Green light stimulation of photo-integrated micro-lattices

Vitaly A. Smirnov[a], Liubov I. Vostrikova*[a,b]
[a]Rzhanov Institute of Semiconductor Physics SB of RAS, Pr. Acad. Lavrentieva 13, Novosibirsk, Russia, 630090; [b]Novosibirsk State University of Economics and Management, Departments of Mathematics and Nature Sciences and Informational Technologies, Kamenskaya st. 52/1, Novosibirsk, Russia, 630099

ABSTRACT

Prolonged time of an existence for the nonlinear photo-integrated micro-lattices is connected not only with a conductivity of a medium but it depends on an illumination of the used samples. In this report the possibility of the green light stimulation of the photo-integrated micro-periodic second-order susceptibility lattices is demonstrated. The micro-lattices with very small amplitudes, which were preliminarily written using bi-chromatic powerful laser light, can be increased in some materials up to sufficiently big magnitudes under the action by green radiation. The optically stimulated growth of the small susceptibility lattices takes place independently on a polarization or a direction of propagation of the optical amplifying radiation. The observed phenomenon is not be explained only by considering of the possible nonlinear waves interactions in investigated samples and also may be related to the microscopic asymmetry processes of the optical transitions between local molecular centers. That leads to the appearance of the stimulated growth of the all-optically induced small micro-periodic separations of charges inside the medium and we analyze the possible processes in our investigations. The studied photo-integrated lattices may be useful in future for micro-optics and bio-photonics.

Keywords: green light stimulation, photo-integrated micro-lattices, optical poling, nonlinear-wave interactions.

1. INTRODUCTION

It is well known that the optical poling (OP) is so called an induction inside a medium the sufficiently long lived space periodic electrostatic polarization P(r) by an influence of the multi-chromatic inter-coherent multiple-frequency radiation that leads to a formation of the nonlinear optical periodicities of the second-order polarizability which is locally photo-integrated in the illuminated zone of the amorphous materials. OP creates a reversible change of the symmetry by the local range transforming of the isotropic medium into an optically uniaxial material inside which the nonlinear tree-wave interactions become possible[1-14]. The reversible changes of the optical properties in the illuminated zone of the investigated mediums are present in many phenomena found experimentally, for example, the photo-induced second harmonic generation (SHG)[1-12] inside the planar and volumetric lattices with the nonlinear polarizability $\chi^{(2)}$ which appear due to the OP of the isotropic mediums, Bragg variant of the diffraction and the distributed Raman-Nate light scattering[13] from the induced periodicities of the refractive index Δn due to the OP, the stimulated increase of the local anisotropy and the parametrical nonlinear process[14] due to the OP. The possibility of the OP of the different isotropic media (glasses and waveguides, fibers, polymer films and hybrids with consisting silicon and organic-non-organic materials) has been shown by now. The observed phenomenon of the OP and the processes of the conversion of a radiation inside the transformed zone of the media attract an attention of the scientists both from the point of view of the fundamental scientific research and due to the possibility of the obtaining new perspective broadband optoelectronic elements, in particular for bio-photonics.

Our experiments shown that, in comparison with the wide used simple method of the OP by means of the plane-parallel beams, the way of the volumetric OP by using the crossed Gaussian beams gives the important advantage connected with possibility of a wide variation of properties for the induced polarizability lattices in volumetric media, allowing to create small-scale micro periodicities of the photo-integrated anisotropy. In our work we used the special case of the OP of amorphous medium by means of the crossed Gaussian beams in volume of the samples under calculated specific incident angles for creation the homogenous micro-periodical lattices with sufficiently big sizes. However the prolonged time of an existence for the nonlinear photo-integrated micro-lattices in medium was connected not only with the big sizes of the photo-integrated lattices or a conductivity of the used medium but it depended also on the illumination of the photo-integrated lattices.

In this report the possibility of the green light stimulation of the photo-integrated micro-periodic second-order susceptibility lattices is demonstrated. The micro-lattices with very small amplitudes, which were preliminarily written using bi-chromatic powerful laser light, can be increased in some materials up to sufficiently big magnitudes under the action by green radiation. The optically stimulated growth of the small susceptibility lattices takes place independently on a polarization or a direction of propagation of the optical amplifying radiation. The observed phenomenon is not be explained only by considering of the possible nonlinear waves interactions in investigated samples and also may be related to the microscopic asymmetry processes of the optical transitions between local molecular centers. That leads to the appearance of the stimulated growth of the all-optically induced small micro-periodic separations of charges inside the medium and we analyze the possible processes in presented investigations.

