

# UV laser direct writing of ferroelectric domain inverted structures in single crystal lithium niobate

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## Introduction

Ferroelectric domain engineering in lithium niobate (LN) is a subject of extensive research mainly for the fabrication of quasi-phase-matched (QPM) nonlinear optical devices but also for the improvement of linear devices and microstructuring. The most common method for ferroelectric domain engineering is by the application of an external electric field, higher than the coercive field ( $E_c$ ), at room temperature. In this method dielectric contrast is introduced by a layer of photoconductive polymer. The strength of the electric field is lower below the photopolymer covered areas hence spatial control over ferroelectric inversion is achieved.

In a modification of the E-field poling method intense laser radiation is used, instead of the photopolymer, during the application of the E-field to create the electric field contrast (most likely through a photoconductivity mechanism) for the spatial control of domain inversion. In this case of light assisted poling (LAP) the effective  $E_c$  in the irradiated areas of the crystal has been reported to drop in some cases by at least an order of magnitude<sup>[1,2]</sup>. The laser wavelengths which have been used in LAP are normally not absorbed by the crystal (800 nm–350 nm) and the inverted ferroelectric domains usually extend throughout the thickness of the crystal.

Here we present UV laser induced inhibition of ferroelectric domain inversion where spatially selective pre-exposure of the +z face of congruent LN samples inhibits domain inversion in this area upon the application of an external electric field. In these experiments the two steps of i) UV illumination and ii) E-field application are separated; the application of the external electric field can take place long after (days-months) after the UV illumination.

## Experiments

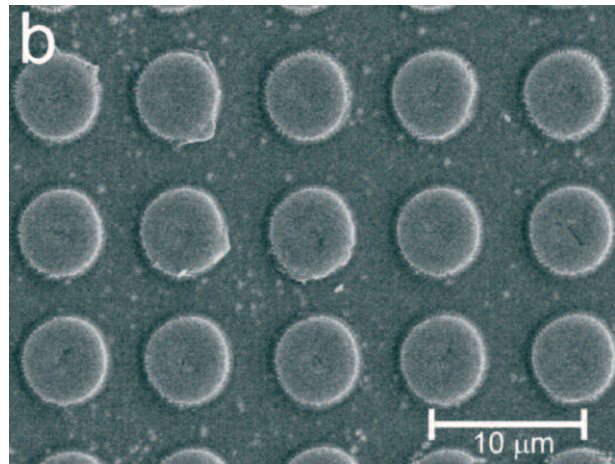
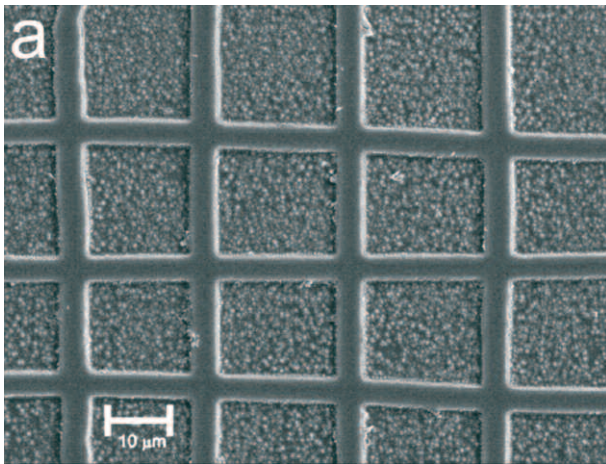
The experimental arrangement for the UV exposure consists of a set of computer controlled x-y-z translation stages on which the crystal sample is mounted and scanned in front of the focused UV laser beam. The laser source which was used in the experiments is a frequency doubled Ar: ion laser at  $\lambda=244$  nm. The laser beam was expanded and spatially filtered and finally focused using a plano-convex lens to a spot size of  $\sim 2$   $\mu\text{m}$ . A computer controlled mechanical shutter was also used to control the exposure

time. LN crystal samples were obtained from 3 inch diameter 500  $\mu\text{m}$  thick standard z-cut wafers purchased from crystal technology inc (USA). In the experiments presented here two types of exposures were performed always on the  $-z$  face of the samples: i) arrays of linear tracks where the crystal is translated linearly at a constant speed and the length of these tracks varied between 0.5mm to 5mm and ii) arrays of exposures where the crystal was static during the exposure the duration of which was controlled by the mechanical shutter.

