

Domain kinetics during polarization reversal in 36° Y-cut congruent lithium niobate

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Abstract. We present experimental study of domain kinetics in 36° Y-cut congruent lithium niobate during polarization reversal by increasing external electric field using liquid electrodes. The motion of the domain walls has been imaged on the Z_s^+ and Z_s^- polar surfaces of the crystal simultaneously. The linear dependence of the threshold field on the field rate has been revealed. The threshold field in low field rate limit has been estimated as 32.8 ± 0.3 kV/mm, which is lower than predicted value. The switching process for polarization reversal in the constant field has dramatically accelerated with increase of external electrical field above threshold. The new approach, which accelerates significantly the *in situ* study of 3D domain kinetics in CLN crystals, has been demonstrated.

1. Introduction

The middle-infrared (mid-IR) wavelength range can be defined as a portion of electromagnetic spectrum varied from edge of near-infrared (2 μ m) to tens of microns. Mid-IR spectral range has a number of so-called “water windows” representing light wavelengths, which can propagate through Earth’s atmosphere without absorption by water vapour [1]. Moreover, most molecules have specific absorption lines in the mid-IR, affording a description that makes it possible to determine them by optical spectroscopy [2].

To create the optical frequency mixers adjusted to the ultra-short pulse sources (below 100 fs) and high average power (above 10 W) in mid-IR, it is necessary to obtain non-linear components with large aperture (about 1 cm²). The periodically poled nonlinear optical crystals of lithium niobate (LN) and lithium tantalate (LT) are well fitted to the optical parametric amplification in the mid-IR due to their wide transparency range and high non-linearity, but the routine poling technique is not suitable for producing the components with appropriate large aperture [3].

In order to overcome this technological barrier, the “slanted poling” (axis-slant quasi-phase matching) has been elaborated [4]. The essence of the method is to produce periodical grating in a plate with slanted polar axis, specifically in 25° X-cut MgO-doped LN. In this work, we have investigated 36° Y-cut congruent LN (CLN) crystals, which are used for surface acoustic wave devices. The main advantages of these crystals are low price and great offer on the market.



2. Experimental

The studied samples represented CLN plates cut in such a way that the angle between normal to the crystal surface and Y-axis was 36° . The 0.5-mm-thick 3-inch wafers were polished from both sides to optical quality and cut on samples with sizes $10 \times 15 \times 0.5 \text{ mm}^3$. All experiments were performed at room temperature.

Polarization reversal was performed by application of external electric field pulses of two shapes: (a) rising field (Fig. 1b) and (b) constant field (Fig. 2b), using liquid electrodes (aqueous solution of lithium chloride). The top electrode 2 mm in diameter was located on the surface corresponding to positive direction of the slanted polar Z-axis (Z_s^+), and the continuous bottom electrode was located on the opposite side (Z_s^-).

Magnitude of the applied external electric field ranged from 30 to 35.2 kV/mm. *In situ* imaging of the domain structure was made by polarizing microscope (Carl Zeiss LMA10, Germany) with magnification 3.2x and recorded by the high-speed CMOS camera (Photron Mini UX100, Japan) with film rate varied from 50 to 250 fps.

Resulting videos were analysed by methods of image processing realized in Wolfram Mathematica software.

3. Results and discussion

Two types of the switching experiments were realized by application of the single pulses of the external field of different waveforms: switching in rising field and in constant field.

The key parameter of polarization reversal in any ferroelectric is excess of the switching field (E_s) above the threshold value (E_{th}) defined as $\Delta E_{ex} = E_s - E_{th}$. It is known that the threshold field in Z-cut CLN is about 21 kV/mm [5]. Hence, the estimated threshold field of the 36° Y-cut CLN should be $E_{th}^N = E_{th}^Z / \sin 36^\circ \approx 35.7 \text{ kV/mm}$.

3.1. Switching in rising field

In order to measure dependence of E_{th} on the field rise rate (dE/dt) (Fig. 1a), several samples were switched by pulses with rising field (Fig. 1b) with dE/dt ranged from 0.2 to $1.3 \text{ kV/mm} \cdot \text{s}^{-1}$. Maximum applied field was 35.2 kV/mm. The application of the pulses with higher field amplitude led to a breakdown. The threshold field was estimated as a field value, at which the first domain became visible. Linear fitting shows that the threshold field for quasi-static switching along the normal to the sample $E_{th}^N = 32.8 \pm 0.3 \text{ kV/mm}$, which is lower than predicted value. This fact can be attributed to formation of the first domains at the electrode edges with higher field value due to a fringe effect.