2. STUDY OF GREEN LIGHT STIMULATION OF PHOTO-INTEGRATED MICRO-LATTICES

The OP of the samples in our experiments was made by two inter-coherent bi-chromatic powerful radiation with the fundamental $E_1=e_1A_1(r)\exp[ik_1r-i\omega t]$ and the frequency doubled $E_2=e_2A_2(r)\exp[ik_2r-i2\omega t]$ laser harmonics of the pulsed YAG:Nd^{3+} light source. As is known, the OP process is the result of the nonlinear wave interaction $\omega + \omega - 2\omega \rightarrow 0$ that leads to the appearance of the local optical polarization[15] or the non-zero space coherent photocurrent[16,17]

$$\mathbf{J} = \mathbf{e}_J A_1^2 A_2 \cos(\Delta\mathbf{kr}), \tag{1}$$

with accumulation of the periodic electrostatic field

$$\boldsymbol{E}(\mathbf{r},t) = \frac{I_1\sqrt{I_2}}{\sigma(I_1,I_2)}\mathbf{A}_0(\mathbf{r})\cos(\Delta\mathbf{kr})\left[1-\exp(-4\pi\sigma t/\varepsilon)\right] \tag{2}$$

Here $\sigma(I_1,I_2)$ is the effective conductivity which can depends on the intensities of the incident light laser radiations of fundamental and doubled frequencies, $\Delta\mathbf{k}=2\mathbf{k}_1-\mathbf{k}_2$ is the spatial periodicity of formed field $\boldsymbol{E}$, ε -permittivity of a medium.

The space separation of charges by means of the coherent photocurrent (1) results in a formation of the spatially periodic electrostatic field $\boldsymbol{E}$ (2) inside the beam interaction zone of the investigated sample. And the accumulated electrostatic field $\boldsymbol{E}$, induced upon the OP of medium, correspondingly causes a reversible change of optical properties inside the investigated sample. Note that we deal here only with the reversible variations in optical properties of a medium, which are caused by the spatial re-distribution of the charge density, and do not consider the question related with change in chemical or orientation compound. Thus, the modulation of the second-order susceptibility (nonlinear lattice's structure of $\chi^{(2)}\sim\chi^{(3)}E$, where the $\chi^{(3)}$ is the third-order susceptibility) appears upon the optical poling of the medium and it has a view

$$\chi^{(2)} = \chi_0^{(2)} f(\boldsymbol{r}) e^{i(\Delta\boldsymbol{kr}+\Delta\phi)} + k.c., \tag{3}$$

Here $f(\boldsymbol{r})$ is the envelope of the arising second-order susceptibility lattice, and the view of the tensor $\chi_0^{(2)}$ depends on the symmetry of medium used. Note that for different cases the modulation of the second-order susceptibility $\chi^{(2)} \sim \chi^{(3)} \boldsymbol{E}$ appearing upon the OP of an isotropic medium can be formed with different periodicities from some micro- up to tens nanometers and it has a tensor view.

So, the spatial periodicity of the photo-induced field $\boldsymbol{E}$ (2) with the lattice's period of $1/\Delta k$ creates the conditions for the existence of the photo-induced nonlinear three-wave interactions[1-14] in the corresponding photo-integrated volumetric micro-periodic structures of the nonlinear second-order susceptibility $\chi_{ijk}^{(2)} \sim \chi_{ijkl}^{(3)} E_l$ and the processes of the nonlinear three-wave interactions for the incident light radiations in such separated lattice's structures can be phase-matched on some phase conditions for the interactions of the light beams with a corresponding periodicity of the formed lattice's structures. It must be noted that there are the non-zero components of the second-order susceptibility tensor $\chi_{ijk}^{(2)}$ which give the existence of the nonlinear three-wave interactions also for the perpendicular polarized incident laser radiations, so that in the general case there are the corresponding polarization dependences for the arising nonlinear wave processes of photo-induced second harmonic generation and parametric down-frequency conversion in investigated isotropic mediums.

The question about a long lifetime and stability to various kinds of influence for the process of OP of isotropic mediums and for manufacturing of steady-stable photo-integrated micro structures of $\chi^{(2)}$ is very important. The simple theoretical estimations of the possible lifetimes for the photo-integrated lattices $\chi^{(2)}$ which were made on the base of taking into account only various possible conductivities of different materials on considering of the exponential dark Maxwell relaxation process are presented in Figure 1.

