

TEMPERATURE DEPENDENCE OF PHOTOINDUCED ABSORPTION IN As_2S_3 GLASS FIBERS

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Chalcogenide glasses are promising materials for different applications in integrated and fiber optics due to a number of advantages in comparison to other optical materials. Chalcogenide glasses exhibit excellent transmission in the near and infrared spectral region up to $\sim 12 \mu\text{m}$, high refractive index ($n \geq 2.4$), low phonon energy, and higher nonlinearity of the refractive index in respect to silica (typically $\times 100$) [1-2]. One of interesting features of chalcogenide glasses is the effect of reversible photoinduced absorption (PA) observed when the chalcogenide glass is illuminated with bandgap light ($h\nu_{exc} \sim E_g$). Photoinduced absorption proved to be an efficient tool for investigation of optical properties in amorphous semiconductor materials [3-6]. PA effect presents interest both as a method for determination of phenomenological parameters of localized states in chalcogenide glasses as well as from the point of view of application in optoelectronics.

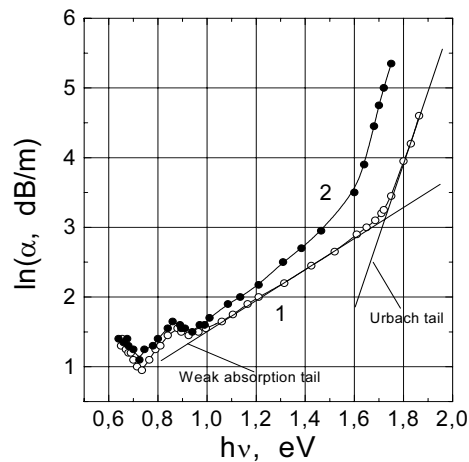


Fig.1. The spectral distribution of the optical absorption in chalcogenide fibers As_2S_3 (1) and As-S-Se (2).

We have investigated the photoinduced absorption in As_2S_3 glass fibers under continuous illumination of the lateral surface of the fibers with band-gap light. The spectral distribution of the optical absorption in chalcogenide glass fibers As_2S_3 is represented in Fig.1. PA measurements have been carried out on unclad fibre samples in the temperature range 77-330 K. The power of the probe beam was adjusted so that its intensity was much less than the intensity of the excitation light not to affect the magnitude of photoinduced absorption. We note that after secession 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 PA coefficient.

At room temperature 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/cm² (Fig. 2). It suggests that in steady state conditions the recombination process of the PA absorbing centres can be described by a bulk bimolecular mechanism. This argument is supported by many experimental results on photoconductivity of chalcogenide glasses, where it was found that in the steady-state the photoconductivity goes up as the square root of the light intensity. The sub-linear intensity dependence is a result of a recombination process, their rate depending upon the excitation light intensity.

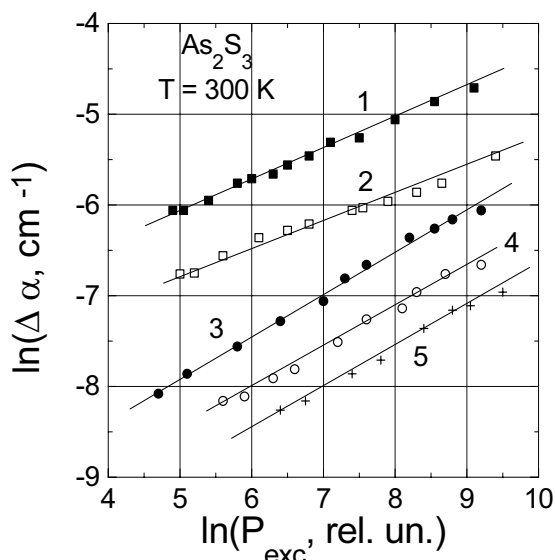


Fig. 2. The intensity dependence of steady state PA coefficient in As_2S_3 glass fibers at room temperature for various probing light photon energy $h\nu_p$ (eV): 1.2 (1); 1.08 (2); 0.95 (3); 0.8 (4); 0.7 (5); The excitation light $\lambda_{\text{exc}} = 0.35$ - 0.75 μm .

