Light-induced domain engineering in ferroelectrics: a route to sub-micron poling

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Introduction

While several techniques to achieve domain inversion in ferroelectric materials such as lithium niobate and lithium tantalate have been successfully demonstrated over the past years, even the 'best' established technique of electric field-induced domain inversion (E-field poling) fails when domain inversion at periodicities of a few microns and below are desired. In order to overcome the limitations associated with E-field poling, we have been investigating the feasibility of a relatively simple single-step technique, which exploits the interaction of intense laser light with ferroelectric materials to engineer their domains at micron and sub-micron scale-lengths. Some light-assisted poling experiments, which take advantage of the ultraviolet light-induced transient change in the coercive field of the illuminated ferroelectric material to transfer a patterned light distribution into an equivalent domain structure in bulk crystals have already been reported for lithium niobate[1,2] and lithium tantalate[3,4] crystals.

We have earlier reported a direct all optical poling (AOP) technique that employs pulsed ultraviolet (UV) laser light to induce surface domain inversion on the +[5] and –[6] z-faces of lithium niobate in a single step. We have been trying to understand the physical principles involved in light-matter interaction with an aim of controlling the shape, size and depth of the randomly orientated light-induced domains with an end goal of tailoring periodic structures in lithium niobate. This report presents the results of our attempts to achieve a degree of control over the nucleation and growth of the domains, via an imposed incident light pattern produced using a phase-mask. It is shown that it is possible to align the surface domains along the maxima of the spatially modulated UV laser light pattern.

Experiments and Results

Optically polished, 500 µm thick, z-cut, undoped congruent lithium niobate crystals supplied by Crystal Technology, USA were used for the AOP experiments.

To investigate the wavelength sensitivity of the AOP process, and also dependence of the depth of the induced domains on the incident wavelength, we have explored a range of laser wavelengths (between 298 nm and 329 nm) that straddle the UV absorption edge of lithium niobate. Laser illumination at these wavelengths was through a tunable Dye laser (NSL-5) available from the Laser Loan Pool of the RAL, and a frequency -quadrupled ND:YVO_4 laser. The NSL-5 laser system was a frequency-doubled Q-switched Nd:YAG laser (Continuum Powerlite 8000) pumping a frequency-doubled dye laser (Sirah PrecisionScan), generating mJ level pulses between 298 nm and 329 nm with pulse durations of ~7-ns. The frequency-quadrupled NdYVO_4 laser operated at 266nm and produced ~5mJ pulses with durations of ~10ns. In order to eliminate the inhomogeneities in the profile of the beam from the dye laser, the beam was passed through an aperture to select a small region of acceptable spatial uniformity. The + z-faces of the crystals were exposed by UV laser light through a phase-mask, which was separated by a distance of 125µm from the crystal face using two identical standard telecom fibers.

As it was known from earlier experiments[5] that AOP of lithium niobate is only possible around its ablation threshold; the single pulse ablation threshold of lithium niobate was ascertained before the AOP experiments. The single pulse ablation thresholds for 266 nm and 298 nm light were established experimentally to be around between 170-190 mJ/cm^2 however, these figures are subject to some degree of uncertainty due to the intrinsic spatial non-uniformity and temporal (pulse to pulse) fluctuation of the dye laser.

Samples were exposed to single and multiple-pulses (up to 10 pulses) at different wavelengths, over a wide range of fluences (between 60 mJ/cm^2 and 360 mJ/cm^2), and then compared with samples exposed under the same conditions but without the phase-mask.

After illumination, the UV exposed sample surface was etched for 20-30 minutes in 48% hydrofluoric acid (HF) and inspected with optical and scanning electron microscopy (SEM).

