EVALUATION OF HOLE DRIFT MOBILITY IN GLASSY As$_2$S$_3$ IN THE TEMPERATURE RANGE 77 - 330 K

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The temperature behaviour of time-averaged hole drift mobility in As$_2$S$_3$ glass was studied in the range 77-330 K on the basis of temperature dependence of steady-state photoinduced absorption. The time-averaged hole mobility was found to be thermally activated at high temperatures with the activation energy ~0.9 eV, of the order of 10^{-10} cm^2/Vs at room temperature, but almost temperature independent below ~ 130 K.

1. Introduction

The drift mobility in low resistivity chalcogenide glasses has been widely investigated during the years [1,2]. The situation is more difficult in the case of As$_2$S$_3$. Investigation of hole drift mobility in As$_2$S$_3$ has been carried out by different research groups mainly on thin film samples. For example, direct measurements have been carried out in As$_2$S$_3$ in the range above room temperature up to +165 °C [2]. The hole drift mobility in pure and doped vapour deposited As$_2$S$_3$ films have been studied in [3] down to - 50 °C. On the other hand there are no experimental results on As$_2$S$_3$ concerning the hole drift mobility at low temperatures. Therefore is very attractive to evaluate the temperature dependence of the time-averaged hole drift mobility in As$_2$S$_3$ glass from the steady-state photoinduced absorption measurements.

2. Experimental details

We have investigated the steady-state characteristics of photoinduced absorption in As$_2$S$_3$ glass fibre under continuous illumination with band-gap light. Employment of fibre samples enables to carry out PA measurements not only at liquid nitrogen temperature but in a wide temperature range, up to 330 K [4,5].

PA measurements have been carried out on unclad fibre samples of As$_2$S$_3$ glass in the temperature range 77 - 330 K. The excitation light source was a 2-W Ar laser. The fibre samples were illuminated with a continuous light with photon energy $h\nu_{exc} \sim E_g$. The probe beam from the spectral range $h\nu_p < E_g$ from a grating monochromator was injected into the input end face of the fibre. When illuminating the lateral surface of the fibre with continuous bandgap light the intensity of probing light at the output of the fibre decreases from its initial value in the dark I_0 to a new one I. The photoinduced absorption coefficient $\Delta\alpha$ is determined as follows:

$$\Delta\alpha = L^{-1} ln(I_0/I) \tag{1}$$

where L is the length of the illuminated segment of the fibre.

The probe beam was adjusted so that the intensity of the probe beam was much less than the intensity of the excitation light, and the probe light intensity never affected the magnitude of photoinduced absorption. We note that after cessation of the bandgap light a full restoration of initial transmittance occurs. Different lengths of the fibre samples (up to 4 m) have been used for measurement of the PA coefficient in the range 77 - 330 K. The intensity of the excitation light was varied up to 10^{-2} W/cm^2.
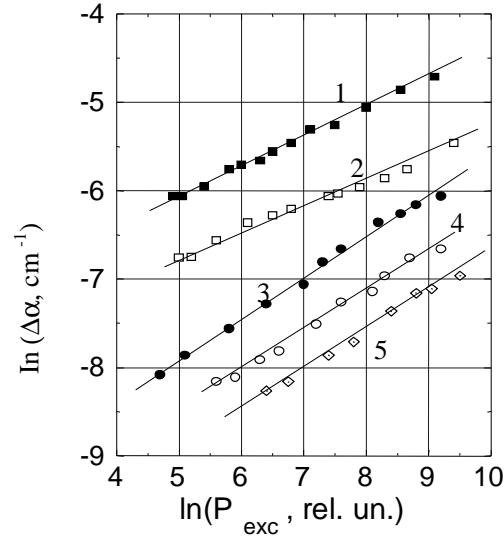
Fig. 1. Intensity dependence of the steady-state PA coefficient in As$_2$S$_3$ glass fibres at room temperature under continuous bandgap excitation ($h\nu_{exc} = 2.41$ eV). The probing light photon energy $h\nu_p$, eV: 1,2 (1); 1.08 (2); 0.95 (3); 0.8 (4); 0.7 (5).

