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TEMPERATURE STUDIES OF OPTICAL ABSORPTION EDGE IN $(Ag_2S)_x(As_2S_3)_{1-x}$ $(x<0.2)$ SUPERIONIC GLASSES

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ABSTRACT

Synthesis of $(Ag_2S)_x(As_2S_3)_{1-x}$superionic glasses for $x<0.2$ are carried out. The spectrometric studies of optical absorption edge in $(Ag_2S)_x(As_2S_3)_{1-x}$glasses are performed in the temperature range 77-390 K. Optical absorption edge of $(Ag_2S)_x(As_2S_3)_{1-x}$ glasses with addition of Ag_2S is strongly smeared and has an exponential shape.The influence of temperature and composition on the absorption edge behavior, parameters of optical absorption edge and electron-phonon interaction as well as ordering-disordering processes in $(Ag_2S)_x(As_2S_3)_{1-x}$ superionic glasses are studied.

Keywords: superionic glasses, absorption edge, Urbach rule, electron-phonon interaction

1. INTRODUCTION

Chalcogenide glasses are the promising materials for the development of new solid electrolytes because of the high values of their electrical conductivity in comparison with oxide glasses [1]. It should be noted that these highly conductive glasses are transparent in the IR region, which is very useful for the creation of functional elements for optical devices. The combination of various properties in chalcogenide glasses [2] and a possibility to change functional parameters during modifications, i.e. a change of chemical composition and production technology, influence of external factors, leads to a wide range of their applications in holography and microlithography, systems of information recording, optoelectronics, infrared and nonlinear optics, sensorics, electronic technology etc. [3,4,5,6,7].

Therefore, Ag-containing ternary Ag-As-S glasses introduce a significant interestfor solid state ionic. As_2S_3 glassy semiconductor, due to theinteresting photostimulated structural changes being revealed in their optical properties, has found wide practical applications as an efficient material for optical data recording, holography, integrated optics. Ag_2S-As_2S_3 chalcogenide glasses differ among the other by high conductivity. The Ag-As-S glasses were extensively studied in the first place due to the potential possibility of their application as a solid electrolyte [8,9,10]. The mechanism of electrical conductivity (ionic or electronic) depends mainly on a silver content in the given ternaryglasses, which in turn influences the other physical properties. Hence, the present work is aimed at conductingtemperature studies of optical absorption edge in superionic glasses $(Ag_2S)_x(As_2S_3)_{1-x}$ with $x<0.2$.

2. METHODOLOGY OF EXPERIMENT

Synthesis of compounds in Ag_2S-As_2S_3 glassy system was carried out in evacuated to a residual pressure 10^{-3} ampoules by direct method from elementary components. The synthesis was performed at a temperature of $700^0$0 C during 24 h with following melt homogenization during 72 h. Melt tempering was held at cooling on the air (at room temperature).

Spectrometric studies of the optical absorption edge were carried out in the temperature range 77-390 K using LOMO KSVU-23 grating monochromator [11,12]. For low temperature studies cryostat of UTREX type was used, stability and accuracy of temperature measurement were maintained at ±0.5 K. The absorption coefficient value α was calculated based on the experimental values of transmission coefficient T_{tr} and reflectivity r using a known formula

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$$\alpha = \frac{1}{d}\ln\left[\frac{(1-r)^2 + \sqrt{(1-r)^4 + 4T_{tr}^2 r^2}}{2T_{tr}}\right], \quad (1)$$

where d is the sample thickness. The relative error in the absorption coefficient determination $\Delta\alpha/\alpha$ have not exceeded 10% at $0.3 \leq \alpha d \leq 3$ [11].

3. EXPERIMENTAL RESULTS AND DISCUSSION

Absorption edge studies of $(Ag_2S)_{0.05}(As_2S_3)_{0.95}$glasshave revealed two temperature ranges: a range of "parallel" red shift of the exponential absorption edge within the temperature interval 77 K ≤ T < 300 K and a range of the Urbach behaviour of the absorption edge at $T \geq 300$ K, in which the dependence of absorption coefficient on the photon energy and temperature is given by the Urbach rule (Fig.1). The Urbach behaviour of the absorption edge is well described by the relation [13]:

$$\alpha(h\nu,T) = \alpha_o \cdot \exp\left[\frac{\sigma(h\nu - E_0)}{kT}\right] = \alpha_o \cdot \exp\left[\frac{h\nu - E_0}{E_U(T)}\right], \quad (2)$$

where $\sigma = kT / E_u$ is the steepness parameter of the absorption edge, E_u is the Urbach energy (energy width of exponential absorption edge), α_0 and E_0 are coordinates of the convergence point of Urbach bundle. For comparison, values of parameters α_0 and E_0 for As_2S_3 and$(Ag_2S)_{0.05}(As_2S_3)_{0.95}$ glasses were listed in Table 1. It is shown that with Ag_2S contentincrease an enhancement of the convergence point coordinates α_0 and E_0 is observed.

