# Large pyroelectric effect in Fe-doped lithium niobate induced by a high-power short-pulse laser

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We demonstrate the pyroelectric effect induced by a short pulse laser with an intensity up to 6 $MW/cm^2$ which is used to heat the Fe-doped Lithium niobate crystal. 0.2 °C temperature rise induced by a 10 ns laser pulse causes a very large current peak (up to 25 mA) since the rate of temperature rise is extremely large comparing with that induced by using an ordinary heater. On the other hand, the pyroelectric current during the cooling is much smaller because of the slow cooling rate by thermal conduction. © *2010 American Institute of Physics*. [doi:10.1063/1.3481380]

The pyroelectric effect is fundamentally based on the temperature dependence of the spontaneous polarization of a polar material. Ferroelectric lithium niobate ($LiNbO_3$, abbreviated as LN) is a typical polar material that exhibits a large pyroelectric effect. Because LN can emit electrons with energies up to $10^5$ eV and can generate electric fields as high as $10^6$ $Vcm^{-1}$,[1] it is the subject of many studies, including a comprehensive review article by Rosenman *et al.*[2] Because of their large pyroelectric effect, LN and lithium tantalate ($LiTaO_3$, abbreviated as LT, a material closely related to LN) are currently used for infrared sensors, x-ray generators, neutron generators, and tabletop nuclear fusion systems.[3–6]

The polarization charges of a ferroelectric crystal are exposed on the surface perpendicular to the direction of the spontaneous polarization. In air, however, these charges are compensated or neutralized by "screening charges," which originate from ionized particles absorbed from the environment.[7] The spontaneous polarization varies with temperature depending on the nature of the pyroelectric material, and the generation of electrons at the surface is explained qualitatively by the appearance of uncompensated charges on the polar faces of the crystal during thermal cycles.

When two opposing polar surfaces are electrically connected, a pyroelectric current can be measured with an ammeter during a temperature change. The pyroelectric current density $J_{pyro}$ can be expressed as

$$J_{pyro} = -\delta Ps/\delta t = -(\delta Ps/\delta T)(\delta T/\delta t) = p_3(\delta T/\delta t), \qquad (1)$$

where $t$ and $T$ are time and temperature, respectively, and $p_3 = \delta Ps/\delta T$ is the pyroelectric coefficient. Equation (1) suggests that $J_{pyro}$ is proportional to the rate of temperature change. The current is thus present only when the crystal temperature is changing, so the rate of change of temperature becomes a key factor in determining the enhancement of the pyroelectric current.

In the conventional method, a thermal heater is used to heat the crystal (e.g., Ref. 3). The heater or heat-gun method can heat the crystal to temperatures as high as 400 °C,[8] but the rate of heating is very slow. Moreover, rapid heating and cooling over such large variations in temperature can result in thermal shock and cracking of the crystal.

An Fe-doped LN crystal (about 600 ppm Fe) was cut and polished to form a plate with polar faces perpendicular to the $Z$ axis and with dimensions of $10 \times 10 \times 0.5$ $mm^3$. Both $+Z$ and $-Z$ faces were coated by 10-nm-thick indium-tin-oxide (ITO) films with no antireflection coating. The sample was clamped vertically on a Teflon plate with two small brass blocks that also served as electrodes. One electrode was connected to an oscilloscope and the other was grounded. A high-power green (wavelength 532 nm) pulsed laser with a pulse width and repetition rate of 10 ns and 10 Hz, respectively, was used to heat the sample. The profile of the pyroelectric current $I_{pyro}$ was measured with sub-nanosecond time resolution using an oscilloscope.

Figure 1 shows that the peak pyroelectric current is linearly related to the light power incident on the crystal, which we define as the peak laser intensity multiplied by the irradiated area. The beam diameter (at $1/e^2$ of the peak power) was 4.25 mm, and the peak power was varied from 0.5 to $8 \times 10^5$ W. At the highest power, we observe a peak current of 25 mA, which is huge compared to the hundreds of nanoamperes or several microamperes that are observed when heating with a heat gun.[8]

Figure 2 shows the relationship between the pyroelectric current and the irradiated area for constant power densities of

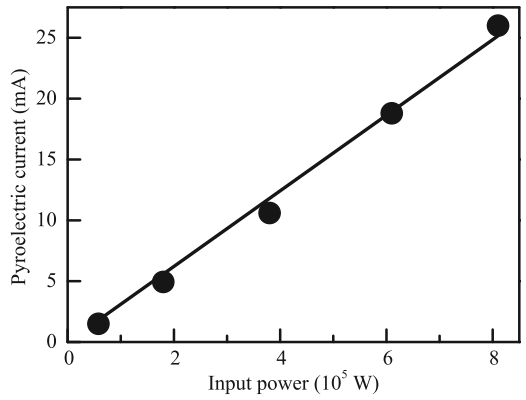


FIG. 1. Pyroelectric current $I_{pyro}$ as a function of laser irradiation power.

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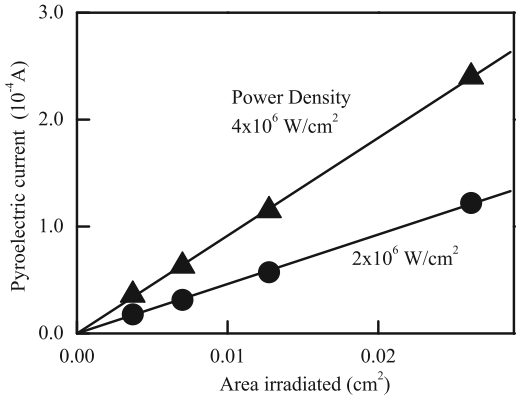


FIG. 2. Dependence of pyroelectric current $I_{\text{pyro}}$ on beam area for two different power densities.

