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Nonlinear interaction of the elliptically polarized light with CdS_xSe_{1-x} quantum dots

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Abstract. Peculiarities of the influence of ruby laser elliptically polarized light on the absorption saturation in glass with CdS_xSe_{1-x} nanocrystals and absorption saturation on light polarization were investigated. It was established that the change of ellipticity from zero to unity does not influence the dependence of transmission on the light intensity. Within the range of linear optics ($I_0 < I_1^*$, where I_0 is the intensity of pumping, I_1 – bleaching threshold) or within the range of absorption saturation ($I_0 > I_2^*$, where I_2^* is the absorption saturation threshold), light polarization at the entrance to glass with CdS_xSe_{1-x} nanocrystals is congruent to light polarization at the exit from glass. Within the range of a nonlinear dependence of absorption on the light intensity $\left(I_1^* < I_0 < I_2^*\right)$, deformation of polarization ellipse increases, when nanocrystals for which $E \perp C$ are bleached, but for nanocrystals where $E \| C$ are not bleached this deformation has a maximum value.

Keywords: nanocrystal, CdS_xSe_{1-x}, light polarization, polarization ellipse, absorption saturation.

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1. Introduction

CdS_xSe_{1-x} nanocrystals the size of which exceeds 6 nm have a hexagonal prism form [1]. They are chaotically placed and chaotically oriented in the volume of glass. If linearly polarized light propagates in this glass, the polarization azimuth φ (φ is the angle between the polarization vector **E** and nanocrystal axis *C*) can take any values within the range $-180° < \varphi < +180°$. In this case, we can describe interaction of linearly polarized light with CdS_xSe_{1-x} nanocrystals in the framework of a two-level model with two types of absorption particles [2]. It allowed us to find relations for describing the empirical dependences of transmission on the linearly polarized light intensity [3]. However, characteristic features of absorption saturation in glasses with CdS_xSe_{1-x} nanocrystals were not researched under conditions when these glasses were irradiated with the elliptically polarized light. The influence of saturation absorption on the form of polarization ellipse was not researched, too. These processes have been considered in this work.

2. Method and samples

Borosilicate glasses doped with $CdS_{0.13}Se_{0.83}$ nanocrystals (glass KC-19, 10 mm in thickness) were researched. Researches of the influence of light ellipticity on absorption saturation and influence of pumping on the form of polarization ellipse were performed using the set-up schematically depicted in Fig. 1.

If glass KC-19 is moved away and the angle of double Fresnel rhomb rotation is fixed, the dependence of transmission $T = I / I_0$ (I_0 and I are the light intensities at the exit from the single Fresnel rhomb and at the exit from the analyzer) on the analyzer rotation angle takes a look as in Fig. 2 (points). Approximation of this dependence (Fig. 2, solid curve) is carried out using the formula [4]

$$\frac{T(\psi)}{T_{\max}} = T_{\min} + (1 - T_{\min})\cos^2\psi , \qquad (1)$$

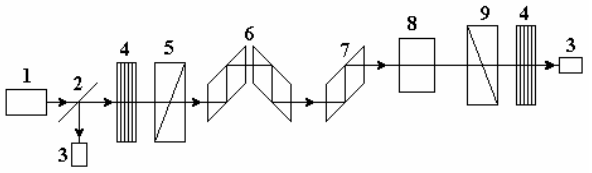


Fig. 1. Experimental set-up used to research the influence of elliptically polarized light on the absorption saturation and saturation absorption on the form of polarization ellipse: *1* – ruby laser, *2* – coupler, *3* – photomultiplier ЭЛУ-ФТ, *4* – set of calibrated neutrally-grey filters, *5* – polarizer (Glan prism), *6* – double Fresnel rhomb, *7* – single Fresnel rhomb, *8* – glass KC-19, *9* – analyzer.

where T_{max}, T_{min} are the maximal and minimum values of transmission. For the fixed value of I_0, we found the dependence $I = TI_0$ on ψ in Carthesian and polar coordinates (Table).

