantum model studied here into a classically frustrated region in which classical theory indicates that the collinear state is already unstable. In addition, our results indicate that there is a second-order phase transition for the ferromagnetic case (J = -1) which is



Fig. 7.12. Phase diagrams of the J-J' model on the square lattice with competing nearest-neighbour bonds for the classical case (left graph) and for the quantum case (right graph). The dashed line indicates a first-order phase transition, while the other transitions are of second order. (Figure taken from [77])

in agreement with classical theory. The collinear-noncollinear transition in the antiferromagnetic case (J = +1) is probably of first order for the quantum model (cf. Fig. 7.10 and discussion in [64]) which compares to a second-order transition for the classical case. Figure 7.12 compares the phase diagrams of this J-J' model for the classical and the quantum cases.

7.5 An Interpolating Kagomé/Triangle Model

We also wish to study another strongly frustrated spin-half Heisenberg model, namely one which interpolates smoothly between the triangular-lattice antiferromagnet (TAF) [59,85–87] and the Kagomé-lattice [65,88,89] antiferromagnet (KAF). We shall refer to this as the interpolating Kagomé/triangle model (illustrated in Fig. 7.13), and the Hamiltonian is given by

$$H = J \sum_{\langle i,j \rangle} \mathbf{s}_i \cdot \mathbf{s}_j + J' \sum_{\{i,k\}} \mathbf{s}_i \cdot \mathbf{s}_k \quad , \tag{7.41}$$

where $\langle i, j \rangle$ runs over all nearest-neighbour (n.n.) bonds on the Kagomé lattice, and $\{i, k\}$ runs over all n.n. bonds which connect the Kagomé lattice sites to those other sites on an underlying triangular lattice. Note that each bond is counted once and once only. We explicitly set J = 1 throughout this paper, and we note that at J' = 1 we thus have the TAF and at J' = 0 we have the KAF.

7.5.1 CCM Treatment of the Interpolating Kagomé/Triangle Model

For the interpolating Kagomé/triangle model described by (7.41), we choose a model state $|\Phi\rangle$ in which the lattice is divided into three sublattices, denoted {A,B,C}. The spins on sublattice A are oriented along the negative



Fig. 7.13. The interpolating Kagomé/triangle model is illustrated in diagram (a), where the bonds of strength J between Kagomé lattice sites are indicated by the thick solid lines and the non-Kagomé bonds of strength J' on the underlying triangular lattice sites are indicated by the "broken" lines. The triangular lattice Heisenberg antiferromagnet (TAF) is illustrated in diagram (b), and we note that the two models are equivalent when J = J'. The quadrilateral unit cells for both cases are also illustrated. The interpolating Kagomé/triangle model contains four sites per unit cell, whereas the TAF has only one site per unit cell. (Figure taken from [65])

z-axis, and spins on sublattices B and C are oriented at $+120^{\circ}$ and -120° , respectively, with respect to the spins on sublattice A. Our local axes are chosen by rotating about the *y*-axis the spin axes on sublattices B and C by -120° and $+120^{\circ}$ respectively, and by leaving the spin axes on sublattice A unchanged. Under these canonical transformations,

$$s_B^x \to -\frac{1}{2} s_B^x - \frac{\sqrt{3}}{2} s_B^z \quad ; \quad s_C^x \to -\frac{1}{2} s_C^x + \frac{\sqrt{3}}{2} s_C^z \quad , \\ s_B^y \to s_B^y \quad ; \quad s_C^y \to s_C^y \quad , \\ s_B^z \to \frac{\sqrt{3}}{2} s_B^x - \frac{1}{2} s_B^z \quad ; \quad s_C^z \to -\frac{\sqrt{3}}{2} s_C^x - \frac{1}{2} s_C^z \quad .$$
(7.42)

The model state $|\Phi\rangle$ now appears mathematically to consist purely of spins pointing downwards along the z-axis, and the Hamiltonian (for J = 1) is given in terms of these rotated local spin axes as,

$$H = \sum_{\langle i \to j \rangle} \left\{ -\frac{1}{2} s_i^z s_j^z + \frac{\sqrt{3\lambda}}{4} (s_i^z s_j^+ + s_i^z s_j^- - s_i^+ s_j^z - s_i^- s_j^z) + \frac{\lambda}{8} (s_i^+ s_j^- + s_i^- s_j^+) - \frac{3\lambda}{8} (s_i^+ s_j^+ + s_i^- s_j^-) \right\} + J' \sum_{\{i \to k\}} \left\{ -\frac{1}{2} s_i^z s_k^z + \frac{\sqrt{3\lambda}}{4} (s_i^z s_k^+ + s_i^z s_k^- - s_i^+ s_k^z - s_i^- s_k^z) + \frac{\lambda}{8} (s_i^+ s_k^- + s_i^- s_k^+) - \frac{3\lambda}{8} (s_i^+ s_k^+ + s_i^- s_k^-) \right\}.$$
(7.43)

Note that *i* and *j* run only over the N_K sites on the Kagomé lattice, whereas *k* runs over those non-Kagomé sites on the (underlying) triangular lattice. *N* indicates the total number of triangular-lattice sites, and each bond is counted once and once only. We also note that we have multiplied all of the off-diagonal terms in the new Hamiltonian by a factor of λ . We shall use this factor in order to determine the perturbation series around the Ising limit ($\lambda = 0$) for the ground-state energy and sublattice magnetisation. The case $\lambda = 1$ corresponds to our isotropic Heisenberg case of (7.41). The symbol \rightarrow indicates an explicit *bond directionality* in the Hamiltonian given by (7.43), namely, the *three* directed nearest-neighbour bonds included in (7.43) point from sublattice sites A to B, B to C, and C to A for both types of bond. We now perform high-order LSUBm calculations for this model via a computational procedure for the Hamiltonian of (7.43).

7.5.2 CCM Results for the Ground-State Properties

We note that for the CCM treatment of the interpolating Kagomé/triangle model presented here (and see [65] for further details) the unit cell contains four lattice sites (see Fig. 7.13). By contrast, previous calculations [59] for the TAF used a unit cell containing only a single site per unit cell. Hence, the interpolating Kagomé/triangle model has many more "fundamental" configurations than the TAF model at equivalent levels of approximation. However, we find that those configurations which are *not* equivalent for the interpolating Kagomé/triangle model but *are* equivalent for the TAF have CCM correlation coefficients $\{S_I, \tilde{S}_I\}$ which become equal at the TAF point, J' = 1. Hence, the CCM naturally and without bias reflects the extra amount of symmetry of the interpolating Kagomé/triangle model at this one particular point. This is an excellent indicator of the validity of the CCM treatment of this model. The results for the interpolating Kagomé/triangle model at J' = 1 thus also exactly agree with those of a previous CCM treatment of the TAF [59].

We now set $\lambda = 1$ for the remainder of this subsection and again we "track" the "trivial" solution for large J' for decreasing values of J' until we reach a *critical* value of J'_c at which the solution to the CCM equations breaks down. Results for J'_c for this model are presented in Table 7.2. A simple "heuristic" extrapolation of these results gives a value of $J'_c = 0.0 \pm 0.1$ for the position of this phase transition point. This result indicates that the classical three-sublattice Néel-like order, of which about 50% remains for the TAF, completely disappears at a point very near to the KAF point (J' = 0).

The results for the ground-state energy are shown in Fig. 7.14 and in Table 7.2. These results are seen to be highly converged with respect to each other over the whole of the region $0 \le J' \le 1$. The results for the ground-state energies of the KAF and TAF model in Table 7.2 agree well with results of other techniques. Indeed, we believe that the extrapolated CCM results



Fig. 7.14. CCM results for the ground-state energy per spin of the interpolating Kagomé/triangle model (with J = 1) using the LSUB*m* approximation with $m = \{2, 3, 4, 5, 6\}$. The boxes indicate the CCM critical points, J'_c , and a simple extrapolation in the limit $m \to \infty$ implies that $J'_c = 0.0 \pm 0.1$. (Figure taken from [65])

Table 7.2. CCM results [65] for the ground-state energy per spin and sublattice magnetisation of the TAF and KAF models using the LSUB*m* approximation with $m = \{2, 3, 4, 5, 6\}$. CCM critical values, J'_c , of the interpolating Kagomé/triangle model (with J = 1), which are themselves indicators of a phase transition point in the true system, are also given. Comparison is made in the last row with the results of other calculations

	KAF		TA	J-J'	
m	E_g/N_K	M^K	E_g/N	M^K	J_c'
2	-0.37796	0.8065	-0.50290	0.8578	-
3	-0.39470	0.7338	-0.51911	0.8045	-0.683
4	-0.40871	0.6415	-0.53427	0.7273	-0.217
5	-0.41392	0.5860	-0.53869	0.6958	-0.244
6	-0.41767	0.5504	-0.54290	0.6561	-0.088
∞	-0.4252	0.366	-0.5505	0.516	$0.0{\pm}0.1$
c.f.	-0.43 ([89])	0.0	-0.551 ([85])	0.5 ($[86, 87]$)	_

are unquestionably among the most accurate results available for the groundstate energies of the TAF and KAF.

We now wish to describe how much of the original classical ordering of the model state remains for the quantum system. If one considers non-Kagomé



Fig. 7.15. CCM results for the sublattice magnetisation of the interpolating Kagomé/triangle model (with J = 1) using the LSUB*m* approximation with $m = \{2, 3, 4, 5, 6\}$. (Figure taken from [65])

lattice sites then the spins on these sites are effectively "frozen" into their original directions (of the model state) at J' = 0. Hence, we believe that the relevant quantity to be considered for this model is the average value of s_k^z (again after rotation of the local spin axes) where k runs only over the N_K Kagomé lattice sites, given by

$$M^{K} = -\frac{2}{N_{K}} \sum_{k=1}^{N_{K}} s_{k}^{z} \quad . \tag{7.44}$$

The results for M^K are presented in Fig. 7.15 and in Table 7.2. The puzzling "upturn" of M^K for the LSUB5 data is an artifact, and typically such behaviour only ever occurs when one enters a phase in which the model state becomes an increasingly bad starting point. Although the extrapolated value for M^K specifically at the KAF point remains non-zero, the LSUB6 result goes to zero very close to the KAF point. CCM results are thus fully consistent with the hypothesis that, unlike the TAF, the ground state of the KAF does not contain any Néel ordering.

7.5.3 Evaluation of the Perturbation Series Using CCM

Finally, it is instructive to make contact with the cumulant series expansions for the anisotropic TAF (i.e., J' = J = 1) with respect to the parameter λ .

Table 7.3. Expansion coefficients in powers of λ up to the 15th order for the ground-state energy per spin, E_g/N , and the sublattice magnetisation, M, for the anisotropic spin- $\frac{1}{2}$ triangular-lattice Heisenberg antiferromagnet obtained from the CCM equations in the LSUB6 approximation. The exact series expansions up to the 11th order obtained by Singh and Huse [85] are also included for comparison. (Table taken from [59])

Order	LSUB6: E_g/N	Exact: E_g/N	LSUB6: M	Exact: M
0	-0.3750000	-0.3750000	1	1
1	0.0000000	0.0000000	0	0
2	-0.1687500	-0.1687500	-0.27	-0.27
3	0.0337500	0.0337500	0.108	0.108
4	-0.0443371	-0.0443371	-0.2726916	-0.2726916
5	0.0204259	0.0204259	0.1717951	0.1717951
6	-0.0283291	-0.0283291	-0.3315263	-0.3315263
7	0.0311703	0.0315349	0.4060277	0.4110737
8	-0.0357291	-0.0476598	-0.5331858	-0.7382203
9	0.0541263	0.0685087	0.8894023	1.1781303
10	-0.0771681	-0.1025446	-1.3927395	-2.0109889
11	0.1294578	0.1565522	2.4179612	3.4012839
12	-0.1848858	?	-4.0426184	?
13	0.2857225	?	6.8086538	?
14	-0.4463496	?	-11.488761	?
15	0.7021061	?	19.388053	?

We have computed the perturbative CCM solutions of E_g/N and the sublattice magnetisation M, as defined in (7.24) with respect to the local spin axes, in terms of the anisotropy parameter λ . In Table 7.3 we tabulate the expansion coefficients from the LSUB6 approximation, together with the corresponding results from exact series expansions [85]. We note that the LSUB6 approximation reproduces the exact series expansion up to the sixth order. We conjecture that the LSUB*m* approximation reproduces the *exact* series expansion to the same *m*th order. Moreover, the fact that the corresponding values of several of the higher-order expansion coefficients from both the CCM LSUB6 perturbative solution and the exact series expansion remain close to each other shows that the exponential parametrisation of the CCM with the inclusion of multi-spin correlations up to a certain order also captures the dominant contributions to correlations of a few higher orders in the series expansions.

7.6 The J_1 – J_2 Ferrimagnet

We now briefly present results for another frustrated model in which we have both nearest- and next-nearest-neighbour antiferromagnetic bonds. (The interested reader is referred to [68,77] for more details.) The Hamiltonian for the square-lattice spin-half/spin-one J_1-J_2 ferrimagnet is given by

$$H = J_1 \sum_{\langle i,j \rangle} \mathbf{s}_i \cdot \mathbf{s}_j + J_2 \sum_{\{i,k\}} \mathbf{s}_i \cdot \mathbf{s}_k \quad , \tag{7.45}$$

where $\langle i, j \rangle$ runs over all nearest-neighbour bonds on the square lattice, $\{i, k\}$ runs over all next-nearest-neighbour bonds. We assume that $J_1 = 1$ and $J_2 > 0$ throughout this article and so this model is *frustrated*. Note that one sublattice (A) of the square lattice is populated entirely by spin-one spins ($s_A = 1$) and the other sublattice (B) is populated entirely by spin-half spins ($s_B = 1/2$). This model is an extension of the well-known spin-half J_1-J_2 model on the square lattice (see e.g. [90–96] and references therein) which serves as a canonical model for the discussion of an order-disorder quantum phase transition driven by the interplay of quantum fluctuations and frustration.

A feature of such ferrimagnetic spin systems is that the Lieb-Mattis theorem may be obeyed (if frustration is excluded) such that the ground state has a magnetic moment per spin of strength $(s_A - s_B) = 1/2$. Note in particular that this property is obeyed for the ferrimagnet of (7.45) at $J_2 = 0$, and thus a macroscopic lattice magnetisation exists for this case.

We note that many ferrimagnetic materials have recently been fabricated and various examples are the "ladder" systems: MnCu(*pba OH*) (H₂O)₃ (where *pba* OH=2-hydroxy-1, 3-propylenebisoxamato) and MnCu(*pba*) (H₂O)₃ \cdot H₂O (where *pba* = 1,3-propylenebisoxamato) [97–99]. These materials contain magnetic atoms Mn ($s_A = 5/2$) and Cu ($s_B = 1/2$).

The classical behaviour of the square-lattice spin-half/spin-one ferrimagnet of (7.45) is also interesting and three distinct phases are predicted. The first such phase for $J_2 \leq 0.25$ is one in which the ground-state is the collinear ferrimagnetic Néel state (shown in Fig. 7.16). A second-order phase transition then occurs within this classical picture to a phase in which the spin-one spins may cant at an angle θ , although the spin-half spins do *not* change their direction (also shown in Fig. 7.16). This state is henceforth referred to as the "spin-flop" state. A first-order phase transition to a collinear state in which next-nearest-neighbour spins are antiparallel (again, see Fig. 7.16) then occurs classically, and this state is referred to here as the "collinear striped" state. Note that this state in the classical model is degenerate with states canting at an arbitrary angle between spins on sublattice A and spins on sublattice B. However, this degeneracy is lifted by quantum fluctuations which select the collinear state [92, 100, 101]. We note that the spin-flop and collinear striped states are "incommensurate" in the sense that no value of the angle θ may be chosen such that the two states are equivalent. We note however that the Néel and spin-flop states are equivalent when $\theta = \pi$.

A further motivation for studying this model is that exact calculations of finite-sized lattices indicate that the behaviour of the quantum ferrimagnet of



Fig. 7.16. The classical ground states used to treat the square-lattice spinhalf/spin-one J_1-J_2 ferrimagnet via the CCM. These states are namely: a) the collinear ferrimagnetic Néel state; b) the collinear "striped" state; and c) the "spinflop" state. Note that long arrows indicate spin-one spins (and their orientation) and short arrows indicate the spin-half spins. The primitive unit cells are shown by the ovals for the Néel and striped states and by the rectangles for the spin-flop state. Full lines indicate the nearest-neighbour bonds and dashed lines indicate the next-nearest-neighbour bonds. (Figure taken from [77])

(7.45) may be much different from the behaviour of its linear chain counterpart and from the square-lattice spin-half J_1-J_2 antiferromagnet. These exact calculations of finite-sized lattices suggest that the behaviour of the squarelattice ferrimagnet is analogous to that of the classical behaviour outlined above.

Three model states are used to treat the square-lattice spin-half/spinone $J_1 - J_2$ ferrimagnet in order to provide results across various regimes of differing quantum order. The first such model state is the collinear Néel state, and the primitive unit cell in this case contains a spin-half site and a spin-one site (shown in Fig. 7.16). The underlying Bravais lattice is formed from two vectors $(\sqrt{2}, \sqrt{2})$ and $(\sqrt{2}, -\sqrt{2})$. There is also an 8-point symmetry group, namely: rotations of 0° , 90° , 180° , and 270° ; and reflections about the lines x = 0, y = 0, y = x, and y = -x. The collinear striped state (also shown in Fig. 7.16) is also used as a model state, in which spins for even values of x along the x-axis point "downwards" and spins for odd values of x point "upwards." The primitive unit cell again contains only two spins, although this time only four of the eight point group symmetry operations are allowed, namely: rotations of 0° and 180° ; and reflections about the lines x = 0 and y = 0. We note that rotations of the local axes of 180° about the y-axis of every spin is carried out such that each spin now appears (mathematically) to point "downwards". Each spin may now be treated equivalently.

It is noted that (in the original unrotated spin coordinates) $s_T^z \equiv \sum_i s_i^z = 0$ is preserved for all CCM approximations for these two models states in order to reduce the number of fundamental configurations at a given LSUB*m* or SUB*m*-*m* approximation level.



Fig. 7.17. Ground-state energy per spin of the square-lattice spin-half/spin-one J_1 – J_2 ferrimagnet versus J_2 for the CCM method using three model states compared to results of exact diagonalisations of finite-sized lattices. (Note that we have set $J_1 = 1$)

The third model state is the spin-flop state. We note that there is no equivalent conserved quantity to s_T^z for the spin-flop model state, although single-body correlations are explicitly excluded from this calculation as they are (in some sense) already included by the rotation of the local axes of spins. It should however be noted that this is an explicit assumption of the calculation for the spin-flop model state.

The amount of ordering on each sublattice is represented by,

$$m_1 = \frac{1}{N_A} \left| \sum_{i_A}^{N_A} \langle \tilde{\Psi} | s_{i_A}^z | \Psi \rangle \right| \qquad ; \qquad m_2 = \frac{1}{N_B} \left| \sum_{i_B}^{N_B} \langle \tilde{\Psi} | s_{i_B}^z | \Psi \rangle \right| \tag{7.46}$$

where i_A runs over all spin-one lattice sites and i_B runs over all spin-half lattice sites. Note that, as usual, all of the spins for all of the models states have been rotated such that all spins appear mathematically to point downwards. The quantities m_1 and m_2 are the expectation values of the magnetic moment in the z-direction on the A and B sublattice, respectively, with respect to a given model state and represent order parameters for this model.

The ground-state energies predicted by the CCM using the three model states are shown in Fig. 7.17 and once again CCM results are in good agreement with results of exact diagonalisations (ED) of finite-sized lattices and spin-wave theory. Results for the sublattice magnetisations shown in Fig. 7.18



Fig. 7.18. Sublattice magnetisations, m_1 and m_2 of (7.46), of the square-lattice spin-half/spin-one J_1-J_2 ferrimagnet versus J_2 for the CCM method using three model states compared to results of exact diagonalisations (ED) of finite-sized lattices. Note that full lines are those results for the sublattice magnetisation using linear spin-wave theory, open circles are those results of exact diagonalisations, and CCM results are indicated by the filled circles. (Figure taken from [68])

also show good correspondence with results of exact diagonalisations (ED) of finite-sized lattices and spin-wave theory. Hence, the CCM yields excellent quantitative accuracy for the ground-state properties of the spin-half/spinone J_1-J_2 ferrimagnet across a wide range of the next-nearest-neighbour bond strength J_2 by the use of three different model states. The CCM thus provides a comprehensive picture of the ordering in the ground state, an accurate prediction of the phase diagram, and even evidence regarding the order of the phase transitions.

The results for the ground-state properties of the spin-half/spin-one J_1 – J_2 ferrimagnet at $J_1 = 1$ and $J_2 = 0$ based on the Néel model state are presented in Table 7.4 and we see that our raw SUB*m*-*m* results appear to converge rapidly. An extrapolation in the limit $m \to \infty$ is also performed for the J_1-J_2 ferrimagnet at $J_1 = 1$ and $J_2 = 0$ in order to provide even better accuracy.

The positions of quantum phase transition points as a function of $\alpha = J_2/J_1$ are also shown in Fig. 7.18. A second-order phase transition is observed at α_{c_1} and CCM results place this at $\alpha_{c_1} = 0.27$, which is slightly above the classical value. By contrast, CCM results predict a first-order phase transition at $\alpha_{c_2} \approx 0.5$ and this result is in good agreement with those results of both spin-wave theory and exact diagonalisations. Another possible phase is also indicated in Fig. 7.18, namely, one is which we have finite and non-zero sublattice magnetisation on the spin-one lattice sites and zero sublattice

Table 7.4. Results for the ground-state energy and amounts of sublattice magnetisations m_1 and m_2 , on the spin-one and spin-half sites respectively, of the square-lattice spin-half/spin-one J_1-J_2 ferrimagnet at $J_1 = 1$ and $J_2 = 0$ based on the Néel model state [77]. Note that N_F indicates the number of fundamental configurations at a given level of LSUB*m* or SUB*m*-*m* approximation. CCM results are compared to exact diagonalisations (ED) of finite-sized lattices. Heuristic extrapolations in the limit $m \to \infty$ are performed

	N_F	E_g/N	m_1	m_2
SUB2-2	1	-1.192582	0.92848	0.42848
SUB4-4	13	-1.204922	0.90781	0.40781
SUB6-6	268	-1.206271	0.90333	0.40334
LSUB6	279	-1.206281	0.90329	0.40330
Extrapolated CCM	-	-1.2069	0.898	0.398
ED $(N = 16)$	-	-1.218134	0.87515	0.37515
ED $(N = 20)$	-	-1.212050	0.88482	0.38483

magnetisation on the spin-half lattice sites. The onset of this phase with increasing J_2 is indicated by the symbol, α^* .

7.7 Conclusion

We have seen in this chapter that the CCM may be applied to various quantum spin systems at zero temperature. In particular, suggestive results for the positions of CCM critical points were observed, and these points were seen to correspond closely to the occurrence of quantum phase transitions in the "real" system. Furthermore, quantitatively accurate results for expectation values with respect to the ground and excited states were determined. These results were seen to be competitive with the best results of other approximate methods.

A possible use of high-order CCM techniques might be to predict excitation spectra of quantum magnets to great accuracy. Furthermore, this would mean that a direct connection might be made to results of neutron scattering experiments. Also, the application of the CCM to quantum spin systems which exhibit novel ordering, such as those with ground states which demonstrate dimer- or plaquette-solid ordering, is another possible future goal. Furthermore, the extension of high-order techniques to bosonic and fermionic systems is possible in future.

High-order CCM techniques might also be applied at even greater levels of approximation with the aid of parallel processing techniques [83]. Indeed, the CCM is well-suited to such an implementation and recent CCM calculations using parallel processing techniques have been carried out for approximately 10^4 fundamental configurations. We believe that an increase of at least ano-

ther order of magnitude in the number of fundamental configurations should easily be possible in the near future by using such techniques.

The extension of the CCM to quantum spin systems at non-zero temperatures might also be accomplished by using thermo-field theory. The application of the CCM at both zero and non-zero temperatures might then help to explain the subtle interplay of quantum and thermal fluctuations in driving phase transitions over a wide range of physical parameters.

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8 Integrability of Quantum Chains: Theory and Applications to the Spin-1/2 XXZ Chain

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Abstract. In this contribution we review the theory of integrability of quantum systems in one spatial dimension. We introduce the basic concepts such as the Yang-Baxter equation, commuting currents, and the algebraic Bethe ansatz. Quite extensively we present the treatment of integrable quantum systems at finite temperature on the basis of a lattice path integral formulation and a suitable transfer matrix approach (quantum transfer matrix). The general method is carried out for the seminal model of the spin-1/2 XXZ chain for which thermodynamic properties like specific heat, magnetic susceptibility and the finite temperature Drude weight of the thermal conductivity are derived.

8.1 Introduction

Integrable quantum chains have continuously attracted attention, because of the possibility of obtaining exact data for the spectrum and other physical properties despite the truly interacting nature of the spins resp. particles. Important examples of these systems are the Heisenberg model [1,2], tJ- [3–5] and Hubbard models [6,7] The computational basis for the work on integrable quantum chains is the Bethe ansatz yielding a set of coupled non-linear equations for 1-particle wave-numbers (Bethe ansatz roots). Many studies of the Bethe ansatz equations were directed at the ground-states of the considered systems and have revealed interesting non-Fermi liquid properties such as algebraically decaying correlation functions with non-integer exponents and low-lying excitations of different types, i.e. spin and charge with different velocities constituting so-called spin and charge separation, see [8–10].

A very curious situation arises in the context of the calculation of the partition function from the spectrum of the Hamiltonian. Despite the validity of the Bethe ansatz equations for all energy eigenvalues of the above mentioned models the direct evaluation of the partition function is rather difficult. In contrast to ideal quantum gases the eigenstates are not explicitly known, the Bethe ansatz equations provide just implicit descriptions that pose problems of their own kind. Yet, knowing the behaviour of quantum chains at finite temperature is important for many reasons. As a matter of fact, the strict ground-state is inaccessible due to the very fundamentals of thermodynamics. Therefore the study of finite temperatures is relevant for theoretical as well as experimental reasons.

The purpose of this review is to introduce to a unified treatment of the ground-state and the finite temperature properties of integrable quantum chains. First, in Sect. 8.2 we introduce the essential concepts of quantum integrability. We show how to construct an infinite set of conserved currents, i.e. local operators commuting with the Hamiltonian. At the heart of these constructions is the embedding of the Hamiltonian into a family of commuting row-to-row transfer matrices of a certain classical model. In Sect. 8.3 we derive a lattice path integral representation for the partition function of a rather large class of integrable Hamiltonians at finite temperature. Here we also introduce a very efficient transfer matrix method based on the quantum transfer matrix. In Sect. 8.4 we review the algebraic Bethe ansatz for the seminal model of the partially anisotropic spin-1/2 Heisenberg chain (XXZ) chain) related to the classical six-vertex model on a square lattice. The results of this section are the construction of eigenstates and eigenvalues of the Hamiltonian as well as the quantum transfer matrix of the system. In Sect. 8.5 we demonstrate how to transform the large number of coupled Bethe ansatz equations into a simple finite set of non-linear integral equations. These equations are studied numerically in Sect. 8.6 where explicit results for the temperature dependence of specific heat and susceptibility of the spin-1/2XXZ chain are given. Finally, in Sect. 8.7 we resume the line of reasoning developed for the proof of integrability and study the thermal conductivity of the XXZ chain a topic that is of considerable current interest.

8.2 Integrable Exchange Hamiltonians

We begin with the definition and discussion of a general class of quantum chains with nearest-neighbour interactions based on (graded) permutations. Consider a one-dimensional lattice with L sites and periodic boundary conditions imposed. A q-state spin variable α_i is assigned to each site i. We generally consider the situation where each spin α has its own grading, i.e. statistics number $\epsilon_{\alpha} = (-1)^{\xi_{\alpha}} = \pm 1$. A spin α with $\epsilon_{\alpha} = +1$ ($\epsilon_{\alpha} = -1$) is called bosonic (fermionic). The Hamiltonian of the "permutation model" can be introduced as

$$H = \sum_{i=1}^{L} P_{i,i+1} \tag{8.1}$$

with the (graded) permutation operator $P_{i,i+1}$

$$P_{i,i+1} |\alpha_1 \cdots \alpha_i \alpha_{i+1} \cdots \alpha_L\rangle = (-1)^{\xi_{\alpha_i \alpha_{i+1}}} |\alpha_1 \cdots \alpha_{i+1} \alpha_i \cdots \alpha_L\rangle, \qquad (8.2)$$

where $\xi_{\alpha_i\alpha_{i+1}}$ is 1 if both α_i and α_{i+1} are fermionic, and 0 otherwise.

Model (8.1) is shown to be exactly solvable on the basis of the Yang-Baxter equation. Many well-known exactly solvable models are of type (8.1), e.g. the spin-1/2 Heisenberg chain with q = 2 and $\epsilon_1 = \epsilon_2 = +1$, the free fermion model with q = 2 and $\epsilon_1 = -\epsilon_2 = +1$, and the supersymmetric t-Jmodel with q = 3 and $\epsilon_1 = -\epsilon_2 = \epsilon_3 = +1$. If m of the ϵ 's are +1 and n(=q-m) are -1, for example $\epsilon_1 = \cdots = \epsilon_m = +1$, $\epsilon_{m+1} = \cdots = \epsilon_q = -1$, we call the model (m, n)-permutation model or just (m, n)-model.

Before sketching the proof of integrability of the general permutation model we look closer at two important special cases. The (2,0)-model ($q = 2, \epsilon_1 = \epsilon_2 = +1$) is the spin-1/2 Heisenberg chain with Hamiltonian

$$H = 2\sum_{i=1}^{L} S_i \cdot S_{i+1} + L/2, \qquad (8.3)$$

in terms of SU(2) spin-1/2 operators S.

The (2, 1)-model (q = 3, $\epsilon_1 = \epsilon_2 = +1$ and $\epsilon_3 = -1$) is the supersymmetric t - J model with Hamiltonian (ignoring a trivial shift)

$$H = -t \sum_{j,\sigma} \mathcal{P}(c_{j,\sigma}^{\dagger} c_{j+1,\sigma} + c_{j+1,\sigma}^{\dagger} c_{j,\sigma}) \mathcal{P} + J \sum_{j} (S_j S_{j+1} - n_j n_{j+1}/4), \quad (8.4)$$

with standard fermionic creation and annihilation operators c^{\dagger} and c, projector $\mathcal{P} = \prod_{j} (1 - n_{j\uparrow} n_{j\downarrow})$ ensuring that double occupancies of sites are forbidden, and 2t = J (with normalization t = 1). The supersymmetric t - J model was shown to be integrable [11, 12] by the well-known Bethe ansatz [13, 14]. The ground-state and excitation spectrum were investigated [15] and critical exponents calculated by finite-size scaling and conformal field theory studies [16, 17]. The thermodynamical properties were studied in [5] by use of the thermodynamical Bethe ansatz (TBA) and in [18, 19] by use of the quantum transfer matrix (QTM).

For proving the integrability of the quantum system, a classical counterpart is defined on a two-dimensional square lattice of $L \times N$ sites, where we impose periodic boundary conditions throughout this paper. We assume that variables taking values $1, 2, \dots, q$ are assigned to the bonds of the lattice. Boltzmann weights are associated with local vertex configurations α , β , μ and ν and are denoted by $R^{\alpha\mu}_{\beta\nu}$, see Fig. 8.1. The classical counterpart to (8.1) is the Perk-Schultz model [20] with the following Boltzmann weights

$$R^{\alpha\mu}_{\beta\nu}(u,v) = \delta_{\alpha\nu}\delta_{\beta\mu} + (u-v)\cdot(-1)^{\xi_{\alpha}\xi_{\mu}}\cdot\delta_{\alpha\beta}\delta_{\mu\nu},\tag{8.5}$$

where u and v are freely adjustable "interaction" parameters assigned to the entire horizontal and vertical lines intersecting in the particular vertex under consideration. These weights satisfy the Yang-Baxter equation which we depict graphically in Fig. 8.2.

In the remainder of this section we want to indicate: (i) how the classical model is related to the quantum chain, and (ii) why the models are integrable. These issues are best discussed in terms of the transfer matrices of the

$$R^{\alpha\mu}_{\beta\nu}(u,v) = \alpha - \frac{\mu}{\sqrt{v}} \beta$$

Fig. 8.1. Graphical depiction of the fundamental *R*-matrix (8.5). The Boltzmann weight assigned to each vertex configuration (configuration of spin variables around a vertex) corresponds to a matrix element of the matrix R



Fig. 8.2. Depiction of the fundamental Yang-Baxter equation (YBE) for the R-matrix with the following graphical rules: Each bond carries a spin variable (ranging from 1 to q), each vertex (via Fig. 8.1) corresponds to a local Boltzmann weight R depending on the local spin configuration. The algebraic term corresponding to each graph of the equation is obtained by multiplying the Boltzmann weights corresponding to the (three) vertices and summing over spin variables on closed (inner) bonds. The values of the spin variables on open (outer) bonds are fixed and the assignment of these values is identical for both sides. The statement of the graphical equation is that both sides of the equation evaluate to the same expression for arbitrary, however identical assignment of spins to the open bonds on either side. A consequence of the YBE is the commutativity of the transfer matrix with respect to the spectral parameter, see Sect. 8.3 and Fig. 8.6

classical model

$$T^{\mu}_{\nu}(u) = \sum_{\{\alpha\}} \prod_{i=1}^{L} R^{\alpha_{i}\mu_{i}}_{\alpha_{i+1}\nu_{i}}(u, v_{i}), \qquad (8.6)$$

where we consider the spectral parameters v_i on the vertical bonds as fixed, i.e. independent of u. In dependence on the spectral parameter u the object T(u) represents a family of commuting matrices (the proof will be given in a slightly more general setting in the subsequent section)

$$T(v)T(w) = T(w)T(v),$$
 for arbitrary $v, w.$ (8.7)

Commutativity holds especially in the case of vanishing spectral parameters $v_i = 0$ on the vertical bonds, which case we refer to as the row-to-row transfer matrix. For this we have the additional simple limiting behaviour

$$T(0) = \text{translation (shift) operator} = e^{iP},$$
 (8.8)

$$\left. \frac{d}{du} \ln T(u) \right|_{u=0} = \text{Hamiltonian} = H, \tag{8.9}$$

also known as the Hamiltonian limit (P: momentum operator) [21, 22].

Apparently, Hamiltonians obtained as members of commuting families of operators possess (infinitely) many conserved quantities. Any element of the family (or higher order derivative) commutes with H. For the case of isotropic SU(m, n)-symmetric systems we have presented the typical proof of integrability based on classical models satisfying the YBE. There are many more models satisfying the YBE with different or reduced symmetries. A famous example of a system with reduced symmetry is the partially anisotropic spin-1/2 Heisenberg chain (also known as XXZ chain). The Hamiltonian and R-matrix corresponding to this will be given in the next section.

Above, we have only shown how to incorporate the momentum operator and the Hamiltonian into the family of commuting operators T(u). As indicated, $\ln T(u)$ is a generating function for conserved quantities

$$\mathcal{J}^{(n)} = \left(\frac{\partial}{\partial u}\right)^n \ln T(u)\Big|_{u=0},\tag{8.10}$$

that appear to be sums of local operators (with or without physical relevance). Quite generally, the current $\mathcal{J}^{(2)}$ is related to the thermal current, whose conservation implies non-ballistic thermal transport, see Sect. 8.7.

Logically, the presentation of diagonalization of the Hamiltonian or, more generally, of the row-to-row transfer matrix should follow directly after the above discussion of integrability. We will see, however, that another class of transfer matrices is worthwhile to be studied. This is a class of commuting staggered transfer matrices (quantum transfer matrices) occurring in the study of the thermodynamics of the quantum system at finite temperature (next section). We therefore postpone the discussion of the algebraic Bethe ansatz for both classes of transfer matrices to Sect. 8.4.

8.3 Lattice Path Integral and Quantum Transfer Matrix

At the beginning of this section we want to comment on the various techniques developed for the study of thermodynamics of integrable systems at the example of the simplest one, namely the spin-1/2 Heisenberg chain.

The thermodynamics of the Heisenberg chain was studied in [1, 2, 23]by an elaborate version of the method used in [24]. The macro-state for a given temperature T is described by a set of density functions formulated for the Bethe ansatz roots satisfying integral equations obtained from the Bethe ansatz equations (8.40). In terms of the density functions expressions for the energy and the entropy are derived. The minimization of the free energy functional yields what is nowadays known as the Thermodynamical Bethe Ansatz (TBA).

There are two "loose ends" in the sketched procedure. First, the description of the spectrum of the Heisenberg model is built on the so-called "string hypothesis" according to which admissible Bethe ansatz patterns of roots are built from regular building blocks. This hypothesis was criticized a number of times and led to activities providing alternative approaches to the finite temperature properties.

The second "loose end" within TBA concerns the definition of the entropy functional. In [1, 2, 23, 24] the entropy is obtained from a combinatorial evaluation of the number of micro-states compatible with a given set of density functions of roots. As such it is a lower bound to the total number of microstates falling into a certain energy interval. However, this procedure may be viewed as a kind of saddle point evaluation in the highly dimensional subspace of all configurations falling into the given energy interval. Hence, the result is correct in the thermodynamic limit and the "second loose end" can actually be tied up. Interestingly, the "second loose end" of the TBA approach was motivation for a "direct" evaluation [25] of the partition function of integrable quantum chains. A straightforward (though involved) calculation leads to the single non-linear integral equation of [26].

