Thermal action on photo-integrated anisotropic micro-lattices

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ABSTRACT

The scientific problems connected with obtaining of the miniature components of the micro- and nanoscale light sources for investigations of various small objects with high spatial resolution and also for stable coherent real-time control during the process of an organization of the high-ordered molecular systems remain actual at present. It is known that exists the possibility for creating of the optically photo-integrated anisotropic micro-lattices with small periodicities down to nanoscale values in different transparent amorphous materials by using the pulses of frequency-multiplex laser radiation. But such photo-integrated anisotropic micro-lattices must be very stable for outside influence. In this paper the thermal action on photo-integrated anisotropic micro-lattices is investigated in phosphate samples and the some specifics of the action of heating are analyzed.

Keywords: photo-integrated micro-lattices, thermal action, nonlinear frequency conversion.

1. INTRODUCTION

It is known that the spatial periodicities of optical anisotropy inside the previously center-symmetrical isotropic materials (some polymer films, volumetric glass samples, fibers, light-guide elements etc.) can be created by using pulses of the inter-coherent frequency-multiplex of optical radiation[1-18]. For example in more used case, two components of YAG:Nd^{3+} laser with coherently connected harmonics of fundamental and doubled frequencies are enough to create the sufficiently long-lived photo-integrated spatially-periodic micro-lattices of the second-order polarizabilities ($\chi^{(2)}$ lattices). The occurrence of the spatial periodicities of optical anisotropy is considered as forming of a stable allocation of the spatially-periodical electric field owing to charges separations by emerging coherent photo-galvanic current[17,18] or also as the local modulated concentrations of the charges with an organization of the long-lived static polarization[16]. As a result, in isotropic materials on the photo-integrated anisotropic micro-lattices there are possibilities for an appearance of the three-wave interactions, such as the nonlinear frequency conversion of a laser radiation with the generation of the second harmonic (SHG)[1-14] and the degenerate parametrical growth of intensity of the sub-harmonic of light[15].

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One of the basic tasks of investigations is to make the matrix samples on a platform of the photo-integrated anisotropic micro-lattices which are perspective for the possible practical applications in different areas, in particular, for obtaining new broadband miniature converters of light signals and other optoelectronic components for laser optics, microscopy and bio-photonics. But, it is necessary to receive the matrix samples not only with high efficiencies of radiation's conversion but also with a sufficiently big lifetime and stability of the photo-integrated anisotropic micro-lattices to various kinds of inside influences.

In this paper the thermal action on photo-integrated anisotropic micro-lattices is investigated in phosphate samples and the some specifics of the action of heating are considered. We analyze the results of the outside influence of heating on process of the induction of the space periodic electrostatic polarization P(r) in medium and, correspondingly, on the formation of the nonlinear optical micro-lattices of second-order polarizability $\chi^{(2)}$ in amorphous material. The attention in process is given to considering the outside influence of the variations of temperature up to high values. In some phosphate samples the perfectly new possibility of the increase of the efficiency of induction of the optical micro-lattices was discovered. The corresponding experiments have been made and the possible mechanisms of the observed and investigated processes are discussed.

2. INVESTIGATION OF TERMAL ACTION ON ANISOTROPIC MICRO-LATTICES

Consider the outside influence of heating on process of the induction of the space polarization P(r) and on the formation of the photo-integrated anisotropic micro-lattices. In classical case for induction in amorphous mediums the space periodic polarizability it's used two inter-coherent bi-chromatic powerful radiation with the fundamental $\boldsymbol{E}_1=\boldsymbol{e}_1E_1(\boldsymbol{r})\exp[i\boldsymbol{k}_1\boldsymbol{r}-i\omega t]$ and the frequency doubled $\boldsymbol{E}_2=\boldsymbol{e}_2E_2(\boldsymbol{r})\exp[i\boldsymbol{k}_2\boldsymbol{r}-i2\omega t]$ laser harmonics. If the light beams propagate due to formation of micro-lattice inside a sample at small angles to the direction of the *y* axis and have the Gaussian profiles along the x and *z* axes that:

