Structural disorder and optical properties of electon-irradiated $As_2S_3(Se_3)$ chalcogenide glasses

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Abstract.

The influence of electron irradiation on a form of the intrinsic absorption edge at various doses was studied. The influence of the processes of structural disorder on the energy position and the absorption edge width was investigated. The optical data are interpreted in a Gody model.

Key words: electron irradiation, absorption edge, chalcogenide glassy semiconductors, the Urbach rule, structural and dynamic disorder.

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Introduction

Glassy chalcogenides of arsenium are characterized by high transparency in a near and middle infrared and belong to a class of materials which are used as active or passive elements in optical engineering [1-3]. Experimental studies of the influence of a radiation load (gamma radiation, X-radiation or electron radiation) enable to determine the character and change of physical properties of these materials, boundary doses of radiation, to learn the nature and mechanisms of radiational defect – formation, reveal conditions of renewing the initial parameters.

The objective of the work is to study the influence of the electron irradiation on a structural and dynamic disorder of $As_2S_3(Se_3)$ glasses in dependence of the dose.

Experiment

Volume ingots As_2S_3 and As_2Se_3 were obtained by the method of alloying the initial elementary components of high purity with subsequent homogenization during continuos isothermal annealing. $As_2S_3(Se_3)$ compounds were checked to stechiometry by X-ray analysis and X-ray photoelectronic spectroscopy methods.

The samples radiation was carried out in usual conditions at room temperature with electrons of an average energy ~ 0.6 meV; the radiation source was SrY-16 of 12.2 Curi activity and the flux density 10^7 electrons/($sm^2 \times s$). The absorbed dose was calculated starting with the expositional dose and the radiation time in a definite point of the source channel. The absorbed dose value was chosen according to well-known results of dose dependences of radiationally-induced effects in glassy chalcogenide glasses of As_2S_3 type [4-6]. The study of the absorption edge was carried out on the device and according to a method described in [7]. For the investigations the samples of different thickness (d=20-400 mkm) were used, and the deviation in determination of the absorption coefficient was $\Delta\alpha/\alpha < 10\%$.

Experimental results and discussion

A reversal or non-reversal nature of the changes in optical parameters of chalcogenide glasses is

usually reffered to inner structurally dependent processes including both local changes in the structure of the short-range order and transformation of the structure in an intermediate (medium) order [8, 9]. It is well known [10-12] that such parameters of the intrinsic absorption edge as the optical pseudogap width E_g* and a characteristic energy of the Urbach (exponential) absorption tail W express the influence of various disorder types on the absorption processes in a longe-wave edge region:

$$E_g^*(T,X) = E_g^*(0,0) - D(\langle W^2\rangle_T + \langle W^2\rangle_S), \quad (1)$$

$$W(T,X) = K_0(\langle W^2\rangle_T + \langle W^2\rangle_S), \quad (2)$$

where D and K_0- are the constants, $E_g^*(0,0)$ is the forbidden band width of an "ideal glass" in the absence of disorder (at T=0). In the general case $\langle W^2\rangle_T$ is a mean square deviation from the electronic potential of glass due to a dynamic (temperature) disorder. A mean square deviation $\langle W^2\rangle_S$ characterizing the structural disorder consists both of intrinsic and induced external sources (radiation, conditions of preparation etc.).

For chalcogenide glasses of the $As_2S_3(Se_3)$ type the exponential part can be described by the expression:

$$\alpha(E,T) = \alpha_0 \exp\left[(E - E_g^*)/W(X,T)\right], \quad (3)$$

where E_g^* is the optical pseudogap, which is determined as an energetic position of the absorption edge at fixed values of the absorption coefficient ($\alpha^* = 10^3 cm^{-1}$) and temperature T; W(T,X) is a characteristic energy or a logarythmic slope $W = \partial\hbar\omega/\partial \ell n\alpha$ is changing in these glasses within the limits of 40-85 meV in dependence on the terms of growth etc. The X parameter in different materials has different values and is determined by the character of disorder. Gody et al. [12] were the first to determine experimentally the proportionality $E_g^*(T,X)$ and $W(T,X)$ for a- Si:H, which was realized in the whole range of W values studied till now. By using the Tauc's concept of "freezed" phonons [13] he spread the idea of the equivalency of the effect of a structural Ws and thermal Wt of disorder onto the band width E_g^* and got a linear relation between E_g^* and W:

$$E_g^*(T,X) = E_g^*(0,0) + D\cdot\langle W^2\rangle_0 - \frac{D}{K}W(T,X) \quad (4)$$

where D is a deformation potential, $\langle W^2\rangle_0$ is a mean-square shift due to zero oscillations. According to this model the optical pseudogap E_g^*(T,X) is determined by the degree of disordering of a glass lattice which is described by $\langle W^2\rangle_S$ parameter, i.e. by changing the sources of different nature it is possible to influence the E_g^* value indirectly.

