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Influence of temperature on charge transports and optical parameters for the $Ge_{15}Sb_5Se_{80}$ and $Ge_{15}Sb_5Te_{80}$ amorphous thin films

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The thin films of chemical composition $Ge_{15}Sb_5Se_{80}$ and $Ge_{15}Sb_5Te_{80}$ were prepared by a thermal evaporation technique. Both the absorbance and the transmittance spectra were measured over the incident photon energy ranges 1.14 to 2.7 and 1.22 to 2.87 eV for the $Ge_{15}Sb_5Se_{80}$ and $Ge_{15}Sb_5Te_{80}$ films respectively. These were used for estimating the reflectance spectra in the same range of the incident photon energy. Both the measured and estimated spectra were carried out in the temperature range 250 to 330 K. With the aids of these spectra, the temperature dependence of charge transports and optical parameters were investigated and analyzed. Using the reflectance spectra, the refractive indices for the $Ge_{15}Sb_5Se_{80}$ and $Ge_{15}Sb_5Te_{80}$ films were determined in the low energy region of the studied incident photon energy range. These refractive index values were plotted as a function of the long wavelengths for the completely investigated range of temperature. These plots were used for elucidating the temperature dependence of both the oscillator and dispersion energies of the refractive index in the $Ge_{15}Sb_5Se_{80}$ and $Ge_{15}Sb_5Te_{80}$ thin films. Thereafter, static refractive index and static dielectric constant were determined. Also the temperature coefficients of the optical band width were determined and discussed for the two mentioned films.

Key words: Optical transports in chalcogenide semiconductors, disordered solids, optical parameters.

INTRODUCTION

Chalcogenide materials were found to exhibit the change in the refractive index under the influence of light, which makes it possible to use these materials to record not only the magnitude but also the phase of illumination. The later is important especially in holographic optical data storage and in the fabrication of various integrated components and devices such as selective optical fiber, mixers, couplers and modulators (Pandey et al., 2005). The optical band gap, refractive index and extinction coefficient are the most significant parameters in the amorphous semi-conducting thin films. It has also been reported that accurate measurements in studying the photo-electrical properties and optical parameters of a material should be carried out for its thin films (Pandey et al., 2005). Impurity effect in chalcogenide materials may have an importance in the fabrication of amorphous semiconductor devices. The most important applications of chalcogenides are now in the field of optics and arise mainly from either their exhibited infrared transmitting properties or photo-induced effects. They have potential uses in integrated optics, optical imaging, optical data storage and infrared optics (Sanghara and Agarwal, 1999; Schwarts, 1993; Pradley, 1989; Tai et al., 1982).

The structure of the thin films strongly influences the electronic properties and is highly dependent upon the preparation technique and deposition conditions (Pathinettan et al., 2000).

The study of the Ge–Sb–Te system has received attention because of the material's use in reversible phase change applications. These applications exploit differences in optical (Noboru et al., 1991) or electrical (Mamon et al., 2003) properties between the crystalline and amorphous phases of the same material. In line with this study, that report the optical changes involved with the Ge–Sb–Te alloys' phase transitions, Yamada et al. found that the change in optical transitivity associated with the tran-

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sitions is an increasing function of over-coordinated Te. Generally, selenide glasses are more stable as compared to telluride glasses. Glasses containing tellurium have worse glass forming ability, smaller glass forming region, lower transmission, and higher refractive index.

They also have higher tendency to crystallization and phase separation (Zavadil et al., 2008). In this work, the obtained $Ge_{15}Sb_5Se_{80}$ and $Ge_{15}Sb_5Te_{80}$ thin films were found to be transparent, where in color, these films exhibits orange and gray colors respectively. The gray films showed lower transmittance spectra than the orange films. The temperature dependence of the photoelectrical transports and optical parameters in these thin films were elucidated in the temperature range 250 - 330 K. In addition, the impacts of the Se and Te contents were discussed in the completely investigated range of temperature.