$$E_1(\boldsymbol{r}) = E_{10}\exp\left(-\frac{x^2+z^2}{2w_1^2}\right), \quad E_2(\boldsymbol{r}) = E_{20}\exp\left(-\frac{x^2+z^2}{2w_2^2}\right), \tag{1}$$

where w_1 and w_2 are the radii of the beams of the used laser radiations at the half-maximum intensity level in the focal plane. This situation appears in the experiment upon focusing practically collimated beams. By that the region with the photo-integrated periodical anisotropic lattice has the micro-size 2D form but it prolonged along *y* direction up to some millimeters.

It is known that the process of the induction of the space polarization P(r) is a result of the coherent phase-matched nonlinear interaction $\omega + \omega - 2\omega \rightarrow 0$ that leads to appearance of the local optical polarization[16] or the non-zero space coherent photo-galvanic current[17,18]

$$\boldsymbol{J} = \boldsymbol{e}_J E_1^2 E_2 \cos(\Delta\boldsymbol{k}\boldsymbol{r}), \tag{2}$$

In such geometry of the formation of the anisotropic micro-lattice the expression for the coherent photo-galvanic current can be described as:

$$\boldsymbol{j}(\boldsymbol{r}) = \boldsymbol{e}_j E_{10}^2 E_{20}\exp\left(-\frac{x^2+z^2}{2a^2}\right)\cos(\Delta ky), \qquad a^2 = \frac{w_1^2 w_2^2}{2w_2^2 + w_1^2}. \tag{3}$$

Where the components of the arising current are connected with the angles on the interaction of optical radiations:

$$\begin{aligned} e_{jz} &= \sigma_1\cos\alpha_1\cos\varphi_0 + \sigma_2\cos\alpha_2 \\ e_{jx} &= \sigma_1\sin\alpha_1\cos\varphi_0 + \sigma_2\sin\alpha_2 \end{aligned}. \tag{4}$$

Here e_{jz} and e_{jx} are the nonzero components of the current, φ_0 is the angle between the vectors e_1 and e_2 , α_1 and α_2 are the angles between the vectors e_1 and e_2 and the *z* axis, respectively, $\Delta k = |\Delta\boldsymbol{k}| = |2\boldsymbol{k}_1 - \boldsymbol{k}_2|$, $\boldsymbol{k}_1$ and $\boldsymbol{k}_2$ are the wave vectors of the laser $\omega+2\omega$ bi-chromatic radiations.

The appearance of the induced photo-galvanic current (see equations (2) and (3)) during the illumination of samples by inter-coherent bi-chromatic light waves (1) after long interval of time leads to accumulation of the long-lived periodic electrostatic field

$$\boldsymbol{E}(\mathbf{r},t)=\frac{\mathrm{I}_1\sqrt{\mathrm{I}_2}}{\sigma(\mathrm{I}_1,\mathrm{I}_2,\mathrm{T})}\boldsymbol{A}(\boldsymbol{r})\cos(\Delta\boldsymbol{kr})\left[1-\exp(-4\pi\sigma(\mathrm{I}_1,\mathrm{I}_2,\mathrm{T})t/\varepsilon)\right] \tag{5}$$

Here σ $(\mathrm{I}_1,\mathrm{I}_2,\mathrm{T})$ is the effective conductivity which depends on intensities of the interacted light harmonics and in our case also on used temperature during the heating of samples, ε -permittivity.

The induced electrostatic field $\boldsymbol{E}$ causes a reversible change of optical properties inside the investigated sample. Thus, the modulation of the second-order polarizability (photo-integrated anisotropic micro-lattice $\chi^{(2)}\sim\chi^{(3)}\boldsymbol{E}$, where the $\chi^{(3)}$ is the third-order polarizability) appears in amorphous medium and it has a tensor view

$$\chi^{(2)}=\chi_0^{(2)}f(\boldsymbol{r})e^{i(\Delta\boldsymbol{kr}+\Delta\phi)}+k.c., \tag{6}$$

Here $f(\boldsymbol{r})$ is the lattice's envelope, and the view of the tensor $\hat{\chi}_0^{(2)}$ depends on a symmetry of medium used.

