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# Effect of Incoherent Illumination on Two-Beam Interaction of Light Waves in a Bismuth Titanate Crystal

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**Abstract**—Experimental investigations and theoretical analysis of the effect of external illumination on the dynamics of formation of photorefractive reflection gratings in a (100)-cut bismuth titanate crystal showed that, in the case of counter interaction of laser beams with a wavelength of 633 nm, incoherent green light may cause a change in the sign of the two-beam gain.

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## INTRODUCTION

The two-beam interaction of coherent light waves in photorefractive crystals is the basis for many applications of dynamic holography, such as optical memory, adaptive correlation filtration and holographic interferometry, and spectral filtration of light [1–6]. Bismuth titanate crystals, due to their relatively fast photorefractive response [1], are often used as a dynamic medium in adaptive holographic devices [2, 4, 5]. Formation of dynamic gratings in reflection geometry in these crystals makes it possible to obtain a high photorefractive response without application of external electric fields [4, 5, 7, 8]. However, high photosensitivity of bismuth titanate leads to photoinduced light absorption, which accompanies counter interaction of light beams [7]. External incoherent illumination of bismuth titanate crystals, which occurs in real devices on their basis (for example, related to natural light), may significantly affect the efficiency of such interaction, causing charge redistribution over the defect centers involved in the formation of reflection holograms.

In this paper, we report the results of experimental investigation and theoretical analysis of the effect of external incoherent illumination on the dynamics of formation of photorefractive reflection gratings and the development of photoinduced light absorption in (100)-cut bismuth titanate crystals. It is found that green illumination of a crystal in which counter interaction of 633-nm light beams occurs may change the sign of the effective two-beam gain. A modification of the band transport model is proposed, which makes it possible to describe the dynamics of charge redistribution in an crystal upon its illumination both by an interference pattern of two coherent beams and by shorter-wavelength incoherent radiation, whose intensity can be changed during experiment. It is shown that the experimentally observed features of development of the efficiency of counter two-beam interaction and photoinduced light absorption can be qualitatively described within the modified model.

## EXPERIMENTAL SETUP

A bismuth titanate ($Bi_{12}TiO_{20}$, BTO) single crystal was grown by the modified Czochralski method from a nonstoichiometric high-temperature solution of bismuth and titanium oxides, taken in the ratio $Bi_2O_3 : TiO_2 = 10 : 1$ [9]. A sample with transverse sizes $4.5 \times 4.5$ mm$^2$ and thickness $d = 1.8$ mm was cut from the grown crystal perpendicularly to the [001] axis. An antireflection coating ($MgF_2$) was deposited on optically polished input and output faces of the sample.

A schematic of the experimental setup for studying the dynamics of formation of photorefractive reflection gratings under incoherent illumination is shown in Fig. 1. An He–Ne laser beam ($\lambda = 633$ nm), controlled by the gate *3*, was used to record reflective gratings in the BTO crystal. The quarter-wave plate QWP and polarizer P made it possible to set a necessary orientation of the laser polarization vector at the input face of the crystal ($x = -d$). The incident beam had the intensity $I_0 \approx 40$ mW cm$^{-2}$; the reflectance from the input and output faces, due to the antireflection coating, did not exceed 1.7%. The interference pattern of the beams incident on the output crystal face ($x = 0$) and reflected from it, at a sufficiently long exposure time (more than 1 s) causes the formation of a photorefractive hologram. The beam intensities $I_P$ and $I_S$ and their variations (caused by the interaction on the reflection hologram and the development of photoinduced light absorption)

