IR laser operation by chalcogenide glass features

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IR induced operation by mechanical parameters in complex chalcogenide glasses with a general formulae $\text{Sb}_2\text{Se}_{3-x}\text{Te}_x\text{-BaCl}_2\text{-PbCl}_2$ (with x = 0.2, 0.8, 1.3) was found under the influence of a pulsed 190 ns CO₂ laser and with light power densities varying within 0.5–1.5 GW/cm² with a wavelength 10.6 µm. The monitoring was performed by acoustic sound velocities. It was established that varying x leads to variations in mechanical properties, which is monitored by acoustic sound velocities. This one is a consequence of Ir laser induced variations of electron and phonon polarizations. Simultaneously changes in pump–probe delaying times in a nanosecond time range were explored. The studies allow to determine the maximally achieved acoustic velocity values. A significant role of an electron–phonon subsystem in the observed IR-induced mechanical treatment is demonstrated. Of a particular interest are studies of sound velocity decay properties after switching off the IR-induced laser beam.

Keywords: optical properties, photoinduced changes.

1. Introduction

The possibility to operate by the properties of chalcogenide glasses presents an enhanced interest both from technological as well as fundamental points of views. In the works [1–4] it was shown that chalcogenide glasses are most promising for IR laser operation due to the enhanced light-induced polarizabilities which are related to the phonon subsystem defining the mechanical features. The crucial role here play the IR-induced changes of optical constants [5], which are extremely sensitive to IR-induced electron–phonon interactions. It should be emphasized that there also exist some other slighter effects; however, normally, their contribution may be neglected as being small [6, 7]. The chalcogenide glasses possess very effective parameters of interactions with light [8, 9]. Most of the studies concerning the chalcogenide glasses were connected with optically operated parameters like transparency, refractive indices or second harmonic generation output. One can expect that very promising may be the IR-induced acoustical effect (IAO) because it includes both the contribution of an electron as well as phonon subsystem. The similar IAO effect was also observed in some other chalcogenides [10], including formation of surface relief [11]. However, they are less applicable due to a low laser threshold damage.

In the present work we will study the optical operation by mechanical properties using the IR-induced nanosecond laser treatment. The main advantage of such treatment consists in a possibility to use the same materials many times without the necessity to synthesize a new one.

For this reason we have explored $\text{Sb}_2\text{Se}_{3-x}\text{Te}_x$ -BaCl₂-PbCl₂ (with x = 0.2, 0.8, 1.3) glasses because, following Refs. [12, 13], such a type of glass possesses a relatively higher degree of electron-phonon anharmonicities, which is a necessary condition for the IR-induced operation. Their synthesis procedure is similar to that described in Refs. [12, 13].

It is necessary to emphasize that IAO efficiency is generally closely related to mechanical plasticity and melting temperature of the chalcogenide glasses [14]. So IAO may be of special interest for such a kind of studies. Moreover, mechanical stresses can cause anisotropic and inhomogeneous space distribution of the refractive indices. In the present work, the principal possibility to operate by IR laser light by simultaneous applications of two fields: dc-electric and optical IR laser one will be shown.

2. Experimental details

The studied samples were synthesized from the melt solution of particular oxide compounds which were taken in the appropriate stoichiometric ratio. The samples were completely amorphous and their space homogeneity was controlled by the optical polarimetry and the X-ray diffraction with Cu K α . Their space non-homogeneity did not exceed 4%. The samples were in a form of parallelopipeds with the sizes of $(0.5 \times 0.5 \times 0.9)$ cm.

The sound velocities determining mechanical features were evaluated by two principal methods: by the echo method and by the transmission method. In the first case, the generator ultrasound (US) and the sound detector were used simultaneously for the sound generation and detection. For the thin films, more suitable is the transmission method, which allows to use the delaying line.

In the present work we have applied the first echo method. Sound velocity was detected using the measurement card connected with an integrated pulse generator and two piezoelectric sensors with frequencies equal to about 4 MHz. Using this method, we have measured propagating sound velocities in materials and corresponding times of their propagation from the generator to the detector for the specimen with known thicknesses. The accuracy of the sound velocity measurements was equal to about 2 m/s.

The time stability of photoinducing laser pulses was deviated not more than 3-6% and the stability of the space distribution of the Gaussian laser beam was less than 0.1%. The average photoinducing beam's diameter of the photoinducing Gaussian-like beam

varied from 7 up to 15 mm to change successively the power density up to 1 GW/cm^2 . The control of the thermostability was performed by the thermocoupling method with precision up to 1 K.

