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Spectral dependence of absorption photoinduced in a $Bi_{12}TiO_{20}$ crystal by 532-nm laser pulses

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Abstract. **The spectral dependences of absorption photoinduced in a pure bismuth titanium oxide crystal by 532-nm laser pulses are studied. It is shown that optical absorption in the crystal in the range from 492 to 840 nm increases with increasing exposure. The photoinduced absorption relaxes in the dark for more than 60 hours. A model of photoinduced absorption is proposed which assumes the population of two trap centres with the normal energy distribution law for the concentrations of electrons photoexcited from donors to the conduction band. This model well describes the spectral dependences of photoinduced absorption by using the average ionisation energies of the traps $E_1 = 1.60$ eV and $E_2 = 2.57$ eV. The model is used to estimate the increase in the photorefractive sensitivity of a bismuth titanium oxide crystal in the near IR region, which was earlier observed after exposing the crystal to visible radiation. It is predicted that the speed of response of dynamic holography devices based on BTO crystals exposed to green light can be increased.**

Keywords: *photoinduced absorption, spectral dependence, bismuth titanium oxide*.

1. Introduction

The development of highly sensitive and stable measuring systems for reliable detection of extremely small physical quantities under real experimental conditions in the presence of uncontrollably varying external factors is an important direction in the problem of technical construction health monitoring, diagnostics of materials, studying biological objects, etc. Such measuring systems based on photorefractive media combine the interferometric principles of highly sensitive detection with adaptation to the noise influence. Among many photorefractive materials, $Bi_{12}SiO_{20}$ (BSO), $Bi_{12}GeO_{20}$ (BGO), and $Bi_{12}TiO_{20}$ (BTO) crystals with the sillenite structure attract great attention. Due to their good photoconduction properties, these crystals are promising for applications in optical memory devices, dynamic holography, integrated and nonlinear optics, and in adaptive information-measuring systems [1 – 7]. The physical properties of a crystal (the impurity type and concentration, the impurity energy level diagram, stoichiometric composition, etc.) determine the two main parameters of devices: their photorefractive sensitivity and response time. However, the material parameters of the crystal are specified during its growth and cannot be changed later.

The mastering of the method for controlling the sensitivity and the speed of response of a photorefractive device should considerably increase its functional possibilities and expand the scope of its applications. These parameters can be controlled by using the photochromic effect consisting in the reversible change of absorption in BSO, BGO, and BTO crystals exposed to visible or near UV light [1, 3, 4, 8 – 18]. The photochromic effect is explained by the charge exchange between defect centres with different photoionisation cross sections, one part of them being donors and another – traps for charge carriers [13, 17 – 19].

The redistribution of charges among donor and trap centres should result in the reversible change in the photorefractive parameters of a crystal. It was experimentally demonstrated earlier that preliminary exposing of BTO crystals to visible radiation resulted in a considerable increase in the efficiency of two-beam interaction in the near IR region (at 1064 nm [20] and 780 nm [21]) on photorefractive holograms formed in pure BTO crystals. The authors of papers [20, 21] concluded that BTO crystals contain deep donor centres from which electrons can be excited to the conduction band, a part of them being then captured by traps. The photon energy required to excite electrons from deep donors was estimated in [20] as ~ 2 eV, whereas according to [21], this energy lies in the range from 1.85 to 1.96 eV.

The increase in the photorefractive sensitivity at a wavelength of 1064 nm caused by the population of traps by electrons indicates a rather high concentration of centres with the energy levels located at distances ~ 1.17 eV from the conduction band bottom. The data on the energy distribution of the concentration of trap centres populated

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by electrons after preliminary exposure of BTO crystals to visible radiation are important for the development of dynamic holography devices operating in the near IR range.

Photoinduced absorption of light in sillenite crystals is usually studied by exposing them to continuous broadband radiation from lamps [1, 3, 8 – 12], monochromatic laser radiation [13 – 16] or quasi-monochromatic radiation from semiconductor light-emitting diodes [15 – 17]. The study of the dynamics of photoinduced absorption in $B_{12}TiO_{20}$: Ca crystals irradiated sequentially by quasi-monochromatic light in the red (660 nm) and green (525 nm) spectral regions revealed a considerable mutual influence of irradiation at one wavelength on the absorption of radiation at the other wavelength [18].

