Non-coherent stimulation of all-optically created nonlinear polarizability in glass

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The long exposure of an isotropic medium to a mutually coherent bi-frequency radiation with the basic and second frequencies of a powerful laser may decrease the optical symmetry of material and give rise to even orders in an expansion of polarization P(r) with respect to the light field. This process is called as all-optical poling that leads to a formation of the nonlinear optical structures of the second-order polarizability $\chi^{(2)}$ inside the amorphous isotropic materials. So, the all-optical poling creates a reversible change of the symmetry by local range transforming of the isotropic medium into an optically uniaxial material inside which the nonlinear tree-wave interactions become possible. The arising changes in optical properties of mediums are presented in many phenomena found experimentally, for example, photo-induced second harmonic generation and degenerate amplification of low light signals of the laser sub-harmonic inside the formed periodicity with the nonlinear polarizability $\chi^{(2)}$, which appear due to the all-optical poling of the isotropic mediums; Bragg self-diffraction and constructive light scattering from the induced modulations of the refractive index Δn due to the all-optical poling; observed light-electric instability of $\chi^{(2)}$structures and etc. The possibility of the all-optical poling of different isotropic media (volumetric glasses and waveguides, polymer consisted and hybrid organic-nonorganic films and fibers) has been experimentally demonstrated. The phenomenon of all-optical poling and the investigations of the photo-integrated periodic polarizability lattices $\chi^{(2)}$ attract an attention of scientists due to the possibility in future for a creating of new broadband optoelectronic elements of micro- and nano-optics, potentially perspective in different areas, in particular, for bio-photonics of molecular and atomic systems.

In this paper it's demonstrated that the nonlinear photo-integrated micro-periodic second-order polarizability lattices with very small amplitudes which were preliminarily recorded by using the bi-frequency powerful pulsed laser light in synthesized phosphate glass can be increased more than order of magnitude under action of non-coherent monochromatic radiation from the side of sample. The optically stimulated growth of the small micro-periodic polarizability lattices takes place independent of the polarization, by the direction of propagation of the optical amplifying radiation from the side and by different its intensities. The observed phenomenon of non-coherent stimulation of all-optically created polarizability in phosphate glass is not be explained only by considering of the possible nonlinear light waves interactions in sample and also may be related to the microscopic asymmetry processes of optical transitions between local centers in isotropic medium that lead to appearance and growth of the all-optically photo-induced small micro-periodic electric charges separations inside the region of interaction in sample, so that the various mechanisms can be responsible for the observed results in phosphate glass.

The experiments in this work were performed by using of the volumetric 1 cm^3 synthesized isotropic sample of phosphate glass. The procedure of experiments consisted of the following. In the first stage, the initial photo-integrated lattice's space-periodic structure of second-order polarizability $\chi^{(2)}$ was prepared using the experimental arrangement depicted in Figure 1.

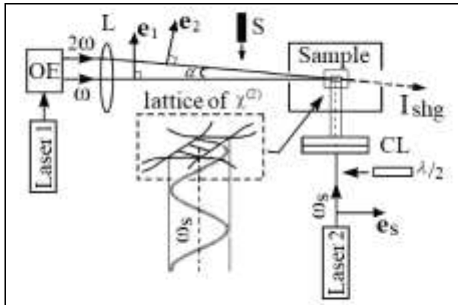

Figure 1. Schematic diagram of the experimental arrangement: Laser 1 - YAG:Nd^{3+}; OE - optical elements for frequency doubling and spatial separation of ω and 2ω beams; L - lens; S - shutter; Sample - phosphate glass; lattice - view of the photo-induced structure of $\chi^{(2)}$; Laser 2 - outside non-coherent source with green light $\lambda_S \approx 0.53$ μm; λ/2 - plate; CL - cylindrical lens.

The all-optical poling of the glass sample was made by two Gaussian inter-coherent bi-frequency powerful radiations with the fundamental $\mathbf{E}_1=\mathbf{e}_1 A_1(\mathbf{r})\exp[i\mathbf{k}_1\mathbf{r}-i\omega t]$ and the frequency doubled $\mathbf{E}_2=\mathbf{e}_2 A_2(\mathbf{r})\exp[i\mathbf{k}_2\mathbf{r}-i2\omega t]$ laser harmonics of pulsed YAG:Nd^{3+} light source (Laser 1 in Fig.1). As is known, the all-optical poling process is the result of the nonlinear wave interaction $\omega + \omega - 2\omega \to 0$ that leads to the appearance of the local optical polarization or non-zero space coherent photocurrent $\mathbf{J} = \mathbf{e}_J A_1^2 A_2 \cos(\Delta\mathbf{k}\mathbf{r})$ with accumulation of periodic electrostatic field inside the sample as

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

Here $\sigma(I_1,I_2)$ is effective conductivity which can depends on intensities of the incident light laser radiations of fundamental and doubled frequencies, $\Delta\mathbf{k}=2\mathbf{k}_1-\mathbf{k}_2$ is spatial periodicity of formed field $\boldsymbol{E}$, ε - permittivity of medium, $\mathbf{A}_0(\mathbf{r})$ - envelope of the lattice shape.

