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Study of nonlinear-optical characteristics of photorefractive BSO and BGO crystals

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***Abstract*. Nonlinear refraction, nonlinear absorption, and optical limitation are studied in $Bi_{12}SiO_{20}$ (BSO) and $Bi_{12}GeO_{20}$ (BGO) crystals at the 532-nm emission wavelength of a picosecond Nd : YAG laser.**

Keywords: *photorefractive crystals, nonlinear refraction, two-photon absorption, optical limitation.*

1. Introduction

Photorefractive media attract great attention in the last years due to the possibility of their various applications in optoelectronics and laser physics. The possibility of using $Bi_{12}SiO_{20}$ (BSO), $Bi_{12}GeO_{20}$ (BGO), $Bi_{12}TiO_{20}$ (BTO), $BaTiO_3$, SBH, and other crystals for data processing and coherent amplification of radiation upon four-wave mixing have aroused considerable interest in them [1 – 3]. In the case of self-focusing of optical beams [4], photorefractive crystals can be used as optical switchers [5]. The possibility of employing BSO crystals as a storage medium with a multilayer optical memory was analysed for the first time in Ref. [6]. The polarisation properties of the crystals, which can be used as optical correlators, were analysed in Ref. [7]. The generation of spatial harmonics upon two-wave mixing in BGO and BSO crystals was investigated in Ref. [8]. At present photorefractive materials are most efficient media for the generation of phase-conjugated waves and real-time holographic recording ('dynamic' holograms). The applications of photorefractive media are analysed in detail in monograph [3]. In addition, the possi-bility of using these media in optical limiters, spatial light modulators, optical computers, etc. is being considered.

All the above potential possibilities of using photorefractive crystals require a detailed study of their optical and nonlinear-optical characteristics. While these characteristics in the field of a low-power light wave were analysed in many papers (see monographs [2, 3]), the optical and nonlinear-optical responses of these crystals in high-power radiation fields are not studied in fact at present.

In this paper, we report the study of nonlinear-optical parameters of BSO and BGO crystals (nonlinear refractive index, nonlinear absorption coefficient, and third-order nonlinear susceptibility) at a wavelength of 532 nm by the *z*-scan method. We also studied optical limitation in BSO and BGO crystals in the field of picosecond radiation and measured optical rotation in these photorefractive crystals in the visible and IR spectral regions.

2. Experimental

The BSO and BGO crystals were grown by the Czochralski technique on a Donets-1 setup. The crystals had the crystal symmetry 23. The BSO crystal had the form of a cube with sides 10 mm, while the BGO crystal had the form of a parallelepiped with sides $17 \times 11 \times 7.3$ mm. The faces of the BSO crystal were oriented with accuracy no worse than 1° along the $\langle 111 \rangle$, $\langle 112 \rangle$, and $\langle 110 \rangle$ directions. The BGO crystal was cut in the directions $\langle 001 \rangle$ (17×11-mm face), $\langle 010 \rangle$ (17×7.3-mm face), and $\langle 100 \rangle$ (11×7.3-mm face). The transmission spectra of the crystals are presented in Fig. 1. The linear absorption coefficients of BSO and BGO crystals at a wavelength of 532 nm were 1.04 and 1.32 cm^{-1}, respectively.

We used the second harmonic of a picosecond Nd : YAG laser ($\tau = 55$ ps, $\lambda = 532$ nm, $E = 0.1$ mJ) with a pulse repetition rate of 2 Hz to study the nonlinear-optical characteristics of the crystals by the *z*-scan method. Laser radiation was focused by lens (*1*) with the focal distance 25 cm (Fig. 2). The laser-beam waist radius was 46 – 48 μm.