Taking advantage of the known relation between the light intensity and electric field E in the light flux [5]

$$E(\mathrm{V/cm}) = 27.46\sqrt{I(\mathrm{W/cm^2})}, \qquad (2)$$

we can find the form of polarization ellipse (Table).

We measured the dependence of the transmission on the light intensity (Fig. 3, points) when a sample was set between the single Fresnel rhomb and analyzer and when the analyzer rotation angle was fixed. The empiric dependence $T = f(I_0)$ was approximated by the curve (Fig. 3, solid curve) calculated using the formula

$$\frac{N_{01}N_{02}(\sigma_1-\sigma_2)^2\tau^2}{(\sigma_1N_{01}+\sigma_2N_{02})\sigma_1\sigma_2(N_{01}-N_{02})^2\tau^2}\times$$
$$\times\ln\frac{\sigma_1N_{01}+\sigma_2N_{02}+2\sigma_1\sigma_2(N_{01}+N_{02})\tau I_0T/(1-T)}{\sigma_1N_{01}+\sigma_2N_{02}+2\sigma_1\sigma_2(N_{01}-N_{02})^2\tau I_0(1-R)}+$$
$$+\frac{\ln T/(1-R)^2}{\sigma_1N_{01}+\sigma_2N_{02}}+\frac{2\tau I_0(1-R)\left\{\left[T/(1-R)^2\right]-1\right\}}{N_{01}+N_{02}}=-d. \qquad (3)$$

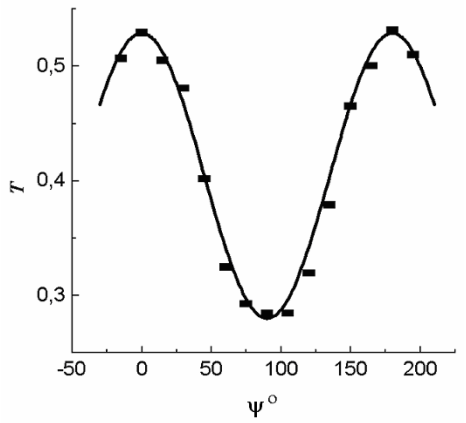


Fig. 2. Dependence of the transmission T (glass KC-19 is moved away) on the angle of analyzer rotation ψ.

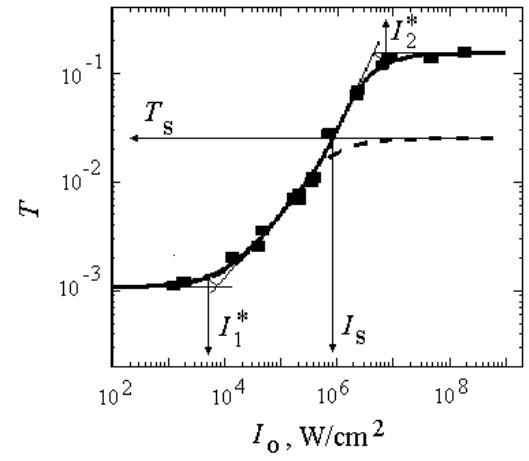


Fig. 3. Dependence of the transmission T for KC-19 glass on the intensity I_0. The value of ellipticity ξ = 0.75, the angle between analyzer polarization plane and light polarization plane at the entrance to the sample is equal 30°. Points represent experimental data, and the solid curve is a result of calculation by using the formula (3). The dependence $T = f(I_0)$ was calculated using the formula (4) and was represented in Fig. 3 by the dotted line. The data of the empiric method to find the bleaching threshold I_1^* and absorption saturation threshold I_2^* are illustrated in Fig. 3 with thin lines.

This formula is obtained in the two-level model with two types of absorption particles [2]. In the formula (3) R is the reflection coefficient, σ_1, σ_2 and N_{01}, N_{02} are the cross-sections and concentrations of centers that take part in absorption, accordingly for nanocrystals with $E \perp C$ and $E \| C$; τ is the lifetime of charge carriers; d is the thickness of a sample. Fulfilling this procedure for the set of analyzer rotation angles, we found the form of the polarization ellipse for a set value of the pumping intensity (Table).