By all-optical poling of bulk medium the coherent photocurrent **J** has two components that are parallel and perpendicular to the planes of the formed field $\boldsymbol{E}$, and moreover, the envelope of the lattice of field $\mathbf{A}_0(\mathbf{r})$ has shape, which is formed by interacting Gaussian beams.

In contrast to the parallel component of current **J**, which causes the accumulation of charges only on the boundary of the region of interaction, the perpendicular to the planes of field $\boldsymbol{E}$ component of current **J** causes the accumulation of charges directly on the lattice planes, and the induced structure of field in this case is uniformly distributed in all region of interaction. The perpendicular case (i.e. volumetric all-optical poling) was maximally practically realized in experiment for big efficiency.

So, the electrostatic field, induced upon the all-optical poling of medium, causes the reversible change of optical properties inside investigated sample. Thus, the modulation of second-order susceptibility (nonlinear lattice of $\chi^{(2)}\sim\chi^{(3)}\boldsymbol{E}$, $\chi^{(3)}$ is the third-order susceptibility) appears upon the poling of medium and it has view $\chi^{(2)} = \chi_0^{(2)} f(\boldsymbol{r}) e^{i(\Delta\boldsymbol{k}\boldsymbol{r}+\Delta\phi)} + k.c.$,

Here $f(\boldsymbol{r})$ is the envelope of the arising lattice $\chi^{(2)}$ and note that the modulation of $\chi^{(2)}$ has a tensor view. The spatial periodicity of photo-induced field $\boldsymbol{E}$ with the lattice's period of $1/\Delta k$ creates the conditions for existence of photo-induced nonlinear three-wave interactions, as second harmonic generation and parametric down-frequency conversion. By that nonlinear processes for incident light radiations in lattice $\chi^{(2)}$ can be phase-matched on some phase conditions for interactions of light beams with corresponding periodicity of formed structure. It must be noted that there are non-zero components of second-order tensor which give the existence of 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 light radiation of powerful pulsed YAG:Nd^{3+} Laser 1 (λ= 1.06μm; pulse duration ~10 ns; repetition rate - 12.5 Hz; pulse energy ~46 mJ) and its second harmonic radiation (with transformation coefficient about 0.1) were separated into two different propagation's channels by special system of optical elements OE (see Figure 1). The fundamental and second harmonic radiation components (ω and 2ω beams in Fig. 1) were linearly polarized in convergence plane and focused by lens to intersect inside glass sample. The convergence angle of laser beams incident onto the glass was about 6.2^0, the beam diameters at focus plane were about 170 and 120 μm, correspondingly, and peak intensities at waist were I_ω~10^{10}W/cm^2 and $I_{2\omega}$~10^{9}W/cm^2.

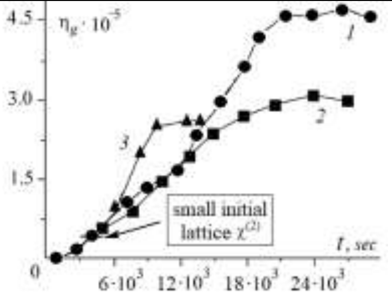

Figure 2. Amplification of SHG efficiency η_g in phosphate glass on growth of photo-induced lattice $\chi^{(2)}$: *1*- dynamics of lattice $\chi^{(2)}$ during preparing by bi-frequency Laser 1 YAG:Nd^{3+}radiation (i.e. traditional all-optical poling process); *2*, *3* - growth of small initial lattice $\chi^{(2)}$ stimulated by non-coherent outside illumination of green beam Laser 2. Curve *2* corresponds to polarization $\mathbf{e}_s$ of non-coherent light which is parallel to plane of convergence of the initial recording beams of Laser 1. Curve *3* corresponds to outside light with perpendicular polarization.

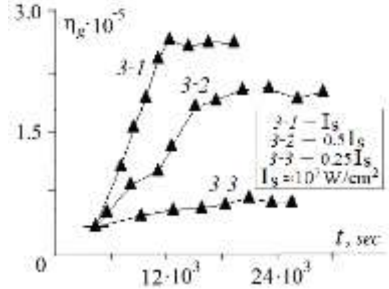

Figure 3. The dependence of amplification of the photo-integrated initial lattice of the second-order polarizability $\chi^{(2)}$ during it's illumination by outside radiation (Laser 2) with different powers. The maximal pulse power for outside beam ω_s was $P_s \approx 40$ mJ, the maximal intensity for this radiation inside lattice $\chi^{(2)}$ was ~10^7 W/cm^2.