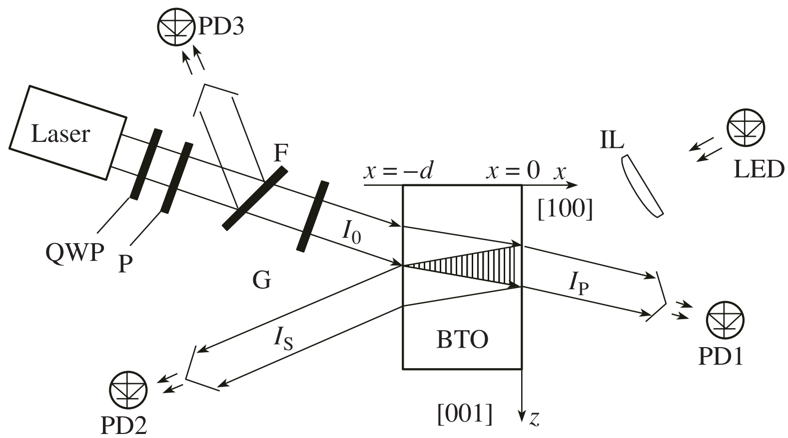


**Fig. 1.** Schematic of the experimental setup for studying the dynamics of formation of photorefractive reflection gratings under incoherent illumination.

were measured by the photodiodes PD1 and PD2. Beam-splitting plate F and photodiode PD3 were used to monitor the laser beam power. Note that light beam reflected from the input face of the crystal (not shown in Fig. 1) was spatially separated from the signal beam $I_S$ due to the slight tapering of the BTO crystal studied.

External incoherent illumination of the crystal was performed using a semiconductor light-emitting diode LED with the average wavelength $\lambda_i \approx 515$ nm and the spectral width $\Delta\lambda_i \approx 30$ nm. Homogeneity of illumination of the crystal region where the reflection hologram was formed was provided by the imaging lens IL. Variation in the current through the LED made it possible to control the illumination intensity in the range from 0.2 to 10 mW cm$^{-2}$.

## DYNAMICS OF REFLECTION GRATING FORMATION

The change in the optical absorption and the efficiency of counter interaction on the reflection grating, occurring during the experiment, were interpreted from the time dependences $I_P(t)$ and $I_S(t)$ within the inexhaustible pump approximation, since the pump ($P$) and signal ($S$) beams interacting in the crystal had intensities satisfying the inequality $\tilde{I}_P(x) \gg \tilde{I}_S(x)$. Generally, these intensities, due to the interaction on the reflection grating, are related by the expression [8]

$$\tilde{I}_S(x)\tilde{I}_P(x) = \tilde{I}_S(0)\tilde{I}_P(0)\exp(-\Gamma_{ef}x), \qquad (1)$$

where the effective gain $\Gamma_{ef}(t) = (2\pi/\lambda)n_0^3 r_{ef} E_{SC}(t)$ is determined by the refractive index $n_0$ of the crystal and the effective parameters, which take into account the features of counter interaction and grating formation dynamics: electro-optic constant $r_{ef}$ and the space charge field $E_{SC}$.

For inexhaustible pump, we assume that $\tilde{I}_P(x) = \tilde{I}_P(0)\exp(-\alpha x)$. As a result, we can find the absorption coefficient for a laser beam from the relation $\alpha(t) = \ln[I_0(1-R)^2/I_P(t)]/d$, which suggests equal reflectances $R$ for the input and output crystal faces. The effective gain can be found in this case from the experimental data: $\Gamma_{ef}(t) = \ln[I_S(t)/RI_P(t)]/d + \alpha(t)$.

The experiments on the dynamics of grating formation were performed in seven stages of the same duration: $\Delta t = 1800$ s. In the first stage of each experiment, the crystal was illuminated by only incoherent light with the intensity $I_{LED} = 0.2$ mW cm$^{-2}$. This procedure made it possible to erase the previously written hologram and obtain approximately the same initial state of the crystal. The dynamics of the development of photoinduced light absorption was not analyzed in detail in this stage; however, estimation showed that the absorption coefficient generally increased from the initial value $\alpha(0) \sim 0.3$ cm$^{-1}$ to $\alpha(t_1) \approx 1.6$ cm$^{-1}$.