In this section we want to introduce the approach to thermodynamics of integrable quantum chains that we believe is the most efficient one, namely the "quantum transfer matrix" (QTM) approach. The central idea of this technique is a lattice path-integral formulation of the partition function of the Hamiltonian and the definition of a suitable transfer matrix [27–35].

In order to deal with the thermodynamics in the canonical ensemble we have to deal with exponentials of the Hamiltonian H. These operators are obtained from the row-to-row transfer matrix T(u) of the classical model in the Hamiltonian limit (small spectral parameter u) (8.9)

$$T(u) = e^{iP + uH + O(u^2)},$$
 (8.11)

with P denoting the momentum operator.

The main idea of the quantum transfer matrix (QTM) method at finite temperature is as simple as follows (for details the reader is referred to the papers [34,35]). First, let us define a new set of vertex weights \overline{R} by rotating R by 90 degrees as

$$\overline{R}^{\alpha\mu}_{\beta\nu}(u,v) = R^{\mu\beta}_{\nu\alpha}(v,u), \qquad (8.12)$$

see Fig. 8.3. We further introduce an "adjoint" transfer matrix $\overline{T}(u)$ as a product of $\overline{R}(-u,0)$ [7,34] with Hamiltonian limit

$$\overline{T}(u) = \mathrm{e}^{-\mathrm{i}P + uH + O(u^2)}.$$
(8.13)

With these settings the partition function Z_L of the quantum chain of length

$$\overline{R} \begin{array}{c} \alpha \mu \\ \beta \nu \end{array} (u,v) = \alpha \begin{array}{c} \mu \\ u \\ v \end{array} \beta \qquad \widetilde{R} \begin{array}{c} \alpha \mu \\ \beta \nu \end{array} (u,v) = \alpha \begin{array}{c} \mu \\ u \\ v \end{array} \beta \\ v \end{array}$$

Fig. 8.3. Graphical depiction of the \overline{R} and \widetilde{R} matrices ((8.12) and (8.19)) associated with R. The Boltzmann weights assigned to the vertex configurations \overline{R} and \widetilde{R} correspond to the Boltzmann weights of the rotated fundamental vertices R

L at finite temperature T reads

$$Z_L = \operatorname{Tr} e^{-\beta H} = \lim_{N \to \infty} Z_{L,N}$$
(8.14)

where $\beta = 1/T$ and $Z_{L,N}$ is defined by

$$Z_{L,N} := \operatorname{Tr} \left[T(-\tau)\overline{T}(-\tau) \right]^{N/2}, \qquad \tau := \frac{\beta}{N}.$$
(8.15)

The r.h.s. of this equation can be interpreted as the partition function of a staggered vertex model with alternating rows corresponding to the transfer matrices $T(-\tau)$ and $\overline{T}(-\tau)$, see Fig. 8.4. We are free to evaluate the partition function of this classical model by adopting a different choice of transfer direction. A particularly useful choice is based on the transfer direction along the chain and on the corresponding transfer matrix T^{QTM} which is defined for the columns of the lattice. The partition function of the quantum chain at temperature $1/\beta$ is given by

$$Z_{L,N} = \operatorname{Tr} \left(T^{\mathrm{QTM}} \right)^{L}.$$
(8.16)

In the remainder of this paper we will refer to T^{QTM} as the "quantum transfer matrix" of the quantum spin chain, because T^{QTM} is the closest analogue to the transfer matrix of a classical spin chain. Due to this analogy the free energy f per lattice site is given just by the largest eigenvalue Λ_{max} of the QTM

$$f = -k_B T \lim_{N \to \infty} \log \Lambda_{\max}.$$
 (8.17)

Note that the eigenvalue depends on the argument $\tau = \beta/N$ which vanishes in the limit $N \to \infty$ requiring a sophisticated treatment.

The QTM as defined above is actually a member of a commuting family of matrices $\mathcal{T}^{\rm QTM}$ defined by

$$\left[T^{\text{QTM}}\right]_{\nu}^{\mu}(v) = \sum_{\alpha} \prod_{j=1}^{N/2} R^{\alpha_{2j-1}\mu_{2j-1}}_{\alpha_{2j}\nu_{2j-1}}(v,\tau) \widetilde{R}^{\alpha_{2j}\mu_{2j}}_{\alpha_{2j+1}\nu_{2j}}(v,-\tau),$$
(8.18)



Fig. 8.4. Illustration of the two-dimensional classical model onto which the quantum chain at finite temperature is mapped. The square lattice has width L identical to the chain length, and height identical to the Trotter number N. The alternating rows of the lattice correspond to the transfer matrices $T(-\tau)$ and $\overline{T}(-\tau)$, $\tau = \beta/N$. The column-to-column transfer matrix T^{QTM} (quantum transfer matrix) is of particular importance to the treatment of the thermodynamic limit. The arrows placed on the bonds indicate the type of local Boltzmann weights, i.e. R and \overline{R} -matrices alternating from row to row. (The arrows indicate the type of Boltzmann weight, they do not denote local dynamical degrees of freedom)

where we introduced yet another vertex weight \tilde{R} as a rotation of R by -90 degrees

$$\widetilde{R}^{\alpha\mu}_{\beta\nu}(u,v) = R^{\nu\alpha}_{\mu\beta}(v,u).$$
(8.19)

Here we have introduced a spectral parameter v such that $\mathcal{T}_{\text{QTM}}(v)$ is a commuting family of matrices generated by v. A proof of this consists of two steps. First, we observe that \tilde{R} matrices and R matrices share the same intertwiner, i.e. the order of multiplication of two \tilde{R} matrices is interchanged by the same R matrix as in the case of the fundamental YBE, see Fig. 8.5. As R and \tilde{R} matrices share the same intertwiner, the transfer matrix obtained from products of arbitrary sequences of R and \tilde{R} with same spectral parameter (say v) on the continuous line constitutes a family of commuting matrices (spanned by v). The proof of this statement is graphically depicted in Fig. 8.6 for the case of the matrix $T^{\text{QTM}}(v)$ (8.18). In other words, $T^{\text{QTM}}(v)$ is integrable which allows us to diagonalize $T_{\text{QTM}}(v)$. The final results, of course, are physically interesting just for v = 0 as the physically meaningful QTM is identical to $T^{\text{QTM}}(0)$.

The main difference to the transfer matrix treatment of classical spin chains is the infinite dimensionality of the space in which T^{QTM} is living (for $N \to \infty$). In formulating (8.17) we have implicitly employed the in-



Fig. 8.5. Graphical derivation of the Yang-Baxter equation for \tilde{R} matrices. In the upper row, the fundamental YBE for R matrices is shown. The YBE for \tilde{R} matrices is obtained through rotation. Note that the intertwiner for R vertices is identical to the intertwiner for \tilde{R} vertices



Fig. 8.6. Railroad proof for the commutativity of transfer matrices: The intertwiner is pulled through from right to left by a successive application of the YBE

terchangeability of the two limits $(L, N \to \infty)$ and the existence of a gap between the largest and the next-largest eigenvalues of T^{QTM} for finite temperature [36,37].

The next-leading eigenvalues give the exponential correlation lengths ξ of the equal time correlators at finite temperature

$$\frac{1}{\xi} = \lim_{N \to \infty} \ln \left| \frac{\Lambda_{\max}}{\Lambda} \right|.$$
(8.20)

Lastly we want to comment on the study of thermodynamics of the quantum chain in the presence of an external magnetic field h coupling to the spin $S = \sum_{j=1}^{L} S_j$, where S_j denotes a certain component of the *j*th spin, for instance S_j^z . This, of course, changes (8.15) only trivially

$$Z_{L,N} := \operatorname{Tr} \left\{ \left[T(-\tau)\overline{T}(-\tau) \right]^{N/2} \cdot e^{\beta hS} \right\}.$$
(8.21)

On the lattice, the equivalent two-dimensional model is modified in a simple way by a horizontal seam. Each vertical bond of this seam carries an individual Boltzmann weight $e^{\pm\beta h/2}$ if $S_j = \pm 1/2$ which indeed describes the action of the operator

$$e^{\beta hS} = \prod_{j=1}^{L} e^{\beta hS_j}.$$
(8.22)

Consequently, the QTM is modified by an h dependent boundary condition. It is essential that these modifications can still be treated exactly as the additional operators acting on the bonds belong to the group symmetries of the model.

We like to close this section with some notes on the relation of the two apparently different approaches, the combinatorial TBA and the operatorbased QTM. In fact, these methods are not at all independent! In the latter approach there are several quite different ways of analysis of the eigenvalues of the QTM. In the standard (and most economical) way, see below, a set of just two coupled non-linear integral equations (NLIE) is derived [34, 35]. Alternatively, an approach based on the "fusion hierarchy" leads to a set of (generically) infinitely many NLIEs [34, 38] that are identical to the TBA equations though completely different reasoning has been applied!

Very recently [26], yet another formulation of the thermodynamics of the Heisenberg chain has been developed. At the heart of this formulation is just one NLIE with a structure very different from that of the two sets of NLIEs discussed so far. Nevertheless, this new equation has been derived from the "old" NLIEs [26, 39] and is certainly an equivalent formulation. In the first applications of the new NLIE, numerical calculations of the free energy have been performed with excellent agreement with the older TBA and QTM results. Also, analytical high temperature expansions up to order 100 have been carried out on this basis.

8.4 Be the Ansatz Equations for the Spin-1/2 XXZ Chain

In the following we consider the anisotropic Heisenberg chain (slightly generalizing (8.3)) with Hamiltonian H_L

$$H = 2\sum_{j=1}^{L} [S_j^x S_{j+1}^x + S_j^y S_{j+1}^y + \Delta S_j^z S_{j+1}^z]$$
(8.23)

with periodic boundary conditions on a chain of length L. Apparently, for $\Delta = +1$ the system specializes to the isotropic antiferromagnetic Heisenberg chain, for $\Delta = -1$ (and applying a simple unitary transformation) the system reduces to the isotropic ferromagnetic case.

The classical counterpart of the XXZ chain is the six-vertex model (Fig. 7). For our purposes the following parameterization of the Boltzmann weights is useful

$$a(w) = 1, \quad b(w) = \frac{\sin(\gamma w/2)}{\sin(\gamma w/2 + \gamma)}, \quad c(w) = \frac{\sin\gamma}{\sin(\gamma w/2 + \gamma)}.$$
 (8.24)

All relations between Hamiltonian of the quantum system, row-to-row trans-



Fig. 8.7. Allowed vertices of the six-vertex models with corresponding Boltzmann weights a, b, c. The remaining vertices are obtained by an exchange of bond-states $1 \leftrightarrow 2$

fer matrix of the classical model and the QTM as introduced in the previous section are also valid in the present case if Δ and γ are related by $\Delta = \cos \gamma$. The only modification concerns (8.9) as here the relation of Hamiltonian (8.23) and the logarithmic derivative of the row-to-row transfer matrix for the weights (8.24) aquires a normalization factor

$$H = 2 \frac{\sin \gamma}{\gamma} \frac{d}{du} \ln T(u) \big|_{u=0}.$$
(8.25)

Monodromy Matrix

Our aim is to diagonalize the row-to-row transfer matrix and the QTM by

means of the algebraic Bethe ansatz. We first review some notation and basic properties of *R*-matrices (as collections of local Boltzmann weights) and the so-called *L*-matrix. The elements of the *L*-matrix at site *j* are operators acting in the local Hilbert space h_j (for the six-vertex model isomorphic to $\simeq \mathbb{C}^2$). The *L*-matrix' element in row α and column β is given in terms of the *R* matrix

$$L_{j\beta}^{\ \alpha}(w) = R_{\beta\nu}^{\alpha\mu}(w)e_{j\nu}^{\ \mu}, \tag{8.26}$$

where e^{μ}_{ν} is a matrix with only non-vanishing entry 1 in row μ and column ν . This reads explicitly for the six-vertex model

$$L_i = \begin{pmatrix} \frac{a+b}{2} + \frac{a-b}{2}\sigma_i^z & c\sigma_i^+ \\ c\sigma_i^- & \frac{a+b}{2} - \frac{a-b}{2}\sigma_i^z \end{pmatrix}$$
(8.27)

with a, b, c given in (8.24).¹ The so-called monodromy-matrix is defined as a product of all *L*-matrices on consecutive sites

$$\mathcal{T} = L_1 \cdots L_N = \begin{pmatrix} A & B \\ C & D \end{pmatrix}.$$
 (8.28)

In essence, the (α, β) -element of the monodromy matrix is the transfer matrix of a system with fixed boundary spins α on the left end and β on the right end of a row.

Algebraic Bethe Ansatz

The procedure of diagonalization can be decribed as follows:

- First we search for a pseudo-vacuum state (reference state) $|\Omega\rangle$ that is a simple eigenstate of the operator-valued diagonal entries A and D of the monodromy matrix \mathcal{T} (and hence an eigenstate of the transfer matrix T = A + D). The lower-left entry C of the monodromy matrix applied to $|\Omega\rangle$ yields zero, the upper-right entry B yields new non-vanishing states. Hence C and B play the role of annihilation and creation operators.
- From the Yang-Baxter equations a quadratic algebra of commutation relations for the entries of the monodromy matrix (notably for A, D, and B) is derived. By use of these relations algebraic expressions of the eigenstates and rather explicit expressions for the eigenvalues are derived.

It turns out that the row-to-row transfer matrix and the QTM can be treated in parallel as the quadratic algebra of the entries of the monodromy matrix is identical for both cases. The only difference lies in the different reference states and the different "vacuum expectation values". We therefore

¹ As a reminder: $\sigma^+ = \sigma^x + i\sigma^y$ and $\sigma^- = \sigma^x - i\sigma^y$, and states $|+\rangle$, $|-\rangle$ correspond to $|1\rangle$, $|2\rangle$.

focus our presentation on the slightly simpler case of the row-to-row transfer matrix and describe the necessary modifications for the QTM in (8.42,8.43).

Reference State

Provided we find local states $|\omega_i\rangle$ that are eigenstates of the diagonal entries of L_i and are annihilated by the left-lower entry, then $|\Omega\rangle$ may be taken as the product of these local states. A glance to (8.27) shows that this is possible just with $|\omega_i\rangle = |2\rangle_i$

$$|\Omega\rangle = \bigotimes_{i}^{N} |2\rangle_{i} \,. \tag{8.29}$$

The monodromy matrix ${\cal T}$ applied to $|\varOmega\rangle$ yields an upper triangular 2×2 matrix of states

$$\mathcal{T} |\Omega\rangle = \begin{pmatrix} a^N |\Omega\rangle & B |\Omega\rangle \\ 0 & b^N |\Omega\rangle \end{pmatrix}$$
(8.30)

or explicitly

$$A |\Omega\rangle = a^N |\Omega\rangle, \quad D |\Omega\rangle = b^N |\Omega\rangle, \quad T |\Omega\rangle = (a^N + b^N) |\Omega\rangle.$$
 (8.31)

Therefore, $|\Omega\rangle$ is an eigenstate of T.

Quadratic Algebra of Operators A, B, and D

We intend to use the operator B as creation operator for excitations, i.e. we demand that the new state $|\Omega_1(v)\rangle := B(v) |\Omega\rangle$ ("one-particle state") be an eigenstate of T(u) = A(u) + D(u). What we need to know is the operator algebra for interchanging B(v) with A(u) and D(u). This algebra can be obtained from the YBE, for a graphical representation see Fig. 8.8. By fixing the exterior spins on the horizontal bonds we obtain all the commutators we need for interchanging the operators. We begin with the relation of any two B-operators illustrated in Fig. 8.9 actually implying commutation

$$[B(u), B(v)] = 0 \tag{8.32}$$

Now we look at products of A and B and again use a graphical representation shown in Fig. 8.10. We algebraically find

$$A(u)B(v) = \frac{a(v-u)}{b(v-u)}B(v)A(u) - \frac{c(v-u)}{b(v-u)}B(u)A(v).$$
(8.33)



Fig. 8.8. Graphical depiction of the YBE



Fig. 8.9. Quadratic relation of *B*-operators obtained from fixing the spins on the left and right open bonds in Fig. 8.8. Note that the summation over the spin variables on the closed bonds between R and \mathcal{T} matrices reduces to just one non-vanishing term on each side of the equation



Fig. 8.10. Bilinear relation of B and A-operators obtained from fixing the spins on the left and right open bonds in Fig. 8.8. Note that the summation over the spin variables on the closed bonds between R and \mathcal{T} matrices reduces to two non-vanishing terms on the left side of the equation and to one non-vanishing term on the right side of the equation



Fig. 8.11. Bilinear relation of B and D-operators obtained in analogy to the above cases

In a similar same way, illustrated in Fig. 8.11, we obtain (after interchanging $u \leftrightarrow v$)

$$D(u)B(v) = \frac{a(u-v)}{b(u-v)}B(v)D(u) - \frac{c(u-v)}{b(u-v)}B(u)D(v).$$
(8.34)

Now we have available all necessary relations for evaluating the application of T(u) to $|\Omega_1\rangle$. By use of the commutation relations we get

$$T(u) |\Omega_1(v)\rangle = \left[\alpha(u)\frac{a(v-u)}{b(v-u)} + \beta(u)\frac{a(u-v)}{b(u-v)}\right] |\Omega_1(v)\rangle - \left[\alpha(v)\frac{c(v-u)}{b(v-u)} + \beta(v)\frac{c(u-v)}{b(u-v)}\right] B(u) |\Omega\rangle, \quad (8.35)$$

where we have used the abbreviations $\alpha = a^N$, $\beta = b^N$. The terms in the first (second) line of the r.h.s. of (8.35) are called "wanted terms" ("unwanted terms") and arise due to the first (second) terms on the r.h.s. of (8.33,8.34).

In order that $|\Omega_1(v)\rangle$ be an eigenstate of the transfer matrix the second term in (8.35) has to vanish and the first one to give the eigenvalue²

$$\frac{\alpha(v)}{\beta(v)} = -\frac{c(u-v)b(v-u)}{c(v-u)b(u-v)} = 1,$$
(8.36)

$$\Lambda(u) = \alpha(u) \frac{a(v-u)}{b(v-u)} + \beta(u) \frac{a(u-v)}{b(u-v)}.$$
(8.37)

We can generalize this to any n-particle state. The argument is quite the same as for just one excitation. We look at the following state

$$|\Omega(v_i)\rangle = \prod_{i=1}^{n} B(v_i) |\Omega\rangle$$
(8.38)

where the numbers v_i will be referred to as Bethe ansatz roots. Demanding that this state be an eigenstate of T(u) we get after successive application of the commutation rules the following set of equations. From the two "wanted terms" the eigenvalue is read off

$$\Lambda(u) = \alpha(u) \prod_{j=1}^{n} \frac{a(v_j - u)}{b(v_j - u)} + \beta(u) \prod_{j=1}^{n} \frac{a(u - v_j)}{b(u - v_j)},$$
(8.39)

and vanishing of the "unwanted terms" yields

$$\frac{\alpha(v_i)}{\beta(v_i)} = \prod_{j(\neq i)}^n \frac{b(v_j - v_i)}{b(v_i - v_j)}, \quad \text{for } i = 1, ..., n.$$
(8.40)

The last constraints are nothing but the famous Bethe ansatz equations. We like to note that we would have obtained the same set of equations by demanding that the function on the r.h.s. of (8.39) be analytic in the whole complex plane

Res
$$\Lambda(u = v_i) = 0 \quad \forall i.$$
 (8.41)

In the case of a general transfer matrix $T\left(v; \{u_k^{(1)}\}; \{u_k^{(2)}\}\right)$ defined as a product of N_1 many vertices of type R and N_2 many vertices of type \tilde{R} with spectral parameter v in the auxiliary space (corresponding to the continuous line) and $u_k^{(1)}$, $u_k^{(2)}$ in the quantum spaces the eigenvalue expression is very similar to (8.39) and (8.40). The only change is the replacement

$$\alpha(u) = e^{+\beta h/2} \prod_{k=1}^{N_1} a(u - u_k^{(1)}) \prod_{k=1}^{N_2} b(u_k^{(2)} - u), \qquad (8.42)$$

$$\beta(u) = e^{-\beta h/2} \prod_{k=1}^{N_1} b(u - u_k^{(1)}) \prod_{k=1}^{N_2} a(u_k^{(2)} - u), \qquad (8.43)$$

² For the sake of transparency of the algebraic structure we do not use the fact that by our convention the Boltzmann weight a is identical to 1.

where we have also introduced $\exp(\pm\beta h/2)$ factors arising from twisted boundary conditions for the transfer matrix as realized in the case of the quantum transfer matrix for a system in a magnetic field h.

We may simplify the following presentation if we perform a rotation by $\pi/2$ in the complex plane, i.e. we introduce a function λ by $\lambda(v) = \Lambda(iv)$ and for convenience we replace $v_j \to iv_j$. The eigenvalue expression for any eigenvalue $\lambda(v)$ of the transfer matrix T reads (see also [22])

$$\lambda(v) = \alpha(\mathrm{i}v) \prod_{j=1}^{n} \frac{a(\mathrm{i}v_j - \mathrm{i}v)}{b(\mathrm{i}v_j - \mathrm{i}v)} + \beta(\mathrm{i}v) \prod_{j=1}^{n} \frac{a(\mathrm{i}v - \mathrm{i}v_j)}{b(\mathrm{i}v - \mathrm{i}v_j)}.$$
(8.44)

Of particular importance is the case of the quantum transfer matrix with $N_1 = N_2 = N/2$ and all $u_k^{(1)} = \tau$, $u_k^{(2)} = -\tau$ with $\tau = 2 \frac{\sin \gamma}{\gamma} \frac{\beta}{N}$, note the normalization factor in (8.25).

Introducing the definition

$$s(v) := \sinh(\gamma v/2), \tag{8.45}$$

we obtain for the functions b, α, β

$$b(iv) = \frac{s(v)}{s(v-2i)},$$

$$\alpha(iv) = e^{\beta h/2} \left[\frac{s(v-i\tau)}{s(v-i\tau+2i)} \right]^{N/2}, \quad \beta(iv) = e^{-\beta h/2} \left[\frac{s(v+i\tau)}{s(v+i\tau-2i)} \right]^{N/2} (8.46)$$

From (8.44) we obtain for the function $\lambda(v)$

$$\lambda(v) = \frac{\lambda_1(v) + \lambda_2(v)}{[s(v - i(2 - \tau))s(v + i(2 - \tau))]^{N/2}},$$
(8.47)

where the terms $\lambda_{1,2}(v)$ are

$$\lambda_{1}(v) := e^{+\beta h/2} \phi(v-i) \frac{q(v+2i)}{q(v)}, \lambda_{2}(v) := e^{-\beta h/2} \phi(v+i) \frac{q(v-2i)}{q(v)},$$
(8.48)

and $\phi(v)$ is simply

$$\phi(v) := \left[s(v - i(1 - \tau))s(v + i(1 - \tau))\right]^{N/2}.$$
(8.49)

The function $q(\boldsymbol{v})$ is defined in terms of the yet to be determined Bethe ansatz roots v_j

$$q(v) := \prod_{j} s(v - v_j).$$
(8.50)

Note that we are mostly interested in Λ which is obtained from $\lambda(v)$ simply by setting v = 0. Nevertheless, we are led to the study of the full v-dependence since the condition fixing the values of v_j is the analyticity of $\lambda_1(v) + \lambda_2(v)$ in the complex plane. This yields

$$a(v_j) = -1,$$
 (8.51)

where the function a(v) (not to be confused with the Boltzmann weight a(w) above) is defined by

$$a(v) := \frac{\lambda_1(v)}{\lambda_2(v)} = e^{\beta h} \frac{\phi(v-i)q(v+2i)}{\phi(v+i)q(v-2i)}.$$
(8.52)

Algebraically, we are dealing with a set of coupled non-linear equations similar to those occurring in the study of the eigenvalues of the Hamiltonian [22]. Analytically, there is a profound difference as here in (8.52) the ratio of ϕ -functions possesses zeros and poles converging to the real axis in the limit $N \to \infty$. As a consequence, the distribution of Bethe ansatz roots is *discrete* and shows an *accumulation point* at the origin, cf. Fig. 8.12. Hence the treatment of the problem by means of linear integral equations for continuous density functions [40] is not possible in contrast to the Hamiltonian case.



Fig. 8.12. Sketch of the distribution of Bethe ansatz roots v_j for finite N. Note that the distribution remains discrete in the limit of $N \to \infty$ for which the origin turns into an accumulation point

8.5 Manipulation of the Bethe Ansatz Equations

The eigenvalue expression (8.47) under the subsidiary condition (8.51) has to be evaluated in the limit $N \to \infty$. This limit is difficult to take as an increasing number N/2 of Bethe ansatz roots v_j has to be determined. In the Hamiltonian case, i.e. for (8.42) with $N_1 = N$ and $N_2 = 0$ (or vice versa), the distribution of roots is continuous and the Bethe ansatz equations (8.40) can be reduced to linear integral equations [22,40]. For the general case, this is no longer possible. In this case, notably for the QTM, the distribution of the roots is discrete and the standard approach based on continuous density functions is not possible. From now on we explicitly discuss the QTM case, i.e. (8.42) with $N_1 = N_2 = N/2$ yielding all information on the free energy at arbitrary temperature T including the limit T = 0. (With some modifications, the final non-linear integral equations also apply in the case of the row-torow transfer matrix and the Hamiltonian. These results, however, are not of prime interest to this review.)

8.5.1 Derivation of Non-linear Integral Equations

The main idea of our treatment is the derivation of a set of integral equations for the function a(v). This function possesses zeros and poles related to the Bethe ansatz roots v_j , see Fig. 8.13. Next we define the associated auxiliary function A(v) by

$$A(v) = 1 + a(v). \tag{8.53}$$

The poles of A(v) are identical to those of a(v). However, the set of zeros is different. From (8.51) we find that the Bethe ansatz roots are zeros of A(v)(depicted by open circles in Fig. 8.14). There are additional zeros farther away from the real axis with imaginary parts close to ± 2 . For the sake of completeness, these zeros are depicted in Fig. 8.14 (open squares), but for a while they are not essential to our reasoning. Next we are going to formulate a linear integral expression for the function $\log a(v)$ in terms of $\log A(v)$. To this end we consider the function

$$f(v) := \frac{1}{2\pi i} \int_{\mathcal{L}} \frac{d}{dv} \log s(v-w) \log A(w) dw$$
(8.54)



Fig. 8.13. Distribution of zeros (\circ) and poles (\times) of the auxiliary function a(v). All zeros and poles $v_j \mp 2i$ are of first order, the zeros and poles at $\pm(2i - i\tau)$, $\pm i\tau$ are of order N/2



Fig. 8.14. Distribution of zeros and poles of the auxiliary function A(v) = 1 + a(v). Note that the positions of zeros (\circ) and poles (\times) are directly related to those occurring in the function a(v). There are additional zeros (\Box) above and below the real axis. The closed contour \mathcal{L} by definition surrounds the real axis and the zeros (\circ) as well as the pole at $-i\tau$

defined by an integral with closed contour \mathcal{L} surrounding the real axis, the parameters v_j and the point $-i\tau$ in anticlockwise manner, see Fig. 8.14. Note that the number of zeros of A(v) surrounded by this contour is N/2 and hence is identical to the order of the pole at $-i\tau$. Therefore the integrand $\log A(w)$ does not show any non-zero winding number on the contour and consequentially the integral is well-defined. By use of standard theorems we see that the function f(v) is analytic in the complex plane away from the real axis (and axes with imaginary parts being integer multiples of $2\pi/\gamma$). Next we perform an integration by parts and apply Cauchy's theorem

$$f(v) = \sum_{j} \log s(v - v_j) - N/2 \log s(v + i\tau) = \log \frac{q(v)}{s(v + i\tau)^{N/2}}, \quad (8.55)$$

Thanks to (8.54) and (8.55) we have a linear integral representation of $\log q(v)$ in terms of $\log A(v)$. Because of (8.52) the function $\log a(v)$ is a linear combination of $\log q$ and explicitly known functions leading to

$$\log a(v) = \beta h + \log \left(\frac{s(v - i\tau)s(v + 2i + i\tau)}{s(v + i\tau)s(v + 2i - i\tau)}\right)^{N/2} + f(v + 2i) - f(v - 2i).$$
(8.56)

From now on we use a shorthand notation for the logarithmic derivative of s(v)

$$d(v) := \frac{d}{dv} \log s(v) = \frac{\gamma}{2} \coth \frac{\gamma}{2} v.$$
(8.57)

By use of the definition of the integration kernel \varkappa

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$$\varkappa(u) := \frac{1}{2\pi i} \frac{d}{du} \log \frac{s(u-2i)}{s(u+2i)} = \frac{1}{2\pi i} [d(u-2i) - d(u+2i)] = \frac{\gamma}{2\pi} \frac{\sin 2\gamma}{\cosh \gamma u - \cos 2\gamma},$$
(8.58)

we may write (8.56) as

$$\log a(v) = \beta h + \frac{N}{2} \log \left(\frac{s(v - i\tau)s(v + 2i + i\tau)}{s(v + i\tau)s(v + 2i - i\tau)} \right) - \int_{\mathcal{L}} \varkappa(v - w) \log A(w) dw.$$
(8.59)

This expression for a(v) is remarkable as it is a non-linear integral equation (NLIE) of convolution type. It is valid for any value of the Trotter number N which only enters in the driving (first) term on the r.h.s. of (8.59). This term shows a well-defined limiting behaviour for $N \to \infty$

$$\frac{N}{2}\log\left(\frac{s(v-\mathrm{i}\tau)s(v+2\mathrm{i}+\mathrm{i}\tau)}{s(v+\mathrm{i}\tau)s(v+2\mathrm{i}-\mathrm{i}\tau)}\right) \to \mathrm{i}N\tau\left[\frac{d}{dv}\log s(v+2\mathrm{i}) - \frac{d}{dv}\log s(v)\right] \\ = \mathrm{i}\beta 2\frac{\sin\gamma}{\gamma}[d(v+2\mathrm{i}) - d(v)], \quad (8.60)$$

leading to a well-defined NLIE for a(v) even in the limit $N \to \infty$

$$\log a(v) = \beta h + \beta \epsilon_0(v+i) - \int_{\mathcal{L}} \varkappa(v-w) \log A(w) dw,$$
(8.61)

where ϵ_0 is defined by

$$\epsilon_0(v) = \mathbf{i}[d(v+\mathbf{i}) - d(v-\mathbf{i})] = 2\frac{\sin^2\gamma}{\cosh\gamma v - \cos\gamma}.$$
(8.62)

This NLIE allows for a numerical (and in some limiting cases also analytical) calculation of the function a(v) on the axes $\Im(v) = \pm 1$. About the historical development we like to note that NLIEs very similar to (8.61) were derived for the row-to-row transfer matrix in [41, 42]. These equations were then generalized to the related cases of staggered transfer matrices (QTMs) of the Heisenberg and RSOS chains [34, 35] and the sine-Gordon model [43].

8.5.2 Integral Expressions for the Eigenvalue

In (8.59) and (8.61) we have found integral equations determining the function a for finite and infinite Trotter number N, respectively. The remaining problem is the derivation of an expression for the eigenvalue λ in terms of aor A.

From (8.47) we see that $\lambda(v)$ is an analytic function with periodicity in imaginary direction (period $2\pi i/\gamma$) and exponential asymptotics along the real axis. Hence, up to a multiplicative constant C, we can write 8 Integrability of Quantum Chains, the Spin-1/2 XXZ Chain 369

$$\lambda_1(v) + \lambda_2(v) = C \prod_l s(v - w_l), \qquad (8.63)$$

where w_l , l = 1, ..., N, are the zeros of $\lambda(v) = \lambda_1(v) + \lambda_2(v)$ which are solutions to $a(v) = \lambda_1(v)/\lambda_2(v) = -1$, i.e. zeros of A(v) = 1 + a(v) that do not coincide with Bethe ansatz (BA) roots! These zeros are so-called hole-type solutions to the BA equations. The holes are located in the complex plane close to the axes with imaginary parts ± 2 , see zeros in Fig. 8.14 depicted by \Box . Thanks to Cauchy's theorem we find for v in the neighbourhood of the real axis

$$\frac{1}{2\pi i} \int_{\mathcal{L}} d(v - w - 2i) [\log A(w)]' dw = \sum_{j} d(v - v_j - 2i) - \frac{N}{2} d(v + i\tau - 2i)$$
(8.64)

as the only singularities of the integrand surrounded by the contour \mathcal{L} are the simple zeros v_j and the pole $-i\tau$ of order N/2 of the function A. Also, for v in the neighbourhood of the real axis we obtain

$$\frac{1}{2\pi i} \int_{\mathcal{L}} d(v-w) [\log A(w)]' dw = \sum_{j} d(v-v_{j}-2i) - \sum_{l} d(v-w_{l}) + \frac{N}{2} d(v+2i-i\tau), \qquad (8.65)$$

where the evaluation of the integral has been done by use of the singularities outside of the contour \mathcal{L} and use of the period $2\pi i/\gamma$ of the integrand. The old contour \mathcal{L} is replaced by a contour $\widetilde{\mathcal{L}}$ such that the upper (lower) part of $\widetilde{\mathcal{L}}$ is the lower part of \mathcal{L} (the upper part of \mathcal{L} shifted by $-2\pi i/\gamma$), and reversed orientation. The surrounded singularities are the simple poles $v_j + 2i - 2\pi i/\gamma$, the zeros w_l with or without shift $-2\pi i/\gamma$, and the pole $i\tau - 2i$ of order N/2of the function A.

Next, we take the difference of (8.64) and (8.65), perform an integration by parts with respect to w, and finally integrate with respect to v

$$\frac{1}{2\pi i} \int_{\mathcal{L}} [d(v-w) - d(v-w-2i)] \log A(w) dw$$

= $\log \frac{[s(v-i(2-\tau))s(v+i(2-\tau))]^{N/2}}{\prod_l s(v-w_l)} + \text{const.}$ (8.66)

Combining (8.63), (8.66) and (8.47) we find

$$\log \lambda(v) = -\beta h/2 - \frac{1}{2\pi i} \int_{\mathcal{L}} \left[d(v-w) - d(v-w-2i) \right] \log A(w) dw, \quad (8.67)$$

where the constant was determined from the asymptotic behaviour for $v \to \infty$ and use of $\lambda(\infty) = \exp(\beta h/2) + \exp(-\beta h/2)$ and $A(\infty) = 1 + \exp(\beta h)$.

These formulas, (8.67) and (8.61), are the basis of an efficient analytical and numerical treatment of the thermodynamics of the Heisenberg chain. There are, however, variants of these integral equations that are somewhat more convenient for the analysis, especially for magnetic fields close to 0. The alternative integral expression for Λ reads [34, 35]

$$\ln \Lambda = -\beta e_0 + \int_{-\infty}^{\infty} K(x) \ln[\mathfrak{A}(x)\overline{\mathfrak{A}}(x)] dx, \quad K(x) = \frac{1}{4\cosh\frac{\pi}{2}x}, \quad (8.68)$$

where $\mathfrak{A}(x)$ and $\overline{\mathfrak{A}}(x)$ are complex-valued functions with integration paths along the real axis. These functions are determined from the following set of non-linear integral equations

$$\ln \mathfrak{a}(x) = -\beta \frac{\sin \gamma}{\gamma} \frac{\pi}{\cosh \frac{\pi}{2}x} + \frac{\pi\beta h}{2(\pi - \gamma)} + \kappa * \ln \mathfrak{A}(x) - \kappa * \ln \overline{\mathfrak{A}}(x + 2i),$$
(8.69)

$$\ln \overline{\mathfrak{a}}(x) = -\beta \frac{\sin \gamma}{\gamma} \frac{\pi}{\cosh \frac{\pi}{2}x} - \frac{\pi\beta h}{2(\pi-\gamma)} + \kappa * \ln \overline{\mathfrak{A}}(x) - \kappa * \ln \mathfrak{A}(x-2i),$$
(8.70)

$$\mathfrak{A}(x) = 1 + \mathfrak{a}(x), \qquad \overline{\mathfrak{A}}(x) = 1 + \overline{\mathfrak{a}}(x).$$
(8.71)

The symbol * denotes the convolution $f * g(x) = \int_{-\infty}^{\infty} f(x-y)g(y)dy$ and the function $\kappa(x)$ is defined by

$$\kappa(x) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \frac{\sinh\left(\frac{\pi}{\gamma} - 2\right)k}{2\cosh k \sinh\left(\frac{\pi}{\gamma} - 1\right)k} e^{ikx} dk.$$
(8.72)

Note that the integrals in (8.69) and (8.70) are well-defined with integration paths just below and above the real axis.

The above equations are obtained from (8.61) by a partial "particle-hole" transformation of the function a(v) only on the axis $\Im(v) = -1$. Replacing $\log A = \log \overline{A} + \log a$ (where $\overline{a} = 1/a$, $\overline{A} = 1 + \overline{a}$) on the lower part of \mathcal{L} in (8.61) leads to an equation involving convolution type integrals with $\log A$, $\log \overline{A}$ and $\log a$. This equation can be resolved explicitly for $\log a$ by straightforward calculations in "momentum space". Finally, $\mathfrak{a}(x) := a(x + i)$ and $\overline{\mathfrak{a}}(x) := \overline{a}(x - i)$.

