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DYNAMICS OF LIGHT-ABSORPTION VARIATIONS INDUCED IN A BISMUTH SILICATE CRYSTAL BY VISIBLE LASER ILLUMINATION

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We present the results of experimental studies of the dynamics of the photoinduced optical absorption in a bismuth silicate crystal subject to continuous laser irradiation with wavelengths of 532 and 655 nm. The semiconductor-laser light beam with the wavelength $\lambda = 655$ nm causes the crystal bleaching at this wavelength, whereas its exposure to a shorter-wavelength irradiation from the optical-spectrum green region with a wavelength of 532 nm increases the optical absorption at both wavelengths, of 532 and 655 nm. The experimental results are interpreted using the theoretical model which assumes that the crystal has deep defect centers of two types so that an electron at each of these centers can be in one of the states characterized by different photoionization cross sections.

1. INTRODUCTION

The sillenite-type $Bi_{12}MO_{20}$ crystals, where M = Si, Ge, and Ti, are widely used as photorefractive materials for dynamic holography [1, 2]. The photoinduced optical-absorption variations inherent in these crystals, i.e., the photochromic effect, are related to the complicated energy structure of their defect centers [1, 3, 4], which in turn affects both the dynamics of formation of the photorefractive holograms and the photorefractive sensitivity. The photoinduced distribution of the charge carriers (electrons) over the defect centers, which is accompanied by the photochromic effect, can be one of the mechanisms of control of the above-mentioned parameters. For example, if the bismuth titanate crystals are irradiated from the visible range of the electromagnetic-radiation spectrum, then not only the photoinduced variations occur in the spectrum of their optical absorption [4], but also the photorefractive sensitivity in the near infrared (IR) region increases [1, 5]. In this respect, the development of the quantitative models which describe the photoinduced effects in the sillenite crystals, is of interest for realizing the dynamic-holography effects in the long-wavelength region of the visible spectrum and the near-infrared region.

In this work, we present the results of the studies of the dynamics of the photoinduced optical absorption, which is observed in the bismuth silicate crystal $Bi_{12}SiO_{20}$ when it is exposed to continuous-wave laser irradiation with wavelengths of 532 and 655 nm.

2. EXPERIMENTAL FACILITY AND TECHNIQUES

The experimental setup for studying the dynamics of photoinduced absorption, which uses two laser sources in an undoped $Bi_{12}SiO_{20}$ (BSO) crystal with section (100) and thickness $d = 7.3$ mm, is shown in

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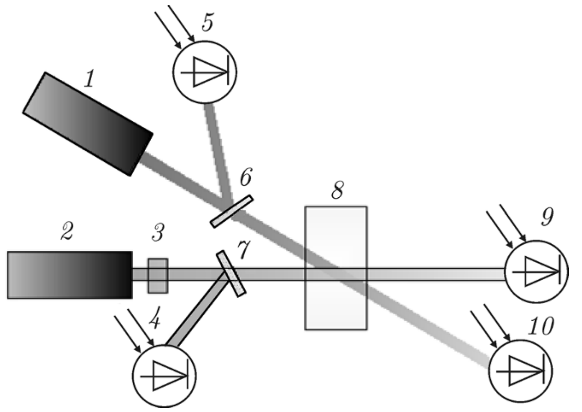


Fig. 1. Optical scheme for observing the dynamics of photoinduced variations in the absorption coefficient: semiconductor laser diode with an operating wavelength of 655 nm (*1*), continuous-wave Nd:YAG laser with a radiation wavelength of 532 nm (*2*), collimator (*3*), beam-splitting plates (*6* and *7*), BSO sample (*8*), and FD-24K photodiodes (*4*, *5*, *9*, and *10*).