For the performance of the photoinduced treatment we have done the laser treatment with the simultaneous measurements of the sound velocities. Such short-time kinetics allowed to avoid the samples overheating. The pumping laser beam was split into two beams with the same intensities which have been incident on the sample's surfaces at the angle 45°. A special proustite delaying line allowed to operate by pump–probe delaying times. The light absorption coefficients were evaluated taking into account samples reflection and transparencies.

Sound velocities versus the time-dependent propagation were measured at different delaying times t of the two photoinducing beams. The averaging statistics over the sample surface was performed to avoid space non-uniformity in the sample distribution within the sample surface.

3. Results and discussion

In Figure 1 the dependences of the sound velocities versus pumping pump-probe delaying time at different x contents are presented. It was established that the maximum of the IR induced velocity is achieved at 39 ns for samples with x = 0.8. It is necessary to add that at further increase in x, the maximum of sound velocity is shifted towards higher delaying times (up to 40 ns) and its value is less. At the same time, at lower x, the maximum is shifted towards less delaying times and the maximum value is at least 20% less than for the x = 0.8.

The second interesting fact consists in an existence of IAO maximum at an intermediate *x* value.

One can see that there exists a significant difference between specimens possessing different *x*. Moreover, the observed time-delaying IAO shift demonstrates some asym-



Fig. 1. Dependences of sound velocities versus the pumping pump-probe delaying time for the studied chalcogenides with different x.

metric shape which is typical of the decay of photoinduced carriers with relatively low electron–phonon anharmonicities [15].

In Figure 2 there are presented some dependences of the IAO versus pumping IR laser densities. One can see that the maximum of the IAO achieves its maximal value at power densities equal to about $0.8-0.95 \text{ GW/cm}^2$. It is crucial that in the power densities higher than 0.6 GW/cm^2 there occur substantial anharmonic phonons stimulated by the external light.

The occurrence of some nonlinearities in the pump power densities (see Fig. 2) may be also a consequence of the occurrence of some metastable nucleation states similar to partial unstable crystallization or nanocrystallization [16, 17]. The latter factor usually favours changes in acoustomechancial features.



Fig. 2. Relative changes in acoustic velocity versus the laser power density for the chalcogenides of different content.



Fig. 3. IR-induced AIO decays for the samples with different content.

After switching off the pumping IR power densities, the observed acoustically induced optics (AIO) effect completely disappears and the mechanical parameters return to their initial states.

In Figure 3 there is presented a time decay of the AIO after switching off the IR laser. One can see that after 60 μ s the photoinduced mechanical parameters for all the *x* contents the glass returned to the initial state. This decay is typical of chalcogenide glasses during their treatment below the damage threshold [15].

The presented results confirm a principal possibility to perform an effective operation by physical and acoustic features of the chalcogenide glasses using the external two-beam pumping treatment. A principal role play here the electron–phonon interactions depending on the glass content. After 60 μ s the photoinduced mechanical parameters for all the *x* contents of the glass returned to the initial state.

The discovered effect is very promising for production of laser operated chalcogenide glasses.

Despite the fact that the enhancement of temperature did not exceed 1 K, our evaluations show that the space distribution of even such small changes may be crucial for the observed effects. It is a consequence of a fact that for the chalcogenide glasses the anharmonic phonons are very sensitive to the increased temperature, contrary to the borate glasses [18]. However, it will be a subject of a separate work in future.

4. Conclusions

We have discovered a principal possibility to operate by mechanical properties of chalcogenide glasses under the influence of external two beams of 190 ns CO₂ with light power densities varying within 0.5–1.5 GW/cm² for Sb₂Se_{3-x}Te_x–BaCl₂–PbCl₂ (with x = 0.2, 0.8, 1.3) chalcogenide glasses. It was established that the maximum of the IR induced velocity is achieved at 39 ns for samples with x = 0.8. It is necessary to add that at further increase of x, the maximum of sound velocity is shifted towards higher delaying times (up to 40 ns) and its value is lower. At the same time at lower x the maximum is shifted towards less delaying times and the maximum value is at least 20% less than for the x = 0.8. One can see that the maximum of the IAO achieves its maximal value at power densities equal to about 0.8–0.95 GW/cm². It is crucial that at the power densities higher than 0.6 GW/cm² there occur substantial anharmonic phonons stimulated by the external light.

References

SHAABAN E.R., Interpretation of the change in optical constatns of different compositions of Ge-Se-In in terms of cohesive energy, Journal of Physics and Chemistry of Solids 73(9), 2012, pp. 1131–1135.

^[2] FLOREA C., BUSSE L., SANGHERA J., SHAW B., AGGARWAL I., A simple phenomenological study of photodarkening in As₂S₃ glasses, Optical Materials 34(8), 2012, pp. 1389–1393.

