Optically Induced Inhomogeneity of the Refractive Index of $LiNbO_3$ Crystals

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The phenomenon of induced inhomogeneity of the refractive index under the action of laser radiation is studied in $LiNbO_3$ and $LiTaO_3$ crystals. The dependence of the refractive index change Δn on crystal composition, temperature, intensity, and diameter of the light beam is determined. It is shown that with variation of the Li/Nb ratio in $LiNbO_3$ between 0.9 and 1.1 and the Li/Ta ratio in $LiTaO_3$ between 0.8 and 1.2, the quantity Δn increases by 5 and 10 times respectively. With increase of the radiation intensity, Δn grows linearly in $LiNbO_3$ whereas it exponentially decreases with increasing temperature. A model of the phenomenon, which can explain the experimental results, is considered.

1. INTRODUCTION

IN certain ferroelectric crystals, such as $LiNbO_3$, $LiTaO_3$[1] and $BaTiO_3$[2], laser radiation produces a change in the refractive index. A light beam passing through the crystal perpendicular to the polar axis z spreads out along this axis, owing to the decrease of the refractive index on the beam axis. After the radiation is stopped, a region with altered refractive index remains in the crystal and can exist for a long time¹⁾. We shall henceforth call this phenomenon induced optical inhomogeneity (IOI). When a specimen with IOI is heated to ~200°C, the region with the altered refractive index vanishes. It was also noted[3] that the change of the refractive index decreases with increasing sample temperature at a constant radiation intensity. The IOI is observed following the action of laser radiation in the visible band, with λ equal to 0.48, 0.53, and 0.63 μ, but is not observed when $\lambda = 1.06\ \mu$. Chen[4] has investigated the dependence of the change Δn of the refractive index under the influence of light from an argon laser ($\lambda = 0.48\ \mu$) on the exposure duration and on the light-beam power. It was shown that the change of the refractive index of the extraordinary wave is 4–5 times larger than that of the ordinary wave. According to the model proposed in[4], the IOI is due to the drift of the photo-excited electrons in the internal electric field directed along the z axis, and by their capture by traps on the periphery of the light beam. The resultant space-charge field leads, owing to the linear electro-optical effect, to a change in the refractive index of the material. The nature of the internal electric field in this model remains unclear.

Johnston[5], developing further the model proposed by Chen[4], suggested that the internal field in a ferroelectric crystal can be due to the change of the spontaneous polarization as a result of photoionization of the charged lattice defects. In $LiNbO_3$, such defects can be, for example, excess Nb ions[5], oxygen vacancies[4], or an Fe^{3+}-ion impurity[6]. The nature of the defects remains unclear to this day.

We report here investigations of the IOI in $LiNbO_3$ and $LiTaO_3$ as a function of the composition, temperature, power, and diameter of the laser beam. A model explaining the experimental results is considered.

2. EXPERIMENTAL PROCEDURE AND RESULTS

The IOI in $LiNbO_3$ crystals was investigated by a polarization method that permits measurement of the birefringence $\Delta n = \Delta(n_e - n_o) \approx \Delta n_e$ (n_e and n_o are the refractive indices of the extraordinary and ordinary waves). The inhomogeneity of the refractive index was produced with a helium-neon laser with $\lambda = 0.63\ \mu$ and a power up to 50 mW. The laser beam was focused into the sample by a lens with f = 5 cm. The samples were cut in the form of polished single-domain plates or bars. The sample z axis was in a plane perpendicular to the beam axis and was inclined 45° to its polarization. Δn was measured with light from the same laser, but attenuated with neutral filters to a level 0.1 mW. The compensator was a quartz wedge of variable thickness. Δn was measured accurate to $\sim 3 \times 10^{-5}$.

Dependence of IOI on the Crystal Composition

The $LiNbO_3$ and $LiTaO_3$ crystals allow wide deviations from stoichiometric composition[7]. We have investigated IOI in $LiNbO_3$ with R = Li/Nb from 0.9 to 1.1 and in $LiTaO_3$ with R = Li/Ta from 0.8 to 1.2.²⁾

Figure 1 shows a plot of Δn against the exposure time for $LiNbO_3$ with different R at a radiation intensity 1500 W/cm². It is seen from the figure that for all values of R, Δn first increases linearly with time and then reaches a stationary value Δn_{st}. The rate of change of Δn, as well as the value of Δn_{st}, increases with increasing R, and the values of Δn_{st} for crystals with R = 1.1 and R = 0.9 differ by a factor of 5.

