Volumetric anisotropic microlattices in compound materials

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ABSTRACT

The volumetric anisotropic microlattices, all-optically integrated into different compound materials, are interesting for investigations since they are potentially perspective for optical micro- and nanoelectronic technologies. In this paper the peculiarities of the properties of the volumetric anisotropic microlattices in different compound materials are considered and the comparisons of the nonlinear processes of light-beams transformations in dependence on the concentrations of different chemical elements in investigated bulk samples are discussed.

Keywords: all-optically integrated volumetric anisotropic microlattices, compound materials, nonlinear processes of light-beam transformations.

1. INTRODUCTION

The exposure of an isotropic medium to the powerful pulses of the mutual coherent multi frequency laser radiation may decrease the optical symmetry of the material and give rise to even orders in the expansion of the polarization of medium with respect to the light field. The occurrence of the anisotropy in samples is considered as a formation of a spatially-periodic electrical field owing to charges separation by an arising coherent photovoltaic current or as a local distribution of the charges with the formation of a long-lived static polarization. This creates the possibilities of the formation of various anisotropic microlattices all-optically integrated into different compound materials (on the base of glass and polymer consisted matrixes, fibers, waveguides and hybrides of the organic-inorganic films ets.[1-23]). The investigations of the linear or nonlinear processes of the light-beams transformations inside the all-optically integrated anisotropic microlattices attract an attention of the scientists due to the possibility in future for a creating of new broadband optoelectronic elements of micro- and nano-optics in visible and infra-red spectral diapazones which are potentially perspective for different areas of applications, in particular, for bio-photonics of molecular and atomic systems.

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In this paper the peculiarities of the properties of the volumetric anisotropic microlattices in different compound materials are considered and the comparisons of the nonlinear processes of light-beams transformations in dependence on the concentrations of different chemical elements in investigated bulk samples are discussed. The influence of the concentrations on the generation of light harmonics in optical media is analyzed. The attention is concentrated on the detailed comparison of results of the investigation of the writing times, relaxation processes and potential efficiencies of the nonlinear generation of light harmonics in different compound materials.

2. FORMATION OF THE VOLUMETRIC ANISOTROPIC MICROLATTICES

The equation for the nonlinear arising current $\boldsymbol{j}(t)$ in an isotropic media by the action of the powerful multi-frequency light field $\boldsymbol{E}(\boldsymbol{r},t)$ is given as the sum of the different order components of $\boldsymbol{j}(t) = \boldsymbol{j}^{(1)} + \boldsymbol{j}^{(2)} + \boldsymbol{j}^{(3)} + \boldsymbol{j}^{(4)} \ldots$, which are described as

$$\boldsymbol{j}^{(n)} = \int_0^\infty \int_0^\infty \ldots \int_0^\infty d\tau_1 d\tau_2 \ldots d\tau_n \hat{\sigma}^{(n)}(\tau_1, \tau_2, \ldots, \tau_n) \cdot \ldots \cdot \boldsymbol{E}(t-\tau_1)\boldsymbol{E}(t-\tau_2)\ldots\boldsymbol{E}(t-\tau_n), \tag{1}$$

and $\left(\hat{\sigma}^{(n)} \cdot \ldots \cdot \boldsymbol{E}(t-\tau_1)\boldsymbol{E}(t-\tau_2)\ldots\boldsymbol{E}(t-\tau_n)\right)_i \equiv \sum_{j_1, j_2, \ldots j_n} \sigma^{(n)}_{i, j_1, j_2, \ldots j_n} \boldsymbol{E}_{j_1}\boldsymbol{E}_{j_2}\ldots\boldsymbol{E}_{j_n}$.

