

Nonlinear response of photorefractive lithium tantalate and niobate at acoustic frequencies

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An amplification of the intensity of pump oscillations is observed experimentally at frequencies from 100 Hz to 1 kHz during photoinduced light scattering and holographic-type parametric scattering in photorefractive lithium tantalate and niobate. Possible ways are analyzed for explaining the existence of a photorefractive response in these crystals over times of 10^{-2} – 10^{-3} s, which are five orders of magnitude shorter than the Maxwell time. © 1997 American Institute of Physics. [S1063-7761(97)02410-4]

1. INTRODUCTION

The phenomenon of photorefraction was observed in the 1960's,^{1,2} but interest in photorefractive media continues up to the present owing to the enormous variety of their properties. For example, a holographic grating inscribed on lithium niobate can be kept for months,³ while the characteristic photorefractive response time in barium titanate is nano- or picoseconds.⁴ According to the generally accepted theory of photorefraction proposed by Glass,⁵ when photorefractive crystals are illuminated a macroscopic current develops in them and rearranges the electric charge density. The electrostatic field of the separated charges changes the refractive index of the medium owing to an electrooptical effect. This theory assumes the existence of a single temporal parameter which characterizes the entire photorefractive process, the Maxwell time,

$$\tau_M = \varepsilon_{st}/4\pi\sigma, \quad (1)$$

where ε_{st} is the dielectric permittivity of the medium and $\sigma = \sigma_T(1 + I/I_s)$ is its conductivity, whose value under illumination varies over a scale length determined by the parameter I_s .⁶

From time to time, however, papers are published which indicate the existence of a photorefractive response with a characteristic time very different from the Maxwell time.^{4,7,8} In our work we have also observed a photorefractive response in copper-doped lithium tantalate and niobate at characteristic frequencies $f \sim 0.1$ – 1 kHz, whereas the Maxwell times for these crystals are tens of minutes.

2. DESCRIPTION OF THE EXPERIMENT

When a coherent light beam strikes a photorefractive medium, wide-angle scattering that is degenerate in frequency may take place over a characteristic time. This scattering is usually associated with optical damage and efforts are made to avoid it, as it greatly reduces the pump intensity. This is photoinduced light scattering, which has been explained in terms of the recording and self-consistent amplification of a large number of holographic gratings.⁹ When certain conditions are met in directions which form a distinct cone whose shape is determined by the four-wave spatial synchronization condition $2\mathbf{k}_p = \mathbf{k}_{s_1} + \mathbf{k}_{s_2}$ (where the sub-

scripts p and s denote the pump mode and scattered light, respectively), in anisotropic crystals it is possible to observe a sudden rise in the scattered light intensity. This effect arises because in these directions two, rather than one, scattered light modes interact on a single holographic grating and is referred to as holographic-type parametric scattering.¹⁰

In a study of the temporal characteristics of the photorefractive response, we have examined $o-e$ -mode photoinduced light scattering and $oo-ee$ -mode holographic-type parametric scattering in $\text{LiTaO}_3:\text{Cu}$ and $ee-oo$ -mode holographic-type parametric scattering in $\text{LiNbO}_3:\text{Cu}$. The pump intensity was modulated at one or two frequencies in the range from 100 Hz to 1 kHz with a small (less than 10%) depth of modulation. The magnitude of the photorefractive response of the crystals at these frequencies was determined from the change in the depth of modulation in the intensity of the light during the scattering process.

The experimental apparatus is shown schematically in Fig. 1. An ordinary polarized beam from a helium-cadmium laser ($\lambda = 442$ nm), for lithium tantalate, or an extraordinary polarized beam from an argon laser ($\lambda = 488$ nm), for lithium niobate, was passed through an electrooptical modulator and was incident on an $X-Z$ -cut crystal. Two photodetectors detected the pump and scattered light intensities. A computer connected to the measurement apparatus through a CAMAC crate processed the data. The depth of modulation in the intensity of the light in both channels was determined from the magnitudes of the Fourier components at the modulation frequencies, and in order to isolate the regular modulation more precisely from the noise, we used the spectrum of the fluctuations in the intensity (rather than the spectrum of the signal), which is the Fourier transform of the autocorrelation function of the luminous intensity, normalized to the square of the average value. Figure 2 shows a typical plot of the autocorrelation function of the pump and scattered light intensities modulated at the two frequencies, together with their Fourier spectra. A coefficient characterizing the magnitude of the photorefractive response at these frequencies was obtained from the ratio of the amplitudes of the Fourier components at the modulation frequencies:

$$K(f) = \sqrt{G_s(f)/G_p(f)}. \quad (2)$$

where

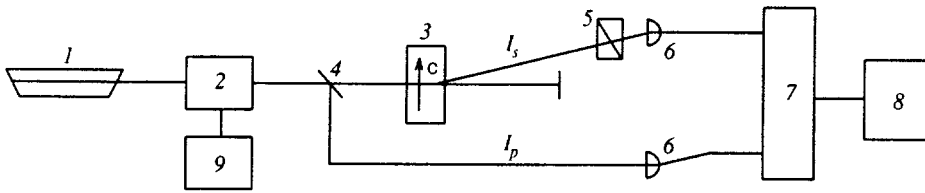


FIG. 1. A sketch of the experimental apparatus: (1) argon or helium-cadmium laser; (2) electrooptical modulator; (3) photorefractive crystal of copper-doped lithium niobate or tantalate (*c* is the optical axis); (4) semitransparent mirror; (5) analyzer; (6) photodetectors; (7) CAMAC crate; (8) computer; (9) electrical pulse generator.

$$G(f) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} b(\tau) e^{2\pi i f \tau} d\tau$$

is the spectrum of the fluctuations and

$$b(\tau) = \langle I(t)I(t+\tau) \rangle / \langle I(t) \rangle^2$$

is the normalized autocorrelation function of the luminous intensity. $K(f) = 1$ corresponds to equality of the relative modulation amplitudes of the pump and scattered light intensities, i.e., to ordinary diffraction on a stationary holographic

grating. It can be seen clearly in Fig. 2, however, that the depth of modulation for the scattered light intensity is much higher than that for the pump light, which means that the photorefractive holographic grating is able to vary as the pump intensity changes, thereby amplifying the modulation in the scattered radiation.

3. DISCUSSION OF RESULTS

Figure 3 shows the scattered intensity and conversion factor for the modulation depth in the pump intensity as