8.6 Numerical Results for Thermodynamical Quantities

By numerical integration and iteration the integral equation (8.61) can be solved on the axes $\Im(v) = \pm 1$ defining functions $a^{\pm}(x) := a(x \pm i)$. Alternatively and particularly convenient for the case of vanishing magnetic field h, equations (8.69) and (8.70) can be used for the functions **a** and $\bar{\mathbf{a}}$. Choosing appropriate initial functions the series a_k^{\pm} with $k = 0, 1, 2, \ldots$ converges rapidly. In practice only a few steps are necessary to reach high-precision results. Moreover, using the well-known Fast Fourier Transform algorithm we can compute the convolutions very efficiently. In fact, some of the convolutions in (8.61) or (8.69,8.70) are delicate to be evaluated in "real space", because of the appearance of a pole of the kernel just at the integration contour. These integrals are automatically handled correctly in "momentum space".

In order to calculate derivatives of the thermodynamical potential with respect to temperature T and magnetic field h one can avoid numerical differentiations by utilizing similar integral equations guaranteeing the same numerical accuracy as for the free energy. The idea is as follows. Consider the function

$$la_{\beta} := \frac{\partial}{\partial \beta} \log a \quad \text{with} \quad \frac{\partial}{\partial \beta} \log(1+a) = \frac{1}{1+a} \frac{\partial a}{\partial \beta} = \frac{a}{1+a} la_{\beta},$$

we directly obtain from (8.61) a *linear* integral equation for la_{β} if we regard the function a as given. Once the integral equation (8.61) is solved for a, the integral equation for la_{β} associated with (8.61) can be solved. In this manner, we may continue to any order of derivatives with respect to T (and h). However, in practice only the first and second orders matter. Here we restrict our treatment to the specific heat c(T) and the magnetic susceptibility $\chi(T)$ (derivatives of second order with respect to T and h), see Figs. 8.15 and 8.16.

Note the characteristic behaviour of c(T) and $\chi(T)$ at low temperatures. The linear behaviour of c(T) and the finite ground-state limit of $\chi(T)$ are manifestations of the linear energy-momentum dispersion of the low-lying excitations (spinons) of the isotropic antiferromagnetic Heisenberg chain. Also, with increase of the repulsion Δ the location of the finite temperature ma-



Fig. 8.15. Specific heat c(T) data versus temperature T for the spin-1/2 XXZ chain with repulsive interaction $0 \le \Delta \le 1$



Fig. 8.16. Susceptibility $\chi(T)$ data versus temperature T for the spin-1/2 XXZ chain with repulsive interaction $0 \le \Delta \le 1$

ximum in c(T) shifts to higher temperatures and the values of $\chi(T)$ drop. In the high temperature limit the asymptotics of c(T) and $\chi(T)$ are $1/T^2$ and 1/T. This and the existence of the finite temperature maximum are a consequence of the finite dimensional local degree of freedom, i.e. the spin per lattice site.

For $\Delta = 1$ note that $\chi(T)$ approaches the ground-state limit $\chi(0)$ in a singular manner, see also Fig. 8.17. The numerical data at extremely low temperatures provide evidence of logarithmic correction terms, see also [44] and later lattice studies [45, 46] confirming the field theoretical treatment by [47]. These logarithmic terms are responsible for the infinite slope of $\chi(T)$ at T = 0 despite the finite ground-state value $\chi(0) = 1/\pi^2$. Precursors of such strong slopes have been seen in experiments down to relatively low temperatures, see e.g. [48]. Unfortunately, most quasi one-dimensional quantum spin systems undergo a phase transition at sufficiently low temperatures driven by residual higher dimensional interactions. Hence the onset of quantum critical phenomena of the Heisenberg chain at T = 0 becomes visible, but cannot be identified beyond all doubts.

8.7 Thermal Transport

The simplest approach to the investigation of transport properties is based on linear response theory leading to the Kubo formulas [49,50] relating conductivities to dynamical correlation functions of local current operators. Hence, the calculation of transport properties is more difficult than the computation of the free energy. In fact, the calculation of correlation functions is a most difficult issue and exact results are rare, even for integrable systems. In some situations, however, the explicit computation of correlation functions can be avoided. For the spin transport of the Heisenberg chain (\equiv electrical



Fig. 8.17. Magnetic susceptibility χ at low temperature T for the isotropic spin-1/2 XXX chain. In the inset $\chi(T)$ is shown on a larger temperature scale

transport of the model in the particle representation) the Drude weight of the dynamical conductivity can be cast into a form involving only spectral properties without explicit recourse to matrix elements. At the time of writing, these expressions are known how to be evaluated for zero temperature, the case of non-zero temperature is still controversial.

A fortuitous case is the thermal transport as the thermal current $\mathcal{J}_{\rm E}$ is one of the conserved currents $\mathcal{J}^{(n)}$ (8.10). The first three conserved currents (n = 0, 1, 2) are related to the momentum operator, the Hamiltonian and the thermal current via

$$P = -i\mathcal{J}^{(0)}$$

$$H = 2\frac{\sin\gamma}{\gamma}\mathcal{J}^{(1)} - \frac{L}{2}\Delta,$$

$$\mathcal{J}_{E} = i\left(2\frac{\sin\gamma}{\gamma}\right)^{2}\mathcal{J}^{(2)} + iL.$$
(8.74)

Note that the spin current (electrical current) is not conserved as it is not contained in the sequence of conserved currents $\mathcal{J}^{(n)}$!

Let us first motivate that $\mathcal{J}^{(2)}$ is indeed the thermal current $\mathcal{J}_{\rm E}$ of the system. To this end we impose the continuity equation relating the time derivative of the local Hamiltonian (interaction) and the divergence of the current: $\dot{h} = -\text{div } j^{\rm E}$. The time evolution of the local Hamiltonian $h_{k,k+1}$ is obtained from the commutator with the total Hamiltonian and the divergence of the local current on the lattice is given by a difference expression

$$\frac{\partial h_{kk+1}(t)}{\partial t} = \mathbf{i}[H, h_{kk+1}(t)] = -\{j_{k+1}^{\mathbf{E}}(t) - j_{k}^{\mathbf{E}}(t)\}.$$
(8.75)

Apparently the last equation is satisfied with a local thermal current operator $j_k^{\rm E}$ defined by

$$j_k^{\rm E} = i[h_{k-1k}, h_{kk+1}].$$
 (8.76)

In fact, up to a trivial scale and shift, the conserved current $\mathcal{J}^{(2)}$ is identical to the r.h.s. of the upper equation, see also [51–56].

The Kubo formulas [49, 50] are obtained within linear response theory and yield the (thermal) conductivity κ relating the (thermal) current $\mathcal{J}_{\rm E}$ to the (temperature) gradient ∇T

$$\mathcal{J}_{\rm E} = \kappa \nabla T, \tag{8.77}$$

where

$$\kappa(\omega) = \beta \int_0^\infty dt \mathrm{e}^{-\mathrm{i}\omega t} \int_0^\beta d\tau \langle \mathcal{J}_\mathrm{E}(-t - \mathrm{i}\tau) \mathcal{J}_\mathrm{E} \rangle.$$
(8.78)

As the total thermal current operator $\mathcal{J}_{\rm E}$ commutes with the Hamiltonian H of the XXZ chain we find

$$\kappa(\omega) = \frac{1}{i(\omega - i\epsilon)} \beta^2 \langle \mathcal{J}_E^2 \rangle, \qquad (\epsilon \to 0+), \tag{8.79}$$

with $\Re \kappa(\omega) = \tilde{\kappa} \delta(\omega)$ where

$$\tilde{\kappa} = \pi \beta^2 \langle \mathcal{J}_{\rm E}^2 \rangle. \tag{8.80}$$

As a consequence, the thermal conductivity at zero frequency is infinite! This is only natural, as the conserved current cannot decay in time. However, the weight of the zero-frequency peak is some finite and non-trivial temperature dependent quantity to be calculated from the second moment of the thermal current.

Quite generally, the expectation values of conserved quantities may be calculated by use of a suitable generating function

$$Z = \text{Tr } \exp(-\beta \mathcal{H} + \lambda \mathcal{J}_{\text{E}}), \qquad (8.81)$$

from which we find the expectation values by derivatives with respect to λ at $\lambda=0$

$$\frac{\partial}{\partial\lambda}\ln Z\Big|_{\lambda=0} = \langle \mathcal{J}_{\rm E} \rangle = 0, \qquad \left(\frac{\partial}{\partial\lambda}\right)^2\ln Z\Big|_{\lambda=0} = \langle \mathcal{J}_{\rm E}^2 \rangle - \langle \mathcal{J}_{\rm E} \rangle^2 = \langle \mathcal{J}_{\rm E}^2 \rangle, \tag{8.82}$$

where we have used that the expectation value of the thermal current in thermodynamical equilibrium is zero. Instead of ${\cal Z}$ we will find it slightly more convenient to work with a partition function

$$\overline{Z} = \operatorname{Tr} \exp(-\lambda_1 \mathcal{J}^{(1)} - \lambda_n \mathcal{J}^{(n)}), \qquad (8.83)$$

notably n = 2. With view to (8.74) we choose

$$\lambda_1 = \beta \left(2 \frac{\sin \gamma}{\gamma} \right), \qquad \lambda_2 = -i\lambda \left(2 \frac{\sin \gamma}{\gamma} \right)^2,$$
(8.84)

and obtain the desired expectation values from \overline{Z}

$$\langle \mathcal{J}_{\rm E}^2 \rangle = \left(\frac{\partial}{\partial \lambda}\right)^2 \ln \overline{Z}\Big|_{\lambda=0}.$$
 (8.85)

We can deal with \overline{Z} rather easily. Consider the trace of a product of N row-to-row transfer matrices $T(u_j)$ with some spectral parameters u_j close to zero, but still to be specified, and the Nth power of the inverse of T(0)

$$Z_N = \operatorname{Tr} \left[T(u_1) \cdot \dots \cdot T(u_N) \cdot T(0)^{-N} \right]$$

= Tr exp $\left(\sum_j [\ln T(u_j) - \ln T(0)] \right).$ (8.86)

Now it is a standard exercise in arithmetic to devise a sequence of N numbers $u_1, ..., u_N$ (actually $u_j = u_j^{(N)}$) such that

$$\lim_{N \to \infty} \sum_{j} [f(u_j) - f(0)] = -\lambda_1 \frac{\partial}{\partial u} f(u) \Big|_{u=0} - \lambda_n \left(\frac{\partial}{\partial u}\right)^n f(u) \Big|_{u=0}.$$
 (8.87)

We only need the existence of such a sequence of numbers, the precise values are actually not important. In the limit $N \to \infty$ we note

$$\lim_{N \to \infty} Z_N = \overline{Z}.$$
(8.88)

We can proceed along the established path of the quantum transfer matrix (QTM) formalism presented above and derive the partition function Z_N in the thermodynamic limit $L \to \infty$

$$\lim_{L \to \infty} Z_N^{1/L} = \Lambda, \tag{8.89}$$

where Λ is the largest eigenvalue of the QTM. The integral expression for Λ reads

$$\ln \Lambda = \sum_{j} [e(u_j) - e(0)] + \int_{-\infty}^{\infty} K(x) \ln[\mathfrak{A}(x)\overline{\mathfrak{A}}(x)] dx, \qquad (8.90)$$

with K(x) already defined in (8.68) and some function e(x) given in [34,35]. In the limit $N \to \infty$ the first term on the r.h.s. of the last equation turns into

$$\lim_{N \to \infty} \sum_{j} [e(u_j) - e(0)] = -\lambda_1 \frac{\partial}{\partial u} e(u) \Big|_{u=0} - \lambda_n \left(\frac{\partial}{\partial u}\right)^n e(u) \Big|_{u=0}, \quad (8.91)$$

a rather irrelevant term as it is linear in λ_1 and λ_n , and therefore the second derivatives with respect to λ_1 and λ_n vanish. The functions $\mathfrak{A}(x)$ and $\overline{\mathfrak{A}}(x)$ are determined from the following set of non-linear integral equations

$$\ln \mathfrak{a}(x) = \sum_{j} [\varepsilon_0(x - iu_j) - \varepsilon_0(0)] + \kappa * \ln \mathfrak{A}(x) - \kappa * \ln \overline{\mathfrak{A}}(x + 2i),$$

$$\ln \overline{\mathfrak{a}}(x) = \sum_{j} [\varepsilon_0(x - iu_j) - \varepsilon_0(0)] + \kappa * \ln \overline{\mathfrak{A}}(x) - \kappa * \ln \mathfrak{A}(x - 2i),$$

$$\mathfrak{A}(x) = 1 + \mathfrak{a}(x), \qquad \overline{\mathfrak{A}}(x) = 1 + \overline{\mathfrak{a}}(x).$$
(8.92)

with a function $\varepsilon_0(x)$ given in terms of hyperbolic functions [34, 35]. Again, the summations in (8.92) can be simplified in the limit $N \to \infty$

$$\lim_{N \to \infty} \sum_{j} [\varepsilon_0(x - iu_j) - \varepsilon_0(x)] = -\lambda_1 \underbrace{\left(-i\frac{\partial}{\partial x}\right)\varepsilon_0(x)}_{=:\varepsilon_1(x)} -\lambda_n \underbrace{\left(-i\frac{\partial}{\partial x}\right)^n \varepsilon_0(x)}_{=:\varepsilon_n(x)}.$$
(8.93)

where the first function can be found in [34, 35] and is simply

$$\varepsilon_1(x) = 2\pi K(x) = \frac{\pi}{2\cosh\frac{\pi}{2}x},\tag{8.94}$$

and hence the second function is

$$\varepsilon_n(x) = \left(-i\frac{\partial}{\partial x}\right)^{n-1} \varepsilon_1(x).$$
(8.95)

We like to note that the structure of the driving term (8.93) appearing in the NLIE (8.92) reflects the structure of the generalized Hamiltonian in the exponent on the r.h.s. of (8.83). We could have given an alternative derivation of the NLIE along the lines of the thermodynamic Bethe ansatz (TBA). In such an approach the driving term is typically the one-particle energy corresponding to the generalized Hamiltonian. Hence it has contributions due to the first as well as the *n*th logarithmic derivative of the row-to-row transfer matrix, i.e. the terms ε_1 and ε_n .

In Fig. 8.18 we show $\tilde{\kappa}(T)$ for various anisotropy parameters γ . Note that $\tilde{\kappa}(T)$ has linear T dependence at low temperatures. At high temperatures $\tilde{\kappa}(T)$ behaves like $1/T^2$.



Fig. 8.18. Illustration of numerical results for the thermal conductivity $\tilde{\kappa}$ as a function of temperature T for various anisotropy parameters $\Delta = \cos \gamma$ with $\gamma = 0, \pi/6, \pi/5, \pi/4, \pi/3, \pi/2$

8.8 Summary

We have reviewed the treatment of integrable quantum systems at zero and finite temperature based on a lattice path integral formulation and the definition of the so-called quantum transfer matrix (QTM). In detail, the thermodynamical properties of the partially anisotropic Heisenberg chain were discussed. As we hope, a transparent analysis of the eigenvalue problem of the row-to-row and quantum transfer matrices has been given, resulting in a set of non-linear integral equations (NLIE). From a numerical solution of these NLIEs at arbitrary temperature the specific heat and magnetic susceptibility data were obtained. Also the Drude weight of the thermal current was calculated. Although we have given the explicit results only for the case of vanishing magnetic field (h = 0), the derivation of the free energy is not restricted to this case. In fact, the NLIEs given above are valid even in the case of non-vanishing fields h. However, the above treatment of the thermal conductivity is limited to the case h = 0. The generalization to arbitrary fields is an open problem as is practically any physical question involving the explicit knowledge of correlation functions at zero and even more at finite temperature.

The purpose of writing this review was to convince the reader that research on integrable quantum systems is an attractive and rewarding field of science. Hopefully, this goal has been reached and the reader has gained from this presentation enough insight to begin or continue his or her own investigations.

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9 Quantum Phases and Phase Transitions of Mott Insulators

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Abstract. This article contains a theoretical overview of the physical properties of antiferromagnetic Mott insulators in spatial dimensions greater than one. Many such materials have been experimentally studied in the past decade and a half, and we make contact with these studies. Mott insulators in the simplest class have an even number of S = 1/2 spins per unit cell, and these can be described with quantitative accuracy by the bond operator method: we discuss their spin gap and magnetically ordered states, and the transitions between them driven by pressure or an applied magnetic field. The case of an odd number of S = 1/2 spins per unit cell is more subtle: here the spin gap state can spontaneously develop bond order (so the ground state again has an even number of S = 1/2 spins per unit cell). and/or acquire topological order and fractionalized excitations. We describe the conditions under which such spin gap states can form, and survey recent theories of the quantum phase transitions among these states and magnetically ordered states. We describe the breakdown of the Landau-Ginzburg-Wilson paradigm at these quantum critical points, accompanied by the appearance of emergent gauge excitations.

9.1 Introduction

The physics of Mott insulators in two and higher dimensions has enjoyed much attention since the discovery of cuprate superconductors. While a quantitative synthesis of theory and experiment in the superconducting materials remains elusive, much progress has been made in describing a number of antiferromagnetic Mott insulators. A number of such insulators have been studied extensively in the past decade, with a few prominent examples being CaV_4O_9 [1], $(C_5H_{12}N_2)_2Cu_2Cl_4$ [2–4], $SrCu_2(BO_3)_2$ [5, 6], $TlCuCl_3$ [7–10], and Cs_2CuCl_4 [11, 12]. In some cases, it has even been possible to tune these insulators across quantum phase transitions by applied pressure [8] or by an applied magnetic field [3,4,7,9]. A useful survey of some of these experiments may be found in the recent article by Matsumoto *et al.* [10].

It would clearly be valuable to understand the structure of the global phase diagram of antiferromagnetic Mott insulators above one dimension. The compounds mentioned above would then correspond to distinct points in this phase diagram, and placing them in this manner should help us better understand the relationship between different materials. One could also classify the quantum critical points accessed by the pressure or field-tuning experiments. The purpose of this article is to review recent theoretical work towards achieving this goal. We will focus mainly on the case of two spatial dimensions (d), but our methods and results often have simple generalizations to d = 3.

One useful vantage point for opening this discussion is the family of Mott insulators with a gap to all spin excitations. All spin gap compounds discovered to date have the important property of being "dimerized", or more precisely, they have an even number of S = 1/2 spins per unit cell [13]. In such cases, the spin gap can be understood by adiabatic continuation from the simple limiting case in which the spins form local spin singlets within each unit cell. A simple approach that can be used for a theoretical description of such insulators is the method of bond operators [14,15]. This method has been widely applied, and in some cases provides an accurate quantitative description of numerical studies and experiments [10, 16]. We will describe it here in Sect. 9.2 in the very simple context of a coupled dimer antiferromagnet; similar results are obtained in more complicated, and realistic, lattice structures. Sect. 9.2 will also describe the quantum phase transition(s) accessed by varying coupling constants in the Hamiltonian while maintaining spin rotation invariance (this corresponds to experiments in applied pressure): the spin gap closes at a quantum critical point beyond which there is magnetic order. Section 9.2.3 will discuss some of the important experimental consequences of this quantum criticality at finite temperatures. A distinct quantum critical point, belonging to a different universality class, is obtained when the spin gap is closed by an applied magnetic field—this is described in Sect. 9.3.

The remaining sections discuss the theoretically much more interesting and subtle cases of materials with an odd number of S = 1/2 spins per unit cell, such as La₂CuO₄ and Cs₂CuCl₄. A complementary, but compatible, perspective on the physics of such antiferromagnets may be found in the review article by Misguich and Lhuillier [17]. Antiferromagnets in this class can develop a spin gap by *spontaneously* breaking the lattice symmetry so that the lattice is effectively dimerized (see discussion in the following paragraph). There are no known materials with a spin gap in which the lattice symmetry has not been broken, but there is a theoretical consensus that spin gap states without lattice symmetry breaking are indeed possible in d > 1 [18]. The study of antiferromagnets with an odd number of S = 1/2 spins per unit cell is also important for the physics of the doped cuprates. These materials exhibit spin-gap-like behavior at low dopings, and many theories associate aspects of its physics with the spin gap state proximate to the magnetically ordered state of the square lattice antiferromagnet found in La₂CuO₄.

Section 9.4 will describe the nature of a spin gap state on the square lattice. We begin with the nearest-neighbor S = 1/2 Heisenberg Hamiltonian on the square lattice—this is known to have a magnetic Néel order which breaks spin rotation invariance. Now add further neighbor exchange couplings until magnetic order is lost and a spin gap appears. We will show that the ground state undergoes a novel, second-order quantum phase transition to a state with *bond order*: translational symmetry is spontaneously broken [19, 20] so that the resulting lattice structure has an even number of S = 1/2 spins per unit cell. So aspects of the non-zero spin excitations in this paramagnet are very similar to the "dimerized" systems considered in Sect. 9.2, and experimentally they will appear to be almost identical. Indeed, it may well be that the experimental materials initially placed in the class of Sect. 9.2, are secretely systems in the class of Sect. 9.4 which have developed bond order driven by the physics of antiferromagets (as in Sect. 9.4.1) at some intermediate energy scale. The host lattice then distorts sympathetically to the bond order, and is effectively dimerized. Such materials will possess many more low-lying singlet excitations than those in the theory of Sect. 9.2: these excitations play an important role in the restoration of translational symmetry as we move towards the Néel state. Unfortunately, such singlet excitations are rather difficult to detect experimentally.

Section 9.5 will address the same issue as Sect. 9.4, but for the case of the triangular lattice. Here the spins are ordered in a non-collinear configuration in the magnetically ordered state, as is observed at low temperatures in Cs_2CuCl_4 [11, 12]. We will argue that in this case there is a route to destruction of magnetic order in which the resulting spin gap state preserves full lattice symmetry [21, 22]. Such a spin gap state has a novel 'topological' order [23] which endows its excitations with charges under an emergent gauge force. Recent experimental measurements of the dynamic structure factor of Cs_2CuCl_4 appear to be described rather well by the excitations of this topologically ordered state at energies above which the magnetic order of the ground state emerges [12, 24].

9.2 Coupled Dimer Antiferromagnet

We begin by describing the quantum phase transition in a simple twodimensional model of antiferromagnetically coupled S = 1/2 Heisenberg spins which has 2 spins per unit cell. The transition is tuned by varying a dimensionless parameter λ . As we noted in Sect. 9.1 different 'dimerized' Mott insulators will correspond to different values of λ , and the value of λ can be tuned by applying pressure [8, 10].

We consider the "coupled dimer" Hamiltonian [25]

$$H_d = J \sum_{\langle ij \rangle \in \mathcal{A}} \mathbf{S}_i \cdot \mathbf{S}_j + \lambda J \sum_{\langle ij \rangle \in \mathcal{B}} \mathbf{S}_i \cdot \mathbf{S}_j, \qquad (9.1)$$

where \mathbf{S}_j are spin-1/2 operators on the sites of the coupled-ladder lattice shown in Fig. 9.1, with the \mathcal{A} links forming decoupled dimers while the \mathcal{B} links couple the dimers as shown. The ground state of H_d depends only on



Fig. 9.1. The coupled dimer antiferromagnet. Spins (S = 1/2) are placed on the sites, the \mathcal{A} links are shown as full lines, and the \mathcal{B} links as dashed lines.

the dimensionless coupling λ , and we will describe the low temperature (T) properties as a function of λ . We will restrict our attention to J > 0 and $0 \le \lambda \le 1$.

Note that exactly at $\lambda = 1$, H_d is identical to the square lattice antiferromagnet, and this is the only point at which the Hamiltonian has only one spin per unit cell. At all other values of λ H_d has a pair of S = 1/2 spins in each unit cell of the lattice. As will become clear from our discussion, this is a key characteristic which permits a simple theory for the quantum phase transition exhibited by H_d . Models with only a single S = 1/2 spin per unit cell usually display far more complicated behavior, and will be discussed in Sects. 9.4,9.5.

We will begin with a physical discussion of the phases and excitations of the coupled dimer antiferromagnet, H_d in Sect. 9.2.1. We will propose a quantum field-theoretical description of this model in Sect. 9.2.2: we will verify that the limiting regimes of the field theory contain excitations whose quantum numbers are in accord with the phases discussed in Sect. 9.2.1, and will then use the field theory to describe the quantum critical behavior both at zero and finite temperatures.

9.2.1 Phases and Their Excitations

Let us first consider the case where λ is close to 1. Exactly at $\lambda = 1$, H_d is identical to the square lattice Heisenberg antiferromagnet, and this is known to have long-range, magnetic Néel phase in its ground state *i.e.* the spin-rotation symmetry is broken and the spins have a non-zero, staggered, expectation value in the ground state with

$$\langle \mathbf{S}_j \rangle = \eta_j N_0 \mathbf{n},\tag{9.2}$$

where **n** is some fixed unit vector in spin space, η_j is ± 1 on the two sublattices, and N_0 is the Néel order parameter. This long-range order is expected to be preserved for a finite range of λ close to 1. The low-lying excitations above the ground state consist of slow spatial deformations in the orientation **n**: these are the familiar spin waves, and they can carry arbitrarily low energy *i.e.* the phase is 'gapless'. The spectrum of the spin waves can be


Fig. 9.2. Schematic of the quantum paramagnet ground state for small λ . The ovals represent singlet valence bond pairs.



Fig. 9.3. (a) Cartoon picture of the bosonic S = 1 excitation of the paramagnet. (b) Fission of the S = 1 excitation into two S = 1/2 spinons. The spinons are connected by a "string" of valence bonds (denoted by dashed ovals) which lie on weaker bonds; this string costs a finite energy per unit length and leads to the confinement of spinons.

obtained from a text-book analysis of small fluctuations about the ordered Néel state using the Holstein-Primakoff method [26]: such an analysis yields *two* polarizations of spin waves at each wavevector $k = (k_x, k_y)$ (measured from the antiferromagnetic ordering wavevector), and they have excitation energy $\varepsilon_k = (c_x^2 k_x^2 + c_y^2 k_y^2)^{1/2}$, with c_x, c_y the spin-wave velocities in the two spatial directions.

Let us turn now to the vicinity of $\lambda = 0$. Exactly at $\lambda = 0$, H_d is the Hamiltonian of a set of decoupled dimers, with the simple exact ground state wavefunction shown in Fig. 9.2: the spins in each dimer pair into valence bond singlets, leading to a paramagnetic state which preserves spin rotation invariance and all lattice symmetries. Excitations are now formed by breaking a valence bond, which leads to a *three*-fold degenerate state with total spin S = 1, as shown in Fig. 9.3a. At $\lambda = 0$, this broken bond is localized, but at finite λ it can hop from site-to-site, leading to a triplet quasiparticle excitation. Note that this quasiparticle is *not* a spin-wave (or equivalently, a 'magnon') but is more properly referred to as a spin 1 *exciton* or a *triplon* [27]. We parameterize its energy at small wavevectors k (measured from the minimum of the spectrum in the Brillouin zone) by



Fig. 9.4. Ground states of H_d as a function of λ The quantum critical point is at [28] $\lambda_c = 0.52337(3)$. The compound TlCuCl₃ undergoes a similar quantum phase transition under applied pressure [8].

$$\varepsilon_k = \Delta + \frac{c_x^2 k_x^2 + c_y^2 k_y^2}{2\Delta},\tag{9.3}$$

where Δ is the spin gap, and c_x , c_y are velocities; we will provide an explicit derivation of (9.3) in Sect. 9.2.2. Figure 9.3 also presents a simple argument which shows that the S = 1 exciton cannot fission into two S = 1/2 'spinons'.

The very distinct symmetry signatures of the ground states and excitations between $\lambda \approx 1$ and $\lambda \approx 0$ make it clear that the two limits cannot be continuously connected. It is known that there is an intermediate secondorder phase transition at [25,28] $\lambda = \lambda_c = 0.52337(3)$ between these states as shown in Fig. 9.4. Both the spin gap Δ and the Néel order parameter N_0 vanish continuously as λ_c is approached from either side.

9.2.2 Bond Operators and Quantum Field Theory

In this section we will develop a continuum description of the low energy excitations in the vicinity of the critical point postulated above. There are a number of ways to obtain the same final theory: here we will use the method of *bond operators* [14,15], which has the advantage of making the connection to the lattice degrees of freedom most direct. We rewrite the Hamiltonian using bosonic operators which reside on the centers of the \mathcal{A} links so that it is explicitly diagonal at $\lambda = 0$. There are 4 states on each \mathcal{A} link $(|\uparrow\uparrow\rangle, |\uparrow\downarrow\rangle, |\downarrow\uparrow\rangle$, and $|\downarrow\downarrow\rangle$) and we associate these with the canonical singlet boson *s* and the canonical triplet bosons t_{α} ($\alpha = x, y, z$) so that

$$|s\rangle \equiv s^{\dagger}|0\rangle = \frac{1}{\sqrt{2}} \left(|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle\right) \quad ; \quad |t_x\rangle \equiv t_x^{\dagger}|0\rangle = \frac{-1}{\sqrt{2}} \left(|\uparrow\uparrow\rangle - |\downarrow\downarrow\rangle\right) \quad ; \quad$$

$$|t_y\rangle \equiv t_y^{\dagger}|0\rangle = \frac{i}{\sqrt{2}} \left(|\uparrow\uparrow\rangle + |\downarrow\downarrow\rangle\right) \quad ; \quad |t_z\rangle \equiv t_z^{\dagger}|0\rangle = \frac{1}{\sqrt{2}} \left(|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle\right).(9.4)$$

Here $|0\rangle$ is some reference vacuum state which does not correspond to a physical state of the spin system. The physical states always have a single bond boson and so satisfy the constraint

$$s^{\dagger}s + t^{\dagger}_{\alpha}t_{\alpha} = 1. \tag{9.5}$$

By considering the various matrix elements $\langle s|\mathbf{S}_1|t_{\alpha}\rangle$, $\langle s|\mathbf{S}_2|t_{\alpha}\rangle$, ..., of the spin operators $\mathbf{S}_{1,2}$ on the ends of the link, it follows that the action of \mathbf{S}_1 and \mathbf{S}_2 on the singlet and triplet states is equivalent to the operator identities

$$S_{1\alpha} = \frac{1}{2} \left(s^{\dagger} t_{\alpha} + t_{\alpha}^{\dagger} s - i \epsilon_{\alpha\beta\gamma} t_{\beta}^{\dagger} t_{\gamma} \right),$$

$$S_{2\alpha} = \frac{1}{2} \left(-s^{\dagger} t_{\alpha} - t_{\alpha}^{\dagger} s - i \epsilon_{\alpha\beta\gamma} t_{\beta}^{\dagger} t_{\gamma} \right),$$
(9.6)

where α, β, γ take the values x, y, z, repeated indices are summed over and ϵ is the totally antisymmetric tensor. Inserting (9.6) into (9.1), and using (9.5), we find the following Hamiltonian for the bond bosons:

$$\begin{aligned} H_{d} &= H_{0} + H_{1} \\ H_{0} &= J \sum_{\ell \in \mathcal{A}} \left(-\frac{3}{4} s_{\ell}^{\dagger} s_{\ell} + \frac{1}{4} t_{\ell \alpha}^{\dagger} t_{\ell \alpha} \right) \\ H_{1} &= \lambda J \sum_{\ell, m \in \mathcal{A}} \left[a(\ell, m) \left(t_{\ell \alpha}^{\dagger} t_{m \alpha} s_{m}^{\dagger} s_{\ell} + t_{\ell \alpha}^{\dagger} t_{m \alpha}^{\dagger} s_{m} s_{\ell} + \text{H.c.} \right) + b(\ell, m) \right. \\ &\times \left(i \epsilon_{\alpha \beta \gamma} t_{m \alpha}^{\dagger} t_{\ell \beta}^{\dagger} t_{\ell \gamma} s_{m} + \text{H.c.} \right) + c(\ell, m) \left(t_{\ell \alpha}^{\dagger} t_{m \alpha}^{\dagger} t_{m \beta} t_{\ell \beta} - t_{\ell \alpha}^{\dagger} t_{m \beta}^{\dagger} t_{m \alpha} t_{\ell \beta} \right) \right] 9.7) \end{aligned}$$

where ℓ , *m* label links in \mathcal{A} , and *a*, *b*, *c* are numbers associated with the lattice couplings which we will not write out explicitly. Note that $H_1 = 0$ at $\lambda = 0$, and so the spectrum of the paramagnetic state is fully and exactly determined. The main advantage of the present approach is that application of the standard methods of many body theory to (9.7), while imposing the constraint (9.5), gives a very satisfactory description of the phases with $\lambda \neq 0$, including across the transition to the Néel state. In particular, an important feature of the bond operator approach is that the simplest mean field theory already yields ground states and excitations with the correct quantum numbers; so a strong fluctuation analysis is not needed to capture the proper physics.

A complete numerical analysis of the properties of (9.7) in a self-consistent Hartree-Fock treatment of the four boson terms in H_1 has been presented in [14]. In all phases the s boson is well condensed at zero momentum, and the important physics can be easily understood by examining the structure of the low energy action for the t_{α} bosons. For the particular Hamiltonian (9.1), the spectrum of the t_{α} bosons has a minimum at the momentum $(0, \pi)$, and for large enough λ the t_{α} condense at this wavevector: the representation (9.6) shows that this condensed state is the expected Néel state, with the magnetic moment oscillating as in (9.2). The condensation transition of the t_{α} is therefore the quantum phase transition between the paramagnetic and Néel phases of the coupled dimer antiferromagnet. In the vicinity of this critical point, we can expand the t_{α} bose field in gradients away from the $(0, \pi)$ wavevector: so we parameterize

$$t_{\ell,\alpha}(\tau) = t_{\alpha}(r_{\ell},\tau)e^{i(0,\pi)\cdot\boldsymbol{r}_{\ell}}$$
(9.8)

where τ is imaginary time, $r \equiv (x, y)$ is a continuum spatial coordinate, and expand the effective action in spatial gradients. In this manner we obtain

$$S_{t} = \int d^{2}r d\tau \left[t_{\alpha}^{\dagger} \frac{\partial t_{\alpha}}{\partial \tau} + C t_{\alpha}^{\dagger} t_{\alpha} - \frac{D}{2} \left(t_{\alpha} t_{\alpha} + \text{H.c.} \right) + K_{1x} |\partial_{x} t_{\alpha}|^{2} + K_{1y} |\partial_{y} t_{\alpha}|^{2} \right. \\ \left. + \frac{1}{2} \left(K_{2x} (\partial_{x} t_{\alpha})^{2} + K_{2y} (\partial_{y} t_{\alpha})^{2} + \text{H.c.} \right) + \cdots \right].$$
(9.9)

Here $C, D, K_{1,2x,y}$ are constants that are determined by the solution of the self-consistent equations, and the ellipses represent terms quartic in the t_{α} . The action S_t can be easily diagonalized, and we obtain a S = 1 quasiparticle excitation with the spectrum

$$\varepsilon_k = \left[\left(C + K_{1x}k_x^2 + K_{1y}k_y^2 \right)^2 - \left(D + K_{2x}k_x^2 + K_{2y}k_y^2 \right)^2 \right]^{1/2}.$$
 (9.10)

This is, of course, the triplon (or spin exciton) excitation of the paramagnetic phase postulated earlier in (9.3); the latter result is obtained by expanding (9.10) in momenta, with $\Delta = \sqrt{C^2 - D^2}$. This value of Δ shows that the ground state is paramagnetic as long as C > D, and the quantum critical point to the Néel state is at C = D.

The critical point and the Néel state are more conveniently described by an alternative formulation of S_t (although an analysis using bond operators directly is also possible [29]). It is useful to decompose the complex field t_{α} into its real and imaginary parts as follows

$$t_{\alpha} = Z(\varphi_{\alpha} + i\pi_{\alpha}), \tag{9.11}$$

where Z is a normalization chosen below. Insertion of (9.11) into (9.9) shows that the field π_{α} has a quadratic term $\sim (C + D)\pi_{\alpha}^2$, and so the coefficient of π_{α}^2 remains large even as the spin gap Δ becomes small. Consequently, we can safely integrate π_{α} out, and the resulting action for φ_{α} takes the form

$$S_{\varphi} = \int d^2 r d\tau \left[\frac{1}{2} \left\{ \left(\partial_{\tau} \varphi_{\alpha} \right)^2 + c_x^2 \left(\partial_x \varphi_{\alpha} \right)^2 + c_y^2 \left(\partial_y \varphi_{\alpha} \right)^2 + s \varphi_{\alpha}^2 \right\} + \frac{u}{24} \left(\varphi_{\alpha}^2 \right)^2 \right]. \tag{9.12}$$

Here we have chosen Z to fix the coefficient of the temporal gradient term, and $s = C^2 - D^2$.

The formulation S_{φ} makes it simple to explore the physics in the region s < 0. It is clear that the effective potential of φ_{α} has a minimum at a non-zero φ_{α} , and that $\langle \varphi_{\alpha} \rangle \propto N_0$, the Néel order parameter in (9.2). It is simple to carry out a small fluctuation analysis about this saddle point, and we obtain the doublet of gapless spin-wave modes advertised earlier.

