

Observation of nonequilibrium diffusion in LiNbO_3 crystals

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Experiments on dynamic self-diffraction in lithium niobate crystals have revealed a new mechanism for the formation of displacement refractive-index gratings with anomalously high wave amplification factors. The mechanism involves the diffusion of highly mobile unthermalized electrons with energies $\varepsilon \gg T$.

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In photorefractive ferroelectric crystals such as LiNbO_3 and LiTaO_3 , the interaction of coherent light beams of identical frequency involves a charge separation due to the photovoltaic effect and the drift and diffusion of photoelectrons.^{1,2} The data from optical experiments constitute a rich store of information on the nature of these kinetic phenomena. In the present letter we show, for the first time, that the diffusion current in lithium niobate crystals is determined primarily by unthermalized electrons with energies $\varepsilon \gg T$.

1. In the simplest two-beam version of the experiment (Fig. 1a), an oscillating electric field is formed by the spatial modulation of the light intensity (in proportion to $\cos qz$):

$$E_q = E' \cos qz + E'' \sin qz = |E| \cos(qz + \Phi). \quad (1)$$

The unshifted component, E' , is related to the photovoltaic effect and the photoconductivity in the external field E_0 (we set $E_0 = 0$ below), while the shifted component, E'' , is related to the diffusion of photoelectrons; Φ is the phase shift between the distributions of the light intensity J and the field E_q . The amplitude $|E|$ and the phase Φ are reconstructed from the measured diffraction efficiency η of the refractive-index grating, which is related to E_q , and the measured energy exchange between the recording beams, ΔJ (Refs. 2 and 3). For ordinary waves with identical initial intensities, $J_1 = J_2$, we have

$$\eta = (n_0^2 r_{13} |E| qh / 4 \sin 2\theta)^2, \quad \Delta J = 2J_1 \sqrt{\eta} \sin \Phi, \quad (2)$$

where n_0 is the refractive index, and r_{13} is the electro-optic constant of the crystal. Expressions (2) hold for an arbitrarily small sample thickness h , under the conditions $\eta, \Delta J/J_1 \ll 1$.

It follows from elementary considerations that over times shorter than the Maxwell time, $t \ll t_M = \varepsilon_{\parallel} / 4\pi\sigma_{\Phi}$ [ε_{\parallel} is the static dielectric constant, and $\sigma_{\Phi}(J_1)$ is the photoconductivity], the field E_q should increase linearly, while the phase shift Φ should remain constant:

$$\text{tg } \Phi = E_{\text{dif}} / E_{\Phi} \quad (3)$$

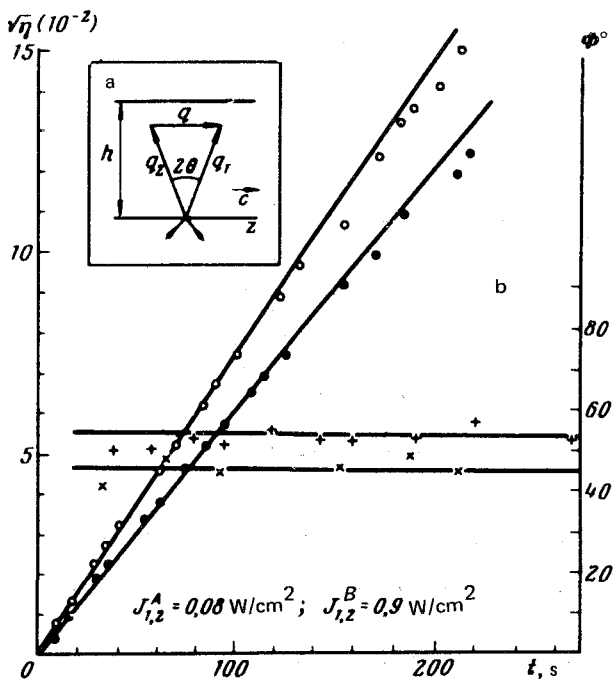


FIG. 1. a: Recording arrangement. b: Time dependence of $\sqrt{\eta}$ and Φ during the initial part of the recording. $\bigcirc - \sqrt{\eta}_A$; $\bullet - \sqrt{\eta}_B$; $\oplus - \Phi_A$; $+$ $- \Phi_B$.

Here the cutoff field E_ϕ , associated with the photovoltaic effect, is a characteristic of the crystal and is easily determined from independent experiments. The so-called diffusion field

$$E_{dif} = qD/\mu \quad (4)$$

is determined by the diffusion coefficient D and mobility μ of the photoelectrons. It is customary to relate D and μ by the Einstein relation $D = \mu T e^{-1}$; then we can write

$$E_{dif} = E_{dif}^T \equiv qT/e, \quad (5)$$

and the phase shift Φ is also fixed. After a long time, $t \gg t_M$, the grating should become a purely displacive grating, $\Phi \rightarrow \pi/2$, and the displaced field component determining the energy exchange becomes $E'' \rightarrow 2E_{dif}$ (Ref. 3).

