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Self-Organization in $LiNbO_3$ and $LiTaO_3$: Formation of Micro- and Nano-Scale Domain Patterns

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We review the study of self-organized formation of several types of quasi-regular micro- and nano-scale domain patterns in single crystalline $LiNbO_3$ and $LiTaO_3$ samples with artificial surface dielectric layers. The domain images revealed by chemical etching have been visualized using optical, scanning electron and scanning probe microscopy (SPM) in contact atomic force mode. SPM piezoresponse imaging mode allows us to investigate the domain structure under the sample surface. We classify the obtained quasi-regular domain structures and discuss the mechanism of their formation and self-maintained growth.

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1. Introduction

Domain engineering in ferroelectric crystals such as $LiNbO_3$ (LN) and $LiTaO_3$ (LT) has revolutionized their use in nonlinear optical applications [1]. It was shown that LN and LT with periodical 1D [2, 3] and 2D [4] domain structure open up a range of possibilities for bulk and waveguide nonlinear optical interactions. During ten years after the first electrical poling of bulk LN samples [5] research on periodically poled LN and LT is under intense interest around the world resulting in production of photonic devices. Breaking the micron-period barrier for periodical domain patterning in LN and LT is very desirable for several new electro-optic applications such as tunable cavity mirrors which need a periodicity of about 350 nm. The most efficient exploitation of engineered sub-micron domain gratings in LN and LT for photonic devices is related with the waveguide fabrication.

The commonly used electric field poling technology based on the application of a high voltage pulse through lithographically defined electrodes cannot be straightforwardly applied to the fabrication of sub-micron gratings. Thus for nano-scale domain engineering such alternative technique as "self-organized domain patterning," due to self-maintained evolution of domain structure, is under consideration [6]. In this work we classify all known self-assembled domain structures produced in LN and LT and suppose the mechanism of self-organization.

2. Experiment

Switching experiments were held in optical-grade single-domain LN and LT wafers with thickness ranged from 0.3 to 2 mm of congruent, stoichiometric and MgO-doped compositions cut perpendicular to the polar axis. The double-crucible modification of Czochralski technique was used for growth of stoichiometric LN and LT [7]. NiCr, In_2O_3:Sn (ITO) and liquid electrolyte (LiCl water solution) electrodes have been used for poling. The controlled artificial insulating layers of photoresist, spin-on-glass, silicon oil and air gap with 0.5 to 2 microns thick have been located between sample surface and one of the electrodes. The single rectangular electric field pulse with amplitude higher than coercive field has been applied to the electrode structure. The controlled backswitching (partial flip-back) was obtained after removing or rapid diminishing of external field [8]. The duration of the switching pulse and the field-diminishing amplitude are the crucial parameters for backswitching kinetics.

For periodical poling the wafers were lithographically patterned with periodic strip NiCr electrodes deposited on Z+ surface only and oriented along one of Y axis [9]. Patterned surface was covered by 0.5-μm thick photoresist layer. For observation of the domain patterns after partial poling Z surfaces were etched by HF without heating. The obtained surface relief was visualized by optical microscope, SEM and AFM techniques. Piezoresponse imaging mode (PRIM) was used for non-destructive domain visualization.

3. Domain Wall Motion and Growth of Isolated Domain

Our approach to domain patterning is based on understanding of key role of bulk screening effects [10, 11]. It is well known that polarization reversal from the single domain state is achieved by nucleation of new domains and their growth [12, 13]. Both processes are

governed by the elementary nucleation at the domain wall (domain growth) and far from the wall (arising of new domains). The local switching field E_{loc} averaged over the volume of nuclei is the driving force of these processes [10, 13]. $E_{loc}(r, t)$ is determined by the sum of external field $E_{ex}(r)$, the depolarization field $E_{dep}(r, t)$ produced by bound charges, and the screening fields due to the charge redistribution at the electrodes–external screening field $E_{scr}(r, t)$ and in the bulk—bulk screening field $E_b(r, t)$ [13, 14]

$$E_{loc}(r, t) = E_{ex}(r) + E_{dep}(r, t) + E_{scr}(r, t) + E_b(r, t)$$

The depolarization field slows the domain growth while the screening process reduces its influence.

Regular motion of plane domain wall is obtained for complete bulk screening, when the strictly oriented walls of a few regular-shape domains (hexagons for SLT and LN) are moved. The predominant switching mechanism is 2D nucleation of the steps at the existing walls with subsequent step motion along the wall by 1D nucleation (Fig. 1a). The domain walls orientation parallel to the crystallographic direction remained during switching.

Domain wall shape instability prevails for incomplete screening, as far as incompletely compensated by external screening depolarization field suppresses the slow regular domain growth as far as the step propagation along the wall is decelerated. The wall shape looses its stability with respect to shape perturbation thus leading to formation and growth of "fingers" (Fig. 1b). Such behavior is very similar to fingering growth in classical experiments with viscous liquids in Hele-Show cell and non-equilibrium growth [15]. It is important to point out that in this case the wall moves as a whole without violation of its continuity.