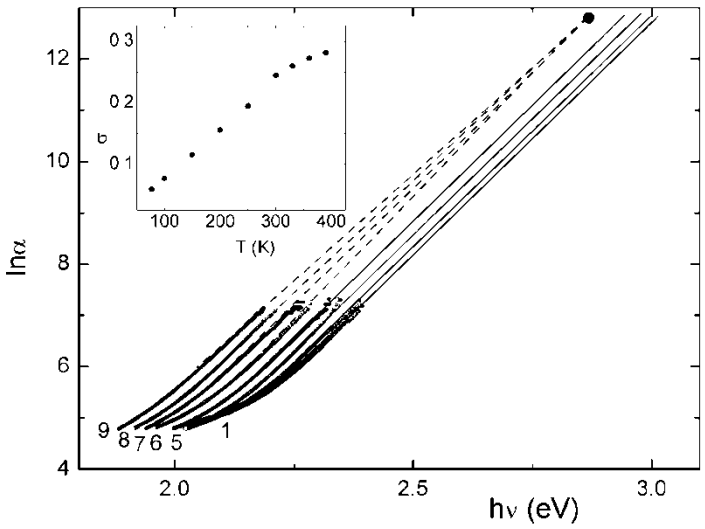

Fig. 1.Spectral dependences of the Urbach absorption edge for $(Ag_2S)_{0.05}(As_2S_3)_{0.95}$ glass at various temperatures: (1) 77, (2) 100, (3) 150, (4) 200, (5) 250, (6) 300,(7) 330, (8) 360 and (9) 390 K. The insert shows the temperature dependence of steepness parameter.

Parameters of the electron-phonon interaction (EPI) σ_0 and $\hbar\omega_p$, were derived from the temperature dependences of the absorption edge slope parameter σ (see insert in Fig. 1) by the equation [14,15]:

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$$\sigma(T) = \sigma_0 \cdot \left(\frac{2kT}{\hbar\omega_p} \right) \cdot \tanh\left(\frac{\hbar\omega_p}{2kT} \right), \quad (3)$$

where: $\hbar\omega_p$ is the effective phonon energy in a single-oscillator model, describing the electron-phonon interaction (EPI), and σ_0 is a parameter related to the EPI constant g as $\sigma_0 = (2/3)g^{-1}$ (parameters $\hbar\omega_p$ and σ_0 are given in Table 1).The parameters $\hbar\omega_p$ and σ_0 for As_2S_3 and $(Ag_2S)_{0.05}(As_2S_3)_{0.95}$ glass are listed in Table 1. It is shown that for the $(Ag_2S)_{0.05}(As_2S_3)_{0.95}$ glass the parameter $\sigma_0<1$, which is an evidence of a strong EPI. Thus, with addition of Ag_2S to As_2S_3 a strengthening of the EPI (i.e. decrease of the σ_0 value) is revealed, whereas the effective phonon energy grows by 57% as compared with As_2S_3 (Table 1).

Table 1. Parameters of the Urbach absorption edge and parameters of EPI for $(Ag_2S)_x(As_2S_3)_{1-x}$ glasses

Compound	**As_2S_3**	**$(Ag_2S)_{0.05}$ $(As_2S_3)_{0.95}$**	**$(Ag_2S)_{0.1}$ $(As_2S_3)_{0.9}$**	**$(Ag_2S)_{0.15}$ $(As_2S_3)_{0.85}$**
α_0 (cm^{-1})	2.97×10^5	3.61×10^5		
E_0 (eV)	2.605	2.868		
E_g^* (300 K) (eV)	2.323	2.252	1.932	1.6640
E_U (300K) (meV)	51.0	105.8	437.7	452.4
σ_0	0.63	0.374		
$\hbar w_p$ (meV)	43.9	68.7		
θ_E (K)	510	797		
$(E_u)_0$ (meV)	35.1	91.7		
$(E_u)_1$ (meV)	70.7	183.8		
$E_g^*(0)$ (eV)	2.395	2.342		
S_g^*	7.6	17.5		