2. Our experiments reveal a behavior which does not conform to the conventional understanding of the nature of the diffusion of the photoelectrons. Figure 1b shows the results found for the initial recording region for two LiNbO_3 crystals. Sample A had 0.03 wt. % Fe, a thickness $h = 0.33$ cm, and a cutoff field $E_\phi = (40 \pm 5) \times 10^3$ V/cm. Sample B was nominally pure with $h = 0.3$ cm and $E_\phi = (8 \pm 1) \times 10^3$ V/cm. The beam from an argon laser, at $\lambda = 5145$ Å, was used for the recording, at the angle $2\theta = 13^\circ$. We find $\sqrt{\eta} \propto t$, $\Phi = \text{const}$, as expected.¹⁾ The energy exchange, however, is anomalously large: The fields E_{dif} calculated from Eq. (3) and the data in Fig. 1b are,

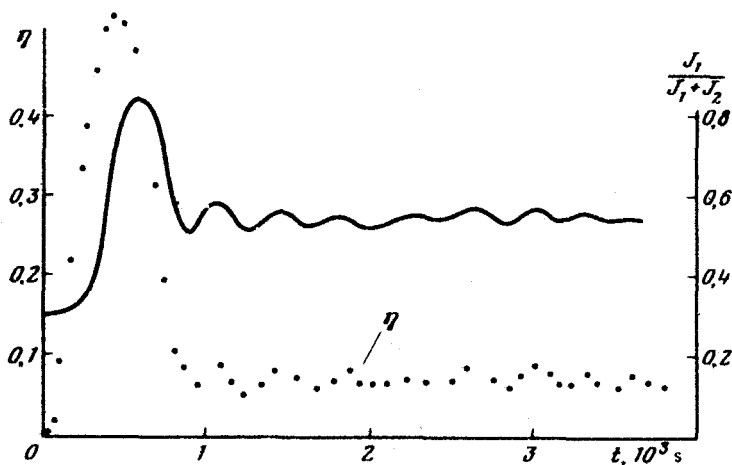


FIG. 2. Energy exchange and diffraction by the grating over long recording times.

respectively 45×10^3 V/cm and 11×10^3 V/cm for crystals *A* and *B*, or 25 and 6 times the diffusion field $E_{\text{dif}}^T = 1.7 \times 10^3$ V/cm.

Figure 2 shows $\eta(t)$ and the relative intensity of one of the beams, $J_1(t)/(J_1 + J_2)$, for a long recording time for crystal *A*. The transient peaks in η and J_1 at $t \sim t_M$ result from the photovoltaic effect.³ The oscillations near the steady state are apparently of instrumental origin, related to the instability of the optical system over long times. In the steady-state part of Fig. 2 the results correspond to $\Phi \cong \pi/2$. From the steady-state energy exchange we can find the independent estimate $E_{\text{dif}} \cong 20 \times 10^3$ V/cm, which agrees well with the value found from the initial part of the recording. The steady-state energy exchange is more than one and a half orders of magnitude greater than that expected (which is determined by the field E_{dif}^T). We wish to emphasize that incorporating such factors as the absorption and scattering of light, the finite beam widths, and several types of charge carriers could only reduce the observed magnitude of the effect.

3. These experimental results find a natural explanation in a model recently proposed for charge transport in photorefractive crystals by Malinovskii and Sturman.⁴ In this model (which is based on experimental data on the photovoltaic effect and the photoconductivity) the unthermalized photoelectrons with energies $\varepsilon \gg T$ are assumed to be relatively important in the current because of the low mobility of the thermalized electrons, μ_T . The mobility μ_T is small because the carriers localize near minima of the fluctuation potential of the crystal. Subsequent experiments on the effect of a magnetic field on the photovoltaic effect have revealed $\mu_e/\mu_T \sim 10^3$ in LiNbO_3 crystals.⁵

For nonequilibrium unthermalized electrons, the diffusion coefficient and the mobility are related by $D_e \cong \varepsilon \mu_e e^{-1}$, instead of the Einstein relation. From (4) we then find the diffusion field

$$E_{\text{dif}} \cong q\varepsilon/e, \quad (6)$$

which is ε/T times E_{dif}^T . Let us compare the contributions of the nonequilibrium and thermalized electrons to the diffusion current:

$$\frac{j_{dif}^e}{j_{dif}^T} \sim \frac{\mu_e}{\mu_T} \frac{\epsilon}{T} \frac{\tau_e}{\tau_{ж}}, \quad (7)$$

where τ_e is the energy relaxation time, and τ_l is the lifetime in the band of thermalized photoelectrons. Assuming $\epsilon/T \sim 10^2$ and $\tau_e \sim 10^{-11}$ s, we find a very slight restriction on the lifetime, $\tau_l \lesssim 10^{-6}$ s, as a condition for the predominance of nonequilibrium diffusion. This restriction is much less stringent than the condition for the predominance of the nonequilibrium photoconductivity.⁴ There thus exists a broad intermediate range of parameters in which σ_ϕ is related to thermalized electrons, diffusion is related to nonequilibrium electrons, and $qT/e \leq E_{dif} \leq q\epsilon/e$. Knowing E_{dif} , we can calculate the energy of the nonequilibrium electrons. For crystal A we find $\epsilon \gtrsim 0.5$ eV.

Since E_{dif} depends strongly on the light frequency and the energy structure of the crystal, we can significantly increase the wave amplification factors. It would also be very interesting to see experiments at large spatial frequencies q , where we would expect to see effects of a pronounced spatial dispersion resulting from the long mean free paths of the non equilibrium electrons.

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¹⁾The points in Fig. 1b correspond to one of the experiments that were carried out. The results are reproducible quite well (within 10–15%) from experiment to experiment.

⁴M. Lines and A. Glass, Principles and Applications of Ferroelectrics and Related Materials, Oxford Univ. Press, 1980 (Russ. transl. Mir, Moscow, 1981).

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