Ferroelectrics, 2002, Vol. 269, pp. 189-194
Reprints available directly from the publisher
Photocopying permitted by license only

DOMAIN KINETICS IN CONGRUENT AND STOICHIOMETRIC LITHIUM NIOBATE

V.Ya. SHUR[a]*, E.L. RUMYANTSEV[a], E.V. NIKOLAEVA[a],
E.I. SHISHKIN[a], R.G. BATCHKO[b], M.M. FEJER[b],
R.L. BYER[b], I. MNUSHKINA[c]
[a]Inst. Phys.& Appl. Math., Ural State University, Ekaterinburg, Russia;
[b]E.L. Ginzton Laboratory, Stanford University, Stanford, CA;
[c]Deltronic Crystal Industries, Dover, NJ

We present the investigation of domain kinetics in single crystals of congruent, stoichiometric and MgO-doped lithium niobate single crystals during polarization reversal using transparent liquid electrolyte and conductive oxide electrodes. Domain structure kinetics has been *in situ* visualized in transmitted light. Domain evolution significantly depends on electrode type. Jerky reconstruction of domain structure obtained for liquid electrodes while formation of charged domain walls was observed for sputtered electrodes in all investigated materials. Electrical measurements are presented for cyclic switching of MgO doped stoichiometric lithium niobate.

Keywords: domain structure; lithium niobate; single crystals; polarization reversal

INTRODUCTION

Recently the problem of searching for the appropriate material for nonlinear optical devices became very important.[1] This material should display not only necessary nonlinear optical properties and high resis-

* Corresponding author e-mail: vladimir.shur@usu.ru

tance to optical damage, but also easily controlled domain structure. In this work we present the detail investigation of domain kinetics in three closely related materials: 1) congruent lithium niobate (CLN), 2) stoichiometric lithium niobate (SLN) and 3) stoichiometric lithium niobate doped by 1% of magnesium oxide (MgO:SLN). These materials are considered to be among the most versatile. Despite different values of coercive field (21 kV/mm in CLN, 6.6 kV/mm in SLN and 3.5 kV/mm in MgO:SLN) all of them demonstrate similar domain kinetics.

EXPERIMENT

We studied single crystalline commercial CLN (Crystal Technology, CA), SLN and MgO:SLN grown by the top-seeded solution method.[2] Single-domain plates were cut perpendicular to the polar **Z** axis (0.2-mm-thick CLN, 0.5-mm-thick SLN and MgO:SLN, area 6x6 mm^2). Two electrode types have been used: liquid electrolyte (LiCl aqua solution) and transparent sputtered In_2O_3:Sn (ITO), same on both **Z** surfaces. Switching were performed at room temperature. Domain evolution in the **whole switching area** (1-mm-diameter) was *in situ* visualized by polarizing microscope in transmitted mode and TV recorded (25 fps) with subsequent image processing. The switching current was simultaneously measured. Thus a unique possibility to establish the correspondence between switching current and domain kinetics was realized.

SWITCHING WITH LIQUID ELECTRODES

Domain evolution in CLN

In situ observation of the domain kinetics has been used for a detailed study of the domain structure evolution during switching with liquid electrode in CLN (Fig. 1). Switching process always starts with nucleation at the electrode edges and/or at the surface defects in the center. Isolated domains grow conserving hexagonal shape with domain walls strictly oriented along **Y** directions (Fig. 1a). Domain wall motion is usually connected with the microscale domain steps propagation along the wall (Fig. 2). The irregular wall propagation and its interaction with

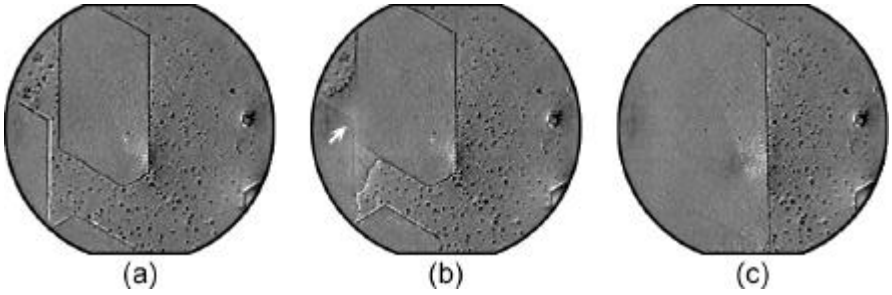

FIGURE 1. Evolution of domain structure during switching in CLN. Time interval from field switch-on: (a) 1.5 s, (b) 2.3 s and (c) 3.3 s. Arrow indicates the domain wall print. Liquid electrodes. E = 22 kV/mm. 1-mm-diameter area.

individual arisen domains (Fig. 1b) are observed. The preferential wall orientation is clearly demonstrated from the beginning to the end of the switching. Each formation of concave shape domains after domain coalescence leads to transformation to regular convex shape by super-fast wall motion. Thus any local deviation from the allowed crystallographic directions disappear rapidly (Fig. 1c). As a result the domain kinetics becomes very jerky.