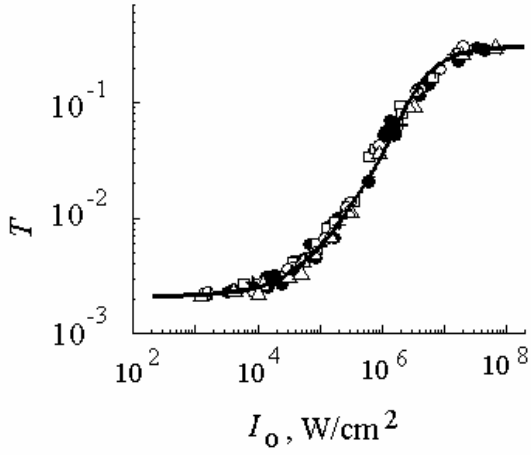
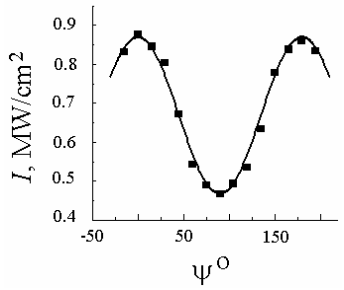
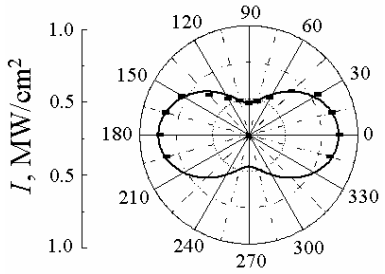
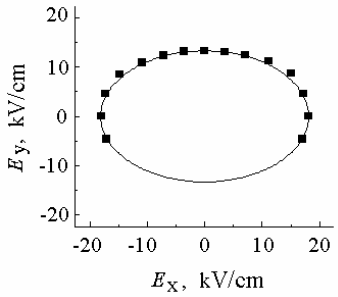
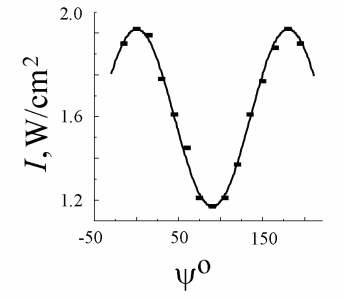
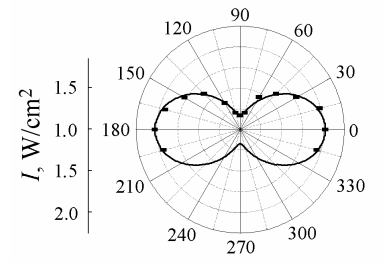
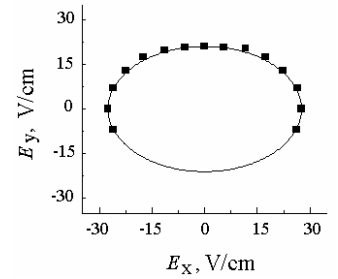
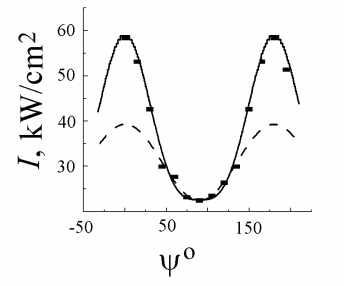
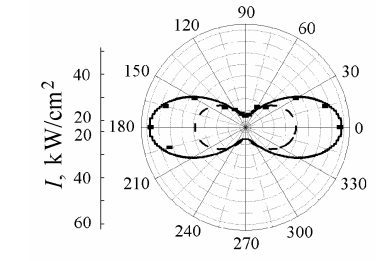
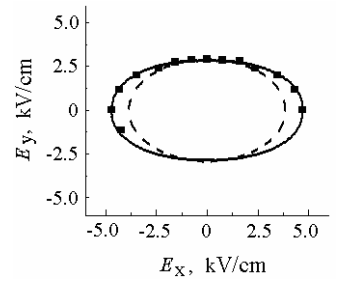
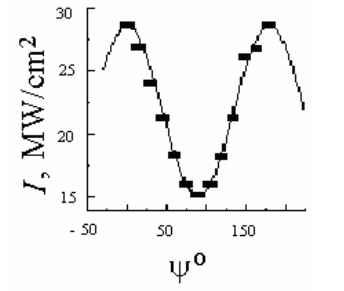
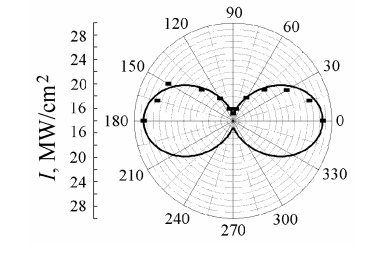
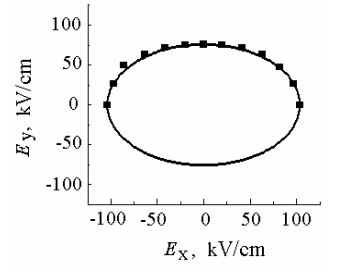