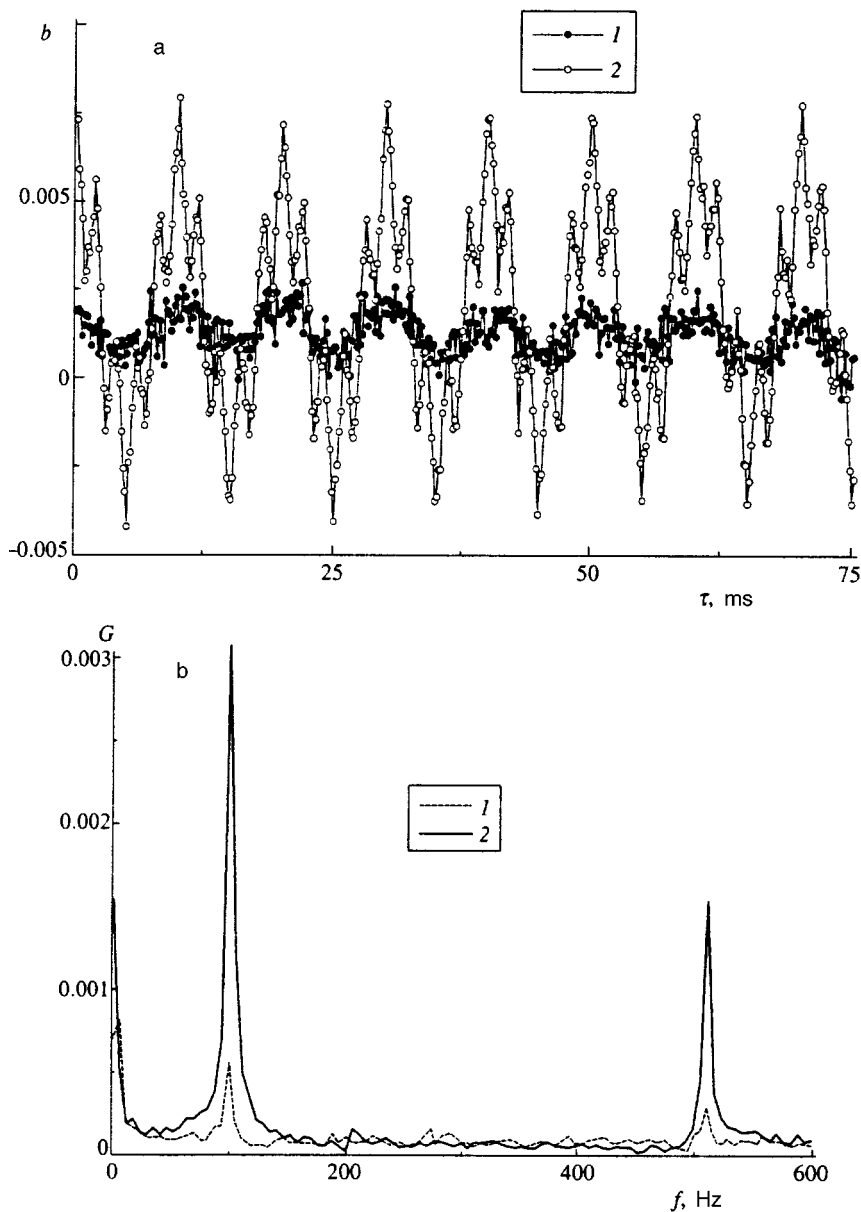


FIG. 2. A typical example of the autocorrelation function (a) and spectrum (b) of the fluctuations in the intensity of a pump modulated at frequencies of 100 Hz and 500 Hz (1) and in the intensity of light scattered as a result of *oo-ee*-mode holographic-type parametric scattering in $\text{LiTaO}_3:\text{Cu}$ (2).

