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Light-induced effects in sillenite crystals with shallow and deep traps

Tatiana Kornienko[a]*, Marina Kisteneva[b], Stanislav Shandarov[b], Alexei Tolstik[a]

[a]*Belarussian State University, 4 Nezavisimosti av., Minsk 220030, Belarus*
[b]*Tomsk StateUniversity of Control Systems and Radioelectronics, 40 Lenina av., Tomsk 634050, Russia*

Abstract

This paper presents the light-induced effects in bismuth silicon and bismuth titanium oxide crystals associated both with the electron transitions into the conduction band and with the filling of shallow and deep traps, which determine the optical and electroconductive properties of these crystals. The dynamics of photoconductivity and light-induced absorption is analyzed under conditions of pulsed laser illumination at the wavelength of 532 nm. The possibility to describe the relaxation processes of a population for trapping levels with the use of two-exponential function is demonstrated. The photoconductivity dynamics is characterized by two relaxation times on the order of 100 ns and 10 μs, whereas for light-induced absorption the lifetimes about 10 μs and several days for short- and long-lived traps, respectively, have been obtained. Because of this, the relaxation transitions may be occurred both to the shallow trap centers with energy located close to the conduction band and to the deep-lying traps, which should be included into a diversified theoretical model adequately describing the light-induced phenomena in photorefractive sillenite-family crystals.

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Keywords: photorefractive crystals; bismuth silicate; bismuth titanate; light-induced effects; trapping levels

* Corresponding author. Tel.: +375-29-590-5751; fax: +375-17-209-5445.
*E-mail address:*tankorni@mail.ru

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1. Introduction

Nomenclature

BTO	a single crystal of bismuth titanium oxide $Bi_{12}TiO_{20}$
BSO	a single crystal of bismuth silicon oxide $Bi_{12}SiO_{20}$

Photorefractive crystals are widely used in various areas of science and technology including the use of such nonlinear media at low (micro- or nanowatt) powers of laser radiation. Among the technological applications of these media may be listed optical image amplification; recording, storage, and information processing with the use of optical methods; phase conjugation; adaptive holography; development of the elements for optical associative memory, microscopy and neurocomputing, etc. (Günter and Huignard (2006), Yeh et al. (1989), Brito et al. (2013), Shandarov et al. (2008)).

Owing to inhomogeneous illumination of photorefractive crystals the charge redistribution between defect centres takes place. A formed space-charge field induces the refractive index perturbations of the crystal due to the linear electro-optic effect. Because of this, one permits to realize the nonlinear effects such as the formation of dynamic diffraction and waveguide structures, including the spatial photorefractive solitons (DelRe et al. (2013) and Gabruseva et al. (2006)).

To describe the physical processes at charge redistribution in photorefractive crystals a great deal of the theoretical models was proposed (see Günter and Huignard (2006), Yu et al. (1999), Tolstik et al. (2007)). Each specific model characterizes one or other types of experimental phenomena. To illustrate, Tolstik et al. (2012) proposed to take into consideration two-photon excitation under exposition of a bismuth titanium oxide crystal by strong picosecond laser pulses. One of the most widely used one-centers model was developed by Kukhtarev (Kukhtarev et al. (1979)). It is based on the band transfer of charge carriers after theirs photoexcitation from deep-lying neutral donor centers accompanied by the recombination of one on the ionized donors, which are considered as the deep-lying traps. The disadvantage of one-center model is an impossibility of explanation for a diversity of phenomena revealed in photorefractive crystals. Among these are the effects of photo-induced changes in the optical absorption and the decay of photorefractive gratings in the dark with temporal behavior different from exponential function, which are associated with existence of a shallow-trap defect centers in the crystal (Brost et al.(1988), Tayebati and Mahgerefteh (1991)).

