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NON-LINEAR ABSORBTION OF As_2S_3 GLASSES AT THE CRITICAL LIGHT INTENSITIES AND FOR TECHNOLOGICAL MODIFICATIONS

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The effect of the linear and non-linear imaginary parts of polarizability or connected with them linear α and two-photon β absorption on transmission and optical damage threshold obtained at the different technology conditions As_2S_3 glasses were investigated. It was established, that with increasing of the melting temperature T_i and velocity of cooling V_i values the forbidden gap width of glasses increase at the decreasing of their density, refractive indices (from 2.71 to 2.48) and two-photon absorption coefficient (from 0.37 to 0,15 cm/MW), that is accompanied by the appropriate increasing of optical damage threshold I_d..

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1. Introduction

Chalcogenide As_2S_3 glasses are promising materials for production of different optical elements for optoelectronics and laser techniques. This is due to the properties of the glasses that are transparent in visible and IR region of spectrum and possess different values of refractive indices and relatively low optical and acousto-optical losses. However, the relatively low damage threshold is the main restriction for their wide applications. As it is known, the damage threshold of the material is mostly defined by the linear and non-linear losses of laser radiation determined by topologies of chalcogenide glass structure, that exhibit medium range order in atomic arrangement [1-5]. The analysis of the effect of structure-topological transformations on refraction coefficient, constants of linear and two-photon absorption, dynamic constants and optical damages of As_2S_3 chalcogenide glasses are presented.

2. Experimental

The rectangular polished to the high optical quality glassy platelets with dimensions of 10x15x (0.2-7) mm^3 were used in experiments. The samples have been fabricated from high grade purity of As and S components by synthesis in evacuated ampoule at different temperatures T_i of the melting treatment and quenching rates V_i. Temperature T_1 = 870 K - is the minimal temperature, at which the interaction As with S occurs in real time scales (150 hours), T_2 =1120 K – is the temperature, at which glassy As_2S_3 is usually synthesised and temperature T_3=1370 K – is the

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maximal temperature, at which of As_2S_3 molecule do not yet dissociates on elementary components. The rates $V_1=10^{-2}$ K/s and $V_{13}=10^2$ K/s are restricted by possible experimental conditions, while $V_2=1$ K/s is optimal for synthesis of glassy As_2S_3.

The electronic microscope EMB-100B and standard criteria for the gradation of micro-dispersion and micro-heterogeneity levels, which were determined by the ratio of quantity of precisely contoured boundaries to quantity of pseudo-granules in the region of 0.1 μm size was used for characterization of the microstructure of glasses.

The Q-switched ruby laser was used as a light source. It emitted the pulses with duration of 25 ns and the energy of 0.4 J. The intensities of incident I_0 and transmitted I through sample light beams have been recorded and measured by photodiodes and oscilloscope.

3. Results and discussion

At the high light intensities the optical properties of the medium are determined by dependent on the electrical field intensity **E** of the electromagnetic wave [6, 7]

$$P_i = \chi_{ij}E_j + \chi_{ijk}E_jE_k + \chi_{ijkl}E_jE_kE_l + \ldots \quad . \tag{1}$$

The imaginary parts of electronic susceptibility tensor χ_{ij} determined the values of the linear losses α, while the imaginary parts of non-linear electronic susceptibilities χ_{ijk} and χ_{ijkll} determined the values of non-linear losses β as a two-step and two-photon absorptions.

The investigation of the linear and non-linear light losses were made by experimental measuring of intensity dependencies of sample transmission [8-9]. They have sublinear shape (Fig. 1, curve 1) and can be approximated by equation

$$I = I_0 \frac{(1-R_0)^2 \exp(-\alpha d)}{1+\beta I_0 \alpha^{-1}(1-R_0)[1-\exp(-\alpha d)]}, \tag{2}$$

where R_0 is the reflection coefficient, d is the sample thickness, α and β are the coefficient of linear losses and two-photon absorption respectively.

The α and β values were calculated from the almost linear reverse transmission I_0/I on incident intensity I_0 (Fig. 1, curve 2). This linearity indicated on the domination of two-photon nature of absorption at the high critical light intensities and may by approximated by equation

$$I_0 / I = \frac{1+\beta I_0 \alpha^{-1}(1-R_0)[1-\exp(-\alpha d)]}{(1-R_0)^2 \exp(-\alpha d)}. \tag{3}$$

The optical damage threshold I_d was determined as the minimal power density that gives a rise to bright flash and, as a result, to sharp decreasing of transmitted beam intensity.

The obtained data on optical parameters of As_2S_3 glasses are presented on the Fig. 2 and are tabulated in the Table 1.

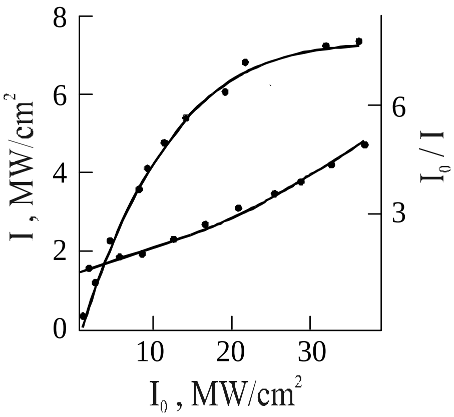

Fig. 1. Intensity dependences of transmitted light intensity and reverse sample transmission.