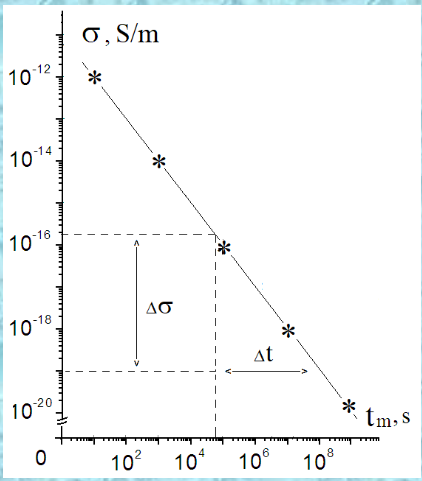

Figure 1. Theoretical estimations for lifetimes of the photo-integrated lattices $\chi^{(2)}$ in presence only pure material conductivities on considering of the dark Maxwell relaxation process.

One can see, there are the possibilities for the creation of sufficiently long-lived photo-integrated lattices of $\chi^{(2)}$ (from days up to years, see interval Δt in figure 1) in case of sufficiently low conductivities inside the interval Δσ in figure 1. However there is the problem connected with a possibility to obtaining the synthesized glasses or other isotropic materials with very low conductivities. For example, a pure silica has the conductivity only about 10^{-16} that gives a lifetime only about some days. But in real experiments the observed kinetic of the three-wave interactions on the photo-integrated lattice $\chi^{(2)}$ in glass materials is more complicate than the simplest theoretical exponential Maxwell curve. It was recently discovered in our experiments that in some glass materials there is the existence of the non-stationary regimes of the processes of three-wave interactions which are accompanied by the increase in time (or decrease) of the transformation efficiencies for the light pulses on the photo-integrated lattices $\chi^{(2)}$. It may be connected with different microscopic processes which are aroused in glasses by OP. Our last experiments are shown also that the photo-integrated anisotropic and nonlinear structures in some amorphous media can be long-lived and stable to the sufficiently powerful light laser illumination.

The experimental set-up on base of the pulsed YAG:Nd^{3+}-laser 1, see Figure 2, was used for OP of different isotropic materials with the creation of the photo-integrated space micro-periodic $\chi^{(2)}$ lattices. The basic (λ=1.064μm) and doubled frequencies of the radiation of laser 1 were separated in space on two channels by use optical elements and filters. Pulse energy of basic radiation was 45mJ, the longitude of pulse was 10ns, frequency repetition was 12,5Hz, and conversion into doubled frequency was 10%. The separated radiations were focused inside isotropic glass samples. During this illumination of sample with powerful inter-coherent two-frequency radiation (in other words during the process of OP) the electrostatic field grating E and, correspondingly, photo-integrated periodicity of the second-order (lattice $\chi^{(2)} \sim \chi^{(3)}E$) had been accumulating in region of the interaction of beams. The nonlinear three-wave conversions took place in sample on the accumulating lattice $\chi^{(2)}$. When only the basic frequency radiation falls on the photo-integrated $\chi^{(2)}$ then the process of nonlinear doubling of light frequency (second-harmonic generation $P_{shg}(2\omega)$ was observed on it in experiment. By using the photomultiplier we registered the radiation transformed on the photo-integrated $\chi^{(2)}$. Growth of signals of peak power of the photo-induced second harmonic $P_{shg}(2\omega)$ was observed on computer in real time.

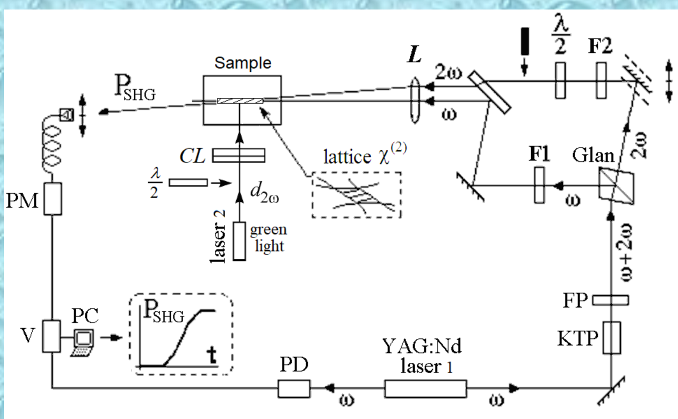

Figure 2. The scheme of the experimental set-up for OP and study of the outside green light stimulation of the photo-integrated micro-periodic second-order susceptibility lattices.