Temporal relaxation in the optical absorption with the availability of several recombination channels can be described, in particular, by the use of rate equations for the concentration of charge carriers occupied the deep and shallow traps with different lifetimes. This approach was used by Matusevich et al. (2009) in modeling the relaxation for additional optical absorption induced preliminary by strong CV radiation with a wavelength of 514 nm in the bismuth titanium oxide crystal, which was observed experimentally with the probing low-intensity laser beam (532 nm). A simpler approach using the decomposition of the observed time dependences into several exponential functions with different relaxation times was used in the description for the dynamics of photoinduced changes in the optical absorption in the bismuth titanium oxide crystal after its exposure by strong laser pulses (532 nm) by Matusevich et al. (2008) and in the bismuth silicon oxide crystal during CV illumination with wavelength of 1064 nm (Kisteneva et al.(2009)).

Photorefractive crystals of the sillenite group $Bi_{12}SiO_{20}$ (BSO) and $Bi_{12}TiO_{20}$ (BTO) show photoconductive properties as well as a long-lived changes in light absorption induced upon exposure to visible and near-infrared radiation (Kargin et al. (2004), Kisteneva et al.(2009)). Dynamics of changes in light absorption only, which was induced by picosecond laser pulses (532 nm) in BTO and BSO crystals has been studied byKisteneva et al. (2010). Furthermore, the picosecond excitation has been used for determine the band mobility of photoexcited electrons in BSO crystal (Biaggo et al.(1997)).

This paper presents the results of experimental studies of photoconductivity and photoinduced absorption kinetics caused in undoped single crystals BSO and BTO by nanosecond laser pulses with a wavelength of 532 nm,

and their interpretation in terms of a model involving the photoexcitation of electrons from deep neutral donors to the conduction band accompanied by population of shallow and deep trap centers. It is shown that the observed time dependences satisfactorily approximated by the sum of two exponential functions, characterizing the fast and slow components of the relaxation processes.

2. Object and method of studies.

In this paper theobjectofstudiesaretheundopedphotorefractive$Bi_{12}TiO_{20}$ and $Bi_{12}SiO_{20}$ singlecrystals. The distinctive features of light interaction and self-interaction phenomena as well as of another photorefractive effect in mentioned medium have been studied during more thirty years. All crystals of sillenite family ($Bi_{12}SiO_{20}$, $Bi_{12}TiO_{20}$, $Bi_{12}GeO_{20}$) are characterized by low Pockels constant as compared to such photorefractive crystals (PRC) as barium titanate or barium-strontium niobate. Certain of their significant advantages are the possibility of operation over a wide temperature range and ease of technological processing. The photorefractive response in sillenites can be improved by external electric fields which could be applied to the samples.

The nominally undoped BSO sample had measuring 4 mm × 2.5 mm× 20 mm along the $[1\,\bar{1}0]$, the [110], and [001] crystallographic directions, respectively. The light radiation was propagated along the $[1\,\bar{1}0]$ axis whereas external electric field was applied along the [110] one with the help of glued silver electrodes.

The light radiation in the nominally undoped BTO sample with thickness d = 3 mm was propagated along the [100] axis. To provide the experimental study for dynamics of photoconductivity an external electric field was applied with the use of silver electrodes glued to the opposite <010> faces of the sample with transverse dimensions of 6.0 mm × 11.2 mm along the [010] and the [001] directions, respectively.

The magnitude of external voltage applied for samples in the experiments relating to photoconductivity ranges up to15 V. For the investigation of temporal relaxation of optical absorption of the BTO sample in darkness we used its preliminary exposition by laser pulses (532 nm) with duration of 15-20 ns, average energy of 1000mJ/cm^2, and repetition rate of 10 Hz during about 300 s. The spectral dependences of optical density of the BTO sample in the range from 400 to 1000 nm were registered with an spectrophotometer SZS-16.

3. Results and discussions

3.1. Temporal behavior of spectral dependences for the changes in light-induced absorption of the BTO crystal after preliminary exposition by nanosecond laser pulses with a wavelength of 532 nm

The spectral dependences for optical density of BTO crystal preliminary exposed by nanosecond pulses (532 nm), which were registered during relaxation to the initial state at different point of time (from 1 to 96 hours after irradiation) are presented in Fig. 1. It can be seen that relaxation of light-induced absorption characterized by very slow component in the scale of time under consideration. The quantitative models of the light-induced absorption in photorefractive crystals are based on the hypothesis of the charge exchange between defect centers with different photoionization cross section (Brost et al. (1988), Temple and Warde(1988), Tayebati and Mahgerefteh(1991), Tolstik et al.(2007), Matusevich et al.(2009), Tolstik and Haider(2012)).

