

$$|\Phi\rangle = \exp \left\{ \sum_k [\alpha_k^+ \alpha_{-k}^+ \gamma_{\alpha\alpha}(k, t) + \alpha_k^+ \beta_{-k}^+ \gamma_{\alpha\beta}(k, t) + \beta_k^+ \beta_{-k}^+ \gamma_{\beta\beta}(k, t)] \right\}.$$

and consequently all the mean values of the type

$$\langle 0 | \alpha_k^+ \alpha_k | \Phi \rangle, \langle 0 | \beta_k^+ \alpha_k | \Phi \rangle, \langle 0 | \beta_k^+ \beta_k | \Phi \rangle, \\ \langle 0 | \alpha_k^+ \beta_k | \Phi \rangle, \langle 0 | \alpha_k^+ \alpha_{-k}^+ | \Phi \rangle, \dots$$

vanish, thus justifying the discarding, mentioned in Sec. 4, of the mean values in the derivation of the fundamental equations.

¹⁾ We note that the classification of the contribution to any quantity by the degrees of the density of the reacting particles is not trivial, since the effective-mass law relates quantities of different order in density.

²⁾ The last formula was derived earlier by Balagurov and Vaks.^[7]

³⁾ Otherwise it is necessary to carry out an extra summation of the ladder diagrams of perturbation theory, to take into account the dynamics of motion with a potential $w_-(x)$. Our approximation is equivalent to the Born approximation in quantum mechanics.

⁴⁾ It will subsequently be shown that

$$a = \frac{N_A(t)}{V} = \left\langle \Phi_0 \left| \sum_k \alpha_k^+ \alpha_k \right| \Phi \right\rangle = \frac{\alpha_0}{V^{1/2}}, \quad \frac{N_B(t)}{V} = b(t) = \frac{\beta_0}{V^{1/2}}.$$

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Induced reflection and bleaching effects in electro-optic crystals

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Partial bleaching of a crystal or total reflection of light were observed during propagation of monochromatic laser radiation parallel to the optic axis of the ferroelectric LiNbO₃. It was found theoretically and experimentally that both effects are due to the appearance of a periodic refractive index grating under the influence of the transmitted and reflected (from the rear face) light waves. The nature of the effect is governed entirely by the direction of the polar axis of the crystal.

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§1. EXPERIMENTAL RESULTS

Action of monochromatic laser radiation of $\lambda = 5145 \text{ \AA}$ wavelength on LiNbO₃:Fe crystals produced the following effects: a beam of $I_0 \approx 1 \text{ W/cm}^2$ intensity incident normally parallel to the optic axis¹⁾ ($\mathbf{k}_0 \uparrow \uparrow \mathbf{c}$) resulted in almost complete transformation into a reflected beam. The intensity of the radiation transmitted by a crystal 0.3 cm thick was less than 5% although its initial value was 60%. The time dependences of the reflection R

$= I_r/I_0$ and transmission $F = I_t/I_0$ coefficients were determined (Fig. 1). The time taken to establish the final pattern decreased on increase of the intensity of light (Fig. 1). A change in the direction of the optic axis ($\mathbf{k}_0 \uparrow \uparrow \mathbf{c}$) altered drastically the nature of the effect. The crystal became bleached, i.e., the transmission coefficient F increased and the reflection coefficient R decreased. The maximum increase in F amounted to 14%. When the direction of incidence deviated from the normal, the reflection and bleaching effects of narrow

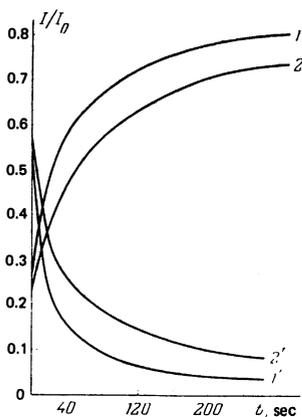


FIG. 1. Time dependences of the reflection (curves 1 and 2) and transmission (curves 1' and 2') coefficients for illumination intensities of 1.5 and 0.6 W/cm². Axis c \parallel k₀.

beams became much weaker. Similar behavior was observed for the reflection and transmission coefficients of a weak wave when the angle of incidence was varied after exposure of a crystal to strong illumination.

The above effects depended weakly on the conditions of the crystal surface; in particular, the experiments took place in air (on an insulated crystal) and in a conducting liquid. In the case of nonmonochromatic light neither effect appeared throughout the investigated range of intensities (up to 1 W/cm²).