Fig. 4. Influence of the light ellipticity ξ on absorption saturation in KC-19 glass. The values of ellipticity are: ● – 0.00; □ – 0.25; ○ – 0.50; + – 0.75; Δ – 1.00.

Table. The influence of absorption saturation on light polarization.

I_0, W/cm^2	$I = f(\psi)$ (Carthesian coordinates)	$I = f(\psi)$ (polar coordinates)	E
$8.7\cdot10^{5*}$			
$1\cdot10^{3}$			
$8.7\cdot10^{5}$			
$1\cdot10^{8}$			

*Light polarization at the entrance to the sample is shown in the first row. We chose the intensity for which at the exit from glass we had the most pronounced changes in light polarization.

3. Influence of the light ellipticity on absorption saturation

In absence of the analyzer, for 0.00, 0.25, 0.50, 0.75, 1.00 values of ellipticity we measured the dependences $T = f(I_0)$ (Fig. 4). With the mean square error ≤±12%, these dependences coincide with each other and are described by the formula (3). It means that the degree of ellipticity does not influence on absorption saturation.

4. Influence of absorption saturation on the form of polarization ellipse

In our opinion, within the range of linear optics ($I_0 < I_1^*$) and range of complete absorption saturation ($I_0 > I_2^*$) (Fig. 3), the dependences of intensity on the angle of analyzer rotation (in Carthesian and polar coordinates) and the form of polarization ellipse at the entrance to glass are congruent to light polarization at the exit from this glass (Table). We think that the absorption saturation most strongly influences on the form of polarization ellipse, when nanocrystals for which $E \perp C$ are bleached, but nanocrystals for which $E \parallel C$ are not bleached. To find the intensity $I = I_S$, when this situation will be realized, we will calculate the dependence $T = f(I_0)$ by using the formula [2]

$$K_1 \ln\frac{K_1 + K_2 + K_2 I_0 T/(1-R) I_1^*}{K_1 + K_2 + K_2 I_0 (1-R) I_1^*} + \\ + K_2 \ln\frac{T}{(1-R)^2} = -K_2 (K_1 + K_2) d\,, \qquad (4)$$

where K_1 and K_2 are the values of the linear absorption coefficient for nanocrystals with $E \perp C$ and $E \parallel C$. In calculations, it was considered that K_2 did not depend on the light intensity. Then, we determined the value of transmission T_S (Fig. 3), after reaching it nanocrystals with $E \perp C$ were bleached. The value T_S corresponds to I_0. For $I_0 = I_S$, we determined in Carthesian and in polar coordinates the dependence of intensity I at the exit from the analyzer versus the angle of its rotation (Table). For comparison, the analogical dependence was found for $I_0 = 1.0 \cdot 10^3$ W/cm^2 (nearby upper range limit of linear optics) (Table), and for $I_0 = 10^8$ W/cm^2 (range of absorption saturation) (Table). It was found that these dependences and forms of the polarization ellipse are congruent.