In (Tolstik et al.(2007)) was shown that slow relaxation of light-induced absorption in preliminary exposed BTO crystal is concerned with depletion of electron occupation for two deep trap centers with average ionization energies of 1.6 and 2.57 eV. It should be noted that illumination of the BTO sample by nanosecond laser pulses (532 nm) changes the light absorption over wide spectral range (see Fig. 1). Because of that the fast components for growth and relaxation of changes in light absorption concerned with filling and depletion of shallow traps by electrons, which should be induced by nanosecond pulses with a wavelength of 532 nm, can be registered by the use of probing CV laser beam with other wavelength.

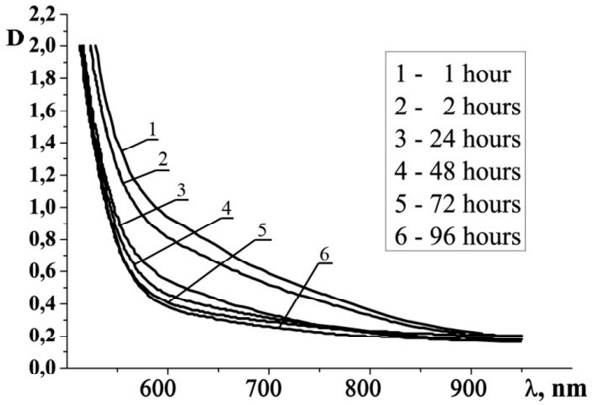

Fig. 1. Optical density spectra D at different instants of time after laser illumination for the BTO sample.

3.2. Dynamics of photoconductivity and light-induced absorption in photorefractive BSO and BTO crystals for exposure by nanosecond laser pulses

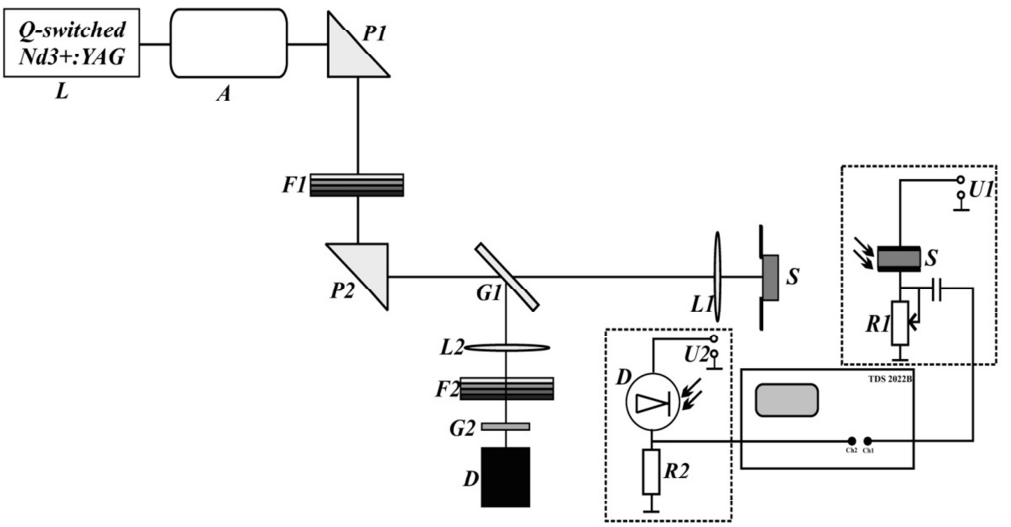

Fig. 2. Schematic of an experimental setup used to study the spectral photoconductivity of photorefractive crystals.

At one of the our work stages we have studied the photoconductivity and photoinduced absorption of the undoped photorefractive single crystals of bismuth silicon and bismuth titanium oxide crystals subjected to exposure to nanosecond laser pulses.