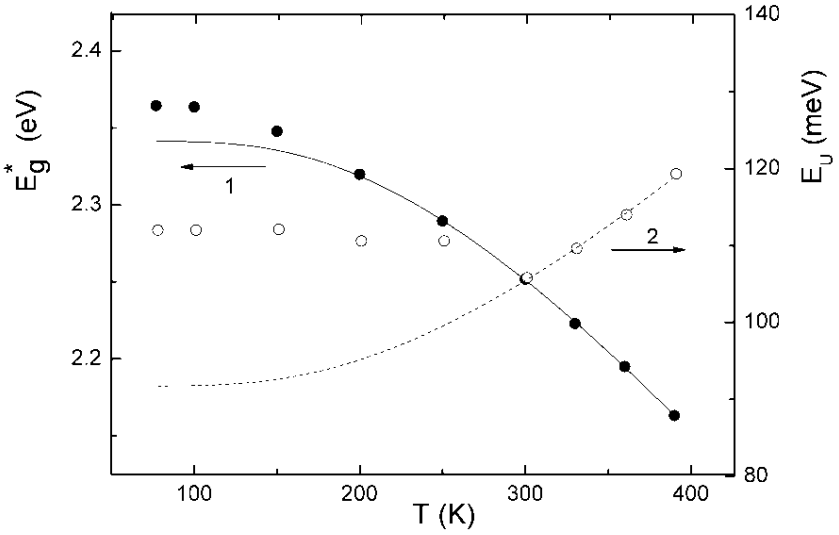

Fig. 2. Temperature dependences of optical pseudogap E_g^* (1) and Urbach energy E_U (2) of $(Ag_2S)_{0.05}(As_2S_3)_{0.95}$ glass: circles – experiment, curves – calculations.

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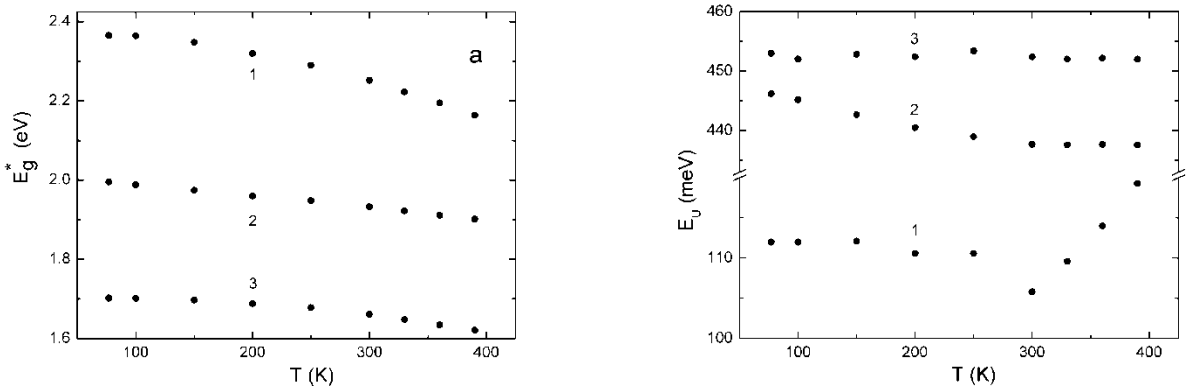
Figure 2 illustrates the temperature dependences of the optical pseudogap E_g^* (spectral position of absorption edge at fixed value of absorption coefficient α=10^3 cm^{-1}) and Urbach energy E_u which are well described in the temperature range of the Urbach behaviour by the equations [16,17,18]:

$$E_g^*(T) = E_g^*(0) - S_g^* k\theta_E \left[\frac{1}{\exp(\theta_E / T) - 1} \right], \quad (4)$$

$$E_U(T) = (E_U)_0 + (E_U)_1 \left[\frac{1}{\exp(\theta_E / T) - 1} \right], \quad (5)$$

where $E_g^*(0)$ and S_g^* are the energy pseudogap at 0 K and a dimensionless constant, respectively; θ_E is Einstein temperature, corresponding to the average frequency of phonon excitations of a system of non-coupled oscillators, $(E_U)_0$ and $(E_U)_1$ are constants. The obtained $E_g^*(0)$, S_g^*, θ_E, $(E_U)_0$, and $(E_U)_1$ parameters for $(Ag_2S)_{0.05}(As_2S_3)_{0.95}$ glass are given in Table 1.

