Domain kinetics in the formation of a periodic domain structure in lithium niobate

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The evolution of the domain structure in $LiNbO_3$ with polarization switching in an electric field is investigated experimentally. Special attention is given to the formation processes of a regular domain applicable to nonlinear optical devices. A new method based on the spontaneous backswitching effect is proposed for creating a regular structure with a period of 2.6 μm in $LiNbO_3$ with a thickness of 0.5 mm. © *1999 American Institute of Physics.* [S1063-7834(99)02410-7]

The formation of a periodic domain structure having micron periods in ferroelectric materials, which are useful in applications, poses a problem whose solution is particularly crucial in order to establish synchronism conditions in nonlinear optical devices.[1] Second harmonic generation in the visible and ultraviolet (UV) ranges require regular domain structures with a period less than 5 μm in substrates having a thickness of at least 0.5 mm. The feasibility of creating a regular domain structure in bulk $LiNbO_3$ placed in an electric field at room temperature was first demonstrated in 1993.[3] This technique has grown in popularity by virtue of its reproducibility and applicability to other nonlinear optical crystals. Several papers have reported applications of this technique to create a domain structure with a period of 3 -4 μm in substrates of thickness $200-300$ μm in $LiNbO_3$ (Ref. 2), $LiTaO_3$ (Refs. 3 and 4), and $KTiOPO_4$ (KTP) (Ref. 5) with the capability of generating blue and UV light. In thicker substrates, however, it is impossible to obtain structures capable of satisfying the quasi-phase synchronism condition for this spectral range. It is essential to note that the domain kinetics in $LiNbO_3$ has been investigated only for very slow switchings,[6] even though it is obvious that a systematic investigation of the stages of evolution of the domain structure in the presence of switching with characteristic times less than one second has particularly important bearing on the refinement of existing methods.

Here we report a detailed investigation of the creation and pinning of a domain structure in bulk $LiNbO_3$ samples in the presence of an electric field applied to periodic stripe electrodes. Visualization of the domain configurations by chemical etching and subsequent examination of the relief with an optical microscope and a scanning electron microscope (SEM) have been used to analyze the evolution stages of the domain structure. Special attention has been devoted to studying the domain structure kinetics in the presence of spontaneous backswitching after the field is turned off.

1. EXPERIMENTAL

The investigated samples were monodomain $LiNbO_3$ wafers of thickness 0.5 mm cut perpendicularly to the polar axis from congruent-grown, optical-quality single crystals. Periodic metal stripe electrodes (NiCr) oriented along the $[10\bar{1}0]$ direction were photolithographically deposited on the (0001) polar plane. To prevent the domains from growing beyond the limits of the electrodes, the electrode-implanted surface was coated with a thin insulator layer (Fig. 1a). The voltage pulses used to generate an above-threshold electric field (21.5 kV/mm) in the bulk of the samples were applied by means of a clamp holder (Fig. 1a) through a liquid electrolyte (LiCl solution).[7,8] Switching was performed at room temperature. The current and voltage pulses were recorded as the domain structure took shape. The parameters of the domain structure could be controlled by varying the shape and duration of the voltage pulse and the current amplitude (Figs. 1b and 1c). To analyze the domain structure obtained after partial or complete switching, the polar planes and cross sections were etched for 5–10 min in hydrofluoric acid at room temperature. The surface relief was observed by the optical and scanning electron microscopes. The preparation of tilted thin sections significantly improved the spatial resolution for examination of the domain structure in the bulk. Detailed information on the evolution of the domains during switching was obtained by comparing the domain images formed after different lengths of the switching pulses.

2. EVOLUTION STAGES OF THE DOMAIN STRUCTURE

An analysis of the domain configurations obtained after partial switchings has revealed several distinct stages in the evolution of the domains[9] (Fig. 2). The switching process begins with ''nucleation'' (the inception of new domains) on the (0001) polar surface along the edges of the electrodes (Fig. 2a) when the field attains a threshold level.[10] The second stage is characterized by growth of the domains in the polar and sidewise directions and their coalescence under the