We close this subsection by noting that all of the above results have a direct generalization to other lattices, and also to spin systems in three dimensions. Matsumoto *et al.* [10] have applied the bond operator method to TlCuCl₃ and obtained good agreement with experimental observations. One important difference that emerges in such calculations on some frustrated lattices [30] is worth noting explicitly here: the minimum of the t_{α} spectrum need not be at special wavevector like $(0, \pi)$, but can be at a more generic wavevector \boldsymbol{Q} such that \boldsymbol{Q} and $-\boldsymbol{Q}$ are not separated by a reciprocal lattice vector. A simple example which we consider here is an extension of (9.1) in which there are additional exchange interactions along all diagonal bonds oriented 'north-east' (so that the lattice has the connectivity of a triangular lattice). In such cases, the structure of the low energy action is different, as is the nature of the magnetically ordered state. The parameterization (9.8) must be replaced by

$$t_{\ell\alpha}(\tau) = t_{1\alpha}(r_{\ell},\tau)e^{i\boldsymbol{Q}\cdot\boldsymbol{r}_{\ell}} + t_{2\alpha}(r_{\ell},\tau)e^{-i\boldsymbol{Q}\cdot\boldsymbol{r}_{\ell}}, \qquad (9.13)$$

where $t_{1,2\alpha}$ are independent complex fields. Proceeding as above, we find that the low energy effective action (9.12) is replaced by

$$\mathcal{S}_{\Phi} = \int d^2 r d\tau \bigg[\left| \partial_{\tau} \Phi_{\alpha} \right|^2 + c_x^2 \left| \partial_x \Phi_{\alpha} \right|^2 + c_y^2 \left| \partial_y \Phi_{\alpha} \right|^2 + s \left| \Phi_{\alpha} \right|^2 + \frac{u}{2} \left(\left| \Phi_{\alpha} \right|^2 \right)^2 + \frac{v}{2} \left| \Phi_{\alpha}^2 \right|^2 \bigg], \qquad (9.14)$$

where now Φ_{α} is a *complex* field such that $\langle \Phi_{\alpha} \rangle \sim \langle t_{1\alpha} \rangle \sim \langle t_{2\alpha}^{\dagger} \rangle$. Notice that there is now a second quartic term with coefficient v. If v > 0, configurations with $\Phi_{\alpha}^2 = 0$ are preferred: in such configurations $\Phi_{\alpha} = n_{1\alpha} + in_{2\alpha}$, where $n_{1,2\alpha}$ are two equal-length orthogonal vectors. Then from (9.13) and (9.6) it is easy to see that the physical spins possess *spiral* order in the magnetically ordered state in which Φ_{α} is condensed. A spiral state is illustrated in Fig. 9.13, and we will have more to say about this state in Sect. 9.5. For the case v < 0, the optimum configuration has $\Phi_{\alpha} = n_{\alpha}e^{i\theta}$ where n_{α} is a real vector: this leads to a magnetically ordered state with spins polarized *collinearly* in a spin density wave at the wavevector Q.

9.2.3 Quantum Criticality

We will restrict our discussion here to the critical point described by S_{φ} . Similar results apply to S_{Φ} for the parameter regime in which it exhibits a second order transition [31]. Experimentally, the results below are relevant to materials that can be tuned across the magnetic ordering transition by applied pressure (such as TlCuCl₃ [8]), or to materials which happen to be near a critical point at ambient pressure (such as LaCuO_{2.5} [32]).

The field theory S_{φ} is actually a familiar and well-studied model in the context of classical critical phenomena. Upon interpreting τ as a third spatial coordinate, S_{φ} becomes the theory of a classical O(3)-invariant Heisenberg ferromagnet at finite temperatures (in general a *d* dimensional quantum antiferromagnet will map to a *d*+1 dimensional classical Heisenberg ferromagnet at finite temperature [33]). The Curie transition of the Heisenberg ferromagnet then maps onto the quantum critical point between the paramagnetic and Néel states described above. A number of important implications for the quantum problem can now be drawn immediately.

The theory S_{φ} has a 'relativistic' invariance, and consequently the dynamic critical exponent must be z = 1. The spin correlation length will diverge at the quantum critical point with the exponent [34] $\nu = 0.7048(30)$. The spin gap of the paramagnet, Δ , vanishes as $\Delta \sim (\lambda_c - \lambda)^{z\nu}$, and this prediction is in excellent agreement with the numerical study of the dimerized antiferromagnet [28].

A somewhat more non-trivial consequence of this mapping is in the structure of the spectrum at the critical point $\lambda = \lambda_c$. At the Curie transition of the classical ferromagnet it is known [35] that spin correlations decay as $\sim 1/p^{2-\eta}$, where p is the 3-component momentum in the 3-dimensional classical space. We can now analytically continue this expression from its p_z dependence in the third classical dimension to the real frequency, ω , describing the quantum antiferromagnet. This yields the following fundamental result for the dynamic spin susceptibility, $\chi(k,\omega)$, at the T = 0 quantum critical point of the coupled-dimer antiferromagnet:

$$\chi(k,\omega) \sim \frac{1}{\left(c_x^2 k_x^2 + c_y^2 k_y^2 - (\omega + i\epsilon)^2\right)^{1-\eta/2}},\tag{9.15}$$

where ϵ is a positive infinitesimal. Note that in (9.15) the momentum k is measured from the (π, π) ordering wavevector of the Néel state. The exponent η is the same as that of the classical Heisenberg ferromagnet, and has a rather small value [34]: $\eta \approx 0.03$. However, the non-zero η does make a significant difference to the physical interpretation of the excitations at the critical point. In particular note that $\text{Im}\chi(k,\omega)$ does not have a pole at any k, but rather a continuum spectral weight above a threshold energy [36,37]

$$\operatorname{Im}\chi(k,\omega) \sim \operatorname{sgn}(\omega) \sin\left(\frac{\pi\eta}{2}\right) \frac{\theta\left(|\omega| - \sqrt{c_x^2 k_x^2 + c_y^2 k_y^2}\right)}{\left(\omega^2 - c_x^2 k_x^2 - c_y^2 k_y^2\right)^{1-\eta/2}},\tag{9.16}$$

where θ is the unit step function. This indicates there are no quasiparticles at the critical point, and only a dissipative critical continuum.

There is also some very interesting structure in the quantum critical dynamic response at nonzero T [36, 37]. Here, one way to understand the physics is to approach the critical point from the paramagnetic side ($\lambda < \lambda_c$). As we noted earlier, the paramagnetic phase has well-defined 'triplon' or 'spin exciton' excitations t_{α} , and these have an infinite lifetime at T = 0. At T > 0, thermally excited t_{α} quasiparticles will collide with each other via their scattering amplitude, u, and this will lead to a finite lifetime [37, 38]. Now approach $\lambda = \lambda_c$. The renormalization group analysis of \mathcal{S}_{ω} tells us that the quartic coupling u approaches a fixed point value in the critical region. This means that u is no longer an arbitrary parameter, and an appropriately defined t_{α} scattering amplitude must also acquire universal behavior. In particular, the t_{α} lifetime is determined by the only energy scale available, which is $k_B T$. So we have the remarkable result that the characteristic spin relaxation time is a universal number times $\hbar/(k_B T)$. More precisely, we can write for the local dynamic spin susceptibility $\chi_L(\omega) = \int d^2k \chi(k,\omega)$ the universal scaling form

$$\operatorname{Im}\chi_L(\omega) = T^{\eta}F\left(\frac{\hbar\omega}{k_BT}\right).$$
(9.17)

Here F is a universal function which has the limiting behaviors

$$F(\overline{\omega}) \sim \begin{cases} \overline{\omega} &, \, |\overline{\omega}| \ll 1\\ \operatorname{sgn}(\omega) |\overline{\omega}|^{\eta} , \, |\overline{\omega}| \gg 1 \end{cases}$$
(9.18)

Note that F has a smooth linear behavior in the regime $|\hbar\omega| \ll k_B T$, and this is similar to any simple dissipative system. The difference here is that the coefficient of dissipation is determined by $k_B T$ alone.

The quantum critical behavior described here is expected to apply more generally to other correlated electron systems, provided the critical theory has non-linear couplings which approach fixed point values.

9.3 Influence of an Applied Magnetic Field

An important perturbation that can be easily applied to antiferromagnets in the class discussed in Sect. 9.2 is a uniform magnetic field. The Zeeman energy in available fields can often be comparable to the typical antiferromagnetic exchange constant J, and so the ground state can be perturbed significantly. It is therefore of interest to understand the evolution of the phase diagram in Fig. 9.4 under an applied field of arbitrary strength. We are interested here in the evolution of the ground state as a function of B where the Hamiltonian H_d in (9.1) is transformed as

$$H_d \to H_d - \sum_j \mathbf{B} \cdot \mathbf{S}_j.$$
 (9.19)

Most of the basic features can actually be understood quite easily in a simple extension of the self-consistent Hartree-Fock theory of bond bosons that was discussed in Sect. 9.2.2. Under the transformation (9.19), it is easily seen from (9.6) that

$$H_d \to H_d + iB_\alpha \sum_{\ell \in \mathcal{A}} \epsilon_{\alpha\beta\gamma} t^{\dagger}_{\ell\beta} t_{\ell\gamma}.$$
 (9.20)

The presence of a non-zero B breaks spin rotation invariance and so all the self-consistent expectation values of operator bilinears have to reflect this reduced symmetry in the Hartree-Fock theory. Apart from this the mechanics of the computation mostly remain the same. However, for stronger fields, it is sometimes necessary to allow for broken translational symmetry in the expectation values, as the ground state can acquire a modulated structure.

We will discuss the results of such an analysis in weak and strong fields in the following subsections.

9.3.1 Weak Fields

For weak fields applied to the paramagnet (specifically, for fields $B < \Delta$, the influence of (9.20) can be understood exactly. The coupling to B involves an operator which commutes with the remaining Hamiltonian (the total spin), and hence the wavefunction of the ground state remains insensitive to the value of B. The same applies to the wavefunctions of the excited states. However, the excited states can have non-zero total spin and so their energies do depend upon B. In particular the triplet t_{α} quasiparticle with energy (9.3) or (9.10) carries total spin S = 1, and consequently we conclude that this triplet splits according to

$$\varepsilon_k \to \varepsilon_k - mB$$
 (9.21)

with $m = 0, \pm 1$. Note that the lowest energy quasiparticle (with m = 1) has a positive energy as long as $B < \Delta$, and this is required for the stability of the paramagnet. So the phase boundary of the paramagnetic phase is exactly $B = \Delta$, and using $\Delta \sim (\lambda_c - \lambda)^{z\nu}$, we can sketch the boundary of the paramagnetic phase as in Fig. 9.5.

What happens beyond the paramagnetic phase? As in Sect. 9.2.2, we answer this question by using the transformation (9.11), and by examining the analog of S_{φ} under a non-zero *B*. Using (9.20), and integrating out π_{α} , we now find that the action S_{φ} in (9.12) remains unchanged apart from the mapping [40]



Fig. 9.5. Evolution of the phases of Fig. 9.4 under a weak field *B* (magnetization plateau at large *B*, appearing in Fig. 9.6, are not shown). The paramagnetic phase has exactly the same ground state wavefunction as that at B = 0. The phase boundary behaves like $B \sim (\lambda_c - \lambda)^{z\nu}$. The *B* field is oriented vertically upwards, and the static moments in the canted phase can rotate uniformly about the vertical axis. The phase boundary at non-zero *B* is described by the z = 2 dilute Bose gas quantum critical theory. The phase diagram of TlCuCl₃ in applied pressure and magnetic field looks similar to the one above [10]. The corresponding phase diagram of the field-induced magnetic ordering transition of a superconductor (rather than a Mott insulator) has been investigated recently [39], and successfully applied to experiments on the doped cuprates; this phase diagram of the superconductor has significant differences from the one above.

$$\left(\partial_{\tau}\varphi_{\alpha}\right)^{2} \to \left(\partial_{\tau}\varphi_{\alpha} + i\epsilon_{\alpha\beta\gamma}B_{\beta}\varphi_{\gamma}\right)^{2}.$$
(9.22)

The action (9.12), (9.22) can now be analyzed by a traditional small fluctuation analysis about $\varphi_{\alpha} = 0$. Let us assume that $\mathbf{B} = (0, 0, B)$ is oriented along the z axis. Then the coefficient of φ_z^2 is s, while that of $\varphi_x^2 + \varphi_y^2$ is $s - B^2$. This suggests that we focus only on the components of φ_{α} in the plane orthogonal to **B**, and integrate out the component of φ_{α} along the direction of **B**. Indeed, if we define

$$\Psi = \frac{\varphi_x + i\varphi_y}{\sqrt{B}} \tag{9.23}$$

and integrate out φ_z , then we obtain from (9.12), (9.22) the effective action for Ψ :

$$S_{\Psi} = \int d^2 r d\tau \left[\Psi^* \partial_{\tau} \Psi + \frac{c_x^2}{2B} \left| \partial_x \Psi \right|^2 + \frac{c_y^2}{2B} \left| \partial_y \Psi \right|^2 - \mu |\Psi|^2 + \frac{u}{24B} |\Psi|^4 \right].$$
(9.24)

Here, $\mu = (s - B^2)/2B$, and we have retained only leading order temporal and spatial gradients and the leading dependence of u. Clearly, this is the theory of a Bose gas in the grand canonical ensemble at a chemical potential μ , with a repulsive short-range interaction [41]. At T = 0, and $\mu < 0$, such a theory has a ground state which is simply the vacuum with no Bose particles. Here, this vacuum state corresponds to the spin gap antiferromagnet, and the *B*-independence of the ground state of the antiferromagnet corresponds here to the μ independence of the ground state of \mathcal{S}_{Ψ} . There is an onset of a finite density of bosons in \mathcal{S}_{Ψ} for $\mu > 0$, and this onset therefore corresponds to the quantum phase transition in the antiferromagnet at $B = \Delta$. So we must have $\mu = 0$ in \mathcal{S}_{Ψ} at precisely the point where $B = \Delta$: the value of μ quoted above shows that this is true at zeroth order in u, and higher order terms in u must conspire to maintain this result.

The above analysis makes it clear that the $\mu \geq 0$ region of S_{Ψ} will describe the quantum phase transition out of the paramagnet at non-zero B. This transition is merely the formation of a Bose-Einstein condensate of the m = 1component of the triplon bosons. For $\mu > 0$ we have a finite density of Ψ bosons which Bose condense in the ground state, so that $\langle \Psi \rangle \neq 0$. From (9.23) we see that this Bose condensation corresponds to antiferromagnetic order in the plane perpendicular to **B**. Indeed, the phase of this Bose condensate is simply the orientation of the spins in the x, y plane, and so here this phase is directly observable. Further, by taking derivatives of (9.19) and S_{Ψ} w.r.t. B, we see that the density of bosons is proportional to the magnetization per spin, Ω , in the direction parallel to **B**:

$$\Omega \equiv \frac{1}{N} \sum_{j} \langle S_{jz} \rangle \propto \langle |\Psi|^2 \rangle, \qquad (9.25)$$

where N is the total number of spins. Consequently, the average magnetic moments in the non-paramagnetic phase are in a 'canted' configuration, as shown in Fig. 9.5. The quantum phase transition between the paramagnet and the canted state is described by the theory of the density onset in a Bose gas: this theory has z = 2, $\nu = 1/2$, and an upper critical dimension of d = 2 [41,42].

We conclude this section by noting that interesting recent work [43] has examined the Bose-Einstein condensation of the m = 1 triplon bosons in a random potential. This is achieved by studying $\text{Tl}_{1-x}\text{K}_x\text{CuCl}_3$, where the stoichiometric disorder among the non-magnetic ions acts as a random potential on the triplons.

9.3.2 Strong Fields

We have seen above that applying a magnetic field eventually leads to the onset of a ferromagnetic moment in the directions of the applied field. How does this moment evolve as we continue to increase the field? Eventually, B will become so large that it pays to have all the spins polarized in the direction of the field: this corresponds to a saturation in the magnetization, and making B even stronger will not change the ground state. In terms of the

t bosons, this fully polarized state, $|FP\rangle$, with $\Omega = 1/2$, is seen from (9.20) or (9.4) to correspond exactly to

$$|FP\rangle = \prod_{\ell} \frac{(t_{\ell x}^{\dagger} + it_{\ell y}^{\dagger})}{\sqrt{2}}|0\rangle.$$
(9.26)

So there must be at least one more quantum phase transition as a B is increased: this is transition from the $|FP\rangle$ state at very large B to a state with a continuously varying ferromagnetic moment which eventually reaches the saturation value from below.

A theory for the transition away from the $|FP\rangle$ state with decreasing B can be developed using methods very similar to those used in Sect. 9.2.2 and 9.3.1. We treat the quartic terms in (9.7) in a Hartree-Fock approximation, and examine small fluctuations away from the $|FP\rangle$ state. These are dominated by excitation which create t_z quanta (which have m = 0) on the dimers, and so the effective theory is expressed in terms of

$$\tilde{\Psi}^{\dagger} \sim t_z^{\dagger}(t_x - it_y). \tag{9.27}$$

Indeed, it is not difficult to see that the resulting theory for $\tilde{\Psi}$ has exactly the same form as (9.24). Now the μ for $\tilde{\Psi}$ decreases with increasing B, and we have $\mu = 0$ at the critical field at which $|FP\rangle$ first becomes the ground state. Furthermore, $\langle |\tilde{\Psi}|^2 \rangle$ now measures the deviation away from $\Omega = 1/2$. Apart from this 'inversion' in the field axis, it is clear that the universality class of the present transition is identical to that discussed in Sect. 9.3.1.

A further possibility for a plateau in the value of Ω with increasing B is worth mentioning [44], as analogs are realized in $\operatorname{SrCu}_2(\operatorname{BO}_3)_2$ [45] and $\operatorname{NH}_4\operatorname{CuCl}_3$ [46]. So far we have found plateaus at $\Omega = 0$ for $B < \Delta$, and at $\Omega = 1/2$ for large B. For the $\Omega = 1/2$ state we had every dimer with a $(t_x^{\dagger} + it_y^{\dagger})/\sqrt{2}$ boson. Now imagine that these bosons form a Wigner-crystalline state so that there are p such bosons for every q dimers; here $0 \leq p \leq q$, $q \geq 1$, are integers. Such a state will have $\Omega = p/(2q)$, and breaks the translational symmetry of the underlying dimer antiferromagnet such that there are q dimers per unit cell (or 2q spins per unit cell). The energy gap towards boson motion in the Wigner crystal (*i.e.* its incompressibility) will ensure that Ω is stable under small variations of B. In this manner we can obtain a magnetization plateau at $\Omega = p/(2q)$ in a state with a unit cell of q dimers.

We summarize the considerations of this subsection in Fig. 9.6, showing a possible evolution of Ω in a model similar to H_d in (9.1). As we have already noted, the plateau onset transitions at $\Omega = 0$ and $\Omega = 1/2$ are both described by the z = 2 dilute Bose gas theory (9.24). The transitions in and out of other fractional plateaus are potentially more complicated because these involve spontaneous breaking of translational symmetry. The translation symmetry could be restored at the same point at which there is onset of superfluid



Fig. 9.6. Magnetization density, Ω , defined in (9.25) as a function of the applied magnetic field. The plateau shown at $\Omega = 0$ is present provided the zero field state is a paramagnet *i.e.* $\lambda < \lambda_c$. The full saturation plateau at $\Omega = 1/2$ is always present. The plateau at $\Omega = 1/4$ is not present in the nearest-neighbor model H_d in (9.1), but it is believed that such a plateau will appear upon including frustrating exchange interactions; this plateau will involve a broken translational symmetry in the coupled dimer antiferromagnet. Such magnetization plateaux are found in SrCu₂(BO₃)₂ [45] and NH₄CuCl₃ [46]

order—this is possibly a first order transition with a jump in the value of Ω . Alternatively, there could be an intermediate 'supersolid' phase, in which case the plateau transition has the same broken translational symmetry on both sides of it, placing it also in the class of (9.24).

9.4 Square Lattice Antiferromagnet

This section will address the far more delicate problem of quantum phase transitions in antiferromagnets with an odd number of S = 1/2 spins per unit cell. We will mainly concern ourselves with square lattice Hamiltonians of the form

$$H_s = J \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + \dots$$
(9.28)

Here J is a nearest-neighbor antiferromagnetic exchange and the ellipses represent further short-range exchange interactions (possibly involving multiple spin ring exchange) which preserve the full symmetry of the square lattice. The model H_d is a member of the class H_s only at $\lambda = 1$; at other values of λ the symmetry group of the square lattice is explicitly broken, and the doubling of the unit cell was crucial in the analysis of Sect. 9.2. With full square lattice symmetry, the paramagnetic phase is not determined as simply as in the small λ expansion, and we have to account more carefully for the 'resonance' between different valence bond configurations.

One ground state of H_s is, of course, the Néel state characterized by (9.2); this is obtained in the absence of the interactions denoted by ellipses in (9.28). Now imagine tuning the further neighbor couplings in (9.28) so that spin rotation invariance is eventually restored and we obtain a paramagnetic ground state. We can divide the possibilities for this state into two broad classes, which we discuss in turn.

In the first class of paramagnets, no symmetries of the Hamiltonian are broken, and the spins have paired with each other into valence bond singlets which strongly resonate between the large number of possible pairings: this is a resonating valence bond (RVB) liquid [47,48]. We will discuss such states further in Sect. 9.5: they have a connection with magnetically ordered states with non-collinear magnetic order, unlike the collinear Néel state of the nearest neighbor square lattice antiferromagnet.

In the second class of paramagnets, the valence bond singlets spontaneously crystallize into some configuration which necessarily breaks a lattice symmetry. A simple example of such a *bond-ordered* paramagnet is the columnar state we have already considered in Fig. 9.2. For the dimerized antiferromagnet H_d , the bond configuration in Fig. 9.2 was chosen explicitly in the Hamiltonian by the manner in which we divided the links into classes \mathcal{A} and \mathcal{B} for $\lambda \neq 1$. For H_s , there is no such distinction between the links, and hence a state like Fig. 9.2 spontaneously breaks a lattice symmetry. Furthermore, there are 3 other equivalent states, obtained by successive 90 degree rotations of Fig. 9.2 about any lattice site, which are completely equivalent. So for H_s , the bond-ordered paramagnet in Fig. 9.2 is four-fold degenerate. Going beyond simple variational wavefunctions like Fig. 9.2, the bond-ordered states are characterized by a bond order parameter

$$Q_{ij} = \langle \mathbf{S}_i \cdot \mathbf{S}_j \rangle; \tag{9.29}$$

the values of Q_{ij} on the links of the lattice in a bond-ordered state have a lower symmetry than the values of the exchange constants J_{ij} in the Hamiltonian. We will develop an effective model for quantum fluctuations about the collinear Néel state in H_s below, and will find that such bond-ordered paramagnets emerge naturally [19].

Let us now try to set up a theory for quantum fluctuations about the Néel state (9.2). It is best to do this in a formulation that preserves spin rotation invariance at all stages, and this is facilitated by the coherent state path integral (see Chap. 13 of [49]). The essential structure of this path integral can be understood simply by looking at a single spin in a magnetic field \mathbf{h} with the Hamiltonian $H_1 = -\mathbf{h} \cdot \mathbf{S}$. Then its partition function at a temperature T is given by

Tr exp
$$(\mathbf{h} \cdot \mathbf{S}/T) = \int \mathcal{D}\mathbf{n}(\tau) \exp\left(i2S\mathcal{A}[\mathbf{n}(\tau)] + S\int_{0}^{1/T} d\tau \mathbf{h} \cdot \mathbf{n}(\tau)\right).$$

(9.30)



Fig. 9.7. The path traced out by a single spin on the unit sphere in imaginary time. After discretizing time, the area enclose by the path is written as the sum over the areas of spherical triangles: A_{ij} is half the area of the triangle with vertices \mathbf{n}_0 , \mathbf{n}_i , \mathbf{n}_j . Different choices for the arbitrary point \mathbf{n}_0 correspond to different gauge choices associated with (9.32) and (9.34).

Here S is the angular momentum of the spin **S** (we are interested primarily in the case S = 1/2) and $\mathbf{n}(\tau)$ is a unit 3-vector with $\mathbf{n}(0) = \mathbf{n}(1/T)$. So the above path integral is over all closed curves on the surface of a sphere. The first term in the action of the path integral is the crucial Berry phase: $\mathcal{A}[\mathbf{n}(\tau)]$ is *half* the oriented area enclosed by the curve $\mathbf{n}(\tau)$ (the reason for the half will become clear momentarily). Note that this area is only defined modulo 4π , the surface area of a unit sphere. The expression (9.30) has an obvious generalization to the lattice Hamiltonian H_s : the action adds up the Berry phases of every spin, and there is an additional energy term which is just the Hamiltonian with the replacement $\mathbf{S}_j \to S\mathbf{n}_j$.

We are now faced with the problem of keeping track of the areas enclosed by the curves traced out by all the spins. This seems rather daunting, particularly because the half-area $\mathcal{A}[\mathbf{n}(\tau)]$ is a global object defined by the whole curve, and cannot be obviously be associated with local portions of the curve. One convenient way to proceed is illustrated in Fig. 9.7: discretize imaginary time, choose a fixed arbitrary point \mathbf{n}_0 on the sphere, and thus write the area as the sum of a large number of spherical triangles. Note that each triangle is associated with a local portion of the curve $\mathbf{n}(\tau)$.

We now need an expression for $\mathcal{A}(\mathbf{n}_1, \mathbf{n}_2, \mathbf{n}_3)$, defined as half the area of the spherical triangle with vertices \mathbf{n}_1 , \mathbf{n}_2 , \mathbf{n}_3 . Complicated expressions for this appear in treatises on spherical trigonometry, but a far simpler expression is obtained after transforming to spinor variables [50]. Let us write

$$\mathbf{n}_j \equiv z_{ja}^* \boldsymbol{\sigma}_{ab} z_{jb}, \tag{9.31}$$

where $a, b = \uparrow, \downarrow$ and we will always assume an implied summation over such indices, σ_{ab} are the Pauli matrices, and $z_{j\uparrow}, z_{j\downarrow}$ are complex numbers obeying $|z_{j\uparrow}|^2 + |z_{j\downarrow}|^2 = 1$. Note that knowledge of \mathbf{n}_j only defines z_{ja} up to a U(1) gauge transformation under which

$$z_{ja} \to z_{ja} e^{i\phi_j}.\tag{9.32}$$

Then, associated with each pair of vertices $\mathbf{n}_i, \mathbf{n}_j$ we define

$$\mathcal{A}_{ij} \equiv \arg\left[z_{ia}^* z_{ja}\right]. \tag{9.33}$$

Under the gauge transformation (9.32) we have

$$\mathcal{A}_{ij} \to \mathcal{A}_{ij} - \phi_i + \phi_j, \tag{9.34}$$

i.e. \mathcal{A}_{ij} behaves like a U(1) gauge field. Note also that \mathcal{A}_{ij} is only defined modulo 2π , and that $\mathcal{A}_{ji} = -\mathcal{A}_{ij}$. For future use, we also mention the following identity, which follows from (9.31) and (9.33):

$$z_{ia}^* z_{ja} = \left(\frac{1 + \mathbf{n}_i \cdot \mathbf{n}_j}{2}\right)^{1/2} e^{i\mathcal{A}_{ij}}.$$
(9.35)

The classical result for the half-area of the spherical triangle can be written in the simple form in terms of the present U(1) gauge variables:

$$\mathcal{A}(\mathbf{n}_1, \mathbf{n}_2, \mathbf{n}_3) = \mathcal{A}_{12} + \mathcal{A}_{23} + \mathcal{A}_{31}.$$

$$(9.36)$$

We chose \mathcal{A} as a *half*-area earlier mainly because then the expressions (9.33) and (9.36) come out without numerical factors. It is satisfying to observe that this total area is invariant under (9.34), and that the half-area is ambiguous modulo 2π .

Using (9.36), we can now write down a useful expression for $\mathcal{A}[\mathbf{n}(\tau)]$. We assume that imaginary time is discretized into times τ_j separated by intervals $\Delta \tau$. Also, we denote by $j + \tau$ the site at time $\tau_j + \Delta \tau$, and define $\mathcal{A}_{j,j+\tau} \equiv \mathcal{A}_{j\tau}$. Then

$$\mathcal{A}[\mathbf{n}(\tau)] = \sum_{j} \mathcal{A}_{j\tau}.$$
(9.37)

Note that this expression is a gauge-invariant function of the U(1) gauge field $A_{j\tau}$, and is analogous to the quantity sometimes called the Polyakov loop.

We are now ready to write down the first form proposed effective action for the quantum fluctuating Néel state. We do need to address some simple book-keeping considerations first:

(i) Discretize spacetime into a cubic lattice of points j. Note that the same index j referred to points along imaginary time above, and to square lattice

points in H_s . The meaning of the site index should be clear from the context. (*ii*) On each spacetime point j, we represent quantum spin operator \mathbf{S}_j by

$$\mathbf{S}_j = \eta_j S \mathbf{n}_j, \tag{9.38}$$

where \mathbf{n}_j is a unit vector, and $\eta_j = \pm 1$ is the sublattice staggering factor appearing in (9.2). This representation is that expected from the coherent state path integral, apart from the η_j factor. We have chosen to include η_j because of the expected local antiferromagnetic correlations of the spins. So in a quantum fluctuating Néel state, we can reasonably expect \mathbf{n}_j to be a slowly varying function of j.

(*iii*) Associated with each \mathbf{n}_j , define a spinor z_{ja} by (9.31).

(*iv*) With each link of the cubic lattice, we use (9.33) to associate with it a $\mathcal{A}_{j\mu} \equiv \mathcal{A}_{j,j+\mu}$. Here $\mu = x, y, \tau$ extends over the 3 spacetime directions.

With these preliminaries in hand, we can motivate the following effective action for fluctuations under the Hamiltonian H_s :

$$\widetilde{\mathcal{Z}} = \prod_{ja} \int dz_{ja} \prod_{j} \delta\left(|z_{ja}|^2 - 1 \right) \exp\left(\frac{1}{\tilde{g}} \sum_{\langle ij \rangle} \mathbf{n}_i \cdot \mathbf{n}_j + i2S \sum_{j} \eta_j \mathcal{A}_{j\tau} \right).$$
(9.39)

Here the summation over $\langle ij \rangle$ extends over nearest neighbors on the cubic lattice. The integrals are over the z_{ja} , and the \mathbf{n}_j and $\mathcal{A}_{j\tau}$ are dependent variables defined via (9.31) and (9.33). Note that both terms in the action are invariant under the gauge transformation (9.32); consequently, we could equally well have rewritten $\widetilde{\mathcal{Z}}$ as an integral over the \mathbf{n}_i , but it turns out to be more convenient to use the z_{ja} and to integrate over the redundant gauge degree of freedom. The first term in the action contains the energy of the Hamiltonian H_s , and acts to prefer nearest neighbor \mathbf{n}_i which are parallel to each other—this "ferromagnetic" coupling between the \mathbf{n}_i in spacetime ensures, via (9.38), that the local quantum spin configurations are as in the Néel state. The second term in the action is simply the Berry phase required in the coherent state path integral, as obtained from (9.30) and (9.37): the additional factor of η_i compensates for that in (9.38). The dimensionless coupling \tilde{q} controls the strength of the local antiferromagnetic correlations; it is like a "temperature" for the ferromagnet in spacetime. So for small \tilde{q} we expect \mathcal{Z} to be in the Néel phase, while for large \tilde{q} we can expect a quantum-"disordered" paramagnet. For a much more careful derivation of the partition function $\widetilde{\mathcal{Z}}$ from the underlying antiferromagnet H_s , including a quantitative estimate of the value of \tilde{g} , see *e.g.* Chap. 13 of [49].

While it is possible to proceed with the remaining analysis of this section using $\widetilde{\mathcal{Z}}$, we find it more convenient to work with a very closely related alternative model. Our proposed theory for the quantum fluctuating antiferromagnet in its final form is [51,52]

$$\mathcal{Z} = \prod_{j\mu} \int_{0}^{2\pi} \frac{dA_{j\mu}}{2\pi} \prod_{ja} \int dz_{ja} \prod_{j} \delta\left(|z_{ja}|^{2} - 1\right) \\ \exp\left(\frac{1}{g} \sum_{j\mu} \left(z_{ja}^{*} e^{-iA_{j\mu}} z_{j+\mu,a} + \text{c.c.}\right) + i2S \sum_{j} \eta_{j} A_{j\tau}\right).$$
(9.40)

Note that we have introduced a new field $A_{j\mu}$, on each link of the cubic lattice, which is integrated over. Like $\mathcal{A}_{i\mu}$, this is also a U(1) gauge field because all terms in the action above are invariant under the analog of (9.34):

$$A_{j\mu} \to A_{j\mu} - \phi_j + \phi_{j+\mu}. \tag{9.41}$$

The very close relationship between \mathcal{Z} and $\widetilde{\mathcal{Z}}$ may be seen [51] by explicitly integrating over the $A_{j\mu}$ in (9.40): this integral can be done exactly because the integrand factorizes into terms on each link that depend only on a single $A_{j\mu}$. After inserting (9.35) into (9.40), the integral over the $j\mu$ link is

$$\int_{0}^{2\pi} \frac{dA_{j\mu}}{2\pi} \exp\left(\frac{(2(1+\mathbf{n}_{j}\cdot\mathbf{n}_{j+\mu}))^{1/2}}{g}\cos(\mathcal{A}_{j\mu}-A_{j\mu})+i2S\eta_{j}\delta_{\mu\tau}A_{j\mu}\right)$$
$$=I_{2S\delta_{\mu\tau}}\left[\frac{(2(1+\mathbf{n}_{j}\cdot\mathbf{n}_{j+\mu}))^{1/2}}{g}\right]\exp\left(i2S\eta_{j}\delta_{\mu\tau}\mathcal{A}_{j\mu}\right),$$
(9.42)

where the result involves either the modified Bessel function I_0 (for $\mu = x, y$) or I_{2S} (for $\mu = \tau$). We can use the identity (9.42) to perform the integral over $A_{j\mu}$ on each link of (9.40), and so obtain a partition function, denoted \mathcal{Z}' , as an integral over the z_{ja} only. This partition function \mathcal{Z}' has essentially the same structure as $\tilde{\mathcal{Z}}$ in (9.39). The Berry phase term in \mathcal{Z}' is identical to that in $\tilde{\mathcal{Z}}$. The integrand of \mathcal{Z}' also contains a real action expressed solely as a sum over functions of $\mathbf{n}_i \cdot \mathbf{n}_j$ on nearest neighbor links: in $\tilde{\mathcal{Z}}$ this function is simply $\mathbf{n}_i \cdot \mathbf{n}_j / \tilde{g}$, but the corresponding function obtained from (9.40) is more complicated (it involves the logarithm of a Bessel function), and has distinct forms on spatial and temporal links. We do not expect this detailed form of the real action function to be of particular importance for universal properties: the initial simple nearest-neighbor ferromagnetic coupling between the \mathbf{n}_j in (9.39) was chosen arbitrarily anyway. So we may safely work with the theory \mathcal{Z} in (9.40) henceforth.

One of the important advantages of (9.40) is that we no longer have to keep track of the complicated non-linear constraints associated with (9.31)and (9.33); this was one of the undesirable features of (9.39). In \mathcal{Z} , we simply have free integration over the independent variables z_{ja} and $A_{j\mu}$. The remainder of this section will be devoted to describing the properties of \mathcal{Z} as a function of the coupling g. The theory \mathcal{Z} in (9.40) has some resemblance to the so-called \mathbb{CP}^{N-1} model from the particle physics literature [50, 53, 54]: our indices a, b take only 2 possible values, but the general model is obtained when $a, b = 1 \dots N$, and we will also find it useful to consider \mathcal{Z} for general N. The case of general N describes $\mathrm{SU}(N)$ and $\mathrm{Sp}(N)$ antiferromagnets on the square lattice [19]. Note also that it is essential for our purposes that the theory is invariant under $A_{j\mu} \to A_{j\mu} + 2\pi$, and so the U(1) gauge theory is *compact*. Finally our model contains a Berry phase term (which can be interpreted as a $J_{\mu}A_{\mu}$ term associated with a current $J_{j\mu} = 2S\eta_j\delta_{\mu\tau}$ of static charges $\pm 2S$ on each site) which is not present in any of the particle physics analyses. This Berry phase term will be an essential central actor in all of our results below for the paramagnetic phase and the quantum phase transition.

The properties of \mathcal{Z} are quite evident in the limit of small g. Here, the partition function is strongly dominated by configurations in which the real part of the action is a minimum. In a suitable gauge, these are the configurations in which $z_{ja} = \text{constant}$, and by (9.31), we also have \mathbf{n}_j a constant. This obviously corresponds to the Néel phase with (9.2). A Gaussian fluctuation analysis about such a constant saddle point is easily performed, and we obtain the expected spectrum of a doublet of gapless spin waves.

The situation is much more complicated for large g where we should naturally expect a paramagnetic phase with $\langle \mathbf{S}_j \rangle = \langle \mathbf{n}_j \rangle = 0$. This will be discussed in some detail in Sect. 9.4.1. Finally, we will address the nature of the quantum phase transition between the Néel and paramagnetic phases in Sect. 9.4.2.

9.4.1 Paramagnetic Phase

The discussion in this section has been adapted from another recent review by the author [55].

For large g, we can perform the analog of a 'high temperature' expansion of \mathcal{Z} in (9.40). We expand the integrand in powers of 1/g and perform the integral over the z_{ja} term-by-term. The result is then an effective theory for the compact U(1) gauge field $A_{j\mu}$ alone. An explicit expression for the effective action of this theory can be obtained in powers of 1/g: this has the structure of a strong coupling expansion in lattice gauge theory, and higher powers of 1/g yield terms dependent upon gauge-invariant U(1) fluxes on loops of all sizes residing on the links of the cubic lattice. For our purposes, it is sufficient to retain only the simplest such term on elementary square plaquettes, yielding the partition function

$$\widetilde{\mathcal{Z}}_{A} = \prod_{j\mu} \int_{0}^{2\pi} \frac{dA_{j\mu}}{2\pi} \exp\left(\frac{1}{e^{2}} \sum_{\Box} \cos\left(\epsilon_{\mu\nu\lambda} \Delta_{\nu} A_{j\lambda}\right) - i2S \sum_{j} \eta_{j} A_{j\tau}\right),\tag{9.43}$$

where $\epsilon_{\mu\nu\lambda}$ is the totally antisymmetric tensor in three spacetime dimensions. Here the cosine term represents the conventional Maxwell action for a compact U(1) gauge theory: it is the simplest local term consistent with the gauge symmetry (9.41) and which is periodic under $A_{j\mu} \rightarrow A_{j\mu} + 2\pi$; closely related terms appear under the 1/g expansion. The sum over \Box in (9.43) extends over all plaquettes of the cubic lattice, Δ_{μ} is the standard discrete lattice derivative ($\Delta_{\mu}f_j \equiv f_{j+\mu} - f_j$ for any f_j), and e^2 is a coupling constant. We expect the value of e to increase monotonically with g.