Further the spatial periodicity of the inserted electrostatic field $\boldsymbol{E}$ with period of $1/\Delta k$ (see the equation (5)) creates the conditions for existence of the electrically induced three-wave interactions in the corresponding induced micro-periodic lattices of nonlinear second-order polarizability $\chi_{ijk}^{(2)}\sim\chi_{ijkl}^{(3)}E_l$ and the processes of the nonlinear conversion of light waves in such separated lattice's structure may be phase-matched for the concrete radiation.

In our experiment we created the volumetric anisotropic micro-periodical modulations of nonlinear second-order polarizability $\chi^{(2)}\sim\chi^{(3)}\boldsymbol{E}$ and investigated the nonlinear light wave conversion processes on these induced spatial micro-lattices during the outside influence by heating. The experimental procedure was as follows. The experiments were performed with the bulk 1 cm^3 amorphous samples on a base of phosphate glass matrix. In the first stage of experiments, the initial anisotropic spatial-periodic micro-lattice of $\chi^{(2)}$ was prepared using the experimental setup for the process of the all-optical induction which presented in Figure 1.

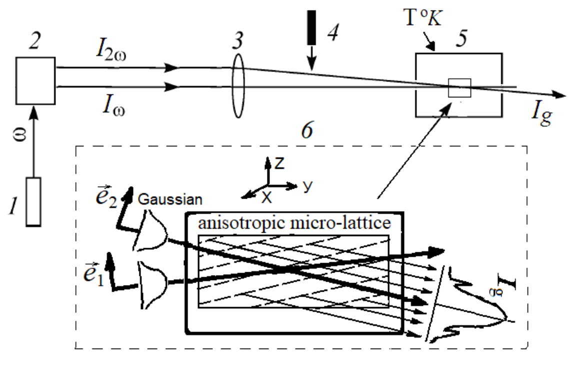

Figure 1. The scheme of the experimental setup: *1* – base powerful YAG:Nd^{3+} laser; *2* - optical elements for frequency doubling and spatial separation of ω and 2ω laser beams; *3* - lens; *4* - shutter; *5* – investigated sample inside the thermal furnace with controlled temperature T^0K; *6* – schematic process of the SHG in photo-integrated micro-lattice of $\chi^{(2)}$.

In experiments the radiation of a pulsed YAG:Nd^{3+} laser (with wave length λ = 1.06 μm; pulse duration about 10 ns; repetition rate - 12.5 Hz; pulse energy of radiation ~ 46 mJ) and its second harmonic (with coefficient of the nonlinear transformation about 0.1) were separated into two channels by a special system of optical elements. By using the filter systems another component of the frequencies of the laser radiation in each channel was attenuated by 10^{10}. The fundamental and second harmonic radiation components (I_ω and $I_{2\omega}$ beams, see in Figure 1) were linearly polarized in the plane of the convergence and focused by a lens to intersect inside the investigated glass sample. The angle of convergence of the laser beams incident on the investigated sample was about 6.2^0, the beam diameters at the focus were about 170 and 120 μm, and the peak intensities of the incident laser radiations at the waist were $I_\omega\sim10^{10}\mathrm{W/cm}^2$ and $I_{2\omega}\sim10^9\mathrm{W/cm}^2$, correspondingly.

Further the long-time exposure by using the bi-chromatic inter-coherent laser light resulted in the formation of the photo-integrated micro-lattice $\chi^{(2)}$ inside the volume of the sample. The amplitude of micro-lattice $\chi^{(2)}$ was monitored by measuring the process of the nonlinear second harmonic generation (SHG) for the I_ω beam passing through the micro-lattice $\chi^{(2)}$ (see I_g beam in Figure 1). The SHG intensity signals detected by a photoelectron multiplier were digitized using an analog-to-digital converter, and further the radiation intensity signals was averaged over 10–40 pulses and fed to a computer for processing and display in real time. The detected power threshold of the measuring system was about 10^{-11} J per pulse. For the measurement of the I_g signal, the incident $I_{2\omega}$ beam was blocked for 5–10 seconds with a 5–10 minutes interval at the sample entrance.

