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Photoconductivity in thin films of a-Ga₄₀Se_xTe_{60-x}

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Résumé. — Cet article présente des mesures de photoconductivité dans des films minces de verres Ga₄₀Se_xTe_{60-x} ($x = 20, 30, 40$). La dépendance en température du photocourant à l'état stationnaire et du courant d'obscurité montrent que les photoconductivités correspondantes sont activées. L'intensité du photocourant montre une dépendance en loi de puissance en fonction de la radiation incidente. On a aussi effectué des mesures de photoconductivité transitoires sur tous les échantillons. On trouve que la décroissance du photocourant après coupure de l'irradiation est exponentielle dans tous les verres. La décroissance de I_{ph}/I_d peut être expliquée par la croissance du nombre de défauts avec la concentration de Te.

Abstract. — Present communication reports on photoconductivity measurements in thin films of Ga₄₀Se_xTe_{60-x} glasses ($x = 20, 30, 40$). Temperature dependence of steady state photocurrent and dark current show that dark and steady state photoconductivity are activated processes. Intensity dependence of photocurrent reveals about power law dependence of photocurrent on incident radiation. Also incorporated are the transient photoconductivity measurements on all the samples. The decay of photocurrent after the cessation of illumination has been found to be exponential in all the glasses. The decrease in I_{ph}/I_d can be explained in terms of the increasing no. of defect states with increasing Te content.

Introduction.

Due to manifold applications in different fields amorphous semiconductors and in particular chalcogenide glasses are drawing a lot of attention. In order to optimize these materials for possible applications it is essential to understand the nature of recombination kinetics taking place in these materials. Chalcogenide glasses are generally characterized by a trap-limited carrier mobility, a Fermi energy in the center of the band gap, diamagnetic behaviour and an insensitivity to preparation conditions. Photoconductivity measurements are relevant from the application point of view as well as to understand the nature of localized states present in these materials.

Photoconductivity measurements have been carried out on a-semiconductors in bulk form or in thin film form [1-8] and a number of models [9-12] have been proposed to explain the results. The diverse nature of photoconducting properties in a-semiconductors is visible from presence of maxima with

respect to temperature in some glasses [1-3] whereas its absence was reported in few others [7, 8]. The intensity dependence of photocurrent on incident radiation has been reported to be linear in some cases [2, 4] whereas it is sq. root in others [4, 6, 8]. Decay of photocurrent, after the incident radiation is switched off has been reported as non exponential in some cases [8, 13, 14], as a power law decay in a few others [15, 16] and also as a sum of exponential decays [4, 17, 18] depending upon the nature of recombination process taking place in the material.

This communication reports about the photoconductivity studies in thin films of Ga₄₀Se_xTe_{60-x} ($x = 20, 30, 40$). Temperature dependence of steady state photoconductivity is studied at various levels of illumination in amorphous thin films of various compositions. Intensity dependence of photocurrent is studied at different temperatures in the range 310-370 K. Also incorporated are transient photoconductivity measurements to analyse the nature of decay process taking place in present glasses.

In order to make comparison, the photoconduc-

tivity results for one particular temperature or intensity are plotted in the figures. The results at other intensities or temperatures were found to be of the same nature. Further included are the temperature dependence of dark current.

Experimental.

Glasses of different compositions of $\text{Ga}_{40}\text{Se}_x\text{Te}_{60-x}$ system were obtained in bulk form by usual quenching technique. Elements of 5 nines purity were sealed in evacuated quartz ampules in desired stichiometric ratios. The ampules were then placed in an electric furnance with rocking arrangement and temperature was raised upto 1 000 °C. The ampules were then kept at ~ 1 000 °C for 10 h under continuous rocking and were then directly quenched into ice cooled water. The DSC measurements confirmed the glassy nature of the alloys and the glass transition temperature (T_g) values for the samples are given in table I.

Table I. — Glass transition temperature (T_g), conductivity (σ), pre-exponential factor (σ_0) and activation energy for different compositions.

Material	T_g (°C)	Dark conductivity (σ) at 325 K, $\Omega^{-1} \text{ cm}^{-1}$	σ_0 $\Omega^{-1} \text{ cm}^{-1}$	ΔE
$\text{Ga}_{40}\text{Se}_{40}\text{Te}_{20}$	181	8.46×10^{-7}	0.90	0.38 eV
$\text{Ga}_{40}\text{Se}_{30}\text{Te}_{30}$	161	1.26×10^{-6}	0.042	0.29 eV
$\text{Ga}_{40}\text{Se}_{20}\text{Te}_{40}$	153	10.05×10^{-6}	2.47×10^{-3}	0.22 eV

Thin films of different glasses were obtained from the bulk material by vacuum evaporation in a standard coating unit. Well degassed glass plates with high purity indium deposited onto them by vacuum evaporation were used as a substrates for depositing amorphous thin films. The coplanar geometry (length ~ 2 cm and electrode gap ~ 0.2 mm) so obtained was used for photoconductivity measurements. For the sake of comparison of results, the thickness of the amorphous films and other deposition parameters were kept about constant.