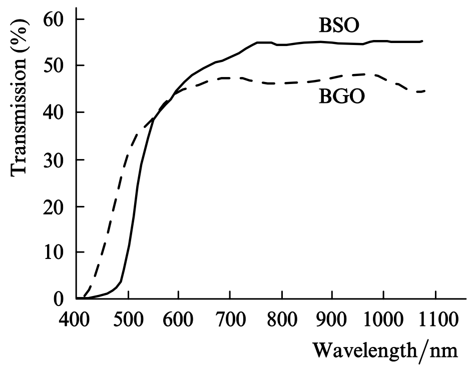

Figure 1. Transmission spectra of BSO and BGO crystals.

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The experiments were performed in the radiation intensity range from 10^8 to 7×10^9 W cm^{-2}. Crystal (*2*) under study (BSO or BGO) mounted on micropositioning stage (*8*) was moved along the optical axis z, passing through the focal region. The energy of laser pulses was measured with calibrated FD-24K photodiode (*4*). Aperture (*11*) of diameter 1 mm transmitting 3 % of laser radiation was placed at a distance of 120 cm behind the focal region (the limiting aperture scheme). An FD-24K photodiode was placed behind the aperture.

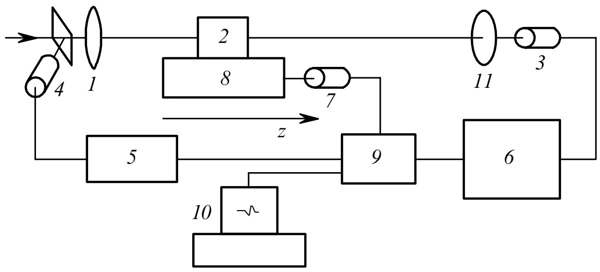

Figure 2. Scheme of the automated setup for measuring the nonlinear-optical parameters of photorefractive media: (*1*) focusing lens; (*2*) sample (BSO or BGO crystal); (*3*, *4*) photodiodes; (*5*, *6*) stroboscopic voltage converters; (*7*) four-phase step motor; (*8*) micropositioning stage; (*9*) controller; (*10*) PC; (*11*) limiting aperture.

The limiting aperture scheme allows one to determine the sign and magnitude of the nonlinear refractive index n_2 of the crystals, as well as the third-order susceptibility $\chi^{(3)}$. To find the two-photon absorption coefficient $\beta_{2\omega}$, the aperture was removed and the dependence of the transmission of a crystal on its position with respect to the focal point (open aperture scheme). Output signals from photodetectors (*3*) and (*4*) were fed to stroboscopic voltage converters (*5*) and (*6*), which were synchronously triggered by a signal from an avalanche photodiode. The digital output signals from both stroboscopic voltage converters were fed to controller (*9*), which was coupled with PC (*10*) through a serial interface.

A micropositioning stage with a crystal was moved with four-phase step motor (*7*) with a step of 20 μm per cycle. To reduce the influence of intensity fluctuations, the radiation intensity transmitted through the crystal was normalised to the radiation intensity detected in front of the focal region. Each point in Figs 3–8 corresponds to averaging over 20 pulses. The scheme and experimental procedures are described in detail in Ref. [9].

The optical rotation angles in BSO and BGO crystals were measured using radiation at 633 and 1150 nm from a He–Ne laser.

3. Results and discussion

Self-focusing of laser radiation (at 457 nm) in photorefractive media was first observed in a SBN crystal [10]. Self-focusing of laser radiation in a BSO crystal in an external electric field was analysed in Ref. [11]. Below, we present the results of studying self-focusing and other nonlinear-optical processes in photorefractive BSO and BGO crystals by the z-scan method. Measuring nonlinear-optical parameters of photorefractive crystals by other methods (four-wave mixing and nonlinear interferometry) involves difficulties due to the necessity to consider their optical activity, especially for such crystals as BSO, BGO, and BTO, which complicates (taking into account optical rotation in these crystals) the interpretation of experimental data. The z-scan method eliminates the influence of polarisation effects on the results of measurements [12].