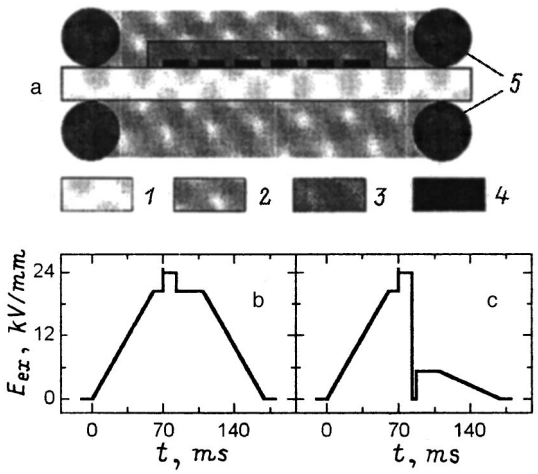

FIG. 1. a) Electrode configuration for the creation of a periodic domain structure: *1*) $LiNbO_3$ wafer; *2*) liquid electrolyte; *3*) insulator layer; *4*) periodic stripe electrodes; *5*) annular spacers; b) Shape of the switching voltage pulse in the conventional method; c) pulse shape with backswitching.

electrodes (Fig. 2b). Upon completion of this stage, laminar domains are formed all the way across the sample (Fig. 2c). In the third stage the planar boundaries of the newly formed domains move beyond the limits of the electrodes (Fig. 2d).

After a rapid decrease in the switching field the switching process stops and, depending on the pulse duration, two scenarios of domain evolution are possible: stabilization of the newly formed domain structure or partial switching of the domains back to the original stage (reverse poling or ''backswitching'').[7,11–13] In backswitching the boundaries of the switched domains move toward the electrodes; in addition, chains of wedge-shaped domains form along the edges of the electrodes (Fig. 2e).

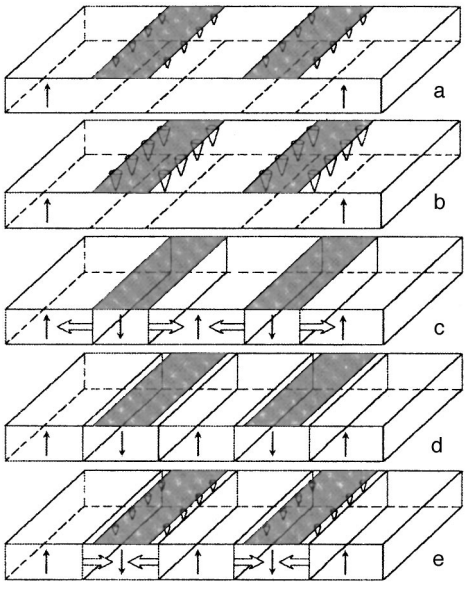

FIG. 2. Main stages of domain evolution in the switching of a monodomain wafer with stripe electrodes on (0001).

3. BASIC APPROACH

Our approach to analyzing the domain structure evolution in ferroelectrics is based on the assumption that screening effects play key role.[14,15] Switching from the single-domain state is known to be induced by the formation and growth of domains of opposite sign. The rates of formation and growth of the domains are governed by the polar component of the local electric field E_z (Refs. 10 and 15) at the nucleation sites and at the domain boundaries, respectively. The spatial distribution of the local field $E_z(r,t)$ is determined by the sum of the *z*-components of fields of various origins: 1) the external field $E_{ex}(r)$ produced by the voltage applied to the electrodes; 2) the depolarizing field $E_{dep}(r,t)$ produced by bound charges of the instantaneous domain configuration; 3) two types of screening fields, which compensate the depolarizing field: one external $E_{escr}(r,t)$, induced by charge redistribution on the electrodes, and the other internal $E_{bscr}(r,t)$, generated by various bulk mechanisms[14,16,17]:

$$E_z(r,t)=E_{ex}(r)-[E_{dep}(r,t)-E_{escr}(r,t)-E_{bscr}(r,t)]. \tag{1}$$

The depolarizing field slows domain growth, but its influence is diminished by the screening effects. As a rule, the external screening time constant is much shorter than the bulk constant and is governed by the parameters of the external circuit. Bulk screening processes are very significant, because even after the cessation of external screening a residual depolarizing field $E_{dr}(r)$ (of the same order as the coercive field) still exists in the bulk, owing to the presence of a surface dielectric gap in ferroelectrics[10,11]:

$$E_{dr}(r)=E_{dep}(r)-E_{escr}(r)\approx 0. \tag{2}$$

For an infinite, monodomain ferroelectric capacitor we have

$$E_{dr}(r)=2L/d(P_S/\varepsilon_L\varepsilon_0), \tag{3}$$

where L is the thickness of the dielectric gap, d is the thickness of the sample, P_S is the spontaneous polarization, and ε_L is the dielectric permittivity of the gap.