As is standard in duality mappings, we first rewrite the partition function in 2+1 spacetime dimensions by replacing the cosine interaction in (9.43) by a Villain sum [56, 57] over periodic Gaussians:

$$\mathcal{Z}_{A} = \sum_{\{q_{\bar{j}\mu}\}} \prod_{j\mu} \int_{0}^{2\pi} \frac{dA_{j\mu}}{2\pi} \exp\left(-\frac{1}{2e^{2}} \sum_{\Box} \left(\epsilon_{\mu\nu\lambda} \Delta_{\nu} A_{j\lambda} - 2\pi q_{\bar{j}\mu}\right)^{2} - i2S \sum_{j} \eta_{j} A_{j\tau}\right), \tag{9.44}$$

where the $q_{\bar{j}\mu}$ are integers on the links of the *dual* cubic lattice, which pierce the plaquettes of the direct lattice. Throughout this article we will use the index \bar{j} to refer to sites of this dual lattice, while j refers to the direct lattice on sites on which the spins are located.

We will now perform a series of exact manipulations on (9.44) which will lead to a dual *interface* model [19, 20, 58]. This dual model has only positive weights—this fact, of course, makes it much more amenable to a standard statistical analysis. This first step in the duality transformation is to rewrite (9.44) by the Poisson summation formula:

$$\sum_{\{q_{\bar{j}\mu}\}} \exp\left(-\frac{1}{2e^2} \sum_{\Box} \left(\epsilon_{\mu\nu\lambda} \Delta_{\nu} A_{j\lambda} - 2\pi q_{\bar{j}\mu}\right)^2\right)$$
$$= \sum_{\{a_{\bar{j}\mu}\}} \exp\left(-\frac{e^2}{2} \sum_{\bar{j}} a_{\bar{j}\mu}^2 - i \sum_{\Box} \epsilon_{\mu\nu\lambda} a_{\bar{j}\mu} \Delta_{\nu} A_{j\lambda}\right) (9.45)$$

where $a_{\bar{j}\mu}$ (like $q_{\bar{j}\mu}$) is an integer-valued vector field on the links of the dual lattice (here, and below, we drop overall normalization factors in front of the partition function). Next, we write the Berry phase in a form more amenable to duality transformations. Choose a 'background' $a_{\bar{j}\mu} = a_{\bar{j}\mu}^0$ flux which satisfies

$$\epsilon_{\mu\nu\lambda}\Delta_{\nu}a^{0}_{\bar{j}\lambda} = \eta_{j}\delta_{\mu\tau}, \qquad (9.46)$$

where j is the direct lattice site in the center of the plaquette defined by the curl on the left-hand-side. Any integer-valued solution of (9.46) is an acceptable choice for $a_{\overline{j}\mu}^0$, and a convenient choice is shown in Fig. 9.8. Using



Fig. 9.8. Specification of the non-zero values of the fixed field $a_{j\mu}^0$. The circles are the sites of the direct lattice, j, while the crosses are the sites of the dual lattice, \bar{j} ; the latter are also offset by half a lattice spacing in the direction out of the paper (the $\mu = \tau$ direction). The $a_{j\mu}^0$ are all zero for $\mu = \tau, x$, while the only non-zero values of a_{jy}^0 are shown above. Notice that the a^0 flux obeys (9.46).

(9.46) to rewrite the Berry phase in (9.44), applying (9.45), and shifting $a_{\bar{j}\mu}$ by the integer $2Sa_{\bar{j}\mu}^0$, we obtain a new exact representation of \mathcal{Z}_A in (9.44):

$$\mathcal{Z}_{A} = \sum_{\{a_{\bar{j}\mu}\}} \prod_{j\mu} \int_{0}^{2\pi} \frac{dA_{j\mu}}{2\pi} \exp\left(-\frac{e^{2}}{2} \sum_{\bar{j},\mu} (a_{\bar{j}\mu} - 2Sa_{\bar{j}\mu}^{0})^{2} -i \sum_{\Box} \epsilon_{\mu\nu\lambda} a_{\bar{j}\mu} \Delta_{\nu} A_{j\lambda}\right). \quad (9.47)$$

The integral over the $A_{j\mu}$ can be performed independently on each link, and its only consequence is the imposition of the constraint $\epsilon_{\mu\nu\lambda}\Delta_{\nu}a_{\bar{j}\lambda} = 0$. We solve this constraint by writing $a_{\bar{j}\mu}$ as the gradient of an integer-valued 'height' $h_{\bar{i}}$ on the sites of the dual lattice, and so obtain

$$\mathcal{Z}_{h} = \sum_{\{h_{\bar{j}}\}} \exp\left(-\frac{e^{2}}{2} \sum_{\bar{j},\mu} (\Delta_{\mu} h_{\bar{j}} - 2Sa^{0}_{\bar{j}\mu})^{2}\right).$$
(9.48)

We emphasize that, apart from an overall normalization, we have $Z_h = Z_A$ exactly. This is the promised 2+1 dimensional interface, or height, model in almost its final form.

The physical properties of (9.48) become clearer by converting the "frustration" $a_{\bar{j}\mu}^0$ in (9.48) into offsets for the allowed height values. This is done by decomposing $a_{\bar{j}\mu}^0$ into curl and divergence free parts and writing it in terms of new fixed fields, $\mathcal{X}_{\bar{j}}$ and $\mathcal{Y}_{j\mu}$ as follows:

$$a_{\bar{j}\mu}^0 = \Delta_\mu \mathcal{X}_{\bar{j}} + \epsilon_{\mu\nu\lambda} \Delta_\nu \mathcal{Y}_{j\lambda}. \tag{9.49}$$

The values of these new fields are shown in Fig. 9.9. Inserting (9.49) into (9.48), we can now write the height model in its simplest form [20]



Fig. 9.9. Specification of the non-zero values of the fixed fields (a) $\mathcal{X}_{\bar{j}}$, (b) $\mathcal{Y}_{j\mu}$, (c) $\epsilon_{\mu\nu\lambda}\Delta_{\nu}\mathcal{Y}_{j\lambda}$ introduced in (9.49). The notational conventions are as in Fig. 9.8. Only the $\mu = \tau$ components of $\mathcal{Y}_{j\mu}$ are non-zero, and these are shown in (b). Only the spatial components of $\epsilon_{\mu\nu\lambda}\Delta_{\nu}\mathcal{Y}_{j\lambda}$ are non-zero, and these are oriented as in (c) with magnitude 1/4. The four dual sublattices, W, X, Y, Z, are also indicated in (c). Note that $\mathcal{X}_W = 0$, $\mathcal{X}_X = 1/4$, $\mathcal{X}_Y = 1/2$, and $\mathcal{X}_Z = 3/4$.

$$\mathcal{Z}_{h} = \sum_{\{H_{\bar{j}}\}} \exp\left(-\frac{e^{2}}{2} \sum_{\bar{j}} \left(\Delta_{\mu} H_{\bar{j}}\right)^{2}\right),\tag{9.50}$$

where

$$H_{\bar{j}} \equiv h_{\bar{j}} - 2S\mathcal{X}_{\bar{j}} \tag{9.51}$$

is the new height variable we shall work with. Notice that the $\mathcal{Y}_{j\mu}$ have dropped out, while the $\mathcal{X}_{\bar{j}}$ act only as fractional offsets (for S not an even integer) to the integer heights. From (9.51) we see that for half-odd-integer S the height is restricted to be an integer on one of the four sublattices, an integer plus 1/4 on the second, an integer plus 1/2 on the third, and an integer plus 3/4 on the fourth; the fractional parts of these heights are as shown in Fig. 9.9a; the steps between neighboring heights are always an integer plus 1/4, or an integer plus 3/4. For S an odd integer, the heights are integers on one square sublattice, and half-odd-integers on the second sublattice. Finally for even integer S the offset has no effect and the height is an integer on all sites. We discuss these classes of S values in turn in the following subsections.

4.1.1 S Even Integer

In this case the offsets $2SX_{\bar{j}}$ are all integers, and (9.50) is just an ordinary three dimensional height model which has been much studied in the literature [57,59]. Unlike the two-dimensional case, three-dimensional height models generically have no roughening transition, and the interface is always smooth [59]. With all heights integers, the smooth phase breaks no lattice symmetries. So square lattice antiferromagnets with S even integer can have a paramagnetic ground state with a spin gap and no broken symmetries. The smooth interface corresponds to confinement in the dual compact U(1) gauge theory [60]: consequently the z_a of \mathcal{Z} are confined, and the elementary excitations are S = 1 quasiparticles, similar to the φ_{α} of S_{φ} . This is in accord with the exact ground state for a S = 2 antiferromagnet on the square lattice found by Affleck *et al.*, the AKLT state [61].

4.1.2 S Half-Odd-Integer

Now the heights of the interface model can take four possible values, which are integers plus the offsets on the four square sublattices shown in Fig. 9.9a. As in Sect. 9.4.1.1, the interface is always smooth *i.e.* any state of (9.50) has a fixed average interface height

$$\overline{H} \equiv \frac{1}{N_d} \sum_{\overline{j}=1}^{N_d} \langle H_{\overline{j}} \rangle, \qquad (9.52)$$

where the sum is over a large set of N_d dual lattice points which respect the square lattice symmetry. Any well-defined value for \overline{H} breaks the uniform shift symmetry of the height model under which $H_{\overline{j}} \to H_{\overline{j}} \pm 1$. In the present context, only the value of \overline{H} modulo integers is physically significant, and so the breaking of the shift symmetry is not important by itself. However, after accounting for the height offsets, we now prove that any smooth interface must also break a lattice symmetry with the development of bond order: this means that Z_A in (9.44) describes spin gap ground states of the lattice antiferromagnet which necessarily have spontaneous bond order.

The proof of this central result becomes clear upon a careful study of the manner in which the height model in (9.50) and (9.51) implements the 90° rotation symmetry about a direct square lattice point. Consider such a rotation under which the dual sublattice points in Fig. 9.9c interchange as

$$W \to X, \quad X \to Y, \quad Y \to Z, \quad Z \to W.$$
 (9.53)

The terms in the action in (9.51) will undergo a 90° rotation under this transformation provided the integer heights $h_{\bar{j}}$ transform as



Fig. 9.10. Mapping between the quantum dimer model and the interface model \mathcal{Z}_h in (9.50). Each dimer on the direct lattice is associated with a step in height of $\pm 3/4$ on the link of the dual lattice that crosses it. All other height steps are $\pm 1/4$. Each dimer represents a singlet valence bond between the sites, as in Fig. 9.2.

$$h_W \to h_X, \quad h_X \to h_Y, \quad h_Y \to h_Z, \quad h_Z \to h_W - 1.$$
 (9.54)

Notice the all important -1 in the last term—this compensates for the 'branch cut' in the values of the offsets $\mathcal{X}_{\bar{j}}$ as one goes around a plaquette in Fig. 9.9c. From (9.54), it is evident that the average height $\overline{H} \to \overline{H} - 1/4$ under the 90° rotation symmetry under consideration here. Hence, a smooth interface with a well-defined value of \overline{H} always breaks this symmetry.

We now make this somewhat abstract discussion more physical by presenting a simple interpretation of the interface model in the language of the S = 1/2 antiferromagnet [62]. From Fig. 9.9a it is clear that nearest neighbor heights can differ either by 1/4 or 3/4 (modulo integers). To minimize the action in (9.50), we should choose the interface with the largest possible number of steps of $\pm 1/4$. However, the interface is frustrated, and it is not possible to make all steps $\pm 1/4$ and at least a quarter of the steps must be $\pm 3/4$. Indeed, there is a precise one-to-one mapping between interfaces with the minimal number of $\pm 3/4$ steps (we regard interfaces differing by a uniform integer shift in all heights as equivalent) and the dimer coverings of the square lattice: the proof of this claim is illustrated in Fig. 9.10. We identify each dimer with a singlet valence bond between the spins (the ellipses in Fig. 9.2), and so each interface corresponds to a quantum state with each spin locked in a singlet valence bond with a particular nearest neighbor. Fluctuations of the interface in imaginary time between such configurations correspond to quantum tunneling events between such dimer states, and an effective Hamiltonian for this is provided by the quantum dimer model [63]. While such an interpretation in terms of the dimer model is appealing, we should also note that it is not as general as the dual interface model: on certain lattices, while the collinear paramagnetic state continues to have a representation as a dual interface model, there is no corresponding dimer interpretation [64].



Fig. 9.11. Sketch of the two simplest possible states with bond order for S = 1/2 on the square lattice: (a) the columnar spin-Peierls states, and (b) plaquette state. Here the distinct line styles encode the different values of the bond order parameter Q_{ij} in (9.29) on the links. This should be contrasted from Figs. 9.1–9.4 where the line styles represented distinct values of the exchange constants in the Hamiltonian. In the present section, the Hamiltonian has the full symmetry of the square lattice, and the orderings represented above amount to a spontaneous breaking of the lattice symmetry. Both states above are 4-fold degenerate; an 8-fold degenerate state, with superposition of the above orders, also appears as a possible ground state of the generalized interface model. Numerical studies of a number of two-dimensional quantum antiferromagnets [66–68, 70, 73–75] have found ground states with spontaneous bond order, similar to the states shown above.

The nature of the possible smooth phases of the interface model are easy to determine from the above picture and by standard techniques from statistical theory [20, 62]. As a simple example, the above mapping between interface heights and dimer coverings allows one to deduce that interfaces with average height $\overline{H} = 1/8, 3/8, 5/8, 7/8$ (modulo integers) correspond to the four-fold degenerate bond-ordered states in Fig. 9.11a. To see this, select the interface with $h_{\bar{j}} = 0$ for all \bar{j} : this interface has the same symmetry as Fig. 9.11a, and a simple computation summing over sites from (9.51) shows that this state has average height $\overline{H} = -(0 + 1/4 + 1/2 + 3/4)/4 = -3/8$ for S = 1/2. The remaining three values of \overline{H} correspond to the three other states obtained by successive 90° rotations of Fig. 9.11a. In a similar manner, interfaces with H = 0, 1/4, 1/2, 3/4 (modulo integers) correspond to the four-fold degenerate plaquette bond-ordered states in Fig. 9.11b. A simple example of such an interface is the "disordered-flat" state [65] in which $h_{\bar{i}} = 0$ on all sites \bar{j} , except for the W sublattice which have $\mathcal{X}_{\bar{i}} = 0$; for these sites we have $h_{\bar{i}}$ fluctuate randomly between $h_{\bar{j}} = 0$ and $h_{\bar{j}} = 1$, and independently for different \bar{j} . The average height of such an interface is H = -((0+1)/2 + 1/4 + 1/2 + 3/4)/4 =-1/2 for S = 1/2, and the mapping to dimer coverings in Fig. 9.10 shows easily that such an interface corresponds to the state in Fig. 9.11b. All values of H other than those quoted above are associated with eight-fold degenerate bond-ordered states with a superposition of the orders in Fig. 9.11a and b.

All these phases are expected to support non-zero spin quasiparticle excitations which carry spin S = 1, but not S = 1/2. Despite the local corrugation in the interface configuration introduced by the offsets, the interface remains smooth on the average, and this continues to correspond to confinement in the dual compact U(1) gauge theory [60]. Consequently the spinons of Fig. 9.3b are confined in pairs. The structure of the resulting S = 1 triplon quasiparticles is very similar to the excitations of the paramagnetic phase of the coupled dimer antiferromagnet of Sect. 9.2, as we already noted in Sect. 9.1.

Support for the class of bond-ordered states described above has appeared in a number of numerical studies of S = 1/2 antiferromagnets in d = 2which have succeeded in moving from the small q Néel phase to the large g paramagnet. These include studies on the honeycomb lattice [66] (duality mapping on the honeycomb lattice appears in [19]), on the planar pyrochlore lattice [67,68] (duality mapping for a lattice with the symmetry of the planar pyrochlore is in [64, 69], with a prediction for the bond order observed), on square lattice models with ring-exchange and easy-plane spin symmetry [70] (duality mapping on spin models with easy plane symmetry is in [52, 71, 72]), and square lattice models with SU(N) symmetry [73] (the theories (9.40), with $a = 1 \dots N$, and (9.50) apply unchanged to SU(N) antiferromagnets). The case of the square lattice antiferromagnet with first and second neighbor exchange is not conclusively settled: while two recent studies [74, 75] (and earlier work [25,76]) do observe bond order in a paramagnetic spin-gap state, a third [77] has so far not found such order. It is possible that this last study is observing signatures of the critical point between the Néel and bond-ordered states (to be described in Sect. 9.4.2) which is expressed in a theory for deconfined spinons in \mathcal{Z}_c in (9.55).

Finally, we also mention that evidence for the spontaneous bond order of Fig. 9.11 appears in recent numerical studies of *doped* antiferromagnets [78,79].

4.1.3 S Odd Integer

This case is similar to that S half-odd-integer, and we will not consider it in detail. The Berry phases again induce bond order in the spin gap state, but this order need only lead to a two-fold degeneracy.

9.4.2 Critical Theory

We turn finally to the very difficult issue of the nature of the quantum phase transition from the Néel state to one of the bond-ordered paramagnetic states in Fig. 9.10 as a function of increasing g. This has been a long-standing open problem, and many different proposals have been made. The two phases break different symmetries of the Hamiltonian, and so are characterized by very different order parameters (one lives in spin space, and the other in real

space). Landau-Ginzburg-Wilson (LGW) theory would imply that a generic second-order transition is not possible between such phases, and one obtains either a first-order transition or a region of co-existence of the two orders. However, the bond-order in the paramagnet was obtained entirely from quantum Berry phases attached to the fluctuating Néel order, and it is not clear that LGW theory applies in such a situation.

Recent work by Senthil *et al.* [80,81] has proposed an elegant resolution to many of these problems, and we will describe their results in the remainder of this subsection. The results are based upon solutions of a series of simpler models which strongly suggest that related results also apply to the SU(2) invariant, S = 1/2 models of interest. The computations are intricate, but the final results are quite easy to state, and are presented below. We will mainly limit our discussion here to the case of antiferromagnets of spin S = 1/2.

First, contrary to the predictions of LGW theory, a generic second-order transition between the Néel state and the bond-ordered paramagnet is indeed possible (let us assume it occurs at $g = g_c$ for \mathcal{Z} in (9.40)). The theory for such a quantum critical point is obtained simply by taking a naive continuum limit of \mathcal{Z} while ignoring both the compactness of the gauge field and the Berry phases. Remarkably, these complications of the lattice model \mathcal{Z} , which we have so far stated were essential for the complete theory, have effects which cancel each other out, but *only* at the critical point. Note compactness on its own is a relevant perturbation which cannot be ignored *i.e.* without Berry phases, the compact and non-compact lattice CP^1 model have distinct critical theories [82]. However, the surprising new point noted by Senthil et al. [80, 81] is that the non-compact CP^1 model has the same critical theory as the compact CP^1 model with S = 1/2 Berry phases. Taking the naive continuum limit of \mathcal{Z} in (9.40), and softening the hard-constraint on the z_{ia} , we obtain the proposed theory for the quantum critical point between the Néel state and the bond-ordered paramagnet for spin S = 1/2 [80,81]:

$$\mathcal{Z}_{c} = \int \mathcal{D}z_{a}(r,\tau) \mathcal{D}A_{\mu}(r,\tau) \exp\left(-\int d^{2}r d\tau \left[|(\partial_{\mu} - iA_{\mu})z_{a}|^{2} + s|z_{a}|^{2} + \frac{u}{2}(|z_{a}|^{2})^{2} + \frac{1}{4e^{2}}(\epsilon_{\mu\nu\lambda}\partial_{\nu}A_{\lambda})^{2} \right] \right).$$
(9.55)

We have also included here a kinetic term for the A_{μ} , and one can imagine that this is generated by integrating out large momentum z_{ja} . On its own, \mathcal{Z}_c describes the transition from a magnetically ordered phase with z_a condensed at $s < s_c$, to a disordered state with a gapless U(1) photon at $s > s_c$ (here s_c is the critical point of \mathcal{Z}_c). Clearly the $s < s_c$ phase corresponds to the Néel phase of \mathcal{Z} in (9.40) for $g < g_c$. However, the $s > s_c$ phase does not obviously correspond to the $g > g_c$ bond-ordered, fully gapped, paramagnet of \mathcal{Z} . This is repaired by accounting for the compactness of the gauge field and the Berry phases: it is no longer possible to neglect them, while it was safe to do so precisely at $g = g_c$. The *combined* effects of compactness and Berry phases are therefore *dangerously irrelevant* at $g = g_c$.

It is important to note that the critical theory of (9.55) is distinct from the critical theory S_{φ} in (9.12), although both theories have a global O(3) symmetry [82]. In particular the values of the exponents ν are different in the two theories, and the scaling dimension of the Néel order parameter φ_{α} under S_{φ} is distinct from the scaling dimension of the Néel order parameter $z_a^* \sigma_{ab}^a z_b$ at the critical point of \mathcal{Z}_c .

It is interesting that Z_c in (9.55) is a theory for the S = 1/2 spinors z_a . These can be understood to be the continuum realization of the spinons shown earlier in Fig. 9.3b. Thus the spinons become the proper elementary degrees of freedom, but *only* at the quantum critical point. Hence it is appropriate to label this as a 'deconfined quantum critical point' [80]. These spinons are confined into a S = 1 quasiparticle once bond order appears for $g > g_c$, for reasons similar to those illustrated in Fig. 9.3b.

A key characteristic of this 'deconfined' critical point is the irrelevance of the compactness of the gauge field, and hence of monopole tunnelling events. A consequence of this is that the flux of the A_{μ} gauge field in \mathcal{Z}_c is conserved. This emergent conservation law, and the associated long-range gauge forces are key characteristics of such critical points.

We summarize in Fig. 9.12 our results for S = 1/2 square lattice antiferromagnets, as described by \mathcal{Z} in (9.40).

The claims above for the conspiracy between the compactness and Berry phases at the critical point are surprising and new. They are central to a complete understanding of square lattice antiferromagnets, and a full justification of the claims appears in the work of Senthil *et al.*. The following subsections illustrate their origin by considering a series of models, of increasing complexity, where similar phenomena can be shown to occur. The reader may also find it useful to look ahead to Tables 1 and 2, which summarize the intricate relationships between the models considered.

4.2.1 Lattice Model at N = 1

This subsection describes a simplified lattice gauge theory model introduced by Sachdev and Jalabert [51]. While the duality analysis presented below was initiated in [51], its correct physical interpretation, and the implications for more general models are due to Senthil *et al.* [80,81].

The model of interest in this subsection is the N = 1 case of \mathcal{Z} . Physically, such a model will be appropriate for an antiferromagnet in the presence of a staggered magnetic field: such a field will prefer z_{\uparrow} over z_{\downarrow} (say). So we write the preferred single component complex scalar simply as $z_j = e^{i\theta_j}$, and obtain from (9.40)



Fig. 9.12. Phase diagram of the model \mathcal{Z} in (9.40) of S = 1/2 antiferromagnets with full square lattice symmetry. There is a Néel phase for $g < g_c$ which breaks spin rotation invariance; it has a doublet of gapless spin wave excitations. The bond-ordered paramagnet for $g > g_c$ preserves spin rotation invariance but breaks square lattice symmetry; it has a gap to all excitations, and the non-zero spin excitations are described by S = 1 triplet quasiparticles which are very similar to the 'triplons' discussed in Sect. 9.2.1. The critical point at $g = g_c$ is described by the theory of S = 1/2 'spinons', \mathcal{Z}_c in (9.55) at its critical point $s = s_c$; note that this mapping to the spinon theory \mathcal{Z}_c does not work away from $g = g_c$, and spinons are confined for all $g > g_c$. A phase diagram like the one above has been used as a point of departure to obtain a phase diagram for doped Mott insulators [22,83], as a description of the cuprate superconductors; evidence for spontaneous bond order in doped antiferromagnets appears in [78,79].

$$\mathcal{Z}_{1} = \prod_{j} \int_{0}^{2\pi} \frac{d\theta_{j}}{2\pi} \int_{0}^{2\pi} \frac{dA_{j\mu}}{2\pi} \exp\left(\frac{1}{e^{2}} \sum_{\Box} \cos\left(\epsilon_{\mu\nu\lambda} \Delta_{\nu} A_{j\lambda}\right) + \frac{1}{g} \sum_{j,\mu} \cos\left(\Delta_{\mu} \theta_{j} - A_{j\mu}\right) + i2S \sum_{j} \eta_{j} A_{j\tau}\right). \quad (9.56)$$

We have chosen here to explicitly include a compact Maxwell term for the gauge field, as that proves convenient in the description of the duality mappings. Note that if we integrate out the θ_j for large g, then we again obtain the model Z_A in (9.43) which was used to describe the paramagnetic phase in Sect. 9.4.1. So bond order appears also in the model Z_1 at large g. This bond order disappears as g is reduced, at a transition we will describe below.

Rather than attack \mathcal{Z}_1 directly, it is useful as a warm-up, and to make contact with previous work, to consider a sequence of simpler models that

have been considered in the literature. As we have emphasized, \mathcal{Z}_1 features the combined complications of compactness and Berry phases, essential for a proper description of quantum antiferromagnets. It is the simplest model in which it can be shown that these complications effectively neutralize one another at the critical point.

In the following subsection, we make things simpler for ourselves momentarily by dropping *both* the compactness and the Berry phases. We will then, in the subsequent subsections, add these complications back in.

A. XY Model with a Non-compact U(1) Gauge Field

Dropping both compactness and Berry phases, \mathcal{Z}_1 reduces to

$$\mathcal{Z}_{\rm SC} = \prod_{j} \int_{0}^{2\pi} \frac{d\theta_{j}}{2\pi} \int_{-\infty}^{\infty} dA_{j\mu} \exp\left(-\frac{1}{2e^{2}} \sum_{\Box} \left(\epsilon_{\mu\nu\lambda} \Delta_{\nu} A_{j\lambda}\right)^{2} + \frac{1}{g} \sum_{j,\mu} \cos\left(\Delta_{\mu} \theta_{j} - A_{j\mu}\right)\right).$$
(9.57)

Notice that the Maxwell term for the gauge field now has a simple Gaussian form. This is simply the lattice, classical, Ginzburg-Landau model (or an XY model) of a superconductor at finite temperatures coupled to electromagnetism. This model has been studied extensively in the past, and the key result was provided by Dasgupta and Halperin [84]. As we review below, they showed that \mathcal{Z}_{SC} exhibited an *inverted XY transition i.e.* it was dual to the theory of a complex scalar ψ in the absence of a gauge field:

$$\mathcal{Z}_{\rm SC,dual} = \int \mathcal{D}\psi(r,\tau) \exp\left(-\int d^2r d\tau \left(|\partial_{\mu}\psi|^2 + \overline{s}|\psi|^2 + \frac{\overline{u}}{2}|\psi|^4\right)\right).$$
(9.58)

The field ψ is a creation operator for *vortices* in the original theory of the Ginzburg-Landau superconductor. These have a short-range interaction (\overline{u} above) because of the screening provided by the electromagnetic flux quantum attached to every vortex in (9.57). So the vortex loops of (9.57) behave like the world lines of the dual boson field of (9.58). The tuning parameter \overline{s} in (9.58) is 'inverted' from the perspective of the direct theory: the $\overline{s} < \overline{s}_c$ phase with $\langle \psi \rangle \neq 0$ has a vortex condensate and so is the normal state of a Ginzburg-Landau superconductor, while the $\overline{s} > \overline{s}_c$ phase with $\langle \psi \rangle = 0$ has the vortices gapped as in the superconducting phase.

We now provide a few steps in the analysis which links (9.57) to (9.58). The steps are very similar to those described in Sect. 9.4.1 below (9.43) and (9.44). We write the cosine in (9.57) in its Villain form, decouple it by the Poisson summation formula using integer currents $J_{j\mu}$, and also decouple the Maxwell term by a Hubbard-Stratonovich field $P_{\bar{j}\mu}$; this yields the analog of (9.45) for \mathcal{Z}_{SC} :

$$\mathcal{Z}_{\text{SC},1} = \prod_{j} \int_{0}^{2\pi} \frac{d\theta_{j}}{2\pi} \int_{-\infty}^{\infty} dA_{j\mu} \sum_{\{J_{j\mu}\}} \int_{-\infty}^{\infty} dP_{\bar{j}\mu} \exp\left(-\frac{e^{2}}{2} \sum_{\bar{j},\mu} P_{\bar{j}\mu}^{2}\right)$$
$$-\frac{g}{2} \sum_{j\mu} J_{j\mu}^{2} + i \sum_{j} J_{j\mu} \left(\Delta_{\mu}\theta_{j} - A_{j\mu}\right) + i \sum_{\Box} \epsilon_{\mu\nu\lambda} P_{\bar{j}\mu} \Delta_{\nu} A_{j\lambda} \left(9.59\right)$$

The advantage of this form is that the integrals over θ_j and $A_{j\mu}$ can be performed exactly, and they lead to the constraints

$$\Delta_{\mu}J_{j\mu} = 0 \quad ; \qquad J_{j\mu} = \epsilon_{\mu\nu\lambda}\Delta_{\nu}P_{\bar{j}\lambda}. \tag{9.60}$$

We solve these constraints by writing

$$J_{j\mu} = \epsilon_{\mu\nu\lambda} \Delta_{\nu} b_{\bar{j}\lambda} \quad ; \quad P_{\bar{j}\mu} = b_{\bar{j}\mu} - \Delta_{\mu} \varphi_{\bar{j}}, \tag{9.61}$$

where $b_{\bar{j}\mu}$ is an integer valued field on the links of the dual lattice, and $\varphi_{\bar{j}}$ is a real valued field on the sites of the dual lattice. This transforms (9.59) to

$$\mathcal{Z}_{\mathrm{SC},2} = \prod_{\bar{j}} \int_{-\infty}^{\infty} d\varphi_{\bar{j}} \sum_{\{b_{\bar{j}\mu}\}} \exp\left(-\frac{e^2}{2} \sum_{\bar{j},\mu} (b_{\bar{j}\mu} - \Delta_{\mu}\varphi_{\bar{j}})^2 -\frac{g}{2} \sum_{\Box} (\epsilon_{\mu\nu\lambda}\Delta_{\nu}b_{\bar{j}\lambda})^2\right); \quad (9.62)$$

precisely this dual form was obtained by Dasgupta and Halperin [84], and used by them for numerical simulations. We proceed further analytically, using methods familiar in the theory of duality mappings [57]: we promote the integer valued $b_{\bar{j}\mu}$ to a real field by the Poisson summation method, and introduce, by hand, a vortex fugacity y_v . This transforms $\mathcal{Z}_{SC,2}$ to

$$\mathcal{Z}_{\mathrm{SC},3} = \prod_{\bar{j}} \int_{-\infty}^{\infty} db_{\bar{j}\mu} \int_{-\infty}^{\infty} d\varphi_{\bar{j}} \int_{-\infty}^{\infty} d\vartheta_{\bar{j}} \exp\left(-\frac{e^2}{2} \sum_{\bar{j},\mu} (b_{\bar{j}\mu} - \Delta_{\mu}\varphi_{\bar{j}})^2 - \frac{g}{2} \sum_{\Box} \left(\epsilon_{\mu\nu\lambda}\Delta_{\nu}b_{\bar{j}\lambda}\right)^2 + y_v \sum_{\bar{j},\mu} \cos\left(2\pi b_{\bar{j}\mu} - \Delta_{\mu}\vartheta_{\bar{j}}\right)\right).$$
(9.63)

Notice that the effect of the vortex fugacity is to yield the least action when $b_{\bar{j}\mu}$ is an integer (ignore $\vartheta_{\bar{j}}$ momentarily): so we have effectively 'softened' the integer constraint on $b_{\bar{j}\mu}$. We have also introduced here a new real valued field $\vartheta_{\bar{j}}$ on the sites of the dual lattice simply to make the $\mathcal{Z}_{SC,3}$ invariant under U(1) gauge transformations of $b_{\bar{j}\mu}$. This is mainly because the physics is clearer in this explicitly gauge-invariant form. We could, if we had wished, also chosen a gauge in which $\vartheta_{\bar{j}} = 0$, and then the field $\vartheta_{\bar{j}}$ would not be present in $\mathcal{Z}_{SC,3}$ (this justifies neglect of $\vartheta_{\bar{j}}$ above). In the complete form in (9.63), it is clear from the first two Gaussian terms that fluctuations of the $b_{\bar{j}\mu}$ gauge

field have been 'Higgsed' by the real field $\varphi_{\bar{j}}$. Indeed, it is more convenient to choose a gauge in which $\varphi_{\bar{j}} = 0$, and we do so. Now the fluctuations of $b_{\bar{j}\mu}$ are 'massive' and so can be safely integrated out. To leading order in y_v , this involves simply replacing $b_{\bar{j}\mu}$ with the saddle point value obtained from the first two Gaussian terms, which is $\bar{b}_{\bar{j}\mu} = 0$. So we have the very simple final theory

$$\mathcal{Z}_{\mathrm{SC},4} = \prod_{\bar{j}} \int_{-\infty}^{\infty} d\vartheta_{\bar{j}} \exp\left(y_v \sum_{\bar{j},\mu} \cos\left(\Delta_{\mu} \vartheta_{\bar{j}}\right)\right),\tag{9.64}$$

which has the form of the dual XY model. We now take the continuum limit of (9.64) by a standard procedure [85] of introducing a complex field ψ conjugate to $e^{i\vartheta_{\bar{j}}}$, and obtain the theory $\mathcal{Z}_{SC,dual}$ as promised. This establishes the duality mapping of Dasgupta and Halperin [84].

B. XY Model with a Compact U(1) Gauge Field

Now we ease towards our aim of a duality analysis of Z_1 , by adding one layer of complexity to Z_{SC} . We make the gauge field in (9.57) compact by including a cosine Maxwell term [86]:

$$\mathcal{Z}_{M} = \prod_{j} \int_{0}^{2\pi} \frac{d\theta_{j}}{2\pi} \int_{0}^{2\pi} \frac{dA_{j\mu}}{2\pi} \exp\left(\frac{1}{e^{2}} \sum_{\Box} \cos\left(\epsilon_{\mu\nu\lambda} \Delta_{\nu} A_{j\lambda}\right) + \frac{1}{g} \sum_{j,\mu} \cos\left(\Delta_{\mu} \theta_{j} - A_{j\mu}\right)\right).$$
(9.65)

The Dasgupta-Halperin duality mapping can be easily extended to this theory. We now write both cosine terms in their Villain forms, and then proceed as described above. The results (9.59) and (9.62) continue to have the same form, with the only change being that the fields $P_{\bar{j}\mu}$ and $\varphi_{\bar{j}}$ are now also *integer* valued (and so must be summed over). Promoting these integer valued fields to real fields by the Poisson summation method following [57], we now have to introduce *two* fugacities: a vortex fugacity y_v (as before), and a monopole fugacity \tilde{y}_m (discussed below). Consequently, $\mathcal{Z}_{SC,3}$ in (9.63) now takes the form

$$\mathcal{Z}_{M,3} = \prod_{\bar{j}} \int_{-\infty}^{\infty} db_{\bar{j}\mu} \int_{-\infty}^{\infty} d\varphi_{\bar{j}} \int_{-\infty}^{\infty} d\vartheta_{\bar{j}} \exp\left(-\frac{e^2}{2} \sum_{\bar{j},\mu} \left(b_{\bar{j}\mu} - \Delta_{\mu}\varphi_{\bar{j}}\right)^2 - \frac{g}{2} \sum_{\Box} \left(\epsilon_{\mu\nu\lambda}\Delta_{\nu}b_{\bar{j}\lambda}\right)^2 + y_v \sum_{\bar{j},\mu} \cos\left(2\pi b_{\bar{j}\mu} - \Delta_{\mu}\vartheta_{\bar{j}}\right) + \tilde{y}_m \sum_{\bar{j}} \cos\left(2\pi\varphi_{\bar{j}} - \vartheta_{\bar{j}}\right)\right).$$
(9.66)

Again, the positions of the $\vartheta_{\bar{j}}$ above are dictated by gauge invariance, and the effect of the vortex and monopole fugacities is to soften the integer value constraints on the $b_{\bar{j}\mu}$ and $\varphi_{\bar{j}}$. Proceeding as described below (9.63), we work in the gauge $\varphi_{\bar{j}} = 0$, and to leading order in y_v , \tilde{y}_m replace $b_{\bar{j}\mu}$ by its saddle point value in the Gaussian part of the action, which remains $\bar{b}_{\bar{j}\mu} = 0$. Then, instead of (9.64), we obtain

$$\mathcal{Z}_{M,4} = \prod_{\bar{j}} \int_{-\infty}^{\infty} d\vartheta_{\bar{j}} \exp\left(y_v \sum_{\bar{j},\mu} \cos\left(\Delta_{\mu}\vartheta_{\bar{j}}\right) + \tilde{y}_m \sum_{\bar{j}} \cos\left(\vartheta_{\bar{j}}\right)\right).$$
(9.67)

We see that the new second term in (9.67) acts like an ordering field on the dual XY model. Taking the continuum limit as was done below (9.64) using [85] a complex field ψ conjugate to $e^{i\vartheta_{\bar{j}}}$, now instead of $\mathcal{Z}_{SC,dual}$ in (9.58) we obtain [87,88]

$$\mathcal{Z}_{M,\text{dual}} = \int \mathcal{D}\psi(r,\tau) \exp\left(-\int d^2r d\tau \left(|\partial_\mu \psi|^2 + \overline{s}|\psi|^2 + \frac{\overline{u}}{2}|\psi|^4 - y_m(\psi+\psi^*)\right)\right).$$
(9.68)

The new term proportional to y_m has the interpretation of a monopole fugacity. The compact gauge field now permits Dirac monopoles, which are points in spacetime at which vortex loops of the 'superconductor' can end: hence y_m is coupled to the creation and annihilation operators for the dual boson ψ *i.e.* the vortices. In the form (9.68) it is also clear that y_m acts like an ordering field in the dual XY model. We expect that such an XY model has no phase transition, and $\langle \psi \rangle \neq 0$ for all \bar{s} . So the presence of monopoles has destroyed the 'superconducting' phase. Comparing the properties of (9.58) and (9.68) we therefore conclude that making the gauge field compact in \mathcal{Z}_{SC} in (9.57) is a strongly relevant perturbation: the inverted XY transition of \mathcal{Z}_{SC} is destroyed in the resulting model \mathcal{Z}_M .