For low temperature ($T = -172$ C) the intensity dependence of PA coefficient exhibits a similar power law behaviour $\Delta\alpha \sim P^n$ with the coefficient n around 0.4. Another feature of photoinduced absorption at low temperature is the “saturation” effect that is illustrated for probing light photon energy $h\nu_p = 2.14$ eV (Fig. 4).

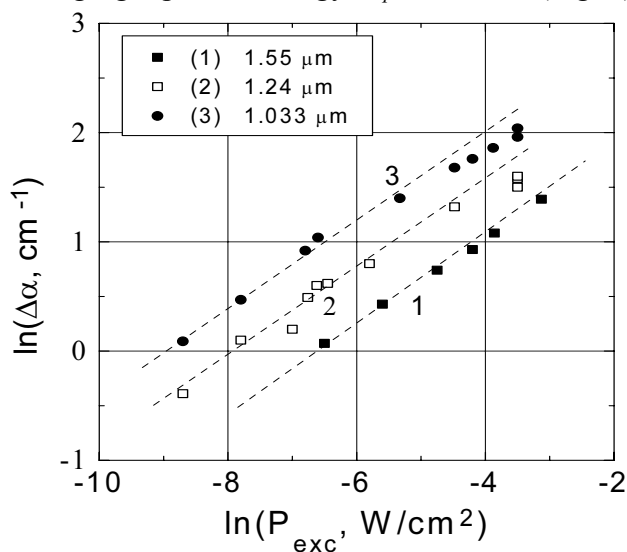


Fig. 3. Intensity dependence of PA steady-state coefficient in As_2S_3 glass fibers at -172 °C. Illumination was carried out with Ar^+ -light, $h\nu_{\text{exc}} = 2.41$ eV. The probing light wavelength $\lambda_p = 1.55$; 1.24 ; 1.033 μm .

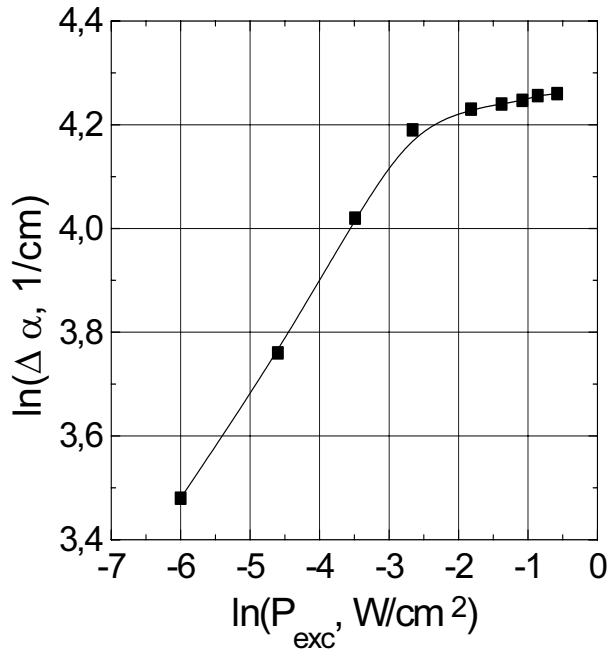


Fig. 4. The intensity dependence of steady state PA coefficient in As_2S_3 glass samples at $T = -172$ C for probing light $h\nu_p = 2.14$ eV. Illumination was carried out with Ar^+ -light $h\nu_{exc} = 2.41$ eV.

When illuminating the fiber sample with the bandgap light the exciting light is absorbed non-uniformly through the fiber cross section. The excess kinetic energy of the electron-hole pair is dissipated through exchange of energy with phonons. In an amorphous semiconductor these electrons and holes are not necessarily free to move, since either one or both of photo-induced charges can be in localized states. Even when both carriers are free they initially move in each other's field and if they can recombine with each other, a process called geminate recombination occurs [7, 8]. If the carriers avoid geminate recombination, they can move by hopping through localized states. The mobile carriers can be also trapped in localized states. Once trapped they can be released again via a trapping of the opposite type of carrier.