After the domain wall shift a "wall print" is observed at the previous wall rest position (place of wall staying for about 0.1 second) (Fig. 1b). Similar behavior has been obtained by Gopalan [3] during wall motion in congruent lithium tantalate. All prints disappear gradually. Print lifetime is proportional to the wall rest time. Observation of prints can be explained by electrooptic effect caused by electric field produced by screening charges, which accumulate in the bulk while domain wall stay in fixed position.

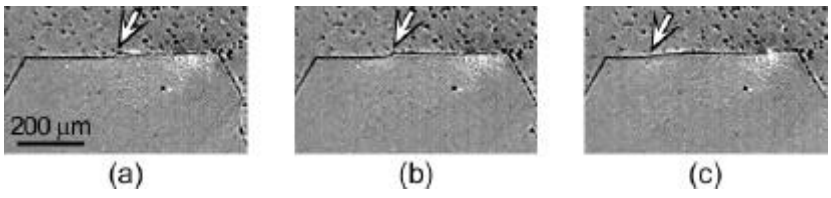

FIGURE 2. Layer-by-layer domain growth by step propagation along the wall in CLN. Time interval from field switch-on: (a) 1.20 s, (b) 1.28 s and (c) 1.36 s. Liquid electrodes. E = 22 kV/mm.

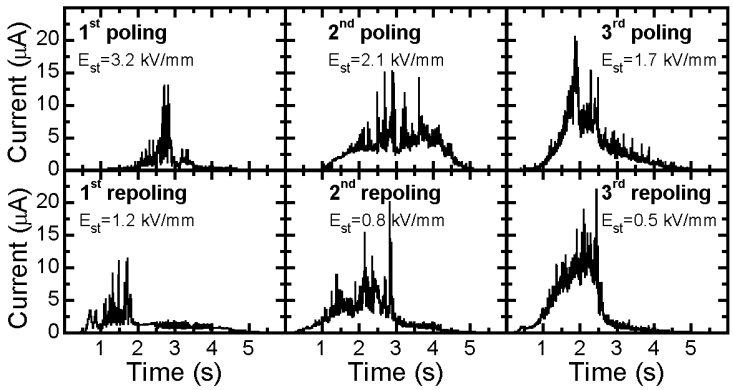

FIGURE 3. Evolution of switching current during cyclic switching in MgO:SLN. Liquid electrodes. E_{max} = 4 kV/mm.

Switching current and hysteresis measurements in MgO:SLN

The kinetics of domain structure in SLN and MgO:SLN while switching with liquid electrodes is very similar to that in CLN. In addition to optical observation we carried out the switching current and hysteresis loop measurements in MgO:SLN. We use half-period sine unipolar pulse of 6 s duration both for poling and repoling with 40 s delay between poling and repoling. It was obtained that cyclic switching leads to significant change of switching current (Fig. 3). The start field E_{st} introduced as field value corresponding to $\mathbf{j} = 0.02\ \mathbf{j}_{max}$, rapidly decreases during cycling, so as switching unipolarity (difference between start fields for poling and repoling). Increasing of current amplitude and total switching charge is the evidence of domain wall motion acceleration.

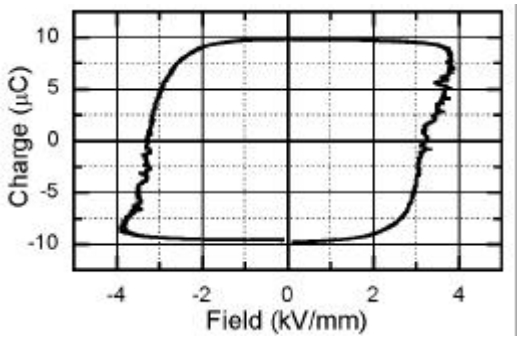

FIGURE 4. Hysteresis loop in MgO:SLN. Liquid electrodes.

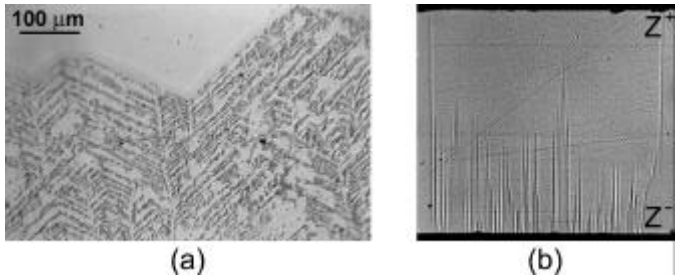

FIGURE 5. (a) Z^- view, (b) Y view of CDW in CLN. ITO electrodes. Optical visualization of etched surface. 0.2-mm-thick sample.