In this paper, we studied the spectral dependences of absorption photoinduced in a pure BTO crystal by laser pulses. We proposed a model of photoinduced absorption caused by the population of two trap centres by photoexcited electrons. This model well approximates experimental spectral dependences in the range from 492 to 840 nm. Changes in the photorefractive sensitivity and response time of the crystal in the near IR region caused by photoinduced absorption are estimated.

2. Experimental method and results

We studied photoinduced absorption in a 2.8-mm-thick pure bismuth titanium oxide crystal irradiated by 532-nm second-harmonic pulses from a Nd : YAG laser. The pulse duration was 30 – 50 ns, the pulse repetition rate was 10 Hz, and the average pulse energy was 0.5 mJ. The spatially homogenous part of the laser beam was selected with a 2.5-mm aperture to produce 100 mW cm^{-2} of average power density incident on the crystal. All experiments were performed at room temperature.

Laser irradiation produced a defect region in the crystal with different transmission in the visible and IR regions. The spectral dependences of the absorption coefficient $k_{exp}(\lambda, t_e)$ obtained for different exposure times t_e are presented in Fig. 1a. One can see that the absorption spectrum of the crystal in the range from 492 to 840 nm changes gradually upon laser irradiation. The effect is noticeable already after irradiation for one minute (exposure ~ 5 J cm^{-2}), while the growth of the absorption coefficient saturates after irradiation for 1 h (exposure ~ 300 J cm^{-2}).

Figure 1b shows the spectral dependences of the absorption coefficient $k_{rel}(\lambda, t_r)$ recorded at different instants t_r after the beginning of relaxation of photoinduced absorption, which lasted in the dark for 60 h. The relaxation of the absorption coefficient began from the values $k_{exp}(\lambda, t_e^f)$ achieved during the exposure time $t_e^f = 3600$ s.

The photoinduced absorption spectra can be obtained as the difference of the absorption coefficients $\Delta k_{exp}(\lambda, t_e) = k_{exp}(\lambda, t_e) - k_{exp}(\lambda, 0)$ and $\Delta k_{rel}(\lambda, t_r) = k_{rel}(\lambda, t_r) - k_{exp}(\lambda, 0)$ for the exposure and relaxation stages, respectively. It is convenient to analyse theoretically the dependence of photoinduced absorption on the photon energy $E = \hbar\omega$ expressed in electronvolts. The corresponding experimental dependences $\Delta k_{exp}(E, t_e)$ and $\Delta k_{rel}(E, t_r)$ are presented in Fig. 2. One can see that photoinduced absorption increases with the photon energy and achieves the maximum value (~ 5.5 cm^{-1}) for $E = 2.52$ eV ($\lambda = 492$ nm).