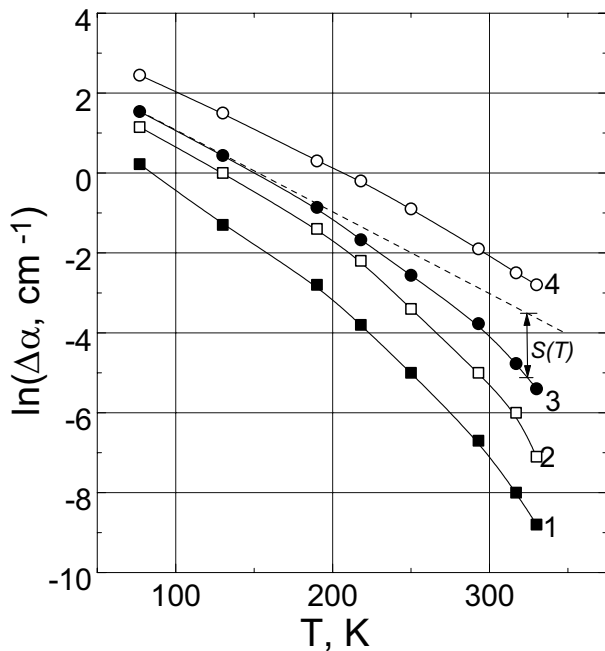


Fig. 5. The temperature dependence of the steady state PA coefficient in As_2S_3 glass fibers. The fiber samples were irradiated with Ar laser light $\lambda_{exc} = 0.46-0.52$ μm of 10^{-2} W/cm^2 . The probing light photon energy $h\nu_p$ (eV): 0.78 (1); 1.08 (2); 1.2 (3); 1.5 (4).

When free to move, the electron and hole diffuse apart to a distance r , determined by the diffusion constant and thermalization time, which is proportional to the excess ki-

netic energy [7, 8]. The electron and hole are bound by the energy $e^2 / 4\pi\epsilon\tilde{\epsilon}r$. According to the theory, such thermalized pair can either recombine (geminate recombination) or escape their mutual Coulomb attraction and separate into a free electron and a free hole [8-10]. If r is larger than the critical distance r_0 ($e^2 / 4\pi\epsilon\epsilon_0 r_0 = kT$), then the electron and hole will diffuse apart. If $r < r_0$ they hop together and form an exciton [7]. Recombination does not take place until carriers thermalize to energy near the mobility edge.

Excitation of chalcogenide glass with band-gap light creates a density of excess electrons and holes, which thermalize more quickly than it takes them to recombine. These excess carriers diffuse through the material until they recombine, or are permanently trapped. Until the carriers recombine or are permanently trapped they may contribute to the conductivity or to photoinduced absorption. The captured carriers take part in the process of photoinduced absorption. It is appropriate to speak of the diffusion of the carriers before they recombine since their average separation is much greater than their hopping distance [8, 9]. For probing light photon energy $h\nu_p$ the PA coefficient is determined by the density of photoinduced absorbing centres localized in the energy range $0 < E < h\nu_p$. On the other hand, for the stationary state conditions the density of photoinduced absorbing centres and, respectively, the photoinduced absorption coefficient are determined by the bimolecular recombination coefficient, which controls the density of localized absorbing centres.

The temporal characteristics of PA under condition of lateral illumination with bandgap light have been studied for the stage after switching on/off the excitation light [11] in a time range $\sim 10^{-2}$ - 10^4 s. The character of PA kinetics depends on experimental conditions, such as temperature, probing light photon energy and excitation light intensity. Illustration of PA time dependence $\Delta\alpha(t)$ for different excitation light intensities is represented in Fig. 6, 7. After the excitation light switching on one can observe a rapid increase of PA coefficient $\Delta\alpha$ up to a quasi-stationary state, with a very slow subsequent change. This quasistationary state PA can be treated with a sufficient approximation as a stationary state PA. Respectively, the time interval after the excitation light switching on, when PA reaches the quasistationary state can be defined as the stationary state time t_{st} (Fig. 6). The quasi-stationary time does not depend on probing light photon energy, but decreases with increasing the excitation light intensity. For low excitation light intensities ($\sim 10^{-6}$ W/cm²) at liquid nitrogen temperature the PA kinetics has a linear character in a wide time interval (10^{-6} W/cm²) (Fig. 7).

