Formation of miniature converters on all-optically inserted nonlinear modification

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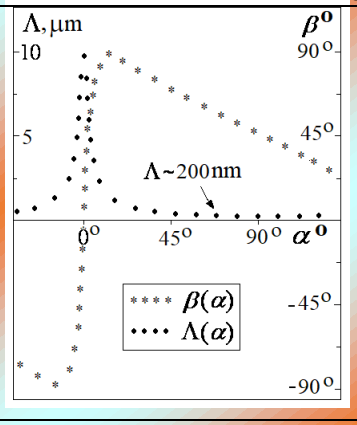
The features of all-optically inserted nonlinear modification in different isotopic media on a base of glass-matrixes for possible formation of the miniature optical frequency micro- and nano-converters are considered. The obtained experimental results of nonlinear frequency conversion of visible laser radiation on the all-optically inserted volumetric lattices of second-order susceptibility, which induce in isotropic samples by action of the pulsed powerful bi-chromatic inter-coherent radiation of YAG:Nd laser, are analyzed. Calculation results in Fig. 1 show that the distinct feature of volumetric optical poling is the existence of sharp dependencies of spacial periods $\Lambda(\alpha)$ and orientation angles $\beta(\alpha)$ for photo-integrated micro-structures of second-order susceptibility $\chi^{(2)}$ in region of small optical poling angles α of intersecting Gaussian laser beams. By changing optical poling angle α, the periods Λ of photo-integrated microstructures $\chi^{(2)}$, arising in medium, vary in broad range (almost by two orders of magnitude, Fig. 1): from $\Lambda_{max} = \lambda_1/(2\delta n) \sim 10^4$ nm to $\Lambda_{min} = \lambda_1/(4n_1) \sim 150$ nm (i.e. down to about tenth fractions of the light wavelength). It means that the very small micro- and nano-periodical nonlinear optical photo-integrated structures of the second-order susceptibility $\chi^{(2)}$ can be created in isotropic glass mediums by process of volumetric all-optical modification. By variation of dispersion in specially synthesized polymer-consisted materials or hybrid materials may be in future it will possible to receive a smaller periodicity within several nanometers. Orientation of the photo-integrated $\chi^{(2)}$ lattice, Fig. 1, also strongly depends on used angle α during optical poling process. When angle α varies within several degrees near zero, angle β takes values inside interval from $-\pi/2$ to $\pi/2$. This means that upon volumetric optical poling it is possible to produce the photo-integrated $\chi^{(2)}$ lattice with almost any (from parallel to perpendicular) orientation of its planes with respect to direction of photo-integrated field E inside isotropic mediums. The important result in Fig. 1 is existence of small all-optical poling angles α in which condition $\beta = \angle(\mathbf{k}_1,\Delta\mathbf{k}) \approx \pi/2$ is fulfilled so that the direction of lattice vector $\Delta\mathbf{k}$ is practically parallel to direction of the photo-integrated field E. The electric charge carriers in this case are separated mainly in direction which is perpendicular to planes of the field lattice E being produced in medium, and separation of electric charge carriers being accompanied by accumulation of effect near these planes of the field lattice E.



It is obvious that this type of separation of charge carriers should lead to formation of the volume periodic electrical field structure which is homogeneously distributed in the entire of all-optical modification region in sample. So, only this case from optical poling parameters is more perspective for a creation of homogeneous large field lattices E and corresponding high effective nonlinear-optical micro-periodic structures of susceptibility $\chi^{(2)}$.

Fig 1. The different space periods $\Lambda=2\pi/\Delta k$ and orientation angles $\beta=\angle(\mathbf{k}_1,\Delta\mathbf{k})$ of photo-integrated microstructures of second-order susceptibility $\chi^{(2)}$ on various all-optical poling angles α between incident laser beams (ω, $\mathbf{k}_1$) and (2ω, $\mathbf{k}_2$), and $\Delta\mathbf{k} = 2\mathbf{k}_1 - \mathbf{k}_2$. For calculations here used values: $\lambda_1=1.06\mu$m, $\delta n=n_2-n_1=5\cdot10^{-2}$, $n_{1,2}\approx1.5$ (n_1 and n_2 are refractive-indexes in pure oxide glass for ω and 2ω radiations of YAG:Nd laser).

The question about lifetime of photo-integrated $\chi^{(2)}$ lattice and it's stability to various kinds of the outside influences during process of photo-modification in different glass media is also very important for investigations since there is basic problem of manufacturing of long-lived and very steady-stable photo-integrated microstructures of second-order susceptibility $\chi^{(2)}$ for possible practical creation on these base of new miniature nonlinear wave converters perspective for laser systems and optoelectronics, and also for bio-photonics. Theoretical estimations show the existence of sufficiently big lifetimes (some days and years, Fig. 2) for amorphous isotropic materials in which the electric conductivities are smaller than the values of $10^{-16}\div10^{-20}$ $(\Omega\cdot m)^{-1}$. So, it gives the restrictions on the potential perspective media. But in really performed experiments the observed kinetic of nonlinear three-wave interactions on photo-integrated periodic structures of second-order susceptibility $\chi^{(2)}$ in investigated glass materials is more complicate than the theoretically considered exponential one. It was discovered in experiments that in some glass materials there is the existence of the non-stationary regimes of processes of three-wave interactions which are accompanied by the increase in time (or decrease) of efficiencies of nonlinear frequency doubling of light laser pulses on photo-integrated lattices $\chi^{(2)}$ and it may be connected with the different microscopic processes which are aroused in glasses during all-optical modification.

