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Electrooptic properties of lithium niobate crystals for extremely high external electric fields

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ABSTRACT The electrooptic effect in lithium niobate crystals ($LiNbO_3$) for extremely high externally applied electric fields of up to 65 kV/mm is investigated. Homogeneous electrooptic refractive-index changes of up to 4.8×10^{-3} are found for ordinarily polarized light. No quadratic electrooptic effect is observed. An upper bound for the quadratic electrooptic coefficient of $|s_{13}| \leq 2.3 \times 10^{-21}\ \mathrm{m^2/V^2}$ is determined. Electrooptic, angular, and wavelength tuning of the Bragg condition of a thermally fixed hologram are demonstrated.

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1 Introduction

In the last few decades, crystalline lithium niobate ($LiNbO_3$) has become one of the most important ferroelectric materials because of its robustness, availability, optical quality, and, of course, its electrooptic and photorefractive properties. Thus, lithium niobate is widely used for various optical devices, like intensity and phase modulators, deflectors, switches, wavelength filters, and holographic data storage systems [1, 2].

In many cases, external electric fields are applied to the crystals to use the electrooptic effect to enhance the photorefractive properties, or to obtain information about the light-induced charge transport [3]. The influence of extremely high externally applied electric fields on the electrooptic properties is of special interest for electrooptic modulators and tunable wavelength filters, as well as for holographic data-storage systems that use electric field multiplexing. Petrov et al. investigated electric field multiplexing of reflection volume holograms in $LiNbO_3$, but only used small electric fields of up to 0.6 kV/mm [4].

In this work, we investigate the electrooptic properties of $LiNbO_3$ crystals for fields of up to 65 kV/mm. Furthermore, electrooptic tuning of a thermally fixed hologram is explored.

2 Theoretical considerations

We use holography as a precise method for studying the electrooptic properties of $LiNbO_3$. Before presenting the method itself, we first outline some fundamental considerations.

2.1 *Influence of an external electric field on the refractive-index changes*

The amplitude of the refractive-index changes Δn is given by

$$\Delta n = -\frac{1}{2} n_0^3 \left(r_{13} E_3 + s_{13} E_3^2 \right) . \tag{1}$$

Here, $n_0 = 2.3486$ [5] is the refractive index for ordinarily polarized light (wavelength $\lambda = 488\,\mathrm{nm}$), and $r_{13} = 11.3\,\mathrm{pm/V}$ [5, 6] is the corresponding linear and s_{13} the corresponding quadratic electrooptic coefficient, using contracted indices. The electric field E_3 is a superposition of the externally applied electric field E_{ext} and a sinusoidal space-charge field $E_{SC} = E_{SC,1} \cos(Kz)$ that might be present for holographic recording with two plane waves. The total field is

$$E_3 = E_{ext} + E_{SC,1} \cos(Kz) , \tag{2}$$

where $E_{SC,1}$ is the amplitude of the space-charge field. Inserting this field into (1), the refractive-index change can be written as

$$\Delta n = \Delta n_0 + \Delta n_1 \cos(Kz) + \Delta n_2 \cos(2Kz) , \tag{3}$$

with

$$\Delta n_0 = -\frac{1}{2} n_0^3 \left(r_{13} E_{ext} + s_{13} E_{ext}^2 + \frac{1}{2} s_{13} E_{SC,1}^2 \right) ,$$
$$\Delta n_1 = -\frac{1}{2} n_0^3 \left(r_{13} E_{SC,1} + 2\, s_{13} E_{ext} E_{SC,1} \right) , \quad \text{and}$$
$$\Delta n_2 = -\frac{1}{4} n_0^3\, s_{13} E_{SC,1}^2 .$$

For an elementary volume phase hologram, Δn_0 contains two components that yield a so-called Bragg-shift that is induced by an external electric field:

$$\Delta n_{0,E_{ext}} = -\frac{1}{2} n_0^3 \left(r_{13} E_{ext} + s_{13} E_{ext}^2 \right) . \tag{4}$$

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The influence of an electric field on the amplitude of a refractive-index grating is given by

$$\Delta n_{1,E_{\text{ext}}} = -n_0^3 s_{13} E_{\text{ext}} E_{\text{SC},1} \,. \tag{5}$$

