Time dependence of ferroelectric coercive field after domain inversion
for lithium-tantalate crystal

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We found the ferroelectric coercive field of LiTaO₃, both in forward and reverse direction, vary with time after the domain is inverted. The coercive field drops when the domain is inverted, then gradually recovers. This phenomenon is light sensitive. The existence of a net time-varying internal electric field after domain inversion is hypothesized. The internal field is composed of the depolarization field, which is due to the spontaneous electric dipole moments, and an opposite direction time-varying space-charge field which is due to the redistribution of free-carriers transport under the influence of the depolarization field. Electro-optical effect caused by the internal electric field has been observed by means of an in situ optical monitoring technique for the domain inversion process. The in situ optical monitoring technique is based on using the LiTaO₃ thin-plate crystal as a low finesse Fabry–Perot interferometer. © 1995 American Institute of Physics.

Quasi-phase matching (QPM) by periodical inversion of the nonlinear optical coefficient can achieve high conversion efficiency for second-harmonic generation (SHG). Periodically inverted ferroelectric domain for achieving this purpose has been demonstrated on laminated and waveguide LiNbO₃ crystals. LiTaO₃ is known for its large nonlinear susceptibility, short wavelength transparency, and better resistance of photorefractive effect. Intensive studies on fabrication, characterization, and performance of SHG device for blue and green lasers on periodically inverted ferroelectric domain LiTaO₃ are taking place, but studies on basic material properties of LiTaO₃ in the domain inversion process are lacking. In this letter, we report our findings and analysis on time dependence of the ferroelectric coercive field after domain inversion of the LiTaO₃ crystal.

In this work, we used 0.5 mm thick SAW grade z-cut LiTaO₃ crystals. Electric field poling method was used to invert the domain. The external field was applied to the crystal plate along the crystallographic c axis through transparent liquid electrodes that were in direct contact with the crystal plate sur faces. The external electric field started from 0 V and ramped up at a rate of 16 V per second. Displacement current was measured during the ramping-up period of the external field. The external field was turned off after the displacement current reached its maximum value and dropped back to zero. The external field strength at which the maximum displacement current occurred was taken as the coercive field. It is known, for LiTaO₃, the ferroelectric hysteresis loop is asymmetric, and the crystallographic c axis is the easy axis. We shall call the direction along which the external field inverts the domain at higher strength the “forward” direction, and the opposite direction along which the external field inverts the domain at lower strength the “reverse” direction. The procedure for measuring the coercive field in forward direc-

In Figs. 1(a) and 1(b), O curves were measured in dark, triangular and square curves were measured under illumination by a fiber-guided tungsten lamp at 40 and 160 mW total optical power level measured at the output end of the fiber, respectively. The illumination was concentrated on a circular area of ~1 cm diam on the sample surface, and it was kept on constantly throughout the measurement process. Figures 1(a) and 1(b) show that the coercive field dropped right after domain inversion, then recovered. The recovery was fast in the first few minutes after the domain was inverted, then gradually slowing down. We have ob-

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served the recovery process in the time frame of months, the coercive field of forward direction slowly recovering toward its original value of 10 900 V/0.5 mm. These figures also show that the phenomenon is dependent on the intensity level of a relatively white-lighted illumination. The light intensity dependence is stronger for the reverse rather than the forward direction of domain inversion. We have also observed the light-sensitive behavior with SUV illumination for both directions, since the intensity level of the SUV source was not determined, the result for SUV illumination is only qualitative.

Since the coercive field drops after domain inversion, a smaller external field can invert the domain in the following inversion operation. It implies that an internal field might exist after previous inversion, such that the internal field would assist the external field in the following inversion operation. The internal field should be in the opposite direction to the spontaneous polarization, and it is time varying; it gradually decreases to zero, so that the coercive field gradually recovers.

Temperature of the sample surface during light illumination was monitored with a direct in-contact thermal couple. Without knowing the strength of the total field, the effect produced by light illumination is not temperature dependent, but it is possible related to effects of photogenerated charge carriers.

On the basis of this reasoning, we hypothesize the origin of the phenomenon as follows: There exists a time-varying internal electric field in the crystal after domain inversion. The internal field is composed of the depolarization field caused by the spontaneous dipole moments and a time-varying space-charge field that is in the opposite direction to the depolarization field. After domain inversion, the space-charge field gradually builds until it becomes equal to the depolarization field in magnitude. The space-charge field is due to redistribution and possibly trapping by impurities of free carriers, which consist of the dark carriers and photogenerated carriers. The carriers transport in the crystal under the influence of the depolarization field. The space charges recombine when the spontaneous polarization is inverted, then regenerate and redistribute after the inversion. For low electric conductivity dielectric crystal, the regeneration and redistribution processes of the charge carriers would be slow, these processes would also be dependent on the concentration of the photogenerated charge carrier, therefore, on the intensity level of the illumination.