In the second stage, incoherent illumination was retained the same as in the first stage. At the onset of the second stage ($t$ = 1800 s) the gate G (Fig. 1) was opened. As a result, we could measure furthermore (using the experimental data for $I_P(t)$ and $I_S(t)$) the efficiency of light-beam interaction under incoherent illumination on the photorefractive grating formed by these beams and changes in the optical absorption of the crystal. In each of the five subsequent stages, the incoherent illumination intensity increased in comparison with the previous stage.

The time dependences $\alpha(t)$ and $\Gamma_{ef}(t)$ calculated from the above relations for a laser beam with a polarization vector having an orientation close to optimal and making the angle $\theta_P(-d) = 50°$ with the $y$ axis at the input face of the crystal (see Fig. 1) are shown in Fig. 2a. The formation of a reflection grating, observed in the second stage of the experiment, is accompanied by an increase in the optical absorption of the crystal by ~0.35 cm$^{-1}$. With an increase in the incoherent light intensity, the photorefractive grating amplitude decreases and, at $I_{LED}$ = 3.0 mW cm$^{-2}$ for $t$ > 11 800 s, the effective gain becomes even negative.

It is known that, in the case of counter interaction in (100)-cut crystals, which is considered here, the sign of the effective electro-optic constant $r_{ef}$ depends on the orientation of the polarization vector [8]. The maximum negative value of $r_{ef}$ in the crystal studied is observed at $\theta_P(-d)$ = 140°. The time dependences $\alpha(t)$ and $\Gamma_{ef}(t)$ for such a polarization of the incident laser beam are shown in Fig. 2b. Comparison of the dependences $\Gamma_{ef}(t)$ in Figs. 2a and 2b, corresponding to effective electro-optic constants with different signs, suggests that the space-charge field $E_{SC}(t)$ of the reflection grating behaves identically in these experiments, although the time dependences of the optical absorption, $\alpha(t)$, are different.

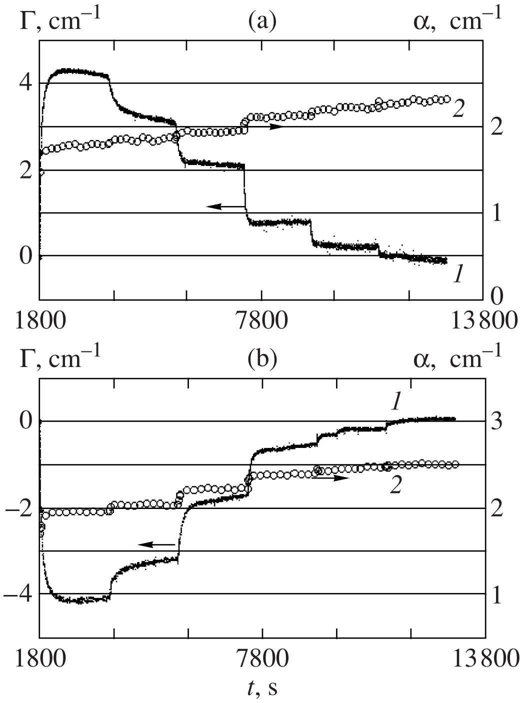


**Fig. 2.** Experimental dependences of the (*1*) effective gain $\Gamma_{ef}(t)$ for a photorefractive reflection grating and (*2*) the absorption coefficient $\alpha(t)$ (2) under incoherent illumination at the average wavelength $\lambda_i \approx 515$ nm, for two orientation angles of the laser beam polarization vector at the input face of the BTO crystal: $\theta_P(-d)$ = (a) 50° and (b) 140°. The illumination intensity $I_{LED}$ = 0.2 ($t$ = 1800–3600 s), 0.4 (3600–5400 s), 0.8 (5400–7200 s), 1.6 (7200–9000 s), 2.4 (9000–10 800 s) and 3.0 (10 800–12 600 s) mW cm$^{-2}$.