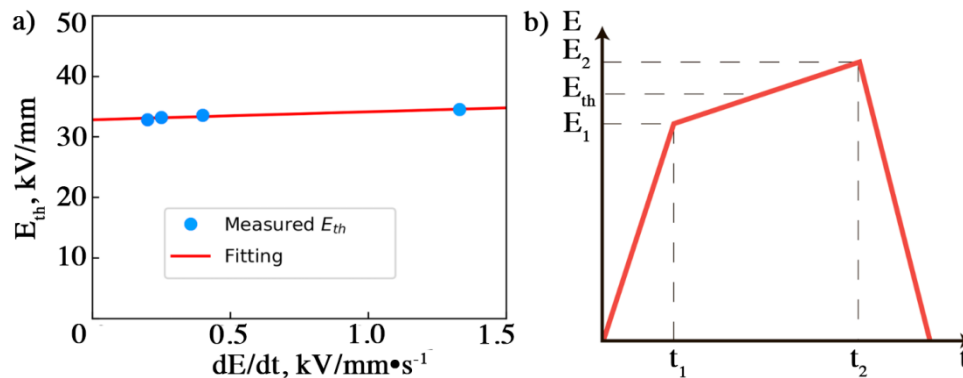


Figure 1. (a) Dependence of the threshold field (E_{th}) defined as a field value at the moment, when the first domain became visible on the field rise ($\Delta E/\Delta t$). Experimental points were fitted by the linear dependence.

(b) The waveform of the rising field pulse. $\Delta t = t_2 - t_1$, $\Delta E = E_2 - E_1$.

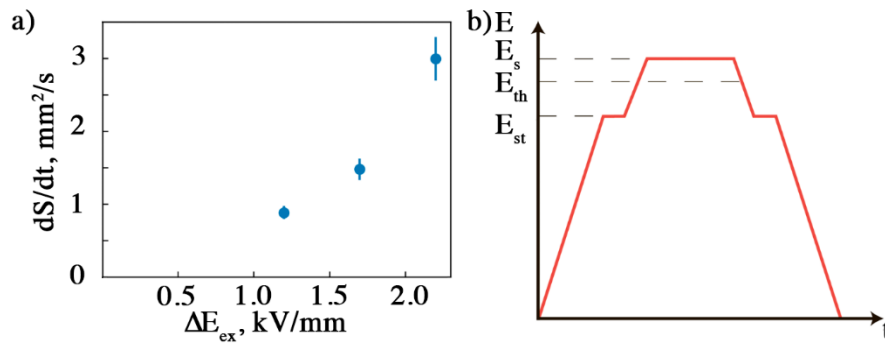


Figure 2. (a) Dependence of the switched area growth rate (dS/dt) on the excess of the switching field over the threshold value (ΔE_{ex}) for polarization reversal in constant field. (b) Waveform of the pulse used for switching in constant field. E_s - switching field, E_{st} - stabilization field, E_{th} - threshold field.

The kinetic maps [6] shown in Figure 3 were obtained by successive overlapping the domain wall positions recorded with frequency 5 Hz during polarization reversal by rising field pulse with $dE/dt = 0.2 \text{ kV}/\text{mm}\cdot\text{s}^{-1}$. The domain wall positions were detected by consistent application of an edge detection pipeline to the video frames. The edge detection pipeline included the following steps: 1) digital noise reduction by subtraction of the first frame (the image of the sample in the initial single domain state) and application of a curvature flow filter [7], 2) application of Canny Edge algorithm [8] to obtain the image with domain wall contrast only. The colour of each wall corresponds to the value of applied external field and the time, at which the wall was recorded. The final kinetic map represents a sum of the frames processed by the edge detection pipeline mentioned above. The maps demonstrate propagation of domain walls on Z_s^+ and Z_s^- surfaces. The clearly seen nonmonotonic change of the wall velocity can be attributed to pinning by defects and domain-domain interactions.

