INFLUENCE OF ABSORPTION SATURATION ON POLARIZATION OF LIGHT PROPAGATING IN DICHROIC SINGLE CRYSTALS

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UDC 535

Relations that describe the polarization state of a light propagating in a uniaxial dichroic single crystal have been analyzed in the case of weak absorption, when Re $\varepsilon \gg$ Im ε, ε being the relative dielectric permittivity of the crystal. It has been found that those relations allow the state of light polarization to be predicted in the validity range of the linear optics laws, as well as provided that saturation and two-photon absorption are in operation. It has also been shown that those relations quantitatively describe the experimentally established regularities in a variation of such parameters of the light polarization ellipse as the angle of rotation of its major semiaxis, ellipticity, focal parameter, and eccentricity.

1. Introduction

For the excitation of uniaxial crystals while studying the saturation of absorption in them, as a rule, a linearly polarized emission with the direction of the polarization vector **E** of the electromagnetic wave being parallel or perpendicular to the optical axis C of the crystal is used. In this case, the anisotropy of absorption does not influence its saturation. If the value of the angle φ between the vector **E** and the axis C (the polarization azimuth) at the front face of the crystal differs from $n\pi/2$, where $n = 0, 1, 2, 3, \ldots$, the anisotropy of absorption reveals itself essentially through the character of the dependence of the transmission factor T on the intensity I_0 of exciting radiation [1]. In particular, this dependence is described in the framework of the two-level model with two kinds of absorbing particles [2].

It is known that, in the range of strong absorption, where the real part of the complex dielectric permittivity can be neglected in comparison with the imaginary one (Im $\varepsilon \gg$ Re ε), a gradual rotation of the vector **E** with respect to the optical axis C occurs as linearly polarized light propagates in a uniaxial crystal [3], but it is linearly polarized light that leaves the crystal. At the same time, in the ranges of crystal transparency and weak absorption, i.e. under the condition Re $\varepsilon \gg$ Im ε, light at the back face of the crystal is polarized elliptically [4]. The dependences of the polarization ellipse parameters (namely, the ellipticity, focal parameter, eccentricity, and angle of rotation of its longer semiaxis) on the polarization azimuth, wavelength, and factor of linear absorption are known in this case [4]. However, the variation behavior of those parameters at the transition from linear absorption to absorption saturation has not been studied enough. It is only known [1] that the polarization azimuth influences the threshold of absorption saturation.

This work aimed at studying the peculiarities of a variation of the light polarization characteristics as the light intensity increases up to values that correspond to the transition of a semiconductor into the state of absorption saturation. We have analyzed the relations that describe the polarization state of light when it, being in the spectral interval of crystal transparency or insignificant absorption, propagates through a uniaxial dichroic crystal; the intensity of light might be either low (linear optics) or high (nonlinear optics). Those relations have allowed us to describe quantitatively the experimental features of variations of the light polarization properties and the polarization ellipse parameters at the transition from the linear absorption to the absorption saturation mode in a typical uniaxial CdS crystal.

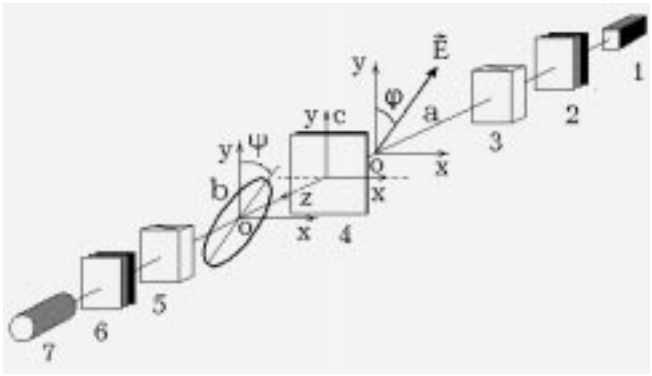

Fig. 1. Block diagram of measurement of the influence of absorption saturation on light polarization: a dye or helium-neon laser (1), sets of calibrated neutral grey filters (*2* and *6*), polarizers (Glan prisms) (*3* and *5*), lamellar CdS single crystal whose optical axis is parallel to the y-axis (*4*), photomultiplier ELU-FT (*7*). Insert *a* illustrates the orientation of the polarization vector $\mathbf{E}$ of an electromagnetic wave with respect to the optical axis C at the front face of the crystal, φ being the polarization azimuth. Insert *b* illustrates the shape of the polarization ellipse at the back face of the crystal, ψ being the angle of rotation of the longer semiaxis of the polarization ellipse with respect to the optical axis C after the light transmission through the crystal

