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Citation: Applied Physics Letters 92, 092903 (2008); doi: 10.1063/1.2890728
View online: http://dx.doi.org/10.1063/1.2890728
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Depolarization field mitigated domain engineering in nickel diffused lithium tantalate


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(Received 7 January 2008; accepted 5 February 2008; published online 4 March 2008)

We demonstrated a domain reversal mechanism on Z-cut congruent-grown lithium tantalate (LiTaO$_3$) composed of nickel (Ni) diffusion followed by pulse field poling. Domain nucleation and forward growth were found confined to the nondiffused regions, where the commonly observed serrated domain fronts in poled LiTaO$_3$ were absent in this work. These observations are ascribed to the formation of domain nucleation barrier by the depolarization field and reveal the divergence effect in the ferroelectric spontaneous polarization at the domain boundary due to Ni diffusion. This mechanism simplifies the fabrication of periodically poled LiTaO$_3$ for second-harmonic generation in the blue spectral regime. © 2008 American Institute of Physics. [DOI: 10.1063/1.2890728]

Ferroelectric crystals are characterized by the reversibility of spontaneous polarization ($P_s$) when subject to an external electric field poling above the crystal’s coercive field $E_c$. The capability for ferroelectrics to retain a periodically poled domain structure with alternating sign change in $P_s$ can substantially modify the material’s tensor properties. This effect can lead to a realization of high-density nonvolatile memory devices due to fast polarization switching in ferroelectric thinfilms. Periodic sign reversal in $P_s$ can further impose on the ferroelectric a structure-related vector to compensate the phase mismatch in nonlinear wave mixing processes. This mechanism is known as quasi-phase-matching (QPM) and has been recently applied to second-harmonic generation (SHG) and third-harmonic generation and optical parametric oscillation (OPO). Compared with the conventional birefringent phase matching scheme, the QPM method offers a higher conversion efficiency due to access to the largest nonlinear susceptibility tensor $\chi^{(2)}$ and absence of beam walk off. Recent studies further clarify the advantages of using two-dimensionally (2D) poled ferroelectric nonlinear photonic crystals to achieve broad acceptance bandwidth in the spectral-tuning and temperature-tuning range, and interesting optical phenomena such as cascade, conic, and Čerenkov generation in the QPM process.

These observations prompt a need of domain engineering to deploy a desirable pattern transfer in the electrically poled ferroelectrics. A typical structure composed of an insulator pattern over gratinglike metallic contact or vice versa has been demonstrated for the poling electrode. The polarization switching process is known to initiate underneath the corrugated edge of the electrode thus made. The corresponding domain nucleation rate has been found proportional to $\sim \exp(-\alpha/E_c)$, where $\alpha$ represents a phenomenological activation term and $E_c$ the poling field. The kinetics of domain reversal is known to consist of a fast domain forward growth followed by a sidewise motion. Conventional wisdom further suggests a use of pulse field shaping such as back switching to mitigate the polarization switching process.

Caution, however, arises from frequent observation of unbounded expansion and merging of inverted domains in periodically poled lithium niobate (LiNbO$_3$) and lithium tantalate (LiTaO$_3$). For example, merging of inverted domains in LiTaO$_3$ can result in a serrated front which displays an order of magnitude increase in the wall velocity. This hampers the development of fine-pitch poled LiTaO$_3$ as the device lateral homogeneity decreases along the crystal’s polar axis. Examination of the electrostatic field distribution indicates that the driving force of domain broadening is due to the fringing field effect. The latter reflects field distortion due to bending of the equal potential line across the corrugated ferroelectric/electrode interface. Such distorted field line can extend beyond the electrode contact window and cause domain broadening.

In this work, we demonstrate a ferroelectric poling scheme on congruent-grown LiTaO$_3$ that can be immune to the aforementioned shortcomings. It is based upon Landauer’s nucleation theory in which bound charges, arising from the divergence of $P_s$ at the domain boundary, can constitute a nucleation barrier to prohibit the nucleation of inverted domain and its forward growth. With a moderate bound charge density $\sim 1 \mu C/cm^2$, a large depolarization field $\sim 10^7 V/cm$ has been estimated at the domain boundary. This number is two orders of magnitude higher than the crystal’s coercive field and can, thus, inhibit the applied field from initiating domain reversal. It has further been noted that between pairs of charged boundaries, the field line due to electrostatic repulsion can facilitate the confinement of lateral domain motion.