The IOI in $LiTaO_3$ was investigated at $\lambda = 0.53\ \mu$, the second harmonic of a cw neodymium-doped aluminum-yttrium garnet laser, since the radiation with $\lambda = 0.63\ \mu$ did not produce a noticeable change in the refractive index of $LiTaO_3$. The intensity of the harmonic at the focus of the lens reached ~500 W/cm². Figure 2 shows the dependence of Δn_{st} on the ratio R = Li/Ta, from which it follows that the susceptibility to IOI increases by 8–10 times when R changes from 0.8 to 1.2.

Temperature Dependence of IOI

The temperature dependence of the IOI in $LiNbO_3$ was investigated in the interval from 77 to 430°K. It follows from the experiment that in the range from 300 to 430°K the change of Δn_{st} with temperature can be described by a relation of the type $\sim\exp(\Delta_{eff}/kT)$, where Δ_{eff} is the effective activation energy, k Boltzmann's constant, and T the absolute temperature. From the slope of the ln Δn line as a function of 1/T

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1) This phenomenon is known in the literature as "optical damage."

2) The values of R pertain to the melt.

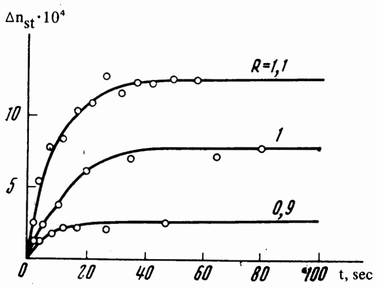


FIG. 1

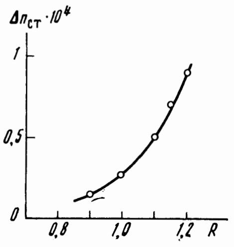


FIG. 2

FIG. 1. Dependence of Δn on the duration of the irradiation for $LiNbO_3$ crystals with different R = Li/Nb.

FIG. 2. Dependence of Δn_{st} on R = Li/Ta in $LiTaO_3$.

(Fig. 3) we can obtain for Δ_{eff} a value $\sim(0.3 \pm 0.06)$ eV for crystals with R equal to 1, 1.1, and 1.15. The low value of Δn in the crystal with R = 0.9 has made it impossible to estimate Δ_{eff} for this crystal. When the $LiNbO_3$ samples were cooled to liquid-nitrogen temperature, an increase of Δn not exceeding 10 times was observed. A larger increase of Δn_{st} would be expected from the relation $\Delta n_{st} \sim \exp(\Delta_{eff}/kT)$.

Apparently, lowering the temperature causes Δn_{st} to increase to a maximum value $\sim 10^{-3}$ [4], beyond which no further increase is possible.

Dependence of IOI on the Power and Diameter of the Laser Beam

Figure 4 shows a plot of Δn_{st} in $LiNbO_3$ against the light-beam diameter D at a constant radiation power. The diameter of the light beam changed from 4.5×10^{-3} to 17×10^{-3} cm when the focal length of the lens was increased from 4.5 to 17 cm. The experiments with lenses having $f \leq 9$ cm were performed on a plate 0.44 mm thick, and with $f > 9$ cm on a plate 2.9 mm thick. This increased the accuracy of the measurement of Δn in the case of long-focus lenses. In all cases, the crystal thickness remained smaller than the length of the focal region of the lens. It is seen from Fig. 4 that $\Delta n_{st} \sim (1/D)^2$.

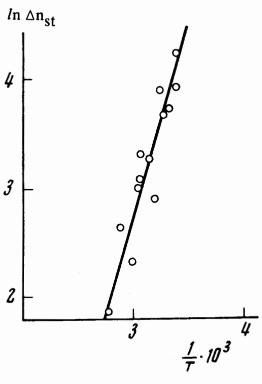
Figure 5 shows the dependence of Δn_{st} on the radiation power at a constant beam diameter $D = 4.5 \times 10^{-3}$ cm. The radiation was attenuated with calibrated neutral filters of varying transmission. As follows from the figure, the value of Δn_{st} increases linearly with the power. Thus, our results lead to the conclusion that Δn_{st} varies linearly with changing intensity up to 500 W/cm². At the helium-neon laser power used in our experiments we did not obtain the saturation of Δn_{st} which was observed in[4].