The experiments in this work were performed with use of the volumetric 1 cm^3 synthesized isotropic samples of a phosphate glass based. The procedure of the experiments consisted of the following. In the first stage, the initial photo-integrated space-periodic lattice $\chi^{(2)}$ with small amplitude was prepared using the experimental arrangement for OP depicted in Figure 2. The fundamental and second harmonic radiation components (ω and 2ω beams correspondingly in Figure 2) were linearly polarized in the plane of convergence and focused by a lens to intersect inside the experimental glass sample. The angle of the convergence of the laser beams incident onto the investigated sample was about 6.2^0, the beam diameters at the focus plane were about 170 and 120 μm, correspondingly, and the peak intensities at the waist were $I_\omega \sim 10^{10}$W/cm^2 and $I_{2\omega} \sim 10^9$W/cm^2. The estimated length of the lattice $\chi^{(2)}$ was about 0.3 cm.

So, the exposure to the bi-chromatic light resulted in the formation of the photo-integrated space-periodic structure $\chi^{(2)}$ inside the glass volume. The amplitude of the photo-integrated lattice $\chi^{(2)}$ was monitored in experiment by measuring the photo-induced second harmonic generation (SHG) efficiency for ω beam passing through the appearing lattice $\chi^{(2)}$ inside the investigated glass sample (see $P_{shg}(2\omega)$ beam in Figure 2). The SHG signals detected by a photoelectron multiplier were digitized using an analog-to-digital converter and fed to a computed for processing and display in real time. The YAG:Nd^{3+} laser 1 radiation energy was averaged over 10–40 pulses. The detection threshold of our measuring system was about 10^{-11} J per pulse. In the process of the measurement of the photo-induced second harmonic generation signal $P_{shg}(2\omega)$, the incident fundamental beam ω at the sample entrance was blocked for 5–10 seconds with a 5–10 minutes time interval.

In the second experimental step (performed after preparation on the small initial photo-integrated second-order susceptibility lattice of $\chi^{(2)}$ inside the investigated glass sample), the region with the formed initial lattice of $\chi^{(2)}$ was illuminated by different light radiations of the sufficiently powerful sources.

In first case, the region with small initial induced second-order susceptibility lattice $\chi^{(2)}$ was illuminated by the separated I_ω beam. By that the change in value of the amplitude of the photo-integrated lattice $\chi^{(2)}$ was measured as the second harmonic conversion efficiency η by calculation of the energy ratio of the $P_{shg}(2\omega)$ and $P(\omega)$ beams. In process of the illumination by the separated I_ω beam we observed the amplification of the initial photo-integrated lattice $\chi^{(2)}$ from small initial efficiency about $0.2 \cdot 10^{-5}$ up to maximal value $\eta_{\parallel}$ about $5.3 \cdot 10^{-5}$ in saturation which was observed after time period $t_{\parallel}$ about $1.9 \cdot 10^4$ s, see Table 1. So, there is the big growth of the photo-integrated lattice $\chi^{(2)}$ by illumination I_ω beam.

Table 1. Data of amplification of lattice $\chi^{(2)}$ by illumination beams.

Beam	$t_{\parallel}, s$	$t_{\perp}, s$	$\eta_{\parallel}$	$\eta_{\perp}$
I_ω	$1.9 \cdot 10^4$	$2.2 \cdot 10^4$	$5.3 \cdot 10^{-5}$	$5.2 \cdot 10^{-5}$
$d_{2\omega}$	$1.1 \cdot 10^4$	$2.3 \cdot 10^4$	$2.4 \cdot 10^{-5}$	$2.8 \cdot 10^{-5}$

We made also the experiment in which the polarization of the illuminated beam I_ω was perpendicular to the plane of the convergence of used by OP radiations which formed the initial lattice $\chi^{(2)}$. See results in Table 1. So, the observed polarization dependence was very small as in efficiency as well in saturation time, the effect of polarization was insignificant.
The obtained in our experiments results showed that the possible contribution of the three-wave processes, leading to the SHG in phase with the induced lattice's structure of the second-order susceptibility $\chi^{(2)}$, is insignificant or absent. It means that the observed phenomenon is not be explained only by considering nonlinear wave interaction in our medium and also may be related to microscopic asymmetry processes of the optical transitions between some local centers in a band gap of the investigated solid isotropic glass material that leads to the appearance and growth with time of the optically induced initial small micro-periodic electric charges separations inside the sample.

