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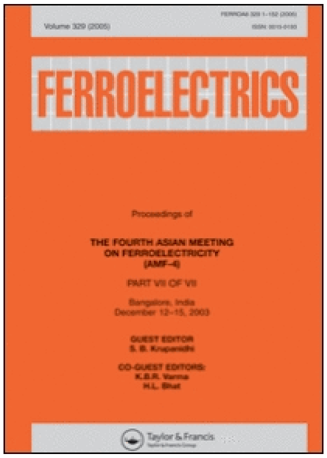

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Physical basis of the domain engineering in the bulk ferroelectrics

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Physical Basis of the Domain Engineering in the Bulk Ferroelectrics

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We present a review of our theoretical and experimental investigations of the domain engineering aspects of domain structure development. Particular attention is paid to the processes governing the fabrication of periodic domain patterns.

Keywords: domain engineering; kinetics; domain structure; periodical domain patterns; screening; depolarization field; switching

INTRODUCTION

Recently a new branch of ferroelectric technology and science named **domain engineering** has been developing rapidly. The target of this activity is to find ways of producing desired domain structures with precise reproducibility of the main parameters in commercially available ferroelectric materials. Recently the particular attention is paid to periodically poled ferroelectrics (lithium niobate[1], lithium tantalate and KTP family crystals[2]) which had a major impact on nonlinear optical frequency conversion. Currently commercial vendors offer periods greater then 6 microns which are acceptable for mid-infrared and green light applications. However the shorter periods necessary for blue light and near ultraviolet applications have only been demonstrated in certain laboratories and appear to be limited to around 4÷5 μm for

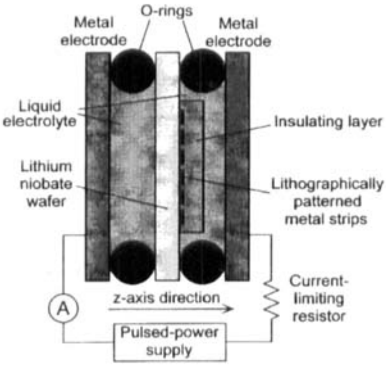
0.5 mm thick devices. Detailed study of the physical nature of nucleation, evolution and stabilization of the domain patterns is required to solve this problem.

It was shown that all existing domain patterns obtained during switching in external field are of kinetic nature[3]. The domain evolution is determined by the spatial distribution of local field produced by various charged systems with wide range of the relaxation times. The knowledge of kinetics of screening processes and sources of the fields driving the domain evolution is the foundation for the progress in domain engineering.

RECENT ACHIEVEMENTS IN BULK DOMAIN PATTERNING

Lithium niobate $LiNbO_3$ has favorable characteristics for development as quasi-phase-matched material. The successful control of domain inversion in the bulk via an applied electric field at room temperature by Yamada et. al.[4] initiated the intensive development of this promising approach.

The periodic bulk domain structure was prepared in $LiNbO_3$ wafers of congruent composition. The single domain 0.2 - 0.5 mm thick samples were photolithographically patterned with periodic metal-electrode structure deposited on $\mathbf{z}^+$ only[4,5]. The electrodes were overcoated with a thin insulating layer to inhibit growth of the domains between these electrodes (Fig.1).



FUGURE 1 Scheme of the experimental setup for poling.

High voltage pulses were applied producing the electric fields greater than threshold value (E_{th} ~ 21 kV/mm)[4,5]. The parameters of the domain structure were controlled by the pulse shape and switching current. After poling the polar surfaces and cross-sections were etched and the domain patterns on z^+, z^- and y surfaces were visualized by optical microscope in reflection mode.

Recently the single pulse poling ("switching") with current limitation was used (Fig.2a).[5,6] The "stabilization" stage of the pulse was added to prevent the spontaneous domain reversion to the original configuration (so-called "backswitching process") after removing of the poling field.

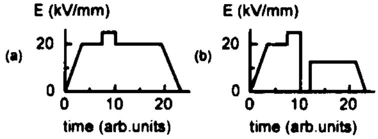


FIGURE 2 Poling voltage waveforms for (a) conventional poling, (b) backswitching one.