Another situation will be realized in the range of nonlinear absorption. When I_0 is increased from I_1^* up to I_2^*, we observe the gradual distortion of the sinusoidal dependence I on ψ and forms of polarization ellipse. When $I_0 = I_S$, the distortions are maximal (Table). These results can be explained using the following model of light interaction with a set of uniaxial crystals that are chaotically oriented inside the bulk.

5. Model

Let linearly polarized light flux falls perpendicularly on the entrance face of an uniaxial single crystal. If the azimuth of polarization $\varphi \neq n\pi/2$, where $n = 0, 1, 2, 3, \ldots$, it is possible to consider that in the same direction two components of light flux travel inside this single crystal. In one of them $E \perp C$, and in the other $E \parallel C$. At the exit from the analyzer, the dependence of the intensity I on φ is described with the equation [3]:

$$I = I_\perp \cos^2\varphi \cos^2\psi + I_\parallel \sin^2\varphi \sin^2\psi + \\ + 0.5\sqrt{I_\perp I_\parallel}\sin(2\varphi)\sin(2\psi)\cos\delta\,, \qquad (5)$$

where $I_\perp$, $I_\parallel$ are intensities of light components at the exit from the crystal, for one of which $E \perp C$ and for the other – $E \parallel C$, $\delta = 2\pi(n_e - n_0)d/\lambda$ is the angle of phase lag, ψ is a rotation angle of the analyzer in respect to the polarizer, n_e and n_0 is the refraction indexes of extraordinary and ordinary waves, respectively. According to the formula (5), two factors determine polarization of light leaving the crystal: phase lag and different value of change in $I_\perp$ and $I_\parallel$, when light travels inside the crystal.

Let elliptically polarized light falls on the glass with nanocrystals. Let the lengths of major semiaxis (a) and minor semiaxes (b) are known (Fig. 5). Rotating the analyzer, find an angle for which the intensity at the exit from the analyzer is maximal. We set this angular position of the analyzer equal to zero. Rotating the analyzer by the angle α in relation to the major semiaxis of the polarization ellipse, we choose the light beam with a certain value of polarization. This angle α defines the position of the electric field vector $\mathbf{E}_\alpha$.

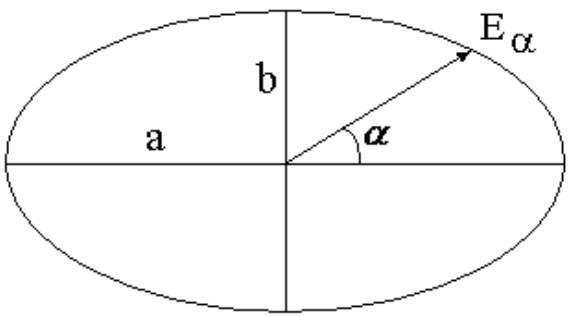


Fig. 5. Ellipse of light polarization.

We will consider that elliptically polarized light consists of a set of linearly polarized light beams that differ one from another only by the angle α. Each of these beams interacts with a large amount of nanocrystals. They can be separated into three large groups. To the first one, we will take nanocrystals for which the polarization azimuth equals to zero, i.e. nanocrystals with $E_\alpha \| C$. The second group contains nanocrystals for which $\varphi = \pm 90°$, i.e. nanocrystals with $E_\alpha \perp C$. The third group consists of all other nanocrystals. In every nanocrystal from the third group, two components of a light beam travel in the same direction. For one of them $E \perp C$, and for the other – $E \| C$. Different velocities of their travel (phase factor) and different values of the decreasing intensity (amplitude factor) are reasons of rotation of the polarization vector in every individual nanocrystal.

Phase factor. If in one nanocrystal from the third group, one component provides phase retardation with respect to other by the angle δ, there always will be another nanocrystal (as a consequence of availability of nanocrystal dispersion in size) with the angle equal to 180°–δ. In this case, phase terms (the third term in the right part of the formula (5)) for these nanocrystals are identical in their value, but opposite in sign. As a result, the total amount of phase terms is equal to zero. Hence, we can think that the set of nanocrystals consists of a pair in each of which the compensation phase factor is realized. It means that in glasses with the chaotically oriented uniaxial crystals, the phase factor does not influence on light polarization.