The temperature dependence of steady-state PA coefficient was measured for different probing light photon energies for excitation light intensity P = 10^{-2} W/cm^2 (Fig. 2). The steady-state photoinduced absorption is strongly affected by the temperature. When decreasing the temperature from room temperature 330 K down to liquid nitrogen temperature PA coefficient increases by several orders of magnitude.
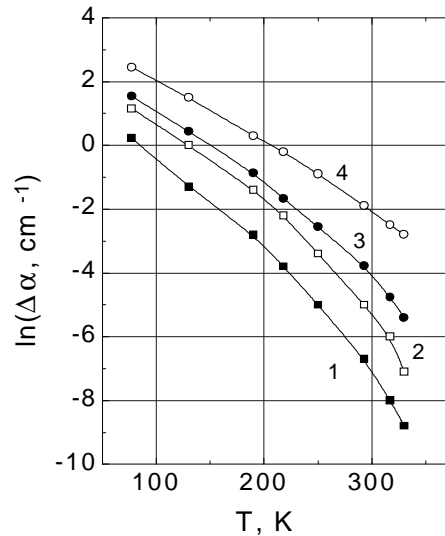
Fig. 2. Temperature dependence of the steady-state PA coefficient in As$_2$S$_3$ glass fibres. The probing light photon energy $h\nu_p$, eV: 1.5 (1); 1.2 (2); 1.08 (3); 0.7 (4). The excitation light intensity P = 10^{-2} W/cm^2, $h\nu_{exc} = 2.41$ eV.

3. Discussion

The dependence of the steady-state PA coefficient on the excitation light intensity exhibits approximately the square-root behaviour when the intensity of the exciting light is varied by about four orders of magnitude, 10^{-6}-10^{-2} W/cm^2 (Fig. 1). It suggests that in the steady state recombination of the PA absorbing centres can be described by a bulk bimolecular mechanism [8]. This argument is supported by many experimental results on photoconductivity of chalcogenide glasses, where it was found that in the steady-state the photocondctivity goes up as the square root of the light intensity.

For probing light photon energy $h\nu_p$ the PA coefficient is determined by the density of photoinduced absorbing centres localised in the energy range $0 < E < h\nu_p$. On the other hand for the stationary state the density of photoinduced absorbing centres, and respectively the photoinduced absorption coefficient, is determined by the bimolecular recombination coefficient b, which controls the density of localised absorbing centres. It is known that only those levels, positioned between the quasi-Fermi levels for trapped holes, and the quasi-Fermi level for trapped electrons are effective in the recombination process. These traps are so deep that thermal re-excitation into the valence/conduction band can be neglected. The rate equation for the density of photoinduced absorbing centres N in the steady-state is as follows:

$$dN/dt = N_f/\tau - bN^2 \tag{2}$$

where N_f is the density of free carriers, and τ is the life-time of excess carriers. The lifetime for excess carriers is determined by $\tau = N_f/G$, where G is the generation rate. Therefore equation (1) will be:

$$dN/dt = G - bN^2 \tag{3}$$

In the steady state $dN/dt = 0$ and therefore $N = (G/b)^{1/2}$. On the other hand, the fractional change in optical transmission is determined by the density of induced absorbing centres N and induced absorption cross section σ.

For simplicity we shall approximate the illuminated segment of the fibre (which represents a cylinder of diameter d and length L) by a prism of the same length and of square cross section d^2. Therefore, we can write for the fractional change of optical transmission $\Delta T/T$:

$$-\Delta T/T = \sigma \int_0^d N(z)dz \tag{4}$$

were z is the distance from the surface, d is the diameter.

In the general case the generation rate of excess carriers is determined as:

$$G(z) = \alpha F\gamma(1-R)exp(-\alpha z) \tag{5}$$

where α is the absorption coefficient for excitation light, F is the incident photon flux, γ is the quantum efficiency and R is the reflectivity for excitation light. The recombination coefficient b can be related to the fractional change in optical transmission through approximated formula [8,9]:

$$b = 4\sigma^2 F\gamma(1-R)/(\Delta T/T)^2\alpha \tag{6}$$

Relation (6) can be used for evaluation of the temperature dependence of the bimolecular recombination rate coefficient. Fig. 3 shows the temperature dependence of the bimolecular rate coefficient, determined from the temperature dependence of steady-state PA coefficient (Fig. 2). The recombination b, determined from Eq. (6) is strongly dependent on the absorption cross section per carrier σ. When deriving $b(T)$ a value of $\sim 10^{-16}$ cm^2 was used for σ estimated from the measurements of light induced ESR and PA in chalcogenide glasses [11]. The absorption coefficient α was taken from the measurements of Tauc et al. [12], corrected for its temperature dependence.