Our experiments shown also that the photo-integrated anisotropic and nonlinear structures of $\chi^{(2)}$ in some investigated glasses can be long-lived (some days, Fig. 3) and stable to sufficiently powerful laser illuminations. But electrical conductivity in these samples is not so small. In means that may be there is the sufficiently influence of some microscopic processes on formation and stability of photo-integrated $\chi^{(2)}$ structures arising inside isotropic glass samples during volumetric optical poling.

The distribution of maximal values of observed efficiencies η for nonlinear frequency doubling on photo-integrated microstructure of second-order susceptibility $\chi^{(2)}$ in various compound glass samples is presented in Fig. 3. The chemical compound in investigated samples plays the important role as well as the some active doping additives that can to increase sufficiently the amplitude of photo-integrated $\chi^{(2)}$ microstructure in glasses and, accordingly, to increase the conversion efficiency η for three-wave interaction up to some order of magnitude. According to experimental results, Fig. 3, the more perspective materials are the multi-compound meta-phosphate glasses doped with some rare-earth elements and multi-compound oxide glasses with preliminarily suppressed UV- and γ- luminescence. The important result Fig. 3 that longest lifetimes coincide with biggest efficiencies. Now the greatest obtained value of conversion efficiency η is about $\sim5\cdot10^{-3}$ but it is smaller than in nonlinear crystals and therefore the additional investigations are necessary.

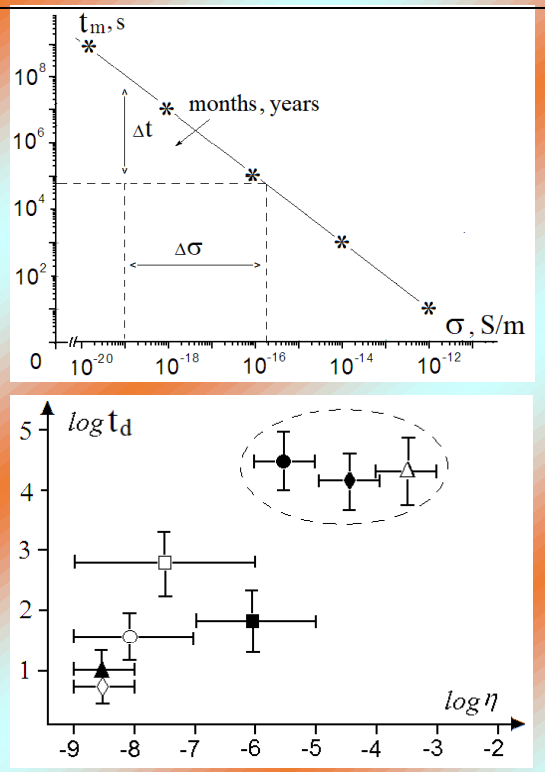


Fig 2. Theoretical estimations for lifetimes t_m of photo-integrated structures of second-order susceptibility $\chi^{(2)}$ in isotropic media with dependent electrical conductivities σ.

Fig 3. The obtained intervals of experimental values for efficiencies $log\eta$ of nonlinear three-wave interaction (for process of nonlinear frequency doubling) on photo-integrated micro-structures of second-order susceptibility $\chi^{(2)}$ and also for observed dark times $log t_d$ of the relaxation of the photo-integrated structures $\chi^{(2)}$ in different compound glass materials: O - pure optical oxide glasses with consist of more than 60% mol. SiO_2; Δ - multi-compound oxide glasses with suppressed UV- and γ-luminescence; □ - silicate glasses doped with germanium (up to 20% mol.); ◊ - glasses on the base of La-Ga as pure as doped with the rare-earth elements; ♦ - pure phosphate glasses; • - multi-compound meta-phosphate glasses doped with the rare-earth elements; ■ - oxide glasses with content of the fourth and the fifth groups of chemical elements (Pb, Ti, Nb, Sb); ▲ - glass matrix with content of crystallite of quartz or Li-Nb and Li-F.

The investigated photo-integrated structures may be useful in future for creation of specific converters for micro-nano-scale optoelectronics. Authors are grateful to groups of Nikolaev Institute of Inorganic Chemistry and Vavilov State Optical Institute for offered some synthesized samples for investigations. The work was carried out as part of Russian State Task FWGW-2025-0019.

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