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Paper number: 1398

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Optical control of electric-field poling in $LiTaO_3$

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Abstract

We present a room temperature technique for optically inducing periodic domain-inverted structures in bulk (0.2mm thick) $LiTaO_3$. By simultaneous application of an electric field and patterned illumination using UV wavelengths (351nm and 364nm) we demonstrate modulation of the resulting domain profile. We discuss the origins of the observed optical effect and describe our results from repeated domain switching, by cycling the electric field.

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Patterning of domain structures in ferroelectric materials has recently received attention both as a means of achieving novel transducer structures for acousto-optic applications, and a way of allowing phase matching of second-order nonlinear optical interactions. In particular, the technique of quasi phase matching (QPM) [1], which involves the periodic spatial modulation of a crystal's nonlinear optical coefficients by periodic domain inversion, has attracted considerable interest in crystals such as $LiNbO_3$ and $LiTaO_3$. To date, many post-growth techniques for achieving periodic domain profiles have been reported in the literature. These techniques comprise various combinations of heat treatment, electric fields and surface chemistry, and involve processes such as in-diffusion of impurities (such as Ti) [4], out-diffusion of LiO_2 [5], proton exchange of Li [6], and electron-beam irradiation [7], as well as the direct application of large electric fields via patterned electrodes [8]. All these methods require either specialist clean-room photolithographic fabrication of patterned electrodes, or lengthy thermal or electron-beam scanning processes. Experiments demonstrating the advantages of direct optical control of ferroelectric domain profiles have previously concentrated on photorefractive materials such as Ce:SBN [9]. Here we present details of a simple room temperature technique for patterning the domain profile of $LiTaO_3$ - a material which is considerably less susceptible to photorefractive damage - by the simultaneous application of optical and electrical fields without the need for photolithographic processing.

At room temperature (~25°C), $LiTaO_3$ is ferroelectric with crystal class 3m. The spontaneous polarisation, P_S, is 50µCcm^{-2} [10] and the nonlinear optical coefficient, d_{33}, is 26pmV^{-1} [11]. Both the direction of P_S and the sign of d_{33} are reversed easily by applying an electric field, along the crystallographic z-axis, in excess of the coercive field, E_C. We measured E_C to be ~225kVcm^{-1} in our samples. The field required to return the crystal to its original state is somewhat lower than this, and its precise value depends upon the time elapsed since the initial reversal [12, 13]. We measured ~120kVcm^{-1} after 2 minutes. The crystals used in our experiments were 0.2mm thick, z-cut optical grade wafers of Czochralski-grown material supplied by Mitsui Corporation, Japan.

Optical illumination was provided by the UV lines (predominantly 364nm and 351nm) of an Ar ion laser. Due to the wide band-gap of $LiTaO_3$, a reasonably small absorption of ~4cm^{-1} was measured at these wavelengths. Using cylindrical optics, the beam was launched into the wafer through the +y face which was polished to minimise beam distortion (see figure 1). The other crystal surfaces were otherwise untreated. A binary grating (mark-to-space ratio of 1:1) was placed in the beam path to allow a spot of elliptical Gaussian profile ($\phi_{\parallel z}$ ~150µm, $\phi_{\parallel x}$ ~3mm) superimposed with dark bands of period 280µm, to be projected onto the entrance face. The peak intensity was ~100Wcm^{-2}. The electric field was then applied to the crystal via liquid electrodes on the ±z-faces. Under computer control, the field was ramped slowly (at a rate of 0.5kVcm^{-1}s^{-1}) past E_C, and the displacement current arising from the polarisation switching was measured as a function of the applied voltage. The integrated area of this graph allowed the total switched charge to be monitored.