2. Specimens and the Method of Measurement

Dichroic CdS single crystals 42 μm in thickness were studied. A linearly polarized light flux stroke normally the front crystal face which was parallel to the optical axis C of the crystal (Fig. 1). Single crystal *4* was placed between polarizer *3* and analyzer *5*. A fixed value of the polarization azimuth, i.e. the angle φ between the optical axis C aligned along the OY axis (inset, *a*) and vector $\mathbf{E}$, was set by rotation of the crystal.

Light sources were (i) a dye laser *1* with a pulse half-height duration of 10 ns and an emission band halfwidth of 0.1 nm and (ii) a helium-neon laser. A photomultiplier ELU-FT (*7*) served as a light detector. A variation of the light intensity was fulfilled by a displacement of neutral grey filters from the filter collection *2* located before the specimen to collection *6* behind it.

The dependence of the light intensity I on the angle of rotation of the analyzer about the coordinate axis z, that coincided with the direction of light propagation, was measured (Fig. 1). The analysis of this dependence allowed us to determine the values of the angle ψ, which correspond to the maximal, $I_{\max}$, and minimal, $I_{\min}$, output light intensities at the back face of the crystal, i.e. to investigate the peculiarities of the influence of the light intensity on a shape of the polarization ellipse.

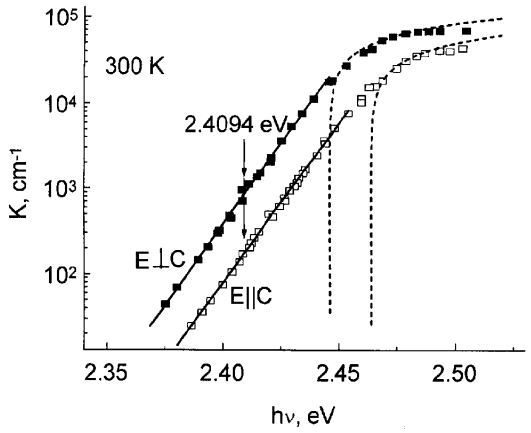

Fig. 2. Absorption spectra of a CdS single crystal. The number above the arrow indicates the photon energy of a laser emission, for which the influence of the light intensity on the light polarization state was investigated. Dashed curves correspond to the approximations of the dependences $K_\perp(h\nu)$ and $K_\parallel(h\nu)$ by the root law. Solid lines denote the Urbach sections of the corresponding absorption spectra

3. Propagation of Linearly Polarized Light in a Uniaxial Dichroic Single Crystal

Let a linearly polarized flux of radiation normally strike the front face of a weakly absorbing uniaxial dichroic crystal (Fig. 1). Formally, weak absorption means that the extinction coefficient is considerably smaller than 1 [4]. For CdS crystals, the condition of weak absorption is satisfied in a spectral interval where the dependence of the absorption factor on the photon energy is described by the Urbach rule (Fig. 2).