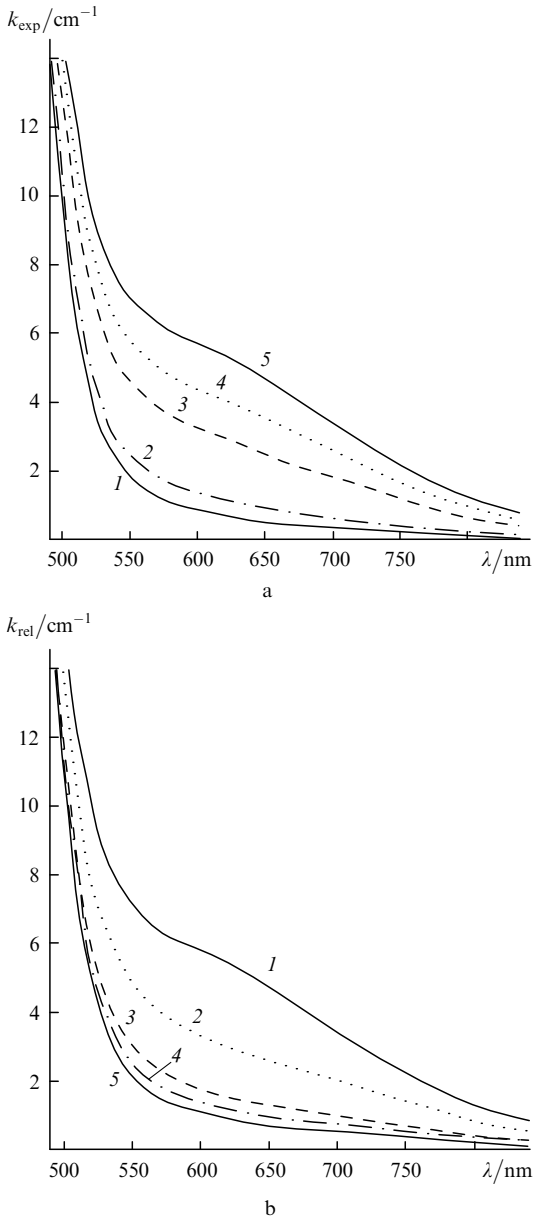

Figure 1. Changes in the absorption spectra of the BTO crystal irradiated by laser pulses after exposure times 0 (*1*), 60 (*2*), 600 (*3*), 1800 (*4*) (a), and 3600 s (*5*) and during relaxation at different instants of time after irradiation: 0 (*1*), 15 (*2*), 34 (*3*), 40 (*4*), and 60 h (*5*) (b).

3. Theoretical model of photoinduced impurity absorption

The quantitative models of the photoinduced absorption of light in sillenite crystals are based on the hypothesis of the charge exchange between defect centres with different photoionisation cross sections [13, 17 – 19]. It is assumed that electrons excited by light into the conduction band from deep donor centres populate traps, which results in a change in optical absorption. It is assumed in [13, 17, 19] that traps are shallow and their depletion due to thermal excitation of electrons to the conduction band provides the relaxation of induced variations in absorption in the dark. However, the authors of [18] have shown that the localisation depth of the trap energy levels in the energy gap of a $Bi_{12}TiO_{20}$: Ca crystal exceeds 1.43 eV from the conduction-

band bottom. The corresponding value for a pure BTO crystal can be estimated from the data presented in [20] as 1.17 eV.

The absorption spectrum in the case of excitation of electrons by photons of energy $\hbar\omega$ from a deep defect centre having a local discrete energy level in the energy gap and the ionisation energy E_i can be written in the form [22]

$$k_\delta(\hbar\omega, E_i) = B_{ei}\frac{(\hbar\omega/E_i - 1)^{1/2}}{(\hbar\omega/E_i)^3}, \quad (1)$$

where the coefficient $B_{ei} = SN_{ei}$ is proportional to the concentration N_{ei} of these centres and their photoionisation cross section S. As follows from Fig. 2, the spectral dependence of induced absorption in the BTO crystal is not consistent with Eqn (1). This can be explained, first, by the presence of traps of several types with different values of the ionisation energy and cross section. Second, a great imperfection of bismuth titanium oxide crystals [3, 12] should lead to considerable random fluctuations in the concentration of defects, resulting in fluctuations of their ionisation potential. In this case, the local levels of defects are overlapped, and the energy distribution of their concentration in the energy gap can be represented by a smooth function $N(E_i)$. This allows one to introduce the coefficient $B(E_i)dE_i = SN(E_i)dE_i$ determining the absorption of light per interval dE_i of the ionisation energy. The total absorption coefficient for radiation with frequency ω can be written in the integral form

$$k(\hbar\omega) = \int_0^{\hbar\omega} SN(E_i)\frac{(\hbar\omega/E_i - 1)^{1/2}}{(\hbar\omega/E_i)^3} \times \frac{1}{1 + \exp[(E_F - E_i)/k_B T]}dE_i, \quad (2)$$

where the energy E_F is the distance from the Fermi level to the conduction-band bottom; k_B is the Boltzmann constant; and T is the absolute temperature.

