Spatially oscillating photovoltaic current in iron-doped lithium niobate crystals

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A new mechanism for space-charge formation, which is attributed to the nondiagonal components of the photovoltaic-effect tensor, has been observed in experiments on recording dynamic phase gratings by beams with a crossed polarization in iron-doped lithium niobate crystals.

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1. One mechanism of photorefraction in ferroelectric crystals is due to the appearance of a steady-state current when a short-circuited crystal is illuminated (photovoltaic effect).[1,2] The theory of the effect, which was developed in Refs. 3 and 4, relates the photovoltaic current to the electric-field intensity of the light wave by means of a third-rank tensor

$$j_i^{ph} = \beta_{ijk} E_j E_k , \ \beta_{ijk} = \beta_{ikj}^* . \tag{1}$$

In the first experiments[1] on recording holographic gratings in $LiNbO_3$:Fe crystals two light beams were used with ordinary polarization, and the wave vector of the grating **K**, which is equal to the difference of the wave vectors of the interacting beams $\mathbf{K} = \mathbf{k}_1 - \mathbf{k}_2$, was parallel to the polar C axis. In this case the photovoltaic current was directed along the spontaneous-polarization axis, and its magnitude varied in accordance with the intensity distribution in the interference pattern with a proportionality constant β_{33}.

A recording was also obtained with beams with ordinary polarization for $K \| C$ (β_{32} component)[5] and for $K \perp C$ (β_{22} component).[6] In all these cases, uniform illumination of the crystal produces a uniform photovoltaic current, and the grating recording is due only to the spatial modulation of the light intensity.

In this paper we are presenting data on the recording of a dynamic grating in $LiNbO_3$:Fe crystals by beams with a crossed polarization (ordinary and extraordinary beams, $K \perp C$). The achievement of such a recording is the first experimental proof of the existence of spatially oscillating currents in $LiNbO_3$:Fe, which are determined by the nondiagonal β_{15} components of the photovoltaic tensor. It has been established from the recording efficiency that the β_{15} component is close in order of magnitude to the strongest diagonal component β_{33}.

2. When the light wave

$$E = e_0 E_0 \exp(i\mathbf{k}_0\mathbf{r}) + e_e E_e \exp(i\mathbf{k}_e\mathbf{r}) \tag{2}$$

acts on an $LiNbO_3$ crystal ($\mathbf{e}_0$ and $\mathbf{e}_e$ are the polarization unit vectors, and E_0 and E_e

are the complex amplitudes of the ordinary and extraordinary waves), an oscillating current appears in the crystal in a direction perpendicular to the C axis[4] (Fig. 1a)

$$j^{ph} = \beta^{c}_{15} E_0 E_e \cos(\mathbf{Kr}) + \beta^{a}_{15} E_0 E_e \sin(\mathbf{Kr}) \tag{3}$$

$$= \sqrt{(\beta^{c}_{15})^2 + (\beta^{a}_{15})^2}\; E_0 E_e \cos[\,\mathbf{Kr} - \mathrm{arc\,tg}(\beta^{c}_{15}/\beta^{a}_{15})\,],$$

This current consists of two parts, which are attributed to the symmetrical and antisymmetrical components of the photovoltaic tensor.

This current results in the appearance of a steady-state space-charge grating with a wave vector **K**. The space-charge field E_{sc} determines the modulation depth of the index of refraction

$$E_{sc} \cong j^{ph}/\sigma_{ph}, \tag{4}$$

where σ_{ph} is the photoconductivity of the crystal, which is much greater than the dark conductivity.

The resulting grating can be detected from the diffraction of one of the recording beams; the polarization of the diffracted radiation in this case must be orthogonal to the reading beam (Fig. 1b).

3. In the experiment we used $LiNbO_3$ crystals with 0.03 wt.% Fe; they were X-cut crystals with dimensions of $X{:}Y{:}Z = 10{:}10{:}3$ mm^3. The radiation beam of an *He-Cd* laser was split into two by means of a birefringent crystal and were cross polarized; these beams then converged symmetrically in the crystal, as depicted in Fig. 1.

A three-dimensional phase grating was recorded in the crystal. The polarization of the diffracted radiation was orthogonal to the reading beam; no diffraction was observed if the polarization plane of the reading beam was rotated by 90°. This shows

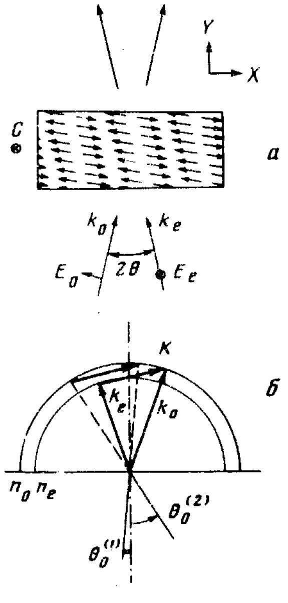

FIG. 1. (a) Experimental setup. Propagation directions of the photovoltaic current are indicated by arrows in the crystal. (b) Vector diagram of the interaction being examined. In addition to anisotropic diffraction (wave vectors are represented by solid arrows) isotropic Bragg diffraction (wave vectors are denoted by dashed arrows) is also possible on the recorded grating.

that the observed effect indeed results from a redistribution of the space charge because of spatially oscillating currents.