The residual depolarizing field can be screened as a result of charge redistribution in the bulk and the orientation of charged dipole defects.[16,17] These processes are comparatively slow ($\tau\sim 10^{-1}-10^{5}$ s) (Ref. 10), so that the usually observed bulk screening delay produces various memory phenomena.[15] For example, the initial domain state can be partially or completely reconstructed after a sufficiently rapid reduction of the external field (spontaneous backswitching). This process is induced by the partially screened residual depolarizing field:

$$E_{bs}(r,t)=-[E_{dr}(r)-E_{bscr}(r,t)]. \tag{4}$$

We have shown previously that the backswitching effect can be used to create periodic domain structures having exceptionally short periods.[9]

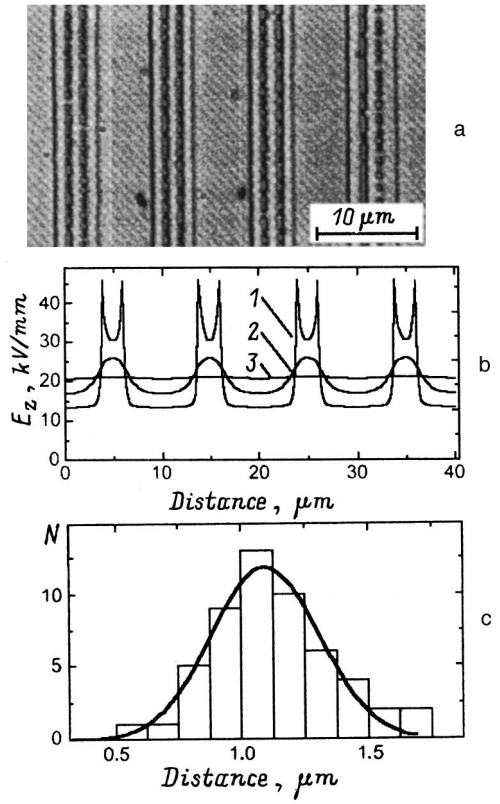

FIG. 3. a) Formation of nucleation centers near the edges of the electrodes (in backswitching); b) theoretically calculated spatial distribution of the polar field component $E_z(x)$ at various distances from the surface Δz (in fraction of the electrode period): *1*) 0.01; *2*) 0.1; *3*) 0.6; c) histogram of the distances between neighboring wedge domains and its approximation by a Gaussian curve.

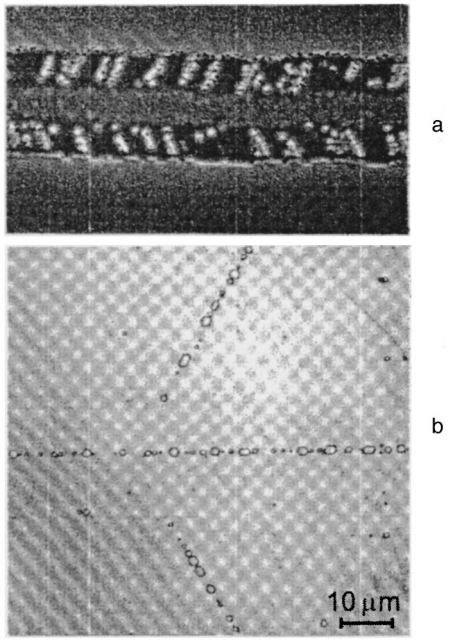

FIG. 4. a) Nanodomains formed near the edge of an electrode in backswitching; b) growth of domain chains on the $(000\bar{1})$ surface in the preferred growth directions with switching in a homogeneous field.

4. DETAILS OF DOMAIN STRUCTURE EVOLUTION

4.1. Formation of domains on the surface

The density of nucleation centers is customarily assumed to be an important parameter, limiting the spatial frequency of periodic domain structures.[8,12] It has been established experimentally that the average density of nucleation centers in $LiNbO_3$ depends weakly on the electrode material.[12,18] Our experiments have shown that the density of newly formed, isolated wedge domains exhibits strong spatial inhomogeneity: Far from the edges of the electrodes it does not exceed $1000\,mm^{-2}$ (Ref. 8), but along the edges of the electrodes the linear density of newly formed domains attains 1100 mm^{-1} (Fig. 3a).[9]

This behavior can be attributed to the singular spatial distribution of the polar component of the local field E_z near the surface at the edges of the electrodes. It should be noted that spatial inhomogeneity of the field $E_z(x)$ exists only near the surface, and its amplitude decays rapidly with increasing depth. The field is essentially homogeneous to a depth of the order of the electrode period (Fig. 3b). Spatially inhomogeneous nucleation is attributable therefore to the inhomogeneity of the field in a thin surface layer, and the growth of the domains in the bulk takes place in a homogeneous field.