2.2 *Piezoelectric influence on a holographic grating*

The grating period Λ of an elementary hologram in a piezoelectric crystal like $LiNbO_3$ can be changed by an externally applied electric field. If the field is applied parallel (or antiparallel) to the ferroelectric axis of the $LiNbO_3$ crystal ($E_1 = E_2 = 0$, $E_3 \neq 0$), and if the holographic grating is also oriented parallel to this axis ($K \parallel c$), the relative change of the grating period along this axis is determined by the 3-component of the stress tensor S:

$$S_3 = \frac{\Delta\Lambda}{\Lambda} \,. \tag{6}$$

For the calculation of the component S_3, we use the following relation between the stress tensor S and the strain tensor T of a piezoelectric crystal [7]:

$$T_{ij} = C_{ijkl} S_{kl} - e_{kij} E_k - \frac{1}{2} l_{klij} E_k E_l \,, \tag{7}$$

where C is the stiffness tensor, e is the linear and l the quadratic piezoelectric tensor, and E is the amplitude of the electric field. In the following, all tensor elements are written with contracted indices. Assuming that the crystal is free, i.e., no mechanical pressure is applied to the crystal, the strain tensor is zero, and numerical analysis of the set of equations shows that the contribution of the quadratic piezoelectric effect is negligible when external electric fields of up to 65 kV/mm are applied. Thus, an excellent approximation for the stress tensor element S_3 is

$$S_3 = \frac{e_{33}}{C_{33}} E_3 \,. \tag{8}$$

With $C_{33} = 2.357 \times 10^{11}\,\text{N/m}^2$ and $e_{33} = 1.785\,\text{C/m}^2$ [6], we get

$$S_3 = 7.57 \times 10^{-12}\,\frac{\text{m}}{\text{V}} E_{\text{ext}} \,. \tag{9}$$

2.3 *Change of the Bragg angle due to an external electric field*

The Bragg condition for a volume phase hologram in the reflection geometry is

$$\Theta_{\text{B}} = \arccos\left(\frac{\lambda}{2n_0\Lambda}\right) \,, \tag{10}$$

where Θ_{B} is the angle between the propagation direction of the light inside the crystal and the surface normal of the sample in the case of Bragg incidence and λ is the wavelength of the light in vacuum. The total differential of the Bragg condition yields

$$\Delta\Theta_{\text{B}} = \frac{\partial\Theta_{\text{B}}}{\partial n_0}\Delta n_0 + \frac{\partial\Theta_{\text{B}}}{\partial\Lambda}\Delta\Lambda \,. \tag{11}$$

Refractive-index changes Δn_0 and changes of the fringe spacing $\Delta\Lambda$ can be caused by the electrooptic and the piezoelectric effect, respectively. From (6) and (10), it follows that

$$\Delta\Theta_{\text{B}} = \frac{\lambda}{2n_0\Lambda\sqrt{1-\left(\frac{\lambda}{2n_0\Lambda}\right)^2}}\left(\frac{\Delta n_0}{n_0} + S_3\right) \,. \tag{12}$$

Thus, the Bragg-shift, which is due to an externally applied electric field, is caused by $\Delta n_{0,\text{Ext}}$ and $S_3(E_{\text{ext}})$, given by (4) and (9), respectively.

3 Experimental methods

A congruently melting iron-doped $LiNbO_3$ crystal with an iron concentration of $c_{\text{Fe}} = 18 \times 10^{24}\,\text{m}^{-3}$ (0.05 wt. % Fe_2O_3) was used for the investigations of the electrooptic material properties. The densities of filled and empty traps, $c_{\text{Fe}^{2+}}$ and $c_{\text{Fe}^{3+}}$, were deduced from absorption spectroscopy [8]. The ratio $c_{\text{Fe}^{2+}}/c_{\text{Fe}^{3+}}$ was 0.008, and the dimensions of the sample were $a \times b \times c = 10.0 \times 10.0 \times 0.22\,\text{mm}^3$. An elementary hologram (a sinusoidal holographic grating) with a fringe spacing of $\Lambda = 105$ nm was recorded and thermally fixed [9, 10].

