Optical nonlinearity of pure lithium niobate crystals and hologram recording at low temperatures

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The photoinduced change in the refractive index increases sharply when nominally pure lithium niobate crystals are cooled to liquid-nitrogen temperature. Study of the diffraction by photoinduced gratings shows that the reason for the sharp increase is the formation of space charge by a photovoltaic effect.

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1. This is a first report of a study of the optical nonlinearity of ferroelectric crystals at low temperatures. In nominally pure recrystallized lithium niobate crystals we have found a substantial increase in the nonlinearity (by more than an order of magnitude). At room temperature, unfocused beam from a helium-cadmium laser (beam diameter ~3 mm, power level of 10 mW) can pass through a 3-mm $LiNbO_3$ crystal essentially without distortion. When the crystal is cooled, the beam divergence increases sharply, and in the far zone there is a preferential increase in the dimension of the spot along the polar axis of the crystal (Fig. 1). There is also a pronounced anisotropic photoinduced scattering, which absorbs much of the light incident on the crystal.

Changes also occur in the nature of hologram recording. There is a sharp increase in the sensitivity of the crystal as a recording medium, and the diffraction efficiency of the resulting holograms increases.

2. The pronounced nonlinearity of wide-gap electrooptic crystals results from

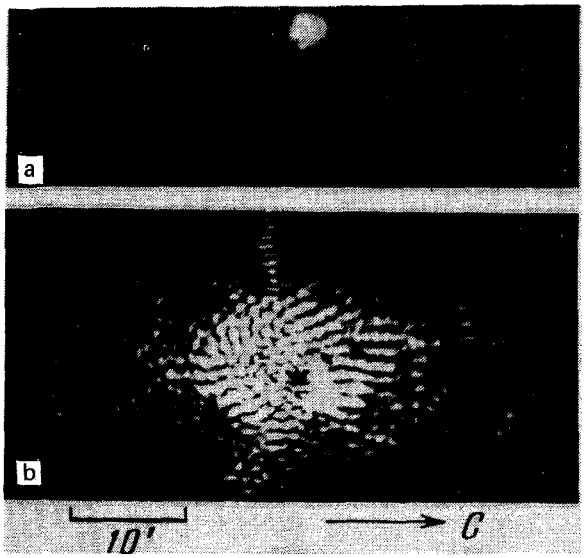

FIG. 1. Angular distribution of an unfocused laser beam after it has passed through a nominally pure recrystallized $LiNbO_3$ crystal. a—At 300 K; b—at 77 K.

the formation of an inhomogeneous space charge during illumination. The field of this charge modulates the refractive index. The magnitude of the space-charge field is determined by the charge-separation mechanism; the separation may result from the application of an external field to the crystal, from the pyroelectric charge at the ends of a broken crystal, from diffusive charge separation, or from the volume photovoltaic effect.[1]

At room temperature in recrystallized crystals, the hologram is recorded primarily through a diffusive separation of charge. The space-charge field is equal to the diffusion field,

$$E_T = \frac{2\pi}{\Lambda} \frac{kT}{e}, \tag{1}$$

where Λ is the scale dimension of the inhomogeneity of the light spot.[2] For an unfocused laser beam, and at room temperature, the field E_T is a few volts, in agreement with the fact that the beam is not distorted as it passes through the crystal (Fig. 1a). As the temperature is lowered, the field E_T should become even weaker, but this conclusion is in qualitative contradiction of experiment (Fig. 1b).

During the recording of holograms it was found that there is no energy exchange between beams of equal intensity, despite the increase in the diffraction efficiency. This result suggests that the nonlinear response is of a local nature at low temperatures, in contrast with the nonlocal diffusive response at room temperature.[2]

All these results show that as the temperature is lowered there is a change in the mechanism for the charge separation and thus for the photorefraction.

3. To determine the mechanism for the charge separation, we used a modified version of the method of "forced" light scattering[3,4] to measure the effective electric

field acting on the photoexcited carriers.

A dynamic grating was recorded by two beams; the beams filled the entire cross section of the crystal and thus eliminated the formation of large-scale optical inhomogeneities which would distort the wavefront, as shown in Fig. 1b. We measured the kinetics of the increase in the diffraction efficiency during the recording and the "non-Bragg" erasure[5] of a grating. These measurements were carried out under conditions such that the hologram efficiency was no more than a few percent, so that we could ignore the effects of photoinduced light scattering, which are seen at large phase shifts.