Let the optical axis of the crystal be parallel to its front face and aligned, as is shown in Fig. 1 (inset *a*). If the polarization azimuth $\varphi \neq n\pi/2$, where $n = 0, 1, 2, 3, \ldots$, we may assume that two components of the light flux, with $\mathbf{E} \perp C$ in one of them and $\mathbf{E} \parallel C$ in the other, propagate in the crystal in the same direction. At the back face of the crystal, the dependence of the light intensity I on φ and ψ is described by the equation [4]

$$I = I_0(1-R)^2 \times$$

$$\times\{\exp(-K_\perp d)\cos^2\varphi\cos^2\psi + \exp(-K_\parallel d)\sin^2\varphi\sin^2\psi +$$

$$+2\exp[-(K_\perp + K_\parallel)d/2]\sin\varphi\cos\varphi\sin\psi\cos\psi\cos\delta\}, \quad (1)$$

where $I_0(1-R)^2\exp(-K_\perp d) = I_\perp$ and $I_0(1-R)^2\exp(-K_\parallel d) = I_\parallel$ are the light intensity of the components with $\mathbf{E} \perp C$ and $\mathbf{E} \parallel C$, respectively, at the back face of the crystal; $K_\perp$ and $K_\parallel$ are the absorption

factors for those components, d is the thickness of the specimen, I_0 is the intensity of light at the front face of the crystal, R is the reflection factor, and δ is the phase incursion angle.

If the absorption factor depends on the light intensity nonlinearly, then, in order to calculate the values of $I_\perp$ and $I_\parallel$ in the range, where the Urbach rule is true, one must use relations that take into account the residual absorption K_h [5]:

$$I_{\perp,\parallel} = I_0(1-R)^2\exp(-K_l d), \quad I_0 < I_l,$$

$$I_{\perp,\parallel} = I_0(1-R)^2\exp(-K_l d)\times$$

$$\times\frac{I_l}{I_h}\left[1+\frac{K_h}{K_l}\left(\frac{I_0}{I_l}-1\right)\right]^{K_l/K_h}, \quad I_h < I_0 < I_l,$$

$$I_{\perp,\parallel} = I_0(1-R)^2\exp(-K_h d)\frac{I_l}{I_0}\left(\exp K_h d+\frac{I_0-I_h}{I_l}\right),$$

$$I_0 > I_h, \qquad (2)$$

where I_l and I_h are the light intensities at the beginning and the end of the absorption factor change interval, respectively; and K_l and K_h are the absorption factors measured in the range of low (linear optics) and high (the range of absorption saturation) intensities, respectively. The method for determination of the parameters I_l, I_h, K_l, and K_h is illustrated in Fig. 3.

In the range of interband transitions, to evaluate the values of $I_\perp$ and $I_\parallel$, one should use the relation [2]

$$(K_l-K_h)\ln\frac{K_l+K_h I/(1-R)I_l}{K_l+K_h I_0(1-R)/I_l}+$$

$$+K_h\ln\frac{I}{I_0(1-R)^2} = -K_l K_h d. \qquad (3)$$

In the range of uniaxial crystal transparency, where two-photon absorption plays an important role, $I_\perp$ and $I_\parallel$ are determined from the formula

$$I = \frac{(1-R)^2 I_0\exp(-K_1 d)}{1+(K_2 I_0/K_1)(1-R)[1-\exp(K_1 d)]}, \qquad (4)$$

where K_T is the coefficient of residual one-photon linear absorption and K_2 is the coefficient of two-photon absorption.

Provided that residual one-photon absorption is absent and the light intensity is low, so that the processes of two-photon absorption still may be neglected, formula (1) is transformed into the well-known relation [4]

$$I = I_0\cos(\varphi-\psi) - I_0\sin 2\varphi\sin 2\psi\sin^2\delta/2. \qquad (5)$$

To define the light intensity at the back face of the specimen in the intermediate intensity range, we ought to use one set of relations (e.g., formulae (1) and (2)) for one of the components and another set (e.g., formulae (1) and (4) or (5)) for the other.