Our detailed investigations of the initial stage of spontaneous backswitching with SEM imaging of the domain configuration on the polar plane has revealed the existence of chains of nanodomains (Fig. 4a). These domains typically have diameters of 50–100 nm at a linear density up to $10^4\,mm^{-1}$. We assume that the usually observed wedge domains are formed as a result of the growth of these nanodomains. It is important to note that isolated needle-shaped microdomains of unknown origin with diameters less than 1 μ have been observed some time ago in $LiNbO_3$ (Ref. 19).

An analysis of the domain configurations in the first stage of backswitching shows that a correlation is observed in the spatial distribution of the wedge domains (Fig. 3c). This characteristic can be identified with a decrease in the local field near a newly formed wedge domain.[10] The modification of the spatial distribution of the electric field suppresses the growth of neighboring nucleation centers. This phenomenon imparts a quasi-regularity to the growing wedge domains, and the number of such domains is considerably smaller than the number of nanodomains.

4.2. Growth and coalescence of wedge domains under the electrodes

The newly formed wedge domains continue to grow as a result of the motion of the domain boundaries in the forward (polar) and sidewise directions. An analysis of the static domain configurations observed on tilted thin sections has shown that the rate of forward growth v_f in $LiNbO_3$ is approximately 100 times the rate of sidewise motion v_s. The velocity ratio dictates the observed vertex angles of the wedge domains, which are less than one degree.

In the case of simultaneous growth of a system of wedges, the local field $E_z(r,t)$ at the vertex of a given wedge depends on the distances to its neighbors. Consequently, if a neighbor lingers or stops altogether (as a result of being slowed down by defects), the local field near the vertex of

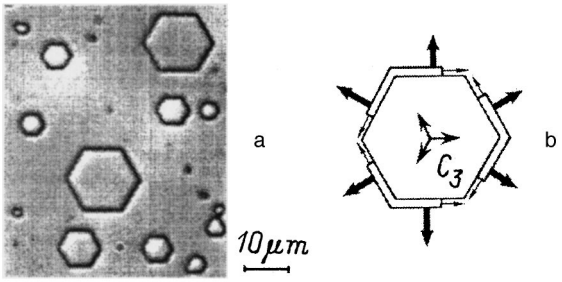

FIG. 5. Hexagonal domains in $LiNbO_3$: a) domain configuration; b) diagram of laminar domain growth. The light arrows indicate the direction of motion of the steps, and the heavy arrows indicate the direction of motion of the domain boundaries.

the moving wedge changes, causing the direction of growth to deviate. This phenomenon should create a difference between the domain configurations on different polar surfaces, as is indeed often observed in experiment.

An analysis of the domain configurations in the first stage of backswitching has shown that the growing domains in $LiNbO_3$ usually have a hexagonal shape (Fig. 5a). To explain the growth of domains having a regular geometry, we exploit the previously mentioned similarity of crystal growth and domain growth.[20,21] Domain growth (sidewise motion of the domain boundaries) is induced by the motion of individual steps along the domain boundaries (Fig. 5b). The trigonal symmetry of $LiNbO_3$ in the plane perpendicular to the polar axis causes the steps to move in the three $[10\bar{1}0]$ directions. Six planar domain boundaries are formed as a result of this growth pattern (Fig. 5b).[22] It is important to note that anisotropy is also encountered in the formation of chains of wedge domains oriented in the three $[10\bar{1}0]$ directions during switching in a homogeneous field (Fig. 4b).

The stripe electrodes are always oriented along one of the preferred growth directions (motion of the steps). Owing to this orientation, laminar growth prevails after coalescence of the chains of domains formed along the edges of the electrodes in the first switching stage, resulting in the formation of pairs of stripe domains having planar boundaries on the (0001) surface. In thick samples the coalescence of isolated domains and the formation of stripes on (0001) terminates before the vertices of the wedges grow all the way across. It has been shown experimentally that for a distance of 0.9 μm between nucleation centers the coalescence of isolated domains terminates when their vertices grow in the polar direction to depths of $50-100\,\mu$m.

The pairs of domain boundaries formed under the edges of the electrodes move opposite to one another until complete switching has taken place under the electrodes on (0001). For samples having a thickness of 0.5 mm and electrodes less than 3 μm wide this coalescence terminates before the vertices of the wedges grow clear across the sample.