Let us analyse our experimental results in the frameworks of this model. In Fig.1 the correlation between E_g^* and W for glassy $As_2S_3(Se_3)$ is dependence of the nature of disorder due to various external factors. This correlation shows that the optical pseudogap E_g^* and, this being more important, the slope of an exponential portion of the edge are changing in dependence of the disorder degree. A linear relation between E_g^* and W for chalcogenide glasses $As_2S_3(Se_3)$ is fulfilled practically in the whole range of the values of W energies which was studied up to this time. Thus it can be stated that in this case for these materials the contribution of the structural ("intrinsic" and induced) and the thermal contributions into a change of disorder potential is adequate, and the change of the slope probably reflects the change of the distribution of the states in the tails of zones.

By means of fitting (4) to experimental results in Fig.1 we obtained dose dependences

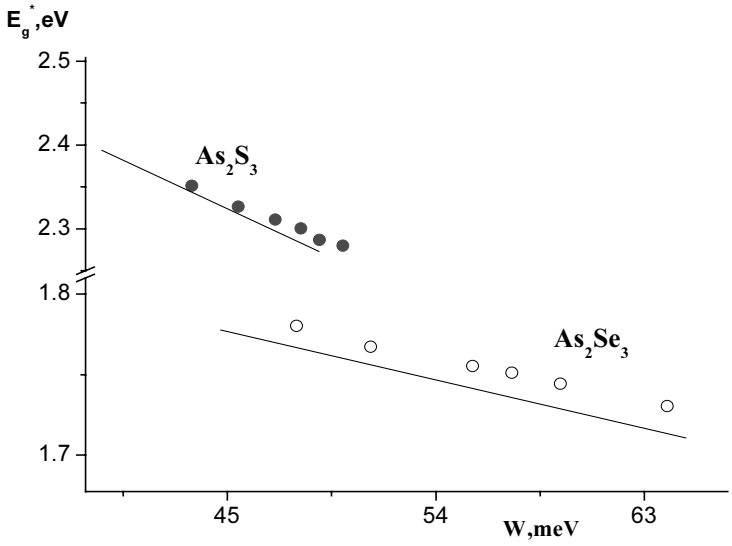

Fig.1. The dependence of optical pseudogap E_g^* on the slope of the exponencial Urbach tail W. Blank and black circules correspond to different phases and conditions of preparation [16].

of structural and temperature contributions into W(T,X) at temperatures 80 and 300K for $As_2S_3(Se_3)$ - Table1.

The increase of the radiation doses leads to an increase a structural contribution into W(T,X) both at nitrogen and room temperature. The change of the value of a thermal component is not large that testifying to a slight change in $\sigma(T) = KT/W(T)$ and, correspondently, weak dependence of parameter σ_0 connected with a constant of the electron-phonon interaction g by the relation $\sigma_0 = 2/3\, g$ [14] on the electronic radiation dose. Thus, it can be stated that in this case the electronic radiation of $As_2S_3(Se_3)$ glasses is not only influencing the intrinsic defect centres existing befor radiation but also creates new electronically induced defects [15].

Conclusions

Dose dependences of energy parameters of the intrinsic absorption edge testify to an electron-induced creation of new defects which change the disorder potential. A characteristic energy of the exponential absorption tail W(T,X) shows not only the temperature but also the structural disordering of other kinds:

a) intrinsic structural disorder of an “ideal” glass;
b) induced structural disorder due to external factors (of radiation or technological nature).

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Table 1

Structural and temperature contribution dependences in W(X,T) for glassy As_2S_3 and As_2Se_3.

Doze	As_2Se_3		As_2S_3	
	W_S,meV	W_T,meV	W_S,meV	W_T,meV
D_0	43	5.7	39	4.6
D_1,10^{13}el/cm^2	50	7.0	44	5.5
D_2,10^{16}el/cm^2	56	8.2	50	6.4

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