The BSO and BGO crystals have large linear refractive indices (2.6 for BSO and 2.5 for BGO). Changes in these parameters in the field of an intense electromagnetic wave (due to a great nonlinear-optical addition of the nonlinear refractive index) lead to variations in the propagation and directivity of the electromagnetic wave. We studied self-focusing, nonlinear absorption, and optical limitation in crystals exposed to laser radiation with the power density $I \sim 10^9$ W cm^{-2}. The optical breakdown thresholds were 2×10^{10} W cm^{-2} for the BGO crystal and 4×10^{10} W cm^{-2} for the BSO crystal.

Figure 3 shows the dependence of the normalised transmission T of the BSO crystal (with the $\langle 110 \rangle$ orientation) obtained by the z-scan method using the limiting aperture scheme. A similar dependence was obtained for the $\langle 111 \rangle$; orientation of the crystal. Figure 3 demonstrates a positive effect of nonlinear refraction, which is characterised by the appearance of self-focusing (the dip in the dependence $T(z)$ in the front of the focal region followed by the peak behind the focal region). Note also that the curve $T(z)$ is asymmetric, its dip being deeper than the height of the peak. This is explained by nonlinear absorption.

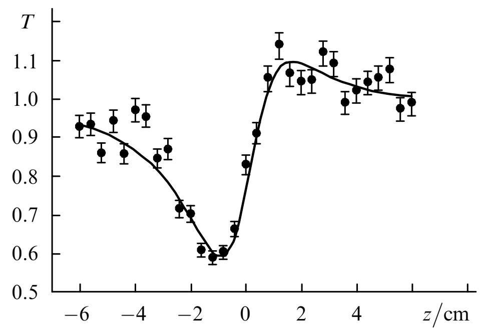

Figure 3. Dependence of the normalised transmission of the BSO crystal (the $\langle 110 \rangle$ orientation) on its position with respect to the focal plane in the limiting aperture scheme (solid curve: theory; circles: experiment).

One of the advantages of the z-scan method is the possibility of separating contributions from several nonlinear-optical processes. In the general case, when nonlinear refraction and nonlinear absorption are simultaneously observed in a sample (Fig. 3), the normalised transmission can be written in the form [13]

$$T(z) = 1 + \frac{4x}{(x^2+9)(x^2+1)}\Delta\Phi_0 - \frac{2(x^2+3)}{(x^2+9)(x^2+1)}\Delta\Psi_0, \quad (1)$$

where $x = z/z_0$; $z_0 = k\omega_0^2/2$ is the diffraction length of the beam; $k = 2\pi/\lambda$ is the wave number; ω_0 is the beam radius in the focal plane; $\Delta\Phi_0 = k\gamma I_0 L_{\rm eff}$ and $\Delta\Psi_0 = \beta_{2\omega} I_0 L_{\rm ef}/2$ are the phase shifts of the electromagnetic field in the focal plane caused by nonlinear refraction and nonlinear absorp-

Table 1. Nonlinear-optical parameters of BSO and BGO crystals at a wavelength of 532 nm.

Crystal	Crystallographic orientation	L/mm	$n_2/10^{-12}$ cgs units	$\mathrm{Re}\chi^{(3)}/10^{-13}$ cgs units	$\beta_{2\omega}/10^{-9}$ cm W^{-1}	$\mathrm{Im}\chi^{(3)}/10^{-13}$ cgs units
BSO	$\langle 110\rangle$	10	3.23 ± 0.64	9.38 ± 1.88	1.91 ± 0.38	11.57 ± 2.31
	$\langle 111\rangle$	10	4.36 ± 0.87	12.6 ± 2.52	1.62 ± 0.32	9.81 ± 1.96
BGO	$\langle 001\rangle$	7.3	7.36 ± 1.47	20.77 ± 4.15	3.65 ± 0.73	38 ± 7.6
	$\langle 010\rangle$	11	6.92 ± 1.38	19.5 ± 3.9	2.87 ± 0.57	29.8 ± 5.96
	$\langle 100\rangle$	17	7.44 ± 1.48	21 ± 4.1	3.62 ± 0.72	37.6 ± 7.52