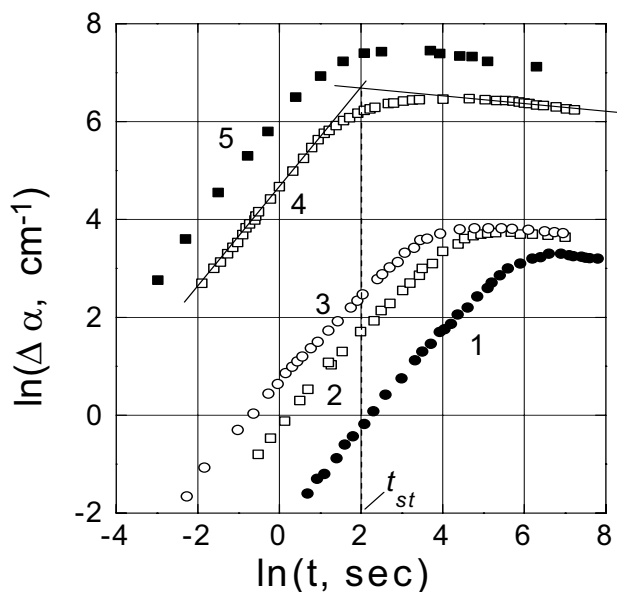


Fig. 6. The kinetics of photoinduced absorption in As_2S_3 fibers at temperature 77 K for probing light photon energy $h\nu_p = 0.7$ eV. The intensity of excitation light P_{exc} : 0.27 (1); 2.9 (2); 7.9 (3); 50 (4); 400 (5) mW/cm². The probing light wavelength $\lambda_p = 0.514$ μm .

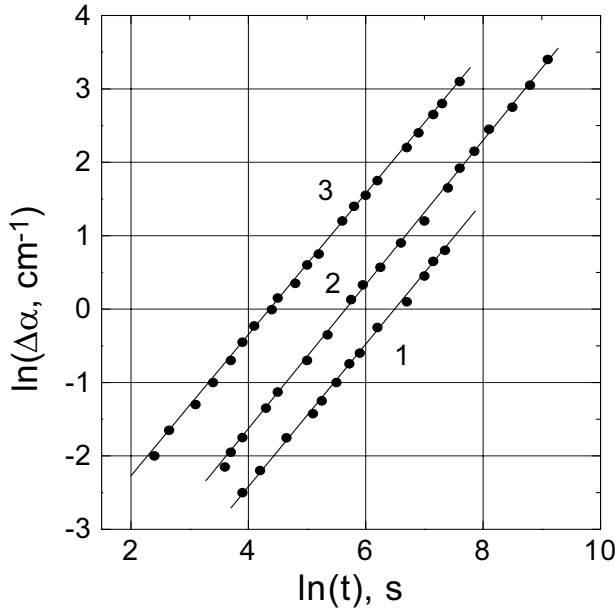


Fig. 7. The PA kinetics in As_2S_3 glass fibers at 77 K under weak excitation of the lateral light, $P_{exc} = 10^{-5} \text{ W/cm}^2$. λ_p : 1.77 (1); 1.15 (2); 0.826 (3) μm .