In order to exclude the possibility of the three-wave processes capable of pumping the lattices of the second-order susceptibility $\chi^{(2)}$, we performed a special experiments using an additional YAG:Nd laser 2, see figure 2, equipped with a frequency converter (with λ= 0.53 μm; pulse duration ~5 ns; repetition rate 12 Hz; maximal pulse power $P_{2\omega} \approx 40$ mJ) which is non-coherent with the incident beams used for the process of OP. The green light beam of this additional laser 2 was focused on the glass sample from the side face by means of a cylindrical lens (see $d_{2\omega}$ beam in figure 2) so as to cover the whole region of the initial photo-integrated lattice $\chi^{(2)}$ with the length of about 0.3 cm. The estimated maximal intensity of the green beam I_g of this additional laser 2 in the focal plane was about 10^9 W/cm^2.

The experimental data of the amplification of the initial photo-integrated lattice $\chi^{(2)}$ by the additional powerful outside green light beam $d_{2\omega}$ are presented in table 1. In this case we also observed the sufficiently big amplification of the initial photo-integrated lattice $\chi^{(2)}$, but the observed saturation value of the efficiency was about two times smaller. As can be seen, the efficiency of the amplification of the initial photo-integrated lattice $\chi^{(2)}$ in this case is also practically independent on the polarization of the outside green beam $d_{2\omega}$ and for it's perpendicular polarization it is comparable with the efficiency of the amplification for parallel polarization. However, the saturation time for the perpendicular variant was about two times smaller. It means that the accumulation process for charge separation in this case has more fast behavior.

So, all our experiments with outside illuminations presented in this work show that in the phosphate glass samples by the action of the outside powerful light field with the frequencies ω or 2ω (also not dependent for additional outside $d_{2\omega}$ source on it's polarization and on coherence with the initial light laser radiations used for the writing of the initial lattice's structure of the second-order susceptibility $\chi^{(2)}$ in a process of the OP) it was not observed a decrease in time for the amplitude of the initial photo-integrated lattices of the second-order susceptibility $\chi^{(2)}$ in our investigated phosphate glass samples. Moreover, in our synthesized glasses there is the sufficient amplification of the amplitude of the photo-integrated lattices $\chi^{(2)}$ by the action of the powerful light sources with frequencies ω or 2ω.

Also we made the experiments with use the different powers of the incident outside illumination $d_{2\omega}$. The obtained values of the efficiency by amplification of the initial photo-integrated lattices $\chi^{(2)}$ on the action of the outside $d_{2\omega}$ laser 2 radiation with different intensities is shown in Figure 3.

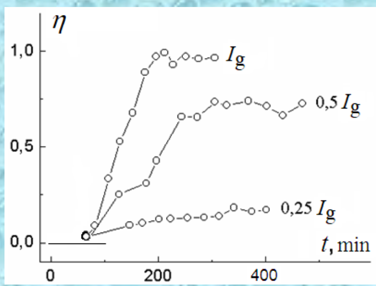

Figure 3. The dependence of the amplification of the photo-integrated initial lattices of the second-order susceptibility $\chi^{(2)}$ during it's illumination by outside YAG:Nd laser 2 radiation $d_{2\omega}$ with different intensities. The maximal pulse power for $d_{2\omega}$ beam was $P_{2\omega} \approx 40$ mJ, the maximal intensity for this radiation in focal place, correspondingly, was $I_g \sim 10^9$ W/cm^2.

One can see in Figure 3 there is the decrease of the maximal value of the amplification of the small initial photo-integrated lattice $\chi^{(2)}$ with decreasing of the power of the additional outside YAG:Nd laser illumination $d_{2\omega}$, but the dependence is not very sharp. So, in our samples there is also some possibility for the increase of the amplitude of the initial lattice of the second-order susceptibility $\chi^{(2)}$ in case when the value of the power of the additional outside YAG:Nd laser illumination $d_{2\omega}$ was chosen smaller in four times than the maximal one.

In conclusion, it can be noted that the obtained in this work experimental results show, in our opinion, the perspectives of the OP of the isotropic glass materials, in particular on the phosphate matrix base, for the creation in future of new broadband optoelectronic elements for the nonlinear frequency wave conversions of light pulses, potentially perspective in different areas of micro- and nano-optics, and also including for bio-photonics. By that, the observed possibility for amplification of the initial photo-integrated lattices $\chi^{(2)}$ can be used for compensation of the small lifetime and for long time support of the existence of lattices $\chi^{(2)}$ in samples.

