Motion of space-charge waves in $LiNbO_3$:Fe

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During the dark relaxation of phase holograms written in electrooptic $LiNbO_3$:Fe crystals, the holograms undergo a spatial displacement. This observational fact is linked with a propagation of trap charge-exchange waves in the crystal. The dispersion of these waves, $\omega \sim K^{-1}$, agrees with the experimental results.

In this study of the dark relaxation of dynamic holograms written in $LiNbO_3$:Fe crystals (0.05% Fe by weight in the melt) we used samples with dimensions of $3\times3\times0.4$ mm along the X, Y, and Z axes, respectively, and with an absorption coefficient of 6.4 cm^{-1} at the wavelength 0.44 μm. The faces of the sample perpendicular to the Y axis were covered with a silver paste, in which a window was left for writing holograms; the dimensions of the window were 2×0.8 mm along the X and Y axes,

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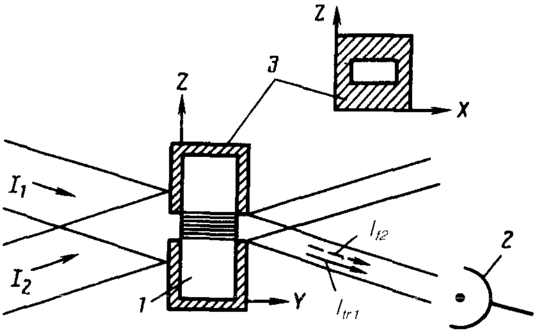

FIG. 1. Experimental arrangement used to study the motion of gratings. The inset is the face of the crystal perpendicular to the Y axis. 1—Sample; 2—photodetector; 3—silver paste.

respectively (Fig. 1). Phase diffraction gratings were written in an arrangement in which two light beams with a wavelength of 0.44 μm, the extraordinary polarization, identical intensities of 0.8 W/cm^2, and diameters of 1.5 mm intersected in the crystal.[1] The bisector of the angle at which the beams met coincided with the Y axis; the wave vector of the grating which was written was directed along the Z axis of the crystal. The diffraction efficiency of the gratings which were written was 1–10%. The gratings were erased by heating the crystal to about 200 °C and then holding it at room temperature for about 2 days.

The dark conductivity of the crystal, $\sigma_d = 6\times 10^{-15}$ S/cm, was measured with a V7–30 electrometer along the Z axis in samples of various dimensions, in order to determine the contribution of the surface conductivity. The latter turned out to be negligible.

To observe the relaxation of the holograms, we used the same light beams as during the writing, but attenuated by neutral light filters to an intensity of 10^{-5} W/cm^2. Special experiments showed that this illumination level has essentially no effect on the relaxation process. Using a calibrated photodetector with an FD-1 photodiode, we measured the intensity of the beam transmitted through the crystal, I_{tr1}, with beam I_2 turned off; the intensity of the diffracted beam, I_{f2}, with beam I_1 turned off; and the intensity of the resultant beam, I_Σ, with I_1 and I_2 turned on (Fig. 1). We found that the intensity I_Σ oscillates sinusoidally in time (Fig. 2a), while I_{tr1} and I_{f2} remain essentially constant over the oscillation period. We observed up to ten oscillation periods, with an amplitude which fell off slowly over time. Figure 2b shows the oscillation period T as a function of the grating wave vector K. We see that the dependence $T(K)$ is linear.

When the holograms were written immediately after the completion of an erasure by heating at 200 °C, we did not observe a linear $T(K)$ dependence: The period T was independent of the wave vector K over the interval 1 $\mu m^{-1} \leqslant K \leqslant 4$ mm^{-1} and had a value of 830 s.

It follows from the diagram in Fig. 1 that when the interference pattern created in the crystal by the intersection of the reading beams is rotated an angle φ with respect to the previously written grating, the intensity of the resultant signal can be written

$$I_\Sigma = I_{tr1} + I_{f2} + 2\sqrt{I_{tr1}\, I_{f2}}\cos(\pi/2-\varphi) = I_{tr1} + I_{f2} + 2\sqrt{I_{tr1}\, I_{f2}}\sin\varphi ,$$

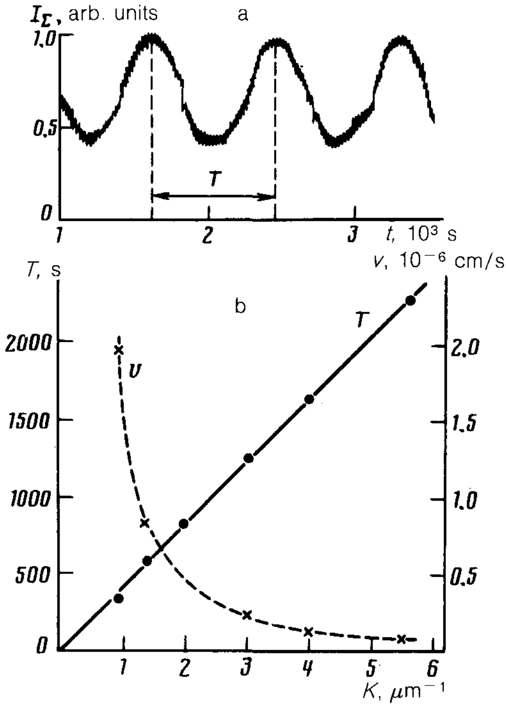

FIG. 2. a—Oscillations of the resultant intensity over time in the case $K = 2.0$ μm^{-1}; b—dependence of the period of these oscillations on the wave vector of the gratings which have been written.