Wall motion through correlated nucleation by propagation of the boundary of switched area consisting of isolated domains appears when the bulk screening becomes absolutely ineffective. In this case the number of arrays consisting of small isolated domains appears in front of the wall (Fig. 1c). In proper conditions such mechanism leads to formation of the number of strip domains oriented along Y direction and parallel to the electrode edge (Fig. 1d).

Correlated nucleation in the vicinity of moving domain boundary dominates due to long-range electrostatic interaction. The crucial role of intrinsic or artificial dielectric surface layer (dielectric gap) is brightly displayed. There is much evidence for correlated nucleation

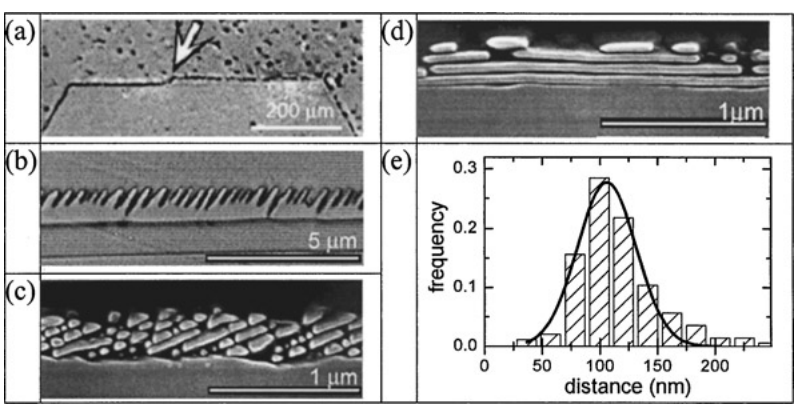

FIGURE 1 Variants of domain wall motion in CLN. (a) Regular motion by step propagation. Instantaneous image. (b) Finger-type instability of domain shape. (c) Propagation of nanodomain arrays. (d) Nanoscale strip domain structure. (a), (b) optical images. (c), (d) SEM images. (e) Distribution function of the domain periods for the strip structure.

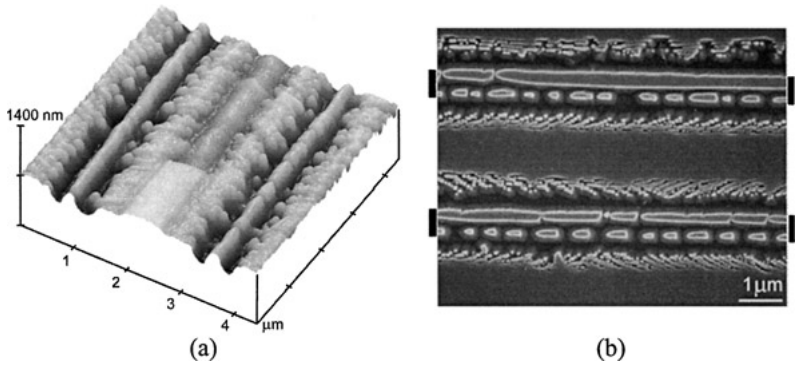

FIGURE 2 Nanodomain arrays in periodically poled CLN oriented along (a) Y^- directions with the fragment of electrode and (b) X directions. Z^+ view. Domain patterns were revealed by etching and visualized by (a) AFM and (b) SEM.

during switching in LN in uniform field. The common manifestation is the nucleation at a short distance from a domain wall, firstly discovered by us in lead germanate [16].

In the framework of our approach, correlated nucleation is due to the spatial distribution of polar component of the local field E_{loc} at the wall due to existence of intrinsic or artificial dielectric gap. Our calculations of the field distribution for a needle-like domain with charged domain walls show the field maximum situated at a distance of about the thickness of the dielectric gap from the wall [6]. This maximum exists just near the surface and determines the position of nucleation sites. The correlated nucleation plays the most important role during backswitching after removing of external field.

The obtained variety of self-organized domain structures arising in LN and LT can be divided in two groups by dimensionality: 1D and 2D structures.

4. 1D Domain Structures

Domain fingers arisen during domain broadening out of electroded area during domain patterning. In this case the fingers propagate in the area covered by artificial insulating layer. The fingers are strictly oriented along three Y^- directions, which are preferential for nucleation in LN and LT (Fig. 1b).

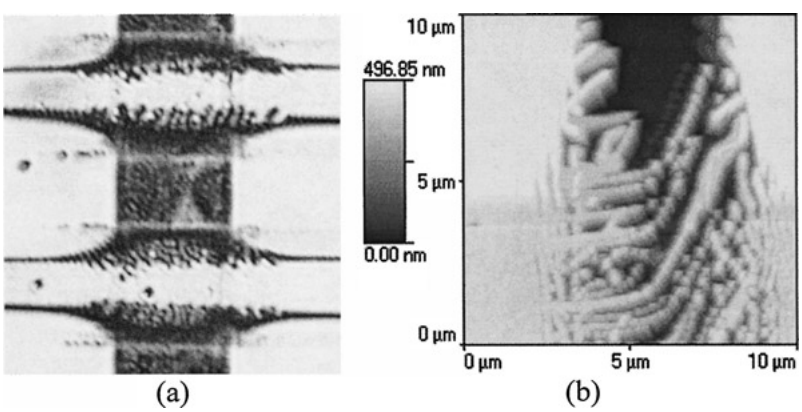

FIGURE 3 Sub-micron domain arrays appearing during poling of CLN with proton exchanged waveguides visualized by (a) optical microscope and (b) by AFM.