The diffraction efficiency increased quadratically with the time during the recording and decayed exponentially during the non-Bragg erasure, so that the following approximate expressions can be used for these measurements[6]:

$$\eta(t)_{\text{rec}} = \frac{4m}{(m+1)^2}\left\{-\frac{\pi r n^3 z E}{2\lambda\cos\theta}\left[1-\exp\left(-t/\tau_M\right)\right]\right\}^2, \tag{2}$$

$$\eta(t)_{\text{read}} = \eta_0 \exp\left(-2t/\tau_M\right), \tag{3}$$

where m is the intensity ratio of the recording beams, 2θ is the angle at which these beams meet, $\tau_M = (\epsilon\epsilon_0/\sigma)$ is the Maxwellian charge-relaxation time, and z is the thickness of the crystal.

Figure 2 shows the effective field E and the relaxation time τ_M for three fixed temperatures.

We see that the effective field increases by a factor of 35 when the sample is cooled from room temperature to liquid-nitrogen temperature, reaching ~7.5 kV/cm

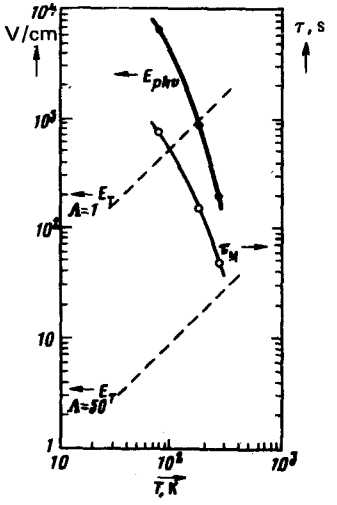

FIG. 2. Temperature dependence of the effective photovoltaic field E_{phv} and of the space-charge relaxation time τ_M. Dashed lines–Calculated diffusion field for an interference pattern with a period $\Lambda = 50$ μm, in accordance with the present experiments, and for a field with $\Lambda = 1$ μm.

and thus explaining the increase in the nonlinearity (Fig. 1). At the same time, the photoconductivity of the crystal decreases significantly.[1)]

4. The correlation between the temperature dependence of E and that of σ suggests that the reason for the drift of the photoexcited carriers is the anomalous photovoltaic effect.[6] This possibility is strengthened by the fact that the photoinduced change in the refractive index is of a local nature, $\Delta n(r) \sim I(r)$, and by the fact that charge is transferred only in the direction along the spontaneous-polarization axis.

The effective photovoltaic field is given by[6]

$$E_{phv} = \frac{k\alpha I}{\sigma_d + \sigma_{ph}}, \tag{4}$$

where α is the absorption coefficient of the crystal, k is the Glass constant, I is the intensity of the bias illumination, σ_d is the dark conductivity of the crystal, and σ_{ph} is the photoconductivity ($\sigma_d \ll \sigma_{ph}$). From independent measurements of σ_{ph} and E_{phv} we found that the temperature-induced changes in $k\alpha$ do not exceed a factor of 3.5 in the specified temperature interval. From the value $\alpha = 1.5$ cm^1 we can calculate the Glass constant; its value at room temperature turns out to be $k = 1.5 \times 10^{-9}$ A $\cdot$ cm^2/W, in good agreement with data in the literature.[8] The temperature dependence of the Glass constant can also be determined. Results of this type, however, are of extremely dubious value, since even at room temperature the ratio of the active and inactive absorption components at the working wavelength is not known exactly (and may, furthermore, depend on the temperature).

In summary, the reason for the change in the mechanism for the optical nonlinearity of $LiNbO_3$ during cooling is that the redistribution of the space charge by thermal diffusion becomes ineffective, while the photovoltaic charge transfer, in contrast, increases sharply, primarily because of an increase in the resistance of the crystal.

These results confirm that nominally pure lithium niobate crystals contain an uncontrolled impurity which may be involved in a photoexcitation, accompanied by the appearance of a photovoltaic current.[1]

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[1)]In the calculation of σ it was assumed, in accordance with Ref. 4, that the value of ϵ remains essentially constant over the working temperature range.

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