C. Berry Phases

We are finally ready to face Z_1 , and add in the final layer of complication of the Berry phases. Again, the Dasgupta-Halperin duality can be extended by combining it with the methods of Sect. 9.4.1 (this was partly discussed in [51]). Now the monopoles carry Berry phases [19, 89], and these lead to cancellations among many monopole configurations. In the long-wavelength limit it turns out that the only important configurations are those in which the total monopole magnetic charge is q times the charge of the elementary monopole [19, 20, 89]. Here q is the smallest positive integer such that

$$e^{i\pi Sq} = 1,$$
 (9.69)

i.e. q = 4 for S half an odd integer, q = 2 for S an odd integer, and q = 1 for S an even integer. Using the physical interpretation of (9.68), we therefore conclude that the monopole fugacity term should be replaced by one in which the monopoles are created and annihilated in multiples of q; the dual theory of \mathcal{Z}_1 in (9.56) then becomes

$$\mathcal{Z}_{1,\text{dual}} = \int \mathcal{D}\psi(r,\tau) \exp\left(-\int d^2 r d\tau \left(|\partial_\mu \psi|^2 + \overline{s}|\psi|^2 + \frac{\overline{u}}{2}|\psi|^4 - y_{mq}(\psi^q + \psi^{*q})\right)\right).$$
(9.70)

An explicit derivation of the mapping from Z_1 to $Z_{1,\text{dual}}$ can be obtained by an extension of the methods described above for Z_{SC} and Z_M . We express the Berry phase term using the 'background field' $a_{j\mu}^0$ in (9.46), and then we find that $Z_{\text{SC},2}$ in (9.62) is now replaced by

$$\mathcal{Z}_{1,2} = \sum_{\{b_{\bar{\jmath}\mu}\}} \sum_{\{\varphi_{\bar{\jmath}}\}} \exp\left(-\frac{e^2}{2} \sum_{\bar{\jmath},\mu} \left(b_{\bar{\jmath}\mu} - \Delta_{\mu}\varphi_{\bar{\jmath}} - 2Sa^0_{\bar{\jmath}\mu}\right)^2 -\frac{g}{2} \sum_{\Box} \left(\epsilon_{\mu\nu\lambda}\Delta_{\nu}b_{\bar{\jmath}\lambda}\right)^2\right). \quad (9.71)$$

Notice that, as in Sect. 9.4.1, the Berry phases appear as offsets in the dual action. We now promote the integer field $b_{\bar{j}\mu}$ and $\varphi_{\bar{j}}$ to real fields by the Poisson summation method (just as in (9.66)), at the cost of introducing vortex and monopole fugacities. The final steps, following the procedure below (9.66), are to transform to the gauge $\varphi_{\bar{j}} = 0$, and to then set the 'Higgsed' dual gauge field $b_{\bar{j}\mu}$ to its saddle point value determined from the Gaussian terms in the action. It is the latter step which is now different, and the presence of the $a_{\bar{j}\mu}^0$ now implies that the saddle point value $\bar{b}_{\bar{j}\mu}$ will be non-zero and site dependent. Indeed, it is crucial that the saddle point be determined with great care, and that the square lattice symmetry of the underlying problem be fully respected. This saddle point determination is in many ways analogous to the computation in Sect. III.B of [20], and it is important that all the modes on the lattice scale be fully identified in a similar manner. The similarity to [20] becomes clear after using the parameterization in (9.49) for $a_{\bar{j}\mu}^0$ in terms of the $\mathcal{X}_{\bar{j}}$ and the $\mathcal{Y}_{j\mu}$ shown in Fig. 9.9. Finally, after transforming $b_{\bar{j}\mu} \to b_{\bar{j}\mu} + 2S\Delta_{\mu}\mathcal{X}_{\bar{j}}$ and $\vartheta_{\bar{j}} \to \vartheta_{\bar{j}} + 4\pi S\mathcal{X}_{\bar{j}}$, we obtain from (9.71)

$$\mathcal{Z}_{1,3} = \prod_{\bar{j}} \int_{-\infty}^{\infty} db_{\bar{j}\mu} \int_{-\infty}^{\infty} d\vartheta_{\bar{j}} \exp\left(-\frac{e^2}{2} \sum_{\bar{j},\mu} \left(b_{\bar{j}\mu} - 2S\epsilon_{\mu\nu\lambda}\Delta_{\nu}\mathcal{Y}_{\lambda}\right)^2 - \frac{g}{2} \sum_{\Box} \left(\epsilon_{\mu\nu\lambda}\Delta_{\nu}b_{\bar{j}\lambda}\right)^2 + y_v \sum_{\bar{j},\mu} \cos\left(2\pi b_{\bar{j}\mu} - \Delta_{\mu}\vartheta_{\bar{j}}\right) + \tilde{y}_m \sum_{\bar{j}} \cos\left(\vartheta_{\bar{j}} + 4\pi S\mathcal{X}_{\bar{j}}\right)\right).$$
(9.72)

Now, the saddle point value of the massive field $b_{\bar{j}\mu}$ is easily determined from the first terms in (9.72), yielding

$$\bar{b}_{\bar{j}\mu} = \alpha \epsilon_{\mu\nu\lambda} \Delta_{\nu} \mathcal{Y}_{j\lambda}. \tag{9.73}$$

where $\alpha \equiv 2Se^2/(e^2 + 8g)$. Note that only the spatial components of $\bar{b}_{\bar{j}\mu}$ are non-zero, and these have the simple structure of Fig. 9.9c. In particular, the magnitude of the $\bar{b}_{\bar{j}\mu}$ are the same on all the spatial links, and the use of (9.49) was crucial in obtaining this appealing result. With this saddle point value, (9.72) simplifies to the following model for the field $\vartheta_{\bar{j}}$ only (this is the form of (9.67) after accounting for Berry phases):

$$\mathcal{Z}_{1,4} = \prod_{\bar{j}} \int_{-\infty}^{\infty} d\vartheta_{\bar{j}} \exp\left(y_v \sum_{\bar{j},\mu} \cos\left(\Delta_{\mu} \vartheta_{\bar{j}} - 2\pi \bar{b}_{\bar{j}\mu}\right) + \tilde{y}_m \sum_{\bar{j}} \cos\left(\vartheta_{\bar{j}} + 4\pi S \mathcal{X}_{\bar{j}}\right)\right). \quad (9.74)$$

The most important property of this dual XY model is the nature of the ordering field in the last term of (9.74). For S = 1/2, notice from Fig. 9.9a that this field is oriented north/east/south/west on the four sublattices in of the dual lattice in Fig. 9.9c. So if we take a naive continuum limit, the average field vanishes! This is the key effect responsible for the cancellations among monopole configurations induced by Berry phases noted earlier; in the dual formulation, the Berry phases have appeared in differing orientations of the dual ordering field. The XY model in (9.74) also has the contribution from $\bar{b}_{\bar{j}\mu}$, which appear as a 'staggered flux' acting on the $\vartheta_{\bar{j}}$ (see Fig. 9.9c), but we now show that this is not as crucial in the continuum limit.

Before we take the continuum limit of $\mathcal{Z}_{1,4}$, we discuss its implementation of the square lattice symmetries. In particular, we are interested in the Z_4 symmetry which rotates the four sublattices in Fig. 9.9c into each other, as the values of $\mathcal{X}_{\bar{\jmath}}$ seem to distinguish between them. Let us consider the symmetry \mathcal{R}_n which rotates lattice anticlockwise by an angle $n\pi/2$ about the direct lattice point at the center of a plaquette in Fig. 9.9c, associated with the transformation in (9.53). It is easy to see that $\mathcal{Z}_{1,4}$ remains invariant under \mathcal{R}_n provided we simultaneously rotate the angular variables $\vartheta_{\bar{\jmath}}$:

$$\mathcal{R}_n: \quad \vartheta_{\bar{\jmath}} \to \vartheta_{\bar{\jmath}} + nS\pi.$$
 (9.75)

It is now useful to introduce complex variables which realize irreducible representations of this Z_4 symmetry. We divide the lattice into plaquettes like those in Fig. 9.9c, and for each plaquette we define variables ψ_p , with pinteger, by

$$\psi_p = \frac{1}{2} \left(e^{i\vartheta_W} + e^{ip\pi/2} e^{i\vartheta_X} + e^{ip\pi} e^{i\vartheta_Y} + e^{i3p\pi/2} e^{i\vartheta_Z} \right). \tag{9.76}$$

Note that we need only use p = 0, 1, 2, 3 because ψ_p depends only on $p \pmod{4}$. Under the symmetry \mathcal{R}_n we clearly have

$$\mathcal{R}_n: \quad \psi_p \to e^{in(2S-p)\pi/2}\psi_p; \tag{9.77}$$

the factor of $e^{inS\pi}$ arises from (9.75), and that of $e^{-inp\pi/2}$ from the real-space rotation of the lattice points. Note that only for p = 2S is ψ_p invariant under \mathcal{R}_n , and this is consistent with the fact that it is ψ_{2S} which appears in $\mathcal{Z}_{1,4}$ as the ordering field term. Let us now write the action in $\mathcal{Z}_{1,4}$ in terms of these new variables. Ignoring the spacetime variation from plaquette to plaquette, the action per plaquette is

$$S_{1,4} = -2y_v \sum_{p=0}^{3} \left[\cos\left(\pi (p-\alpha)/2\right) |\psi_p|^2 \right] - \tilde{y}_m \left(\psi_{2S} + \psi_{2S}^*\right) + \dots$$
(9.78)

Here the ellipses represent other allowed terms, all consistent with the symmetry (9.77), which must be included to implement the (softened) constraints on ψ_p arising from (9.76) and the fact that the $e^{i\vartheta_{\bar{j}}}$ are unimodular. Apart from ψ_{2S} , for which there is already an ordering field in the action, the condensation of any of the other ψ_p breaks the lattice symmetry (9.77), and so drives a quantum phase transition to the bond-ordered state. The choice among the ψ_p is controlled by the coefficient of the y_v term in (9.78), and we choose the value of $p \neq 2S$ for which $\cos(\pi(\alpha + p)/2)$ is a maximum. We are interested in the large g paramagnetic phase, and here α is small, and the appropriate value is p = 0. The resulting continuum theory for $\psi = \psi_0$ then must be invariant under (9.77), and it is easily seen that this has just the form $\mathcal{Z}_{1,\text{dual}}$ in (9.70) with q determined by (9.69). Other choices of p for the order parameter lead to different types of bond order, with a ground state degeneracy smaller or larger than the q in (9.69); such states have partial or additional bond order, and are clearly possible in general. However, our analysis of the paramagnetic states in Sect. 9.4.1 indicates that a choice $\psi = \psi_{p\neq 0}$ is unlikely for the models under consideration here, and we will not consider this case further here.

We have now completed our promised derivation of the model $\mathcal{Z}_{1,\text{dual}}$ in (9.70) dual to the N = 1 lattice gauge theory model \mathcal{Z}_1 in (9.56). Rather than being an XY model in a field (as in (9.68)), $\mathcal{Z}_{1,\text{dual}}$ is an XY model with a *q*-fold anisotropy. This anisotropy encapsulates the *q*-fold binding of monopoles claimed earlier. In the language of (9.74) the average ordering **Table 9.1.** Summary of the duality mappings for N = 1. Only the Lagrangean's are specified, and a summation/integration of these over spacetimes is implicit. The fixed field $\eta_j = \pm 1$ in the Berry phase in the third row is the sublattice staggering factor in (9.2). The integer q in the third row is specified in (9.69). For S = 1/2, we have q = 4, and then the y_{mq} perturbation is dangerously irrelevant. Hence the critical theory for the model with monopoles and Berry phases in the third row, is identical to that for the first row

N = 1	
Direct lattice model	Dual model
$\mathcal{L}_{SC} = (1/(2e^2)) \left(\epsilon_{\mu\nu\lambda} \Delta_{\nu} A_{j\lambda}\right)^2 - (1/g) \cos\left(\Delta_{\mu} \theta_j - A_{j\mu}\right)$	$\mathcal{L}_{SC, ext{dual}} = \partial_{\mu}\psi ^2 + \overline{s} \psi ^2 + \frac{\overline{u}}{2} \psi ^4$
$\mathcal{L}_{M} = -(1/e^{2})\cos\left(\epsilon_{\mu\nu\lambda}\Delta_{\nu}A_{j\lambda}\right) - (1/g)\cos\left(\Delta_{\mu}\theta_{j} - A_{j\mu}\right)$	$egin{aligned} \mathcal{L}_{M, ext{dual}} &= \partial_{\mu}\psi ^2 + \overline{s} \psi ^2 + rac{\overline{u}}{2} \psi ^4 \ &- y_m(\psi+\psi^*) \end{aligned}$
$\mathcal{L}_{1} = -(1/e^{2})\cos\left(\epsilon_{\mu\nu\lambda}\Delta_{\nu}A_{j\lambda}\right) - (1/g)\cos\left(\Delta_{\mu}\theta_{j} - A_{j\mu}\right) - i2S\eta_{j}A_{j\tau}$	$\mathcal{L}_{1,\text{dual}} = \partial_{\mu}\psi ^2 + \overline{s} \psi ^2 + \frac{\overline{u}}{2} \psi ^4 - y_{mq}(\psi^q + \psi^{*q})$

fields on the $\vartheta_{\bar{j}}$ oscillate from site to site and cancel out, and only the qth moment of the field survives. Now the combined effect of the monopoles and Berry phases in \mathcal{Z}_1 is decided by the term proportional to y_{mq} . In the paramagnetic phase of the direct model, which is $\bar{s} < \bar{s}_c$ and $\langle \psi \rangle \neq 0$, this q-fold anisotropy is certainly very important. For S = 1/2, q = 4 it orders the ψ field along four particular angles, and these are easily shown to be [51] one of the four degenerate bond-ordered states in Fig. 9.11. However, at the critical point $\bar{s} = \bar{s}_c$ it is known that this 4-fold anisotropy is irrelevant [90]: so in $\mathcal{Z}_{1,\text{dual}}$ the monopoles can be neglected at the critical point $s = s_c$, but not away from it.

We have now achieved the desired objective of this subsection. Compactness alone was a strongly relevant perturbation on the model of a scalar field coupled to electromagnetism in Z_{SC} . However, when we combined compactness with the Berry phases in Z_1 , then we found that the monopoles effectively cancelled each other out at the critical point for S = 1/2. Consequently the
theory for the critical point in Z_1 is identical to the theory for the critical point in Z_{SC} , and this is the simple inverted XY model $Z_{SC,dual}$ in (9.58). The results of this subsection are summarized in Table 1.

4.2.2 Easy Plane Model at N = 2

A second explicit example of the remarkable phenomenon described above is provided by the physically relevant N = 2 case of the model of central interest, \mathcal{Z} in (9.40), but in the presence of an additional spin-anisotropy term preferring that the spins lie within the XY plane. In such a situation, we may write the complex spinor z_{ja} as

$$z_{ja} = \frac{1}{\sqrt{2}} \begin{pmatrix} e^{i\theta_{j\uparrow}} \\ e^{i\theta_{j\downarrow}} \end{pmatrix}, \qquad (9.79)$$

so that the action is expressed in terms of *two* angular fields, θ_{\uparrow} and θ_{\downarrow} . Inserting (9.79) in (9.40), we obtain a generalization of the N = 1 model Z_1 in (9.56):

$$\mathcal{Z}_{2} = \prod_{j} \int_{0}^{2\pi} \frac{d\theta_{j\uparrow}}{2\pi} \int_{0}^{2\pi} \frac{d\theta_{j\downarrow}}{2\pi} \int_{0}^{2\pi} \frac{dA_{j\mu}}{2\pi} \exp\left(\frac{1}{e^{2}} \sum_{\Box} \cos\left(\epsilon_{\mu\nu\lambda} \Delta_{\nu} A_{j\lambda}\right) + \frac{1}{2g} \sum_{j,\mu,a} \cos\left(\Delta_{\mu} \theta_{ja} - A_{j\mu}\right) + i2S \sum_{j} \eta_{j} A_{j\tau}\right).$$
(9.80)

As in (9.56), we have chosen to explicitly include a Maxwell term for the U(1) gauge field as it proves convenient in the subsequent duality analysis. The model Z_2 provides a complete description of the phases of the square lattice antiferromagnet (9.28) with an additional easy-plane anisotropy term.

We can now proceed with a duality analysis of (9.80) using methods precisely analogous to those discussed in Sect. 9.4.2.1: the only difference is we now have two angular fields $\theta_{a=\uparrow,\downarrow}$, and so certain fields come with two copies. We will therefore not present any details, and simply state the series of results which appear here, which closely parallel those obtained above for N = 1.

• Neglecting *both* compactness of the U(1) gauge field and the Berry phases, it is straightforward to take the continuum limit of Z_2 in its direct representation, and we obtain the theory Z_c in (9.55), but with an additional spin-anisotropy term

$$\mathcal{Z}_{2c} = \int \mathcal{D}z_{a}(r,\tau) \mathcal{D}A_{\mu}(r,\tau) \exp\left(-\int d^{2}r d\tau \left[|(\partial_{\mu} - iA_{\mu})z_{a}|^{2} + s|z_{a}|^{2} + \frac{u}{2}(|z_{a}|^{2})^{2} + v|z_{\uparrow}|^{2}|z_{\downarrow}|^{2} + \frac{1}{4e^{2}}(\epsilon_{\mu\nu\lambda}\partial_{\nu}A_{\lambda})^{2} \right] \right),$$
(9.81)

where v > 0 prefers spins in the easy plane. We can carry through the analog of the duality mapping between (9.57) and (9.58), and instead of (9.58) we now obtain a theory for *two* dual fields ψ_a representing vortices in θ_{\uparrow} and θ_{\downarrow} [82]

$$\mathcal{Z}_{2c,\text{dual}} = \int \mathcal{D}\psi_a(r,\tau)\mathcal{D}B_\mu(r,\tau) \exp\left(-\int d^2r d\tau \left[|(\partial_\mu - iB_\mu)\psi_\uparrow|^2 + |(\partial_\mu + iB_\mu)\psi_\downarrow|^2 + \overline{s}|\psi_a|^2 + \frac{\overline{u}}{2}(|\psi_a|^2)^2 + \overline{v}|\psi_\uparrow|^2|\psi_\downarrow|^2 + \frac{1}{4\overline{e}^2}(\epsilon_{\mu\nu\lambda}\partial_\nu B_\lambda)^2 \right] \right).$$
(9.82)

Note that there is now a non-compact U(1) gauge field B_{μ} which survives the continuum limit: this field arises from the analog of the field $b_{\bar{j}\mu}$ in (9.63), and here it is not completely Higgsed out. The most remarkable property of (9.82) is that it is identical in structure to (9.81): the actions are identical under the mapping $z_{\uparrow} \to \psi_{\uparrow}, z_{\downarrow} \to \psi_{\downarrow}^*$, and $A_{\mu} \to B_{\mu}$. In other words, the theory \mathcal{Z}_{2c} is self-dual [82].

• As in Sect. 9.4.2.1, we next make the A_{μ} gauge field compact, but continue to ignore Berry phases *i.e.* we perform a duality analysis on (9.80), in the absence of the last term in the action. Now, instead of (9.68), (9.82) is modified to

$$\mathcal{Z}_{2M,\text{dual}} = \int \mathcal{D}\psi_a(r,\tau)\mathcal{D}B_\mu(r,\tau) \exp\left(-\int d^2r d\tau \left[|(\partial_\mu - iB_\mu)\psi_\uparrow|^2 + |(\partial_\mu + iB_\mu)\psi_\downarrow|^2 + \overline{s}|\psi_a|^2 + \frac{\overline{u}}{2}(|\psi_a|^2)^2 + \overline{v}|\psi_\uparrow|^2|\psi_\downarrow|^2 + \frac{1}{4\overline{e}^2}(\epsilon_{\mu\nu\lambda}\partial_\nu B_\lambda)^2 - y_m(\psi_\uparrow\psi_\downarrow + \psi_\downarrow^*\psi_\uparrow^*)\right] \right).$$
(9.83)

The last term represents the influence of monopoles, and these now have the effect of turning a ψ_{\uparrow} vortex into a ψ_{\downarrow} vortex [80–82]. Again, as in (9.68), the y_m term in (9.83) is clearly a strongly relevant perturbation to $\mathcal{Z}_{2c,\text{dual}}$ in (9.82). It ties the phases of ψ_{\uparrow} and ψ_{\downarrow} to each other, so that (9.83) is effectively the theory of a *single* complex scalar coupled to a non-compact U(1) gauge field B_{μ} . However, we have already considered such a theory in the *direct* representation in (9.57). We can now move from the dual representation in (9.83) back to the direct representation, by the mapping between (9.57) and (9.58). This leads to the conclusion, finally, that the theory (9.83) is dual to an ordinary XY model. In other words, the theory \mathcal{Z}_2 in (9.80) without its Berry phase term is an XY model. However, this is precisely the expected conclusion, and could have been easily reached without this elaborate series of duality mappings: just integrating over $A_{j\mu}$ for large e^2 yields an XY model in the angular field $\theta_{\uparrow}-\theta_{\downarrow},$ which represents the orientation of the physical in-plane Néel order.

• Finally, let us look at the complete theory Z_2 . An explicit duality mapping can be carried out, and as in (9.70), the action (9.83) is replaced by [52, 71, 80, 81]

$$\begin{aligned} \mathcal{Z}_{2M,\text{dual}} &= \int \mathcal{D}\psi_a(r,\tau) \mathcal{D}B_\mu(r,\tau) \exp\left(-\int d^2 r d\tau \left[|(\partial_\mu - iB_\mu)\psi_\uparrow|^2 + |(\partial_\mu + iB_\mu)\psi_\downarrow|^2 + \overline{s}|\psi_a|^2 + \frac{\overline{u}}{2}(|\psi_a|^2)^2 + \overline{v}|\psi_\uparrow|^2|\psi_\downarrow|^2 + \frac{1}{4\overline{e}^2}(\epsilon_{\mu\nu\lambda}\partial_\nu B_\lambda)^2 - y_{mq}\left((\psi_\uparrow\psi_\downarrow)^q + (\psi_\downarrow^*\psi_\uparrow^*)^q\right) \right] \right), (9.84) \end{aligned}$$

where the integer q was defined in (9.69). The subsequent reasoning is the precise analog of that for N = 1. For S = 1/2 and q = 4, the term proportional to y_{mq} representing q-fold monopole is irrelevant at the critical point (but not away from it in the paramagnetic phase). Consequently, the critical theory of (9.84) reduces to (9.82). So just as at N = 1, the *combined* influence of monopoles and Berry phases is dangerously irrelevant at the critical point, and for the critical theory we can take a naive continuum limit of Z_2 neglecting both the Berry phases and the compactness of the gauge field.

We have now completed our discussion of the N = 2 easy plane model and established the existence of the same remarkable phenomenon found in Sect. 9.4.2.1 for N = 1, and claimed more generally [80,81] at the beginning of Sect. 9.4.2 as the justification for the critical theory (9.55). As we saw in some detail in Sect. 9.4.1, monopoles, and attendant Berry phases, are absolutely crucial in understanding the onset of confinement and bond order in the paramagnetic phase. However, for S = 1/2, the Berry phases induce a destructive quantum interference between the monopoles at the quantum critical point, leading to a critical theory with 'deconfined' spinons and a non-compact U(1) gauge field which does not allow monopoles. These results are summarized in Table 2.

The results in Tables 1 and 2 can be generalized to arbitrary values of N, for models with the analog of an 'easy plane' anisotropy: as in (9.79), all the z_a have equal modulus and are expressed in terms of $a = 1 \dots N$ angles θ_a . The dual models have N vortex fields ψ_a , and N - 1 non-compact U(1) gauge fields $B_{b\mu}$, $b = 1 \dots (N-1)$. For $a = 1 \dots (N-1)$, the field ψ_a has a charge +1 under the gauge field with b = a, and is neutral under all gauge fields with $b \neq a$. For a = N, the field ψ_N , has a charge -1 under all N - 1 gauge fields. (This gauge structure is similar to that found in 'moose' field theories [91].) The dual representation of the monopole operator is $\prod_{a=1}^{N} \psi_a$, and this appears as the coefficient of y_m (notice that this operator is neutral under all the gauge fields). The q^{th} power of this operator appears as the

Table 9.2. As in Table 1, but for the N = 2 easy plane case. The index *a* extends over the two values \uparrow , \downarrow . Again for S = 1/2, q = 4, the critical theory for the third row is the same as that for the first row. The dual model in the second row is effectively the theory of a single complex scalar coupled to a non-compact U(1) gauge field B_{μ} ; by the inverse of the duality mapping in the first row of Table 1, this theory has a direct XY transition

N = 2, easy plane					
Direct lattice model	Dual model				
$\mathcal{L}_{2,SC} = (1/(2e^2)) \left(\epsilon_{\mu\nu\lambda} \Delta_{\nu} A_{j\lambda}\right)^2 - (1/g) \cos\left(\Delta_{\mu} \theta_{ja} - A_{j\mu}\right)$	$\mathcal{L}_{2SC,\text{dual}} = (\partial_{\mu} - iB_{\mu})\psi_{\uparrow} ^{2} + (\partial_{\mu} + iB_{\mu})\psi_{\downarrow} ^{2} + \overline{s} \psi_{a} ^{2} + \frac{\overline{u}}{2} \left(\psi_{a} ^{2}\right)^{2} + \overline{v} \psi_{\uparrow} ^{2} \psi_{\downarrow} ^{2} + \frac{1}{2\overline{e}^{2}} (\epsilon_{\mu\nu\lambda}\partial_{\nu}B_{\lambda})^{2}$				
$\mathcal{L}_{2M} = -(1/e^2) \cos \left(\epsilon_{\mu\nu\lambda} \Delta_{\nu} A_{j\lambda}\right) - (1/(2g)) \cos \left(\Delta_{\mu} \theta_{ja} - A_{j\mu}\right)$	$\mathcal{L}_{2M,\text{dual}} = \left (\partial_{\mu} - iB_{\mu})\psi_{\uparrow} \right ^{2} + \left (\partial_{\mu} + iB_{\mu})\psi_{\downarrow} \right ^{2} + \overline{s} \psi_{a} ^{2} + \frac{\overline{u}}{2} \left(\psi_{a} ^{2} \right)^{2} + \overline{v} \psi_{\uparrow} ^{2} \psi_{\downarrow} ^{2} + \frac{1}{2\overline{e}^{2}} (\epsilon_{\mu\nu\lambda}\partial_{\nu}B_{\lambda})^{2} - y_{m} \left(\psi_{\uparrow}\psi_{\downarrow} + \psi_{\uparrow}^{*}\psi_{\downarrow}^{*} \right)$				
$\mathcal{L}_2 = -(1/e^2)\cos\left(\epsilon_{\mu\nu\lambda}\Delta_{\nu}A_{j\lambda}\right) -(1/g)\cos\left(\Delta_{\mu}\theta_{ja} - A_{j\mu}\right) - i2S\eta_j A_{j\tau}$	$\mathcal{L}_{2,\text{dual}} = (\partial_{\mu} - iB_{\mu})\psi_{\uparrow} ^{2} + (\partial_{\mu} + iB_{\mu})\psi_{\downarrow} ^{2} + \overline{s} \psi_{a} ^{2} + \frac{\overline{u}}{2} (\psi_{a} ^{2})^{2} + \overline{v} \psi_{\uparrow} ^{2} \psi_{\downarrow} ^{2} + \frac{1}{2\overline{c}^{2}}(\epsilon_{\mu\nu\lambda}\partial_{\nu}B_{\lambda})^{2} - y_{mq} ((\psi_{\uparrow}\psi_{\downarrow})^{q} + (\psi_{\uparrow}^{*}\psi_{\downarrow}^{*})^{q})$				

coefficient of y_{mq} . Note that the monopole operators involves a product of N fields, and for large enough N, both y_m and y_{mq} can be expected to be irrelevant perturbations at the quantum critical point.

Finally, these analyses of \mathcal{Z} in (9.40) can be complemented by a study of its $N \to \infty$ limit, without any easy-plane anisotropy. This was carried out some time ago [51, 92], and it was found that monopoles were dangerously irrelevant at the quantum critical point, *both* with or without Berry phases (as noted above for large N in the easy plane case). It is important to note that the situation at large N is subtly different from that for N = 1, 2: in the latter case, monopoles are dangerously irrelevant in the presence of S = 1/2 Berry phases, but relevant without Berry phases. The key understanding of this distinction emerged in the recent work of Senthil *et al.* [80, 81], which finally succeeded in placing the earlier large N results within the context of dual theories of topological defects in statistical mechanics.

9.5 Triangular Lattice Antiferromagnet

We continue our analysis of quantum antiferromagnets with an odd number of S = 1/2 spins per unit cell, but consider a class qualitatively different from those in Sect. 9.4. One of the defining properties of the models of Sect. 9.4 was that the magnetically ordered Néel state was defined by (9.2): the average magnetic moment on all sites were collinear, and only a single vector **n** was required to specify the orientation of the ground state. This section shall consider models in which the moments are *non-collinear*; the triangular lattice is the canonical example. However, similar results should also apply to other two-dimensional lattices with non-collinear ground states, such as the distorted triangular lattice found in Cs₂CuCl₄ [11].

We consider the model (9.28), but with the spins residing on the sites of the triangular lattice. This has a magnetically ordered state illustrated in Fig. 9.13; for this state (9.2) is replaced by

$$\langle \mathbf{S}_j \rangle = N_0 \left(\mathbf{n}_1 \cos(\mathbf{Q} \cdot \mathbf{r}) + \mathbf{n}_2 \sin(\mathbf{Q} \cdot \mathbf{r}) \right).$$
(9.85)

Here $\mathbf{Q} = 2\pi(1/3, 1/\sqrt{3})$ is the ordering wavevector, and $\mathbf{n}_{1,2}$ are two arbitrary orthogonal unit vectors in spin space

$$\mathbf{n}_1^2 = \mathbf{n}_2^2 = 1$$
 ; $\mathbf{n}_1 \cdot \mathbf{n}_2 = 0.$ (9.86)

A distinct ground state, breaking spin rotation symmetry, is obtained for each choice of $\mathbf{n}_{1,2}$.

We now wish to allow the values of $\mathbf{n}_{1,2}$ to fluctuate quantum mechanically across spacetime, ultimately producing a paramagnetic state. As in Sect. 9.4, we should account for the Berry phases of each spin while setting up the effective action: an approach for doing this is presented in Sect. VI of [52]. However, the full structure of the critical theory is not understood in all cases, as we describe below.

One possible structure of the paramagnetic state is a confining, bondordered state, similar to that found in Sect. 9.4. However, there is no complete theory for a possible direct second-order transition from a non-collinear magnetically ordered state to such a paramagnet. Ignoring Berry phases, one could define the complex field $\Phi_{\alpha} = n_{1\alpha} + in_{2\alpha}$, which, by (9.86), obeys



Fig. 9.13. Quantum phase transition described by \mathcal{Z}_w in (9.90) as a function of s. The state on the left has non-collinear magnetic order described by (9.85). The state on the right is a 'resonating valence bond' (RVB) paramagnet with topological order and fractionalized S = 1/2 neutral spinon excitations (one spinon is shown above). Such a magnetically ordered state is observed in Cs₂CuCl₃ [11, 12], and there is evidence that the higher energy spectrum can be characterized in terms of excitations of the RVB state [24].

 $\Phi_{\alpha}^2 = 0$, and then proceed to write down an effective action with the structure of (9.14). However, it is clear that such a theory describes a transition to a paramagnetic phase with a doublet of S = 1 triplet quasiparticles, and we can reasonably expect that such a phase has spontaneous bond order (in contrast to the explicit dimerization in the models of Sect. 9.2). Berry phases surely play an important role in inducing this bond order (as they did in Sect. 9.4.1), but there is no available theory for how this happens in the context of (9.14). Indeed, it is possible that there is no such direct transition between the non-collinear antiferromagnet and the bond-ordered paramagnet, and resolving this issue remains an important open question.

In contrast, it is possible to write down a simple theory for a direct transition between the non-collinear antiferromagnet and a paramagnetic phase not discussed so far: a resonating valence bond liquid [47,48,93] with *deconfined spinons and topological order*. This theory is obtained by observing that the constraints (9.86) can be solved by writing [94,95]

$$\boldsymbol{n}_1 + i\boldsymbol{n}_2 \equiv \epsilon_{ab} w_b \boldsymbol{\sigma}_{ac} w_c, \qquad (9.87)$$

where w_a is a 2 component complex spinor obeying $|w_{\uparrow}|^2 + |w_{\downarrow}|^2 = 1$. It is useful to compare (9.87) with (9.31), which parameterized a single vector also in terms of a complex spinor z_a . Whereas (9.31) was invariant under the U(1) gauge transformation (9.32), notice that (9.87) is only invariant under the Z_2 gauge transformation

$$w_a(r,\tau) \to \varrho(r,\tau) w_a(r,\tau),$$
(9.88)

where $\rho(r, \tau) = \pm 1$ is an arbitrary field which generates the gauge transformation. This Z_2 gauge transformation will play an important role in understanding the structure of the paramagnetic phase [21–23, 96, 97].

We can now study fluctuations of the non-collinear antiferromagnet by expressing the effective action in terms of the w_a . Apart from the familiar constraints of spin rotational invariance, and those imposed by (9.88), the effective action must also obey the consequences of translational invariance which follow from (9.85); the action must be invariant under

$$w_a \to w_a e^{-i\boldsymbol{Q}\cdot\boldsymbol{a}/2},\tag{9.89}$$

where a is any triangular lattice vector. In the continuum limit, this leads to the following effective action

$$\mathcal{Z}_{w} = \int \mathcal{D}w_{a}(r,\tau) \exp\left(-\int d^{2}r d\tau \left[|\partial_{\mu}w_{a}|^{2} + s|w_{a}|^{2} + \frac{u}{2}(|w_{a}|^{2})^{2}\right]\right);$$
(9.90)

notice there is a free integration over the w_a , and so we have softened the rigid length constraint. Comparing this with (9.55), we observe that the U(1) gauge field is now missing, and we simply have a Landau-Ginzburg theory for a 2 component complex scalar. The Z_2 gauge invariance (9.88) plays no role in this continuum critical theory for the destruction of non-collinear magnetic order, but as we discuss below, it will play an important role in the analysis of the paramagnetic phase. The theory (9.90) has a global O(4) invariance of rotations in the 4-dimensional space consisting of the real and imaginary parts of the 2 components of w_a : consequently the critical exponents of (9.90) are identical to those of the well known 4-component φ^4 field theory. Notice that there is no O(4) invariance in the microscopic theory, and this symmetry emerges only in the continuum limit [95,98]: the simplest allowed term which breaks this O(4) invariance is $|\epsilon_{ab}w_a\partial_{\mu}w_b|^2$, and this term is easily seen to be irrelevant at the critical point of the theory (9.90).

Let us now turn to a discussion of the nature of the paramagnetic phase obtained in the region of large positive s in (9.90). Here, the elementary excitations are free w_a quanta, and these are evidently S = 1/2 spinons. There is also a neutral, spinless topological excitation [21, 22, 99] whose importance was stressed in [97]: this is the 'vison' which is intimately linked with the Z_2 gauge symmetry (9.88). It is a point defect which carries Z_2 gauge flux. The vison has an energy gap in the paramagnetic phase, and indeed across the transition to the magnetically ordered state. This was actually implicit in our taking the continuum limit to obtain the action (9.90). We assumed that all important spin configurations could be described by a smooth, singlevalued field $w_a(r, \tau)$, and this prohibits vison defects around which the w_a are double-valued. It is also believed that the vison gap allows neglect of Berry phase effects across the transition described by (9.90): after duality, the Berry phases can be attached to monopoles and visons [97, 100], and these are suppressed in both phases of Fig. 9.13.

9.6 Conclusions

This article has described a variety of quantum phases of antiferromagnetic Mott insulators, and the transitions between them.