Here $\sigma^{(n)}$ is a tensor with rang n+1 for the nonlinear conductivity by a reaction of isotropic medium. Note, for the considered isotropic center-symmetrical media (with symmetry of class type as $\infty\infty$m) the second-order tensor $\sigma^{(2)}$ is absent ($\sigma^{(2)} = 0$), but the components of the tensor $\sigma^{(3)}$ are non-zero values. It means that, in traditional optical poling process by means of the intersecting beams of the two-frequency inter-coherent laser light radiations with the fundamental and the doubled frequencies, there is the coherent photovoltaic current which is described as

$$\boldsymbol{j}_i = \sigma^{(3)}_{ijkl}(0; \omega, \omega, -2\omega)\boldsymbol{E}_{1j}\boldsymbol{E}_{1k}\boldsymbol{E}^*_{2l} \exp[i\boldsymbol{r}(2\boldsymbol{k}_1 - \boldsymbol{k}_2) + (2\psi_1(t) - \psi_2(t))] + k.c. \tag{2}$$

The separation of charges by the arising current results in the formation of a spatially periodic electrical field in a sample. The picture of the spatial distribution of field is very complicated because the envelope of the space-periodic current (2) has a shape which is formed by the intersection of the laser beams and the arising current has the components that are parallel and perpendicular to the planes of the formed space-periodic electrical field. In a simple case with using of the incident collinear fronts, it is obvious that the perpendicular current causes the accumulation of charges directly on the planes of the space-periodic electrical field in contrast to the parallel component of the current which causes the accumulation of charges along the border of the overlap region of the interacting light beams. But in some cases of the sufficiently small angles of the intersection of the interacting light beams on condition that $1/(|2\boldsymbol{k}_1 - \boldsymbol{k}_2|a) \ll 1$ (where a is the spatial size of the overlap region of the interacting light beams), the perpendicular current in a sample gives the main contribution into the formation of the spatial periodicity of the electrical field $\mathbf{E}=\boldsymbol{e}_q(j_\perp/\sigma)$, where $\boldsymbol{e}_q$ is the unit vector perpendicular to the lattice planes and σ is the linear electric conductivity of a medium. We suppose below, according to our estimations, that the arising electrical field **E** in our case is described by this expression and it causes the reversible change of the optical properties inside the medium with the formation of the volumetric anisotropic microlattices. Here we deal with the reversible variations in the optical properties of the medium which are caused by the spatial redistribution of the charge density and the arising of the optical polarization in medium. So, the arising optical polarization for the incident light beam $\boldsymbol{E}_\Omega(\boldsymbol{r},t)$ in a presence of the electrical field $\mathbf{E}(\boldsymbol{r}, t)$ is appeared as a sum of the corresponding order components $\boldsymbol{P}(t) = \boldsymbol{P}^{(1)} + \boldsymbol{P}^{(2)} + \ldots$, which are described as

$$\boldsymbol{P}^{(n)} = \int_0^\infty \int_0^\infty \ldots \int_0^\infty d\tau_1 d\tau_2 \ldots d\tau_n \, \hat{\chi}^{(n)}(\tau_1, \tau_2, \ldots, \tau_n) \cdot \ldots$$

$$\ldots \cdot \left(\boldsymbol{E}_\Omega(t-\tau_1) + \mathbf{E}(t-\tau_1)\right)\left(\boldsymbol{E}_\Omega(t-\tau_2) + \mathbf{E}(t-\tau_2)\right)\ldots\left(\boldsymbol{E}_\Omega(t-\tau_n) + \mathbf{E}(t-\tau_n)\right), \tag{3}$$

where $\left(\hat{\chi}^{(n)}\cdot\ldots\cdot\left(\boldsymbol{E}_{\Omega}(t-\tau_1)+\mathbf{E}(t-\tau_1)\right)\left(\boldsymbol{E}_{\Omega}(t-\tau_2)+\mathbf{E}(t-\tau_2)\right)\ldots\left(\boldsymbol{E}_{\Omega}(t-\tau_n)+\mathbf{E}(t-\tau_n)\right)\right)_i \equiv$

$$\equiv \sum_{j_1,j_2,\ldots,j_n}^{\infty}\chi^{n}_{i,j_1,j_2,\ldots,j_n}(\boldsymbol{E}_{\Omega}+\mathbf{E})_{j_1}(\boldsymbol{E}_{\Omega}+\mathbf{E})_{j_2}\ldots(\boldsymbol{E}_{\Omega}+\mathbf{E})_{j_n}.$$