Fig. 1. The light beams from continuously operating semiconductor laser *1* with the wavelength $\lambda_r = 655$ nm and the radiation-flux density $I_r = 30$ mW/cm^2 at the crystal input face and continuously operating solid-state laser *2* with $\lambda_g = 532$ nm and $I_g = 200$ mW/cm^2 were used for both illumination of crystal *8* and monitoring the photoinduced variations in its optical absorption. The induced variations in the crystal transmission at a wavelength of 655 nm were measured by photodiodes *5* and *10* and glass beam-splitting plate *6*. Photodiodes *4* and *9* and the beam-splitting plate *7* were used for such measurements at a wavelength of 532 nm. Intersection of the used light beams across the entire crystal thickness was ensured by collimator *3* and adjustment of their relative locations. The time dependences of the photodiode currents were measured using the three-channel data-processing computer system [6], which allowed us to specify the intervals from 0.1 to 10 s between the readouts. The identical FD-24K photodiodes, which were used in the experimental setup (Fig. 1) for both wavelengths, had different values of the current monochromatic sensitivity S_{ph}. However, the time variation $\Delta k_d(t)$ in the absorption coefficient for each wavelength was calculated according to the relationship

$$\Delta k_d(t) = \frac{1}{d} \ln \frac{I_S(t)/I_K(t)}{I_S(0)/I_K(0)}, \tag{1}$$

which rules out the influence of the differences in S_{ph} on the measurement results. In Eq. (1), I_S is the intensity of the radiation transmitting through the crystal, I_K is the radiation intensity for the check channel, and d is the crystal thickness.

Two techniques were used in the experiments on the study of the absorption-coefficient dynamics in the BSO crystal. In the first technique, the photoinduced absorption variations were caused by a light beam with the wavelength $\lambda_g = 532$ nm, while the induced absorption variations were monitored by the sample transmission of radiation with another wavelength $\lambda_r = 655$ nm using optical diode *10*. Optical diode *5* ensured control over the laser output power.

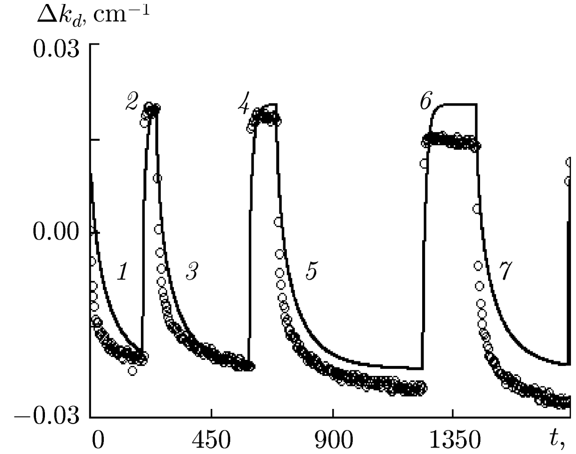
The second technique allowed us to study the dynamics of the natural photoinduced variations for a single wavelength. In this case, the crystal transmission was recorded using photodiodes *5* and *10* for radiation with the wavelength $\lambda_r = 655$ nm and photodiodes *4* and *9* for the wavelength $\lambda_g = 532$ nm.

3. EXPERIMENTAL RESULTS

The dynamics of the absorption coefficient Δk_d for the $Bi_{12}SiO_{20}$ crystal is shown in Fig. 2. The experiment started with irradiation of the crystal by the sounding laser radiation with the wavelength $\lambda_r = 655$ nm at time $t_0 = 0$ after which its bleaching began to occur (Fig. 2, region *1*). At $t_1 = 200$ s, the illumination radiation with a wavelength of 532 nm was switched on, which acted on the crystal up to the time $t_2 = 250$ s and comparatively rapidly increased the optical absorption (Fig. 2, region *2*). The subsequent experimental stages in regions *3*, *5*, and *7* are characterized by bleaching of the crystal, when it is affected only by the sounding radiation from the red region of the spectrum ($\lambda_r = 655$ nm), and its darkening at this wavelength due to the effect of the radiation with $\lambda_g = 532$ nm (Fig. 2, regions *4* and *6*).

