Restrictive domain motion in polarization switching of lithium niobate

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We investigate the electrostatic control of domain nucleation and motion for periodical polarization (P_s) switching on Z-cut congruent-grown lithium niobate (LiNbO$_3$). A tenfold reduction in the tangential field (E_x) can be resolved as the perturbed, positive charge density intervening otherwise the single-domain crystal exceeds $10^{-3} P_s$. This concept is realized in a two-step procedure consisting of a high-temperature treatment followed by pulsed field poling. Bulk periodic polarization switching is then realized underneath the oxidized electrode with the lateral 180° domain motion confined to the edge of the charged domain boundary. The forward domain growth can extend through the 500-μm-thick substrate and reach an aspect ratio as large as 150. These observations are attributed to the constraint on domain nucleation and the suppression of E_x by the positive charge originating from the divergence of P_s at the inverted domain boundary near the subsurface regime. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1503169]

The use of a quasi-phase-matching (QPM) technique[1] in nonlinear wave interactions has been actively pursued in the field of nonlinear optics. In ferroelectric nonlinear crystals, the QPM condition can be fulfilled by periodical domain reversal at every coherent length $l_c = \lambda_\omega / 4(n_{2\omega} - n_\omega)$ such that the sign change in the spontaneous polarization (P_s), and therefore the second-order nonlinear susceptibility $\chi^{(2)}$, can be used to compensate destructive phase interference.[2] Efficient parametric processes,[3] optical and acoustic applications,[4] just to name a few, are vivid examples benefiting from such investigations. Such a plethora of research activities are ascribed to the availability of QPM structures of periodically poled lithium niobate (PPLN);[5] lithium tantalate (PPLT);[6] potassium titanyl phosphate (PPKTP),[7] and its isomorphic family;[8] and more recently, the commercialization of these materials. A common practice is to invoke chemical exchange in the fabrication of QPM structures. Albeit simple in nature, employment of such methods inevitably results in surface phases of H$_x$Li$_{1-x}$NbO$_3$ (Ref. 9) and H$_x$Li$_{1-x}$TaO$_3$ (Ref. 10) or introduces surface refractive index dispersion in ion-changed KTP,[11] which are known to be detrimental to conversion efficiency.

Pulsed field poling, on the other hand, offers an alternative domain reversal approach by delivering a pulsed field across the crystal's polar (Z) axis to overcome the coercive field (E_c) and initiate the polarization switching process.[12] Based upon early studies of BaTiO$_3$, it is suggested that the surface nucleation rate, sidewise expansion, and coalescence of the domain constitute three major steps in the kinetics of polarization switching.[13,14] The situation, however, becomes complicated on congruent-grown LiNbO$_3$ and LiTaO$_3$ due to the existence of a large internal field and the associated axial anisotropy in the polarization switching process.[15] Improved domain poling techniques such as the nichrome (NiCr) electrode[16] and backswitching of the poling wave form[17] have been adapted to enhance the surface nucleation density

and to control the domain relaxation in these widely used nonlinear materials.

In the electrical poling scheme, one takes advantage of current injection from the external circuit to compensate the internal and depolarization field so as to initiate the polarization switching process.[17] Irregularity[18] and broadening[19] of inverted domains, however, have been constantly observed in the poling process and result in the loss of conversion efficiency. One such cause has been ascribed to the electrostatic interaction among the charged domain walls and nucleation steps.[18] A more fundamental issue, however, can be related to tangential field (E_x) enhanced current spreading in the unpatterned regime, which is normally coated with a layer of insulating material such as SiO$_2$ (Ref. 20) or spin on glass.[16]

As illustrated in Fig. 1 of a conventional poling configuration, a relatively large $E_x \sim 4E_c$ can exist underneath the insulating layer. Even though E_x only affects a thin region up to $z \sim 0.25 \, \Lambda$ and fades away at larger distance, what is important is its field line pointing to an *inward* direction from both sides underneath the insulating layer. This phenomenon is believed to result in field screening and compensating in the unpatterned regime, and brings up the issue of domain
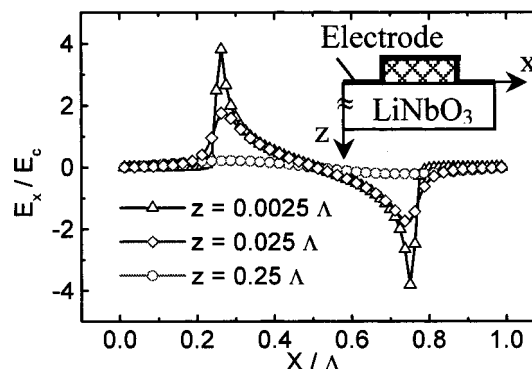