Laminar through domains having a regular configuration do not form until the completion of forward growth. However, the evolution of the domain structure does not always end with this event. Pronounced unwanted broadening of the laminar domains beyond the limits of the electrodes is always observed in the final stage.

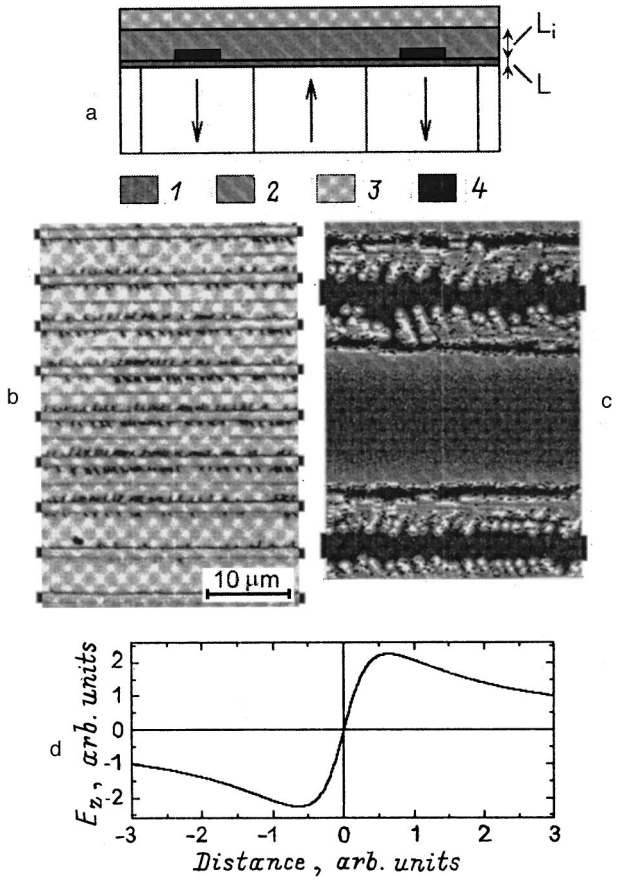

FIG. 6. a) Diagram of the surface zone of the wafer with electrodes: *1*) dielectric gap; *2*) insulator layer; *3*) liquid electrolyte; *4*) metal electrodes; b, c) domain configurations with anomalous broadening of the domains beyond the limits of the electrodes, viewed by the optical microscope (b) and SEM (c); d) theoretically calculated spatial distribution of the polar component of the local field near the surface in the vicinity of a planar domain boundary.

4.3. Motion of domain boundaries beyond the electrodes

It is obvious that, when periodic stripe electrodes are used, spatial inhomogeneity of the external field E_{ex} occurs only in a shallow surface layer having a thickness of the order of the period of the electrode structure Λ, so that the motion of the newly formed planar domain boundaries beyond the limits of the electrodes in thick samples ($\Lambda \ll d$) takes place in an essentially homogeneous electric field.

The rate of motion of a domain boundary beyond the electrodes is determined by the polar component of the local electric field E_z (Ref. 23). The degree of compensation of the depolarizing field by external screening (redistribution of charges in the liquid electrodes) is substantially lower than under the metal electrodes (Fig. 6a). The presence of a strong residual depolarizing field and its comparatively slow bulk screening in $LiNbO_3$ at room temperature cause E_z to diminish when the boundary moves a distance Δx beyond the limits of an electrode, owing to the increase in the uncompensated fraction of the depolarizing field. The domain boundary stops when

$$E_z(\Delta x) - E_{th} \approx 0, \tag{5}$$

where E_{th} is the threshold field for sidewise movement of a planar domain boundary.

The decrease in E_z during movement of the boundary is caused by increases in $E_{dr}(\Delta x)$ and the field generated by ''attached'' charges formed during screening of the initial monodomain state, because charge redistribution can be ignored for rapid motion of the boundary. Disregarding the conductivity of the insulator layer and allowing for the fact that the thickness of the insulator layer L_i is much greater than the thickness of the dielectric gap L, for our experimental geometry (Fig. 6a) we can find the dependence on Δx of the average field in the bulk E_z if we approximate the spatial distribution of bound and screening charges by a strip of width Δx with charge density σ:

$$E_z(\Delta x) = E_{ex} - E_{dep}(\Delta x)$$
$$= U/d - \sigma(\varepsilon_i \varepsilon_0)^{-1} F(\Delta x/d), \quad (6)$$

where U is the applied voltage, ε_i is the dielectric permittivity of the insulator, $F(\Delta x/d) = 1/\pi[2\arctan(\Delta x/d) + \Delta x/d \ln(1+d^2/\Delta x^2)]$, and $\sigma = LP_S d^{-1} \varepsilon_b \varepsilon_L^{-1}(1+k)$.