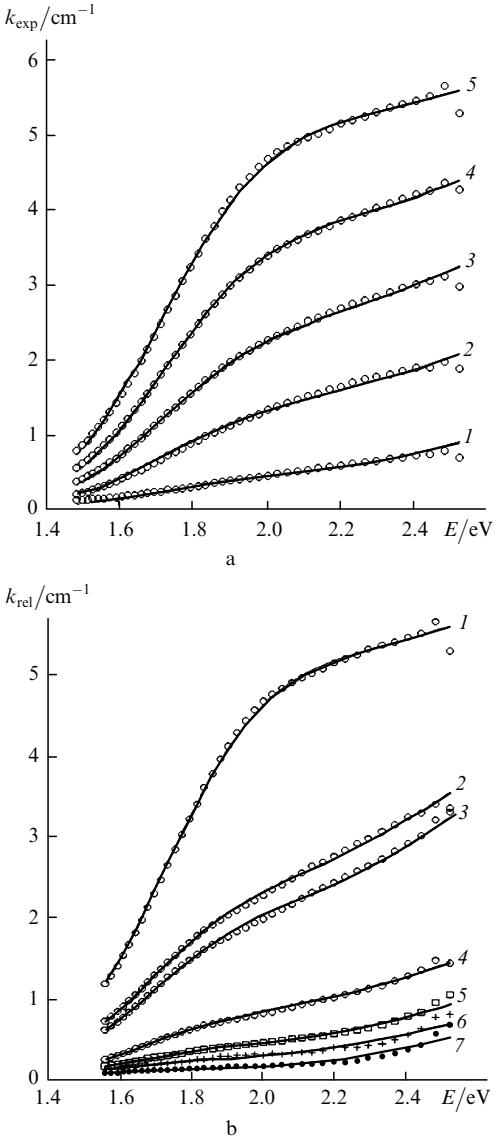

Figure 2. Spectral dependences of the induced absorption in the BTO crystal irradiated by laser pulses for exposure times 60 (*1*), 300 (*2*), 600 (*3*), 1800 (*4*), and 3600 s (*5*) (a) and during relaxation at different instants of time after irradiation: 0 (*1*), 15 (*2*), 17 (3), 34 (*4*), 40 (*5*), 43 (*6*), and 60 h (7) (b). Points are the experiment, solid curves are the theory.

4. Approximation of the spectral dependences of photoinduced absorption

We will assume that the energy distribution $N_n(E_i)$ of the concentration of trap centres of the n type is described by the normal law. We also assume that the parameters of each distribution remain invariable upon the occupation and depletion of traps, so that the dependence of B on the ionisation energy and time can be written in the form

$$B(E_i, t) = \sum_n b_n(t)\exp\left[-\frac{(E_n - E_i)^2}{\Delta E_n^2}\right], \quad (3)$$

where the function $b_n(t)$ takes into account the time dependence of the filling of the n centre by electrons.

The experimental dependences of photoinduced absorption $\Delta k_{exp}(E, t_e)$ and $\Delta k_{rel}(E, t_r)$ are fitted by solid curves in Fig. 2, which are constructed based on expressions (2) and (3) taking into account two centres with the average ionisation energies $E_1 = 1.597 \pm 0.092$ eV and $E_2 = 2.566 \pm 0.028$ eV. The corresponding distribution half-widths and the Fermi level position are $\Delta E_1 = 0.277 \pm 0.018$ eV, $\Delta E_2 = 0.580 \pm 0.031$ eV and $E_F = 1.027$ eV.

5. Discussion of the results

The dependences $b_1^{exp}(t_e)$ and $b_2^{exp}(t_e)$ found upon approximation by expression (3) (Fig. 3a) show that the electron occupation of the second centre with the energy levels located near the top of the valence band begins to saturate at exposure times $t_e \sim 900$ s, whereas for the first centre with the higher energy no saturation is observed even for $t_e = 3600$ s. During the dark relaxation described by the function $b_1^{rel}(t_r)$ (Fig. 3b), the contribution of the first centre to photoinduced absorption decreases nearly exponentially with the time constant ~ 18 h. We can assume that the

occupation of such traps with the average ionisation energy $E_1 \sim 1.6$ eV occurs only through the conduction band, while their depletion occurs by tunnelling followed by the recombination of electrons on ionised donors and other lower-energy trap centres. Such tunnelling can occur, as confirmed in [23] for lithium niobate containing iron at high concentrations, due to a high concentration of structural defects in BTO [3, 12]. The capture of a part of tunnelling electrons by centres with the average energy $E_2 \sim 2.57$ eV can explain a nonmonotonic time dependence observed upon relaxation of photoinduced absorption caused by these traps [the dependence $b_2^{\mathrm{rel}}(t_\mathrm{r})$ in Fig. 3b].