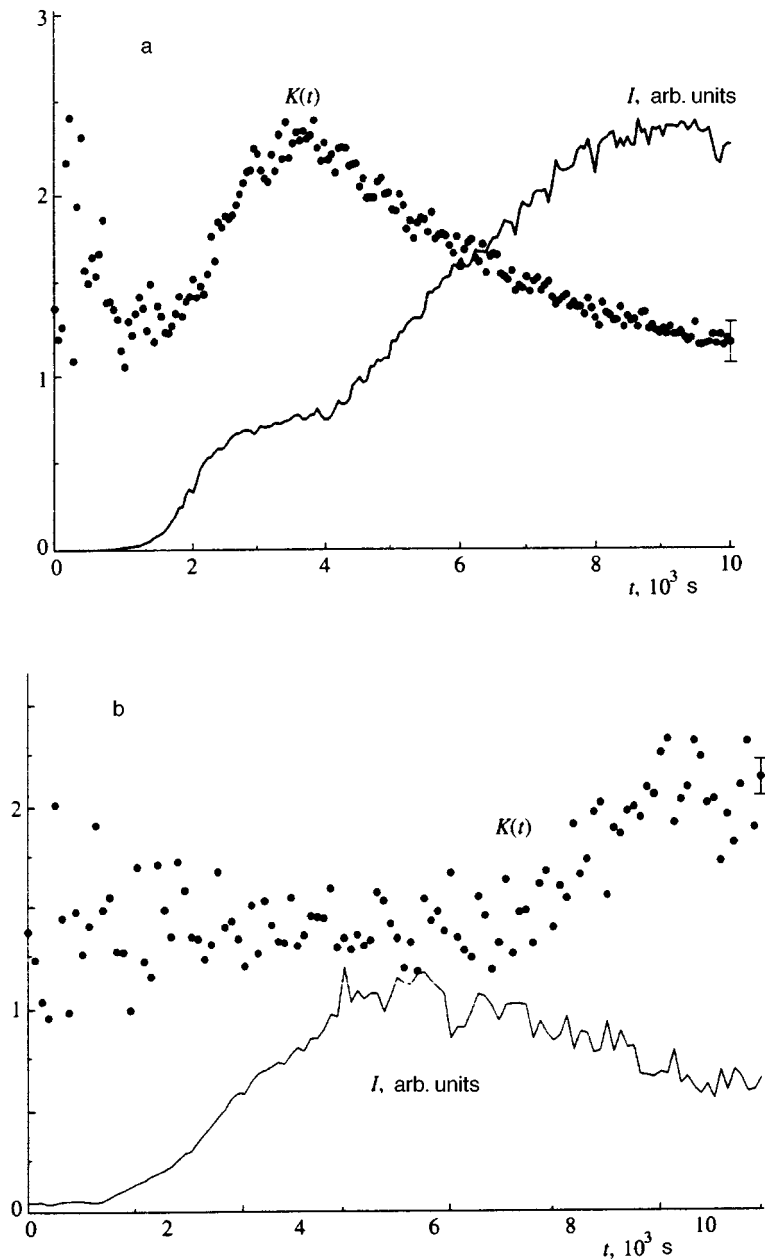


FIG. 3. Time dependence of the scattering intensity and conversion factor for the depth of modulation in the pump intensity at frequencies of $f=300$ Hz (a) and $f=100$ Hz (b) during detection of $ee-oo$ -mode holographic-type parametric scattering in $\text{LiNbO}_3:\text{Cu}$ (a) and $oo-ee$ -mode holographic-type parametric scattering in $\text{LiTaO}_3:\text{Cu}$ (b).

functions of time during the detection of holographic-type parametric scattering in lithium niobate and tantalate. By the end of the first hour for the lithium tantalate and the second hour for the lithium niobate, the scattering intensity has stopped rising and has attained an approximately constant level, i.e., the recording of the holographic gratings saturates with the effect of the electrostatic field of the separated charges balancing the photogalvanic current. The time over which the process of recording the holographic-type parametric scattering reaches saturation is of the same order as the Maxwell time $\tau_M \sim 10^3$ s. At the same time, however, the holographic grating is able to amplify pump oscillations at frequencies on the order of a hundred hertz, i.e., for a oscillation period, $T \sim 10^{-2}$ s, which is five orders of magnitude shorter than the Maxwell time.

The time dependence of the conversion factor for the depth of modulation is different for lithium niobate and tan-

talate. In Fig. 3a for $\text{LiNbO}_3:\text{Cu}$ the coefficient $K(t)$ reaches a maximum in the middle of the dynamic regime where the scattering intensity is rising rapidly, while it falls off to unity in the saturation regime. Thus, in lithium niobate the holographic grating is able to react to the changes in the pump intensity only during the period when the grating is increasing even without this change, while in the saturation regime, as might be expected, it becomes stable. The $K(t)$ curve is substantially different for lithium tantalate, as can be seen in Fig. 3b. It has no distinct maxima, as in Fig. 3a; the conversion factor differs little from $K \approx 1.4$ throughout the dynamic regime. Then in the saturation regime the conversion factor not only fails to drop to unity, but increases to $K \approx 2$. Thus, even in the stationary state, a photoinduced holographic grating in lithium tantalate is able to react to changes in the pumping over times much shorter than the Maxwell time.

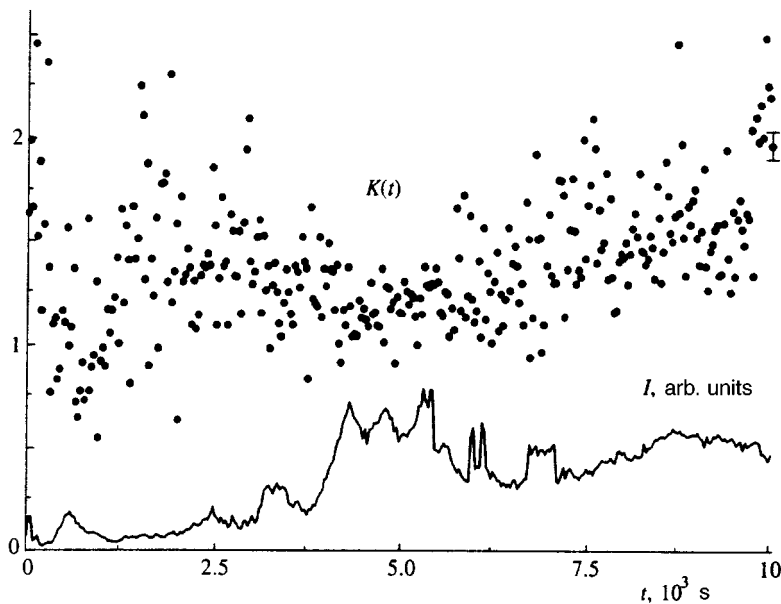


FIG. 4. Time dependence of the scattering intensity $I(t)$ and conversion factor for the depth of modulation of the pump, $K(t)$ at a frequency of $f=100$ Hz during detection of $o-e$ -mode photoinduced light scattering in $\text{LiTaO}_3:\text{Cu}$.