The temperature dependence of steady-state PA coefficient was measured for different probing light photon energies for excitation light intensity $P_{exc} = 10^{-2} \text{ W/cm}^2$. It was shown that the steady state PA coefficient exhibits a very strong temperature dependence. While decreasing the temperature from 330 K to 77 K the PA coefficient increases by several orders of magnitude. In the framework of multiple trapping model the dependence $\Delta\alpha(T)$ can be approximated by a power-law function (Fig.5) with a small additional term $S(T)$, which weakly depends on temperature and decreases with increasing the probing light photon energy [11]:

$$\ln(\Delta\alpha) = \ln C_1 - (kT/E_0)\ln A - S,$$

where C is a constant, $A > 1$ is a parameter that depends on the light excitation G , S is an additional negative term which depends weakly on temperature: $\ln S = -h\omega/E_0 + (kT/E_0)\ln A$. By plotting the magnitude of term $\ln S$ against T and extrapolating it to $T = 0$ one can find the magnitude of the characteristic energy of the distribution of traps E_0 [11].

P. O’Connor and J. Tauc [12] have shown that PA characteristics can be applied for evaluation of temperature dependence of the bimolecular rate coefficient (b) and the time averaged hole drift mobility (μ_p). For the stationary state conditions the density of photoinduced absorbing centers and respectively the PA coefficient are determined by the bimolecular recombination coefficient, which controls the density of localized absorbing centers. Under moderate excitation light intensities ($P_{exc} < 10^{-2} \text{ W/cm}^2$) the recombination process in As_2S_3 glass is found to be bimolecular and diffusion limited. This let us relate the PA coefficient to the bimolecular recombination rate coefficient, generation rate, and the fractional change in optical transmission $\Delta T/T$ through an approximate equation [12]:

$$b = 4\sigma^2 F(1-R)/(\Delta T/T)^2 \alpha.$$

where $\Delta T/T$ is the fractional change in optical transmission; R is the reflectivity for excitation light; F is the incident photon flux; b is the induced absorption cross section; α is the absorption coefficient for excitation light. Further, one can show that the bimolecular recombination coefficient b can be related to the time averaged hole drift mobility μ_p , as follows [12]:

$$\mu_p = (\epsilon\epsilon_0/e) b.$$

This approach has been applied to PA in chalcogenide glass fibers [13] for deriving the time averaged hole drift mobility in glassy As_2S_3 from the temperature dependence of the stationary state PA coefficient (Fig. 8). By this technique the time averaged hole drift mobility

was found to be temperature activated at high temperatures and almost temperature independent below approximately 130 K. The time-averaged hole drift mobility at room temperature value was found to correlate with the results obtained from electrical measurements [14]. The temperature dependence of the drift mobility of photoinduced holes at room temperature has an activation energy of about 0.9 eV.

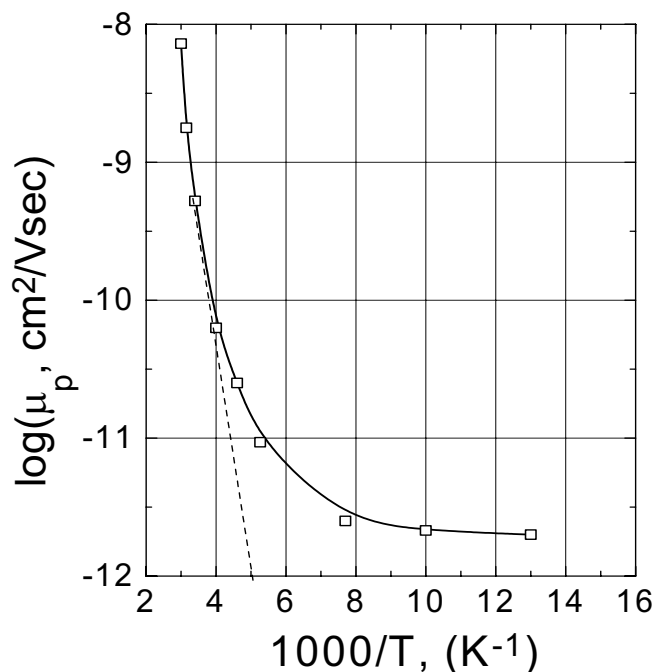


Fig. 8. Temperature dependence of the time-averaged drift mobility (μ_p) in As_2S_3 glass derived from the temperature dependence of steady state PA coefficient (*Andreish, Culeac, Owen and Ewen, 1998*).

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