Figure 2 shows that the crystal bleaching $-\Delta k_d$, which is reached under the action of radiation of the red spectrum region, increases from 0.021 cm^{-1} in region *1* to 0.028 cm^{-1} in the last region *7*. At the

Fig. 2. Dynamics of variations in the light-absorption coefficient in the $Bi_{12}SiO_{20}$ crystal for a sounding beam with a wavelength of 655 nm, which has an input flux density of 30 mW/cm^2 during the entire experiment ($t = 0$–1800 s), with the switch-on of the laser irradiation with a wavelength of 532 nm and a flux density of 200 mW/cm^2 on the input edge of the crystal for the time intervals $t = 200$–250 s (region *2*), 600–700 s (region *4*), and 1250–1 450 s (region *6*). The circles and the solid curve denote the experimental data and the numerical-simulation results, respectively.



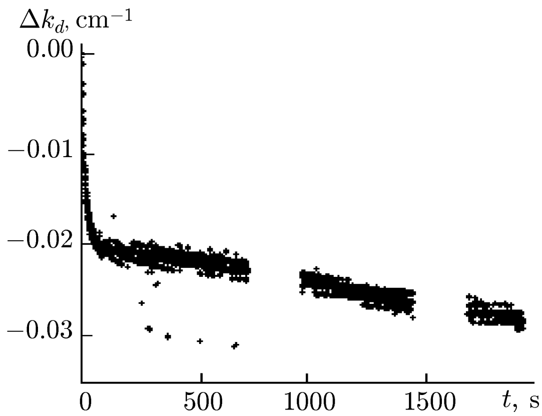


Fig. 3. Dynamics of variations in the light-absorption coefficient for the fixed wavelength $\lambda_r = 655$ nm.

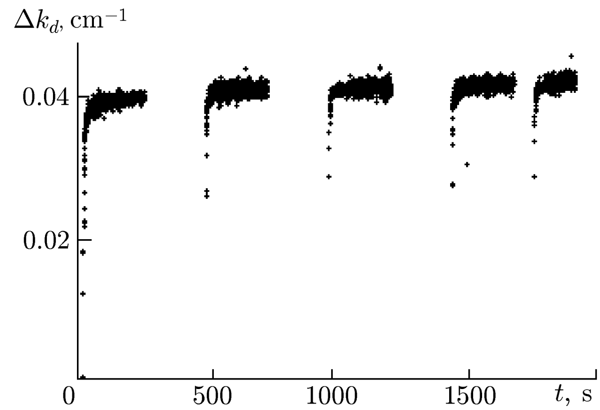


Fig. 4. Dynamics of variations in the light-absorption coefficient for the fixed wavelength $\lambda_g = 532$ nm.

same time, the crystal darkening caused by the short-wavelength laser radiation decreases from 0.020 to 0.015 cm^{-1}.

In the experiments on the study of the dynamics of the natural photoinduced absorption variations due only to light with the wavelength $\lambda_r = 655$ nm, radiation with flux density increased up to about 120 mW/cm^2 was used. The results of one of such experiments are depicted in Fig. 3. It is seen in the figure that the absorption coefficient rapidly (for about 30 s) decreases by a value of about 0.019 cm^{-1} after the crystal-irradiation onset at $t = 0$. Then a slower crystal bleaching occurs and continues in the absence of irradiation, which was switched off at the times $t_{off} = 720$ and 1440 s and again switched on at $t_{on} = 960$ and 1680 s. During the entire experimental time (1920 s), the absorption-coefficient variations reached -0.029 cm^{-1}.

Figure 4 shows the dynamics of the natural variations in the absorption coefficient of the studied crystal $Bi_{12}SiO_{20}$ for green light with $\lambda_g = 532$ nm and the input flux density about 200 mW/cm^2. It is seen in the figure that a photoinduced increase in optical absorption is observed for the above-mentioned wavelength. However, in contrast to the observed behavior of absorption at a wavelength of 655 nm (Fig. 3), the end of the crystal irradiation with $\lambda_g = 532$ nm at the times $t_{off} = 240$, 720, 1200, 1680 and 1920s is accompanied by the dark relaxation of the induced variations by about 0.015 cm^{-1} for the time intervals about 250 s, which last up to the next switch-on of the irradiating laser beam. As in the previous case, fast absorption variations at the initial region (its increase amounts to 0.037 cm^{-1} for 15 s) are replaced by a comparatively slow absorption increase.