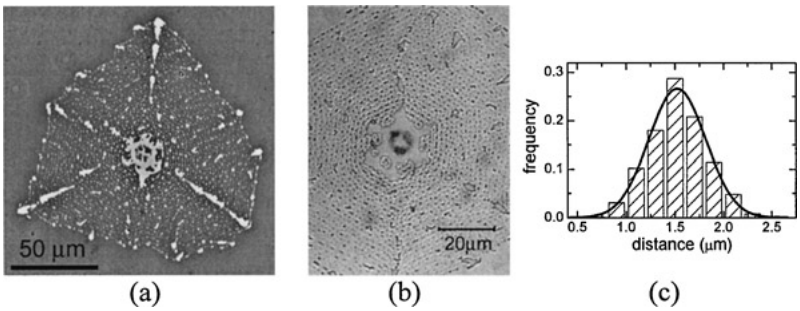

FIGURE 4 (a), (b) Web-structures in SLT with artificial dielectric layer revealed by etching. Optical observation. (c) Distribution function of the distances between neighboring domains.

Strip structures forms as a result of fast growth of domains in CLN parallel to the strip electrode edge (Fig. 1d). The statistic treatment of the several domain images allows to reveal that the structure is strongly organized with average period about 110 nm (Fig. 1d). This value can be related to the thickness of the intrinsic dielectric gap in CLN.

Nanodomain arrays appear easily during backswitch poling after field switch-off in LN in the vicinity of strip electrodes under artificial dielectric layer [6]. The arisen arrays consist of 30–100-nm-diameter needle-like domains with charged domain walls (Fig. 2). All arrays are strictly oriented along crystallographic directions. Change of switching conditions allows to produce the structures oriented along X (Fig. 2a) and Y (Fig. 2b) directions. Similar quasi-regular nano-scale structures appear in LN with waveguides in the area covered by nonpolar surface layer formed by proton exchange (Fig. 3).

5. 2D Domain Structures

Web structures formation has been observed during switching in stoichiometric LT with artificial dielectric layer. Web growth starting from pinhole in dielectric layer leads to development of highly symmetrical ordered domain structure (Fig. 4). It must be pointed that shape of the switched area covered by needle-like domains is similar to the hexagonal shape of individual domain during regular growth in SLT. The structure is highly organized

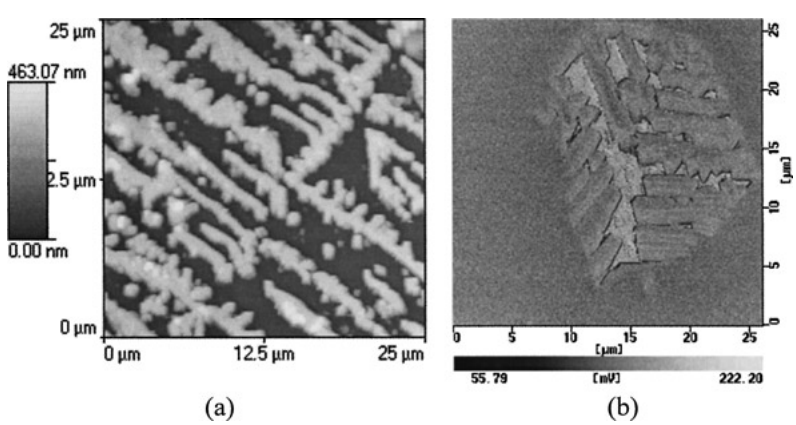

FIGURE 5 Dendrite domain structures in MgO:CLN visualized by SPM (a) in contact AFM and (b) PRIM.

(Fig. 4b) with average distance between neighboring domains 1.5 microns (Fig. 4c). This value is very close to the thickness of used dielectric layer. It opens a possibility to control the period of correlated domain structure by using the artificial dielectric surface layer.

Dendrite domain structures with low spatial ordering appear in CLN and MgO doped LN after removing of external field near the edges of the switched area covered by air gap or silicon oil layer (Fig. 5a). PRIM observation of the structure allows to claim that it consists of shallow domains (Fig. 5b). These dendrites propagate through the sample due to growth of domain array branches and may cover the areas about square millimeters. The orientation of individual branches sometimes essentially deviates from the crystallographic directions in rather free manner in contrast with nanodomain arrays.

6. Conclusion

The formation of quasi-regular patterns by self-maintained domain evolution has been investigated in LN and LT single crystals. It was supposed that the structure period is determined by the thickness of the dielectric layer and can be controlled while using the artificial insulating layers. The revealed regularity of formation and growth of 1D and 2D structures allows to suppose the new methods for creation of the submicron and nano-scale domain patterns in LN and LT for production of photonic crystals.

Acknowledgments

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