Temperature studies of the optical absorption edge in $(Ag_2S)_{0.1}(As_2S_3)_{0.9}$ and $(Ag_2S)_{0.15}(As_2S_3)_{0.85}$ glasses showed that in the temperature interval 77 K ≤ T ≤ 390 K a red shift of the exponential absorption edge is observed with the temperature increase. The Urbach energy for $(Ag_2S)_{0.1}(As_2S_3)_{0.9}$ glass in the temperature interval 77 K ≤ T < 300 K decreases, while at $T \geq 300$ K remains unchanged; the Urbach energy for $(Ag_2S)_{0.15}(As_2S_3)_{0.85}$ glass in the temperature interval under investigation is a constant value (Fig.3). The non-Urbach behaviour of the optical absorption edge in $(Ag_2S)_{0.1}(As_2S_3)_{0.9}$ and $(Ag_2S)_{0.15}(As_2S_3)_{0.85}$ glasses can be explained using the formalism of separation of the contributions from static and dynamical structural disordering types. It is shown that in the temperature interval under investigation only the short-range order in the atomic arrangement $(Ag_2S)_{0.1}(As_2S_3)_{0.9}$ and $(Ag_2S)_{0.15}(As_2S_3)_{0.85}$ glasses is present. With the temperature increase, the medium-range order is gradually established, resulting in a decrease of dynamic structural disordering contribution $(E_U)_{X,dyn}$. The decrease of $(E_U)_{X,dyn}$ along with the increasing contribution of the temperature-related disordering $(E_U)_T$ at the constant contribution of static structural disordering contribution $(E_U)_{X,stat}$ results in a temperature independence of the Urbach energy E_U in the $(Ag_2S)_{0.1}(As_2S_3)_{0.9}$ and $(Ag_2S)_{0.15}(As_2S_3)_{0.85}$ glasses.



(a) (b)

Fig. 3. Temperature dependences of optical pseudogap E_g^* (a) and Urbach energy E_U, (b) for $(Ag_2S)_{0.05}(As_2S_3)_{0.95}$ (1), $(Ag_2S)_{0.1}(As_2S_3)_{0.9}$ (2) and $(Ag_2S)_{0.15}(As_2S_3)_{0.85}$ (3) glasses.

Compositional studies of $(Ag_2S)_x(As_2S_3)_{1-x}$ superionic glasses showed that with increasing of Ag_2S content the optical absorption edge is shifted to the low-energy range and strongly smears (Fig.4). The nonlinear decrease of E_g^* as well as the sharp increase of Urbach energy (almost into 4 times) is observed at the transition from $(Ag_2S)_{0.05}(As_2S_3)_{0.95}$ to $(Ag_2S)_{0.1}(As_2S_3)_{0.9}$ (Fig.5).

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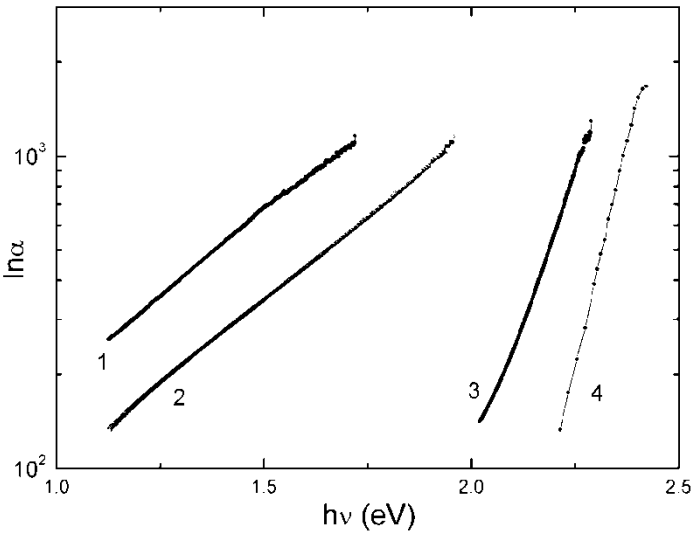

Fig. 4. Spectral dependences of the absorption edge for As_2S_3 (1), $(Ag_2S)_{0.05}(As_2S_3)_{0.95}$ (2), $(Ag_2S)_{0.1}(As_2S_3)_{0.9}$ (3) and $(Ag_2S)_{0.15}(As_2S_3)_{0.85}$ (4) glasses at temperature T = 300 K.