tion, respectively; γ is the nonlinear refractive index; I_0 is the intensity of focused laser radiation; $L_{\mathrm{eff}} = [1 - \exp(-\alpha_0 L)]/\alpha_0$ is the effective length of the sample; α_0 is the linear absorption coefficient; L is the sample length; n_2 is the nonlinear refractive index related to γ by the expression $\gamma = 40\pi n_2/(cn_0)$ (γ is measured in m^2 W^{-1}, n_2 in the cgs units); c is the speed of light in m s^{-1}; n_0 is the linear refractive index of the sample; and $\beta_{2\omega}$ is the two-photon absorption coefficient. By introducing the notation $\rho = \beta_{2\omega}/(2k\gamma)$, we obtain the relation between $\Delta\Phi_0$ and $\Delta\Psi_0$ ($\Delta\Psi_0 = \rho\Delta\Phi_0$). In this case, Eqn (1) takes the form

$$T = 1 + \frac{2(-\rho x^2 + 2x - 3\rho)}{(x^2+9)(x^2+1)}\,\Delta\Phi_0\,. \tag{2}$$

Expression (2) gives the theoretical dependence of the normalised transmission of the crystal taking into account experimental parameters (Fig. 3). The best agreement between the theory and experiment was obtained for $\Delta\Phi_0 = 1.2$ for the $\langle 110\rangle$ orientation of the crystal. The values of the nonlinear refractive index n_2 and the real part $\mathrm{Re}\chi^{(3)}$ of the third-order nonlinear susceptibility for the $\langle 110\rangle$ and $\langle 111\rangle$ orientations of the BSO crystal are presented in Table 1.

The open-aperture study showed that nonlinear absorption was the same within the experimental error for both crystal orientations. Figure 4 shows the dependence of the normalised transmission of the BSO crystal with the $\langle 111\rangle$ orientation on its position obtained by the z-scan method with the open aperture. A similar dependence was obtained for the $\langle 110\rangle$ crystal orientation.

The absorption coefficient α of a medium can be written in the form

$$\alpha(I) = \alpha_0 + \beta_{2\omega} I. \tag{3}$$

The theoretical curve shown in Fig. 4 was plotted taking expression (3) into account. A slight deviation of the experimental points from the theoretical curve can be due to the error in measuring the beam-waist radius and the influence of refractive nonlinearities on the general picture of the normalised transmission in the open aperture scheme. This circumstance was taken into account in the estimate of experimental errors (Table 1). The two-photon absorption coefficients of the BSO crystal and the imaginary parts of the third-order susceptibility for different crystallographic directions are also presented in Table 1. Note that the difference between the values of nonlinear-optical parameters measured for different crystal orientations is very small.

The presence of nonlinear, in particular, two-photon absorption in the BSO crystal makes this crystal promising for the optical limitation of laser radiation. Figure 5 shows the dependences of the normalised transmission of the photorefractive BSO crystal on the laser radiation intensity.

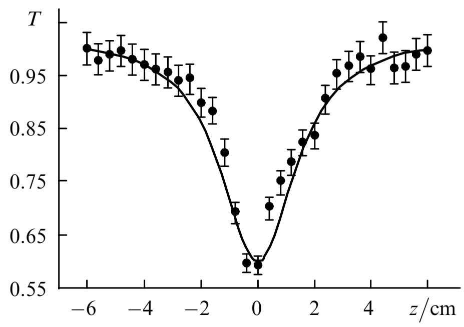

Figure 4. Dependence of the normalised transmission of the BSO crystal (the $\langle 111\rangle$ orientation) on its position with respect to the focal plane in the open aperture scheme (solid curve: theory; circles: experiment).