Dependence of the domain wall velocity along positive and negative projections of Y-axis on Z_s^+ surface on applied external electric field is shown in Figure 3c. The switching process always starts by appearance of isolated domains at the electrode edges. Two mechanisms are observed later: (1) slow jump like growth of the domains and (2) anisotropic wall motion after domain merging. For applied field just above the threshold (ΔE_{ex} ranged from 0.3 to 0.8 kV/mm), the walls stopped at the defects with local increasing of the threshold field (pinning centres). The walls restarted, when the applied field overcame the local threshold field value [9].

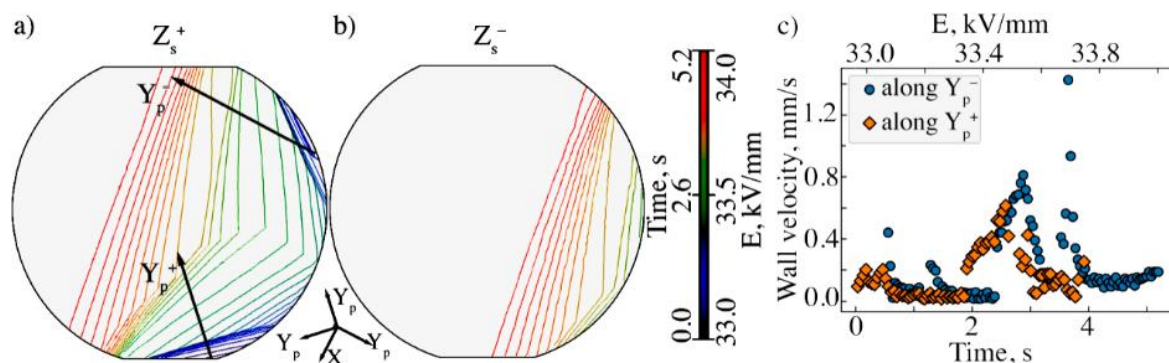


Figure 3. Kinetic maps of the domain structure evolution for polarization reversal in 36° Y-cut CLN by single pulse of rising field with field rate of $0.2 \text{ kV}/\text{mm}\cdot\text{s}^{-1}$ on (a) Z_s^+ and (b) Z_s^- surfaces. The domain wall colour corresponds to the time and applied external field. The time starts from appearance of the first visible domain in the field of vision. (c) The field dependence of the domain wall velocity along positive and negative projections of Y-axis shown by arrows on (a) on Z_s^+ polar surface.

It is known that the domain walls orientations in Z-cut CLN are defined by C_{3v} crystal symmetry. For equilibrium switching conditions characterized by effective screening of depolarization field, the domain walls are strictly oriented along Y-axis [10-13]. In our experiment, domain walls were oriented along projections of Y-axes at low fields ($\Delta E_{ex} \leq 0.8$ kV/mm) and their orientation on Z_s^+ and Z_s^- surfaces tended to X-axis with increasing of ΔE_{ex} . The obtained deviation from crystallographic directions is caused by increasing of the kink concentration at the wall [14,15].

3.2. Switching in constant field

The domain kinetics was studied in 36° Y-cut CLN in constant fields ranged from 34 to 35.2 kV/mm. The stabilization steps $E_{st} = 30$ kV/mm were added to the waveform before and after the switching step (Fig. 2b). The first stabilization step allowed preventing the crystal dielectric breakdown and the second one allowed minimizing the backswitching process [16,17].

Dependence of the switched area growth rate on the excess of the switching field above the threshold value (ΔE_{ex}) is shown in Figure 2a. It is seen that the relative increasing of the external electrical field in 0.0035 accelerated the polarization reversal three times.

4. Conclusion

The domain kinetics in 36° Y-cut CLN during polarization reversal by rising external electric field using liquid electrodes has been studied. The polarization reversal in tilted cut of CLN crystal gives information, which accelerates significantly the *in situ* study of 3D domain kinetics due to a possibility to observe domain walls motion on the Z_s^+ and Z_s^- polar surfaces of the crystal simultaneously. The kinetic maps containing complete information about wall motion have been used for analysis of data obtained during polarization reversal in rising field. Observed deviation of the domain walls from crystallographic axis is attributed to the increase of the kink concentration at the wall. The measured linear dependence of the threshold field on the field rate allows estimating the threshold field in low field rate limit as, $E_{th}^N = 32.8 \pm 0.3$ kV/mm which is lower than predicted value due to the fringe effect. The increase of external electrical field above threshold leads to a dramatic acceleration of the switching process for polarization reversal in the constant field. The obtained information will be used for further development of electric field poling technique in CLN crystals for high-power laser sources.

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