AIP | Applied Physics Letters

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Citation: Applied Physics Letters **98**, 062902 (2011); doi: 10.1063/1.3553194
View online: http://dx.doi.org/10.1063/1.3553194
View Table of Contents: http://scitation.aip.org/content/aip/journal/apl/98/6?ver=pdfcov
Published by the AIP Publishing

Direct writing of ferroelectric domains on the *x*- and *y*-faces of lithium niobate using a continuous wave ultraviolet laser

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(Received 10 December 2010; accepted 18 January 2011; published online 7 February 2011)

Ferroelectric domain reversal has been achieved by scanning a tightly focused, strongly absorbed ultraviolet laser beam across the *x*- and *y*-faces of lithium niobate crystals. The domains were investigated by piezoresponse force microscopy. The emergence and width of any domain was found to depend on the scanning direction of the irradiating laser beam with respect to the polar *z*-axis. Full width and half width domains or no domain formation at all could be achieved for scanning along specific directions. We interpret the results by a direct correlation between the local temperature gradient and the resulting polarization direction. © *2011 American Institute of Physics*. [doi:10.1063/1.3553194]

Lithium niobate ($LiNbO_3$) has recently become the material of first choice for nonlinear optical applications in the visible and near-infrared wavelength range.[1] In many cases, patterning of ferroelectric domains at a length scale of sub-to-few microns is required. For instance, frequency conversion employing quasiphase matching relies on a periodically poled domain pattern.[2] Domain patterning is in general performed by electric field poling with structured electrodes, which reverses the direction of the spontaneous polarization by locally applying an electric field along the polar *z*-axis of the crystal, exceeding the so-called coercive field (E_c).[3]

A different approach for domain patterning uses UV laser beam writing. It was found that irradiating the $-z$ face of $LiNbO_3$ with a tightly focused, strongly absorbed UV laser beam directly poles domains along the laser-written track without any application of an external electric field.[4] The very same procedure on the $+z$ face results in poling inhibition of the irradiated area.[5] In both cases, i.e., of direct poling and poling inhibition, the resulting domains were found to follow the laser track, irrespective of the writing direction with respect to the crystallographic axes, and to expand a few microns into the crystal depth.

Indeed, for some applications, such as specific waveguide configurations[6,7] or efficient frequency doubling in whispering gallery mode (WGM) resonators,[8] domain patterning via the *x*- and *y*-faces of $LiNbO_3$ is also desired. In the case of ridge waveguides, the issue was addressed using a cumbersome fabrication process for the electrodes and thereby poling across the ridges (and thus again along the *z*-axis).[9,10] For WGM resonators, utilizing radially poled *z*-cut crystals might be a possible alternative.[11] Although isolated solutions for specific applications have been developed, an easy-to-apply method for domain patterning via the nonpolar faces is still needed. A first approach addressing this issue has been reported recently, creating domains on the *y* face of $LiNbO_3$ crystals by electron beam writing.[12]

In this contribution, we present UV laser-induced ferroelectric domain inversion by irradiating the *x*- and *y*-faces of $LiNbO_3$ crystals. Interestingly, the emergence and width of any domain formed was found to depend on the scanning direction of the irradiating laser beam with respect to the direction of the polar *z*-axis.

For our experiments, we used 500 μm thick *x*- and *y*-cut crystals of congruent lithium niobate (CLN) from Crystal Technology, Inc. and congruent lithium niobate with 5 mol % magnesium doping (MgCLN) from Yamaju Ceramics Co., Ltd. UV laser light of 275 nm wavelength with an incident power of 35 mW was focused on the crystal surfaces to a spot focal diameter of 5 μm (full width at half maximum). The absorption length is of the order of 1 μm only,[13] giving rise to strong local heating of the crystal at the laser-irradiated area. In order to write laser tracks, the crystals were moved with a velocity of $v=100$ μm/s using a high-precision computer-controlled translation stage. Laser tracks were written either parallel, antiparallel, or perpendicular to the *z*-axis of the crystal, denoted later as $v \rightrightarrows z$, $v \leftrightarrows z$, and $v \perp z$, respectively. The generated domain patterns were investigated by piezoresponse force microscopy (PFM).[14]

Figure 1 shows the result of a UV-irradiated track written along the *y* direction on an *x*-cut sample. The width of the UV-irradiated track manifests itself in the topography as a shallow indentation of ≈6 nm depth caused by a brief polishing step, which more efficiently removed the part of the crystal affected by the strong laser irradiation [Fig. 1(a)].

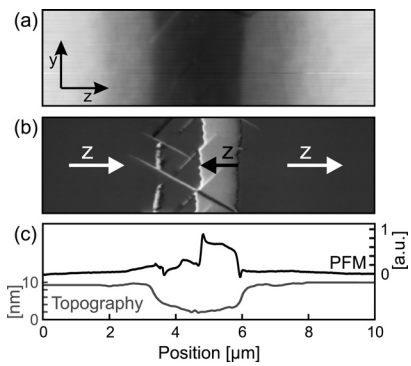

FIG. 1. Topography (a) and simultaneously recorded PFM image (b) of a Mg-doped *x*-cut $LiNbO_3$ crystal previously irradiated with a UV laser beam scanned along the *y*-axis. As best seen by the line scans in (c), a domain of half the width of the laser beam has been generated. The narrow line features seen in (b) are thermoinduced defects.