Thin film samples were annealed in vacuum at 373 K for about two hours. The incident radiation used for photoconductivity measurements was obtained from a 200 W tungsten lamp. The photoconductivity measurements were carried out under a vacuum of 10^{-3} Torr. The effect of monochromatic radiations obtained by using interference filters in the range 420-660 nm was observed and the results were found to be of the same nature as with the white light. The reported results are with white light

owing to better photon flux. Proper care was taken to avoid the thermal effects of incident radiation.

The voltage applied across the samples was 1.5 V obtained by a dry cell. The samples were however found to be ohmic upto 30 V in the absense as well as in the presence of light. The current measurements were accurate upto 1 pA. Temperature measurements were done by a calibrated copper-constantan thermocouple attached to a 3 1/2 digital temperature indicator and were accurate upto 0.1 °C.

Results and discussion.

Temperature dependence of dark current of different compositions of $\text{Ga}_{40}\text{Se}_x\text{Te}_{60-x}$ system was observed in the temperature range 310-373 K. Figure 1 represents the values of I_d (dark current) plotted logarithmically against $1000/T$ for thin films of the system with $x = 20, 30$ and 40 %. In I_d vs. $1000/T$ plots are straight lines for all the samples and show that dark conductivity in these samples is an activated process having a single activation energy (ΔE) in the observed temperature range. The conductivity of the samples decreases as Se content increases. Similar behaviour in the dark conductivity has been reported in As-Se-Te glasses [19] with the variation of Se : Te ratio though the preexponential factor σ_0 shows different behaviour in the two systems. Values of σ_0 and ΔE were computed and are given in table I. It is found that in present samples σ_0 decreases with increase in Te percentage.

Fig. 1. — $\ln I_d$ (dark current) against $1000/T$ for $\text{Ga}_{40}\text{Se}_x\text{Te}_{60-x}$ system ($x = 20, 30, 40$).

In chalcogenide glasses σ_0 in the range $10^3\text{--}10^4\text{ }\Omega^{-1}\text{cm}^{-1}$ indicates the conduction in extended states, whereas a lower value of σ_0 is an indication of a wide range of localized states [20]. From the results of table I it seems that increasing Te content leads to an increase in the density of localized states.

The temperature dependence of photocurrent was observed at various illumination intensities in all samples. Figure 2 represents the variation of the photocurrent $\ln(I_{ph})$ against $1000/T$ at three different illumination intensities for Ga₄₀Se₃₀Te₃₀ glass. Figure 2 shows that photoconductivity is an activated process and that the activation energy for photoconduction is almost the same for the three levels of illumination.

Fig. 2. — $\ln I_{ph}$ against $1000/T$ for Ga₄₀Se₃₀Te₃₀ glass at three levels of illumination.

Photocurrent does not reach a maximum with temperature in the explored temperature range in any of the samples. Absence of maxima in photoconductivity has been predicted in amorphous semiconductors [20] if photogenerated carrier densities are less than in the dark ($I_{ph} < I_d$) at room temperature. Absence of maxima w.r.t. temperature for $x = 20$, and $x = 30$ samples is in agreement with the above prediction as $I_{ph} < I_d$ in these samples. Similar absence of maximum has been reported by Shiah and Bube [21] in a number of glassy alloys. Though $I_{ph} > I_d$ at room temperature for $x = 40$ sample, yet no maximum could be observed in this sample. A behaviour similar to that of Ga₄₀Se₄₀Te₂₀ has been reported for a number of glasses [8, 21, 22] where photocurrent is greater than dark current at room temperature. Activation energy for photoconduction was computed for different x values and are given in table II.

Table II. — Various photoconductivity parameters for different compositions.