A crucial part of device processing as proposed in this work is to prepare a periodic distribution of positively bound charges underneath the +Z surface of LiTaO$_3$. A “head-to-head” configuration of $P_s$, viz., $\nabla \cdot P_s = -\rho$, as inferred from the electrostatics analysis. Such bound charges, when not screened by the injected carriers, can produce a depolarization field to constrain the nucleation and motion of the inverted domain. A head-to-head domain con-
figuration can be realized by introducing a shallow inverted domain in the virgin ferroelectric crystal. This type of domain configuration has been reported on the +Z face of LiNbO$_3$ by using titanium (Ti) in diffusion~\cite{25} or lithium (Li) out diffusion~\cite{27} at a temperature higher than 1050 °C. However, similar heat treatment on LiTaO$_3$ at ~600 °C, which is below the crystal’s Curie temperature, failed to create such inverted domains.~\cite{28} The reason has been attributed to the lack of activated diffusion process with which the impurity-gradient induced field is insufficient to initiate the polarization switching.

To resolve this issue, we seek solutions from a nickel (Ni) diffusion process which has been applied to make waveguides in LiTaO$_3$.~\cite{29} Testing electrodes made of inverted domains.~\cite{28} The reason has been attributed to the lack of activated diffusion process with which the impurity-gradient induced field is insufficient to initiate the polarization switching.

First shown in Fig. 1(a) is the SEM micrograph taken from the etched −Z face of a poled LiTaO$_3$ Sample A whose right half plane in the crystal’s +Z face was diffused with Ni prior to proceed with electric poling. Referring to Fig. 1(a), a sparse distribution of triangular shape of hillocks corresponding to the inverted domains are discernible in the right half plane of the sample’s −Z face; whereas uniformly poled domains are observed in the other left half plane. Further evidence revealing the Ni-affiliated domain poling process can be found from the magnified SEM micrograph of etch pits shown in Fig. 1(b). In the latter case, the micrograph was taken from the right half plane of Sample A’s +Z face. The etch pits shown here exhibit a size distribution in the nanometer to submicron regime and an invert-pyramid shape complementary to that observed in Fig. 1(a). They represent nuclei of inverted domains which respond to electric poling in the Ni-diffused region over the +Z face LiTaO$_3$. It further suggests that the fissurelike structure in an oxidized metallic film can act an electric path to the underlying ferroelectric surface and serve as nucleation site to initiate domain reversal.~\cite{25} This mechanism is confirmed in Fig. 1(c) where the cross-sectional optical micrograph taken from the right half side of sample A exhibits a straightforward growth of inverted domains through the 0.5 mm thick substrate. One can therefore conclude from Fig. 1 that for regions diffused with Ni there appears a barrier to inhibit the occurrence of polarization switching.

A possible link between the Ni diffusion and the polarization switching of LiTaO$_3$ is via the depolarization field to inhibit the nucleation of inverted domain. This proposed mechanism can be verified by examining the etch profile of periodically poled LiTaO$_3$ samples diffused with Ni electrodes. As example, in Fig. 2, we overlay the (a) AFM surface profilometry data with (b) SEM micrograph taken upon the etched +Z face of a periodically poled LiTaO$_3$ sample B diffused with a ~29.5 μm period of Ni grating. We first note a presence of the virgin LiTaO$_3$ crystal’s polarization state (+P$_s$) in the non-diffused and unpoled region I. The latter serves as a level reference as an etch depth of ~0.55 μm can be inferred from all of the Ni-diffused regions II regardless it has been electrically poled or not. This observation suggests that the Ni-diffusion region is characterized with negative polarization state. In comparison, a ~1.7 μm etch depth associated with electrically poled domains is observed in the non-Ni diffused region III. It reveals a spatial confinement of the electrically poled domains (region III) bounded by the Ni-diffusion layer (region II), suggesting that the latter acts as a barrier to prohibit lateral domain motion. This analysis is further confirmed in Figs. 2(b) and 2(c) where the +Z and −Z face optical micrographs of another set of periodically poled LiTaO$_3$ Sample using Ni-diffused grating electrodes exhibit a ~50% duty cycle in the poled domain structure. Furthermore, by closely examining the surface morphology of Fig. 2(a), one can identify the etch stop surface in the Ni-diffused layer (region II) to be associated with the positive domain side in the +Z face of LiTaO$_3$. We also found Ni diffusion in the −Z face of LiTaO$_3$ (data not shown here) can still retain the etching characteristics of negative polarization state. These analyses verify that the Ni diffusion process in LiTaO$_3$, like the case of Ti diffusion in LiNbO$_3$,~\cite{27} can result