The exposure to the bi-frequency light resulted in formation of the photo-integrated space-periodic structure $\chi^{(2)}$ inside glass volume. The amplitude of arising structure $\chi^{(2)}$ was monitored in experiment by measuring the photo-induced second harmonic generation (SHG) efficiency η_g for $I_1(\omega)$ beam passing through appearing lattice $\chi^{(2)}$ inside glass sample. The SHG signals (I_{SHG} beam in Fig.1) detected by photoelectron multiplier were digitized using analog-to-digital converter and fed to computed for processing and display in real time. The threshold of measuring system was ~ 10^{-11} J per pulse. In process of measurement of the photo-induced second harmonic generation signal I_{SHG}, the incident beam 2ω at sample entrance was blocked for 10 seconds with 10 minutes time interval.

In the second experimental step (performed after preparation on the 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 light radiation of outside non-coherent source Laser 2. The dynamics of the photo-induced SHG signal on growth of the photo-induced lattice $\chi^{(2}$ was monitored by periodically passed beam ω through the lattice for several seconds. During this several seconds period of time, only the radiation of laser beam $I_1(\omega)$ was passed through the amplifying lattice of $\chi^{(2)}$ and the energy of the aroused beam $I_{SHG,}$ photo-induced in this lattice of $\chi^{(2)}$, was registered on computer. The growing photo-integrated structures of $\chi^{(2)}$ were measured on second harmonic conversion efficiency up to η_g ~10^{-5}, which was determined as energy ratio of the I_{SHG} and $I_1(\omega)$ beams.

Figure 2 shows the time variation of the conversion efficiency η_g for the process of photo-induced SHG for performed experiments. Curve *1* describes the process of the recording of the photo-integrated space-periodic structure of $\chi^{(2)}$ in our glass sample under the action of bi-frequency light up to saturation (i.e. the traditional all-optical poling). The maximum photo-induced second harmonic conversion coefficient in this case was about η_g ~ 4,5·10^{-5}. The characteristic lifetime of the photo-integrated lattice's structure of second-order susceptibility $\chi^{(2)}$ in sample studied was 7–10 days.

Curves *2–3* in Figure 2 show the characteristics of obtained results of experiments during amplification of the photo-induced small lattice $\chi^{(2)}$ in our sample by action of outside light (source Laser 2). As can see, the amplification of the initial photo-integrated lattice's structure of $\chi^{(2)}$ exists for different polarizations of the outside light beam and for it's perpendicular polarization it is comparable value with the amplification for parallel polarization (see curves *2* and *3*), but writing times are different.

Also it was made the experiment with using different powers of the incident outside illumination. The obtained in experiment dependences of amplify-cation of the photo-integrated lattice $\chi^{(2)}$ by action of outside Laser 2 radiation with different powers is shown in Figure 3. One can see there is the decrease of maximal value of amplification of small initial lattice $\chi^{(2)}$ with decreasing of power of outside illumination Laser 2, but dependence is not very sharp. Thus, it was demonstrated that in phosphate glass sample there is possibility for increasing and maintaining of amplitude of initial lattice of second-order susceptibility $\chi^{(2)}$ when the power value of the outside illumination was chosen smaller in four times than the maximal one.

In conclusion, the investigation of the non-coherent stimulation of all-optically created nonlinear polarizability has been presented. It was demonstrated that nonlinear polarizability in phosphate glass in form of all-optically created periodic second-order susceptibility lattice with small amplitude, which were preliminarily written by using the bi-frequency powerful laser light, can be increased more than order of magnitude under action of non-coherent radiation from side of sample. The optical increase of small polarizability lattice takes place independent of polarization of outside optical amplifying radiation. The observed phenomenon is not be explained only by nonlinear wave's interaction in sample and also may be related to the microscopic asymmetry processes of optical transitions between active centers in band gap of the amorphous glass medium that leads to the appearance and growth of the small periodical electrical charges separations that all-optically induced inside sample. The investigations are interesting due to the possibility in future for creating of new photo-electronical elements on base of all-optically created nonlinear polarizability in glasses and other synthesized amorphous materials, potentially perspective for laser physics and also for bio-photonics. I am grateful to V.A. Smirnov, former researcher of Rzhanov Institute of Semiconductor Physics SB RAS for partial participation. Work was made as part of Russian State Task FWGW-2025-0019.

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