Let us first summarize the phases obtained in zero applied magnetic field, and transitions that can be tuned between them by varying the ratio of exchange constants in the Hamiltonian (experimentally, this can be achieved by applied pressure). The magnetically ordered states discussed were the collinear Néel state (shown in Figs. 9.4 and 9.12), and the non-collinear 'spiral' (shown in Fig. 9.13). We also found paramagnetic states which preserved spin rotation invariance and which had an energy gap to all excitations: these include the dimerized states (shown in Figs. 9.2 and 9.4), the related bond-ordered states which spontaneously break lattice symmetries (shown in Figs. 9.11 and 9.12), and the 'resonating valence bond' paramagnet with topological order and deconfined spinons (shown in Fig. 9.13). The continuous quantum phase transitions we found between these states were:

(a) the transition between the dimerized paramagnet and the collinear Néel state (both states shown in Fig. 9.4) was described by the theory S_{φ} in (9.12); (b) the transition between the dimerized paramagnet and a non-collinear magnetically ordered state was described by S_{Φ} in (9.14);

(c) the transition between the collinear Néel state and the paramagnet with spontaneous bond order (shown in Fig. 9.12) was described for S = 1/2 antiferromagnets by Z_c in (9.55);

(d) the transition between the state with non-collinear magnetic order and the RVB paramagnet (both states shown in Fig. 9.13) was described by \mathcal{Z}_w in (9.90).

We also mention here other quantum transitions of Mott insulators, which involve distinct paramagnets on *both* sides of the critical point. These we did not discuss in the present paper, but such transitions have been discussed in the literature:

(e) the transition between a paramagnet with spontaneous bond order (Fig. 9.12) and a RVB paramagnet (Fig. 9.13) is described by a compact U(1) lattice gauge theory with charge 2 Higgs fields (closely related to Z_1 in (9.56)), and is discussed in [81, 100, 101];

(f) transitions between paramagnets with different types of spontaneous bond order can be mapped onto transitions between different smooth phases of height models like (9.50), and are discussed in [102, 103]. Section 9.3 also considered quantum transitions that could be tuned by an applied magnetic field. We mainly considered the case of the coupled-dimer antiferromagnet, but very similar theories apply to the other states discussed above. The general theory has the structure of S_{Ψ} in (9.24), describing the Bose-Einstein condensation of the lowest non-zero spin quasiparticle excitation of the paramagnet. For the coupled dimer model this quasiparticle had S = 1, but an essentially identical theory would apply for cases with S = 1/2 spinon quasiparticles.

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10 Spin – Orbit – Topology, a Triptych

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Abstract. Transition metal oxides that realize s=1/2 or s=1 quantum spin systems with low dimensionality or geometrically restricted connectivity are often described using simple concepts. We will discuss superexchange rules, the realization of depleted or open topologies using the lone pair concept and the effect of charge/orbital ordering. These considerations will be widened by a brief overview of important materials based on copper-oxygen, vanadium-oxygen and titanium-oxygen coordinations trying to highlight systematic dependencies with respect to structural and electronic elements or properties, respectively.

10.1 Introduction and General Remarks

Providing an extensive and somewhat complete review on structural aspects and electronic properties of low-dimensional quantum spin systems would, in itself, require a whole book. This chapter will focus on inorganic materials avoiding the rather large number of quantum spin systems prepared by coordination chemistry.

Transition metal compounds (oxides and halides) have been investigated intensively in the community of solid state chemists and physicists for many decades. Structurally, this interest is based on the large number of local transition metal - oxygen coordinations and the flexibility in linkage of these groups [1]. Discussing the electronic or magnetic properties of these systems it is obvious that even this rich structural component cannot explain the large variability and richness of the observed phase diagrams and electronic properties. Ranging from insulating to metallic, from antiferromagnetic (AF), superconducting (SC) to even ferromagnetic (FM) phases and more exotic charge, spin or orbital ordering phenomena can be observed [2–4]. Important aspects of these systems that will be further discussed below are visualized in Fig. 10.1. The nanotube based on Na₂V₃O₇ shown in the center will be discussed in Sect. 10.4.7.

In first approximation the electronic properties of transition metal compounds are defined by the special character of the nonclosed d-electron shells of the transition metal ion. In Table 10.1 electronic configurations of transition metal ions are given. The crystalline electric fields of the surrounding oxygen atoms split the respective energy levels leading to a hierarchy of relevant



Fig. 10.1. Important aspects of quantum spin systems together with the projection of a nanotube based on $Na_2V_3O_7$ as an example of the structural versatility of transition metal oxides and halides [5]

Table 10.1. Most relevant transition metals and related electronic configurations

spin $1/2$						spin 1						
$\begin{array}{c} {\rm Cu}^{2+} & {\rm f} \\ {\rm V}^{4+} & {\rm f} \\ {\rm Nb}^{4+} & {\rm f} \\ {\rm Ti}^{3+} & {\rm f} \end{array}$	$1s^2 2 1s^2 2 1s^2 2 1s^2 2 1s^2 2 $	$s^{2} 2p^{6}$ $s^{2} 2p^{6}$ $s^{2} 2p^{6}$ $s^{2} 2p^{6}$ $s^{2} 2p^{6}$	3s2 3s2 3s2 3s2 3s2	${3 p^6 \over 3 p^6 \\ 3 p^6 \\ 3 p^6 \\ 3 p^6 }$	$\begin{array}{c} 3d^9\\ 3d^1\\ 3d^1\\ 3d^1\\ 3d^1\end{array}$	$\begin{array}{c} \mathrm{V}^{3+}\\ \mathrm{Ni}^{2+} \end{array}$	$\frac{1s^2}{1s^2}$	$\frac{2s^2}{2s^2}$	$\begin{array}{c} 2p^6\\ 2p^6 \end{array}$	$\frac{3s^2}{3s^2}$	$\frac{3p^6}{3p^6}$	$\begin{array}{c} 3d^2\\ 3d^8 \end{array}$

energy scales. This hierarchy, depending on the local geometrical and electronic configurations, sets the stage for effective low-energy spin/orbital models, e.g. s=1/2 Heisenberg spins chains or other quantum spin systems, like spin ladders or spin plaquettes. In Fig. 10.1. a sketch of crystalline electric field energy levels is given for $Cu^{2+} - 3d^9$. Please note that the transition metal ions discussed here all lead to the realization of a small (s=1/2, s=1) spin moment and a small or negligible orbital moment. These are preconditions for an overweight of quantum fluctuations at low temperatures.

10.1.1 Antiferromagnetic Correlations and Superconductivity

A large part of the motivation to study transition metal compounds with small spin and low dimensionality is based on the discovery of high temperature superconductivity (HTSC) by Bednorz and Müller in the cuprates [6,7]. This extraordinary effect, however, is just on example of correlation-induced phenomena. The essentially two-dimensional HTSC are based on a square-



Fig. 10.2. Crystalline electric field scheme or Jahn-Teller diagram of $Cu^{2+} - 3d^9$ in an isotropic, octahedral and tetragonally distorted local surrounding. The hole in the $d_{x^2-y^2}$ orbital determines its electronic properties

planar Cu-O-coordination with $3d^9 - s = 1/2$ of Cu^{2+} . The parent insulating compounds show long range Néel order. Hole doping, either by changing the oxygen content or the cation stoichiometry, destroys long range order and enables superconductivity with enormous T'_c s of up to 140 K [7]. The suppression of long range order in a doped 2D quantum antiferromagnet is based on the frustrating line of spin flips that a moving hole carries behind itself. The physics of HTSC is therefore not so much related to electron-phonon coupling as in conventional metallic superconductors [8] but to strong electronic correlations [9–12] on the restricted geometry of the CuO₂-plane. However, full details of this mechanism are presently not understood. For further theoretical aspects we refer, e.g. to other chapters of this book. An overview of inelastic light scattering experiments are given in [13].

The interplay of antiferromagnetic local correlations and superconductivity is also highlighted by the observation of an antiferromagnetic resonance mode in neutron scattering on several HTSC's [14–16]. This collective excitation with the scattering vector $q=(\pi,\pi)$ is strongly enhanced with decreasing temperature, but only in the superconducting state. In low dimensional doped systems with appreciable electronic correlations also charge ordered or spin-charge phase separated phases are expected. Evidence for such an electronic phase separation has been observed and intensively studied as stripe correlations in underdoped HTSC's [17, 18]. Also the related but non-superconducting square plane nickelate La_2NiO_4 shows stripe correlations. However, in the latter case these are more static [19, 20]. Stripe formation may also be understood as a spontaneous reduction of dimensionality of the system. The dimensionality of a spin system plays an important role for the effectiveness of fluctuations to destroy long range order [21, 22]. This argument is valid for a classical system. It becomes even more important for quantum spin systems [23]. In the presence of additional interactions as spin-phonon coupling or longer range (intersite) Coulomb interaction alternative instabilities determine the ground state. As a result spin dimerization or charge ordering may be observed.

10.1.2 Quantum Criticality and the Low Energy Spectrum of Quantum Spin Systems

Certain parts in the phase diagram of such systems attract our special attention [24–26]. These are given by the close proximity of competing phases with long range order or short range correlated states, e.g. Néel order and a spin liquid [4]. If a parameter like doping leads to a diminishing of long-range order and a dropping of the order temperature to zero, quantum critical behavior shows up. The dynamics of the system is governed even at finite but low temperatures by quantum fluctuations instead of thermal fluctuations (Fig. 10.3).

In Table 10.2 properties of such a system are summarized. Quantum criticality or the quantum critical point is a concept of broader validity [24–26]. In quantum spin systems the parameters doping, pressure, stoichiometry, magnetic field or exchange coupling parameters control the system. Competing interactions due to spin frustration are of special importance. Within our concept spin frustration in an antiferromagnet is a very appealing ansatz as a small change of the topology of the spin system or coupling constants may lead to a crossover or transition (strictly only at T=0) to a drastically different ground state.



Fig. 10.3. Sketch of a phase diagram of a quantum spin system in the proximity of the quantum critical point at $g = g_c$, with g a controlling parameter. The competing Néel and short range ordered spin liquid phase enclose a quantum critical regime with cross over phenomena at finite temperatures. Elementary triplet and spin wave excitations of the system are sketched in the respective phases. In the quantum critical regime bound states or longitudinal magnons may be observed due to strong quantum fluctuations

Table 10.2. Properties of the ground state and its excitations close to the quantum critical point of a spin chain with interchain coupling and next nearest neighbor interaction. The spacial decay of spin correlations, the dispersion, degeneracy and quantum statistics of the elementary excitation and the bound or collective modes are given

ground state properties	Néel state	singlet state/spin liquid
GS and its symmetry	symmetry broken two sublattices	symmetry preserved
decay of spin correlations elementary excitation	algebraic gapless spin wave	exponential gapped des Cloizeaux-Pearson triplet
dispersion $\omega(\mathbf{q}_{\parallel})$	$J \cdot \sin(q_{\parallel}) $	$\Delta(\mathbf{q}_{\parallel}) + \pi \cdot \mathbf{J} \cdot \sin(\mathbf{q}_{\parallel}) $
quantum statistics bound/collective state	Boson longitudinal magnon	Fermion singlet bound state

Close to the quantum critical point the susceptibility for the formation of local spin states with s=0 is very large leading to a modification of the low energy excitation spectrum [27]. In contrast the high energy spectrum does not so much depend on the control parameters. For the discussed systems the low energy excitation spectrum consists of elementary and bound states, see Table 10.2. The effect of this transition on the bound states is very drastic. It has been proposed that in the proximity of the quantum critical point on the singlet side a large number of singlet bound states (s=0) form that strongly soften in energy approaching the quantum critical point. On the Néel site of the phase diagram strong quantum fluctuations reduce the sublattice magnetization and longitudinally polarized magnons develop and gain spectral weight close to the quantum critical point. Due to the mixing of competing phases for $T \neq 0$ the resulting critical behavior is anomalous. This scenario of low-energy excitations and bound states has been investigated theoretically for chains [28–30], ladders [31, 32], 2D plaquettes [33–35] and the Kagome lattice [4, 36]. The importance of frustration [37, 38] and the coupling to non adiabatic phonons [39–41] has been especially highlighted. Compounds that have been studied with respect to a quantum critical point are the 1D systems $BaCu_2Si_2O_7$ and $KCuF_3$ [42,43], the 2D systems $SrCu_2(BO_3)_2$ [44–47], CaV_4O_9 [48], $Cu_2Te_2O_5Br_2$ [49–52] and the quasi-3D system $Cu_6Si_6O_3 \cdot 6H_2O$ [53]. The former two 2D systems represent short range ordered states while the latter 2D and quasi-3D systems are in a long range ordered state. Further details will be discussed below.

In Fig. 10.4 the results of a systematic investigation of bound states in quantum spin systems using inelastic Raman scattering are shown [27,54,56]. All investigated systems have a spin gap either due to frustration and/or a dimerization of the spin system. Strong triplet-triplet interaction leads to a



Fig. 10.4. a) Low temperature Raman scattering on the 2D frustrated $SrCu_2(BO_3)_2$, the charge ordered NaV_2O_5 and the frustrated and dimerized spin chain system $CuGeO_3$ as function of the reduced energy scale. In the bottom panel the effect of Zn substitution on the low energy excitation spectrum of $Cu_{1-x}Zn_xGeO_3$ is shown, with x=0, 0.0022, 0.0066 (bottom to top) [54, 55]. The data have been scaled and shifted for clarity. The effect of substitutions on Δ_{01} in this concentration range is negligible. b) Binding potential of a coupled spin chain system with the elementary triplet Δ_{01} , the s=0 singlet bound state Δ_{00} and other composite states of higher order. For energies $E > 2\Delta_{01}$ a dense continuum of single and multiparticle excitations sets in [29,41]

large binding energy with respect to the energy of two elementary triplets $2\Delta_{01}$. Consequently the compound with many modes at lowest energy and largest spectral weight is the 2D frustrated SrCu₂(BO₃)₂ [46]. Altogether 5 modes show up at low temperatures with energies E_{singlet} ranging from $1.1 - 4.1 \ \Delta_{01}$, with $\Delta_{01}=34 \text{ K} (24 \text{ cm}^{-1})$. The latter two systems, NaV₂O₅ and CuGeO₃, show three and one bound states, respectively [57,58].

In CuGeO₃ Zn substitution on the Cu site that induces local spin defects lead to additional bound states with lower energy corresponding to stronger binding [55, 59, 60]. These results are supported by recent NMR/NQR experiments that show enhanced singlet correlations near the substitution center [61]. It is remarkable how sensitive the intensity of primary singlet bound state depends on an even small doping level. Bound states in 1D spin system have been described within the concept of soliton states with magnetoelastic interaction. The binding is then given due to interchain interaction and coupling to dynamic phonons [41, 62, 63]. In Fig. 10.4b a sketch of the resulting linear binding potential is shown. The opening angle decreases for weak coupling leading to an infinity of bound states [30].

Raman scattering is the only spectroscopic measurement technique that is sensitive to singlet bound states. Only very close to a quantum critical point these states also contribute to the specific heat of the system [37]. Singlet bound states are observed in light scattering via an exchange mechanism and are related to the density of states-like two-magnon scattering observed in long range ordered AF [64]. The underlying Hamiltonian is identical to the Heisenberg model [57]. Spin orbit coupling which would lead to one magnon scattering and the observation of triplet states does not play an important role for the systems discussed here.

10.1.3 Frustration

Frustration and quantum criticality are two aspects that are intimately related in quantum spin systems. Frustration describes the state of any system that can not satisfy or comply with all existing interactions. This is evident for an AF spin system where the spins are arranged on triangles and tetrahedra as shown in Fig. 10.5. Also cases of next nearest neighbor (nnn)interactions have often to be considered on a square plaquette or a spin chain with a nonlinear exchange path. We emphasize that only situations are considered where the full translational symmetry of the lattice is preserved, i.e. lattice disorder is not taken into account.

In a classical system frustration would lead to a multitude of possible states with very close or degenerate energies. This results in ground state degeneracy and large low- or zero-temperature entropy. For a further evaluation of the investigated system it is then important to go beyond the local topology and range of interactions that is shown in Fig. 10.5 and also consider the global linkage of these elements and further perturbations or anisotropies.

An historical but still interesting example is ice (H₂O for T<273 K) [65]. The entropy deficit observed at low temperatures is related to the undercon-



Fig. 10.5. Spin frustration as realized in spin systems with a) triangular, b) tetrahedral, c) spin chain, and d) square plaquette topology. A wavy line denotes *nnn* interaction. In a), b) and d) the oxygen in the superexchange path has been omitted. The ratio of copper and oxygen radii in c) has been reversed for clarity



Fig. 10.6. a) Structure of ice (I_h). Each oxygen (larger sphere) has two closer protons and two protons further apart. b) Specific heat and entropy of the spin ice $Dy_2Ti_2O_7$ in the upper and lower panel, respectively. The insets show $\chi^{-1}(T)$ and the topology of the Dy sites, respectively. In the lower panel the entropy of the Ising system (Rln2) and the experimentally observed entropy with deficit are given by dotted and dashed curves. In a magnetic field of 0.5 T (open dots) a part of the entropy is recovered [66]

straint of the proton sites on the tetragonal oxygen sublattice by the local stoichiometry. In Fig. 10.6a this structure is shown. Each oxygen has two closer and two neighbor protons further apart. These ice rules, however, do not determine an unambiguous global structure as for any oxygen site a sixfold degeneracy of these states exists. Therefore the dielectric constant of ice remains very large down to temperatures as low as 100K, related to larger scale proton reorientation processes that slowly freeze-in towards lower temperatures.

Water ice has a magnetic analogue called *spin ice* based on corner shared tetrahedra of Rare Earth ions with FM interactions and Ising anisotropy. Such a spin arrangement is realized in the pyrochlore $Dy_2Ti_2O_7$, with Dy^{3+} in a $4f^9 - s=9/2$ state [67]. The anisotropy leads to "two spin in - two spin out" ice rules within one tetrahedron and a similar entropy deficit. Respective thermodynamic data with a maximum at about 1K and the derived entropy are shown in Fig. 10.6b). The advantage of the study of such systems compared with the nonmagnetic cases is the higher cleanliness of the magnetic field [66, 68]. The latter effect might have some potential for applications as the compounds can be used as cooling media in high magnetic fields [69].

Even more fascinating and richer is the behavior of a quantum spin systems with frustration. Here, the ground state can be formed by any linear combination of these degenerate states leading to possible new ground states without symmetry breaking. An example is the resonating valence bond state that is stabilized on a Kagome lattice [4, 70]. Although proposed earlier the RVB state is not a ground state of the square plane of high temperature superconductors [71–73]. In contrast the Kagome lattice with its combination of depletion and frustration in 2D shown in Fig. 10.11 supports the RVB and its rich low energy singlet excitation spectrum.

Spin systems with strong frustration have been investigated for many years and even recently many more interesting compounds have been found and studied [74–79]. Important question that are in the center of interest are, e.g. the relation between the effect of frustration and the dimensionality of the system [80] or the above discussed low energy excitation scheme in the proximity to a quantum critical point. Frustration may lead to very low energy singlet excitations of a spin liquid [4,37,81–83]. Further related aspects will be discussed below.

10.1.4 Orbital Related Effects

As shown in Fig. 10.2, energy and occupation of 3d orbitals depend sensitively on the local symmetry and distortions as an effect of the crystalline electric fields of the surrounding atoms. On the other side the spatially anisotropic charge distribution can form varying exchange paths and resulting spin configurations or topologies.

This is evident if we compare the alignment of the e_g orbitals $(d_{x^2-y^2})$ and $d_{3z^2-r^2}$ with the t_{2g} orbitals (d_{xy}, d_{yz}) and d_{zx} . While the former point along the crystallographic axes, the latter point along the bisecting line. If these orbital degrees of freedom are taken into account on equal footing spin-orbital models [84,85] develop that have their own dynamics and an extraordinary richness of phases and possible coupled excitations [86–89].

In the limit of large energies of the orbital system with respect to both the spin system and the investigated temperature range the orbital system is ordered and establishes a constant background on which the spin system works. The related lattice distortions that may have static or dynamic character are Jahn-Teller distortions and the overall positive energy balance comes from the lifting of related degeneracies.

An example for such a system is KCuF₃ with Cu²⁺ in a 3d⁹ configuration. A e_g d_{x²-y²} orbital is occupied [43, 90]. The Jahn-Teller distortion of the basic CuF₆ octahedra alternates in *a* and *b* axis direction along the *c* axis. The distortion is intimately coupled to the orientation of one lobe of the d_{x²-y²} orbital along the *c* axis while the other lobe alternates in a and b axis direction. This forms an homogeneous chain of superexchange coupled s=1/2 with J=200 K due to the linear Cu-F-Cu superexchange path. The superexchange path in the other directions is disrupted due to the orbital ordering in alternating directions. In Fig. 10.7 a projected view on chains of CuF₆ octahedra is given together with a sketch of the ordered planes of d_{x²-y²} orbitals. The related Jahn-Teller distortions take place at T_{OO} = 800 K



Fig. 10.7. a) Projected view on distorted CuF_6 octahedra forming chains along the c axis of KCuF₃. Potassium sites have been omitted for clarity. b) connecting the shortest F-F distances the planes of the ordered $d_{x^2-y^2}$ orbitals are depicted. Chains are highlighted by dashed lines

clearly marking a decoupling of the energy scales of spin and orbital system, respectively.

Obviously, even more interesting situations arise if the orbital ordering is destabilized by quantum fluctuations (e.g. t_{2g} orbital degeneracy), competing interactions or a coupling to other degrees of freedom. LaTiO₃ and YTiO₃ with Ti³⁺ and a 3d¹ electronic structure embedded in a cubic perovskite structure are proposed to be examples of such a scenario [91–93]. While in the former compound quantum effects select a particular orbital state depending on the spin configuration, in the latter a weak orbital order is stabilized by an order from disorder mechanism. The discussion of the related effect is nontrivial and goes beyond the scope of this chapter. However, it should be noted that in 2D and even 3D compounds orbitals may form dynamical quasi-one dimensional configurations.

There exist interesting concepts for spin-orbital models on a chain or plane of sites. A coherent spin-orbital structure in a system with orbital degeneracy can lead to a novel mechanism for obtaining a spin-gap. These gapped phases are in the proximity to ferromagnetic and antiferromagnetic phases. In a certain parameter range doubly degenerate ground states are realized which form alternating spin and orbital singlets [94]. Compounds that optimally realize this scenario are recently in the center of interest as the expected anomalies of magnetic and structural properties should be extraordinary.

Promising candidates that show the opening of a comparably large spin gap together with a lattice distortion are found recently within the titanates, such as, e.g. $MgTi_2O_4$ [95], TiOCl [96], $Na_2Ti_2Sb_2O$ [97], and $NaTiSi_2O_6$ [98]. At least for the latter two systems a one-dimensional character of the electronic structure is evident. A further discussion of these systems will be given in Sect. 10.5.3.

10.2 Interplay of Structural and Electronic Properties

It is the challenge of recent activities in solid state chemistry and physics to find new transition metal compounds that allow studying effects of electronic correlations and reduced dimensionality in a model system-like approach. Especially the direct observation of a temperature or substitution-induced crossover into a quantum critical regime is a unique goal. Important aspects of quantum spin systems that develop from the interplay of electronic correlation with topology are summarized in Fig. 10.1.

From a solid state chemist point of view, there is no obvious possibility to design solid structures in a predictive and systematic way and this is of course true for low-dimensional quantum spin systems. One has therefore to rely on few concepts, like for example dimensional reduction [99], or naïve pictures that have emerged based on experiences with structurally related materials (cristallo-chemical considerations). This approach has been quite successful in the case of vanadates. Structural databases [100], e.g. via the extensive use of web resources, may be an important help if expressive search criteria can be developed. As an example minerals with the transition metal ions given in Table 10.1 can be considered.

Mineral compounds provide interesting spin topologies and examples are now numerous on compounds which also demonstrate unusual magnetic properties. Let us mention, the volborthite $Cu_3V_2O_7(OH)_2 \cdot 2H_2O$ [101, 102], a S = 1/2 Kagomé lattice; the azurite $Cu_3(OH)_2(CO_3)_2$, a one-dimensional diamond chain [103]; the malachite $Cu_2(OH)_2CO_3$, a S = 1/2 quantum antiferromagnet with a spin gap [104]; the antlerite $Cu_3(OH)_4SO_4$ and brochantite $Cu_4(OH)_6SO_4$ [105]; the paramelaconite Cu_4O_3 [106,107], the stibivanite Sb_2VO_5 , a S = 1/2 1D chain [108] and finally the dioptase $Cu_6Si_6O_3 \cdot 6H_2O$, a S = 1/2 dimer system with its spin topology structure shown in Fig. 10.13 [53].

In the following we will discuss further, more practical concepts to describe quantum spin systems and apply these systematically to spin systems with increasing complexity, going from chains, dimerized chains and dimers, coupled chains or ladders to 2D topologies.

10.2.1 Concepts Based on Structure and Chemistry

Several concepts can be used to develop a basic understanding of lowdimensional quantum spin systems concerning their exchange topologies and to model effective low-energy Hamiltonians. This understanding is, e.g. an indispensable condition for an effective search for new systems using the above mentioned structural databases [100]. A simple approach that has been used very successfully identifies simple magnetic base units in the crystal structure that are then coupled to more complicated structures. In Table 10.3 typical transition metal ion-oxygen polyhedra are given as function of the oxidation state of the respective ion. This concept of searching for local coordination

TMI			
Vanadium	V ³⁺ octahedron	V ⁴⁺ trig. bipyramid square pyramid octahedron	V ⁵⁺ tetrahedron trig. bipyramid square pyramid octahedron
Copper	Cu ¹⁺ linear	Cu ²⁺ square plane tetrahedron trigon. bipyramid square pyramid pentag. bipyramid	Cu ³⁺ octahedron square pyramid
Titanium	Ti ³⁺ octahedron	Ti ⁴⁺ tetrahedron square plane octahedron	

 Table 10.3.
 Typical transition metal ion (TMI) oxygen coordination polyhedra

 versus oxidation state.
 Characteristic coordinations are highlighted

and their linkage works best in systems with low connectivity, where the transition metal ions have only one or two next neighbor (nn) and next nearest neighbor (nnn) sites with distinctly smaller distance compared to the distance to other magnetic ions. This means that large variations between smallest and larger transition metal ion distances should exist that can easily been evaluated. Using e.g. a layered system as a starting point to reduce the dimensionality of the spin system to D=2 [99], a further depletion of magnetic sites can lead to interesting spin topologies with very close nn. This concept may lose sense if competing antiferromagnetic exchange paths exist that lead to spin frustration. Nevertheless even such systems are interesting due to the stabilization of short range ordered states and a potential ground state degeneracy.

Other approaches use specific structural/chemical properties of nonmagnetic groups that lead to very small hopping integrals and a resulting negligible magnetic exchange. A structural analysis of known quantum spin systems indicates that in many phases nonmagnetic building bricks are made of triangles or tetrahedra in the form of carbonates and borates groups (coordination number: CN3) or silicates, germanates and borates (CN4). To follow this trend the lone-pair concept has been proposed. This concept is based on the fact that lone pair elements as Te^{4+} , Se^{4+} , As^{3+} , possess an electronic charge distribution (noted E) - with a volume close to the one of an oxygen atom - that is not involved in a chemical bond. Indeed, the classical



Fig. 10.8. a) Tetrahedron formed by a lone pair - Te - O coordination. b) Crystal structure of $Cu_2Te_2O_5Br_2$. The sticks with open end mark the approximate position of the lone pair charge density. The crystallographic unit cell is outlined

coordination polyhedron of a lone pair element is a tetrahedron formed by three oxygen atoms and the lone pair E as shown in Fig. 10.8. Comparably "open" structure with low connectivity have been obtained when these elements are mixed with transition metal ions in presence of halide counter anions - the latter are used for charge balance purpose and pseudo template effects (structure directing agents) [49,109,110]. An example is the compound $Cu_2Te_2O_5Br_2$ which has weakly connected Cu^{2+} tetrahedra and the magnetic properties evidence a proximity to a quantum critical point [27,50–52]. In Fig. 10.8b the lone pair positions are given for this system.

10.2.2 Angle Dependence of Superexchange

A microscopic approach [3] uses the strong dependence of superexchange on the angle of the transition metal ion-oxygen-transition metal ion bond. The Goodenough-Kanamori-Anderson (GKA) superexchange rules [111–114] concern sign and magnitude of this exchange via orthogonal orbitals. In a very simplified picture a linear 180 degree superexchange group connecting two 3d eg orbitals by an oxygen 2p orbital is dominantly AF. The strength of this exchange decreases strongly if the contained angle gets close to 90 degree and other FM contributions may dominate.

In Fig. 10.9 it is shown how a 2D square plane can be divided into two magnetically insulated half-planes. Shifting the unit cell of part by half a lattice constant edge-sharing plaquettes with a disrupted vertical exchange path are formed. If two such operations are preformed with cutting lines parallel to each other we realize a ladder-like exchange topology. The number of rungs of the embedded ladder depends on the number of unit cells that are enclosed. Compounds that follow such a scheme are the two- and three-leg ladder



Fig. 10.9. Cutting a 2D square plane and shifting one segment by half a unit cell leads to a disruption of the vertical exchange paths. A 1D exchange topology is formed as the AF coupling between the two halves is frustrated. The 90-degree coupling via edge-coupled plaquettes may also lead to weak FM coupling

systems $SrCu_2O_3$, $Sr_2Cu_3O_5$ and the chain/laddersystem $Sr_{14-x}Ca_xCu_{24}O_{41}$ [115]. Similar structures also exist in the vanadates [116–118].

10.3 Copper-Oxygen Coordinations

In the following we will discuss systems based on copper-oxygen coordinations starting with the simplest superexchange building block, the linear Cu-O-Cu bond that forms an infinite chain systems. The Sr cuprates, e.g. Sr_2CuO_3 , SrCuO₂, and SrCu₂O₃, are a prototype family of compounds closely related to HTSC in their structural and electronic properties [115]. They form spin chains with linear exchange, zig-zag chains and spin ladders, respectively. A discussion of thermodynamic and structural properties of these and related systems can be found in [119, 120] and [115], respectively. The linear chain Sr_2CuO_3 [121–125] has a comparably large exchange coupling constant $(J_{\parallel}=1300\text{-}1400~\mathrm{K})$ and shows Néel ordering at only $T_{\mathrm{N}}=11~\mathrm{K}.$ In SrCuO_2 a zig-zag chain $(J_{\parallel} = 2100 \text{ K})$ is formed by a corner-sharing arrangement of two close chains [121, 126]. In Fig. 10.10 the crystal structure of these systems and the typical temperature dependence of the magnetic susceptibility for s=1/2 spin systems is shown. The maximum in the susceptibility $\chi(T)$ for a homogeneous chain system is expected at $T_{max}^{chain} \approx 0.641$ J. This value is close to $T_{max}^{dimer} \approx 0.63$ J for a spin dimer system. The susceptibility of 2D and 3D systems are given for comparison.

The ladder system $SrCu_2O_3$ is formed by two coupled chains and has exchange coupling constants of $(J_{\parallel} = 850\text{-}2000 \text{ K}, J_{\perp} = 750\text{-}1000 \text{ K} [123])$. As $SrCu_2O_3$ extrapolates in some sense between a chain and a plane of spin moments it represents a very important class of systems. It shows a spin li-



Fig. 10.10. a) Structure of the spin chain Sr_2CuO_3 and b) the zig-zag spin chain system $SrCuO_2$. c) Magnetic susceptibility $\chi(T)$ of a spin s=1/2 system with J=70 K for spin dimers, 1D spin chains, 2D planes and of a 3D system [127–129]

quid ground state and a large singlet-triplet gap of $\delta \approx 0.5J$ [130, 131]. This class of topologies has been intensively investigated as a minimum model for electronic correlation-induced superconductivity [131–135]. Doping these systems, however, has proven to be extremely difficult due to electronic localization and competing instabilities, e.g. a charge density wave. Only in $\mathrm{Sr}_{14-x}\mathrm{Ca}_{x}\mathrm{Cu}_{24}\mathrm{O}_{41}$ a sizable hole content on the ladders can be induced by simultaneous Ca substitution and hydrostatic pressure. The complexity of this material is enormous. Therefore we refer to [54] and given references therein.

10.3.1 3D Dimerized Systems and Effects in Large Magnetic Fields

The simplest realization of a spin gap, however, is a spin dimer. In real compounds interdimer interactions of different strength and dimensionality (1D - 3D) always exist. However, it may lead to drastically different physical properties.

Spin dimer systems with appreciable 3D interdimer interactions are realized in the copper halides KCuCl₃, TlCuCl₃ and NH₄CuCl₃. The dimers are formed by edge-sharing CuCl₆ dioctahedra running parallel to the crystallographic *a*-axis. These ladder-like groups are separated by the cations. Appreciable couplings between sites on different ladders exist as shown in Fig. 10.11a [136]. The compounds KCuCl₃ and TlCuCl₃ have a singlet ground state and a spin gap of $\Delta_{01} = 32$ K and $\Delta_{01} = 7$ K, respectively, as expected for a dimer system [136–141]. The different magnitude of the spin



Fig. 10.11. Exchange topology of a) a coupled dimer system as derived for KCuCl₃, b) the Shastry-Sutherland, c) the 1/5 depleted square lattice and d) the Kagome lattice (a 1/4 depleted triangular lattice). The oxygen sites of the superexchange path have been omitted. The dashed line in a) correspond to magnetic sites on a different *ac* plane

gap is related to a larger interdimer interaction in TlCuCl₃. The exchange topology of NH_4CuCl_3 is less clear and still under debate. The compound shows a collective ordering at $T_N = 1.3$ K of about 1/4 of the spins determined from the magnetic entropy released at the transition [142].

In magnetic fields KCuCl₃ and TlCuCl₃ show transitions into 3D magnetically ordered gapless phases with isotropic cusp-like minima $H_c=22.3$ T and 5.7 T [136, 138, 143], respectively. The effect in the magnetization is quite small ($\Delta m/m \approx 10^{-3}$) and does not depend on the orientation of the field. Furthermore, the related phase lines show an upward curvature towards higher fields for higher temperature. This transition is described as a Bose-Einstein condensation of dilute magnons [144]. Recent NMR, ultrasonic and Raman scattering experiments point to an appreciable spin-lattice coupling and a first order contribution to the character of the transition [145–147]. The idealized case would be an interaction-free system with a T=0 transition of 3rd order. In Fig. 10.12 sound attenuation as function of the magnetic field is shown. The sharp peak associated with the transition shows a hysteresis [146]. The broad maximum corresponds to a matching of the relaxation rate of the triplets with the frequency of the sound wave. Sound attenuation



Fig. 10.12. Magnetic field dependence of the ultrasonic attenuation in $TlCuCl_3$. The insets show the derived phase diagram compared with earlier thermodynamic data and the transition regime after subtracting an intrinsic attenuation background with a weaker field dependence

in such a compound is sensitive to the temperature and field-induced triplet distribution on the spin dimer lattice.

The compound NH₄CuCl₃ shows larger changes of the magnetization in magnetic fields as a sequence of five well-defined transitions ($\mu_0 H_c = 5$, 12.8, 17.9, 24.7, 29.1 T) with plateaus at m= 1/4, 3/4 and 1, with the magnetization m per copper site in units of $g\mu_B$ [148–150].

Plateaus in m(H) of a 1D s=1/2 spin systems are due to a topologically induced quantization and fulfill the Oshikawa-Yamanaka-Affleck condition n(s-m) = integer, with n the spatial period of the spin ground state (number of spins per unit cell), s the magnitude of the spin and m the magnetization [151]. In a two-leg ladder or a frustrated/dimerized Heisenberg chain this condition leads to plateaus at m=0 and 1/2 [151–154]. The plateau at m=1/4 in NH₄CuCl₃ is not understood so far.

A spin dimer system bridging 2D and 3D is the mineral dioptase with the composition $\text{Cu}_6\text{Si}_6\text{O}_3 \cdot 6\text{H}_2\text{O}$ [53]. The magnetic structure is based on dimers of Cu^{2+} that are located in between a stack of Si_6O_{18} rings, see Fig. 10.13. They form chiral chains along c, placed on a ab-honeycomb lattice. Two coupling constants are defined as an inter-chain J_1 , which couples the chiral Cu^{2+} chains along c and an intra-chain coupling J_2 . Such a lattice demonstrates how a 3D lattice can be established by just two coupling constants. For small J_1 the Cu-sublattice complies with weakly coupled spin-1/2 chiral-chains along c which are at sufficiently low temperatures in a long



Fig. 10.13. a)+b) Structure of the magnetic sites of dioptase in the *ab* plane and along the *c* axis, respectively. c) Phase diagram giving the Néel temperature and the spin gap as function of the alternation δ of the two coupling parameters, with $J_1 = J(1 + \delta) (J_2 = J(1 - \delta))$ given by open (full) lines. An arrow marks the estimated position of dioptase in the phase diagram [53]

range ordered state. In the opposite limit, $J_2 = 0$, the Cu-sublattice reduces to planes of isolated dimers with a spin-gap and no long-range order. In an evaluation of thermodynamic and Raman scattering data J = 57 K and $\delta = 0.1$ have been estimated, i.e. the system is in proximity to quantum critical point [53].

10.3.2 2D Dimerized Systems

To realize a gapped or gapless spin liquid ground state in a 2D exchange topology [54] additional frustrating exchange terms or a depletion of the magnetic lattice must exist. Examples are the Shastry-Sutherland lattice [155], a square plane with additional diagonal (nnn) couplings, the square plaquette lattice, consisting of a 1/5 depleted square plane or the Kagome lattice that corresponds to a 1/4 depleted triangular lattice [79]. In Fig. 10.11 b)-d) these topologies are sketched.