## THEORETICAL MODEL OF THE SPACE-CHARGE FIELD FORMATION FOR A PHOTOREFRACTIVE REFLECTION GRATING

Figure 2 indicates that an increase in the illumination intensity leads to an increase in optical absorption, followed by a decrease, down to a change in the effective gain sign. Such a change in the sign of $\Gamma_{ef}$, observed both at the positive value of the effective electro-optic constant $r_{ef}$ (for $\theta_P(-d)$ = 50°) and at $r_{ef} < 0$ (for $\theta_P(-d)$ = 140°), may occur only due to the change in the sign for the electric field of the grating.

For theoretical analysis of the dynamics of the field amplitude and photoinduced light absorption, we will use the band transport model, which suggests the presence of closely located donor-trap pairs in a crystal [10]. In the case of observation of photoinduced changes at a wavelength of 633 nm, which is considered here, it is sufficient to consider the two levels corresponding to donors ($E_D$) and traps ($E_T$), from which electrons can be excited by red light to the conduction band (Fig. 3). Incoherent green light excites electrons from the $E_D$ and $E_T$ levels and from the deep second donor center with the ionization energy $E_L$. We will take into account the effects of recombination of free charge carriers to the centers unoccupied by electrons; tunnel transitions of electrons between donor and trap centers, $E_D$ and $E_T$; and the transitions from traps ($E_T$) to empty (ionized) donors ($E_L$).

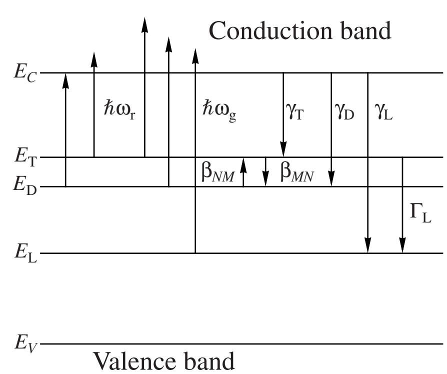


**Fig. 3.** Energy-band diagram of a BTO crystal for the band transport model with closely located donor–trap pairs and deep donor centers and a schematic diagram of electronic transitions (shown by arrows) under red (photon energy $\hbar\omega_r$) and green ($\hbar\omega_g$) illumination.

For a photorefractive grating with the vector $\mathbf{K} = (2\pi/\Lambda)\mathbf{x}^0$, directed along the $x$ axis, the material equations corresponding to the model under consideration have the form

$$\frac{\partial N}{\partial t} = -S_D(I_R + I_G)N + \gamma_D(N_0 - N - M)n + \beta_{MN}M - \beta_{NM}N, \quad (2)$$

$$\frac{\partial M}{\partial t} = -S_T(I_R + I_G)M + \gamma_T(N_0 - N - M)n - \beta_{MN}M + \beta_{NM}N - \Gamma_L ML, \quad (3)$$

$$\frac{\partial L}{\partial t} = S_L I_G(L_0 - L) - \gamma_L Ln - \Gamma_L ML, \quad (4)$$

$$\rho = e(L - M - N + N_0 - N_A - n), \quad (5)$$

$$\frac{\partial E}{\partial x} = \frac{\rho}{\varepsilon}, \quad (6)$$

$$\frac{\partial}{\partial x}\left(e\mu nE + \mu k_B T\frac{\partial n}{\partial x}\right) = -\frac{\partial \rho}{\partial t}, \quad (7)$$

where $N$ and $n$ are the concentrations of electrons at donor centers and in the conduction band, respectively; $S_D$ and $\gamma_D$ are, respectively, the photoexcitation cross sections of electrons from such donors and the constant for recombination to the donor center in an empty donor–trap pair; $M$ is the electron concentration in traps, $S_T$ and $\gamma_T$ are, respectively, the photoionization cross section and the recombination constant for trap centers; and $L$ is the concentration of ionized deep donor centers, which have the photoexcitation cross section $S_L$ in the neutral state and are characterized by the recombination constant $\gamma_L$. The coefficient $\Gamma_L$ characterizes the probability of tunnel transition of electrons from traps to deep donors, and $\beta_{NM}$ and $\beta_{MN}$ are the probabilities of tunnel transitions of electrons between the donor and trap in a donor–trap pair (Fig. 3). The parameters $N_0$, $L_0$, and $N_A$ are, respectively, the total concentrations of such pairs, deep donors, and the acceptors compensating for the charge of empty donor–trap pairs in darkness; $I_R$ is the laser intensity; $I_G$ is the incoherent light intensity; $E$ is the space-charge field of the grating; $e$ is the elementary charge; $\varepsilon$ is the static dielectric permittivity of the crystal; $\mu$ is the electron mobility; $k_B$ is the Boltzmann constant; and $T$ is temperature.