FIG. 1. Calculated tangential field (E_x) distribution at various depths beneath the insulating layer. Inset shows a QPM device in a conventional poling configuration.
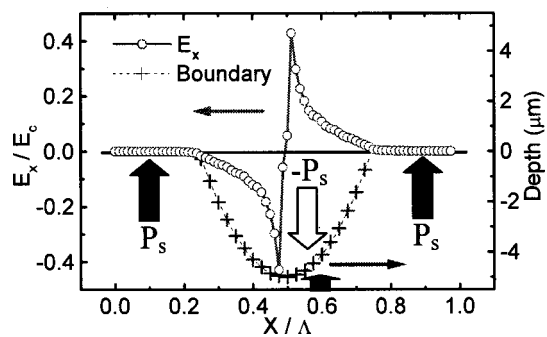
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FIG. 2. Calculated tangential field (E_x) at $z = 0.0025\Lambda$ underneath a positively charged parabola intervening between the electrode. The arrows indicate the polarity of P_s. Note the reduction of E_x and outward field line with respect to those in Fig. 1.

FIG. 3. $-y$ face micrograph of a PPLN-QPM device with 20 μm period after (a) the first heat treatment at 1050 °C for 5 h and (b) followed up by a pulsed field poling at 21 kV/mm. The substrate thickness is 500 μm.

broadening.[21] Our calculations of E_x and E_z are based upon a finite-difference method capable of solving the Poisson equation with a spatial distribution of charge and dielectric.[22] The structure parameters for evaluating the poling fields on the 500-μm-thick congruent-grown LiNbO$_3$ consist of an applied voltage of 10.5 kV, a dielectric constant of 30, a QPM periodicity Λ of 20 μm, and an insulating layer of photoresist with 1 μm thickness and dielectric constant of 3, respectively.

In this work, we report a proof-of-concept mechanism in which we design a spatial distribution of P_s in the subsurface regime to restrain the domain nucleation and motion in a subsequent field poling process. One such example is illustrated in Fig. 2, upon which a uniformly and positively charged parabola is introduced to represent the polarization-induced charge in a head-to-head configuration of P_s. We thereby evaluate the E_x at a depth $z = 0.0025\ \Lambda$ ($\Lambda = 20\ \mu$m) along the parabolic domain boundary. As the induced charge density increases to $10^{-3} P_s$, we note the replacement of the insulating layer by the positively charged domain boundary not only can cause a substantial E_x reduction (e.g., by one order of magnitude down to $0.4 E_c$ in the peak value), but also can result in a field line pointing to an *outward* direction. The latter signifies a promising mechanism to oppose the current spreading effect that would be valuable for electric poling.

We then proceeded with periodical polarization switching on Z-cut, 500-μm-thick, double-side polished, congruent-grown undoped LiNbO$_3$ substrates obtained from Crystal Technology. To derive a positive charge distribution via the divergence effect on P_s, we seek a solution by perturbing the subsurface domain structure with a polarity of P_s opposite to that of the virgin crystal.[23] A typical procedure is to let the patterned sample be placed inside a quartz tube furnace and undergo heat treatment at 1050 °C for 5 h in an air ambience. The sample was then transferred to a poling apparatus consisting of a pulsed voltage amplifier made from Trek (model 20/20A for 20 kV and 20 mA output). The poling setup was similar to that originally designed by Myers *et al.*,[24] in which a lithium chloride liquid electrode was made to contact the sample surfaces via a pair of viton O rings with 3 mm diam. To stabilize the domain reversal process, a fast turn-on rectifying diode was put in series with the poling apparatus such that relaxation of the inverted domain can be inhibited at termination of the pulsed field.[25]
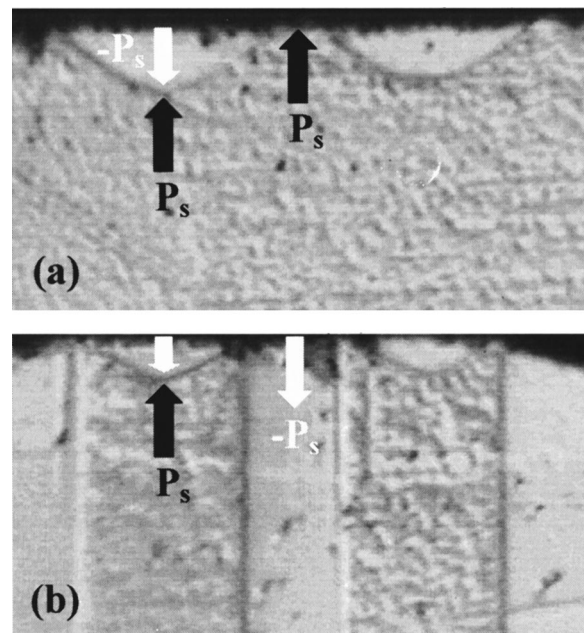
Illustrated in Fig. 3(a) is the $-y$ face micrograph of an etched 20-μm-period QPM structure after the first high-temperature treatment at 1050 °C. Albeit the thin ($\sim$50 nm) Al electrode oxidizes and transforms into an Al$_2$O$_3$ pattern during the heat treatment, we note it can *preserve* the underlying LiNbO$_3$ domain in its original polarization state. In addition, we find the porosity associated with microcracks in the Al$_2$O$_3$ electrode (not shown)[26] can provide a current limiting mechanism for the next step of pulsed field poling. Moreover, in the uncovered LiNbO$_3$ regime, shallow surface domain inversion takes place in a triangular spatial distribution, whose thickness can be as deep as 0.25Λ (5 μm). This phenomenon is in contrast to a recent study on SiO$_2$-coated LiNbO$_3$ (Ref. 27) and KTP,[28] upon which the stress-induced piezoelectric effect is reported to cause domain inversion under the SiO$_2$ clad. These observations suggest that the origin of surface domain inversion in the present study is dominated by the space-charge effect arising from the out-diffusion and defect gradient associated with the high-temperature process.[29–31]