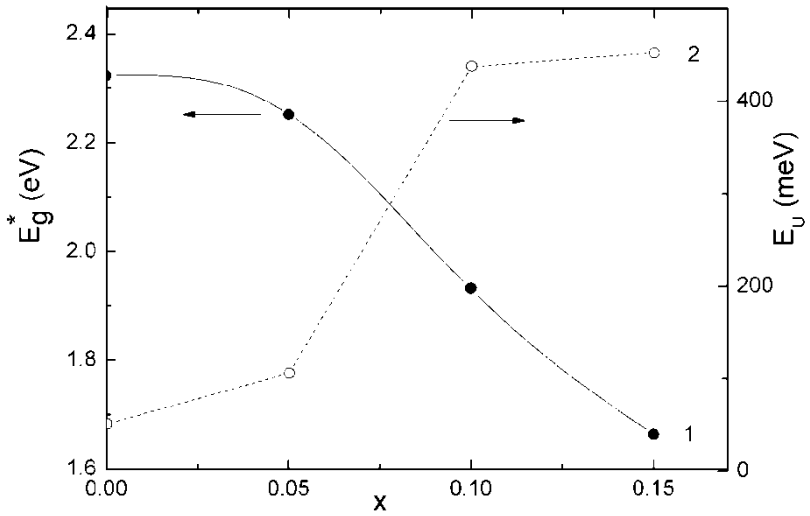

Fig. 5. Compositional dependences of optical pseudogap E_g^* (1) and Urbach energy E_U (2) for $(Ag_2S)_x(As_2S_3)_{1-x}$ glasses.

It should be noted that besides the temperature and structural disordering in $(Ag_2S)_x(As_2S_3)_{1-x}$ glasses the compositional disordering, caused by the addition of Ag_2Sin As_2S_3, is realized. The structural disordering in glasses under investigations is caused by (i) the thermal vibrations of atoms and structural elements, and (ii) by the defects and impurities of a structure and absence of a long-range order in an atomic arrangement. According to [19], the effects of different types of disordering influence on the Urbach energy in solid solution are described by the relation

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$$E_U = (E_U)_T + (E_U)_X + (E_U)_C = (E_U)_T + (E_U)_{X+C}, \quad (1)$$

where $(E_U)_T$, $(E_U)_X$ and $(E_U)_C$ are the contributions of temperature, structural and compositional disordering to (E_U), respectively. It is seen from Eqs. (5) and (6) that $(E_U)_{X,C} \equiv (E_U)_0$ and $(E_U)_T \equiv (E_U)_1/(\exp(\theta_E/T) - 1)$. For the estimation of the contribution of the different types of disordering into Urbach energy E_U we used the procedure described in Ref. [20,21,22]. Thus, the contributions of temperature in dependent $(E_U)_{X,C}$ (structural and compositional) and temperature dependent $(E_U)_T$ disordering were calculated. It is shown that with addition of Ag_2S the contribution of the sum of structural and compositional disordering into the Urbach energy grows up from 69 % to 87% for $(Ag_2S)_{0.05}(As_2S_3)_{0.95}$ glass.

4. CONCLUSIONS

It has been shown that with addition of Ag_2S the absorption edge of $(Ag_2S)_x(As_2S_3)_{1-x}$ glasses is strongly smeared and has an exponential shape. For $(Ag_2S)_{0.05}(As_2S_3)_{0.95}$ glass two regions of absorption edge temperature behaviour were revealed: a range of "parallel" red shift of the exponential absorption edge within the temperature interval 77 K $\leq T <$ 300 K and a range of the Urbach behaviour of the absorption edge at $T \geq$ 300 K; for $(Ag_2S)_{0.1}(As_2S_3)_{0.9}$ and $(Ag_2S)_{0.15}(As_2S_3)_{0.85}$ glasses in the temperature interval 77 K $\leq T \leq$ 390 K a red shift of the exponential absorption edge is observed with the temperature increase.It was found outthat with addition of $Ag_2S$to $As_2S_3$the EPI becomes stronger, whereas the effective phonon energy in $(Ag_2S)_{0.05}(As_2S_3)_{0.95}$ glass increases by 57% comparing with pure As_2S_3. With Ag_2Scontent increase one can observe a nonlinear decrease ofE_g^*. Moreover, the Urbach energy E_U grows by almost four times at the transition from $(Ag_2S)_{0.05}(As_2S_3)_{0.95}$ to $(Ag_2S)_{0.1}(As_2S_3)_{0.9}$. The contributions of the temperature independent (i.e. structural and compositional) and temperature dependent disordering to the Urbach energy were estimated for $(Ag_2S)_{0.05}(As_2S_3)_{0.95}$ glass.

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