Figure 4 shows the time dependence of the scattering intensity and conversion factor of the oscillations for photoinduced light scattering in lithium tantalate. As can be seen from this graph, it is meaningless to introduce the concepts of dynamic and saturation regimes for photoinduced light scattering, since the scattering intensity experiences rapid drops throughout the entire observation period. This is caused, first of all, by the competition between photoinduced light scattering and holographic-type parametric scattering which causes a large part of the pump intensity to be removed by holographic-type parametric scattering and, second, by the possibility of electrical breakdowns in the crystal, and, finally, by the fact that energy exchange between two interacting waves is forbidden under stationary conditions.¹¹ Evidently, for the same reasons the conversion factor for the depth of the intensity modulation has a large scatter, although on the average, as for holographic-type parametric scattering, it is undoubtedly greater than unity.

Let us finally examine Fig. 5 which shows the conversion factor for the modulation depth of the pump intensity for two frequencies (not multiples) at once, $f_1=100$ Hz and $f_2=430$ Hz, during holographic-type parametric scattering in $\text{LiTaO}_3:\text{Cu}$. The picture is much more complicated for two modulation frequencies: although the scattering intensity varies significantly only within the first hour after the onset of recording, as in Fig. 3b, the conversion coefficients undergo rapid changes for eight hours and only after that do they approach a roughly constant level of $K_1 \approx K_2 \approx 1.5$. Evidently, the amplifications of the intensity oscillations at the two frequencies are interrelated and cannot be treated independently. Otherwise it would be difficult to explain the fact that a time almost an order of magnitude longer than the time to record the gratings is necessary to attain a stationary situation, not to mention that twice during the observation period there was a time when one of the conversion factors had a local maximum while the other was less than unity (i.e., the

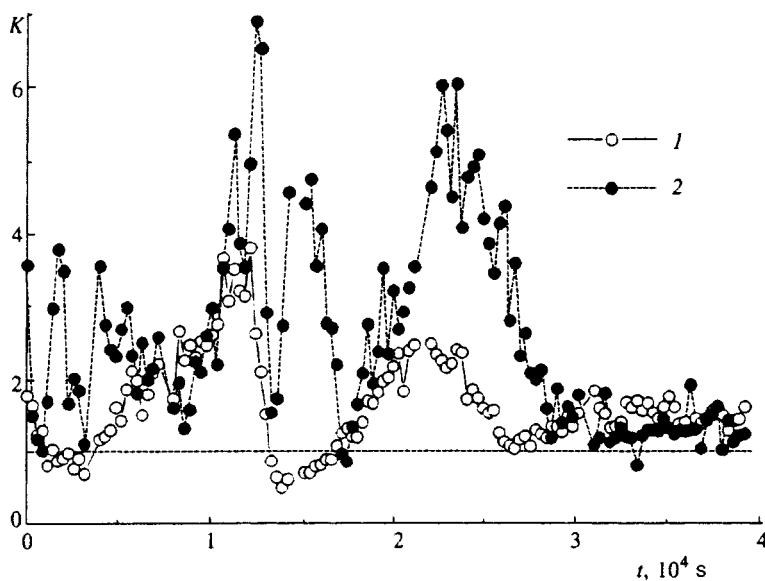


FIG. 5. Time dependence of the conversion factor for the depth of modulation of the pump intensity, $K(t)$ at frequencies of $f_1=100$ Hz (I) and $f_2=430$ Hz (2) during detection of $oo-ee$ -mode holographic-type parametric scattering in $\text{LiTaO}_3:\text{Cu}$.

relative amplitude of the oscillations decreased), an effect observed in the preceding plots.