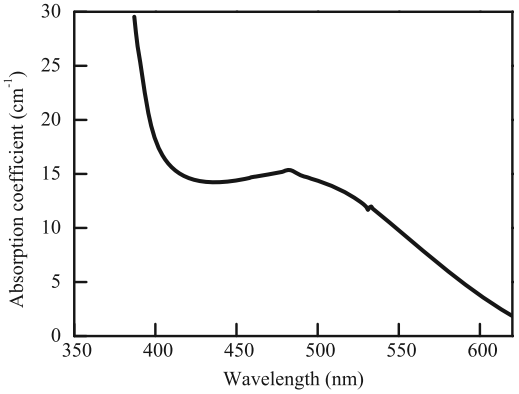


FIG. 4. Absorption spectrum of Fe-doped LN coated with ITO.

4 and $2\times10^6$ W/cm$^2$. The beam waist was varied from 0.7 to 1.8 mm, which translates into a variation in irradiated area on the crystal surface from 0.4 to 2.5 mm$^2$. These data demonstrate that for constant power density, the pyroelectric current is proportional to the irradiated area. The sample size does not influence the pyroelectric current in this method, which means that only the irradiated area is heated by the laser beam. The portion of the crystal not irradiated is not heated and serves as a heat sink.

Figure 3 shows the time-dependent pyroelectric current when the crystal is irradiated by a pulse with intensity $6\times10^6$ W/cm$^2$. During the 10 ns irradiation period, the crystal is heated. The data show that even after the 10 ns irradiation, the current decreases but exists, which indicates that the crystal temperature is still increasing some 30 ns after the laser irradiation.

In addition, we find that the current in the opposite direction, which is usually observed while the crystal cools, is very small. We detected this small, oppositely directed current from about 30 ns after irradiation, and it lasts until approximately 300 ns before dropping below the noise level. In other words, upon irradiation with a 10 ns laser pulse, the heating and cooling cycle for LN is approximately 300 ns.

When a crystal with absorption coefficient $\alpha$ and thickness $L$ is heated by light with intensity $I_0$ over an area $A$, Eq. (1) can be modified to

$$J_{\text{pyro}} = p_3(T/\delta t) = I_{\text{pyro}}/A = p_3 I_0(1-\exp^{-\alpha L})/(Lc_p\rho) \sim p_3 I_0 \alpha/(c_p\rho) \quad (\text{when } \alpha L \ll 1), \tag{2}$$

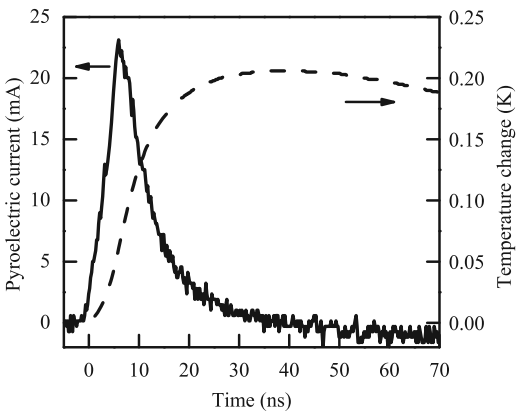


FIG. 3. Time dependence of pyroelectric current and calculated temperature change.

$$T = \frac{1}{Ap_3}\int I_{\text{pyro}}dt, \tag{3}$$

where $c_p$ is the specific heat at constant pressure and $\rho$ is the specific gravity. With the measured $I_{\text{pyro}}$ and material constants such as $p_3=-83$ $\mu$C m$^{-2}$ K$^{-1}$,[9] $c_p=602$ J K$^{-1}$ kg$^{-1}$, and $\rho=4650$ kg m$^{-3}$ taken from the literature, the absorption calculated with Eq. (2) is 14.8 cm$^{-1}$ for the experimental conditions of $I_0=5.2\times10^5$ W·cm$^{-2}$ and $A=0.56$ cm$^2$ (reflection at the incident surface is not considered). Figure 4 shows an absorption spectrum of the Fe-doped LN crystal used with 10-nm-thick ITO films on both faces. The absorption coefficient $\alpha$ at 532 nm is about 15 cm$^{-1}$, which is in fair agreement with the value calculated above.

The temperature change can be calculated from Eq. (3). The maximum temperature rise is calculated to be 0.2 °C 30 ns after the laser irradiation, as shown in Fig. 3, following which the temperature slowly decreases. In our experiment, the temperature change is very small but the heating rate $\delta T/\delta t$ is extremely fast—of the order of $10^7$ K/s. Because the heating rate is around 1 K/s with the conventional heat-gun method, only several hundred nanoamperes are observed.[8] Such a large difference in currents between heating with a short laser pulse (tens of milliampere) and with an ordinary heater (hundreds of nanoampere) is due to the difference in the factor $\delta T/\delta t$ between the two cases.

Figure 3 also shows that only a very small current is observed in the opposite direction while the crystal cools. Normally, the current in the opposite direction that is observed during cooling is of the same magnitude as (or larger than) the current observed during heating because the factor $\delta T/\delta t$ for cooling is almost same as, or larger than, for heating. In our case, heating by laser-pulse irradiation is very fast but cooling based on thermal conduction is very slow.

Although LN exhibits a comparatively large photovoltaic effect, the coefficient is much smaller (one fifth to one tenth) than the pyroelectric coefficient.[10] In the present study, the photovoltaic current was not taken into account.

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