In order to assess the effect of the UV illumination during the poling process, the resulting structure was examined under a polarising microscope. As with $LiNbO_3$, stress birefringence adjacent to domain boundaries makes visualisation of the structure possible. In the unilluminated regions, uniform poling occurs. In regions exposed to UV, however, the switching appears to have been impeded. The contrast of this stress pattern becomes more pronounced as the crystal is switched repeatedly. The photograph in figure 2 shows the resulting structure, viewed along the z-axis, after five successive switches. The stress pattern clearly shows the influence of UV light during poling : the dark vertical bands (in the top half of the figure) coincide with the strips of UV illumination under the electrodes. The horizontal boundary indicates the edge of the electrode area. Structure resulting from the stresses of domain formation also appears to extend into the region of crystal outside the electrode area (in the lower half of figure 2) through which the UV also passed but no uniform electric field was applied. The saw-tooth appearance of this structure would seem intuitively consistent with the crystal's trigonal symmetry. It is clear from figure 2, however, that the most pronounced effect occurs under the electrode area in the regions of highest UV intensity towards the centre of the Gaussian beam. This spatial intensity profile allows us to approximate an intensity threshold, of about 50Wcm^{-2}, for the patterning effect. UV light polarised perpendicular and parallel to the crystal z-axis yielded qualitatively similar results. The apparent disruption of the domain switching is borne out by a reduction in the total switched charge measured during successive cycles when compared to a sample repeatedly switched without optical illumination. The resulting domain profile was also confirmed by etching the sample in an acid mixture HF:HNO$_3$ (ratio 1:2) for 6 hours at room temperature where preferential etching clearly indicates the domain orientation. The etched sample is photographed in transmitted unpolarised light in figure 3a. Etching at small cracks (due to stress) and defects is also apparent. Figure 3b shows the surface profile, under the electrode area, along the x-direction. The peaks correspond to the illuminated areas where switching has been impeded, and the direction of P_S has remained predominantly in the original crystal orientation after five switches. The underlying concave feature is due, we believe, to an enhancement of the overall etch rate at defects induced by the repeated switching in the region of the optically-induced domain disruption.

As the maximum temperature difference that can exist between light and dark fringes is only a few degrees [14] due to the low optical absorption, it is clear that laser-induced heating close to the crystal's phase transition temperature, T_C~938K, cannot account for any resulting modulation of the domain structure. Besides, crystal heating would be expected to enhance switching (rather than inhibit it, as observed here), by lowering E_C and making switching easier [15]. It seems probable that the effect occurs as a result of the interaction between the externally applied electric field and the internal electric field (arising from P_S and surface charge), which is itself a function of local temperature and intensity variations. Charge mobility near the surface and the pyroelectric effect both affect this internal field, as does the presence of thermally and photo-generated carriers. Although the crystal samples used in our experiments were nominally pure, the possibility of impurity inclusions of only a few ppm (which are known to enhance significantly the photovoltaic effect and the resulting space charge fields [16]) cannot be ruled out. Defects (such as vacancies) and local lattice distortions (such as those due to the inclusion of hydrogen during crystal growth [17]) would also complicate this internal field. Recent work by Chao *et al.* [13] would seem to confirm that photoinduced charges do indeed play a compensating roll. All these parameters, currently the subject of further research, influence the resulting internal electric field distribution which determines the domain orientation and stability. Further work is also underway to assess the resolution and flexibility of this optical patterning technique by tuning the illumination period by varying the angle of intersection of interfering coherent beams at both UV and visible wavelengths.

In summary, we have presented an optical technique for defining domain-inverted structures in bulk $LiTaO_3$. UV illumination of the crystal sample during electrical poling appears to impede domain switching, allowing an incident optical pattern to be replicated as a stable domain profile. This work was supported by the UK Engineering and Physical Science Research Council (EPSRC) under grant number GR/K28251.