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The broad bright stripe (shown in light gray) in the PFM image corresponds to a directly poled domain [Fig. 1(b)]. Obviously, domain inversion occurred only within approximately half the width of the UV-irradiated track [Fig. 1(c)]. This characteristic is confirmed by the presence of the thermoinduced damage due to the irradiation procedure across the full width of the irradiated track [Fig. 1(b)]. Wedge polishing of the *y* face reveals that the poled domains are ≈ 1.5 μm deep, whereas the thermoinduced damage reaches a depth of up to 3 μm.

In order to comprehend the mechanism of UV laser direct poling via the nonpolar faces of $LiNbO_3$, we refer to experiments where crystals were subjected to temperature gradients along the *z*-axis either as bulk samples, by annealing in an oven exhibiting a nonuniform temperature profile,[15] or locally, by irradiation with a strongly absorbed infrared (IR) laser beam.[16] In these experiments, the direction of the spontaneous polarization was observed to follow the projection of the temperature gradient on the *z*-axis: For the annealing procedure, the $+z$ face developed at the warmer end of the crystal and for the case of local laser irradiation, a head-to-head domain boundary formed at the center of the laser beam due to the opposing temperature gradients. Both publications presume that the temperature gradient in the crystal causes an electric field whose origin is of thermoelectric nature. It is important to note that the experiments mentioned above were carried out at temperatures close to, if not above, the Curie point of $T_c = 1415$ K. Indeed, at temperatures very close to T_c, the value of the coercive field is well below 100 V/mm[17] and consequently, small electric fields are sufficient to pole the crystal.

We now propose that the mechanism described above applies also to our case of UV laser irradiation. As in the case of IR laser irradiation,[16] the crystal is locally heated close to or even above T_c, and the opposing temperature gradients lead to a head-to-head domain boundary formed at the center of the laser beam. Consequently, one half of the width of the laser-irradiated track is domain-inverted (see Fig. 1). For our experimental situation, we can estimate the temperature gradient to be $\Delta T \approx 200$ K/μm, whereby the width of the temperature profile is larger than the width of the laser profile.[18] Together with the appropriate value of the thermoelectric power tensor $Q_{33} = 0.8$ mV/K,[16] we obtain a thermoelectric field of 160 V/mm, sufficient to set the direction of spontaneous polarization at temperatures close to T_c.

The proposed scenario is summarized in Fig. 2: Strongly absorbed laser irradiation gives rise to a temperature profile, (a), which implies opposed temperature gradients, (b). These result in a bipolar electric field, (c), which causes domain inversion in half of the heated volume, (d): The width *d* of the resulting inverted domain is expected to depend on the temperature profile and thus on the focusing and the power of the irradiating laser beam.

Figures 3(b)–3(d) show PFM images obtained on a *y*-cut CLN crystal where laser writing has been performed along *x*- and *z*-directions. The individual laser tracks are numbered in chronological order by ① to ④. When the laser beam moves along the *x*-axis, which is perpendicular to the *z*-axis ($v \perp z$; ① and ②), domains of only half the width of the laser beam are generated and the direction of the laser beam movement has no influence on the generated domains. This corresponds exactly to the situation which has been described above (Fig. 2). However, when the laser beam moves parallel to the

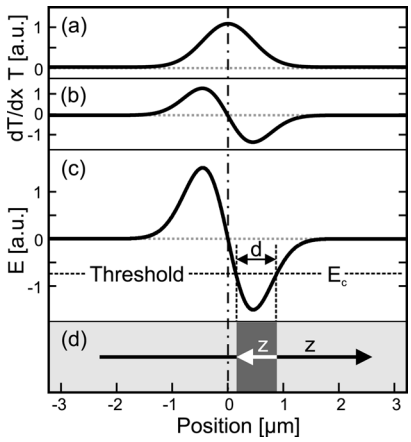

FIG. 2. Schematic illustration of the mechanism for domain inversion of an *x*- or a *y*-cut $LiNbO_3$ crystal upon UV laser irradiation. Due to the high absorption of the material, the crystal is locally heated to its melting point (a), leading to opposing temperature gradients (b). A bipolar electric field builds up (c), which, in a region of width *d*, exceeds the coercive field E_c and therefore leads to poling. The resulting domain pattern is shown in (d).

z-axis, either a domain of the full width of the laser beam is generated ($v \leftrightarrows z$; ④) or no domain at all emerges ($v \rightrightarrows z$; ③). Note that previously written domains (from the laser tracks ① and ②) have been poled back from laser writing along track ③ [Fig. 3(c)].