We further exhibit in Fig. 3(b) the etched $-y$ face micrograph of a periodically poled LiNbO$_3$ QPM structure that has undergone both the heat treatment and pulsed field poling at 21 kV/mm. Instead of resuming a forward domain growth along the existing 180° triangular domain boundary,[13] we now have the bulk periodical polarization switching nucleate under the oxidized Al$_2$O$_3$ electrode on the $+Z$ face. These observations confirm the use of micropores from the oxidized electrode to establish an electric contact and form nucleation sites to the underlying LiNbO$_3$. In comparison, the inverted triangular domains formed during the first heat treatment remain unchanged and stay outside the electrode regime after the pulsed field poling.

The most important fact, however, is the observation of lateral 180° domain motion *confined* to the edge of the triangular domain boundary. This phenomenon differs from a re-
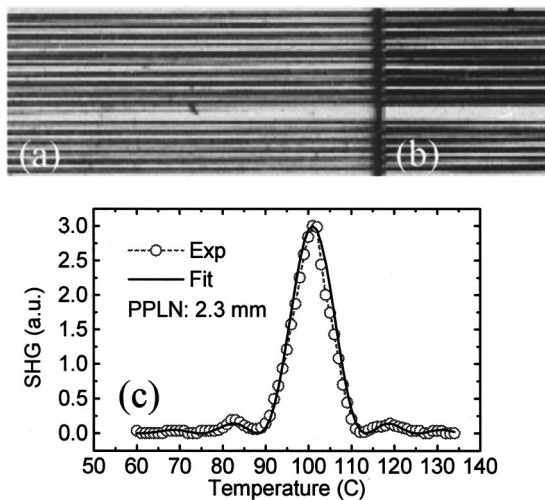
FIG. 4. Micrograph of a PPLN-QPM device with a nominal 6.7 μm period in the (a) $-y$, and (b) $-Z$ faces, and (c) the temperature-tuning curves of the SHG power measurement and the fitting result. The substrate thickness is 500 μm.

cent investigation upon which a nonpolarized phase of cubic perovskite was introduced to suppress the sidewise domain motion in LiTaO$_3$.[6] The above observations suggest the positive charge, arising from the divergence of P_s along the triangular domain boundary, can constitute a potential barrier against current spreading into the unpatterned regime and inhibit the propagation of domain forward growth along the existing boundary. In addition, it also supports the use of a repelling field line from the positive charge to reduce the tangential field and restrict the lateral 180° domain motion upon field poling.

In extending this two-step poling procedure toward the realization of second-harmonic-generation (SHG) lasers in the green and blue spectral regimes, we illustrate in Fig. 4 the etched micrograph of the (a) $-y$, and (b) $-Z$ faces of a QPM device with a nominal 6.7 μm period on 500-μm-thick LiNbO$_3$. A close examination of the switched domains reveals a mirror image correspondence between the etched patterns in the $-y$ and $-Z$ faces, respectively. This observation confirms the forward growth of inverted domains and their restrictive motion bound by the positive charge. Using a cw Nd:YAG laser as the pumping source, data shown in Fig. 4(c) reveal a good temperature tuning curve for the SHG power measurement. The fitting result indicates the QPM device has reached a phase-matching length of 2.3 mm, which is limited by the contact O ring used in the poling apparatus.

In summary, we report the use of electrostatic control via $-\nabla \cdot P_s = \rho$ to manipulate the interaction among domain walls and nucleation sites during the pulsed field poling process. By introducing a positive charge distribution intervening between the patterned electrodes, bulk periodic polar-

ization switching in LiNbO$_3$ is found to be confined to the edge of the charged domain boundary. These observations are ascribed to the constraint on domain nucleation and suppression of tangential field due to the polarization-induced charge at the inverted surface domain boundary.

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