Measured hysteresis loop demonstrates coercive field E_c = 3.3 kV/mm (Fig. 4). At the same time the switching is practically "without threshold" – the threshold field $E_{th} \sim 0.1\ E_c$. We distinguish two stages during both poling and repoling. First, "smooth stage" – up to the half of switching charge the process goes without any sufficient noise in the signal. Second, "noisy stage" is accompanied by essential noise in the loop. It can be explained by high current amplitude in the circuit caused by fast domain processes, which leads to field "jumps".

CHARGED DOMAIN WALLS

Switching by ITO electrodes leads to formation of charged domain walls (CDW),[4] which propagates in CLN and SLN by motion of boundaries strictly oriented along crystallographic directions (Fig. 5a, 6), while in MgO:SLN the boundaries are more irregular (Fig. 7). CDW is visualized in crossed polarizers due to strong electrooptic effect.

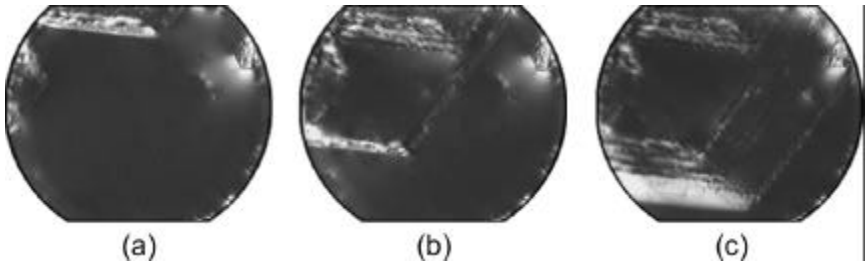

FIGURE 6. CDW in SLN. Time interval from field switch-on: (a) 2.4 s, (b) 2.6 s, (c) 2.8 s. ITO electrodes. E = 4.8 kV/mm. 1-mm-diameter area.

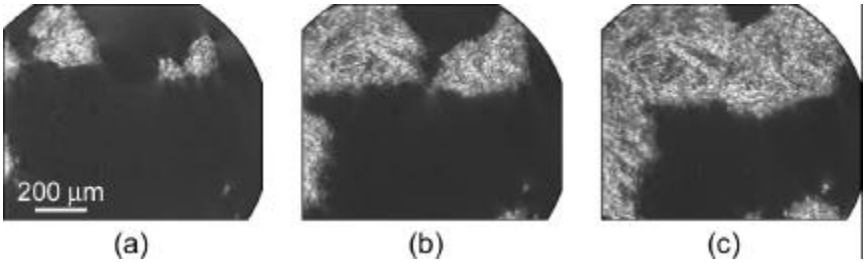

FIGURE 7. Formation of CDW in MgO:SLN. Time interval from field switch-on: (a) 0.9 s, (b) 1.5 s and (c) 2.1 s. ITO electrodes. E = 3.4 kV/mm.

Our observation shows that in used experimental conditions the domains nucleated at $\mathbf{Z}^+$ surface, grow in forward direction and coalesced at $\mathbf{Z}^+$, but never reach $\mathbf{Z}^-$ surface, thus forming an exotic dented domain pattern (Fig. 5b), which does not disappear after field switch-off and during subsequent switching cycles.

CONCLUSION

Domain kinetics has been investigated in congruent, stoichiometric and magnesium doped LN single crystals using transparent liquid and ITO electrodes. The domain evolution in all investigated materials is qualitatively similar. For liquid electrodes it is defined by jerky motion of the strictly oriented domain walls while for ITO the formation of the charged domain walls is obtained. The essential role of bulk screening process for stabilization of the charged domain walls is revealed.

The research was made possible in part by Programs "Basic Research in Russian Universities" (Grant No.5563) and “Priority Research in High School. Electronics”, by Grant No. 01-02-17443 of RFBR, and by Award No.REC-005 of CRDF.

References

[1] R.L. Byer, *J. Nonlinear Opt. Phys.& Mater.* **6**, 549 (1997).
[2] L. Huang, D. Hui, D.J. Bamford, S.J. Field, I. Mnushkina, L.E. Myers, and J.V. Kayser, *Appl. Phys. B* **72**, 301 (2001).
[3] V. Gopalan and T. Mitchell, J. Appl. Phys. **85**, 2304 (1999).
[4] V.Ya. Shur, E.L. Rumyantsev, E.V. Nikolaeva, and E.I. Shishkin, *Appl. Phys. Letters* **77**, 3636 (2000).