3. DISCUSSION OF RESULTS

We shall derive below the dependence of the stationary change of the refractive index Δn_{st} on the laser beam intensity, sample temperature, and the physical parameters of the sample.

We consider a ferroelectric crystal exposed to a laser beam with limited diameter D and intensity I; the beam is perpendicular to the polar axis z. We assume, just as in[4], that the sample contains electron traps with concentration N. A fraction N_t of the traps is filled with electrons and can be ionized by the light, whereas the fraction $(N - N_t)$ is free and can serve as centers for electron capture. According to[5], the change ΔP_s of the spontaneous polarization in the illuminated region is proportional to the concentration of the non-ionized traps ($N_t - n_t$, where n_t is the concentration of the filled traps in the illuminated region). Consequently, the change of the refractive index takes the form

FIG. 3. Plot of Δn_{st} of $LiNbO_3$ vs. temperature.



$$\Delta n_3 = -f_{33}n_3^3\Delta P_s = -f_{33}n_3^3 p(N_t - n_t), \tag{1}$$

where n_3 is the refractive index along the z axis, f_{33} is the electric-optical coefficient, and p is the change of the dipole moment of the lattice following ionization of one trap.

At the same time, the change of the spontaneous polarization ΔP_s leads to the appearance of a polarization charge with density $\rho_p = -\nabla P_s$ and a corresponding field with intensity $E_p = -4\pi\Delta P_s$, directed towards the negative end of the polar axis. The free electrons can drift in this field towards the boundary of the light beam, forming a space charge whose field E_{sp} is opposite to E_p.

The change of the concentrations of the free electrons n_e and of the filled traps n_t in the illuminated region can be described by the equations

$$\frac{dn_e}{dt} = \alpha In_t + \beta n_t - Bn_e(N - n_t) - \frac{n_e}{\tau_d}, \tag{2}$$

$$\frac{dn_t}{dt} = -\alpha In_t - \beta n_t + Bn_e(N - n_t), \tag{3}$$

where α and β are the probabilities of photo- and

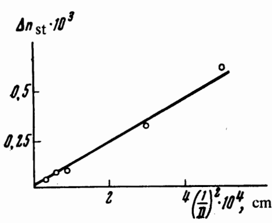


FIG. 4

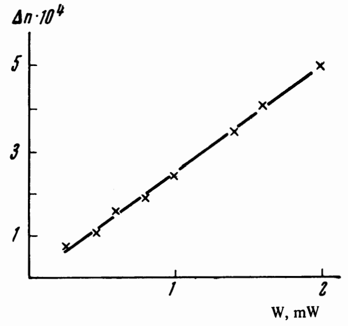


FIG. 5

FIG. 4. Dependence of Δn_{st} in $LiNbO_3$ on the diameter of the light beam.

FIG. 5. Dependence of Δn_{st} in $LiNbO_3$ on the radiation power W.

thermo-ionization of the trap, B is the recombination probability, and τ_d is the time of electron drift from the illuminated region to the boundary of the beam. The drift time is $\tau_d = D/2\mu E_d$, where μ is the mobility and $E_d = E_p - E_{sp}$.

According to (1), to determine the change Δn of the refractive index it is necessary to determine n_t from (2) and (3). It is impossible in the general case to solve the system (2) and (3), but it is possible to find the stationary value $\Delta n_{st} \sim (N_t - n_t)_{st}$.

Chen[4] obtained an expression for Δn_{st} from a system of equations similar to (2) and (3). He assumed that the stationary conditions set in when the recombination rate exceeds the carrier drift velocity i.e., $B(N - n_t) \geq 1/\tau_d$. However, the expression obtained for Δn_{st} from this condition does not contain the experimentally observed temperature dependence. We start from the assumption that the carrier drift ceases and that a stationary value $(N_t - n_t)_{st}$ will be reached when the carrier density in the illuminated region, due to the photo- and thermo-ionization, becomes comparable with the thermal concentration n_0 of the electrons in the remaining part of the crystal.