§ 2. THEORY

These effects may be explained by assuming that two light waves interact in an electro-optic crystal.^[1,3] We must bear in mind that the presence of transmitted and reflected (from the rear face) light waves modulates the intensity of light with depth in the crystal. The inhomogeneous distribution of the intensity then produces variations of the refractive index which forms a volume phase hologram. A redistribution of the intensities of the light waves traveling through such a hologram results in further changes in this hologram and, finally, in localization of light near one of the faces of the crystal. The final distribution of the intensity is then unrelated to the presence of the initial (reflected from the rear face) wave, i.e., a self-conjugate holographic layer governed by the parameters of the crystal is formed. The appearance of such a steady-state pattern may be regarded as the result of the instability of a plane monochromatic wave in the investigated medium.

We shall now consider in greater detail the self-reflection and self-bleaching effects. We shall first obtain the equation for the amplitudes of the transmitted and reflected light waves. The initial one-dimensional wave equation is

$$\frac{\partial^2 \epsilon_{\perp} E}{\partial t^2} - c^2 \frac{\partial^2 E}{\partial z^2} = 0, \quad (1)$$

where ϵ_{\perp} is the transverse part of the permittivity tensor. If we assume that the change in ϵ_{\perp} is small,

$$\epsilon_{\perp} = \epsilon_0 + \Delta\epsilon(z, t), \quad \Delta\epsilon \ll \epsilon_0,$$

we can go over in Eq. (1) to the usual envelopes^[4]

$$E = \psi_+ e^{ikz - i\omega t} + \psi_- e^{-ikz - i\omega t}. \quad (2)$$

The expansion of $\Delta\epsilon$ may contain spatial periods which are multiples of the light-intensity period:

$$\Delta\epsilon = \frac{1}{2} \epsilon^0 + \epsilon^1 e^{2ikz} + \epsilon^2 e^{4ikz} + \dots + \text{c.c.}, \quad (3)$$

where ϵ^1 are slowly varying functions of z and t .

Substituting Eqs. (2) and (3) into Eq. (1), we obtain a system of coupled equations^[2]:

$$\frac{2i}{k} \frac{\partial \psi_+}{\partial z} + \frac{\epsilon^1}{\epsilon_0} \psi_- = 0, \quad \frac{2i}{k} \frac{\partial \psi_-}{\partial z} - \frac{\epsilon^{1*}}{\epsilon_0} \psi_+ = 0. \quad (4)$$

We have ignored the time derivatives of ψ_{\pm} assuming that the quasistationary condition $\omega t_0 \gg kz_0$ is satisfied (t_0 and z_0 are the characteristic time and scale of change of ψ_{\pm}); we have also ignored the absorption of light.

We can easily see that at every moment the system (4) retains the total wave energy flux, i.e.,

$$|\psi_+(z, t)|^2 - |\psi_-(z, t)|^2 = f(t). \quad (5)$$

The system (4) should be supplemented by the material equations for $\epsilon^1(\psi_{\pm})$. For crystals exhibiting the linear electro-optic effect we can write $\Delta\epsilon = \alpha E$, where α is the electro-optic constant and $E(z, t)$ is the electrostatic field. The nature of the nonlinearity of the medium is then governed by the expression for the current. We shall describe the latter by the formula^[2]

$$j = \frac{e \kappa \epsilon_0^{1/2}}{\pi \hbar \gamma} \left\{ \mu (E - E_{\infty}) + D \frac{\partial}{\partial z} \right\} |\psi_+ e^{ikz} + \psi_- e^{-ikz}|^2, \quad (6)$$

which allows for the diffusion and drift of electrons, and also for the photogalvanic effect.^[5,6] In Eq. (6), γ^{-1} is the carrier lifetime, μ is the mobility, $D = \mu T/e$ is the diffusion coefficient, and κ is the absorption coefficient. An expression for the current of the type (6) has often been used to analyze holographic records in electro-optic crystals.^[2,3,5]

Using Eq. (6) and a combination of the Maxwell and continuity equations

$$\frac{\partial}{\partial z} \left(\frac{\partial E}{\partial t} + \frac{4\pi j}{\epsilon_{\parallel}} \right) = 0 \quad (7)$$

(ϵ_{\parallel} is the static permittivity), we can find the field $E(\psi_{\pm}, z, t)$ and—having calculated its first harmonic—to complete the system (4).

We note that the higher harmonics of the electrostatic field $E^{2,3,\dots}$ are generally not small compared with E^1 . However, this does not give rise to higher spatial harmonics of light, i.e., the waves with $2k, 3k$, etc., since the Bragg (resonance) conditions are satisfied only by the first components of ψ_{\pm} .^[1,2] The small parameter for the higher harmonics is $\Delta\epsilon/\epsilon_0$.