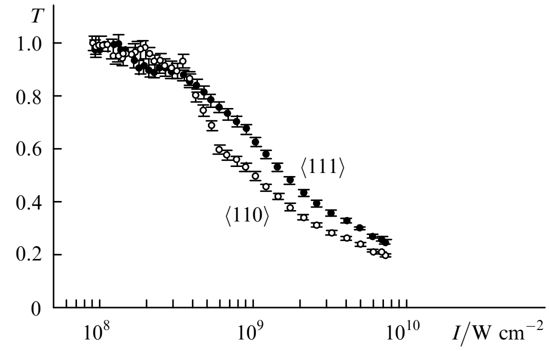

Figure 5. Dependences of the normalised transmission of the BSO crystal in the $\langle 111\rangle$ and $\langle 110\rangle$ orientations on the laser radiation intensity.

One can see that the transmission of the crystal decreases with the radiation intensity. In particular, we observed the decrease in the transmission by factors of 4.3 and 5.5 for the crystal orientations $\langle 111\rangle$ and $\langle 110\rangle$, respectively, for the intensity $I = 7 \times 10^9$ W cm^{-2}.

In the case of the BGO crystal, no qualitative differences were observed between the theoretical curves and experimental normalised transmission both for the limiting and open aperture schemes (Figs 6 and 7). Note that the signs of the real and imaginary parts of the nonlinear third-order susceptibility were invariable for the $\langle 001\rangle$, $\langle 010\rangle$, and $\langle 100\rangle$ crystal orientations. The nonlinear refractive indices, two-photon absorption coefficients, and the real and imaginary parts of the nonlinear third-order susceptibility measured for the $\langle 001\rangle$, $\langle 010\rangle$, and $\langle 100\rangle$ orientations of the BSO crystal are presented in Table 1.

Figure 8 shows the dependences of the normalised transmission of the BGO crystal on the laser radiation intensity. In this case, as for the BSO crystal, the transmission of laser radiation noticeably decreased when the laser radiation intensity exceeded 10^8 W cm^{-2}. We obtained

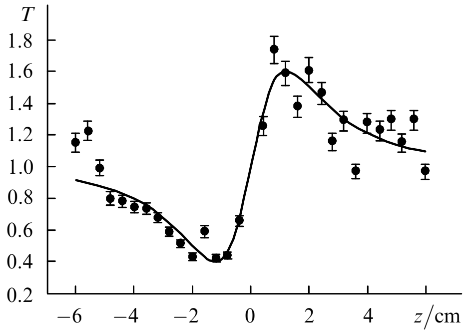

Figure 6. Dependence of the normalised transmission of the BGO crystal (the ⟨001⟩ orientation) on its position in the limiting aperture scheme (solid curve: theory; circles: experiment).

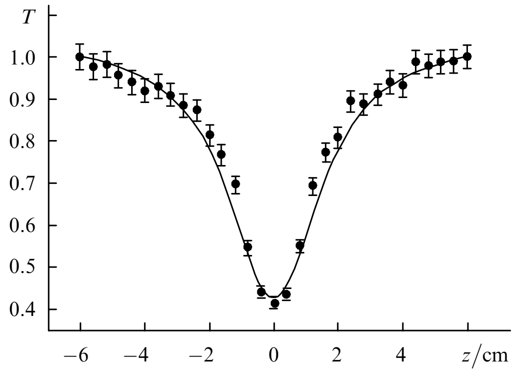

Figure 7. Dependence of the normalised transmission of the BGO crystal (the ⟨100⟩ orientation) on its position in the open Dependence of the normalised transmission of the BGO crystal (the ⟨001⟩ orientation) on its position in the limiting aperture scheme (solid curve: theory; circles: experiment).