ACKNOWLEDGEMENTS

The authors are grateful to group of Prof. V.V. Sokolov, Nikolaev Institute of Inorganic Chemistry SB RAS, and to group of Prof. O.S. Schavelev, Vavilov General Optical Institute of Saint-Petersburg, for offered number of synthesized samples for investigations. The work was supported by the Russian State Project No. 0242-2021-0012 "Coherent and nonlinear phenomena in homogeneous and structured media and in elements of photonics when interacting with intense laser radiation and charged particle beams".

REFERENCES

[1] Hickstein, D.D., Carlson, D.R., Kowligy, A., Diddams, S.A., Papp, S.B., Mundoor, H., Smalyukh, I.I., Khurgin, J.B., Srinivasan, K., Westly, D. “Self-organized nonlinear gratings for ultrafast nanophotonics,” Nature Photonics. 13(7), 494-499 (2019).

[2] Balakirev, M. K., Kityk, I. V., Smirnov, V. A., Vostrikova, L. I., Ebothe, J., “Anysotropy of the optical poling of glass,” Physical Review A 67, 023806-8 (2003).

[3] Odane, C., Tsutsumi, N., ”Phase-matched noncentrosymmetric polarization in a polymeric waveguide induced by all-optical poling,” JOSA B 20(7), 1514-1519 (2003).

[4] Smirnov, V.A., Vostrikova, L.I., “Volumetric anisotropic microlattices in compound materials,” Proc. SPIE 11846, 118460E (2021).

[5] Hirao, K., Qian, G., Wang, M., et. al., “Second-order nonlinearity in bulk azodye-doped hybrid inorganic-organic materials by nonresonant all-optical poling,” Chem. Phys. Lett. 381, 677-682 (2003).

[6] Smirnov, V.A., Vostrikova, L.I., “Impact of rare-earth doping upon micro-periodical anisotropy,” Proc. SPIE 11846, 118460L (2021).

[7] Daengngam, C., Heflin, J.R., Hofmann, M., Wang, A., Xu, Y., Liu, Z., “Demonstration of a cylindrically symmetric second-order nonlinear fiber with self-assembled organic surface layers,” Opt. Express 19(11), 10326-35 (2011).

[8] Kassab, L.R.P., Ozga, K., Slezak, A., Da Silva, D.M., Miedzinski, R., “Influence of gold nano-particles on optically stimulated in TeO_2–ZnO and GeO_2–PbO amorphous thin films,” Opt. Commun. 283(19), 3691-3694 (2010).

[9] Balakirev, M.K., Vostrikova, L.I., Smirnov, V.A., Kityk, I.V., Plucinski, K.J. “Limitation of optical poling in germanium-silicate glasses,” Opt. Commun. 230(1-3), 211-218 (2004).

[10] Valeev, A.I., Kundikova, N.D., Petrovsky, G.T., Churikov, V.M., Schavelev, K.O., Schavelev, O.S., Jakobson, N.A., ”New class of the glasses for the second-harmonic generation,” Journal of Optical Materials 68, 49-54 (2001).

[11] Huang, Z., Chen, A., Chen, Z., Deng, L., “Control of absorption and Kerr nonlinearity based on quantum coherence without driving field,” Modern Physics Letters B 24(30), 2921-2930 (2010).

[12] Liu, Y. L., Wang, W.J., Gao X.X., Zhang B.Y., Li, H., “Preparation and second-order nonlinearity of organic/inorganic hybrid materials doped with organic chromophore,” J. At. Mol. Sci. 2, 334-341 (2011).

[13] Balakirev, M. K., Smirnov, V. A., Vostrikova, L. I., “Self-diffraction of light upon optical poling of glass,” Sov. Quantum Electronics 32(5), 546-549 (2002).

[14] Smirnov, V. A., Vostrikova, L. I., “Parametric process during the optical poling of glass,” Bulletin of the Russian Academy of Sciences: Physics. 79(2), 176-180 (2015).

[15] Antonyuk, B. P., “All optical poling of glasses,” Opt. Commun. 174, 427-429 (2000).

[16] Baskin, E. M., Entin, M. V., “Hopping mechanism of coherent photo-voltaic effect and photoinduced polar anisotropy in glass”, [Proceedings of an International Workshop, Chicago 19-22 May, USA], Kluwer Academic Publishers, London, 191-202 (1998).

[17] Kovalev V.M., Sonowal K., Savenko I.G., “Coherent photogalvanic effect in fluctuating superconductors,” Physical Review B. 103(2), 024513 (2021).