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Effect of Sequential Exposition to Short- and Long-Wavelength Radiation on the Optical Absorption in the Bismuth Titanium Oxide Crystal Doped by Aluminum

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Abstract

Changes in the spectral dependences of the optical absorption induced in the bismuth titanium oxide crystal doped by aluminum as a result of sequential exposition to cw laser radiation first with the wavelength \( \lambda_1 = 532 \text{ nm} \) and then with the longer wavelength \( \lambda_2 = 633, 655, 663, 780, 871, \) or 1064 nm are investigated. Our experiments show that after the short-wavelength exposition to radiation with \( \lambda_1 = 532 \text{ nm} \), the optical absorption in the crystal increases, and in the range 470–1000 nm, yields the spectrum whose form is independent of the initial crystal state. The subsequent exposition to longer-wavelength radiation leads to enhanced transmittance of the crystal in the examined spectral range. A maximum decrease of the optical absorption in the crystal is observed upon exposure to radiation with the wavelength \( \lambda_3 = 663 \text{ nm} \).

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1. Introduction

Sillenite \( \text{Bi}_4\text{SiO}_{12} \) (BSO), \( \text{Bi}_4\text{GeO}_{12} \) (BGO), and \( \text{Bi}_4\text{TiO}_{12} \) (BTO) crystals possess high sensitivity to optical radiation manifested through the photoconductivity and photorefractive and photochromic effects [Petrov, Stepanov,
Khomenko (1991), Malinovskii, et al. (1990), Kargin et al. (2004)]. Due to high refractive index, fast photorefractive response, and good electrooptical, photochromic, and photoconductive properties, these crystals are often used as photosensitive media in dynamic holography [Petrov, Stepanov, Khomenko (1991), Malinovskii, et al. (1990), Kargin et al. (2004)].

According to [Egorysheva (2005)], the edge of the fundamental absorption band for the BSO, BGO and BTO crystals at 300 K is located at $\lambda_f = 385$, 385, and 403 nm, respectively. Before the main edge, the intensive band at $\lambda = 380–500$ nm, called shoulder in the literature, is located, where the absorption coefficient at a wavelength of 420 nm is equal to 20, 40, and 100 cm$^{-1}$ for BSO, BGO, and BTO, respectively [Malinovskii, et al. (1990), Kargin et al. (2004)]. It is considered that this intensive impurity absorption band is caused by the presence of intrinsic structural defects [Malinovskii, et al. (1990), Kargin et al. (2004)]. In the sillenite crystals exposed to light of visible and near-UV ranges, the photochromic effect is observed due to electron photoexcitation from defects representing deep donor centers to the conduction band and subsequent recombination on traps whose photoionization cross section is larger than that of donors [Shandarov et al. (2007)]. In this case, the reversible change of the photorefractive crystal parameters can also be observed. For example, in [Odoulov, Shcherbin, Shumeljuk (1994)] it was shown experimentally that preliminary illumination of undoped bismuth titanium oxide crystals by visible radiation increased significantly the efficiency of two-wavelength interaction on photorefractive holograms formed in them by light beams of the near-IR range.

At the same time, as indicated in [Malinovskii, et al. (1990), Kargin et al. (2004)], the impurities affect considerably the photorefractive parameters of the sillenite crystals. This can be due to the fact that crystal doping can cause not only formation of new deep levels, but also change of the parameters of structural defects characteristic for undoped samples [Kargin et al. (2004)]. Thus, a high value of the optical absorption for the bismuth titanium oxide (Bi$_2$TiO$_4$:Ca) crystals doped by calcium in the blue-green range of the spectrum was attributed to the presence of the shoulder in the absorption spectrum supposed to be caused by the structural defects in the crystal lattice [Malinovskii, et al. (1990), Kargin et al. (2004)]. The significant absorption in the Bi$_2$TiO$_4$:Ca crystal limits its application as a photorefractive material in devices using laser radiation with wavelengths of 532, 514.5, and 488 nm. The absence of the shoulder or its insignificant intensity is observed for the BTO crystals doped by Al, Ga, Zn, P, Cd, and V elements [Kargin et al. (2004)].

In this regard, it is of interest to investigate changes in the optical absorption spectra for both undoped and doped crystals under external irradiation that will allow the energy parameters of the corresponding structural defects to be determined and the existing model of the photorefractive effect in the sillenite crystals to be refined.

The present work is devoted to experimental investigations of the spectral dependences of the optical absorption in the Bi$_2$TiO$_4$:Al crystal and their changes after sequential exposition to cw laser radiation to the shortest wavelength $\lambda_i = 532$ nm in the first stage followed by exposition to the longer wavelength $\lambda_n$ taking in different experiments value of 633, 655, 663, 780, 871, or 1064 nm, respectively.