So to investigate photoconductivity a pulse solid-state yttrium aluminum garnet laser operating in the second harmonic generation mode (wavelength of generation is 532 nm) as the radiation source and a parametric generator provided homogeneous illumination of the sample at the wavelengths from 440 to 700 nm and with power of radiation from 5 kW/cm^2 to 300 kW/cm^2 was used (Fig. 2). The exposing radiation intensity and polarization could

be varied by system of light filters F1 and F2 and polarizers. The photorefractive sample was incorporated into the electric circuit.

One of the difficulties in described scheme is the high-resistance of photorefractive sample. To record a photoconductivity signal load resistor was connected in series with this sample. The signal from load resistor was recorded by a TDS 2022V two-channel digital oscilloscope. The electric circuit was designed so that all its elements were in the closest proximity to the refractive sample to exclude the transitive electric processes. But another difficulty is limit of load resistance value: the transitive-process time constant should be smaller than times characterizing the relaxation transitions: «conduction band – traps» and «traps – traps». As a result, different sets of the crystal responses were obtained which constitute dependences of load resistor voltage under exposing radiation with various wavelengths and different intensities. Figure 3 presents the curves for such kinetics. Taking into consideration the electric circuit design, the initially measured kinetics could be recalculated as a function of the crystal voltage U_{cr} or current I_{cr} of time t, and also as a function of the crystal resistance $R_{c\,r}$ of time t.

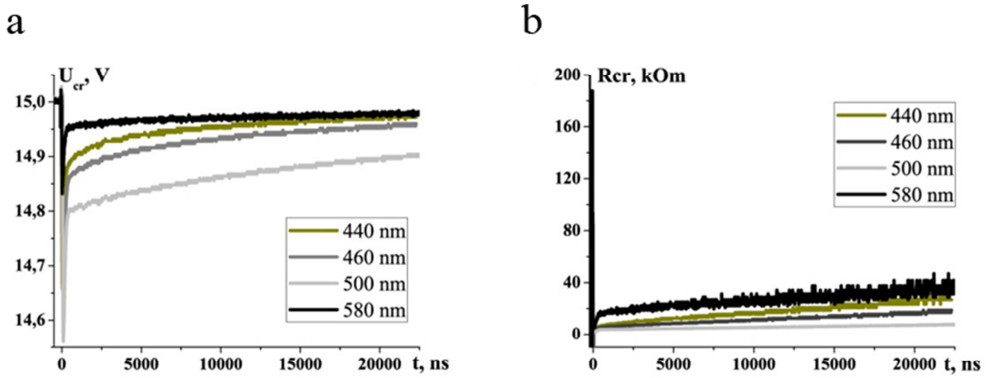

Fig. 3. The typical curve for response of the photorefractive bismuth silicon oxide crystal subjected to illumination with nanosecond laser radiation with intensity I=2 kW/cm^2 and the corresponding kinetics of the electrical resistance.

Generally the kinetics presented in Figure 3 are described by a complex non-single-exponential function:

$$U = U_0 + U_1 e^{-(t-t_0)/\tau_1} + U_2 e^{-(t-t_0)/\tau_2} + U_3 e^{-(t-t_0)/\tau_3} + \ldots + U_i e^{-(t-t_0)/\tau_i} \quad (1)$$

where t_0, U, U_0, U_1, U_2, U_3, U_i – mathematical approximation coefficients; τ_1, τ_2, τ_3, τ_i – relaxation times.

However, even approach of two-exponential function gives a good agreement with the experimental results, for example, it is rather good approximation for describing dependence of absorption on the relaxation time for different wavelengths after laser exposition during different periods of time (Matusevich et. al. (2008) and Kisteneva et.al. (2009)). Using such approximation the photoconductivity kinetics could be divided into the fast and slow components by two exponential functions. Thus for a bismuth silicon oxide crystal, time of a slow component depend on the radiation wavelength and power (Figure 4) and comes to 10 – 50 μs. At the same time fast component of signal is about of 100 ns. In the case of a bismuth titanium oxide crystal times of a fast component ranges from 15 to 40 ns and the same values of a slow component are from 1 to 4 μs. It is obviously that number of electrons in the conduction band is proportional to the product between the illuminating radiation power and the absorption coefficient of the sample. Figure 4 presents the lifetime of a slow component as a function of the number of electrons in the conduction band at the different wavelength. As seen, an increase in the number of electrons within the conduction band is responsible for shortening of the relaxation time.