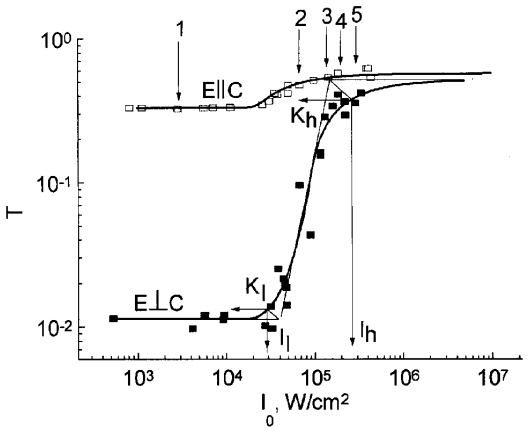

Fig. 3. Experimental (points) and theoretical (solid curves) dependences of the transmission factor T of a CdS single crystal 42 μm in thickness on the intensity of incident light I_0 for polarizations $\mathbf{E}\perp C$ and $\mathbf{E}\parallel C$. The theoretical curves were calculated using formula (2). The arrows mark the light intensity values I_0, at which the light polarization characteristics were measured: 2.97×10^3 (*1*), 6.72×10^4 (*2*), 1.38×10^5 (*3*), 1.95×10^5 (*4*), and 2.97×10^5 W/cm^2 (*5*). I_l and I_h are the intensities that correspond to the edges of the growing transmittance interval, K_l and K_h are the corresponding absorption factors

4. Influence of Absorption Saturation on Light Polarization

The formulae quoted above are valid in the range of weak absorption, where Re $\varepsilon \gg$ Im ε. Therefore, the wavelength $\lambda = 514.5$ nm ($h\nu = 2.4094$ eV), which corresponds to the central part of the Urbach section of the absorption spectrum (Fig. 2), has been selected for this research. For this wavelength, a typical dependence of the transmittance factor T on the light intensity is observed both for the $\mathbf{E}\perp C$ and $\mathbf{E}\parallel C$ polarizations (see Fig. 3). First, T does not depend practically on I_0. However, after the intensity I_0 having reached some threshold value I_l, an abrupt growth of the specimen transmittance is observed. After the intensity I_0 having reached another characteristic value I_h, T becomes

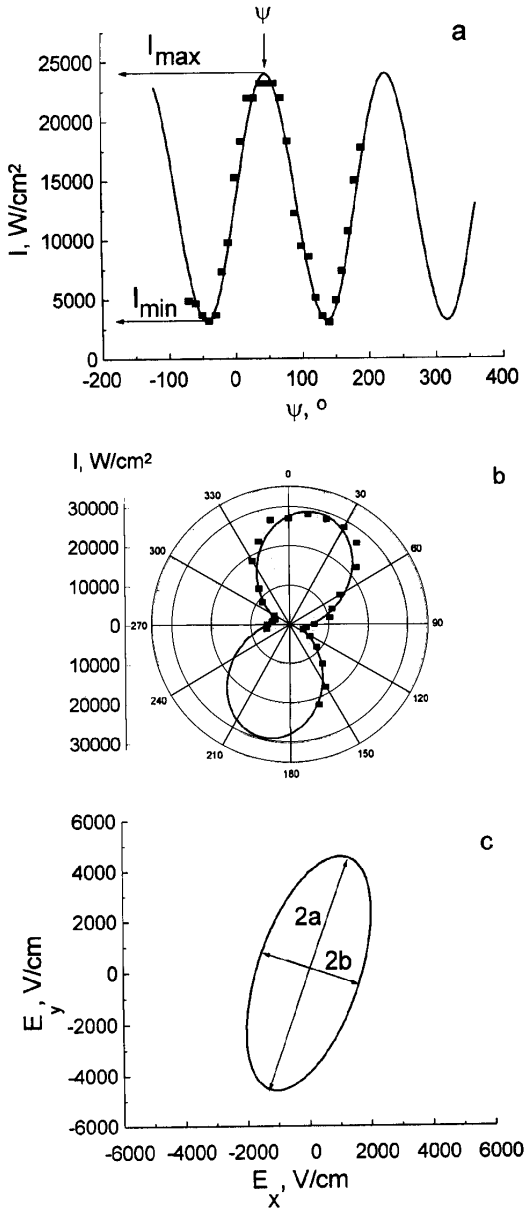

Fig. 4. Experimental (points) and theoretical (solid curves) dependences of the light intensity I_0 at the back face of the crystal on the angle of polarizer rotation ψ, plotted in Cartesian (a) and polar (b) coordinates, and a view of the polarization ellipse (c). The theoretical curves were calculated by formulae (1) and (2). The polarization azimuth $\varphi = 30°$. $I_0 = 1.95 \times 10^5$ W/cm^2

almost independent of I_0, which testifies to that absorption has come to its saturation value.