The crystal was mounted on a rotation stage to control the angle of incidence. The ordinarily polarized light of an Ar^+-laser (wavelength $\lambda = 488$ nm, light intensity of only $I = 0.55\,\text{W/m}^2$ to prevent dynamic effects) was partly diffracted. The intensities, I_{t} and I_{d}, of the transmitted and diffracted beams were measured. The angular dependence of the ratio $I_{\text{d}}/(I_{\text{t}} + I_{\text{d}})$, which determines the diffraction efficiency η, is called the selectivity curve. The maximum value of this ratio is observed for Bragg incidence (angle Θ_{B}), and the amplitude of the refractive-index changes Δn_1 can be deduced from η using Kogelnik's formula for the reflection geometry [11]:

$$\eta = \tanh^2\left(\frac{\pi\,\Delta n_1\,d}{\lambda\cos\Theta_{\text{B}}}\right) \,, \tag{13}$$

where d is the thickness of the crystal. Although the crystal used was only 0.22 mm thick, this formula for a volume phase

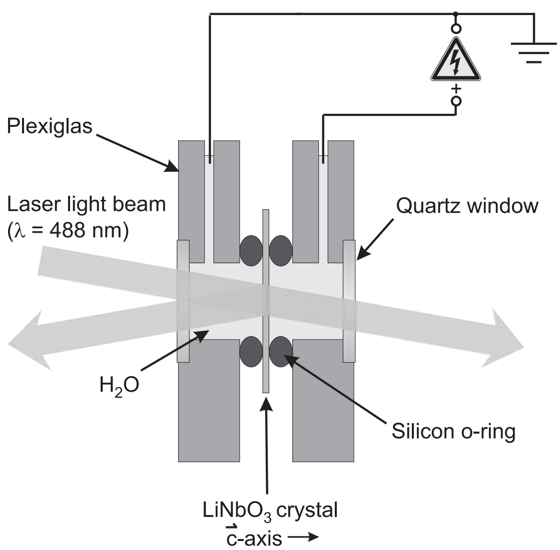

FIGURE 1 Schematic drawing of the crystal holder used

hologram is fully applicable because of the short period length ($\Lambda = 105$ nm) of the reflection hologram.

The crystal was placed in a Plexiglas holder (see Fig. 1). The surfaces perpendicular to the *c* axis are contacted with liquid and transparent electrodes. The selectivity curves were measured with externally applied electric fields of up to 65 kV/mm, with the positive voltage applied to the negative dipole end of the crystal. If larger fields were applied, there would be a risk of electric breakdown that could destroy the sample. This occurs typically between 70 and 80 kV/mm. An external electric field in the opposite direction would reverse the direction of the ferroelectric *c* axis of the crystal.

4 Experimental results

Application of an external electric field causes a shift of the Bragg angle. Figure 2 shows the selectivity curves for different externally applied electric fields. A shift of the Bragg angle $\Delta\Theta_B$ of $-0.55°$ was observed for an external electric field of 65 kV/mm, as shown in Fig. 3. Note that this is the angle inside the material. This Bragg-shift corresponds to an electrooptic tuning range of $\Delta\lambda = 0.75$ nm for the blue light ($\lambda = 488$ nm). This external field of 65 kV/mm caused a homogeneous refractive-index change of $\Delta n_0 = -4.8 \times 10^{-3}$. Using other crystal cuts or geometries [4], it may be possible to extend the tuning range further for the same external electric fields.

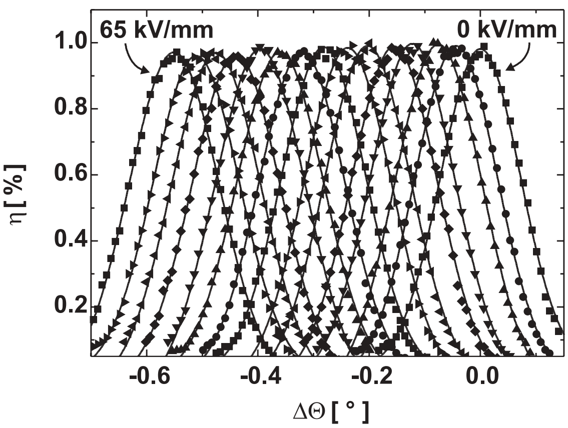

FIGURE 2 Selectivity curves for different external electric fields (0–65 kV/mm): diffraction efficiency η vs. incident angle $\Delta\Theta$. The *solid curves* are guides for the eye