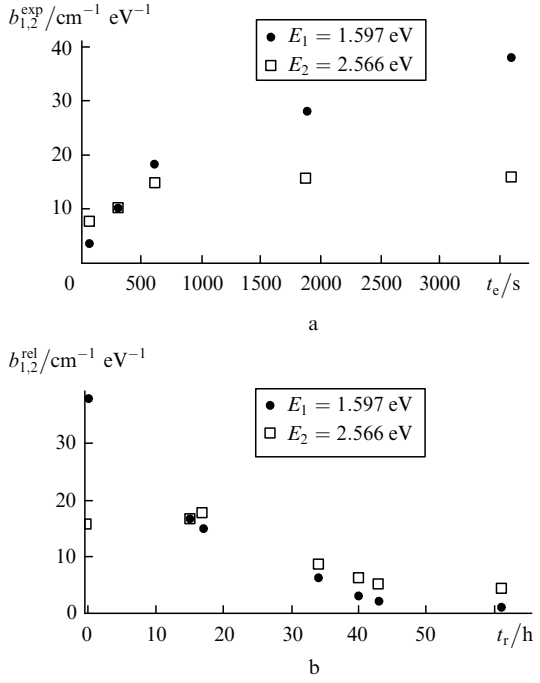

Figure 3. Time dependences of functions $b_1(t)$ and $b_2(t)$ characterising the population of traps with the ionisation energies $E_1 = 1.597$ eV and $E_2 = 2.566$ eV by electrons upon pulsed laser irradiation of the crystal (a) and during relaxation (b).

It can be assumed that electrons captured by these traps may be involved in the processes of photoexcitation and redistribution of the volume charge resulting in the formation of photorefractive holograms upon illuminating a crystal by the interference pattern produced by the reference and signal light waves. In this case, the increase in the sensitivity of the BTO crystal to the recording of photorefractive holograms in the near IR range, which was observed after preliminary exposing the crystal to visible radiation [20, 21], is mainly determined by the population of the trap with the average energy $E_1 = 1.60$ eV by electrons. Using the known definition [2], we express the holographic sensitivity as the ratio of the growth rate of the amplitude of modulation Δn of the crystal refractive index by the photorefractive grating in the initial region of its formation to the intensity modulation amplitude mI_0:

$$S_\mathrm{ph} = \frac{1}{mI_0}\frac{\mathrm{d}|\Delta n|}{\mathrm{d}t}, \tag{4}$$

where m is the contrast and I_0 is the average light intensity in the interference pattern. In the case of the diffusion mechanism of formation of photorefractive gratings, which was used in papers [20, 21], we obtain from relations presented in [2] the holographic sensitivity in the form

$$S_\mathrm{ph}(\omega) = \frac{n^3 r_\mathrm{eff} E_\mathrm{d}}{2(1 + K^2 L_\mathrm{d}^2)}\frac{e\mu\tau_\mathrm{rec}}{\varepsilon}\frac{\phi(\omega)k(\omega)}{\hbar\omega}, \tag{5}$$

where n is the refractive index of the crystal; r_eff is the effective electrooptical constant; $E_\mathrm{d} = K(k_\mathrm{B}T/e)$ is the diffusion field; $L_\mathrm{d} = (\mu\tau_\mathrm{rec}k_\mathrm{B}T/e)^{1/2}$ is the diffusion length; μ and τ_rec are the mobility and recombination time of charge carriers; $K = 2\pi/\Lambda$; Λ is the spatial period of the grating; ε is the static permittivity of the crystal; e is the elementary electric charge; and ϕ is the quantum yield.

The spectral dependence of the photorefractive sensitivity, which is described by the last factor in expression (5), changes after irradiation of the crystal due to a change in its optical absorption $k(\omega)$. The absorption coefficient of the BTO crystal measured at 780 nm before crystal irradiation was 0.14 cm^{-1}, while after irradiation for 3600 s it became equal to 1.56 cm^{-1}. The initial absorption at 840 nm was 0.02 cm^{-1}, while after irradiation it was 0.76 cm^{-1}.

Before irradiation of the crystal the absorption coefficient in the spectral region above 840 nm decreases with increasing wavelength; however, its values calculated from the transmission data are within the experimental error. Variations in the spectral dependence of the photorefractive sensitivity of the BTO crystal, which are caused by the increase in absorption during irradiation, in particular, in the spectral range above 840 nm can be estimated from the theoretical model presented above. Figure 4 shows the spectral dependences of induced absorption in the region from 700 to 1100 nm calculated by using expressions (2) and (3) and the found average values of the model parameters and coefficients $b_{1,2}^{\mathrm{rel}}(t_\mathrm{r})$ for relaxation times $t_\mathrm{r} = 17$ and 34 h.