The nonlinear processes of the light-beams transformations (as second harmonic generation and parametric down conversion of light) are take place as a reaction in second-order polarization of medium on incident light field $\mathbf{E}_{\Omega}(\boldsymbol{r},t)$. For nonlinear process of the second harmonic generation we have the integrated polarization $\boldsymbol{P}^{(2)}(2\Omega)$ inside an isotropic medium and the corresponding components of the arising anisotropic microlattices of second-order susceptibility $\chi^{(2)}$ described as

$$\boldsymbol{P}^{(2)}(2\Omega)=\chi^{(3)}(2\Omega;\Omega,\Omega,0)\cdot\cdot\mathbf{E}\boldsymbol{E}_{\Omega}\boldsymbol{E}_{\Omega},$$

$$\chi^{(2)}_{ijk}(2\Omega;\Omega,\Omega)=\chi^{(3)}(\mathbf{E}_i\delta_{jk}+2\mathbf{E}_k\delta_{ij}),\quad \chi^{(3)}=2\pi\chi^{(3)}_{iiii}(\Omega,\Omega,0). \qquad (4)$$

If the direction of the arising electrical field **E** coincides with the direction of the coordinate axis *x* then $\mathbf{E}=(E_x,0,0)$ and there are the equations: $\chi^{(2)}_{xxx}=3\chi^{(3)}E_x,\ \chi^{(2)}_{iix}=2\chi^{(3)}E_x,\ \chi^{(2)}_{xii}=\chi^{(3)}E_x,\ (i=y,z)$. The maximal nonlinear conversion of light on formed $\chi^{(2)}$ will take place for light radiation polarized along the direction of the induced electrical field **E**. But there are the other non-zero components of tensor $\chi^{(2)}$ which give the existence of the nonlinear conversions also for perpendicular polarized radiation, so that in the general case there are the corresponding polarization dependences for the arising nonlinear processes of light-beams transformations as second harmonic generation and parametric down conversion. So, the specific properties of the formation of the induced electrical field **E** in isotropic materials reveal the some variations of the properties for the processes of the nonlinear light-beams transformations and, in particular, for the investigated in this paper the nonlinear process of the generation of second harmonic which is observed in experiments in different volumetric mediums.

3. EXPERIMENTAL INVESTIGATIONS

In typical experiments upon all-optical poling the set-up on a base of a powerful pulsed laser is used for the creation of the photo-induced second-order tensor susceptibility $\chi^{(2)}$ in investigated samples. The beams of the basic $\lambda_1(\omega)$ and doubled $\lambda_2(2\omega)$ frequencies of a laser radiation are separated in space on two channels by use the optical elements and filters. In our case we used the powerful YAG:Nd^{3+} laser ($\lambda_1(\omega)$ = 1.064μm, $\lambda_2(2\omega)$ = 0.532μm), the pulse energy of the basic radiation was 45 mJ, the longitude of pulse was 10 ns, frequency repetition was 12,5 Hz, and conversion into doubled frequency was 10%. The separated beams of the radiations are focused at small angles inside the samples in spots from 250 down to 5 micro meters. The exposure by using this bi-chromatic light results in the formation of the induced space-periodic tensor microstructure $\chi^{(2)}$ inside the samples. After the writing of the volumetric anisotropic microlattices $\chi^{(2)}$ in samples up to saturation we overlapped the incident radiation of the doubled frequency and further in photomultiplier registered the radiation of the second harmonic from the basic laser radiation generated on the created susceptibility $\chi^{(2)}$. Growth of signals of peak power $P_{shg}(2\omega)$ of the generated second harmonic was observed on computer in real time. Note that other authors in works had similar typical experimental schemes for investigations of the process of harmonic generation in induced susceptibility $\chi^{(2)}$ but with some variations. So, in experiments can be estimated the efficiency of the nonlinear process of the generation of the second harmonic in photo-induced lattices $\chi^{(2)}$ as $\eta_g=P_{shg}(2\omega)/P(\omega)$ (where $P(\omega)$ is the power of the incident light with basic frequency).