We shall now consider the steady-state solutions of the system (4). For an isolated crystal the steady-state

condition implies $j=0$; we can then easily obtain from Eq. (6) the first Fourier harmonic of the electrostatic field.

$$E' = -2iE \frac{\psi_+ \psi_-^*}{|\psi_+|^2 + |\psi_-|^2 + |f|}. \quad (8)$$

Here, $E_{\text{diff}} = 2kT/e$ is the characteristic field governed by the diffusion process.

We shall first consider the reflection effect assuming that $\mathbf{c} \uparrow \uparrow \mathbf{k}_0$. In this case we may assume a crystal to be semi-infinite and postulate that under steady-state conditions we have $|\psi_+|^2 = |\psi_-|^2 \equiv |\psi|^2$. We then readily find from Eqs. (4) and (8) that

$$|\psi|^2 = |\psi_0|^2 \exp(-\alpha E_{\text{diff}} k z / \epsilon_0), \quad (9)$$

where $\psi_0 = \psi_+(z=0)$ is the amplitude of the incident wave. It follows from Eq. (9) that the intensity distribution is localized at a depth of the order of

$$l_{\text{diff}} = \epsilon_0 e / 2\alpha k^2 T.$$

At a temperature of $T = 300^\circ\text{K}$ for $\lambda = 5000 \text{ \AA}$ incident on lithium niobate crystals, we have $l_{\text{diff}} \approx 0.05 \text{ cm}$. The magnitude of the modulation of ϵ^1 is found, in accordance with Eq. (8), to be independent of z .

We shall finally consider the bleaching effect: $\mathbf{k}_0 \uparrow \uparrow \mathbf{c}$. When the direction of the crystal axis (the sign of α) is altered, solutions of the (9) type lose their meaning and the finite thickness of the crystal has to be allowed for explicitly. In this case, we can use the boundary condition on the rear face of the crystal ($z=l$):

$$|\psi_-(l)|^2 = \beta |\psi_+(l)|^2$$

$\beta = [(\epsilon_0^{1/2} - 1) / (\epsilon_0^{1/2} + 1)]^2$ is the reflection coefficient from the rear face} and we then obtain from Eq. (4)

$$\frac{|\psi_+|^2}{|\psi_0|^2} = \frac{1 - \beta [1 - \exp\{\alpha E_{\text{diff}} k (z-l) / \epsilon_0\}]}{1 - \beta [1 - \exp(-\alpha E_{\text{diff}} k l / \epsilon_0)]} \quad (10)$$

The light emerging from the crystal has the intensity

$$|\psi_{\text{out}}|^2 = (1 - \beta) |\psi_+(l)|^2 = \frac{(1 - \beta) |\psi_0|^2}{1 - \beta [1 - \exp(-\alpha E_{\text{diff}} k l / \epsilon_0)]} \quad (11)$$

i.e., smaller than $|\psi_0|^2$ but larger than $(1 - \beta) |\psi_0|^2$. For $l > l_{\text{diff}}$,

$$|\psi_+(l)|^2 \approx |\psi_0|^2 / (1 - \beta), \quad |\psi_-(0)|^2 \approx 0$$

and, therefore, the rear face of the crystal becomes completely bleached.³⁾

If a crystal is not insulated but is, say, placed in a conducting liquid, the condition $j = j_0 = \text{const}$ should apply in the steady-state case. The current j_0 is then found from the condition of absence of a potential difference between the faces of the crystal:

$$\int_0^l E dz = 0.$$

However, it is remarkable that neither allowance for

the short-circuiting of the faces of the crystal nor the influence of the photogalvanic effect alter, within the framework of the model (6), the steady-state distributions of light given by Eqs. (9) and (10). This can be demonstrated directly by calculating E^1 by means of Eqs. (6) and (12).

§3. DISCUSSION

The thickness of the reflecting layer can easily be estimated from the experimental data of Fig. 1 by the application of Eq. (9). If we assume that the intensity of light decreases exponentially, we find that

$$l_0 \approx 0.1 \text{ cm},$$

i.e., the result is in reasonable agreement with the theory. Bearing also in mind that in the case of lithium niobate we have

$$\beta = [(\epsilon_0^{1/2} - 1) / (\epsilon_0^{1/2} + 1)]^2 \approx 0.15,$$

we find that the observed increase in the transmission (by 14%) is also in agreement with the results of § 2.