The researches of variations of the polarization properties of light at the transition from its linear absorption to the state of absorption saturation were carried out for several values of the intensity I_0 (marked by arrows in Fig. 3) and the polarization azimuth φ. Within the whole investigated intervals of I_0 and φ, the dependences of the light intensity I at the back face of the crystal on the angle of analyzer rotation ψ look like that shown in Fig. 4,a. The same dependence in polar coordinates is presented in Fig. 4,b. To put the results of calculations by formulae (1) and (2) (solid curves in Figs. 4,a and b) in agreement with experimental ones (points), we changed the phase incursion angle δ. At its optimal value, the error of approximation of the experimental dependences by the theoretical ones did not exceed 5%.

It was found that $\delta = 0$ at low intensities ($I_0 < I_l$) within the Urbach range of absorption. Within the same range, but provided absorption saturation ($I_0 < I_l$), and within the range of crystal transparency, $\varphi \neq 0$. In the general case, the phase incursion angle $\Delta = 2\pi n + \delta$, where $n = 0, 1, 2, 3, \ldots$, $\delta = 2\pi d(n_o - n_e)/\lambda$, n_o and n_e are the refractive indices for the ordinary and extraordinary rays, respectively, λ is the wavelength. Making use of the optimal value of δ, we estimated the difference $n_o - n_e$. It turned out that, for $n = 1$ in the formula $\Delta = 2\pi n + \delta$ and $\lambda = 632.8$ nm, $n_o - n_e = 1.65 \times 10^{-2}$, which differs from the value of 1.7×10^{-2}, quoted in [6] by no more than 3%; while for $\lambda = 514.5$ nm and at absorption saturation ($I_0 = 2.97 \times 10^5$ W/cm^2), $n_o - n_e = 1.37 \times 10^{-2}$, which differs from 1.7×10^{-2} [6] by no more than 20%. Some greater error in the latter case may be resulted from the influence of a large concentration of nonequilibrium current carriers which are generated by light at the intensities sufficient for attaining the state of absorption saturation, on n_o and n_e. In Eqs. (1) and (2), the influence of light intensity on the refractive index was not taken into account.

Making use of the known relation between the intensity I expressed in W/cm^2 units and the strength of the electric field E expressed in V/cm units [7]

$$E = 27.46 I^{0.5}, \qquad (6)$$

we found the shape of the polarization ellipse (Fig. 4,c).

The data presented in Fig. 4 allow the following parameters of the polarization ellipse to be determined: the phase incursion angle δ, obtained from the best fitting of the experimental dependences $I(\psi)$ by theoretical ones (Fig. 4, panels a and b), and the values of the angle ψ between the longer semiaxis of the ellipse and the axis C which correspond to the minimal ($I = I_{\max}$) and maximal ($I = I_{\min}$) amplitudes of the light intensity at the back face of the crystal (Fig. 4,a). An ensemble of these parameters obtained at different values of φ and I_0 allows the variation of light polarization properties at the transition from the

linear absorption to the absorption saturation mode to be traced.

The shape evolution of the polarization ellipse, which accompanies the variation of φ and I_0, is shown in the table. It is evident that the dependence of the shape of the polarization ellipse on the azimuth considered for the wavelength within the range of crystal transparency (the first row of the table) is similar to that obtained in the state of absorption saturation (the second row). The basic difference between them is different signs of polarization ellipse rotation. It is caused by the circumstance that a CdS single crystal is negative ($n_e > n_o$) at $\lambda = 632.8$ nm and positive ($n_o > n_e$) at $\lambda = 514.5$ nm (see [6]).