So, here in Table 1 we present the obtained summary results of the investigations of the nonlinear light harmonic generation on the anisotropic microlattices of the second-order susceptibility $\chi^{(2)}$ photo-induced in various compound materials with different concentrations.

Table 1. Experimental results in various materials

Compound, mol.%	***a*, μm** ***l*, cm**	I_ω, 10^9 W·cm^{-2} $I_{2\omega}$, 10^9 W·cm^{-2}	**Laser pulse data**	t_w, **min**	t_d; t_ω; $t_{2\omega}$, **min**	η_g
$SiO_2$20,5%PbO16%Na_2O+ 6%$TiO_2$1,7%CeO_2	50 0,05	7 0,05	ML, QS*	10		10^{-4}
76%$Pb(PO_3)_2$5,5%CeO_2; {38%PbO;4%P_2O_5}	10 0,01	35 0,05	pulse	30		10^{-4}
BS7 {50%$SiO_2$45%PbO}	10 0,006	17 0,07	ML, QS		15 15 5	10^{-5}
$Ba(PO_3)_2Pb(PO_3)_2$ +1,5%CeO_2; {16%PbO}	10 0,01	35 0,05	pulse	≥40	≥10^4 ≥60 -	10^{-5}
SiO_2 (>85%)	10 0,01	30 2	ML, QS	10	10^4 >20 >20	10^{-5}
$SiO_2$20,5%PbO16%Na_2O+ (7÷22%)TiO_2	50 0,05	7 0,05	ML, QS*	10		10^{-5}÷10^{-6}
$SiO_2$20,5%PbO16%Na_2O	50 0,05	7 0,05	ML, QS*	10		10^{-6}
JS4 {66%$SiO_2$16%PbO}	50 0,05	7 0,05	ML, QS*	10	$2\cdot10^4$ - -	10^{-6}
$SiO_2$8%GeO_2	35 0,02	25 2	ML, QS	20÷30		10^{-6}
$Pb(PO_3)_2$-$R(PO_3)_2$; R=Ca, Sr, Ba; {5÷40%PbO}	10 0,01	25 0,042	pulse	60÷10	≥100 ≥60 -	10^{-8}÷10^{-6}
$SiO_2$3%GeO_2+0,5%P	90 0,1	50 0,8	ML, QS	>30		$2\cdot10^{-7}$
SK5{39%$SiO_2$40%BaO +15%$B_2O_3$5%Al_2O_3}	100 0,1	13 3	ML, QS	10	$3\cdot10^3$ - -	10^{-7}
BS4{SiO_2, PbO}	10 0,006	17 0,07	ML, QS			$8\cdot10^{-8}$
F8{50,2%$SiO_2$39,7%PbO+ 3,8%Na_2O5,6%K_2O}	100 0,1	13 $5\cdot10^{-3}$	ML, QS	10	60 - -	$5\cdot10^{-8}$
JZS19 {62%SiO_2+ 35%PbO}	50 0,05	7 0,05	ML, QS*	10		$4\cdot10^{-8}$
PS5 {69%$SiO_2$26%PbO}	50 0,05	7 0,05	ML, QS*	10		$3\cdot10^{-8}$
BS8 {63%$SiO_2$26%PbO}	50 0,05	7 0,05	ML, QS*	10		$3\cdot10^{-8}$
GeO_2(10÷40%)PbO	50 0,05	7 0,05	ML, QS*	10		10^{-8}
K8 {PM15, ≥70%SiO_2}	10 0,006	17 10^{-6}	ML, QS	10÷20	120 120 -	10^{-8}
≥60%$SiO_2$15%(K_2O+ Na_2O)6%$B_2O_5$5%BaO	250 0,7	4 0,4	pulse*	40	30 >10^4 >10^3	10^{-8}
HS24, S24 {crystal}	50 0,05	7 0,05	ML, QS*	10		10^{-8}
BS3{SiO_2, PbO}	10 0,006	17 0,07	ML, QS			$5\cdot10^{-9}$