The experimental spectral dependences of the optical absorption in the Bi$_2$TiO$_4$:Al crystal after sequential exposition to laser radiation first with the wavelength $\lambda_i = 532$ nm and then with the wavelength $\lambda_n = 633, 663, 871, \text{ or } 1064$ nm, respectively, are shown in Fig. 1.
It was obtained that though the optical absorption spectrum \( k(\lambda) \) of the examined Bi\(_{12}\)TiO\(_{20}\):Al sample in the initial state changed from experiment to experiment, illumination of the input face of the crystal with light having the wavelength \( \lambda_i = 532 \) nm and the intensity \( I_i = 15.4 \) mW/cm\(^2\) for 1930 s in the first stage each time led to its invariable form (curve 1 in Fig. 1) with maximum values of \( k(\lambda) \). The crystal illumination in the subsequent stages lasted until saturation of the optical absorption spectra \( k(\lambda) \). Radiation with \( \lambda_i = 633 \) nm \( (I_i = 6 \) mW/cm\(^2\), 11 stages with total illumination time \( t_n = 3060 \) s; curve 3 in Fig. 1); 655 nm \( (I_i = 10 \) mW/cm\(^2\), 11 stages, \( t_n = 3060 \) s; not shown); 658 nm \( (I_i = 20 \) mW/cm\(^2\), 6 stages, \( t_n = 1620 \) s; not shown); 663 nm \( (I_i = 270 \) mW/cm\(^2\), 4 stages, \( t_n = 960 \) s; curve 2); 780 nm \( (I_i = 4 \) mW/cm\(^2\), 22 stages, \( t_n = 6420 \) s; not shown); 871 nm \( (I_i = 6 \) mW/cm\(^2\), 14 stages, \( t_n = 3960 \) s; curve 4), or 1064 nm \( (I_i = 64 \) mW/cm\(^2\), 7 stages, \( t_n = 1860 \) s; curve 5) was used. As can be seen from Fig. 1, illumination of the Bi\(_{12}\)TiO\(_{20}\):Al crystal by longer-wavelength radiation after its irradiation by light with \( \lambda_i = 532 \) nm leads to its enhanced transmittance in the entire examined wavelength range from 470 to 1000 nm.

Fig. 1. Experimental spectral dependences of the optical absorption coefficient for the Bi\(_{12}\)TiO\(_{20}\):Al crystal exposed first to laser radiation with a wavelength of 532 nm (1) and then to radiation with a wavelength of 663 (2), 633 (3), 871 (4), or 1064 nm (5), respectively, until saturation.

Fig. 2. Experimental spectral dependences of the enhanced transmittance of the Bi\(_{12}\)TiO\(_{20}\):Al crystal preliminary illuminated with laser radiation at a wavelength of 532 nm observed after subsequent illumination with radiation at a wavelength of 663 (1), 655 (2), 633 (3), 780(4), 871 (5), or 1064 nm (6) until saturation.
Figure 2 shows the spectral dependences characterising the maximum changes in the optical absorption $\Delta \kappa(\lambda)$ for the Bi12TiO20:Al crystal with enhanced transmittance, preliminary illuminated by green light ($\lambda_i = 532$ nm), reached by its subsequent illumination with longer-wavelength radiation at $\lambda n = 663$ (curve 1), 655 (curve 2), 633 (curve 3), 780 (curve 4), 871 (curve 5), or 1064 nm (curve 6).

It should be noted that the maximum decrease of the optical absorption in the Bi 12TiO20:Al crystal was observed after crystal illumination with radiation at the wavelength $\lambda_0 = 663$ nm.

3. Discussions

An analysis of the dependences shown in Figs. 1 and 2 and their comparison with the results presented in [9, 10] demonstrates that they can be described by the model considering the contribution to the impurity absorption of the electrons photoexcited from the deep donor centers, whose concentrations obey a normal distribution over the ionization energy, to the conduction band and of the intracenter transitions [8]. However, unlike the data presented in [9, 10], the contribution of the intracenter transitions which must be resonant in character and the spectral characteristics of the optical absorption independent of the conditions of sample illumination can be described only by three Gaussian curves with maxima at $\lambda_{m1} = 827$ nm (quantum energy $\hbar \omega_{m1} = 1.5$ eV), $\lambda_{m2} = 761$ nm ($\hbar \omega_{m2} = 1.63$ eV), and $\lambda_{m3} = 704$ nm ($\hbar \omega_{m3} = 1.76$ eV). In [Akrestina et al. (2012), Shandarov et al. (2013)], to describe the resonant character of the dependences $\Delta \kappa(\lambda)$ for the enhanced transmittance of the Bi12TiO20:Al crystal illuminated by radiation at the wavelengths $\lambda_0 = 660$ and 1064 nm, two other intracenter transitions with maxima for energy quanta of 2.13 and 2.44 eV (582 and 508 nm) were also taken into account. From our analysis of the enhanced transmittance of the sample illuminated with six radiation wavelengths it follows that consideration of the given transitions was caused by the insufficiency of the experimental data in [9, 10]. The maxima observed for dependences 1, 2, 3, and 6 of $\Delta \kappa(\lambda)$ at $\lambda \approx 577$ nm and for dependences 4 and 5 at $\lambda \approx 600$ nm (Fig. 2) can be described by changes of the contribution of the electron photoexcitation processes to the conduction band associated with their photoinduced redistribution over the deep donor centers.

4. Conclusions

Thus, illumination of the Bi12TiO20:Al crystal with cw laser radiation at the wavelength $\lambda_i = 532$ nm yields maximum values of the optical absorption in the range 470–1000 nm and the spectral dependence $k(\lambda)$ independent of the initial crystal state. Subsequent illumination of the examined sample with longer-wavelength laser radiation at the wavelength $\lambda_0 = 633, 655, 663, 780, 871,$ or 1064 nm leads to its enhanced transmittance that reaches saturation depending on the employed wavelength. The minimum values of $k(\lambda)$ were observed after illumination of the Bi12TiO20:Al crystal by light at a wavelength of 663 nm.

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