The efficiency of the recorded gratings was quite low compared with that obtained from the recording by beams with identical polarization. We must therefore show that this effect is not a consequence of anisotropic diffraction[6] by a weak grating that is recorded by beams with an identical polarization (because of depolarization by elements of the experimental setup and imprecise crystal orientation).

The normal isotropic diffraction (without polarization rotation) by the grating, which is recorded by beams with orthogonal polarization when the incidence angles of the reading beam are specially chosen, can be detected in a control experiment. The wave vector of the grating, which is recorded in the symmetrical scheme, is inclined with respect to the crystal surface by an angle

$$\psi = \frac{1}{2}\left[\sqrt{(n_0/\sin\theta)^2 - 1} - \sqrt{(n_e/\sin\theta)^2 - 1}\right]. \tag{5}$$

In this case the conditions of normal Bragg diffraction for ordinary waves at angles $\theta_0^{(1,2)}$ (see Fig. 1b)

$$\theta_0^{(1,2)} = \arcsin\left\{ n_0 \sin\left[\arcsin\frac{|\mathbf{K}|\lambda}{4\pi n_0} \pm \psi\right.\right\} \tag{6}$$

can be satisfied over a certain interval of recording angles.

Figure 2 shows the calculated dependences of ψ and $\theta_0^{(1,2)}$ on the covergence half-angle θ of the recording beams and the experimental points corresponding to the diffraction angles of the ordinary wave. An excellent agreement with calculation shows that in our experiment the grating recording is indeed determined by the nondiagonal β_{15} component of the photovoltaic tensor.

3. The diffraction efficiency of the dynamic grating in the initial recording stage is given by

$$\eta = \frac{I_g}{I_0} \approx \left(\frac{\pi z n^3 r E_{sc}}{\lambda \cos\theta}\,\frac{t}{\tau}\right)^2, \tag{7}$$

where I_0 and I_g are the intensities of the reading and diffracted radiation, and τ is the

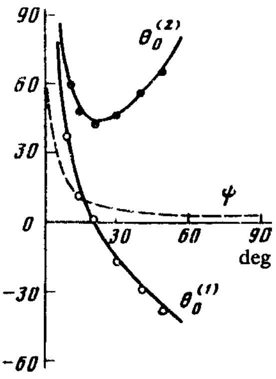

FIG. 2. Dependences of the isotropic Bragg diffraction angles $\theta_0^{(1,2)}$ and of the inclination angle ψ of the grating wave vector with respect to the crystal faces on the convergence half-angle θ of the recording beams.

Maxwellian charge relaxation time. In the traditional recording $r_{33} = 3.08 \times 10^{-9}$ cm/V and $E_{sc} = \beta_{33} I_3/\sigma_{\rm ph}$ appear in Eq. (7), and in our case $r_{51} = 0.28 \times 10^{-9}$ cm/V and $E_{\rm sc} = \sqrt{(\beta^c_{15})^2 + (\beta^a_{15})^2}\sqrt{I_0 I_e/\sigma_{\rm ph}}$. Therefore, after measuring the intensity of the diffracted radiation for identical recording and reading conditions we find

$$\left(\sqrt{(\beta^c_{15})^2 + (\beta^a_{15})^2} / \beta_{33}\right) \approx (r_{33}/r_{51})(I^{\perp}_g/I^{\parallel}_g)^{1/2}\ . \tag{8}$$

For a fixed exposure time of the order a several seconds ($t \ll \tau$) the intensity of the diffracted radiation in the scheme of Fig. 1 was 2×10^{-3} of the value for normal recording. It follows from this that $\sqrt{(\beta^c_{15})^2 + (\beta^a_{15})} \approx 0.25\beta_{33}$.1)

The imaginary components β^c_{15} and real β^c_{15} components cannot be determined separately from the diffraction-efficiency measurements.2) Information about the imaginary component can be obtained, in principle, from a measurement of the energy transfer between the recording beams.[7] For the conditions of our experiment the total diffraction efficiency was too small to observe the energy transfer. This can presumably be accomplished later by using thicker crystals.

In summary, we can conclude that a spatially oscillating photovoltaic current with an amplitude of the same order of magnitude as the known scalar photovoltaic current[1] is excited in iron-doped lithium niobate crystals which are stimulated by beams with a crossed polarization.

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1)The experimental recording was performed by using finite-dimension beams, and nonlinear distortions, known as "optical damage", appeared in the crystal. As a result, the quoted number is assumed to be only an estimate with order-of-magnitude accuracy.

2)The method used in Ref. 8 to estimate β^C_{15} gives an erroneous result, since the change in the polarization state of the light beam as it propagates through the crystal is ignored in it.

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