where the shift of $\pi/2$ is added because of the diffraction by the grating. We thus see that temporal oscillations in I_Σ could result from only a change in φ over time. Since the interference pattern is fixed, this conclusion leads to the further conclusion that the written grating must be moving. The velocity at which the grating moves is described by $v \sim K^{-2}$ (Fig. 2b), and it ranges from 10^{-7} to 2×10^{-6} cm/s.

The average amplitude of the oscillations is about 80% of the maximum possible amplitude, $2(I_{tr1} I_{f2})^{1/2}$. This result may imply that an immobile grating exists along with the moving grating in the crystal.

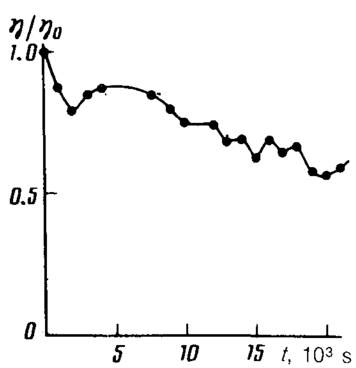

FIG. 3. Time evolution of the diffraction efficiency during dark erasure ($\eta_0 = 10\%$ is the diffraction of the grating at the time at which the writing is terminated; $K = 2.1$ μm^{-1}).

To study the time dependence of the diffraction efficiency of the gratings, we measured the intensity I_{f2} when only beam I_2 was turned on, for 10 s at intervals of 1000 s. Figure 3 shows the time evolution of the diffraction efficiency (for $K = 2.0$ μm^{-1}). This dependence turns out to be nonmonotonic. Measurements carried out for K values from 0.9 to 3.9 μm^{-1} show that holograms with larger values of K undergo a slower relaxation. In all cases, however, the lifetime of the gratings is more than two orders of magnitude greater than the Maxwell relaxation time $\tau_M = \epsilon/4\pi\sigma_d \approx 600$ s. These results contradict the present understanding of the situation—that a hologram written in a crystal should, after the end of the illumination, undergo a monotonic erasure with a time scale corresponding to a Maxwell relaxation and independent of the period of the grating. It is pertinent to note that Kanaev and Malinovskiĭ[2] have observed an increase in the storage time of holograms.

We know that the process by which holograms are erased is linked with the relaxation of a nonuniform filling of deep impurity centers in the crystal by electrons. In a static field E_0, trap charge-exchange waves can propagate through a crystal of this sort; such waves would be manifested as oscillations in the bound charge.[3] Accordingly, a hologram in a field E_0 should constitute a charge-exchange wave which is propagating along E_0. The periodic field of this wave should modulate the refractive index of the crystal. Furman[4] has pointed out the possibility that holograms will move during uniform illumination of a crystal to which a static field E_0 is applied from a voltage source.

As was shown in Ref. 3, under the condition $Kl_0 > 1$ ($l_0 = \mu\tau_l$; t_l is the electron lifetime in the conduction band; and μ is the mobility of these electrons) the dispersion of the wave is $\omega = 2\pi/T = (\tau_M K l_0)^{-1}$. This result agrees within a numerical factor with the experimental dependence in Fig. 2b.

The time constant of the damping of the trap charge-exchange waves, τ may be one or two orders of magnitude greater than τ_M, according to the theory, and it is described by[3] $\tau \sim K^2$. This behavior agrees qualitatively with the experimental observation that gratings with relatively large values of K relax more slowly. It is pertinent to note that Belabaev *et al.*,[5] have observed a decay law $\tau \sim K^2$ for gratings in crystals subjected to a uniform illumination.

The existence of a quasisteady field E_0, required for the propagation of trap charge-exchange waves in a crystal, can be linked with the presence of a slowly damped component of the relaxation current J_R in a crystal without an inversion center. Such a current would arise, for example, from the production of a nonequilibrium population of impurity centers during the writing of gratings. The field of the space charge created by this current, $E_0 = -J_R/\sigma_d$, should be sufficient to satisfy the condition $Kl_0 > 1$. With $\mu\tau_l \sim 10^{-8}$ cm$^2/V$ we find $E_0 \gtrsim 10^4$ V/cm; also using the measured value of σ_d, we find $J_R \gtrsim 6 \times 10^{-11}$ A/cm^2. Since the crystal parameters μ and τ_l are not known well, this estimate is extremely crude.

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