Material	γ at 310 K	I_{ph}/I_d at 310 K	ΔE_{ph}	τ at 310 K (s)
Ga ₄₀ Se ₄₀ Te ₂₀	0.75	1.42	0.12 eV	69
Ga ₄₀ Se ₃₀ Te ₃₀	0.96	0.88	0.11 eV	71
Ga ₄₀ Se ₂₀ Te ₄₀	1.00	0.23	0.18 eV	65

Intensity dependence of photoconductivity has been studied for all the samples. Values of $\ln I_{ph}$ against $\ln F$ (A.U.) are plotted in figure 3 for different samples at 310 K. Figure 3 shows that photoconductivity follows power law on incident radiation ($\sigma_{ph} \propto F^\gamma$). Values of γ were obtained from the slope of the curves in figure 3 and are inserted in table II. Value of γ seems to increase as Te content is increased in present system. The value of γ (Tab. II) for $x = 20, 30$ is nearly one (~ 1.0) indicating that the recombination is monomolecular.

Fig. 3. — $\ln I_{ph}$ against $\ln F$ (A.U.) for different x values in the Ga₄₀Se_xTe_{60-x} system.

For the sample with $x = 40$, the γ has dropped to 0.75. According to Rose [23] the value of γ between 0.5 and 1.0 can be justified by assuming the existence of continuous distribution of states in the band gap and the resulting recombination will be bimolecular. Arene and Baixeras [24] and others [17, 25] have observed the values of γ lying between 0.5 and 1.0 in different amorphous materials which has been attributed to the distribution of localized states in the energy gap. This behaviour is also quite understandable in the light of the relative values of photocurrent and dark current. For $x = 40$, the photocurrent is

larger than the dark current and thus the recombination has to be bimolecular [20] whereas, for $x = 20, 30$ photogenerated carriers are less than dark carriers ($I_{ph} < I_d$, see Tab. II) and recombine into the pool of thermally produced recombination centres and thus monomolecular recombination takes place [20].

I_{ph}/I_d for different x values calculated at 310 K and given in table II show that I_{ph}/I_d decreases with increasing Te content. This parameter depends on the life time of photogenerated carriers, which in turn depends upon the density of localized states in the energy gap. Higher density of localised states leads to lower value of carrier life time resulting in low value of I_{ph}/I_d [25]. A similar effect of Te content on the localized states concentration has been observed by us during dielectric measurements of these samples reported elsewhere [26].

Transient photoconductivity measurements were made on all the samples. Rise and decay of photocurrent was observed by using incident radiation of fixed intensity (170 A.U.) and is found to be quite slow in all the samples. In a material with traps in the energy gap, the recombination time of carriers is same as carrier life time in the case when free carrier density is much more than the trapped carrier density [27]. If the density of free carriers is much less than the trapped carriers, the resulting recombination is effectively dominated by the rate of trap emptying and is much larger than the carrier life time, resulting in a slow decay. A comparison of the results in figure 4 indicates that the rise and decay of photocurrent for $x = 40$ sample is faster as compared to other x values.

Figure 4 shows that there is an increment in the value of current in all the samples even after a long time when the incident light was cut off. It is believed that this increment may not be simply due to carriers trapped in localized states [28]. For simplification of the decay analysis, persistent current was subtracted from the observed values and the corrected values of I_{ph} (decay) are plotted logarithmically against time (t) for different x values in figure 5.

In disordered materials presence of traps plays an important role in the photoconductivity through trap filling and emptying processes. The traps which are filled during excitation will empty, at a rate depending upon their cross-section and ionization energy. If the retrapping of carriers freed from traps can be neglected then an exponential decay is expected [27] as guided by the relation $p \sim p_0 \exp(-t/\tau)$ where τ is the decay time constant. Results in figure 5 show that decay of photocurrent is exponential in all the samples except near the onset of transients where recombination is faster and this fast component goes on increasing with increasing x values.

Using the plots in figure 5, decay time constant

Fig. 4. — Rise and decay of photocurrent (expressed in I_{ph}/I_d) with time (t) for different x values at 310 K and at intensity 170 A.U.

Fig. 5. — $\ln I_{ph}$ (decay) against time (t) for different x -values.

(τ) was calculated for different x values and are given in table II. Value of decay time constant (τ) does not show a regular dependence on x values. Similar behaviour has been observed in the macroscopic relaxation time values (τ_0) obtained during the electric measurements [26].

To conclude, photoconductivity with respect to

temperature is an activated process in all the samples. There is power law dependence of photocurrent ($I_{ph} \propto F^\gamma$) on incident radiation. I_{ph}/I_d values calculated for different samples increase with increasing x values. This behaviour is consistent with the dark

conductivity observations which indicate the increasing number of defect states with increasing Te %. Similar effect of increasing Te % in the present system has been observed by us during the dielectric measurements [26].

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