EXPERIMENTAL SETUP

The $Ge_{15}Sb_5Se_{80}$ and $Ge_{15}Sb_5Te_{80}$ amorphous thin films of the same thickness (1µm) were prepared by using a thermal evaporation technique. Under this method, starting compounds were evaporated and collimated under vacuum (~ 10^{-6} torr) onto cleaned glass substrates. Powders of the starting materials in their amorphous states were used for evaporating the above-mentioned films. The glass substrates were, sequentially, carefully cleaned using hot water, NaOH solution, chromic acid, distilled water and isopropyl alcohol. Thermal evaporation was undertaken using Edwards E306A coating unit. The unit has attached its own two-stage rotary pump /diffusion pump system. For each sample under preparation, a molybdenum boat is charged with the starting compounds in granular form and stoichiometric ratio, and the vacuum chamber is pumped down to about ~ 10^{-6} torr. Under a shutter, the boat is gradually heated until the contained material starts to evaporate. At this point the shutter is removed, allowing the vapor to coat the glass substrates. The obtained samples that used in the present measurements were mounted on the cold finger inside a cryostat "Oxford DN1704-type", which was evacuated to about 10^{-4} torr. A digital temperature controller "Oxford ITC601-type" controlled the temperature inside the cryostat. A Shimadzu model UV-1650PC UV-visible double beam spectrophotometer was employed to record the reflectance and transmittance spectra over the incident photon energy ranges 1.14 to 2.7 and 1.22 to 2.87 eV for the $Ge_{15}Sb_5Se_{80}$ and $Ge_{15}Sb_5Te_{80}$ films respectively. With the aids of these spectra, the absorption coefficient, extinction coefficient and refractive index were estimated.

RESULTS AND DISCUSSION

The optical band gap, refractive index and extinction coefficient are the most significant parameters in amorphous semi-conducting thin films. So, an accurate measurement of the optical constants is extremely important. Chalcogenide glasses have been found to exhibit the change in refractive index under the influence of light, which makes it possible to use these materials to record not only the magnitude but also the phase of illumination. The latter is especially important in holographic optical data storage and in the fabrication of various integrated components and devices such as selective optical filters, mixers, couplers and modulators (Andriesh et al., 1996; Kityk et al., 1995; Wasylak et al., 1999. The photoelectrical transports in the $Ge_{15}Sb_5Se_{80}$ and $Ge_{15}Sb_5Te_{80}$ thin films and their optical parameters were investigated and discussed in this part with the aids of the optical measurements. The transmittance and reflectance spectra were measured for the $Ge_{15}Sb_5Se_{80}$ and $Ge_{15}Sb_5Te_{80}$ thin films in the temperature range 250 to 330 K. In the mentioned spectra, the ranges 1.14 to 2.7 and 1.22 to 2.87 eV of the incident photon energy was covered for the $Ge_{15}Sb_5Se_{80}$ and $Ge_{15}Sb_5Te_{80}$ thin amorphous films respectively. Figures 1a, 1b, 2a and 2b suggest typical behaviors in the completely investigated range of temperature, but there is a considerable effect of temperature on the above-mentioned films. As the optical measurements are productive tools for understanding the band structure, energy gap width and optical parameters of both crystalline and amorphous non-metallic materials, the optical absorption coefficients α of the films under investigation can be calculated in terms of the transmittance spectra T by using the following relation:

$$\alpha = \frac{1}{d} \, \mathrm{Ln}\left(1/T\right) \tag{1}$$

In accordance with this relation, the α – E was described for the investigated films (see figures 3a and 3b). The values of the absorption coefficient were calculated for the whole investigated temperatures and over the investigated ranges of the incident photon energy 1.14 to 2.7 and 1.22 to 2.87 eV respectively for the $Ge_{15}Sb_5Se_{80}$ and $Ge_{15}Sb_5Te_{80}$ films. Figure 3b suggests that the absorption coefficient of the $Ge_{15}Sb_5Te_{80}$ films exhibits a long band tail at the long wavelength region of the incident photon energy range. This week absorption tail is most probably originated from defects and impurity states within the band gap. Tailing of the band states into the gap width may be induced from a large concentration of free carriers resulting from screened Coulomb interaction between carriers that perturbs the band edges. Thereafter it steeply increases with increase in the photon energy near the fundamental edge. But in the $Ge_{15}Sb_5Se_{80}$ films, tailing of the band states into the gap width may be induced from a small concentration of free carriers resulting from screened Coulomb interaction between carriers that perturbs the band edges, where there is no absorption tail exhibited in figure 3a. In the region of the photon energy, that has values greater than those of the exponential edge region do, the absorption coefficient linearly increases with increasing the incident photon energy (high absorption region) for both the amorphous films under investigation.