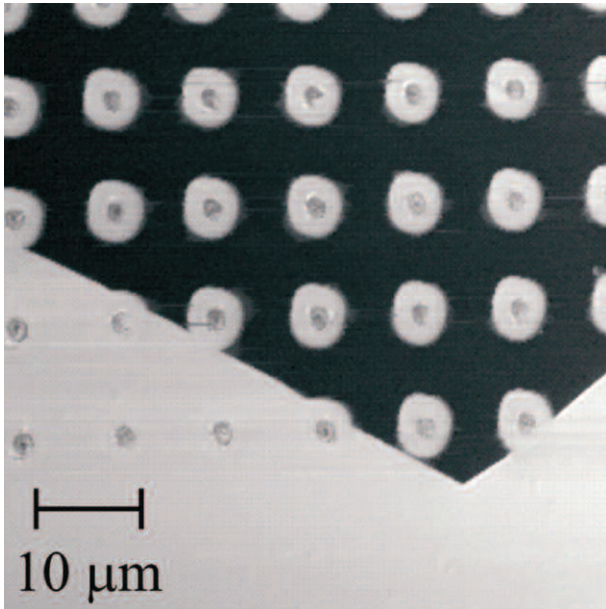
After UV illumination the samples were domain inverted by applying a uniform external electric field exceeding the  $E_c$  of the LN crystal ( $E_c \sim 22$  kV/mm). The domain inversion was performed in an arrangement similar to the one described in<sup>[3]</sup>. In this poling arrangement the LN crystal sample is mounted between two rubber O-rings and sandwiched between two transparent fused silica plates. Water held in the space between the silica plate and the crystal surface is used to provide electrical contact with the surface of the crystal. In this poling arrangement it is possible to monitor the domain inversion process using a CCD camera and a set of two linear polarizers. In order to verify the presence of inverted ferroelectric domains and to further investigate their topography in more detail using scanning electro microscopy (SEM) the samples were briefly etched ( $\sim 15$  minutes) in pure HF acid. The topography of such etched samples is shown in the SEM image in figure 1. Figure 1a shows an array of cross hatched linear tracks while figure 1b shows an array of dots which corresponds to static exposures.

The features (linear tracks and dots) which are shown in the SEM images in figure 1 correspond to the UV treated part of the surface and seem to have maintained its original domain orientation. The area surrounding these features is domain inverted and corresponds to a  $-z$  face. This is shown clearly in figure 1b where the dot features are surrounded by a hexagonal newly inverted domain.

Further investigation of these features was performed using piezo-response microscopy (PFM) which detects the displacement of the surface caused by the inverse piezoelectric effect and can in this way distinguish between opposite ferroelectric domain orientations. A PFM scan on a poled sample carrying sets of dot-type exposures confirmed the hypothesis that the features which were



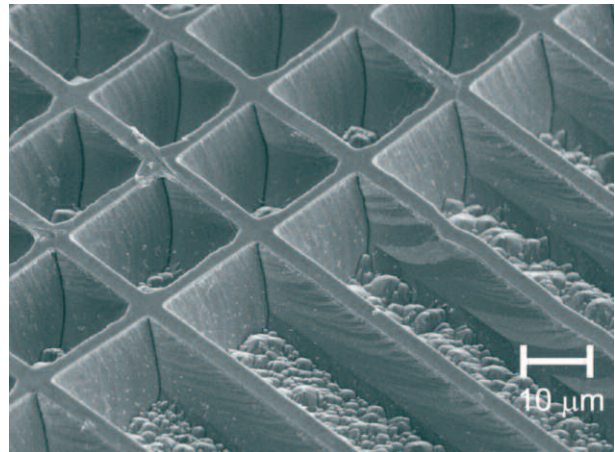
**Figure 1.** SEM images of the +z face of a UV exposed poled and briefly etched showing differential etch contrast in the corresponding UV exposed areas a) cross hatched linear pattern and b) array of dots.