This method allows the fabrication of nonlinear device chips with high yield and reproducible characteristics in 0.5 mm thick by 50 mm long samples based on processing of 3 inch diameter $LiNbO_3$ wafers. The obtained domain periods of 10 to 30 microns are suitable for infrared devices[1,6,7]. Recently the optimization of this method makes it possible to achieve the periods up to 6.5 microns with duty cycles about 0.3[5].

CURRENT PROBLEMS

The main problems are connected with reproducibility of the domain structure parameters, especially for short periods and for thick samples. The nucleation density defining the period limit usually is less then required. The typical domain pattern on z^+ shows the significant shift of the domain walls out of electroded area which is difficult to control. Moreover this shift can lead to merging of domains on z^+. The pattern at z^- always differs to some extent with that on z^+ and for a short period electrode structure the periodic domain patterns on z^- are not obtained at all.

MAIN APPROACH

Our approach to these remaining problems in domain patterning in the bulk is based on the assumption of key role of bulk screening effects. It is well-known that polarization reversal from the single domain state is achieved by nucleated of new domains and their growth[6]. Both processes are governed by the elementary nucleation in the bulk ("nucleation" of the domains) and at the domain wall (domain growth)[3,8]. The local electric field E_s averaged over the volume of nucleus is the driving force of this process[3,9]. The local field E_s (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 – the external screening field E_{escr}(r,t) and in the bulk – the bulk screening field E_{bscr}(r,t)[3,10,11].

$$E_s(r,t) = E_{ex}(r) - [E_{dep}(r,t) - E_{escr}(r,t) - E_{bscr}(r,t)] \qquad (1)$$

In the case of strip electrodes the external field is essentially spatially nonuniform. The depolarization field slows the domain growth while the screening process reduces its influence. After complete external screening the bulk residual depolarization field E_{dr} remains due to the surface dielectric gaps of the thickness L[10,11].

$$E_{dr} = E_{dep} - E_{escr} = 2L\, P_S\, (\varepsilon_L \varepsilon_o d)^{-1} \qquad (2)$$

where P_S - spontaneous polarization, ε_L - dielectric permittivity of the gap.

This residual field can be screened only by the charge redistribution in the bulk and/or aligning of polar defects[10,11]. After removing the external field, the spontaneous backswitching process will be obtained if

$$E_s(r,t) = - [E_{dep}(r,t) - E_{escr}(r,t) - E_{bscr}(r,t)] > E_{th} \qquad (3)$$

Thus the initial single domain state can be even completely reconstructed. Manipulation of these screening effects can be used to enhance the fidelity of fine-pitch domain structures.

THE STAGES OF DOMAIN EVOLUTION

Let us consider the domain kinetics during switching from the single domain state in a spatially inhomogeneous field. The whole patterning process in $LiNbO_3$, as in any ferroelectric single crystal during switching from the single domain state[8], can be divided into main stages: 1) nucleation of new domains at the surface (Fig.3a), 2) forward growth in polar direction and enlargement of the nucleated domains and their coalescence(Fig.3b), 3) plane domain wall motion (Fig.3c), and 4) stabilization of the domain structure.

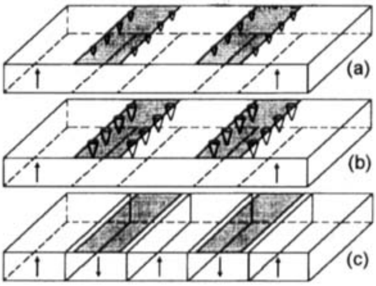
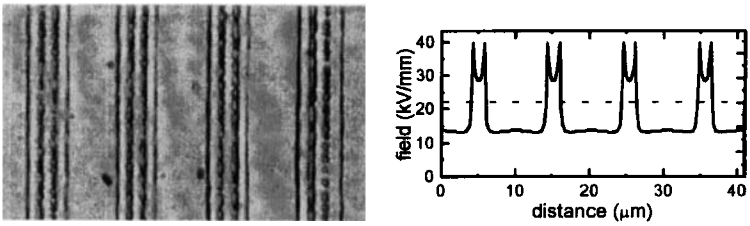


FIGURE 3 The main stages of domain evolution during switching in single domain plate with stripe electrodes.