4. ANALYSIS AND DISCUSSION OF THE RESULTS

The dynamics of the photoinduced light absorption in the $Bi_{12}SiO_{20}$ crystal is described by the model [7, 8] which assumes the presence of two types of the electrically neutral deep defect centers 1 and 2 such that an electron at each center can be in one of the states D and T characterized by the smaller and greater values of the photoionization cross section, respectively. According to the band diagram in Fig. 5, the first-type center (with the electron energies D_1 and T_1) is deeper than the second one (with the electron energies D_2 and T_2).

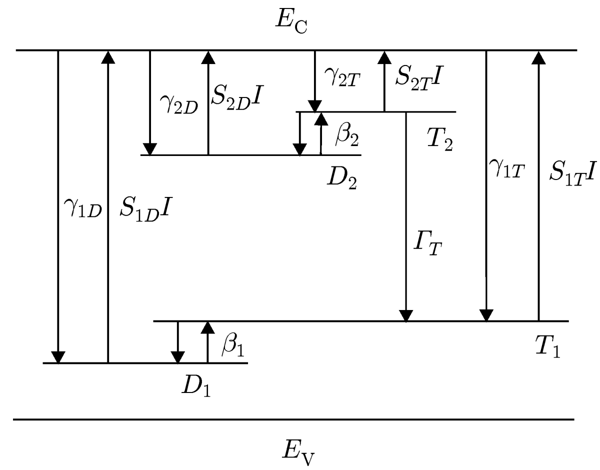


Fig. 5. Diagram of the energy levels in the forbidden energy band of the crystal.

The probability of the electron localization in the states D or T, each of which is characterized by its own potential well, is determined by the crystal temperature. Due to the electron–phonon interaction and the close spatial location of these two wells, the electron can tunnel through the separating barrier. It is assumed that some of the above-mentioned centers are ionized under the dark conditions and their charge is compensated by the radiation-insensitive acceptors with number density N_A. Under the action of light, the electrons move from the nonionized centers to the conduction band and then recombine with the ionized deep centers to the states D or T. Within the framework of this model, for the light with a wavelength of 532 nm whose energy quantum $\hbar\omega$ allows one to excite electrons to the conduction band from all centers D_1, D_2, T_1, and T_2, the absorption coefficient is determined by the expression

$$\alpha_G = \hbar\omega_G \, (S_{1D}N_1 + S_{1T}M_1 + S_{2D}N_2 + S_{2T}M_2), \tag{2}$$

where N_1 and N_2 or M_1 and M_2 are the number densities of the nonionized centers at which the electrons are in the states D_1 and D_2 or T_1 and T_2, respectively, and S_{1D} S_{2D}, S_{1T}, and S_{2T} are the photoionization cross sections corresponding to the above centers and states. For the light with a wavelength of 655 nm, whose quanta are capable of ionizing only the centers D_2 and T_2, the absorption coefficient is determined by the expression

$$\alpha_R = \hbar\omega_R \, (S_{2D}N_2 + S_{2T}M_2). \tag{3}$$

Using the system of the rate equations [7] corresponding to this model, we numerically analyzed the dynamics of the observed effects for the experiment in which the absorption variations are induced by a light beam with the wavelength $\lambda_g = 532$ nm and the absorption variations are tracked by the sample transmission of radiation with another wavelength $\lambda_r = 655$ nm. The numerical-simulation results are shown by the solid curve in Fig. 2 where the experimental data are also presented. The figure shows that our model developed in [7, 8] satisfactorily describes the dynamics of the development and relaxation of the photoinduced light absorption in the studied bismuth silicate crystal.

Figure 6 shows the numerical-simulation results for the dynamics of the number densities of the nonionized centers N_1 and M_1, as well as N_2 and M_2, subjected to the red and green laser irradiation.