To perform the experiments of the outside influence of the heating on the formation of the micro-lattice $\chi^{(2)}$ we used a thermal furnace in setup with control of temperature. The experiments were concentrated on the investigations of the outside thermal influence on our samples with induced micro-lattice of $\chi^{(2)}$. In doing so during the process of the induction of the micro-lattice $\chi^{(2)}$ the samples of the phosphate glasses were located inside the small furnace and the temperature of the investigated sample during the process were changed. The special temperature control system has a feedback and was very sensitive and stable. We had the possibility for variation of the temperature of studied sample during formation of the second-order space-periodic micro-lattice $\chi^{(2)}$ in big interval of values from room temperature up to 523K with sufficiently small variations about 2,5K. The stability and registering precision of the temperature for the investigated sample inside the small furnace were high for each fixed level.

In our experiments we investigated the step kinetics of the induction process at different temperatures of the glass sample by study of the behavior of the signal of the intensity of the nonlinear SHG during the writing of the space-periodic micro-lattice $\chi^{(2)}$. The averaged step results of our experimental investigations are presented in Table 1.

Table 1. Data by heating of micro-lattices.

Steps of formation of micro-lattice $\chi^{(2)}$ without heating (at room temperature 293K)			**Steps of formation of micro-lattice $\chi^{(2)}$ during heating (from room temperature up to 523K)**			
t*, *min	**I_g, *a.u.***	**$\chi^{(2)}$**	***t*, *min***	***T*, *K***	**I_g, *a.u.***	**$\chi^{(2)}$ (T)**
0	0.05	Growth	0	293	0.05	Growth
25	0.05		25	298	0.05	
50	0.07		50	303	0.08	
75	0.11		75	313	0.1	
100	0.33		100	323	0.24	Slowly growth
125	0.49		125	333	0.31	
150	0.62		150	343	0.37	
175	0.76		175	353	0.4	
200	0.89		200	363	0.42	
225	0.95		225	373	0.44	
250	0.98	Saturation	250	383	0.5	
275	1		275	393	0.54	
300	0.99		300	403	0.59	
325	0.98		325	413	0.61	
350	0.99		350	423	0.65	
375	1		375	433	0.69	
400	0.97		400	443	0.72	
425	1		425	453	0.75	
450	0.98		450	463	0.77	Saturation 80%$\chi^{(2)}$
475	1		475	473	0.8	
500	1		500	483	0.78	
525	0.97		525	493	0.05	Collapse $\chi^{(2)} = 0$
550	0.99		550	503	0.04	
575	1		575	513	0.06	

The observed process of the writing of the micro-lattice $\chi^{(2)}$ at room temperature in our phosphate samples has as traditional view when the signal of the intensity of the SHG increases with time and reaches the same steady-state value which is its maximum one. The time of the writing of the micro-lattice $\chi^{(2)}$ up to steady-state level was about 4 hours (see Table 1) and the maximal observed efficiency at saturation of the SHG $\eta_g = I_g / I_\omega$ was about $4{,}5 \cdot 10^{-5}$. The room lifetime of the recorded micro-lattice $\chi^{(2)}$ in the investigated phosphate glass samples was about 5-7 days. Theoretically estimated by us the saturation level, which corresponds to maximal value of the modulation amplitude of micro-lattice $\chi^{(2)}$ in our sample, was ~ 70 times less than it in the well-known optical nonlinear crystals.