Results and discussion

Areas irradiated through the phase-mask at fluences significantly below the ablation threshold showed no evidence of domain formation. For a narrow range of fluences between (~190 to ~250 mJ/cm^2) domain formations parallel to the phase-mask lines were possible, however with a noticeable degree of associated ablation. The effect of UV illumination with and without the use of a phase-mask is shown in Fig. 1. Fig. 1a is an SEM image of an etched surface exposed without the use of a phase-

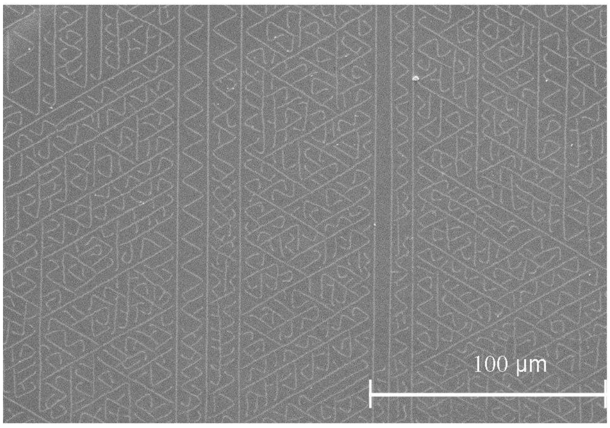

Figure 1a. SEM image of a sample exposed without a phase-mask.

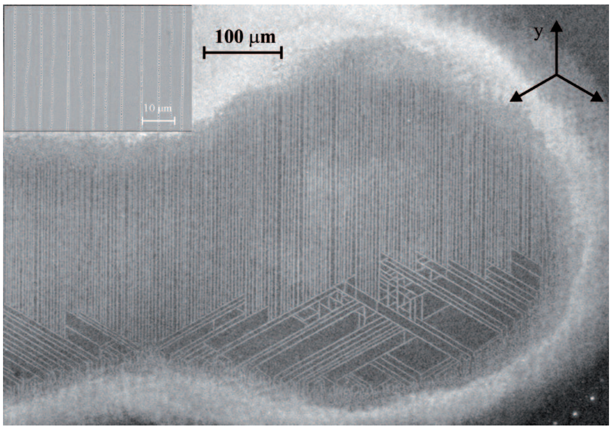

Figure 1b. SEM image of a sample exposed with a phase-mask. Inset shows a magnified image of a section of Fig. 1b.

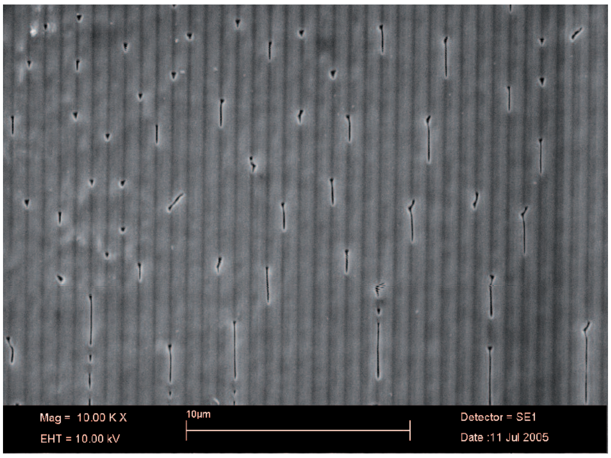

Figure 2. SEM image showing an edge of an illuminated area.

mask, and shows random nucleation and propagation of light-induced domains along the three equivalent symmetry directions of the crystal. This effect has been extensively discussed before in reference[5]. Fig. 1b shows an SEM image of the crystal surface that has been irradiated using a phase mask. The difference between the two images is distinctly apparent, with Fig. 1b showing the majority of the light-induced domains aligned along a specific symmetry direction, (vertical in the figure). This alignment however has been imposed by the orientation of the phase-mask, which coincides with one of the three crystallographic y-axes. As expected domains induced by AOP nucleate and propagate only in the presence of light of near damage threshold optical intensity along the same symmetry direction.