Amplitude factor. Influence of the electric field on polarization of light traveling inside glass with nanocrystals should be considered for the case of intensities that do not influence (region of linear optics and region of absorption saturation) and influence (region of nonlinear absorption) on the value of the light absorption coefficient.

Let the light flux from the region of linear optics ($I_0 < I_1^*$) or from the region of absorption saturation ($I_0 > I_2^*$) falls on the arbitrarily chosen nanocrystal, the polarization azimuth being $\varphi \neq n\pi/2$ ($n = 0, 1, 2, 3, \ldots$). In the set of nanocrystals, there always will be a nanocrystal with the azimuth 180°– φ. Select this value of φ, then at the entrance to the nanocrystals $E_{1\perp} = E_{2\perp} = E_{1\|} = E_{2\|}$ (Fig. 6a). As light travels inside the nanocrystal, the value of this component decreases in accord to the

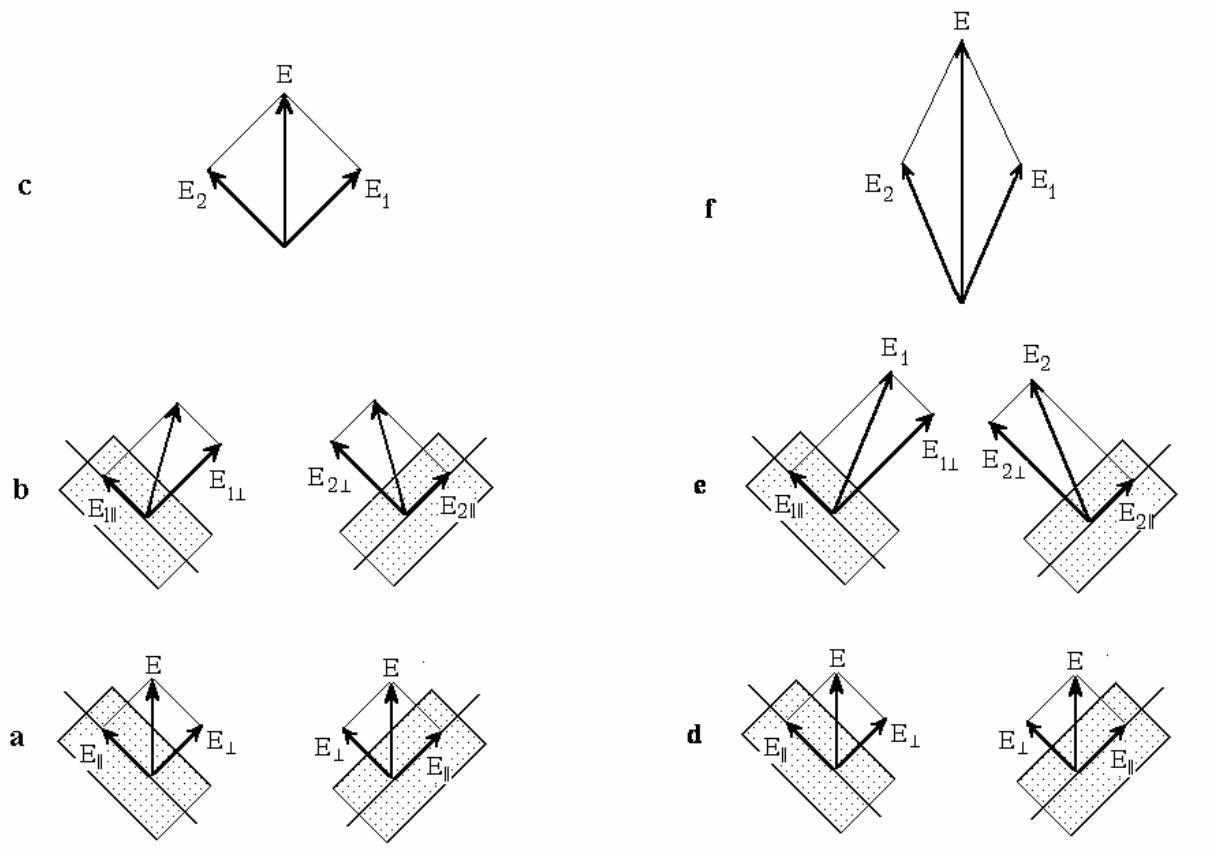


Fig. 6. Illustration of the influence of amplitude factor on changing the electric field value. **a**, **b**, **c** corresponds to the region of linear optics; **d**, **e**, **f** – to the region of light-intensity influence on the absorption coefficient; **a**, **d** is the electric field near the entrance surface of the sample; **b**, **e** is the electric field near the output surface of the sample; **c**, **f** is the total electric field after light passing through two nanocrystals.

Lambert-Buger law and is determined by the absorption coefficients $K_\perp$ and $K_\parallel$. As a result, at the exit from the nanocrystals of $E_{1\perp} = E_{2\perp}$, $E_{1\parallel} = E_{2\parallel}$, but $E_{1\perp} \neq E_{2\perp}$, $E_{1\parallel} \neq E_{2\parallel}$ (Fig. 6b). It means that in the first nanocrystal the total vector rotates by a certain angle and, in the second one, it rotates by the same angle but in the opposite direction (Fig. 6b). The total light vector (Fig. 6c) that goes out from both nanocrystals has the angular position identical to that at the entrance to nanocrystals (Fig. 6a). Thus, linear absorption coefficients do not change angular position of light beams from which the polarization ellipse consists of. As a result, in the course of light absorption the amplitude of each of these beams decreases at the same time. It means that, in the region of linear optics and region of absorption saturation, the form of polarization ellipse at the exit from glass with CdS_xSe_{1-x} nanocrystals is congruent to that of ellipse at the entrance.