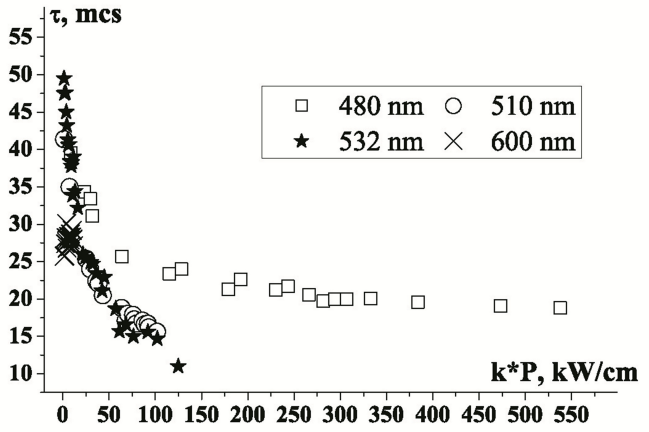

Fig. 4. The characteristic times of a slow component on two-exponential expansion for the photoexcitation relaxation kinetics in the bismuth silicon oxide crystal depending on the quantity proportional to the number of electrons in the conduction band, obtained for various wavelengths of illuminating radiation.

Fig. 5, a, b shows the photoconductivity signal as a function of the laser illumination intensity and wavelength. The spectral dependences of photoconductivity in the case of laser illumination are varying monotonically for the wavelengths above 500 nm. A decrease in the absorption coefficient with the increased wavelength leads to the corresponding decreasing of the photoconductivity. But when the wavelength of exposing illumination approaches to the energy of interband transition there is drastic increase in the absorption coefficient of the sample and so the observed decrease of the conductivity signal for the bismuth silicon oxide crystals in the short-wavelength spectral region at the wavelengths below 500 nm. Increase in the crystal absorption is illustrated by the calculated spectral dependence for the sample penetration depth (solid line in Fig. 5, *a*). For the sample of 4 mm thick complete exposure illumination could be distributed evenly across the sample and the maximum of photoconductivity spectral dependences close to the wavelength 500 nm. In the case of the bismuth titanium oxide crystals absence of maximum associates greater than the crystal thickness feasible penetration depth of radiation over the whole studied range from 500 to 600 nm.

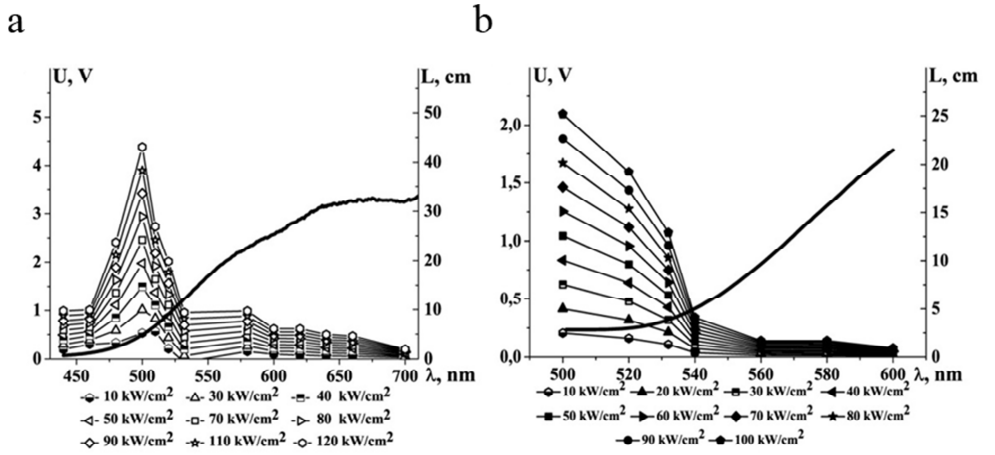

Fig. 5. Spectral dependence of the photoconductivity for the photorefractive single crystals of bismuth silicon oxide (a) and titanium oxide (b) crystals.