In completing our discussion of the experimental data, we must note that the results were completely irreproducible during this study of the temporal dependence of the conversion factor for the intensity modulation depth: two curves obtained under the same conditions can have substantially different features aside from the fact that the gain coefficient is almost always greater than unity. This suggests a stochastic mechanism for amplification of the intensity oscillations at these frequencies.

In sum, our results yield the following conclusions:

1. Amplification of oscillations (or fluctuations) in the pump intensity during scattering in photorefractive lithium tantalate and niobate has been observed at frequencies from 100 Hz to 1 kHz. We were unable to detect amplification of the oscillations reliably at higher frequencies.

2. Amplification of the oscillations is observed in lithium tantalate under any conditions, while in lithium niobate it occurs only during the dynamic regime, when there is a rapid increase in the holographic gratings, i.e., in the nonstationary case.

3. The amplification of oscillations during photoinduced light scattering and holographic-type parametric scattering indicates that the holographic gratings recorded in a crystal vary with changes in the pump intensity. As a result, the oscillations in the scattered radiation are determined by a combination of the oscillations in the pump intensity and of the amplitude of the holographic grating.

4. The amplification of oscillations with a period much shorter than the Maxwell time of the medium indicates the existence of a new, as yet unknown mechanism for photorefractive with a substantially shorter characteristic time.

5. The mechanism for the amplification of the oscillations appears to be stochastic in nature and leads to strong fluctuations in the gain coefficient, a lack of reproducibility in the results, and, perhaps, interactions among the amplification processes for oscillations at different frequencies.

We now consider some possible explanations for the observed effect based on earlier work.^{4,7} The existence of a photorefractive response in barium titanate over times on the order of tens of picoseconds has been observed by recording holograms with picosecond pulses,⁴ while at least a nanosecond is required to redistribute the electron density in a crystal of this type. This rapid photorefractive response was explained by the fact that electrons photoexcited in the conduction band of the crystal change its refractive index owing to Drude transfer and, thereby, record a grating based on free carriers.⁴ This grating spreads out owing to diffusion of electrons in the conduction band, i.e., its lifetime is bounded above by the Maxwell time. The idea of the existence of a free carrier grating can be used to explain the amplification of pump fluctuations. In fact, a change in the carrier concentration in the conduction band takes place essentially immediately (over times on the order of picoseconds), i.e., a free carrier grating is capable of reacting immediately to a change in the intensity of a pump at frequencies on the order of hundreds of hertz.

Another possible explanation of the observed effect is

based on data obtained in Ref. 7. There a photo-Hall effect was observed which involved the appearance of a photo-Hall current in a $\text{LiNbO}_3:\text{Fe}$ crystal perpendicular to the magnetic field vector and to a photogalvanic current that is proportional to the intensity of illumination of the crystal. The mobility of the carriers forming this current was determined from the magnitude of the photo-Hall current and turned out to be three orders of magnitude higher than the ordinary carrier mobility in lithium niobate. It was proposed that the measured mobility corresponds to nonthermal electrons that have not yet been clothed in a polaron "coat" and, therefore, have a much lower effective mass and a higher mobility. Since the mobility of a medium is proportional to the mobility of its carriers, while the concentration of nonthermal electrons may increase under nonstationary conditions (for example, in the case of fluctuations in the pump intensity), the Maxwell time (1) corresponding to the nonthermal carriers may be much smaller than usual, which would also explain the amplification of the light intensity fluctuations during scattering at acoustic frequencies.

4. CONCLUSION

We have observed a response in photorefractive lithium tantalate and niobate over times five orders of magnitude shorter than the Maxwell time that leads to nonlinear conversion of fluctuations in the pump intensity. We have proposed two possible ways of explaining the observed effect which, in principle, are not mutually exclusive. The existing phenomenological theory of photorefractive processes (for example, the recording of holograms, the development of holographic-type parametric scattering), does not account for the existence of free electrons on gratings or of nonthermal carriers in photorefractive crystals. Thus, in order to clarify the true causes of the amplification of pump intensity oscillations, a more rigorous theory of photorefractive must be developed which includes the coupling of various processes which have very different characteristic times in photorefractive crystals: from the photoexcitation times for carriers in the conduction band (times on the order of picoseconds) to the Maxwell time for redistribution of the electron density in a crystal.

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