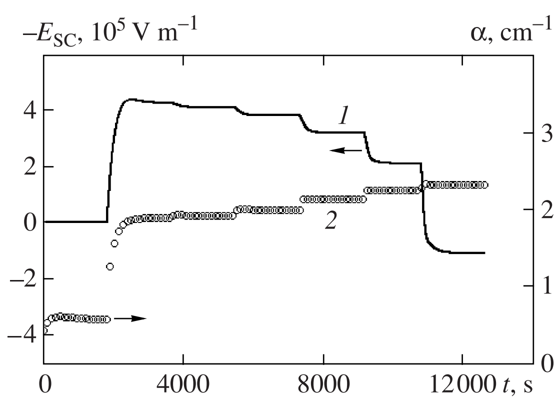


**Fig. 4.** Time dependences of the optical absorption coefficient $\alpha(t)$ and the space charge field $E_{SC}(t)$ for a photorefractive hologram in a BTO crystal, calculated in accordance with the experimental conditions.

Within the approximation of low contrast ($m \ll 1$) of the interference pattern forming the photorefractive grating, the system of equations (2)–(7) can be linearized by expanding unknown functions in Fourier series. The use of the approximations of adiabaticity and low light intensity leads to a closed system of equations for zero spatial harmonics, which was solved by numerical methods allowing simulation of incoherent illumination of a crystal. The functions found make it possible to calculate the time dependence of the optical absorption

$$\alpha(t) = \hbar\omega[S_D N^{(0)}(t) + S_T M^{(0)}(t)], \quad (8)$$

where $\hbar\omega$ is the laser photon energy; in addition, they can be used for numerical analysis of the system of equations for the amplitudes of the first spatial harmonics, $N^{(1)}(t)$, $M^{(1)}(t)$, $L^{(1)}(t)$, and $n^{(1)}(t)$. The dynamics of these charge gratings, according to Eqs. (5) and (6), determines the time dependence of the grating electric field:

$$E_{SC}(t) = \frac{e}{m\varepsilon|\mathbf{K}|} \times [L^{(1)}(t) - N^{(1)}(t) - M^{(1)}(t) - n^{(1)}(t)]. \quad (9)$$

## CALCULATION RESULTS AND DISCUSSION

The time dependences of the optical absorption and the space-charge field of a photorefractive hologram, calculated in accordance with the experimental conditions, using the above-described technique and relations (8) and (9), are shown in Fig. 4. Numerical analysis was performed for a grating with the period $\Lambda$ = 120 nm and the following material parameters of a crystal: $N_0 = 1.2 \times 10^{25}$ m$^{-3}$, $L_0 = 4.3 \times 10^{25}$ m$^{-3}$, $N_A = 2.6 \times 10^{24}$ m$^{-3}$, $\gamma_D = 8 \times 10^{-18}$ m$^3$ s$^{-1}$, $\gamma_T = 1.3 \times 10^{-15}$ m$^3$ s$^{-1}$, $\gamma_L = 3 \times 10^{-19}$ m$^3$ s$^{-1}$, $\Gamma_L = 2.54 \times 10^{-27}$ m$^3$ s$^{-1}$, $\beta_{NM} = 10^{-4}$ s$^{-1}$, $\beta_{MN} = 1.6 \times 10^{-3}$ s$^{-1}$, $S_T = 8.5 \times 10^{-5}$ m$^2$ J$^{-1}$, $S_D = 1.05 \times 10^{-5}$ m$^2$ J$^{-1}$, $S_L = 4 \times 10^{-5}$ m$^2$ J$^{-1}$, $\mu = 2 \times 10^{-6}$ m$^2$ V$^{-1}$ s$^{-1}$, and $\varepsilon = 4.16 \times 10^{-10}$ F m$^{-1}$.