The key parameters that characterize the polarization ellipse are the orientation of the longer semiaxis of the ellipse with respect to the crystal optical axis C, whose quantitative characteristic is the angle ψ, ellipticity, eccentricity, and focal parameter. In the range of weak absorption, where the inequality $\mathrm{Re}\,\varepsilon \gg \mathrm{Im}\,\varepsilon$ holds good, the values of those parameters depend on the polarization azimuth and the light intensity. As the absorption factor reduces when absorption saturates, the values of the parameters

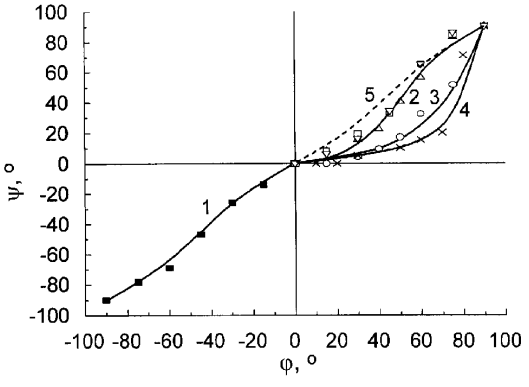

Fig. 5. Dependences of the angle of rotation ψ of the longer semiaxis of the polarization ellipse with respect to the optical axis C on the polarization azimuth φ for various values of the exciting light intensity $I_0 = 0.141$ ($\lambda = 632.8$ nm, solid squares), 2.97×10^5 ($\lambda = 514.5$ nm, hollow squares), 1.95×10^5 ($\lambda = 514.5$ nm, deltas), 1.38×10^5 ($\lambda = 514.5$ nm, nablas), 4.72×10^4 ($\lambda = 514.5$ nm, circles), and 2.97×10^3 W/cm^2 ($\lambda = 514.5$ nm, crosses). Curves *1* and *5* were calculated by formula (8), curves *2–4* by formula (7), with the absorption factors $K_\perp$ and $K_\parallel$ being estimated from formula (2)

Influence of the polarization azimuth and the intensity of exciting light on the shape of the light polarization ellipse at the back face of the CdS crystal

I_o, W/cm^2 (E_X; E_Y, V/cm)	Azimuth of polarization, degree						
	0	15	30	45	60	75	90
1.41×10^{-1} (1×10; 1×10) $n_o<n_e$							
2.97×10^5 (1×10^4; 1×10^4) $n_o>n_e$							
1.95×10^5 (1×10^4; 1×10^4) $n_o>n_e$							
1.38×10^5 (1×10^4; 1×10^4) $n_o>n_e$							
6.72×10^4 (5×10^3; 5×10^3) $n_o>n_e$							
2.97×10^3 (1×10^3; 1×10^3) $n_o>n_e$							

Notations. I_0 is the intensity of exciting light; E_X and E_Y are the scale of the Cartesian coordinates, in which the polarization ellipse is depicted.

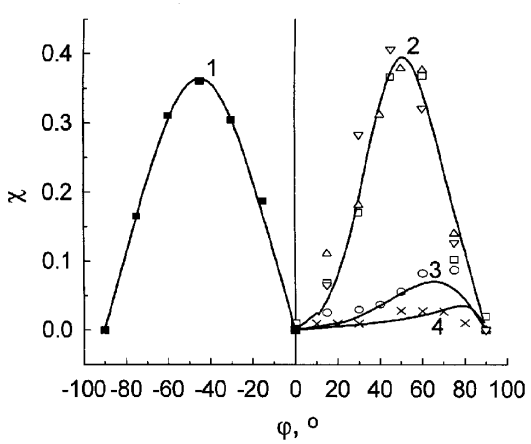

Fig. 6. Dependences of the ellipticity χ of the polarization ellipse on the polarization azimuth φ for various values of the exciting light intensity I_0 (the notations are the same as in Fig. 5). Curves *1–4* were calculated by formula (9), in which the values of $I_{\min}$ and $I_{\max}$ were determined by formula (1), where, in turn, the absorption factors $K_\perp$ and $K_\parallel$ were calculated by formula (2) and the angle ψ by formula (7)

concerned should also be expected to change. This conclusion is confirmed qualitatively by the table data, which illustrate the shapes of the polarization ellipse in a Cartesian coordinate system, the scale of the axes being indicated in the first column. To estimate these variations quantitatively, one has to calculate the relevant parameters and compare them with experimental values.