FIG. 1. (Color online) SEM micrographs of the etched (a) −Z and (b) +Z faces of poled LiTaO$_3$ sample A with right half of its +Z face diffused with Ni prior to proceed with electric poling. (c) Cross-sectional optical micrograph taken from the right half side of sample A showing a straight growth of inverted domains through the 0.5 mm thick substrate.

FIG. 2. (Color online) (a) AFM surface profilometry taken from the +Z face of periodically poled LiTaO$_3$ sample B and overlaid with the SEM micrograph. Region I: nondiffused, nonepoled. Region II: Ni-diffused, electrically poled. Region III: nondiffused, electrically poled. Region IV: Ni-diffused, nonepoled. (b) +Z and (c) −Z face micrograph of an electrically poled LiTaO$_3$ sample with Ni-diffused electrode of 29 μm period.
in shallow surface domain inversion in the +Z face and retain the virgin ferroelectric crystal’s polarity in the −Z face. It further supports our proposal by invoking the depolarization field to mediate the polarization switching process and to manipulate the nucleation and motion of inverted domains. In comparison, we note that the commonly used proton exchange method can only result in surface domain inversion on the −Z face of LiTaO₃.¹ It, however, can severely modify the ferroelectric LiTaO₃ or LiNbO₃ +Z face into a cubic perovskite structure by losing the crystal’s spontaneous polarization.²³,3³

As a further proof of this proposed mechanism in shaping the ferroelectric domain structures, we prepared another set of 2D periodically poled LiTaO₃ sample C with a periodicity of 5.1 × 5.1 μm². Here, the optical micrograph in the inset of Fig. 3 depicts the periodic domain structure of sample C in the −Z face where 2D array of equilateral triangular domains are clearly observed to reside on the tetragonal lattice sites that have been electrically poled through the 0.5 mm thick substrate. These data provide evidence that the serrated wall front, constantly observed in poled LiTaO₃ due to fast step growth at the merged domain boundaries,¹⁹,3⁴ is absent in our poling scheme. Last but not the least, we illustrate in Fig. 3 the QPM-SHG wavelength tuning spectra of sample C which matches the generation of 465 nm blue laser. The experiments were conducted by using a home-built green laser pumped OPO with a temperature-tuned periodically poled stoichiometric LiTaO₃ to operate in the 930 nm band. At normal incidence (θ = 0°) the QPM-SHG process can be fulfilled at a wavelength of 925.5 and 923.5 nm, respectively, by matching the condition of 2kg₂ + Gmn = k₂mn at a reciprocal vector of G₁,0 and G₁,±1. The smaller Fourier component of χ⁽²⁾(G₁,±₁) accounts for a lower SHG conversion efficiency compared with its counterpart of χ⁽²⁾(G₁,0). By slightly rotating the sample at 4°, the degeneracy of QPM-SHG wavelength at G₁,1 and G₁,−1 splits to 915 and 935 nm. Detailed optical analysis of these 2D poled LiTaO₃ nonlinear photonic crystals will be presented in a forthcoming publication.

In summary, we have demonstrated a method of creating surface inverted domain on the +Z face of LiTaO₃ by Ni diffusion. By invoking the divergence effect of spontaneous polarization and judiciously manipulating the depolarization field, not only can one suppress the domain nucleation probability but also confine the motion of electrically poled domains to the boundary formed by the Ni diffusion process. This mechanism allows us to fabricate periodically poled LiTaO₃ structures for visible SHG laser applications.

The authors wish to acknowledge supporting from the National Science Council, Grant No. NSC 96-2221-E-002-098.

²For a recent review, see, Micro/nano Engineering and Characterization of Ferroelectric Crystals For Applications in Photonics, edited by P. Ferraro, S. Grilli, and P. De Natale (Springer, New York, 2008).
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