Comparison of the curves in Figs. 2a and 4 shows that the experimentally observed dynamics of induced optical absorption and the efficiency of counter laser-beam interaction under incoherent illumination is in qualitative agreement with the results of the calculations within the model considered here. The analysis performed shows that the increase in the optical absorption with an increase in the illumination intensity is related to the preferred occupation of the trap centers having the photoionization cross section $S_T > S_D$ by electrons (see formula (8)). In this case, the amplitude of the trap-charge grating $M^{(1)}$ decreases, whereas, for the donor-charge grating $N^{(1)}$, which is in antiphase with $M^{(1)}$, the amplitude increases. The amplitude $L^{(1)}$ of the deep-donor grating, which is in phase with the trap-charge grating, increases with the illumination intensity, however, to a lesser extent, than $N^{(1)}$. As a result, the space-charge field, determined by relation (9), changes its sign at some illumination intensity. Note that the amplitude of the free-charge-carrier grating satisfies the inequality $n^{(1)} \ll N^{(1)}, M^{(1)}, L^{(1)}$.

## CONCLUSIONS

Thus, the possibility of controlling the efficiency and sign of the photorefractive reflection grating formed in a bismuth titanate crystal by 633-nm laser radiation under incoherent green illumination has been experimentally demonstrated. A modification of the band transport model has been proposed, according to which closely located donor–trap pairs (allowing photoexcitation of electrons to the conduction band by red light) and deep donor centers (the latter can be photoexcited only by incoherent light) are present in the crystal. The numerical analysis performed here shows that the experimentally observed dynamics of optical absorption and effective gain of the photorefractive reflection grating is in qualitative agreement with the predictions of the proposed modification of the theoretical model.

## ACKNOWLEDGMENTS

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## REFERENCES

1. Petrov, M.P., Stepanov, S.I., and Khomenko, A.V., *Fotorefractivnye kristally v kogerentnoi optike* (Photorefractive Crystals in Coherent Optics), St. Petersburg: Nauka, 1992.
2. Kamshilin, A.A., Jaaskelainen, T., and Kulchin, Yu.N., *Appl. Phys. Lett.*, 1998, vol. 73, p. 705.
3. Kamshilin, A.A. and Prokofiev, V.V., *Opt. Lett.*, 2002, vol. 27, p. 1711.
4. Romashko, R.V., Shandarov, S.M., Kul’chin, Yu.N., et al., *Izv. Akad. Nauk, Ser. Fiz.*, 2005, vol. 69, no. 8, p. 1143.
5. Romashko, R.V., Kul’chin, Yu.N., and Kamshilin, A.A., *Izv. Akad. Nauk, Ser. Fiz.*, 2006, vol. 70, no. 9, p. 1296.
6. Shamrai, A.V., Kozlov, A.S., Il’ichev, I.V., and Petrov, M.P., *Kvantovaya Élektron. (Moscow)*, 2005, vol. 35, no. 8, p. 734.
7. Ageev, E.Yu., Shandarov, S.M., Veretennikov, S.Yu., et al., *Kvantovaya Élektron. (Moscow)*, 2001, vol. 31, p. 343.
8. Plesovskikh, A.M., Shandarov, S.M., Mart’yanov, A.G., et al., *Kvantovaya Élektron. (Moscow)*, 2005, vol. 35, p. 163.
9. Prokofiev, V.V., Carvalho, J.F., Andreeta, J.P., et al., *Cryst. Res. Technol.*, 1995, vol. 30, p. 171.
10. Shandarov, S.M., Polyakova, L.E., Mandel, A.E., et al., *Proc. SPIE*, 2007, vol. 6595, p. 124.