The parameter k allows inclusion of the prehistories for a shift from the fully screened initial state, $k=1$, and after long residence in the biased state, $k=-1$.

The derivation of Eq. (7) and its application to analyzing experimental data of the motion of a planar domain boundary in gadolinium molybdate are described in detail in our previous work.[24]

For the given electrode geometry, Eq. (7) and condition (3) can be used to account for the movement of the domain boundaries to a certain distance beyond the limits of the electrodes and to determine the field dependence of the maximum displacement $\Delta x_{max}(E_{ex} - E_{th})$. It should be noted that the finite conductivity of the imperfect insulator layer leads to a major increase in the displacement of the boundaries. The observed displacement of the domain boundary must depend therefore on the composition of the insulation layers and their deposition technology.

4.4. ''Anomalous motion'' of the domain boundaries

It has been observed experimentally that the displacement Δx_{max} in the formation of domain structures with a small period often changes from one electrode to the next, and extraordinarily large boundary displacements are observed in some regions (Figs. 6b and 6c). An attempt to ascribe this behavior to inhomogeneity of the insulating layers has been thwarted by observations showing that the motion of the domain boundary is ruled by an anomalous mechanism in this case.

To investigate the early stages of evolving of the domains in this case, we have analyzed the domain configurations obtained after very short partial backswitching. This possibility is based on the experimentally confirmed similarity of the positions of the nucleation centers in forward and reverse polings. Observations of the domain structures (etched surface relief) in the optical microscope shows that the domain kinetics in anomalous evolution consists in the formation, growth, and eventual coalescence of chains of domain ''fingers'' oriented in one of the $[10\bar{1}0]$ directions (Fig. 6b). Observations with increased spatial resolution (in the SEM) show that the optically observed domain fingers comprise strings of wedge domains approximately 10 nm in diameter (Fig. 6c; cf. Fig. 4b). The linear density of nucleation centers in the strings is greater than $10^4\ mm^{-1}$. In the formation of small-period domain structures the anomalous mechanism can lead to merging of the boundaries between the electrodes and break up the periodicity of the domain structure.

The anomalous mechanism of domain boundary motion can be analyzed by analogy with the ''correlated nucleation'' effect, which we have observed previously in lead germanate.[25,26] The effect is attributable to the long-range influence of a moving domain boundary. A calculation of $\Delta E_z(x)$ near a planar domain boundary (Fig. 6d) reveals a pronounced maximum at a distance of the order of the thickness of the surface dielectric gap.[10] In $LiNbO_3$ this distribution results in the formation of strings of wedge nanodomains in the surface and layer and their subsequent growth in preferred directions (Fig. 6c). The coalescence of the strings produces an inhomogeneous, anomalously large displacement of the boundaries and modifies their shape.

4.5. Evolution of domain structure in backswitching

In a departure from the conventional approach we have demonstrated the possibility of using spontaneous backswitching to create periodic domain structures. This nontraditional approach is based on notions that have been confirmed experimentally in our previous studies.[10,13,15]

The shape of the voltage pulse is changed for polarization utilizing backswitching (Fig. 1c). Under the influence of the first part of the pulse (strong field) laminar domains are formed with a width exceeding that of the electrodes. In the switched region during the stage of a weak applied field, the domain boundaries begin to move in the reverse direction (backswitching). Moreover, as in ordinary switching, wedge-shaped domains form and grow along the edges of the electrodes (Figs. 2 and 3a). The domain structure obtained through backswitching is pinned during the stage of stabilization of the polarizing voltage pulse, but in a much weaker field than in the conventional technique (Fig. 1c). Domain structures corresponding to different degrees of backswitching can be pinned by varying the duration of the weak-field stage. The monitored reverse movement of planar boundaries makes it possible to obtain domain structures clear through the sample with unprecedented short periods in thick samples by virtue of the compensation of unwanted broadening of the domains in the switching stage.

The backswitching method has enabled us to create a domain structure with a period of 2.6 μm in $LiNbO_3$ having a thickness of 0.5 mm (Fig. 7). It should be noted that the conventional procedure is incapable of yielding structures of this kind in such thick substrates.