JS18{SiO_2, PbO}	20 0,005	1 $2\cdot10^{-3}$	ML, QS	3	- 0,05 -	$5\cdot10^{-9}$
JS18 {SiO_2, PbO}	5 0,001	50 1,2	ML, QS			$2\cdot10^{-9}$
BS12{SiO_2, PbO}	10 0,006	17 0,07	ML, QS			$<10^{-9}$
$Al_2O_3 : Cr^{3+}$ {ruby}	6 0,001	35 0,7	ML, QS	0,08		$5\cdot10^{-10}$
GeO_2(10÷50%)PbO	5 0,001	30 0,05	ML, QS			$<10^{-11}$

a - diameter of the region of interaction of light radiations (diameter of waist in focus)

l - length of the region of interaction

I_ω - intensity of the basic frequency radiation in region of interaction of beams

$I_{2\omega}$ - intensity of the doubled frequency radiation in region of interaction of beams

t_w - times of record of a lattices $\chi^{(2)}$ in samples before saturation, t_d - times of a dark relaxation of lattices

t_ω - relaxation time in presence of radiation of basic frequency, $t_{2\omega}$ - relaxation time in presence of radiation of doubled frequency

η_g - efficiencies of light generation of the second harmonic on the photo-induced lattices $\chi^{(2)}$

pulse - pulsed YAG: Nd-laser, τ=30÷50ns, f=10Hz (τ - pulse length, f - repetition frequency)

ML,QS - pulsed YAG: Nd mode-locked and q-switched laser, τ=100÷600ps, f=76÷125MHz, t=200÷300ns, F=1÷6кHz (t - duration of bending around a set of impulses, F - frequency of repetition of a set of impulses)

ML,QS* - τ=30ps, f=76MHz, t=60ns, F=12,5Hz; pulse* - τ=10ns, f=12,5Hz

JS4, JZS19, BS7, BS8, PS5 - samples from a set of light filters (GOST-9411-91)

The experiments were performed in various samples, see Table 1, with different concentrations of the oxides of chemical elements in base matrix as SiO_2, PbO, Na_2O, K_2O, TiO_2, GeO_2 , BaO, B_2O_3, Al_2O_3, $Ba(PO_3)_2$, $Pb(PO_3)_2$ and also with presence of the small percents of different additions from active elements. The potential perpendicular sizes of the integrated volumetric anisotropic microlattices $\chi^{(2)}$ in investigated samples were from ~250 down to ~5 micro meters, the lengths were from ~7000 down to ~10 micro meters. The experimental results show that the observed efficiencies of the nonlinear light harmonic generation on the integrated microlattices of the second-order susceptibility $\chi^{(2)}$ in different compound materials have the values from 10^{-11} up to 10^{-4}. The sufficiently big values of the efficiencies about 10^{-4} were obtained in lead-natrium-silicate mediums with content of the oxide additions CeO_2 and P_2O_5 but they practically coincide with the values of the efficiencies 10^{-5} early obtained in pure silicate materials. Also in lead-phosphate mediums the efficiencies were about 10^{-8}÷10^{-4} but in germanium consisted materials the efficiencies have the smaller 10^{-9}÷10^{-6} values. So, now we have no yet the defined data about the full set of the potentially effective elements for creation of high magnitudes of induced microlattices of the second-order susceptibility $\chi^{(2}$ and, correspondingly, for the obtaining of the high effective nonlinear processes of the light-beams transformations with the generation of the nonlinear second harmonic in investigated mediums.

The writing times for the forming of the volumetric anisotropic microlattices $\chi^{(2)}$ up to maximum saturation in different investigated samples are from some minutes up to hour. The obtained lifetimes of the integrated microlattices $\chi^{(2)}$ in investigated materials are still sufficiently small from minutes up to some days and this is a basic problem which must be solved in following researches. The maximal obtained lifetime was about 10^4 minutes. On the base of the experimental results presented in this paper can be performed the estimated comparative analysis of the influence of the different concentrations of chemical elements in studied samples. The integrated volumetric anisotropic microlattices of the

second-order susceptibility $\chi^{(2)}$ may be used in future for creations of the broadband sources of the nonlinear harmonic generation for micro- and nano-optoelectronics. But for practical applications of such compound materials the additional investigations must be performed for obtaining of more high efficiencies and lifetimes.

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