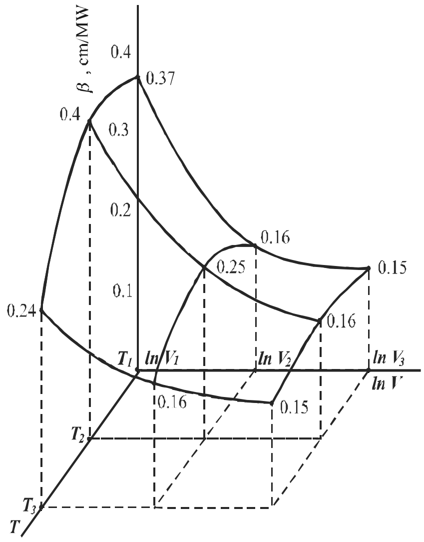

Fig. 2. Dependeces of two-photon absorption coefficient on technology parameters T_iV_i.

Table 1. Optical parameters of As_2S_3 glasses fabricated at different values of T_iV_i.

Meltig tempera-ture T , K	Velosity of quenching *V*, K/c	Den-sity, г/см^3	Forbid-den gap Eg, eV	Refrac-tion index At λ = 633 нм	Linear losses coefficien t α, см$^{-1}$	Two-photon ansorption coefficient β, см/МВт	Optical damage, I_d МВт/ /см^2
T_1=870	V1=10^{-2}	3.201	2.12	2.712	2.16	0.37	30
	V2=1.5	3.195	2.15	2.69	1.17	0.16	45
	V3=150	3.192	2.21	2.664	2.22	0.15	55
T_2=1120	10^{-2}	3.193	2.18	2.705	1.96	0.4	30
	1.5	3.190	2.22	2.65	2.53	0.25	36-40
	150	3.186	2.26	2.602	1.355	0.18	30-40
T_3=1370	10^{-2}	3.192	2.22	2.602	1.90	0.24	30
	1.5	3.184	2.30	2.59	1.855	0.17	36-40
	150	3.176	2.38	2.580	1.73	0.15	50-60

The basic set of the possible structural groups of As-S were realized by the selection of different T_iV_i. They can be divided into two basic types: A type - homogeneous, where main motive is bipyramidal $AsS_{3/2}$ structural units, inherent to glasses with micro-dispersion structure of different degree of connectivity and continuous structural network; and B type which is formed by heteroatomic pseudo-molecular units $As_2S_{4/2}$, As_3S_3, As_2S_5 and the homogeneous aggregations of sulphur S_8 [10, 11]. The same results are received also from the comparative analysis of Raman spectra of arsenic trisulphide glasses in the region of valence vibrations. The realization of the first type of the structural units take place at the minimal values of T_iV_i, while the second one - at the maximal T_iV_i. The band gap width increasing under the decreasing of the density, refraction index from 2.71 up to 2.48 and two-photon absorption coefficient from 0.37 up to 0.15 cm/MW, at respective increasing of the value of the threshold optical damage I_d are observed [12].

On the basis of the comparative analysis of Raman spectra of trisulphide glasses in the region of the valence vibrations was shown, that with the increase of the melting temperature and the cooling rate the concentration of structural units $As_2S_{4/2}$, As $_{3/3}$, S_8, S_n in a matrix of a-As_2S_3 structure is increased. At such increase the expansion of the matrix of a-As_2S_3 structure, the density and speed of ultrasound in glass and, respectively, dynamic strength of a glass expressed by elastic module, decrease.

As it is known, the sharp thermal heating of the illuminated region of the semiconductors plays a leading role in the optical damage by the laser pulse emission of nanosecond duration [1].

The process is connected with excitation of free carriers under the linear and two-photon absorptions and subsequent thermalisation and non-radiation recombination of them. The increase of temperature ΔT are determined by the thermodynamic equation

$$\Delta T = E/mC, \tag{4}$$

where the E - is the light energy absorpted in illuminated region, m – is the mass of this region, C – is the thermal capacity.

For light intensity decreasing i.e. at the $\Delta I << I_0$ e.g. at the $\alpha d<<1$ and $=\beta I_0 d<<1$, the ΔT values could be appreciated from the equations:

$$\Delta T_1 = \alpha I_0 \tau / \rho C \tag{5}$$

$$\Delta T_2 = \beta I_0^2 \tau / \rho C, \tag{6}$$

where τ - is the duration of the laser pulse, ρ - is the density of substance. As the damage process starts at the achieving the critical values of temperature T_{cr} and spatial gradient ΔT_{cr}, the optical damage threshold is related inversely to the values of linear α and two-photon β absorption coefficients.

4. Conclusions

There was investigated the effect of linear and non-linear imaginary parts of the polarizability or connected with it linear α and two-photon β absorption on transmission and optical damage threshold of As_2S_3 glasses obtained in different technological conditions.

It was established, that with the raise of the melting temperature, T_i, and velocity of cooling, V_i, the forbidden gap width of glasses increase, while a decrease is observed for density, refractive indices (from 2.71 to 2.48) and two-photon absorption coefficient (from 0.37 to 0.15 cm/MW). These modifications are accompanied by the appropriate increasing of the optical damage threshold I_d.

The theoretical estimations of the I_d of As_2S_3 in adiabatic approximation show, that the observable increasing of the I_d values are due to the decreasing of the linear and non-linear losses coefficients for the corresponding increasing of the forbidden gap value.

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