One can see that irradiation of the BTO crystal by 532-nm laser pulses can considerably enhance its photorefractive sensitivity to IR radiation at wavelengths up to 1100 nm, i.e. can noticeably expand the spectral range of this crystal as a dynamic medium, thereby expanding applications of holographic devices based on photorefractive BTO crystals.

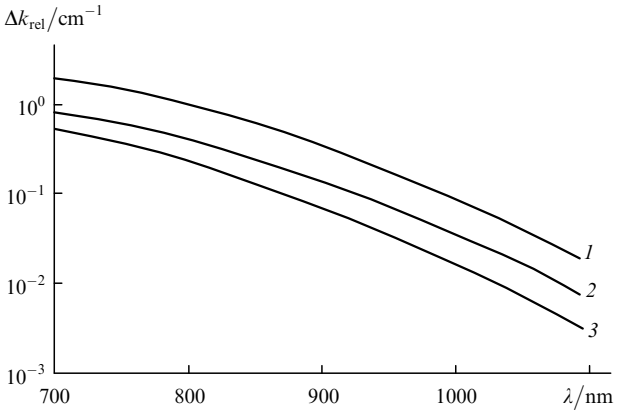

Figure 4. Calculated spectral dependences of additional absorption in the BTO crystal in the spectral region from 700 to 1100 nm induced by laser pulses (*1*) and after relaxation for 17 (*2*) and 34 h (*3*).

Another important parameter of photosensitive media for dynamic holography devices is their response time [2, 24, 25]. The minimal possible formation time of dynamic holograms in photorefractive crystals is equal to the Maxwell relaxation time τ_M [2]. The intensities of the reference (I_R) and signal (I_S) beams commonly used for BTO crystals allow one to neglect the dark conductivity of the crystal compared to its photoconductivity σ_{ph}. In this case, the time τ_M is inversely proportional both to the average intensity $I_0 = I_R + I_S$ of the beams producing the hologram and to the absorption coefficient $k(\omega)$:

$$\tau_M(\omega) = \frac{\varepsilon\hbar\omega}{e\mu\tau_{rec}\phi(\omega)k(\omega)I_0}. \quad (6)$$

Thus, the preliminary illumination of the crystal, resulting in the increase in its absorption coefficient, can considerably reduce its photorefractive response time. It follows from the experimental data presented in Fig. 1 that, when the operating wavelength $\lambda = 633$ nm is used (which is typical for BTO devices), the speed of response should increase by a factor of 8.8 after irradiation of the crystal by 532-nm laser pulses up to saturation.

The absorption coefficient of BTO crystals in the red region also considerably increases after their irradiation by 525-nm semiconductor light-emitting diodes [18]. In accordance with the model of photoinduced impurity absorption presented above, this is caused by the population of traps with average ionisation energies $E_1 = 1.60$ eV and $E_2 = 2.57$ eV by photoexcited electrons and should increase both the speed of response and the efficiency of dynamic holography devices based on photosensitive BTO crystals. Note here that irradiation of crystals in such devices by radiation from semiconductor light-emitting diodes (for example, in adaptive correlation filters [24, 26] and adaptive holographic interferometers [27]) can be readily performed.

6. Conclusions

We have shown that irradiation of the bismuth titanium oxide crystal by 532-nm laser pulses leads to a change in its absorption coefficient, which increases with the exposure time. The photoinduced absorption relaxes in the dark for more than 60 h. The spectral dependence of the induced absorption is well described by the model of capturing photoexcited electrons by traps of two types with the normal energy distribution of their concentration and average ionisation energies $E_1 \sim 1.6$ eV and $E_2 \sim 2.57$ eV. The population of the trap with the average energy $E_1 \sim 1.6$ eV by electrons enhances the photorefractive sensitivity of the irradiated crystal in the near IR range, which can be estimated from the induced absorption. In dynamic holography devices using laser radiation in the red spectral region and photorefractive BTO crystals, the speed of response can be increased by several times after irradiation of the crystal by green light due to the capture of electrons by traps of both types. In addition, such irradiation expands the spectral sensitivity region to the near IR range.

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