The dependence of the angle ψ on the polarization azimuth φ is described by the relation

$$\mathrm{tg}(2\psi) = \frac{\sin 2\varphi \cos\delta \exp[-(K_\perp + K_\parallel)d/2]}{\cos^2\varphi \exp(-K_\parallel d) - \sin^2\varphi \exp(K_\perp d)}, \qquad (7)$$

which results from Eq. (1). The values of the parameters $K_\perp$ and $K_\parallel$ at a fixed intensity I_0 were estimated with the help of formula (2). In the range of crystal transparency and in the state of absorption saturation, where $K_\perp = K_\parallel = 0$, Eq. (7) is simplified to the expression

$$\mathrm{tg}\,2\psi = \mathrm{tg}\,2\varphi \cos\delta. \qquad (8)$$

At $\delta = 0$, according to Eq. (8), a linear relation between the angles ψ and φ is to be expected. The phase incursion only modulates this dependence. The modulation depth grows with δ. It has been found empirically, and the calculation by formula (8) confirms that, in the range of the CdS single crystal transparency, the modulation depth is small at $\lambda = 632.8$ nm and $\delta = 40°$ (Fig. 5, curve *1* and the points around it). If the absorption saturates, $\delta = 42°$ at $\lambda = 514.5$ nm. In this case, the dependence of the angle ψ on the angle φ deviates to some extent from linearity. As the intensity I_0 decreases, the absorption factors $K_\perp$ and $K_\parallel$ affect it strongly. The larger the values of $K_\perp$ and $K_\parallel$, the stronger the deviation of the dependence of the angle ψ on the angle φ from a linear law (Fig. 5, curves *2–4* and the points around them).

The measure of the ellipse deviation from a circle is the ellipticity

$$\chi = b/a = (I_{\min}/I_{\max})^{1/2}, \qquad (9)$$

where b and a are the lengths of the shorter and longer semiaxes of the polarization ellipse, respectively (see Fig. 4,*c*), $I_{\min}$ and $I_{\max}$ are the minimal and maximal values, respectively, of the light intensity at the back face of the crystal (see Fig. 4,*a*). The intensities $I_{\min}$ and $I_{\max}$ were estimated with the help of formula (1), where the values of $K_\perp$ and $K_\parallel$ were calculated from formula (2) and the value of the angle ψ from formula (7). According to expression (9), the ellipticity χ, when cosidering the range of transparency of a uniaxial crystal and supposing the absorption saturation in it, is completely determined by the third term on the right-hand side of Eq. (1), the value of which depends on δ. In full accordance with formula (9), the ellipse degenerates into a straight line at $\varphi = 0$ and 90° (see the table). If the angle φ differs from either 0 or 90°, the ellipse approaches a circle by shape, as the parameter δ increases. For a fixed value of δ, which is 40° for $\lambda = 632.8$ nm and 42° for $\lambda = 514.5$ nm, the ellipticity grows as I_0 reduces or, to say it more correctly, as the absorption factors $K_\perp$ and $K_\parallel$ increase (see the table and Fig. 6).

The ellipse eccentricity

$$\xi = \frac{\sqrt{a^2 - b^2}}{a} = \sqrt{\frac{I_{\max} - I_{\min}}{I_{\max}}} \qquad (10)$$

depends on the value of the phase incursion angle, the polarization azimuth, and the light absorption factors for the $\mathbf{E} \perp C$ and $\mathbf{E} \parallel C$ orientations. The character of the dependence of ξ on two latter parameters is illustrated in Fig. 7. A tending of the eccentricity to unity as I_0 diminishes (Fig. 7) or, more correctly, as the absorption factors $K_\perp$ and $K_\parallel$ increase correlates with the growth of the ellipticity, because the values of both those parameters are determined by the ratio $I_{\min}/I_{\max}$.

