Influence of ultraviolet illumination on the poling characteristics of lithium niobate crystals

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The poling characteristics of lithium niobate crystals under ultraviolet laser illumination are investigated. Light with a wavelength around 334 nm permanently decreases the coercive field of the crystal and is used to demonstrate optical control of domain patterning. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1606504]

Creating ferroelectric domain patterns is crucial for a broad range of applications, such as, e.g., nonlinear frequency conversion, photonic band-gap devices, electro-optic Bragg switches, and data storage.[1–4] Using electrodes patterned by optical lithography, periodically poled components with period lengths down to 3 μm were fabricated. Smaller structures, however, are difficult to achieve with present techniques, and thus alternative methods are under investigation. For lithium tantalate crystals ''optical periodic poling'' was reported.[5,6] This technique can directly transfer a light pattern into a domain structure using unstructured electrodes. The underlying mechanism is a light-induced transient change of the coercive field after a recent poling event. However, no similar effects were found for the more widely used lithium niobate crystals ($LiNbO_3$).[7] Recently photostructuring of domains in $LiNbO_3$ crystals highly doped with magnesium has been demonstrated.[8] In this letter, we present a method to control domain patterns in undoped $LiNbO_3$ crystals using high-intensity ultraviolet (UV) illumination with unstructured electrodes.

Figure 1 depicts the poling setup used in our experiments. The electric field is applied to the crystal with the help of transparent liquid electrodes. Together with fused silica windows, this allows for UV illumination by the unexpanded beam of an Ar^+ laser (beam diameter about 3 mm). Ferroelectric domains in $LiNbO_3$ can be inverted by applying an electric field along the c axis of the crystal, if this field exceeds the so-called coercive field (about -20 kV/mm for congruently melting $LiNbO_3$ crystals). This poling, however, is not instantaneous. It starts with the nucleation of domains which afterward grow by domain-wall movement.[9] During our poling experiments, the electric field is continuously increased at a rate of 30 V/(s mm) up to values well above the coercive field.

To get spatially resolved information about the poling process, the crystal holder is integrated into a Mach–Zehnder interferometer: In $LiNbO_3$ the orientation of a ferroelectric domain determines the sign of the electro-optic coefficient. Therefore, if a homogeneous electric field is applied, the sign of the electro-optic refractive index change depends on the orientation of the domain. When the orientation of the c axis is reversed, this leads to a noticeable discontinuity in the interference pattern.[7] By placing photodiodes in different parts of the interference pattern, this discontinuity can be used to precisely measure the field at which domain reversal occurs in different locations of the crystal. All $LiNbO_3$ crystals used are congruently melting undoped z cuts with a thickness of 0.5 mm (supplier: Crystal Technology Inc.).

A typical measurement of the coercive field versus the number of forward and backward poling cycles is shown in Fig. 2 for the forward poling direction. The sample is illuminated with light of the wavelengths $\lambda = 351$ nm (with the intensities $I = 3$ W/cm^2 and $I = 6$ W/cm^2) and $\lambda = 334$ nm ($I = 3$ W/cm^2) for eight to ten poling cycles each. The coercive field is measured inside the area illuminated by the laser beam (circles), as well as outside this region (triangles). There is always a 6 min waiting time between two poling processes to avoid transient effects.[7] Missing data points indicate that the interferometer could not clearly resolve the phase jump during poling.

The data show that illumination with the light of the wavelength $\lambda = 351$ nm changes the coercive field only temporarily, most probably because the crystal temperature increases due to absorption. Illumination at the wavelength $\lambda = 334$ nm, however, yields a strong decrease in the coercive field, which is still significant (more than 800 V/mm) even after 1 h without illumination. Light from the wavelength $\lambda = 333$ nm yields the same results. This effect is present only if the illumination takes place during the poling process. Illumination before or after the poling has no lasting impact on the coercive field.
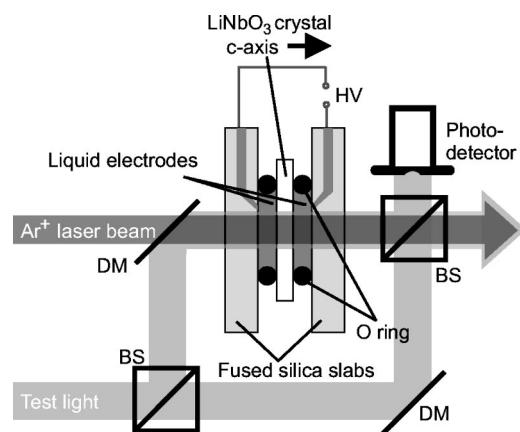

FIG. 1. Schematic depiction of the poling setup used for investigating the influence of light on the coercive field (DM: Dielectrical mirror, BS: Beam splitter, and HV: High voltage).
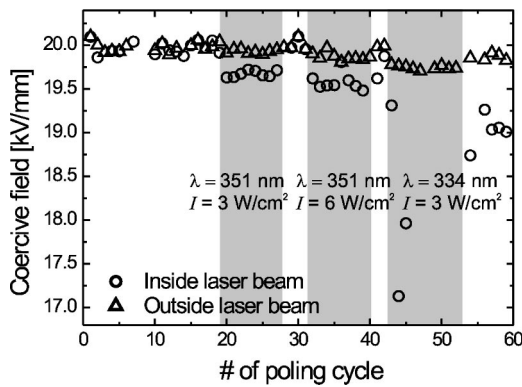
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FIG. 2. Coercive field vs number of poling cycles for the forward poling direction measured interferometrically inside (circles) and outside (triangles) of the illuminated area. Throughout the cycles marked in gray, the crystal is illuminated by a laser beam with wavelength λ and intensity I. After illumination with λ = 334 nm light, the coercive field is noticeably lowered.