Arising of new domains at the surface

It was shown[5,6] that the density of spike-like domain nuclei averaged over the electroded area depends on the electrode material and preparation methods. At the same time experiments show the dramatic increase (by the orders of magnitude) of the nucleation density near the edges of electrodes (Fig. 4a). This behavior is due to the known singular spatial distribution of the polar component of switching field E_z at the surface near the edges of finite electrodes (Fig. 4b). So the required nucleation density can be obtained by improving the technology of optimal electrode edges. The experimentally observed patterns of the first stage of poling and analysis of the switching current data show pronounced correlation in the nuclei distribution along the edges of the electrode (Fig. 4). This domain-domain interaction appears to be due to suppression of the local field in the vicinity of the domain wall.



(a) (b)

FIGURE 4 (a) - Nucleation near the edges of the electrodes (during backswitching) and (b) - calculated spatial distribution of the field polar component E_z at the surface.

Forward growth and enlargement of the nucleated domains

The enlargement of the spike-like domains is achieved through the forward and sideways domain wall motion. The forward growth velocity v_f is always much higher (about 100 times) then the sideways one v_s and is dictated by the value of local electric field. In the case of simultaneous growth of the great number of spikes the value of $E_S(r,t)$ depends on the distances between the neighboring domains. This interaction between the domain tips during the forward growth in real systems (with the pinning on the defects) is the cause of the difference between observed z^+ and z^- domain patterns.

The growing domains in $LiNbO_3$ are of hexagonal shape. This situation is similar to regular crystal growth under small oversaturation[14,15]. In analogy the domain kinetics can be explained by layer-by-layer growth through 1D nucleation (Fig. 5).

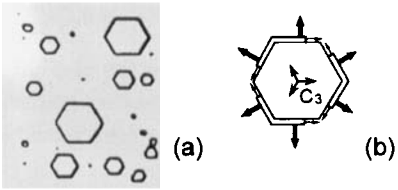


FIGURE 5 Layer-by-layer growth in LiNbO3: (a) – the pattern, (b) – the scheme of domain growth (thin arrows – the directions of step motion; thick arrows – the domain wall motion).

In $LiNbO_3$ the trigonal anisotropy of the surface energy must lead to the preferable motion of the steps on the domain walls in three directions. In this case six plane domain walls are formed (Fig. 5). The prevalence of layer-by-layer growth after coalescence leads to the formation and growth of the lamellar domains (Fig. 3).

Plane domain wall motion

The plane domain wall motion under the nonelectroded areas is nonuniform because of pinning effect due to defects and domain-domain interactions. In addition the effect of correlated nucleation in the vicinity of these walls[16] is very pronounced in $LiNbO_3$. The combined action of all these mechanisms leads to observed variations of wall shape and local duty cycle. The inhibition of external screening of depolarization field in between electrodes by optimization of the insulating layer will prevent the undesirable sideways motion.

Stabilization of the domain structure

The stabilization of obtained domain structure in the conventional approach was achieved by suppression of the backswitching process. It was demonstrated experimentally that the stabilization step duration more then 50 ms is enough to effectively block the backswitching process[6]. Under the assumption that the depolarization field is the driving force of backswitching this time must be of the order of the time constant of screening[3,11].

Backswitching

In contrast to the conventional approach to suppressing backswitching we are investigating to use of the backswitching effect for domain patterning. This radical change of approach to the problem is based on ideas experimentally confirmed in earlier works[3,17]. Rapid removal of the external field leads to backswitching process. In this case the nucleation of new domains starts under the edges of the electrodes. The domain evolution is similar to the conventional switching approach but the produced domain structure is more regular.

The backswitching method opens new possibilities in domain patterning. First, frequency multiplication of the domain pattern compared to the electrode pattern can be obtained by modification of the voltage waveform (Fig. 2b). The backswitching takes place during the zero-applied-field stage.

As a result, additional backswitched domains appear under the electrodes on **z**$^+$ (Fig. 6b). In this case these domains often do not go through to **z**$^-$. Their length depends on the pulse parameters and typically is about 50-100 μm (Fig. 6a). Moreover we have demonstrated that a second stage of frequency multiplication can be obtained in this way (Fig. 6d). In this case the additional domain stripes arise just under the edges of the electrodes. Their length is about 20 - 50 μm (Fig. 6c).