4.6. Stabilization of the domain structure after switching

In conventional switching, the domain structure crated in the electric field is pinned by fairly prolonged exposure to a field somewhat below threshold (Fig. 1b). Measurements of

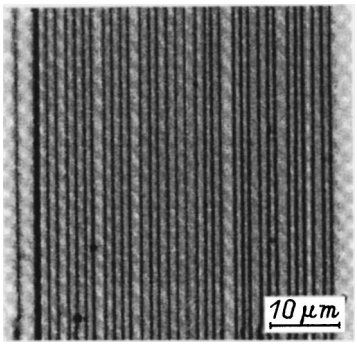

FIG. 7. Domain structure with a period of 2.6 μm obtained with backswitching in a $LiNbO_3$ wafer having a thickness of 0.5 mm; $(000\bar{1})$ surface.

the backswitching fraction (ratio of the reverse and forward poling charges, which are determined by integrating the currents) as a function of the stabilization time, $Q_{bs}(t)/Q_s$, indicate that it decreases exponentially with a time constant of 10–30 ms (Fig. 8). If backswitching is assumed to be driven by the partially screened depolarizing field, the stabilization kinetics is determined by the bulk switching time constant.[13,15,16] It is evident that a stabilization phase lasting more than 50 ms is essentially sufficient for completely blocking backswitching after the field has been turned off.[12]

4.7. Frequency multiplication

The results of our investigation of the domain kinetics associated with backswitching has enabled us to develop qualitatively new methods for the formation of regular structures with multiplication of the spatial frequency of the domain structure in contrast to the structure of the electrodes. We have achieved domain structure frequency doubling through the formation of bands of additional nonthrough domains under the electrodes on (0001) in the presence of backswitching (Fig. 9a). In the polar direction the length of the domains is usually $50-100$ μm and depends on the parameters of the pulse and the width of the electrodes (Fig. 9b). Frequency tripling can be achieved by changing the switching conditions (Fig. 9c). In this case the additional stripe domains are formed under the edges of the electrodes and grow to a depth of $20-50$ μm (Fig. 9d).

A detailed analysis of the domain configurations in the transverse cross section indicates two possible scenarios of domain evolution with backswitching: ''erasure'' and ''splitting.'' The erasure process entails the formation of domains inside a previously switched domain without altering its outward shape (Fig. 9e). In the splitting process a domain growing in the presence of backswitching cleaves a switched domain, significantly altering its shape while approximately preserving its volume (Fig. 9f).

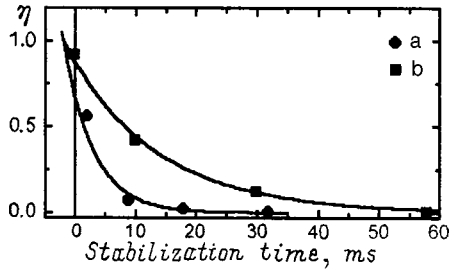

FIG. 8. Time dependence of the backswitching fraction with stabilization in various fields. The experimental points are approximated by exponential functions. a) $E_z=24$ kV/mm, $\tau=11$ ms; b) $E_z=20.6$ kV/mm, $\tau=34$ ms.

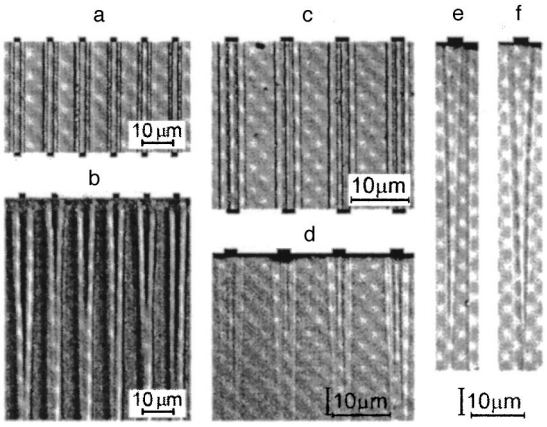

FIG. 9. Spatial-frequency multiplication of the domain structure in the presence of backswitching. Frequency doubling: a) (0001 surface; b) transverse cross section. Frequency tripling: c) (0001) surface; d) transverse cross section. e) Erasure; f) splitting (transverse cross sections).