**Figure 2.** Piezo-response map of the +z face of a UV dot-exposed and partially poled LN sample. The white dots correspond to the pole-inhibited UV exposed areas.

revealed by the acid indeed correspond to ferroelectric domains which maintained their original orientation. This is shown in the PFM scan which is presented in figure 2 where the newly inverted domain appears as black while the dots appear white and has the same contrast as the non-inverted area of the crystal which also appears white in the scan. The v-shaped contrast divide corresponds to the domain wall between the newly poled domain (black) and the original domain white. The dot-exposed array extends on both sides of the domain wall.

Interestingly the area in the centre of the exposed dot appears grey which indicates the presence of an amorphous layer possibly due to melting which occurs in the more intense central area of the Gaussian beam profile.



**Figure 3.** SEM image of a cross hatched pole inhibited pattern etched in HF acid for 39 hours.

### Discussions

In LAP experiments so far the consistent repeatable result is that the effective coercive field reduced under the influence of light. However, the results presented here show the opposite behavior, namely the effective coercive field increases in the UV exposed areas. The difference in the case which is presented here is that short wavelength UV radiation which is highly absorbed by the crystal is used. UV radiation at  $\lambda=244$  nm is absorbed in the first few 10s of nm<sup>[3]</sup> in LN and it is of course capable of producing photo-excited charge and heat. Our hypothesis is that the photo-excited charge diffuses through the affected volume and gets trapped in crystal defects (this is congruent material hence lithium deficient containing large amount of lithium vacancies) thus creating suitable electrostatic termination points for the propagating bulk domain which has nucleated on the original -z face (during the poling process) and propagates towards the original +z face. As a result of this process the surface and the volume of the crystal below the UV exposed areas maintain the original domain polarity.

The depth of this pole-resisting part of the surface can be determined by chemical etching of the y-face of the cross sections of the poled UV tracks. Observation of the PFM scan in figure 2 shows that the pole resisting domains on the right hand side have the same contrast as the bulk non-inverted domain on the left hand side of the image. The depth sensitivity of the PFM is of order  $\sim 100$  nm which is roughly 5 times the radius of curvature of the AFM tip which is used<sup>[4]</sup>. Any domain deeper than 100 nm will appear as bulk hence this sets a minimum depth for the features which we observe.

However, the depth of the pole inhibited area does not compromise the utility of this method for the fabrication of high aspect ratio surface relief structures because a surface domain layer, however thin, that produces a +z face to the HF acid will resist etching anyway. This is clearly demonstrated in figure 3 where an SEM image of a long etched cross hatched sample is shown.

### Conclusions

The exposure of the +z surface of congruent lithium niobate single crystals to UV laser radiation ( $\lambda=244$  nm) is shown here to inhibit domain inversion locally during the process of E-field poling. The effect was investigated by SEM of HF etched samples which showed differentially etched feature corresponding to the UV irradiated areas. The domain inhibition was further verified by piezo-response microscopy. PFM also provides a minimum depth of these pole resisting domains of  $\sim 100$  nm as deduced from the depth sensitivity of the instrument. UV induced poling inhibition is attributed to the redistribution of diffused photoinduced charges and subsequent trapping in crystal defect sites which provide electrostatic termination points for propagating bulk domains during the E-field poling process.

Depending on the depth of these domain structures they can be used in nonlinear waveguide devices however, this effect when combined with chemical etching provides a very versatile tool for the fabrication of surface micro/nano-structuring of lithium niobate such as ridges and gratings.

### References

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