The results shown in Figs. 3(b)–3(d), although surprising at first, match the phenomenological model we described above, as is schematically shown in Fig. 3(a) under this experimental configuration. The poling behavior seems to com-

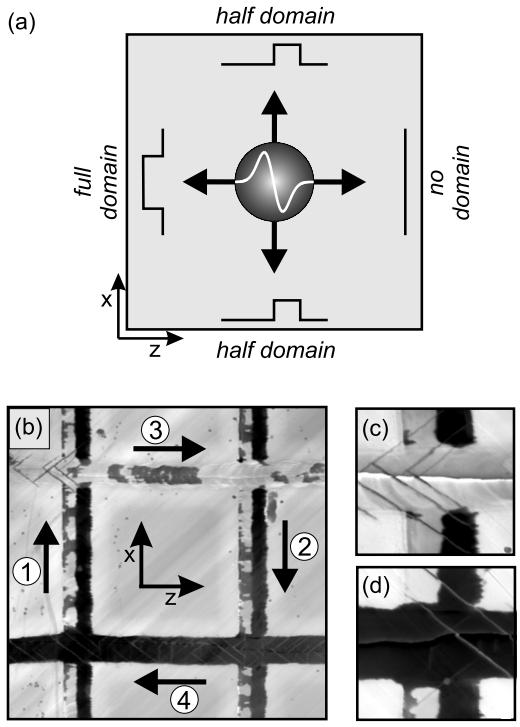

FIG. 3. Dependence of the poling on the scanning direction of the laser beam. (a) shows a schematic with the laser beam at the center and the bipolar electric field distribution inside. The arrows indicate the scanning direction of the laser beam. [(b)–(d)] PFM images showing the influence of both the direction and the chronology (from ① to ④) of laser irradiation on a *y*-cut CLN crystal. The horizontal lines, best seen in (c) and (d), are thermoinduced cracks also visible in the topography. The image sizes are 30 μm × 30 μm in (b) and 5 μm × 5 μm in (c) and (d).

ply with the following two rules: (i) Depending on the orientation of the temperature gradient with respect to the direction of the z-axis, only the part of the temperature gradient that produces the appropriate field polarity will result in ferroelectric domain inversion, as shown in the schematic of Fig. 3 when the laser is scanned perpendicularly to the z-axis; and (ii) the final polarization state is defined during the cooling stage as the temperature drops below T_c. Hence, when the laser scanning is along the z-axis of the crystal, it is the trailing edge of the UV laser beam with respect to the direction of the z-axis that defines whether the cooling part of the crystal will domain-invert or not.

In order to further support our interpretation of our experimental findings, we carried out a series of additional experiments, which we briefly summarize in the following: (i) The x- and the y-faces show the same poling behavior, (ii) the width of the domains was found to decrease with decreasing laser power (keeping the focusing constant), (iii) their depth was measured to be of a few microns, (iv) with strongly reduced laser intensity (by a factor of two), poling was no longer observed, and (v) poling works for both CLN and MgCLN crystals, which is an important issue in terms of the applicability of this technique. All observations described above are in accordance with the model described previously.

The experiments on the nonpolar x- and y-faces of $LiNbO_3$ presented in this paper are a further facet of our investigations on laser-induced domain formation with a tightly focused, strongly absorbed UV laser beam using $LiNbO_3$ crystals. We would therefore like to put our findings on the x- and y-faces in the context of the results previously obtained on the polar z faces,[19] the irradiating parameters being the same in all cases. Generally, it can be said that on the x- and y-faces and the $-z$ face, direct poling upon laser irradiation is observed whereas on the $+z$ face, poling inhibition, whereby domain formation occurs only after subsequent electric field poling, was found. According to our current understanding, domain formation on all faces relies on the heat profile generated in the crystal by laser irradiation. This is supported by the fact that on all faces, the depth of the generated domains is of the same order (a few microns). In the case of x- and y-faces and the $-z$ face, the direction of the spontaneous polarization was confirmed to follow the temperature gradient, as has been observed previously.[15,16] The presumed thermoelectric explanation for the buildup of an electric field causing domain reversal likely needs to be expanded by also taking the pyroelectric effect into account, an issue certainly demanding further investigation. In the case of poling inhibition that is observed on the $+z$ face, lithium redistribution due to heating was found to play the crucial role.[20]

In conclusion, we have shown that ferroelectric domain reversal can be performed by UV irradiation on the nonpolar faces of lithium niobate crystals, thereby generating domains of a few microns depth. We suggest that the mechanism for domain formation could be attributed to a bipolar electric field that builds up during laser irradiation. Together with the already established poling by UV irradiation on the polar z faces, it is therefore now possible to generate domain patterns on all crystal faces, thereby allowing for a wealth of possibilities for application-relevant domain structuring.

We thank D. Jundt for fruitful discussions. The project was supported by a Materials World Network (MWN) grant funded by the Deutsche Forschungsgemeinschaft and the U.S. National Science Foundation (Grant No. NSF-DMR 60 0602986). Further financial support by the Deutsche Telekom AG is also gratefully acknowledged. Financial support from the EPSRC in the framework of Grant No. EP/C515668/1 Portfolio Partnership in Photonics is gratefully acknowledged

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