The focal parameter

$$p = \frac{b^2}{a} = \frac{27.46 \cdot I_{\min}}{\sqrt{I_{\max}}} \qquad (11)$$

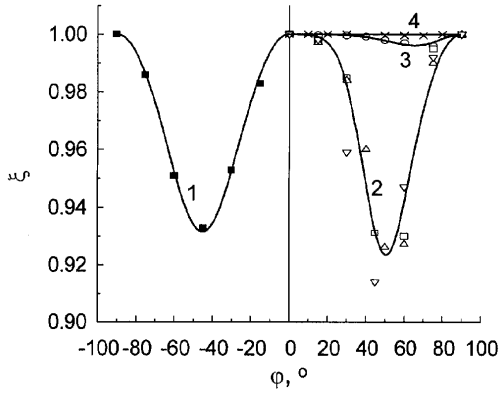

Fig. 7. The same as in Fig. 6 but for the eccentricity ξ of the polarization ellipse. The curves were calculated by formula (10)

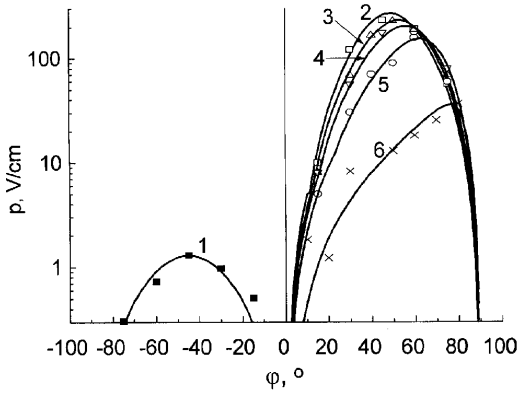

Fig. 8. The same as in Fig. 6 but for the focal parameter p of the polarization ellipse. The curves were calculated by formula (11)

defines the variation of the polarization ellipse at the points of its intersection with a chord drawn through a focus in parallel to the shorter axis, which is connected to the changes of the ellipse parameters that are caused by the action of the phase incursion, the polarization azimuth, and the light intensities in the range of the transition from a linear absorption to its saturation. The character of that variation is shown in Fig. 8. One can see that, at the gradual transition from the absorption saturation to the linear absorption mode, there occurs a gradual reduction of the focal parameter for all values of the polarization azimuth except for the points multiple of $\pi/2$.

Conclusions

For the simulation of the influence of the intensity of light that propagates in a uniaxial single crystal on the state of light polarization, we have used relations that made account of the absorption nonlinearity. They have allowed us to describe quantitatively the variation of the light polarization state at the transition from the range of linear absorption to the state of absorption saturation. These features should be taken into account when developing the absorption optical switches and while processing optical information.

It has been shown that the concordance of the calculation results with experimental ones can be achieved by selecting the optimal value for the phase incursion angle, the amplitude of which makes it possible to estimate the difference between the refractive indices for the ordinary and extraordinary rays both in the range of crystal transparency and in that part near the edge of the fundamental absorption band, where the absorption saturation is realized.

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Received 27.10.04.
Translated from Ukrainian by O.I.Voitenko

ВПЛИВ НАСИЧЕННЯ ПОГЛИНАННЯ НА ПОЛЯРИЗАЦІЮ СВІТЛА, ЩО ПОШИРЮЄТЬСЯ В ДИХРОЇЧНИХ МОНОКРИСТАЛАХ

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Р е з ю м е

Для випадку слабкого поглинання, коли $\mathrm{Re}\varepsilon \gg \mathrm{Im}\varepsilon$ (ε — відносна діелектрична проникність) проаналізовані співвідношення, що описують стан поляризації світла, яке поширюється в одновісному дихроїчному кристалі. Встановлено, що ці співвідношення дозволяють прогнозувати стан поляризації світла як в області виконання законів лінійної оптики, так і за наявності насичення та двофотонного поглинання, а також показано, що вони кількісно описують експериментально встановлені закономірності зміни таких параметрів еліпса поляризації світла, як кут повороту його великої півосі, еліптичність, фокальний параметр та ексцентриситет еліпса.