In summary, the reported comprehensive investigations have yielded new information about the domain structure kinetics in $LiNbO_3$ with rapid polarization switching (with a substantial delay of bulk screening) in a spatially inhomogeneous electric field. We have uncovered the phenomenon of anomalous domain broadening beyond the limits of the electrodes. We have reported the first investigation and qualitative explanation of the specific characteristics of domain structure evolution in the presence of spontaneous backswitching after removal of the external field. The level of insight achieved here has enabled us to exploit this traditionally ''undesirable'' effect as the basis of new methods for the formation of regular domain structures.

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[1] R. L. Byer, J. Nonlinear Opt. Phys. Mater. **6**, 549 (1997).
[2] M. Yamada, N. Nada, M. Saitoh, and K. Watanabe, Appl. Phys. Lett. **62**, 435 (1993).
[3] K. Mizuushi and K. Yamamoto, Opt. Lett. **21**, 107 (1996).
[4] J.-P. Meyn and M. M. Fejer, Opt. Lett. **22**, 1214 (1997).
[5] W. P. Risk and S. D. Lau, Appl. Phys. Lett. **69**, 3999 (1996).

[6] Yu. S. Kuz'minov, *The Electrooptical and Nonlinear Optical Lithium Niobate Crystal* (Nauka, Moscow, 1987).
[7] L. E. Myers, R. C. Eckardt, M. M. Fejer, R. L. Byer, and W. R. Bosenberg, Opt. Lett. **21**, 591 (1996).
[8] G. D. Miller, R. G. Batchko, M. M. Fejer and R. L. Byer, in *SPIE Proceedings*, Vol. 2700: *Solid State Lasers and Nonlinear Crystals* (SPIE, Bellingham, Wash., 1996), p. 34.
[9] V. Ya. Shur, E. L. Rumyantsev, R. G. Batchko, G. D. Miller, M. M. Fejer, and R. L. Byer, Ferroelectrics (in press).
[10] V. Ya. Shur, in *Ferroelectric Thin Films: Synthesis and Basic Properties* (Gordon & Breach, New York, 1996), Chap. 6, p. 153.
[11] V. Ya. Shur and E. L. Rumyantsev, J. Korean Phys. Soc. **32**, S727 (1998).
[12] L. E. Myers, *Quasi-Phasematched Optical Parametric Oscillators in Bulk Periodically Poled Lithium Niobate*, Ph.D. Thesis (Stanford University, 1995).
[13] V. Ya. Shur, V. V. Leguchev, and I. V. Ovechkina, Fiz. Tverd. Tela (Leningrad) **26**, 3474 (1984) [Sov. Phys. Solid State **26**, 2091 (1984)].
[14] V. Ya. Shur, V. V. Leguchev, and N. V. Korovina, Fiz. Tverd. Tela (Leningrad) **26**, 781 (1984) [Sov. Phys. Solid State **26**, 471 (1984)].
[15] V. Ya. Shur and E. L. Rumyantsev, Ferroelectrics **191**, 319 (1997).
[16] P. V. Lambeck and G. H. Jonker, Ferroelectrics **22**, 729 (1978).
[17] V. M. Fridkin, *Ferroelectric Semiconductors* (Nauka, Moscow, 1976).
[18] G. D. Miller, Ph.D. Thesis (Stanford University, 1998).
[19] N. Ohnishi and T. Iisuka, J. Appl. Phys. **46**, 1063 (1975).
[20] V. Ya. Shur and E. L. Rumyantsev, Ferroelectrics **142**, 1 (1993).
[21] W. K. Burton, N. Cabrera, and F. C. Frank, Philos. Trans. R. Soc. London **243**, 299 (1951).
[22] V. Ya. Shur, A. L. Gruverman, V. V. Letuchev, E. L. Rumyantsev, and A. L. Subbotin, Ferroelectrics **98**, 29 (1989).
[23] V. Ya. Shur, A. L. Gruverman, V. P. Kuminov, and N. A. Tonkachyova, Ferroelectrics **111**, 197 (1990).
[24] V. Ya. Shur, E. L. Rumyantsev, V. P. Kuminov, A. L. Subbotin, and E. V. Nikolaeva, Fiz. Tverd. Tela (St. Petersburg) **41**, 126 (1999) [Phys. Solid State **41**, 112 (1999)].
[25] V. Ya. Shur, A. L. Gruverman, N. Yu. Ponomarev, E. L. Rumyantsev, and N. A. Tonkacheva, JETP Lett. **53**, 615 (1991).
[26] V. Ya. Shur, A. L. Gruverman, N. Yu. Ponomarev, and N. A. Tonkachyova, Ferroelectrics **126**, 371 (1992).

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