Orenstein and Kastner [9] have shown that PA and carrier mobility are governed by the same localized carriers. In highly disordered semiconductors, where the mean free path of the carrier in the conducting states is of the order of the interatomic distance, the carriers motion is diffusive [13]. For low mobility semiconductors the diffusion limited recombination is characterised by the Langevin equation [8,13]:
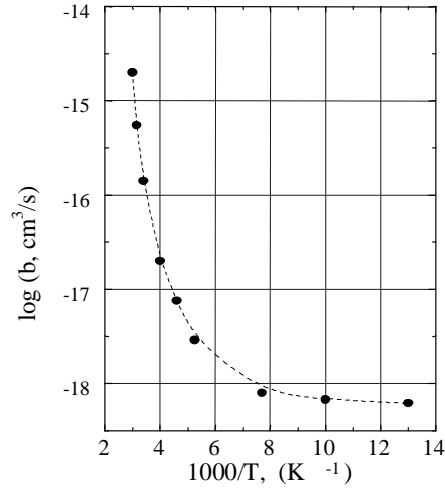

Fig. 3. Temperature dependence of the bimolecular rate coefficient in As_2S_3 glass derived from PA measurements.

$$b = (e/\varepsilon\varepsilon_0)(\mu_n + \mu_p) \tag{7}$$

were μ_n and μ_p are the drift mobility for electrons and holes, respectively, in their mutual Coulomb field. It is known that for chalcogenide glasses the drift mobility for electrons is much less than the holes mobility, $\mu_n << \mu_p$, and μ_n can be neglected in Eq. (7). Therefore, we can estimate the temperature dependence of time-averaged drift mobility from the curve $b(T)$ in Fig. 3.

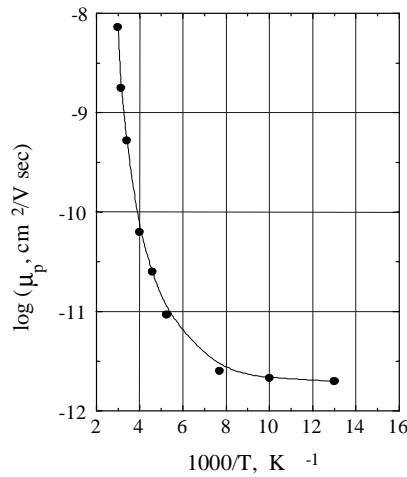$$\mu_p = (\varepsilon\varepsilon_0/e)b \tag{8}$$

Fig. 4. Temperature dependence of the time-averaged drift mobility in As_2S_3 glass derived from temperature dependence of steady-state PA coefficient.

The temperature dependence of hole drift mobility obtained from converting $b(T)$ is represented in Fig. 4. We can mention that the room temperature drift mobility value, obtained in this way is comparable with the drift mobility obtained for As_2S_3 in [2] from electrical measurements.

From the results in Fig. 6 we can observe that the temperature dependence of drift mobility of photoinduced holes exhibits an activation character at high temperatures. The activation energy estimated from the curve in Fig. 4 is approximately 0.9 eV.

4. Conclusions

We have investigated the temperature dependence of time-averaged hole drift mobility in As$_2$S$_3$ glass on the basis of the temperature dependence of steady-state photoinduced absorption in the range 77 - 330 K. For a wide range of excitation light intensity the PA coefficient was found to vary as a square root of the excitation intensity, indicating on the bimolecular recombination mechanism. The temperature dependence of time-averaged hole drift mobility was derived through the Langevin formula for highly-disordered semiconductors, which relates the bimolecular recombination coefficient to carriers mobility.

The time-averaged hole drift mobility in As$_2$S$_3$ was found to be temperature activated at high temperatures, and almost temperature independent below approximately 130 K. The hole mobility at room temperature was found to be of the order of 10^{-10} cm^2/Vs, which correlates with the results obtained from electrical measurements elsewhere. The results presented here have been obtained with a number of approximations, however at this time no other experimental data on carriers mobility in As$_2$S$_3$ down to 77 K were available.

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