Further in our experiments we investigated the process of the writing of the micro-lattice $\chi^{(2)}$ in our phosphate sample during the outside influence of heating at different temperatures. The writing of the space-periodic structure of micro-lattice $\chi^{(2)}$ in this case was carried out when the glass sample was periodically heated by the step increase of the temperature inside the furnace (in which the sample was located one). So, the Table 1 contains also the typical experimental results of the step kinetics during the process of the induction with the formation of the space-periodic micro-lattice $\chi^{(2)}$ in investigated sample at different temperatures. One can see that the increase of the temperature of the studied glass sample during the induction process is no accompanied by a decay of the intensity of the SHG signal. Conversely, we observe the increase of the intensity of the SHG signal that indicates the presence of the writing and amplification of the amplitude of the space-periodic micro-lattice $\chi^{(2)}$ in our investigated glass sample. The process of the growth of the amplitude of micro-lattice $\chi^{(2)}$ in the heating glass is some different from earlier observed at room temperature. It is more slowly than in case at room temperature and the full time of the writing of the micro-lattice $\chi^{(2)}$ is longer approximately in two times. The process of the writing of the micro-lattice $\chi^{(2)}$ takes place up to high temperature about $483K$ and then, at the temperature more than $483K$, the sharp relaxation of the SHG signal is appeared (see by result data in Table the collapse moment). It is obviously that the observed behavior means that the micro-lattice $\chi^{(2)}$ is destroyed only at temperatures more than $483K$. We heated the sample further up to $523K$ and did not more observe the presence of the grows of SHG signal from minimal level which corresponded about $1{,}5 \cdot 10^{-6}$ of the efficiency η_g.

There is also the fundamental question about full reversibility of the process of the induction with the formation of the space-periodic micro-lattice $\chi^{(2)}$ after the heating of the sample up to such high temperatures. May be the heating of the sample with the induced inside space-periodic structure $\chi^{(2)}$ leads not only to the destruction of the micro-lattice $\chi^{(2)}$ but it results also in a local damage of the glass material or its chemical compound and further such damaged sample is not suitable for the full re-writing process? For answer to this important question we made the re-writing of the induction process after one day of the cooling of the used glass sample at room temperature. The obtained experimental results show that there is the full reversibility of the induction process in our phosphate samples. We observed the usual view of the writing of the induced micro-lattice $\chi^{(2)}$ as it was observed earlier in clear glass sample before heating. It means that the heating of the investigated phosphate glass samples higher than of $483K$ leads only to the reversible destruction of the micro-lattice $\chi^{(2)}$ inside the material and, accordingly, the decay of the induction process.

In conclusion we concentrate the attention on some basic obtained results in this paper. In contrast to the usual nonlinear crystals widely popular in lasers and optoelectronics, in which the nonlinear wave conversion of the radiation into the SHG signal is very sensitive to the outside influence of the heating, because of the rapid destruction of a phase synchronism in these materials, in our studied here phosphate glasses samples the presence of the induction process and the existence and increase of the micro-lattice $\chi^{(2)}$ in time with the presence of the nonlinear wave conversion process of the generation of the SHG signal are observed even at very high temperatures. It means that the all-optically induced modulation of the second-order nonlinearity $\chi^{(2)}$ in such glass materials can exist at high temperatures and the phase synchronism is slaved with the change of the temperature and the small amplitude of the micro-lattice $\chi^{(2)}$ can be increased by the outside heating influence. The heating of the sample up to critical temperature (about $483K$ in our case) leads only to the decay of the process of induction and the corresponding destruction of the induced micro-lattice $\chi^{(2)}$, it is not accompanied by the structural reorganization in the medium, and there are the possibilities of the full re-repeat of the induction process in sample. Moreover, in our experiments with heating of the samples we discovered the perfectly new possibility of the sufficiently increase of the efficiency of the induction of the micro-lattice $\chi^{(2)}$ in phosphate glass materials by the high temperature effect. In our opinion, the obtained results show the perspectives of the induction process in amorphous media, in particular in glasses, for the creation of new class of the nonlinear materials in future also including for sufficiently high thermal equipment in laser optics.

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