The observed decrease of the coercive field by illumination with UV light is used to realize light-controlled domain patterning in LiNbO$_3$ crystals. A binary mask (mark-to-space-ratio 1:1, period length ≈1 mm) is inserted into the laser beam about 6 cm in front of the crystal. Since the mask is not imaged on the crystal through a lens, a Fresnel diffraction pattern appears with a fine structure much smaller than the structure of the mask (Fig. 3). The crystal is illuminated during four poling cycles with multiline UV light of an Ar$^+$ laser ($I = 55$ W/cm^2, thereof $I \approx 6$ W/cm^2 with λ ≤ 334 nm), with a waiting time between two subsequent poling processes of 200 s. The laser light is turned off, and a subsequent forward poling process is aborted immediately after the domains start to switch.

Selective etching of the crystal with hydrofluoric acid reveals the domain pattern as a surface topography. With a light microscope, domain structures showing the characteristic double lines from the light pattern [Fig. 3(b)] can be seen in the etched crystal. Therefore, the domain pattern is obviously correlated with the light pattern used for illumination. It also indicates that domain inversion indeed took place in the illuminated regions of the crystal. Figure 4 shows a magnified image obtained with an atomic force microscope (AFM) of the domain structures. The finer domain structure is indeed similar to that of the light pattern [Fig. 3(c)]. There
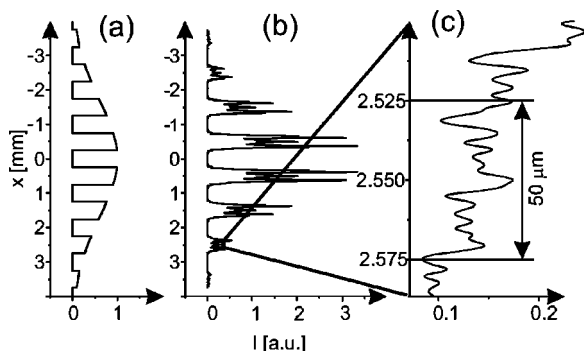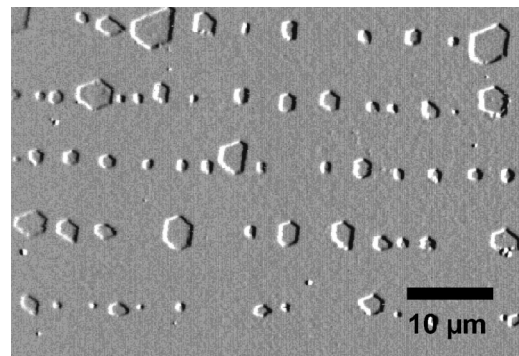

FIG. 4. AFM image of a photoinduced domain-patterned crystal after etching in hydrofluoric acid. A stripelike domain pattern is obvious.

is a small deviation from the calculated diffraction pattern since the multiline laser operation and the refraction due to the different refractive indices of the fused silica windows and the liquid electrodes were not taken into account.

The origin of the effect described here is still not completely clear. Relaxation effects of the coercive fields, like in LiTaO$_3$, can be ruled out. Thermal effects also cannot be the origin, since the coercive field change persists for hours after illumination. One can speculate that the intense UV radiation generates additional defects in the crystals which facilitate the domain-nucleation process and, therefore, enable the domain-wall movement to start at lower electric fields. This may also explain the lack of data points in Fig. 2 during the last process. If the UV light generates significantly more nucleation points, there are more but necessarily smaller domains during the early stages of the poling process, which are below the spatial resolution of the interferometric setup used.

Anyhow, illumination through the mask shows that this effect can be used for optical control of domain patterning. In accordance with Fig. 2, ferroelectric domains have started to grow in the illuminated areas where the coercive field is lowest. A stripe pattern with a period length of 8 μm can be observed (Fig. 4), indicating that the effect is indeed suitable for generating domain patterns on a micrometer scale. However, it is also obvious that the poling process was aborted prematurely before the domain nuclei could coalesce into a single stripe.

By optimization of the effect, one should be able to generate domain patterns on the micrometer scale utilizing an interferometrical light pattern and homogeneous electric fields. In doing so, it is especially important to time the aborting of the final poling process, so that the domains in the illuminated regions have time enough to coalesce, but before the domain growth extends into the unilluminated parts of the crystal.

In this letter, we showed that illumination of congruently melting LiNbO$_3$ crystals during a poling process with light of a wavelength of 334 nm changes the coercive field for subsequent domain reversals. By structured illumination, a domain pattern with 8 μm period length has been fabricated. The effect presented here, together with interferometric patterning, may allow one to realize periodically poled LiNbO$_3$ crystals with period lengths well below 3 μm. Such crystals are of great interest, e.g., for the generation of short-

FIG. 3. Schematic depiction of the generation of micrometer UV light patterns (wavelength λ = 334 nm) with a binary mask with a period length of 1 mm. Shown is the intensity distribution $I(x)$ of a Gaussian beam (a) directly behind the mask and (b) about 6 cm behind the mask. In the magnified portion (c) of the beam, a light modulation with a period length of about 8 μm is visible.

wavelength light, nonlinear photonic crystals, and electro-optically controlled Bragg light switches.

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