From Eq. (2) with $\tau_d = \infty$ and $dn_e/dt = 0$ we obtain

$$n_e = (\alpha I + \beta) n_t / B(N - n_t). \tag{4}$$

Equating $n_e = n_0$, we obtain

$$n_t = BNn_0 / (\alpha I + \beta + Bn_0). \tag{5}$$

The thermal concentration of the electrons n_0 in the non-illuminated region of the crystal can be obtained from the detailed balancing principle, according to which the rate of thermal ionization is equal to the recombination rate

$$n_0 = \beta N_t / B(N - N_t). \tag{6}$$

Substituting (6) in (5) and recognizing that n_0 is given by the well-known expression

$$n_0 = N_t^{1/2} \left(\frac{2\pi mkT}{h^2}\right)^{3/4} e^{-\Delta\varepsilon/2kT}, \tag{7}$$

we obtain ultimately for the stationary concentration of the ionized traps

$$(N_t - n_t)_{st} = \alpha I N_t \left[\alpha I + B\frac{N}{N_t^{1/2}}\left(\frac{2\pi mkT}{h^2}\right)^{3/4} e^{-\Delta\varepsilon/2kT}\right]^{-1}. \tag{8}$$

We obtain an expression for Δn_{st} by substituting (8) in (1)

$$\Delta n_{st} = -f_{33} n_3^3 p \alpha I N_t \left[\alpha I + \frac{BN}{N_t^{1/2}}\left(\frac{2\pi mkT}{h^2}\right)^{3/4} e^{-\Delta\varepsilon/2kT}\right]^{-1}. \tag{9}$$

Expression (9) describes the dependence of the stationary change of the refractive index on the radiation intensity, sample temperature, and trap concentration.

For light of low intensity we can neglect the term αI in the denominator of (9); then

$$\Delta n_{st} = -\alpha I N_t^{3/2} e^{\Delta\varepsilon/2kT} / BN (2\pi mkT / h^2)^{3/4}. \tag{10}$$

It follows from (10) that Δn_{st} depends linearly on the light intensity and exponentially on the temperature, in agreement with experiment.

Comparing expression (10) with the experimental temperature dependence of Δn_{st}, we find that the energy of thermal ionization of the traps in $LiNbO_3$ is $\Delta\epsilon = 2\Delta_{eff} \approx 0.6$ eV. From the equality of the thermal-activation energies for crystals with different R it follows that the IOI in these crystals is apparently connected with electron traps of the same type.

For a high light intensity, when

$$\alpha I \gg B\frac{N}{N_t^{1/2}}\left(\frac{2\pi mkT}{h^2}\right)^{3/4} e^{-\Delta\varepsilon/2kT},$$

we obtain from (10)

$$\Delta n_{st} = -f_{33} n_3^3 p N_t, \tag{11}$$

from which it follows that the change of the refractive index is determined by the total ionization of the traps by the light and does not depend on the intensity. A saturation of Δn_{st} of this type was observed in[4] following irradiation of $LiNbO_3$ with an argon-laser beam of approximate intensity 500 W/cm². The maximum value of Δn_{st} measured in[4] is $\sim 10^{-3}$. Using this value, we can estimate from (11) the maximum value of $\Delta P_s = pN_t \approx 3 \times 10^{-3} P_s$. If we assume that p is of the same order as the average dipole moment per ion of the main lattice, then we obtain $N_t \approx 10^{18}$–10^{19} cm^{-3}. Such a trap concentration can be connected with defects of the main lattice, and not with the presence of uncontrollable impurities in the crystal, since the concentration of the latter is as a rule lower. This was indicated in[5].

It was noted earlier that the traps leading to optical damage in $LiNbO_3$ may be oxygen vacancies or excess Nb ions. The content of these defects, as shown in[7], changes when the crystal composition (the value of R) changes. The increase of Δn_{st} with increasing R, observed in our experiments, suggests that the defects leading to IOI are more readily connected with oxygen vacancies than with excess Nb ions.

The different endurance of $LiNbO_3$ and $LiTaO_3$ crystals to the action of the radiation may be connected both with the difference in the concentrations of such vacancies (i.e., with the different values of N_t) and with the different trap ionization energies $\Delta\epsilon$.

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