- [3] CHMIEL M., PIASECKI M., MYRONCHUK G., LAKSHMINARAYANA G., RESHAK A.H., PARASYUK O.G., KOGUT YU., KITYK I.V., Optical and photoconductivity spectra of novel Ag₂In₂SiS₆ and Ag₂In₂GeS₆ chalcogenide crystals, Spectrochimica Acta Part A: Molecular and Biomolecular Spectroscopy 91, 2012, pp. 48–50.
- [4] XUE B., NAZABAL V., PIASECKI M., CALVEZ L., WOJCIECHOWSKI A., RAKUS P., CZAJA P., KITYK I.V., Photo-induced effects in GeS₂ glass and glass-ceramics stimulated by green and IR lasers, Materials Letters 73, 2012, pp. 14–16.
- [5] SHPOTYUK O.I., KASPERCZYK J., KITYK I.V., Mechanism of reversible photoinduced optical effects in amorphous As₂S₃, Journal of Non-Crystalline Solids 215(2–3), 1997, pp. 218–225.
- [6] MENDES A.C., MAIA L.J.Q., MESSADDEQ S.H., MESSADDEQ Y., RIBEIRO S.J.L., SIU LI M., *Photoexpansion and photobleaching effects in oxysulfide thin films of the* $GeS_2+Ga_2O_3$ system, Physica B: Condensed Matter **406**(23), 2011, pp. 4381–4386.
- [7] GRUHN W., Infrared second-order nonlinear optical effects in Sb₂Te₃-SrBr₂-PbCl₂, Optica Applicata 35(3), 2005, pp. 329–337.
- [8] TINTU R., NAMPOORI V.P.N., RADHAKRISHNAN P., SHEENU THOMAS, Nanocomposite thin films of Ga₅Sb₁₀Ge₂₅Se₆₀ chalcogenide glass for optical limiting applications, Optical Materials 33(8), 2011, pp. 1221–1225.
- [9] LYUBIN V., KLEBANOV M., BRUNER A., SHITRIT N., SFEZ B., Transient photodarkening and photobleaching in glassy GeSe₂ films, Optical Materials 33(6), 2011, pp. 949–952.
- [10] DAVYDYUK G.YE., MYRONCHUK G.L., LAKSHMINARAYANA G., YAKYMCHUK O.V., RESHAK A.H., WOJCIECHOWSKI A., RAKUS P., ALZAYED N., CHMIEL M., KITYK I.V., PARASYUK O.V., *IR-induced features of AgGaGeS₄ crystalline semiconductors*, Journal of Physics and Chemistry of Solids **73**(3), 2012, pp. 439–443.
- [11] GERTNERS U., TETERIS J., Surface relief formation in amorphous chalcogenide thin films during holographic recording, Optical Materials 32(8), 2010, pp. 807–810.
- [12] KITYK I.V., *IR-stimulated second harmonic generation in Sb₂Te₂Se–BaF₂–PbCl₂ glasses*, Journal of Modern Optics **51**(8), 2004, pp. 1179–1189.
- [13] KITYK I.V., IR-induced second harmonic generation in Sb₂Te₃-BaF₂-PbCl₂ glasses, Journal Physical Chemistry B 107(37), 2003, pp. 10083–10087.
- [14] BORMASHENKO E., POGREB R., SUTOVSKY S., LUSTERNIK V., VORONEL A., Mechanical and thermodynamics properteis of infrared transparent low melting chalcogenide glass, Infrared Physics and Technology 43(6), 2002, pp. 397–399.
- [15] FEDORCHUK A.O., GORGUT G.P., PARASYUK O.V., LAKSHMINARAYANA G., KITYK I.V., PIASECKI M., IR operated novel Ag_{0.98}Cu_{0.02}GaGe₃Se₈ single crystals, Journal of Physics and Chemistry of Solids 72(11), 2011, pp. 1354–1357.
- [16] MAJCHROWSKI A., KITYK I.V., MANDOWSKA E., MANDOWSKI A., EBOTHE J., LUKASIEWICZ T., Several features of emission spectra of Pr^{3+} ions incorporated into $Li_2B_4O_7$ glasses matrices, Journal Applied Physics **100**(5), 2006, article 053101.
- [17] KITYK I.V., Nonlinear optical phenomena in the large-sized nanocrystallites, Journal of Non-Crystalline Solids 292(1–3), 2001, pp. 184–201.
- [18] KITYK I.V., MAJCHROWSKI A., Second-order non-linear optical effects in BiB₃O₆ glass fibers, Optical Materials 25(1), 2004, pp. 33–37.

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