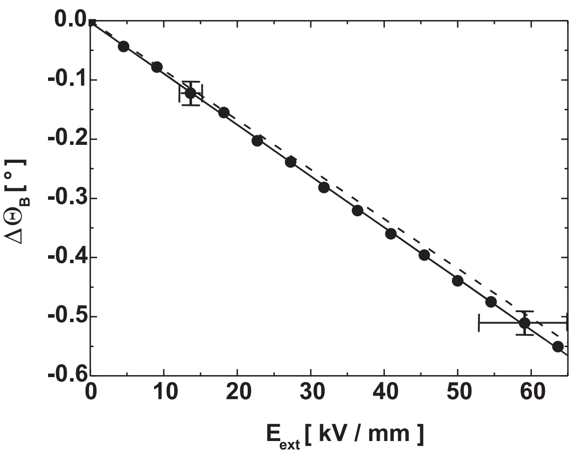

FIGURE 3 Shift of the Bragg angle $\Delta\Theta_B$ vs. external electric field E_{ext}. The *solid curve* is a linear fit to the experimental data, and the *dashed curve* shows the calculated dependence (see (12))

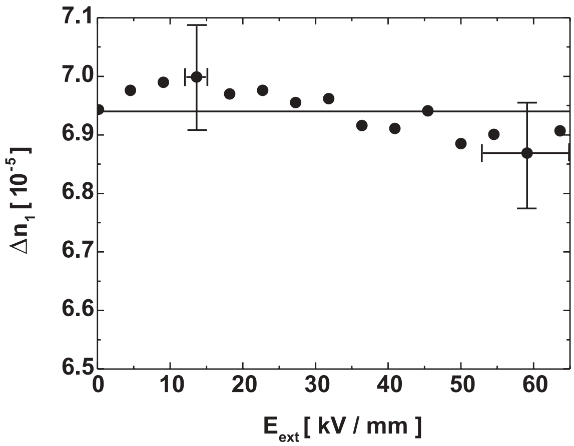

FIGURE 4 Amplitude of the refractive-index grating Δn_1 vs. external electric field E_{ext}. The *solid line* corresponds to the average value

Besides the Bragg angles, the absolute values of the hologram diffraction efficiencies were also evaluated. Figure 4 shows the amplitude Δn_1 of the refractive-index grating versus the externally applied field. The amplitude Δn_1 is almost constant. It varies up to 1.4×10^{-6}, but this is within the measurement uncertainties.

5 Discussion

If a non-negligible quadratic electrooptic effect is present in $LiNbO_3$, the Bragg angle would show a nonlinear behavior with respect to the external electric field. This is not observed. The experimental data can be described excellently by considering only the linear electrooptic effect and the piezoelectric effect, as it is shown by the dashed line in Fig. 3.

Another indicator of a non-negligible quadratic electrooptic coefficient would be a dependence of the amplitude Δn_1 of the refractive-index grating on E_{ext}. The expression (5) predicts a linear relationship between Δn_1 and E_{ext} if a quadratic electrooptic coefficient must be taken into consideration. Although our experimental data shows a moderate linear dependence of Δn_1 on E_{ext} (Fig. 4), the effect is comparable to the noise level, and we can only specify an upper limit for the quadratic electrooptic coefficient (see (5)): $|s_{13}| \leq 2.3 \times 10^{-21}\ \mathrm{m^2/V^2}$.

Thus, for external electric fields of up to 65 kV/mm, no quadratic electrooptic effect has to be considered.

For ordinarily polarized light, we observed electrooptic refractive-index changes of $\Delta n_0 = -4.8 \times 10^{-3}$ (wavelength $\lambda = 488$ nm). The corresponding value for extraordinarily polarized light would be $\Delta n_0 = -1.3 \times 10^{-2}$, taking into account the larger electrooptic coefficient r_{33} [5].

The findings are of practical importance: electrooptic phase modulators, tunable wavelength filters, and fieldmultiplexed holograms can benefit from the fact that $LiNbO_3$ crystals allow the application of external fields as large as 65 kV/mm while still showing a linear electrooptic response.

6 Conclusions

We have observed homogeneous electrooptic refractive-index changes of up to $\Delta n_0 = -4.8 \times 10^{-3}$ (ordinarily polarized light, wavelength $\lambda = 488$ nm, external field $E_{ext} = 65$ kV/mm). This is of special interest, for example, for compact electrooptic modulators, tunable holographic wavelength filters, and field multiplexing of holograms.

Even when an external electric field of up to 65 kV/mm was applied to $LiNbO_3$ crystals, no significant quadratic electrooptic effect was observed. An indication of a weak quadratic electrooptic effect was found by measuring a field-induced change of the amplitude of a refractive-index grating. This sets an upper limit for the quadratic electrooptic coefficient of $|s_{13}| \leq 2.3 \times 10^{-21}\ \mathrm{m^2/V^2}$.

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