We have observed that charged toner particles attached strongly to the newly domain-inverted area on the crystal plate. This observation provided a direct qualitative evidence for the existence of an internal field after domain inversion.

An electro-optical effect should be observed if an internal field exists. We have designed a simple in situ optical monitoring technique to optically monitor the domain inversion process and time evolution of the internal field. The setup is a simple, low finesse Fabry–Perot interferometer. We used a low power He–Ne laser beam incident with the crystal plate at normal angle along the c axis through the transparent liquid electrode. The reflected light from the top and bottom surface of the crystal plate interfere because of the path difference $\beta$

$$\beta = 2\pi nd/\lambda,$$  \hspace{1cm} (1)

where $n$ is the ordinary refractive index of the crystal, $\lambda$ is the wavelength of the He–Ne laser, and $d$ is the thickness of the crystal plate. The change in the ordinary refractive index $\Delta n$, caused by the electro-optical effect follows the relationship\textsuperscript{14}

$$\Delta n = -\frac{1}{2} \gamma_{13} n^3 E_t - \frac{1}{2} S_{13} n^3 E_t^2,$$  \hspace{1cm} (2)

where $\gamma_{13}$ is the linear electro-optical coefficient, $S_{13}$ is the quadratic electro-optical coefficient, and $E_t = E_{ex} + E_{in}$. The total field $E_t$ is composed of the external field $E_{ex}$ and the internal field $E_{in}$. All fields are in either + or − c-axis direction. Without knowing the strength of the total field, the quadratic electro-optical effect should not be ignored.

The reflectance $R$ of the system is given as\textsuperscript{15}.
The reflectance was changing as the external field was increasing. Around time $t_1$, displacement current occurred and maximum displacement current occurred at the time when the reflectance underwent an abrupt change, indicating a fast sign change of $E$, relative to the inversion of the spontaneous polarization, and the direction of the $c$ axis such that the reflectance traced backward abruptly according to Eqs. (1)–(3). At time $t_2$, the external field was turned off and it dropped back to zero in about 5 s. The reflectance kept on changing after the external field was turned off. According to Eqs. (1)–(3), the changing reflectance after turning-off of the external field indicated the existence of a time-varying internal field.

We have also observed that with oblique incidence, the reflectance and transmittance of both polarization of the incident He–Ne laser beam undergoes large changes after domain inversion and removal of the external field. This observation indicated that both ordinary and extraordinary refractive index are changed by the internal field through the electro-optical effect.

One can obtain the magnitude of the internal field and its time evolution directly from Figs. 1(a) and 1(b) for time interval greater than $\sim$100 s, which is about the shortest time interval during which we can reset our instruments for the next domain inversion operation. In principle, it is also possible to calculate the magnitude of the internal field and its evolution by using data in Fig. 2(b) and Eqs. (1)–(3), providing that the quadratic electro-optical coefficient is known. It would also be important to further explore the wavelength dependence of this phenomenon. We expect that the wavelength dependence would rely on the band structure and the impurity levels’ distribution in the band gap of LiTaO$_3$.

In conclusion, we have found that the coercive field of LiTaO$_3$ crystal drops right after domain inversion and recovers gradually. This phenomenon is light sensitive. We postulated the existence of an internal field to account for this phenomenon. We have observed the electro-optical effect caused by the internal field by means of an in situ optical monitoring technique for the domain inversion process.

\begin{equation}
R = |r - re^{-j2\beta}|^2 |1 - r^2e^{-j2\beta}|^2,
\end{equation}

where $r$ is the Fresnel reflection coefficient of the liquid electrode-crystal plate interface.

Because of the large difference between $d$ (0.5 mm) and $\lambda$ (6328 Å), a change of $6.3 \times 10^{-4}$ in $n$ would produce a change of $\pi$ in $\beta$, according to Eq. (1). Therefore, according to Eqs. (1)–(3), a small change in $n$ would produce a large variation in reflectance as the total field is varying.

Figure 2(a) shows a typical measured reflectance versus time curve for which the external field was in the direction of spontaneous polarization, such that there is no domain inversion throughout the ramping-up of the external field. The reflectance followed a sinusoidal according to Eqs. (1)–(3) as the external field was ramping up. The external field was turned off at time $t_0$. The reflectance traced backward rapidly as the external field abruptly decreased to zero in about 5 s. Figure 2(b) shows a typical measured reflectance versus time curve for which the external field was opposite to the spontaneous polarization at the beginning of the ramping-up. The reflectance was changing as the external field was increasing. Around time $t_1$, displacement current occurred and maximum displacement current occurred at the time when the reflectance underwent an abrupt change, indicating a fast sign change of $E$, relative to the inversion of the spontaneous polarization, and the direction of the $c$ axis such that the reflectance traced backward abruptly according to Eqs. (1)–(3). At time $t_2$, the external field was turned off and it dropped back to zero in about 5 s. The reflectance kept on changing after the external field was turned off. According to Eqs. (1)–(3), the changing reflectance after turning-off of the external field indicated the existence of a time-varying internal field.