If only the sounding red irradiation is switched on (regions *1*, *3*, *5*, and *7*), the electron number densities N_2 and M_2 in the states D_2 and T_2, respectively, decrease, while the electron number densities N_1 and M_1 at deeper centers in the states D_1 and T_1, respectively, increase. As a result, the crystal absorption for the illumination with a wavelength of 655 nm decreases. If irradiation with a wavelength of 532 nm (regions *2*, *4*, and *6*) is switched on, then the electron number density N_1 in the state D_1 decreases, while the electron filling of the "shallow" centers in the states D_2 and T_2 increases. The electron number density at the center T_1 varies nonmonotonically. First, for approximately 15–20 s (curve M_1; regions *2*, *4*, and *6*),

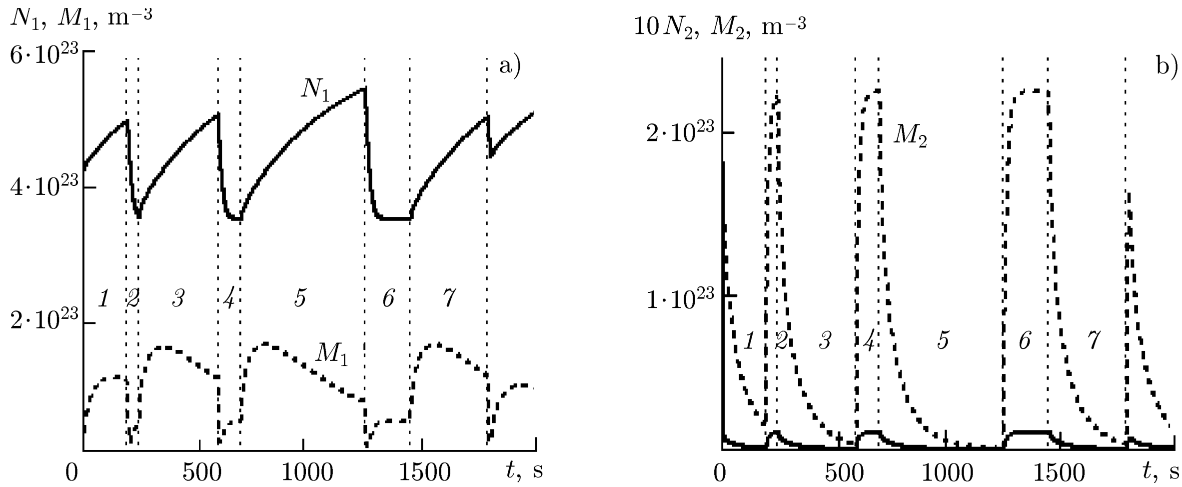


Fig. 6. Dynamics of the electron number density at the centers in the states D and T with small and large photoionization cross sections, respectively, for deeper centers 1 (*a*) and shallower centers 2 (*b*). The sounding beam with a wavelength of 655 nm, which has a flux density of 30 mW/cm^2, is assumed to be constantly switched on ($t = 0$–2000 s). The switch-on of the laser irradiation with a wavelength of 532 nm and a flux density of 200 mW/cm^2 in the crystal was simulated for the time intervals $t = 200$–250 s (region *2*), 600–700 s (region *4*), and 1 250–1450 s (region *6*).

irradiation with a wavelength of 532 nm leads to a decrease in M_1. Then the electron filling of the centers T_1 increases, which is related to the electron transitions from the state T_2 to the state T_1. This leads to an increase in optical absorption in the BSO crystal under irradiation with a wavelength of 532 nm, which is experimentally observed for the two used wavelengths 655 and 532 nm.

5. CONCLUSIONS

Thus, it has been established that during illumination of the undoped bismuth silicate crystal by laser radiation with wavelengths of 532 and 655 nm, the reversible changes in its optical absorption are observed. They can influence the photorefractive characteristics of this crystal, which are important for realization of the dynamic-holography devices. It has been found out that the red irradiation bleaches the crystal, whereas its irradiation by the shorter-wavelength (e.g., green) radiation increases optical absorption in both long- and short-wavelength visible-range intervals.

The presented theoretical model, which assumes the presence of two types of deep defect centers so that each center is characterized by two states with different photoionization cross sections, satisfactorily describes the experimental dependences of the absorption coefficient in the BSO crystal on the radiation affecting the crystal.

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