A detailed investigation of the domain formation shows that the effect is more complex than initially expected. The inset of Fig. 1b shows a magnified SEM image of a section of Fig. 1b. Although the domain lines are formed parallel to the phase mask lines, it is however clear that the periodicity imposed by the phase mask ($\Lambda = 0.726\mu m$) has not been faithfully reproduced in the resultant domain spacing. This is indicative of an electrostatic interaction between the photo-generated surface charges generated at the highly UV absorbing surface. The resulting electrostatic repulsion and re-organisation (e.g. clustering around surface defects) imposes a characteristic electrostatic interaction length that overrides the imposed periodicity of the light intensity pattern. The material is unable to both nucleate and sustain such closely packed domains via a strictly AOP process only. The inset of Fig. 1b also suggests electrostatic interaction between individual domains, namely that the domain lines produced are not continuous, but consist of irregular sections along the y-axis direction.

It was also observed that a slight variation of local intensity levels across the span of the incident laser spot significantly affects the AOP domain formation. As shown in Fig. 2, this variation subsequently results in regions within the illuminated spot showing different stages of nucleation and propagation of the light-induced domains as a function of intensity. Fig. 2 shows an edge of an illuminated area which has experienced therefore a slightly lower fluence. The area was illuminated through a phase-mask with two pulses at 298 nm at a fluence of ~210 mJ/cm^2. It is evident from the SEM image that nucleation, and occasional limited subsequent propagation of domains, always occurs only at positions of maximum light intensity. The intensity variation dictates whether the light-induced domains nucleate only or nucleate and further grow along the symmetry direction. An area where both nucleation and growth (propagation) can also be observed in the same figure (bottom right) however, these expanded domains are spaced further apart resembling the situation shown in the inset of Fig. 1b where the domain lines maintain a critical distance of 3.6 µm to 7.2 µm.

The effect of temperature on the formation of AOP domains was also investigated. The crystal/phase-mask assembly was placed on a hot plate, and exposures were performed at different temperatures ranging from room temperature up to 200°C. No qualitative difference was observed at temperatures below 100°C. However for higher temperatures the domain density was observed to be significantly reduced as domain lines tend to develop even further apart than in the room temperature case. A major difference as seen in Fig. 3 is that the domain lines are not composed of individual sections but instead are continuous lines. Fig. 3 shows an SEM micrograph of continuous domain lines resulting from illumination at a temperature of 154°C.

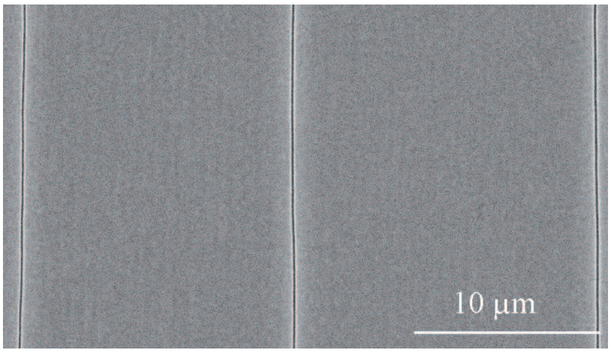

Figure 3. SEM image of a sample illuminated via a phase-mask at 154°C.

Conclusions

We have demonstrated an ordered alignment of AOP domains. The ordering was achieved through a spatially modulated UV light pattern UV laser radiation produced using a phase mask. The light-induced domains nucleate along the optical intensity maxima, and are further encouraged to grow along a specific y-direction of the crystal by a deliberate alignment of the periodic optical intensity pattern along that direction. A total replication of the light pattern was not achieved possibly because of an electrostatic repulsion between adjacent domains, which limits the minimum distance between them to few microns. Furthermore, for experiments performed at room temperature the ordered domain lines consist of smaller discrete domains, while at higher temperatures domain lines are continuous but tend to grow even further apart from each other. Experimental results suggest however that although long domain lines cannot grow in close proximity due to electrostatic limitations it would be possible to achieve denser packing of individual domains (domains which have just nucleated but not expanded) by illuminating with a 2D periodic intensity pattern.

The results presented here are an important first step towards successful manipulation and control of AOP ferroelectric domains in congruent lithium niobate.

References

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