Solid state chemistry provides two compounds, $SrCu_2(BO_3)_2$ [47] and CaV_4O_9 [48, 158, 159], that approximate these topologies. In the compound $SrCu_2(BO_3)_2$ the diagonal (nnn) couplings (J) are significant compared with the square plane exchange (J'), giving a ratio $J'/J \approx 0.6 - 0.68$ [35, 47] and strong frustration. This compound is very close to a quantum critical point as for J'>0.67-0.7J a transition into a Néel phase is expected [160, 161]. A phase diagram including a 3D interaction J" that destabilizes the singlet dimer phase is shown in Fig. 10.14a).

Experimentally $SrCu_2(BO_3)_2$ has a spin gap of $\Delta_{01}=34$ K and very localized triplet excitations. In large magnetic fields plateaus of the magnetization are observed at m/m_s=0, 1/4, 1/3, corresponding to quasi-localized superstructures of the magnetization in space [162, 163]. In neutron scattering the spin gap Δ_{01} (elementary triplet branch) shows only a very weak dispersion as



Fig. 10.14. a) Phase diagram of $SrCu_2(BO_3)_2$ as function of the interplane (J'/J) constants and an intraplane (J''/J) coupling constant, respectively [156]. The bold and solid lines correspond to quantum phase transitions, the dashed region correspond to a representative parameter set for $SrCu_2(BO_3)_2$. The 2D quantum critical point is additionally marked by a circle. b) Dispersion of the elementary triplet branch compared to singlet and triplet pair states as derived from Neutron and Raman scattering [44, 46, 157]. The energies of the singlet pair states have been determined for q=0

shown in Fig. 10.14b). Furthermore, a second triplet branch and singlet pair states (given at q=0) are observed [44–46,157]. This rich excitation spectrum follows in some sense the scenario of the evolution of multiparticle states in the proximity of a quantum critical point as discussed above.

10.3.3 Dimerized Spin Chains

The compound CuGeO₃ [54, 164–166] allows to study a spin gap formation in a dimerized chain as function of temperature. A spin-Peierls transition is observed at $T_{SP} = 14.3$ K that leads to a spin gap of $\Delta_{01} = 24$ -30 K and a reduced gap ratio $2\Delta_{01}/k_BT_{SP} \approx 3.4$ at low temperatures. This compound has a rich physics that includes spin frustration due to sizable *nnn* interaction along the chain and a Cu-O-Cu bonding angle of approximately 98 degrees, see Fig. 10.5c). A further interesting aspect of this compound is the strong nonadiabatic coupling of certain phonon modes to the spin system [39, 167].

10.3.4 Triangular Lattices and Tetrahedra

Trimer and tetrahedra systems are frustrated and may show short range or long range order with reduced order parameters. Examples are the diamond or trimer chain and related topologies with a spin gap and, in contrast, the 2D triangular plane that may order long ranged.



Fig. 10.15. Magnetic susceptibility of Cu₂Te₂O₅X₂, with X = Br, Cl. The inset shows the derivative $\partial \chi / \partial T$ at B = 0.1 and 5 T with the transitions assigned by arrows [50]

The triangular lattice with classical spins is a good example to illustrate the effect of frustration on a spin system [4]. To construct a ground state of a AF correlated spin triangle a good starting point is a $\sum s=0$ state [79,103]. This, however, does not totally fix the orientation of each spin as a joint rotation of the three partners is still allowed. Connecting triangles to planes this *underconstraint* is partially lifted. The remaining degrees of freedom lead to new low energy excitations. In 2D systems that show long range ordering under such circumstances a new universality class of chiral critical behavior can be observed. This has been found in CsMnBr₃, an essentially 2D compound with triangular planes [168–170].

A tetrahedron unit based on four AF coupled quantum spin s=1/2 has 16 states divided into two singlets, three triplets, and a quintuplet. The degeneracy of the two singlets, a dimer product and a plaquette singlet state, is controlled by the coupling constants and the quantum case may lead to interesting low energy singlet degrees of freedom that are easier to describe than the complex situation of the Kagome lattice [79].

The lone pair system $Cu_2Te_2O_5Br_2$ with it structure shown in Fig. 10.8 and discussed earlier in Sect. 10.2.1 is regarded as representing weakly coupled s=1/2 tetrahedra [49,51,52]. It shows weak Neel order at $T_N^{(Br)} = 11.4$ K [50]. An substitution of Br by Cl decreases the unit cell volume and shifts the transition up to $T_N^{(Cl)} = 18.2$ K. It is therefore tempting to regard the system as being close to a quantum critical point. This is supported by the temperature dependence of the magnetic susceptibility shown in Fig. 10.15. The susceptibility of $Cu_2Te_2O_5(Br, Cl)_2$ has a maximum at 30 K (Br) and 23 K (Cl) and then a large part of the susceptibility is vanishing with decreasing temperature. Due to the reduced order parameter for T<T_N a longitudinal

Table 10.4. Overview of low-dimensional vanadium compounds. The classification is based on the spin arrangements deduced from crystal structure determinations - it does not necessarily reflect the spin topology corresponding to the relevant exchange coupling constants. In some compounds cations like X=Li,Na,Ca,Mg,Sr and M=Cu(1+),Na,Ca,Ag can be introduced. LiVGe₂O₆ is a vanadium 3+ compound with s=1

$\begin{array}{llllllllllllllllllllllllllllllllllll$	isolated polyhedra	chains	layers	folded layers
	$\begin{array}{c} \mathrm{K}_{2}(\mathrm{VO})\mathrm{V}_{2}\mathrm{O}_{7}\\ \mathrm{Li}_{2}(\mathrm{VO})\mathrm{SiO}_{4}\\ \mathrm{Li}_{2}(\mathrm{VO})\mathrm{GeO}_{4}\\ (\mathrm{VO})\mathrm{MoO}_{4}\\ \mathrm{Cs}\mathrm{V}_{2}\mathrm{O}_{5}\\ (\mathrm{VO})\mathrm{SeO}_{3}\\ (\mathrm{VO})_{2}\mathrm{P}_{2}\mathrm{O}_{7}\end{array}$	$\begin{array}{c} MgVO_3\\ Sb_2VO_5\\ LiVGe_2O_6\\ XV_2O_5\\ \alpha\prime\text{-NaV}_2O_5\\ \eta\text{-Na}_{1.286}V_2O_5\\ \gamma\text{-LiV}_2O_5 \end{array}$	$\begin{array}{c} CaV_{3}O_{7} \\ CaV_{4}O_{9} \\ (Bi_{2}O_{2})V_{n}O_{2n+1} \\ XV_{2}O_{5} \\ CaV_{2}O_{5} \\ MgV_{2}O_{5} \\ \beta, \beta'-MV_{2}O_{5} \end{array}$	Na ₂ V ₃ O ₇

magnon is observed in Raman scattering experiments [51]. Such an excitation has previously only been observed in neutron scattering experiments on chain systems [42,43,171].

10.4 Vanadium-Oxygen Coordinations

Within the transition metal oxides, vanadates exhibit a unique structural chemistry due to their capacity to realize different vanadium-oxygen coordination schemes associated with different vanadium oxidation states 3+, 4+ and 5+ (Table 10.3 and [1]). In addition, it is possible to isolate a very large number of mixed-valence compounds. In the frame of quantum spin systems, the number of vanadium based compounds (V⁴⁺, S = 1/2) has increased tremendously now to overtake the cuprates. A review of spin-gap systems based on the vanadate family can be found in [116].

Important phases currently under intense investigation are listed in Table 10.4. They are classified according to their local spin arrangements. From the structural point of view, it is possible to obtain phases formed by isolated polyhedra (in the form of a vanadium 4+ square pyramid) like in $K_2(VO)V_2O_7$ [172], dimers formed of edge connected square pyramids pointing up and down as observed in VOSeO₃ [173], isolated chains like in MgVO₃ [174,175], 2D layers as in CaV₄O₉ [176,177] or more exotic structure as for example for Na₂V₃O₇ [5]. In addition to this structural versatility, vanadium oxides present also unusual electronic properties that are based on the interplay of charge, spin, orbital and lattices degrees of freedom. Furthermore, the possibility to obtain exchange coupling constants ranging from few Kelvin as in Li₂VOSiO₄ [178] to hundreds of Kelvin in CaV₂O₅ [179] is highlighted. This distinguishes vanadium compounds from copper based materials.

10.4.1 MV₂O₅ and Related Compounds with Charge Ordering dInstabilities

The family of vanadium oxide compounds of general formula MV_2O_5 (M= Li, Na, Ca, Mg) has been extensively studied in terms of low-dimensional quantum spin systems. Their crystal structures are closely related to the layered compound V_2O_5 which is built up in one direction by infinite double strings of $[VO_5]$ square pyramids that share edges and corners along the short parameter (≈ 3.7 Å, y axis in Fig. 10.16a) and that are held together by corner sharing pyramids in x direction. The M cations are located in between $[V_2O_5]_n$ layers. Different spin arrangements are found depending on the charge of the cation M. Monovalent cations such as Li and Na lead to a vanadium mixed valence state with two different crystal structures. In the case of α' -NaV₂O₅ all vanadium ions are in the valence state 4.5+ while in γ -LiV₂O₅ an electronic localization is established, characterized by two independent $[V^{5+}O_5]$ and $[V^{4+}O_5]$ square pyramids. Furthermore in order to accommodate the high value of the buckling angle observed in the latter structure (see Table 10.5 and Fig. 10.16) a structural mechanism which involves a rotation of blocks of two square pyramids takes place leading to two double strings of V^{4+} corner connected by double strings of V^{5+} . This system can then be depicted as a 1D double chain [180]. As a result of this transformation the oxygen atoms octahedrally coordinate Li atoms. Divalent cations like Ca and Mg lead to compounds with vanadium atoms in the valence state 4+ which are reminiscent to the spin ladder system $SrCu_2O_3$. The higher polarization of Mg compared to Ca results in a higher buckling angle for Mg: 21° compared to 11.3°, respectively.



Fig. 10.16. Idealized projection of MV_2O_5 in the direction [001] showing a) the ladders and b) a projection in the direction [010] indicating the buckling angle μ and the bridging oxygen x

Phase	d_{\perp} (Å)	$\mu \ (deg)$	space group	Reference
V_2O_5	$V^{5+}, 0.470$	0.1	Pmmn	[181]
γ -LiV ₂ O ₅	$V^{4+}, 0.636$	62.0	Pnma	[182]
	$V^{5+}, 0.539$			
α' -NaV ₂ O ₅	$V^{4.5+}, 0.548$	3.2	Pmmn	[183, 184]
MgV_2O_5	$V^{4+}, 0.666$	21.0	Cmcm	[185]
$\mathrm{CaV}_{2}\mathrm{O}_{5}$	$V^{4+}, 0.648$	11.8	Pmmn	[179]

Table 10.5. Vanadium - oxygen distance with respect to the square pyramid basal

plane (d_{\perp}) and buckling angle (μ) of some selected MV₂O₅ phases



Fig. 10.17. Crystal field levels of a V^{4+} ion in a regular VO₅ pyramid as a function of the distance of the V^{4+} ion from the basal plane (from [116])

Important structural parameters that need to be addressed with regards to the magnetic properties are (i) the distance d_{\perp} of the vanadium atoms to the basal plane of the square pyramid and (ii) the buckling angle μ . As mentioned in [186], the ground state of d levels is directly connected to d_{\perp} . In Fig. 10.17 it is shown that for d_{\perp} larger than 0.35 Å, the ground state is d_{xy} in this vanadate family [116]. The angle μ affects the overlap between the Vd_{xy} and O2_p orbitals of the bridging oxygen atom X and therefore the exchange integrals as observed in the frustrated coupled ladder CaV₂O₅ and MgV₂O₅ [187].

10.4.2 α' -NaV₂O₅

The compound α' -NaV₂O₅ is undoubtedly the phase that has received the most attention among the vanadates because it was first thought to be the second example of an inorganic Spin-Peierls system [188]. Now the transition

is understood as a charge ordering with an accompanied spin gap opening. The basis is a quarter-filled spin-ladder system with at high temperature all vanadium in the valence state $V^{4.5+}$ and a centrosymmetric space group Pmmn [183,184]. At $T_{CO} = 34$ K it exhibits a charge ordering corresponding to $2V^{4.5+} \rightarrow V^{5+} + V^{4+}$ and related to a structural as well as a magnetic phase transition with the opening of an energy gap for spin excitations (modulation wave vector q = (1/2, 1/2, 1/4) and $\Delta = 9.8$ meV, respectively) [189–191]. Recent X-ray anomalous scattering investigations on the charge ordering show that it is of zig-zag type in all vanadium ladders [192,193] and that different stacking sequences along the c direction coexist in the structure. These results are in agreement with a X-ray diffraction investigation of the temperaturepressure (T-P) phase diagram of α' -NaV₂O₅, which show the occurrence of a Devil's Staircase type phase transition, i.e., the development of a series of modulation wave vectors along the c^* direction [194]. Ab initio calculations indicate that the total charge ordering occurring on the modulated rung is small with $2\delta_{tot} = 0.05 e^{-1}$ [195].

10.4.3 β - and β' -MV₂O₅

 β - and β' -MV₂O₅ (M = Li⁺, Na⁺, Ag⁺, Cu⁺ and Ca²⁺, Sr²⁺, Cd²⁺, Pb²⁺) vanadium bronzes have been known from the fifties with the structure determination of β -Na_xV₂O₅ [196]. These phases can accommodate a wide range of cations over an extended range of composition (0.15 $\leq x \leq 0.65$). The average structure is characterized by tunnels running parallel to the [010] direction (see Fig. 10.18).

A range of different sites for intercalated cations (denoted M1, M2, M3) has been observed dependent upon the nature and size of intercalated cations with for example copper at the site M3. First studied for their po-



Fig. 10.18. Projection of β and β' -type MV₂O₅ onto the (010) plane. The location of different sites for various intercalant cations are also shown
tential use as positive electrodes in lithium batteries and in the context of bipolaron formation, they have found a renewed interest as quantum spin systems [117, 197–200]. $\beta - Na_{0.33}V_2O_5$ with a mixed valence of $V^{4+}/V^{5+} =$ 1/5 has received a lot of interest. It exhibits a metal-insulator transition of a charge ordering type at $T_{CO} = 135$ K followed by an AF transition at T_N = 25 K. Worth to mention is that the metal-insulator transition is suppressed under hydrostatic pressure and a superconducting phase with $T_c = 9 \text{ K}$ appears at P = 8 GPa. This phase is then the first superconducting vanadate [118]. For other monovalent cation $M = Li^+$ and $Ag^+ T_{CO}$ and T_N are 90 K, 180 K, and 27 K, 7 K, respectively. The magnetic properties indicates that the charge ordered state of V^{4+} correspond to a linear chain [199]. For divalent cations such as Ca^{2+} and Sr^{2+} a charge order transition is observed at 150 K and 170 K, respectively, followed at low temperature by a spin-gap behavior suggesting that the magnetic V^{4+} ions form a two-leg ladder system. β - and β' -MV₂O₅ can then be considered as the archetype system with the occurrence of charge ordering, spin-liquid state and superconductivity in the same structural type.

10.4.4 η -Na_{1.286}V₂O₅

Recently a detailed study of the V⁴⁺ richer zone of the sodium-vanadiumoxygen phase diagram has led to the synthesis and structural characterization of a new vanadium oxide bronze η -Na_{1.286}V₂O₅ or Na₉V₁₄O₃₅ [202,203]. Its crystal structure is built up of layers consisting of VO₅ square pyramids sharing edges and corners with their apical oxygen pointing up and down. These form double strings in the [100] direction of stair-like shape (a step every tenth VO₅ square pyramids). These double strings are isolated in the [001] direction via VO₄ tetrahedra, see Fig. 10.19.

The analysis of the magnetic susceptibility reveals a spin-gap behavior [204] and an additional small kink at T \approx 100 K. A careful X-ray diffraction experiment indicates the presence of weak superlattice reflections corresponding to the doubling of the unit cell b axis below this temperature. The main structural change associated to this phase transition is the appearance of charge ordering (Fig. 10.19b) characterized by two different vanadium oxidation states V⁴⁺ and V⁵⁺ at low temperatures compared to a formal valence state V^{4.5+} at room temperature [204]. This phase provides another example of a double transition consisting of a magneto-distortion of the lattice following a charge ordering of the unpaired electrons.

This behavior seems to be a common feature of many vanadium compounds. A better understanding of these phenomena is only achieved if the local electronic and structural properties are understood to a certain degree. Then the problem of charge redistribution and the origin of charge ordering can be addressed.



Fig. 10.19. a) Projection of η -Na_{1.286}V₂O₅ onto the (010) plane. b) Charge ordering is evidenced at T<100 K with two crystallographic sites V⁴⁺ and V⁵⁺ at 15 K instead of one (open circles) with the formal valence of 4.5+ at room temperature

10.4.5 Depleted Lattices – Playing with Valences (V^{4+}/V^{5+})

Vanadates compounds provide good examples of depleted spin lattices. The very important phase CaV_4O_9 , the first two-dimensional material with a spingap in the spin excitation spectrum [177] belongs to the family CaV_nO_{2n+1} (n = 2, 3, 4) discovered and structurally characterized in the seventies [174, 205, 206]. Its crystal structure depicted in Fig. 10.20 is formed of layers of VO₅ square pyramids: two adjacent VO₅ square pyramids that share one edge pointing alternately up and down. The spin arrangement forms a 1/5 depleted square lattice as observed in Fig. 10.11c. The spin-gap in this compound was found to result from a plaquette RVB state [159, 207, 208].

A further interesting spin topology is observed for the isostructural compounds $K_2(VO)(V_2O_7)$ [209] and $(NH_4)(VO)(V_2O_7)$ [210]. The layered structure presented in Fig. 10.21 is composed of corner-sharing VO₅ square pyramids and tetrahedra: two tetrahedra being connected via one corner to form $[V_2O_7]$ groups. One particularity of such structure is that all VO₅ square pyramids point in the same direction (up in Fig. 10.21) as a result of the uniaxial noncentrosymmetric space group P4bm. The main difference between the two phases is the longer c axis for the NH₄-compound compared to K, i.e; 5.56 Å and 5.22 Å, respectively. The spin arrangement can be described as a diluted 2D square lattice. The fact that VO₅ square pyramids are slightly tilted with respect to each other (due to the lack of an inversion center) leads to an appreciable Dzyaloshinskii-Moriya interaction and an ad-



Fig. 10.20. Projection of CaV_4O_9 onto the (001) plane



Fig. 10.21. Projection of $K_2(VO)V_2O_7$ onto the (001) plane. The V⁴⁺ square pyramids and $[VO_4]$ tetrahedra are given

ditional c axis anisotropy. In the case of $K_2(VO)(V_2O_7)$, an ordering phase transition with a primarily AF ordered state accompanied by weak ferromagnetism and a novel field-induced spin reorientations have been observed at $T \leq T_N = 4 \text{ K}$ [211].

10.4.6 J1-J2 Model on a Square Lattice

For many years the J1-J2 model on a square lattice has been the object of intense theoretical research [212–214] and a relatively clear picture has emerged regarding the magnetic phase diagram as a function of the ratio J_2/J_1



Fig. 10.22. Schematic phase diagram of a frustrated 2D QHAF on a square lattice as a function of the ratio J_2/J_1 of the superexchange couplings



Fig. 10.23. a) Projection of Li_2VOXO_4 (X=Si, Ge) onto the (001) plane, b) exchange integrals for the J1-J2 model on a square lattice, and, c) perspective view of this layered structure. VO₅ square pyramids are in dark grey, XO₄ tetrahedra in light grey

 $(J_1 \text{ nearest neighbor exchange and } J_2 \text{ second neighbor (diagonal) exchange, see Fig. 10.22) [215–219].$

Recently two phases Li_2VOSiO_4 and Li_2VOGeO_4 have been isolated [220] that represent an experimental realization of this model. The structure is formed of VO₅ square pyramids pointing alternating up and down (Fig. 10.23a), and isolated in the [100] and [010] directions by XO₄ tetrahedra.

Worth to mention is that the S=1/2 vanadium atoms are slightly displaced above the basal square plane of the square pyramids by nearly 0.6 Å leading to two vanadium sub-lattices (Fig. 10.23c) and hence introduce a sizeable frustration. The compounds Li₂VOSiO₄ and Li₂VOGeO₄ undergo a low temperature phase transition to a collinear order as predicted for J₂/J₁ >0.65 at $T_N = 2.86$ K and 2.1 K for Si and Ge, respectively. Specific heat and magnetization measurements indicate a ratio $J_2/J_1 = 1.1 \pm 0.1$ with $J_1 + J_2 = 8.2 \pm 1$ K for Li₂VSiO₅ and $J_1 + J_2 = 6$ K for Li₂VGeO₅ [178,221,222]. A discrepancy remains concerning the ratio J_2/J_1 since a value close to 12 has been obtained recently [223]. In-plane exchange integrals $J_1 + J_2 = 9.5 \pm 1.5$ K and inter-plane exchange integral 0.2-0.3 K have been extracted for Li₂VOSiO₄ from a tight binding model fitted to the LDA band structure and mapped onto a Heisenberg model [223–225]. A last determination of the exchange energies in Li₂VOSiO₄ obtained from a high temperature series analysis indicates a lower ratio J_2/J_1 close to 5 [226].

Another interesting result obtained from ²⁹Si NMR spectroscopy is the observation of a structural distortion just above T_N . The phase transition to the collinear phase seems to be triggered by this distortion which could affect the superexchange coupling and lift the degeneracy among the two ground state configurations (I and II in Fig. 10.22); the system selects finally configuration I for temperatures below T_N .

Recent low temperature neutron diffraction measurements on polycrystalline Li₂VOSiO₄ confirm the presence of collinear magnetic order with a propagation vector (1/2, 1/2, 0). The refined ordered moment at 2 K is 0.46(5) $\mu_{\rm B}$ [227]. Another experimental realization of the J1-J2 model on the square lattice is the phase (VO)MoO₄, the structure of which being closely related to Li₂VOSiO₄. It is built up of VO₅ square pyramids isolated by MoO₄ tetrahedra. The value J₁ + J₂ is equal to 155 K in agreement with the value derived from electronic structure calculations [228]. Around 100 K a structural distortion possibly driven by frustration is evidenced and the size of the distorted domains progressively grows on cooling as the temperature reaches the transition to the magnetic ground state at T_c ≈ 42 K.

10.4.7 Exotic Topologies

A very peculiar spin arrangement has been obtained in the case of $Na_2V_3O_7$ (all vanadium in the valence state 4+) [5]. As can be observed in Fig. 10.24a, VO_5 square pyramids share edges and corners to form nanotubes oriented along the *c* axis. The cohesion of the network is ensured by sodium atoms which are located inside - the inner diameter of the nanotube is of approximately 5 Å - and around individual nanotubes. The idealized representation of the unfolded nanotube depicted in Fig. 10.24b allows a simple description of the vanadium polyhedra arrangement. The basic structural unit is formed of three vanadium square pyramids in grey and three of these units are connected by edges to form a slice noted (A) which is therefore constituted of nine square pyramids. The tube is then obtained by a repetition (A), (B), (A), (B), ... of corner sharing slices, Fig. 10.24c.

This compound provides a unique example of a folded layer leading to 1D tubular structure. From the magnetic point of view, this is a rather complex system that involves at least six different coupling constants. The results of dc-susceptibility measurements indicate that upon reducing the temperature



Fig. 10.24. a) Projection of $Na_2V_3O_7$ onto the (001) plane, b) Idealized representation of the unfolded vanadium-oxygen nanotube. The basic unit is formed by V1,V2, V3 in dark gray (shortest V-V distances) and 3 units form one ring, c) The repetition of the sequence (A)(B)(A).. along the c axis forms the nanotube



Fig. 10.25. Anomalies in $1/T_1(T)$ at T_a , T_b , T_c for $Na_2V_3O_7$. These characteristic temperatures corresponds to field-dependent phase transitions as indicated by the arrows. The state below T_a involves spin degrees of freedom. T_a , T_b and T_c shift towards T = 0 K with decreasing external field H [229]

to below 100 K V⁴⁺ moments are gradually quenched with at 10 K only one moment out of nine active [230]. At much lower temperatures a phase transition occurs at a field-dependent transition temperature T_a (Fig. 10.25).

 $Na_2V_3O_7$ may be considered as a low S = 1/2 system in the quantum critical limit [229].

10.5 Titanium-Oxygen Coordinations

As discussed in Sect. 10.1.4 the interplay of orbital and spin degrees of freedom in a transition metal oxide may lead to unconventional behavior. In the following we will discuss three titanates with Ti^{3+} - t_{2g} - s = 1/2 configuration that demonstrate this effect from structural and electronic point of view. All compounds show the opening of a comparably large spin gap as the result of a phase transition that involves a lattice distortion. The established low temperature spin system is essentially isotropic. Nevertheless, these instabilities are not consistent with the context of spin-Peierls transitions [231] or strong coupling scenarios via a nonadiabatic coupling to phonons which worked quite well for CuGeO₃ [39, 232].

Severe deviations from such models show up as an extended fluctuation regime above the transition temperature, a very large spin gap and strong phonon anomalies. The compounds will be discussed in a sequence with increasing dimensionality of the effective spin system that is established at low temperatures going from the 1D NaTiSi₂O₆ via the 2D TiOCl to the 3D MgTi₂O₄.

10.5.1 The Pyroxene NaTiSi₂O₆

The compound NaTiSi₂O₆ is based on the precious stones or minerals NaAlSi₂O₆ and LiAlSi₂O₆ of the pyroxene family. Chains of edge-shared TiO₆ octahedra along the *c* axis of the structure exist that are well insulated by SiO₄ groups [233]. These chains promote an antiferromagnetic exchange between the Ti³⁺ as shown in the high temperature magnetic susceptibility in Fig. 10.26 following a Curie-Weiss dependence with $\Theta_{\rm CW} = -255$ K and C = 0.375 emu.K/mol [98]. For smaller temperatures magnetic correlations develop that can be modelled by a 1D AF Heisenberg spin system with J = 295 K. However, before the maximum of the susceptibility is reached a sharp drop of the susceptibility at T_c=210 K sets in.

This transition is connected with a lattice distortion and a dimer formation of the Ti³⁺ sites along the *c* axis [234]. The Ti-Ti distances alternate from the homogeneous 3.17 Å to distances of 3.05 Å and 3.22 Å, i.e. by 5.3%. A critical exponent of $\beta(2\Theta) \approx 0.16$ has been determined [234]. A broad maximum in the specific heat corresponds to a change in the entropy of $\Delta S \approx 2.8$ J/mol K, about half of the magnetic entropy that would have been released due to a purely magnetic phase transition, $\Delta S = R \ln(2S + 1) = 5.76$ J/mol K.

The sharp drop of the susceptibility can be modelled in the low temperature regime assuming a spin gap of $\Delta_{01} \approx 500$ K. Scaling the transition



Fig. 10.26. Susceptibility $\chi(T)$ of a) NaTiSi₂O₆ and b) TiOCl together with a fit $\chi_{1D}(T)$ to a AF Heisenberg spin chain with J = 295 K and 660 K (full lines), respectively [96,98]. The dashed line in (a) shows the high temperature dependence following a Curie-Weiss law $\chi_{CW}(T)$ with $\theta_{CW} = -255$ K. The inset compares the low temperature susceptibility with a fit using $\Delta \approx 500$ K. The inset in (b) shows the susceptibility in the proximity of the transitions after subtracting a defect contribution

temperature to this gap a reduced gap ratio of $2\Delta/k_{\rm B}T_{\rm SP} = 4.8$ is determined for NaTiSi₂O₆ [98]. This ratio is much larger compared to earlier observations and theory of spin-Peierls transitions with $2\Delta/k_{\rm B}T_{\rm SP} = 3.53$ derived using the BCS formula in a weak coupling regime [231]. For the transition in NaV₂O₅ also a larger value of $2\Delta/k_{\rm B}T_{\rm CO} = 6.4$ has been determined [190,235]. Here, however, the spin gap formation is related to charge ordering and strong electronic correlations [54, 193]. The gap Δ_{01} of TiOCl is also large with respect to the exchange coupling constant J leading to a ratio $\Delta_{01}/J \approx \Delta_{01}/\Theta_{\rm CW} \approx 0.5$ otherwise only achievable in spin ladders.

Large optical phonon anomalies are observed in NaTiSi₂O₆ at $T_c=210$ K as frequency shifts and line widths broadenings [236]. These effects point to a common origin of the spin gap formation and lattice distortion as a orderdisorder transition of the orbital orientation and consecutive Jahn-Teller distortion. Figure 10.27 shows the temperature dependence of some modes that show large anomalies and two relevant orbital configurations. A deeper understanding of this transition is still in progress. The coupling of high energy electronic to low energy spin degrees of freedom should be made responsible for the deviations from mean field behavior.

10.5.2 The Bilayer System TiOCl

The 2D titanate TiOCl has a layered structure formed by $Ti^{3+}O^{2-}$ bilayers, separated by Cl⁻ bilayers. The basic TiCl₂O₆ octahedra are strongly distor-



Fig. 10.27. a) Frequencies of anomalous phonon modes in NaTiSi₂O₆ as function of temperature and two orbital configurations of Ti^{3+} that are involved in the orbital ordering and spin gap formation [236]. The diagonals of the cube faces correspond to the crystallographic *c* axis



Fig. 10.28. a) Projection of the crystal structure of TiOCl along the *a* axis [237], b) electronic level scheme and a sketch of the relevant t_{2g} orbitals on a net of Ti ions. The intralayer d_{xy} orbitals constitute chains along the crystallographic *b* axis [96]. The interlayer d_{xz} , d_{yz} orbitals connect upper and lower Ti sites of the bilayer (larger and smaller dots)

ted and build an edge-shared network in the ab plane of the orthorhombic unit cell (Fig. 28).

The magnetic susceptibility $\chi(T)$ of TiOCl is shown in Fig. 10.26. It is very flat and has a broad maximum at $T_{max} = 400$ K [96]. The realization of a RVB ground state has therefore been proposed for this system [238]. A fit of a 1D Heisenberg model to the susceptibility, however, leads to a satisfactory modelling of the data with J = 660 K for T>130 K.

To understand this behavior the magnetic exchange paths have to be identified. In *a* axis direction a bridging oxygen exist with large bonding angles of $\approx 153^{\circ}$ and Ti-Ti distances of 3.79 Å. In *b* axis direction the Ti-Ti distance 3.38 Å is much smaller. Only in the ac-plane a shorter distance of 3.21 Å within the bilayer exist. Considering the t_{2g} orbitals d_{xy}, d_{xz}, d_{yz} that point to the edges of the distorted octahedra then d_{xz}, d_{yz} are oriented in the latter direction and are quasi-degenerate. In *b* axis direction, however, a linear chain of d_{xy} orbitals form that may be the basis of the 1D susceptibility. A band structure calculations supports this view as the d_{xy} orbital is occupied and is at lowest energy. Using Ti-Ti d_{xy} hopping matrix elements along this direction an exchange coupling of $J_{LDA+U}=t^2/U=720$ K [96] is roughly estimated neglecting the problems that a direct Ti-Ti overlap for such calculations imposes [3, 240].

At lower temperatures the susceptibility in Fig. 10.26 markedly changes. Two kinks and a sharp drop mark phase transitions of first and second order at $T_c^{1st} = 66$ K and $T_c^{2nd} = 94$ K. The first order phase transition also involves a static structural component as a doubling of the *b* axis is observed below this temperature [241]. The first microscopic information about these instabilities comes from recent NMR/NQR experiments on 35 Cl and 47,49 Ti.

Single NMR lines are observed for both Ti and Cl sites for temperatures above T_c^{2nd} indicating only one kind of Ti and Cl site in TiOCl. Below this temperature the lines broaden significantly and finally split into two lines. As shown in Fig. 10.29b both transition temperatures show up in the frequencies. The behavior of the relaxation rates for the two sites is definitely different. $1/T_1$ at ³⁵Cl sites has a high temperature onset ≈ 200 K and shows a cusp at $T_c^{2nd}=94$ K. It should be attributed to dynamic lattice distortion that gradually develop below this temperature scale.

NMR at the 47,49 Ti sites shows a decrease in $1/T_1$ at high temperatures that forms a maximum in $1/T_1$ T at T<T*=135 K. This implies a decrease of the spin fluctuations already in the homogeneous state of the spin system and defines T* as a fluctuation or pseudo gap temperature of TiOCl.

From a fit to $1/T_1T\propto \exp(-\Delta_{01}/k_BT)$ a spin gap can be deduced with $\Delta_{01}\approx 430$ K [239]. This gap with a reduced gap ratio of $2\Delta/k_BT_{SP} = 10{\text -}15$ is extraordinarily large and not consistent with a spin-Peierls mechanism in the weak coupling limit [231]. The pseudo gap phenomena and large fluctuation effects resemble to high temperature superconductors.

The important role of the lattice system is supported by pronounced anomalies of the optical phonons with interlayer displacements that involve Ti and Cl sites [242]. In Fig. 10.30 Raman spectra of TiOCl are shown for $T>T_c^{2nd}$ where NMR evidences the opening of the pseudo gap. Three inter-



Fig. 10.29. a) $1/T_1$ and b) NMR frequencies at ³⁵Cl. c) $1/T_1$ and $1/T_1T$ at ^{47,49}Ti sites of TiOCl [239]



Fig. 10.30. Phonon Raman scattering in TiOCl in the temperature interval from 100-160 K with $\Delta T=5$ K. Phonon lines that origin from the Brillouin zone center (boundary) are marked by full (dashed) arrows. Please note the discontinuous evolution of the scattering intensity for T<T*=135 K [242]

layer phonon modes with c axis displacements are dominant, a Cl-Ti in-phase mode at 203 cm⁻¹, a O-Ti out-of-phase mode at 365 cm⁻¹ and a Ti-Cl out-of-phase mode at 430 cm⁻¹. These frequencies agree reasonably well with results from a shell lattice model. The Raman spectra, however, are dominated for

 $T>T_c^{1st}$ by a very broad excitation that drastically softens approaching T^* from high temperatures. This softening leads to a shift from 160 cm⁻¹ to approximately 130 cm⁻¹.

Based on these effects it has been suggested, that the dominant mode at $130 - 160 \text{ cm}^{-1}$ is a fluctuation-induced zone-folded mode corresponding to the Γ -point phonon at 203 cm⁻¹ [242]. A thermal population of this "softmode" admixes $t_{2g} d_{xz}$, d_{yz} orbital states to the d_{xy} ground state and enhances the role of additional competing exchange paths [240]. This admixture is proposed to be responsible for the extremely large pseudo gap evidenced in NMR and other anomalies in ESR [243]. This scenario is also in agreement with the stepwise increase of intensity of the folded phonon mode at $T \approx 135$ K in good agreement with the fluctuation scale defined by the NMR experiments. Interesting to note is that at low temperatures $T < T_c^{1st}$ the broad anomalies are replaced by a larger number of very sharp phonons that clearly origin from long-range lattice distortions. Now the compound is again more close to a conventional spin-Peierls system. The situation resembles to the vanadate YVO₃ where a crystallographic distortion prepares a similar orbital structure as in TiOCl with a d_{xv} ground state and higher but still fluctuating d_{xz} , d_{vz} states. This leads in YVO₃ to a peculiar magnetization reversal in weak magnetic fields [244].

10.5.3 The Pyrochlore $MgTi_2O_4$

The compound MgTi₂O₄ realizes a 3D strongly frustrated pyrochlore lattice with s=1/2. It belongs to the spinel oxides AB₂O₄ that exhibit a large variety of interesting ground states with superconductivity, charge ordering or heavy fermion behavior [95]. For antiferromagnetically coupled Heisenberg spins on the B cations, as given for MgTi₂O₄ with Ti³⁺, a lattice of corner-shared tetrahedra is formed with maximum frustration and ground state degeneracy. Only recently a transition from a metallic to an insulating spin liquid ground state has been observed and partly attributed to orbital degrees of freedom [95].

In Fig. 10.31 susceptibility and resistivity of MgTi₂O₄ is shown. At T_c = 260 K $\chi(T)$ decreases strongly and $\rho(T)$ increases simultaneously [95]. This transition is also related to a structural distortion from cubic to tetragonal symmetry. The distortion may either suggest an orbital ordering or a Peierls-like scenario for the transition. The latter one can be compared to the metal-insulator transition in the rutile VO₂ [245]. The role of a 1D topology might be played by chains of edge-charing BO₆ octahedra. A local trigonal distortion may then lift the t2g orbital degeneracy into a_{1g} singlet and e_g doublet. An interplay of these bands and splitting of the a_{1g} due to electronic correlations should be responsible for the transition and the singlet ground state [95].



Fig. 10.31. Susceptibility and resistivity of the 3D pyrochlore MgTi₂O₄. At $T_c = 260$ K a strong decrease in $\chi(T)$ and a sudden increase in $\rho(T)$ are observed [95]. The insets show the susceptibility after subtracting a defect contribution and the logarithm of the conductivity vs. 1/T

10.6 Conclusion

For the presented cuprates, vanadates and titanates a peerless richness of phases and quantum spin phenomena has been highlighted. While for the cuprates recent interest is centered on spin liquid states based on unconventional topologies, the quantum nature of vanadates and titanates are often realized due to the interplay of orbital configurations with magnetism. The presented advance of understanding is related to the significant progress made in materials science of complex transition-metal oxides as well as to the motivating interplay between experiment and theory that is very characteristic for this area of physics.

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