The above ideas allow us to interpret also in a simple manner the other experimental results on the influence of light on LiNbO_3 crystals. Of special interest are the results of measurements of the currents flowing along the polar axis of a crystal under short-circuit conditions (Fig. 2). We can see that the current has transient and constant components. The constant component j_0 , corresponding to the photogalvanic current,^[5] depends strongly on the optic axis orientation. For a given illumination intensity the currents in the reflection and bleaching conditions are in the ratio 1:3. This result can be understood if we bear in mind that in the course of bleaching the photogalvanic current is generated by light of intensity $\sim I_0$ throughout the crystal, whereas in the reflection case it is created by light of intensity $\sim 2I_0$ (transmitted and reflected waves) in a relatively small part of the whole crystal. The ratio of the currents is readily estimated to be

$$j_r / j_t \approx 2l_0 / l.$$

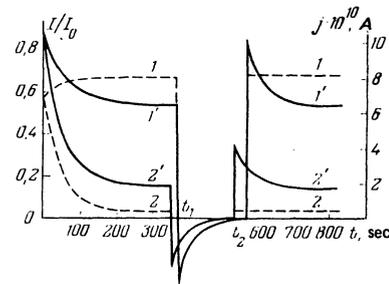


FIG. 2. Time dependences of the transmission coefficients (dashed curves) and currents (continuous curves) in the short-circuit regime obtained for various orientations of the polar axis of a crystal. Curves 1 and 1' correspond to the $\mathbf{k}_0 \uparrow \uparrow \mathbf{c}$ case and curves 2 and 2' to the $\mathbf{k}_0 \uparrow \uparrow \mathbf{c}$ case. The times t_1 and t_2 represent the end of one and the beginning of the next illumination. The intensity of the incident light was 0.6 W/cm^2 .

For the data of Fig. 2 and a crystal $l=0.3$ cm thick the reflecting layer is $l_0 \approx 0.05$ cm thick.

The large difference between the transient currents obtained under reflection conditions during the first and subsequent exposures can be explained in a similar manner. We must simply allow for the fact that after the end of illumination the grating of ϵ^1 remains because of the faster relaxation of the conductivity of the crystal.

We shall conclude by pointing out that the strong dependence of the nonlinear interaction of waves on the polar axis orientation is not specific to the problem under discussion. Such a dependence exists, for example, in the interaction of two waves incident on the surface of a crystal parallel to the optic axis.^[2-3] A common feature in such problems is the amplification of waves with a negative projection of the wave vector on the polar axis. In the final analysis, this result is due to the phase shift between the recorded gratings, i. e., it is due to the quarter-period mismatch between the modulations of the intensity and refractive index.^[8]

- ¹The direction of the c axis is determined by the fact that $E \parallel c$ alters the permittivity by an amount $\Delta\epsilon < 0$.
- ²The zeroth harmonic ϵ^0 is excluded from the system (4) by the phase shift.
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Formation of He^+ ions in various electronic states in $\text{He}^{+2} + \text{He}$ collisions

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One-electron capture ($\text{He}^{+2} + \text{He} \rightarrow \text{He}^+ + \text{He}^+$) and capture with ionization ($\text{He}^{+2} + \text{He} \rightarrow \text{He}^+ + \text{He}^{+2} + e^-$) in $\text{He}^{+2} + \text{He}$ collisions have been investigated for He^{+2} kinetic energies T_0 from 2 to 60 keV. To recognize the processes, both particles from a given collision were recorded by a coincidence technique. For the first time ever, the charge states of the particles were analyzed and the kinetic energy losses and scattering angle of the fast particle were measured for the same collisions. This made it possible to measure the cross sections for elementary processes involving changes in the charge states with the production of particles in definite electronic states and taking place at a fixed impact parameter. It is shown that in the capture process one of the He^+ ions is formed in an excited state with overwhelming probability ($\sim 75\%$ at $T_0 = 60$ keV and nearly 100% at $T_0 = 2$ keV). The energy distributions of the electrons released in capture-with-ionization processes corresponding to definite impact parameters were determined. The impact-parameter dependence of the distributions shows that the capture-with-ionization process is a result of the decay of an autoionization state of the He_2^{+2} quasimolecule. The internuclear distances at which the inelastic transitions take place (as determined from the scattering of the fast He^+ ions) agree with those expected on the basis of the terms of the He_2^{+2} system. It is concluded that the quasimolecular model of the interaction of atomic particles can account well for the features of the $\text{He}^{+2} + \text{He}$ collision.

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I. INTRODUCTION

The two-electron system $\text{He}^{+2} + \text{He}$ provides one of the simplest examples of the interaction of multiply charged ions with atoms and has been repeatedly investigated,

both experimentally and theoretically. Most of the experimental studies, however, were not based on the direct investigation of the processes taking place in $\text{He}^{+2} + \text{He}$ collisions, but on an analysis of the charge states of one of the particles (either the fast one or the slow