Detailed analysis of the individual domain behavior during backswitching reveals two distinct variants of their evolution: erasing and splitting. The erasing process leads to formation of the backswitched domains in earlier switched area without any variation of the external shape of switched domain (Fig. 7a). During splitting the growing backswitched domain cuts the initial switched domain conserving its volume (Fig. 7b).

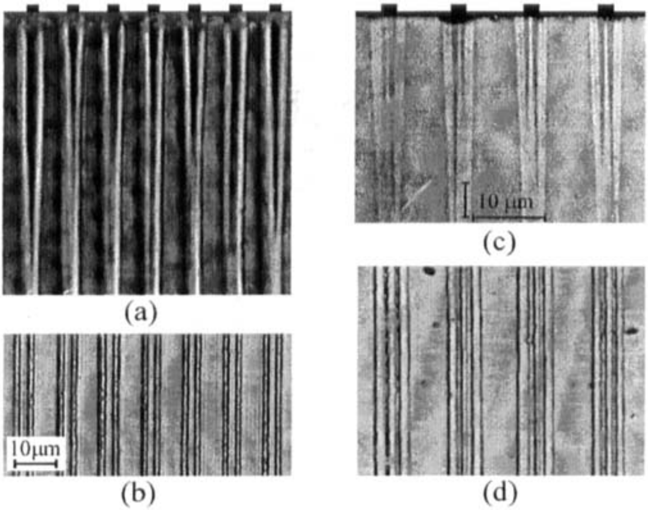


FIGURE 6 Single frequency multiplication: (a) cross section (y surface) and (b) z$^+$ view. Double frequency multiplication: (c) cross section (y surface) and (d) z$^+$ view.

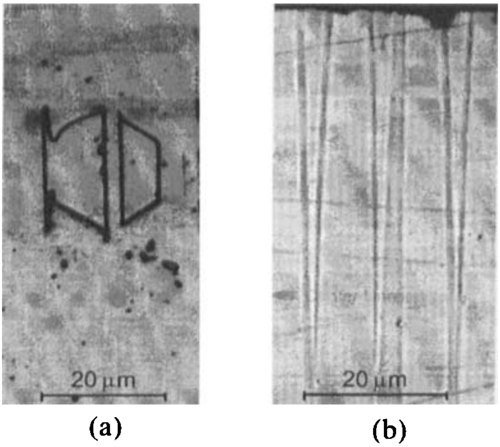


FIGURE 7 Domain changing by backswitching: (a) erasing (z^+ view) and (b) splitting (y surface).

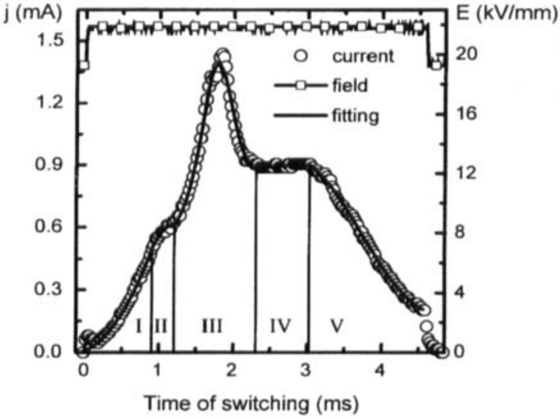


FIGURE 8 Switching current during poling (without current limit). Experimental points fitted by theoretical equations.

Switching current data

It is clear that the obtained domain patterns are defined by all stages of domain evolution. But the information about domain kinetics can not be obtained by conventional methods. We have shown that mathematical treatment of the switching current data allows separation and study of the stages of domain evolution during patterning[18,19].Using this approach for poling in $LiNbO_3$ (Fig. 8) the switching current can be divided into five parts corresponding to the stages of the domain development (the details of these analyses will be published elsewhere). It allows determination of the domain kinetics for domain engineering: time of forward growth, sideways motion velocity and nucleation density. Control of these parameters is critical for optimization of the domain patterning.

CONCLUSION

We have analyzed the main stages of domain patterning important for domain engineering. The domain evolution during spontaneous backswitching after removing of the poling field was investigated for the first time. The important role of backswitching as a powerful tool for producing short domain patterns has been demonstrated.

Acknowledgments

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