Somewhat different situation is realized within the region of nonlinear light absorption ($I_1^* < I_0 < I_2^*$). We consider again that there are two nanocrystals at the entrance of one of which the polarization azimuth is equal to φ and at the entrance of the other is equal to 180°– φ. The electric fields at the entrances of these crystals are identical, i.e. $E_{1\perp} = E_{2\perp} = E_{1\parallel} = E_{2\parallel}$ (Fig. 6d). Linear and nonlinear absorption will cause this change of the electric field that $E_{1\perp} \neq E_{2\perp}$, $E_{1\parallel} \neq E_{2\parallel}$, but $E_{1\perp} = E_{2\perp}$, $E_{1\parallel} = E_{2\parallel}$ (Fig. 6e), i.e. in the case of nonlinear absorption for the angular position of beams of polarization ellipse at the output (Fig. 6f) of glass with CdS_xSe_{1-x} nanocrystals identical in their angular position at the entrance (Fig. 6d). As the value of changes in absorption for these nanocrystals depends on the light intensity, the value of changes in the amplitude of **E** vector depends on the light intensity. Hence, the electric field for those beams changes as a consequence of nonlinear absorption. It means that with increasing the light intensity in nanocrystals with $E \perp C$, bleaching is gradually increased but in nanocrystals with $E \parallel C$ this bleaching is absent. It causes changes in the length of **E** vector in the major semiaxis of polarization ellipse, i.e. the polarization ellipse is deformed. This deformation is maximal when nanocrystals with $E \perp C$ are fully bleached, but bleaching of nanocrystals with $E \parallel C$ is absent. The further increase in the light intensity leads to decreasing $K_\parallel$. As a result, deformation of the polarization ellipse gradually decreases. When I_0 becomes equal to I_2^*, the form of polarization ellipse at the output of glass becomes congruent to the form at its entrance. When the intensity continues to increase, congruence is kept.

6. Conclusions

It is found that linear absorption does not influence on the polarization state of light travelling in medium doped with uniaxial nanocrystals. The form of polarization ellipse is deformed, when the light intensity is able to change the absorption coefficient. These results must be taken into account when researching the polarization properties of materials that operate under conditions of high levels of light intensities.

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