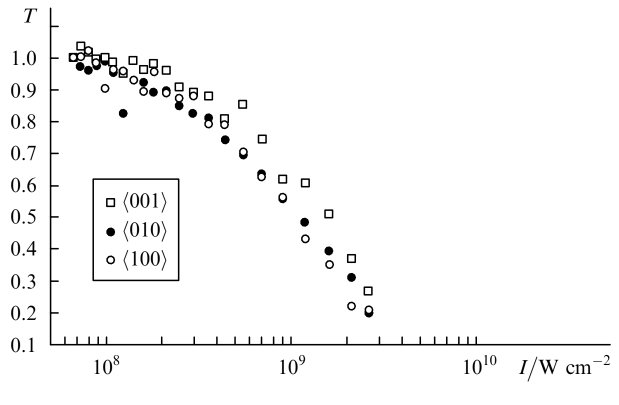

Figure 8. Dependences of the normalised transmission of the BGO crystal in the ⟨001⟩ and ⟨010⟩ orientations on the laser radiation intensity.

approximately five-fold optical limitation for the three orientations of the BGO crystal for the laser intensity $I = 3 \times 10^9$ W cm^{-2}.

The accuracy of our measurements of the normalised transmission was 3 %. The scatter in the experimental data was quite large (see Figs 3 and 6) and was reproduced from experiment to experiment. This question requires a special study. In this aspect, the dependences of the normalised transmission of photorefractive crystals differed considerably from 'smooth' dependences that we obtained earlier for other materials by the z-scan method. It is possible that the optical activity of crystals, their birefringence or metal impurities play some role in this case.

Some of the photorefractive media (in particular, BSO and BGO crystals) have a high optical activity, which is manifested in the optical rotation of linearly polarised radiation propagating in them. The optical activity is characterised by the optical rotation coefficient ρ_0, which determines the angle of optical rotation of light that has travelled a distance 1 mm in a photorefractive medium and is caused by the difference in the refractive indices for the left-hand and right-hand circular polarisations of light. The parameter ρ_0 is related to the gyration constant G by the expression

$$\rho_0 = \frac{\pi G}{\lambda n_0}. \tag{4}$$

The optical rotation angle is related to the parameter ρ_0 by the expression

$$\varphi = \rho_0 L. \tag{5}$$

Table 2 presents the values of the parameter ρ_0 measured for BSO and BGO crystals at wavelengths 632.8 and 1150 nm and the values of this parameter in the short-wavelength part of the spectrum taken from Ref. [3]. The data show that the rotational activity of these crystals in the UV region is higher than that in the IR region. The absorption of these crystals in the UV region is also higher than the IR absorption.

Table 2. Optical rotation coefficients for BSO and BGO crystals at some wavelengths.

Crystal	λ/nm	Crystallographic orientation	ρ_0/deg mm^{-1}	References
BGO	1150	⟨100⟩	5.3	This paper
	1150	⟨010⟩	4.5	This paper
	1150	⟨001⟩	4.8	This paper
	632.8	⟨100⟩	21.4	This paper
	632.8	⟨010⟩	20.4	This paper
	632.8	⟨001⟩	20.4	This paper
	632.8	–	21	[3]
	515	–	39	[3]
	488	–	46	[3]
BSO	1150	⟨111⟩	3.5	This paper
	632.8	⟨111⟩	23	This paper
	632.8	–	21	[3]
	515	–	38	[3]
	488	–	45	[3]

Therefore, crystals that have nonlinear-optical properties and optical activity considered above can be used as intracavity laser radiation limiters (or the elements of a passive negative feedback) in picosecond lasers. Note that such an application of these crystals has not been discussed yet. Semiconductor GaAs, CdSe, etc. crystals were used as such elements to generate stable, long trains of picosecond compressed pulses based on two-photon absorption and nonlinear refraction [14, 15]. The use of optical rotation of laser radiation along with processes considered above would allow the application of optical and nonlinear-optical properties of photorefractive crystals to solve this problem in the IR region.

4. Conclusions

We have studied the nonlinear-optical parameters of photorefractive BSO and BGO crystals at a wavelength of 532 nm by the z-scan method. We have measured the nonlinear refractive indices, nonlinear absorption coefficients, and the corresponding nonlinear susceptibilities of these crystals. The optical limitation of laser radiation was observed due to two-photon absorption at 532 nm in BSO and BGO crystals.

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