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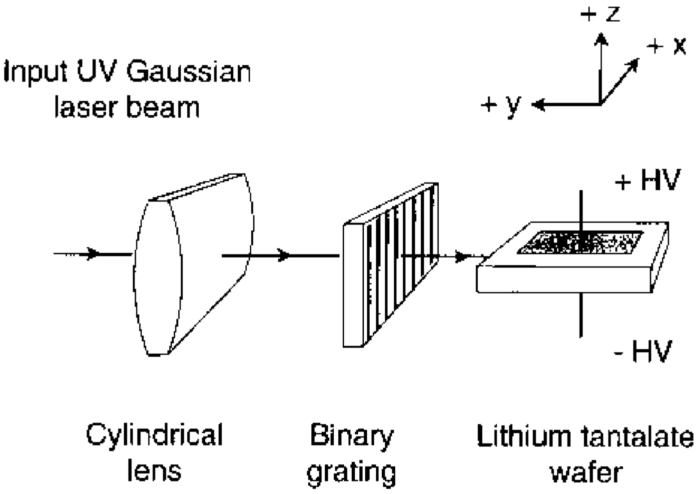
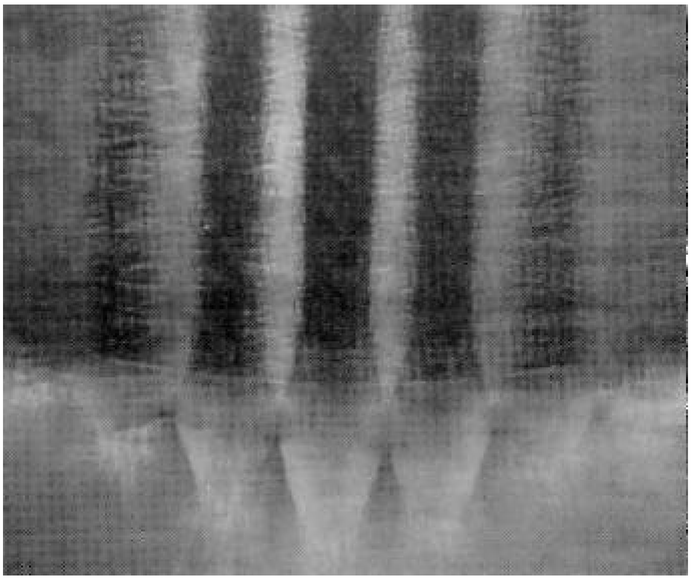
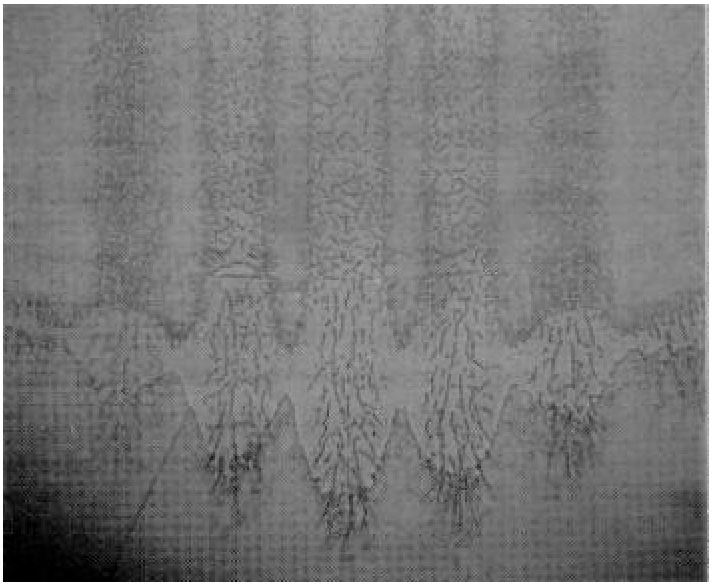
Figures



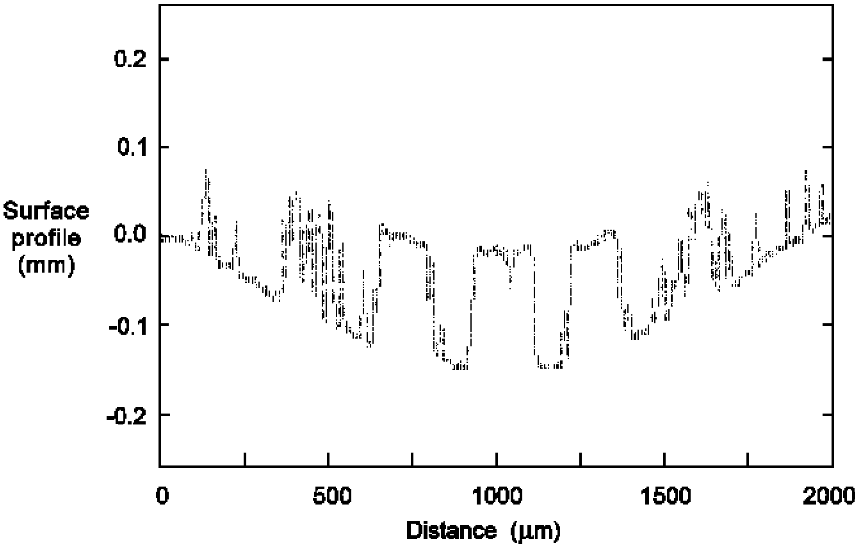
1. Schematic illustration showing experimental arrangement and crystal orientation. The UV beam is incident on the +y face of the $LiTaO_3$ sample and electrodes are applied to the ±z faces. The binary mask is in close proximity to the crystal.



2. UV-induced structure imaged through a polarising microscope. In the unilluminated regions, uniform poling occurs. The dark vertical bands (in the top half of the figure) coincide with the strips of UV illumination. The horizontal boundary coincides with the edge of the electrode.



3a. The UV-induced structure revealed by etching.



3b. Surface relief profile after etching.