In the process of studies into the photo-induced absorption kinetics the photorefractive single crystal was illuminated with picoseconds laser pulses at the wavelength $\lambda = 532$ nm, the peak power at the front face of the crystal was from 10 kW/cm^2 to 100 MW/cm^2. When crystal is illuminated absorption decreases and in experimental setup it is fixed by changes in the signal amplitude of probing beam of a helium-neon laser at the wavelength $\lambda = 632.8$ nm.

The typical responses recorded with TDS-2022V two-channel oscilloscope and is given in Figure 6. The lower trace (channel 1) describes dynamics of the probe radiation from the helium-neon laser transmitted through the bismuth titanium oxide crystals; the upper trace (channel 2) is associated with picoseconds laser pulses.

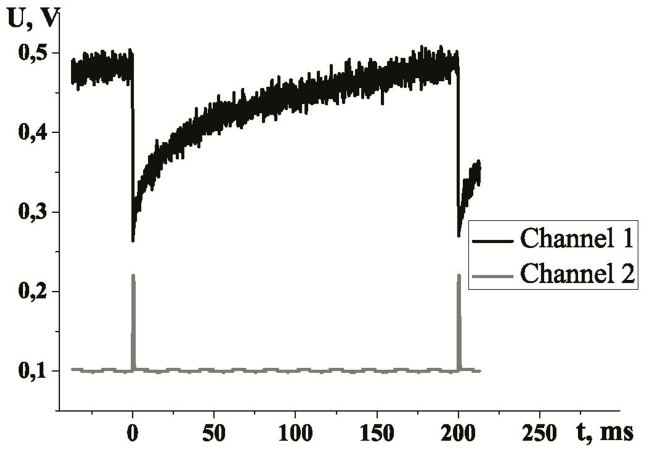

Fig. 6. Oscillograms for probe radiation (1st channel) and picoseconds pulses (2nd channel) at the repetition rate of laser pulses 5 Hz.

It is seen that picosecond excitation leads to a marked decrease in the crystal transmission that retains to its starting level until the next pulse. When the number of the absorbed pulses are increased the permanent transmission level become lower. To receive significant increase of transmission level photorefractive sample needs illumination by a great number of pulses ($\sim 10^3 - 10^4$).

In this case the kinetic dependences also have two components of relaxation time: slow component and fast component. A good quantitative agreement has been achieved for the characteristic times about 1ms for a fast component and to about 50 ms – for a slow component. These times are typical for the short-lived traps of bismuth titanium oxide crystals. In the paper by Matusevich et. al. (2009) and also by Kisteneva et.al. (2009) it has been noted that, apart from such traps, there are not only such type of traps but also deep traps having the lifetimes on the order of several hours or even days (Fig. 1).

In this way the physical mechanisms responsible for the charge transfer processes under light exposure is described as follows. When a photorefractive crystal is exposed to the visible or near ultra-violet light, the electrons are excited and move from the valence band and donor levels to the conduction band, leaving positively charged holes. These free electrons drift under the effect of the electric field applied to the crystal and are trapped by the short- or long-lived traps. A distinctive feature of the light-induced processes in the crystals of sillenite family is involvement of the traps with significantly differing lifetimes from several hundreds of nanoseconds to several days. Because of this, relaxation transitions are possible to both the shallow centers located close to the conduction band and to deep traps.

Conclusions

In this way the characteristics of relaxation kinetics have been established for the photoconductivity and photoinduced absorption processes. The obtained experimental results demonstrate the need for inclusion of shallow and deep trapping levels into description of the photoconductivity and induced absorption dynamics. Such results could be used in the development of a complex theoretical model for description of the light-induced transitions in the photorefractive crystals of sillenite family.

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