Chalcogenide semiconductors containing Ge – Se and Ge – Te have good glass formation ability over a wide compositional range (Esquerre et al., 1978; Gokhale et al., 1990), which make them good candidate as arsenic free IR transparent. The common feature of chalcogenide glasses is the presence of localized states in the m obility gap due to the absence of long-range order as well as

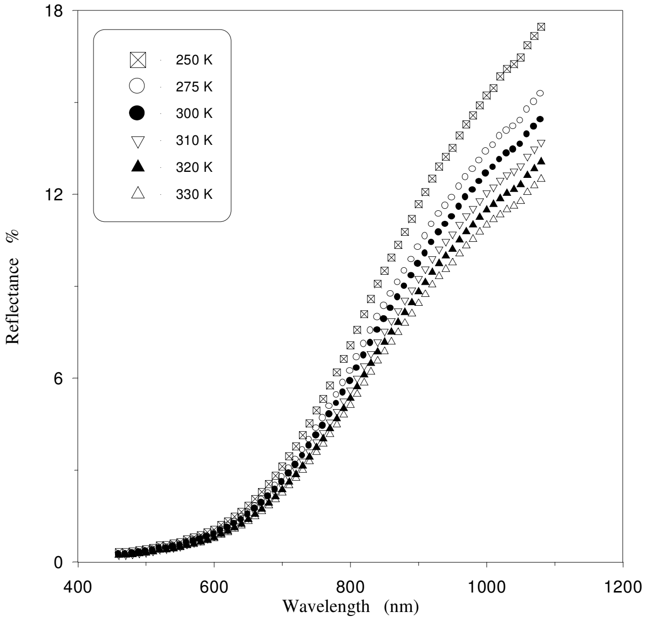

Figure 1a

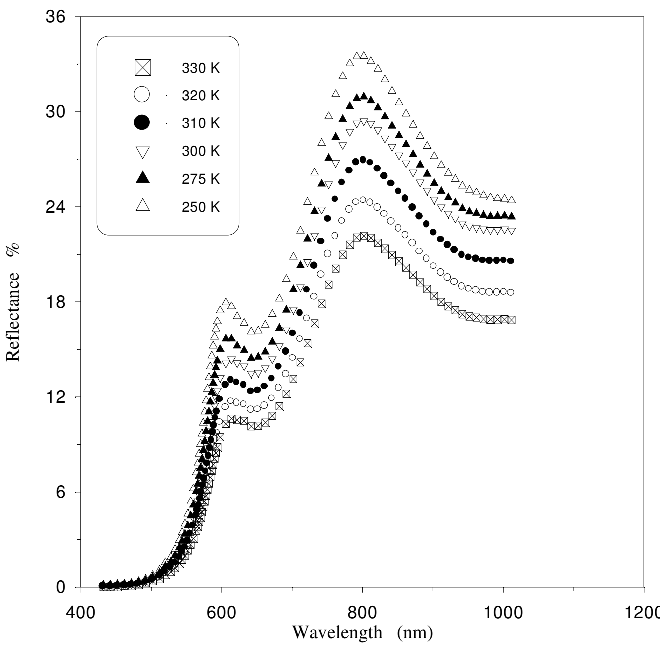

Figure 1b

Figure 1. The temperature dependence of transmittance spectra for the investigated films.

as various inherent structural defects. Films with non-stoichiometric Ge–Sb–Te compositions are of practical interest, and their optical and structural properties have recently become subject of intensive studies. It has been shown that for non-stoichiometric Ge–Sb–Te glasses with constant amount of Te atoms the thermal stability depends on the amount of Sb atoms (Zaluska et al., 1980), while the glass transition temperature decreases by decreasing the coordination number of these glasses (Belhadji et al., 1997). Still, it is an open question how the structural and optical properties of non-stoichiometric Ge–Sb–Te chalcogenide glasses and thin films change by illumination. In this work the optical properties of films with non-stoichiometric $Ge_xSb_{20-x}Te_{80}$ composition are studied. The above mentioned relationship between the absorption coefficient and the incident photon energy ($h\upsilon$) in the high absorption regions (linear increase of α with increase in incident photon energy) is governed by the relation (Pankove, 1971; Tauc, 1974):

$$\alpha = \frac{1}{h\upsilon}\left[A\left(h\upsilon - E_g\right)^m\right] \quad (2)$$

Where, A is constant depends on the transition probability, E_g is the width of the optical band gap, and m is an index that characterizes the optical absorption processes in the investigated films. The analysis of experimental results showed that a proportionality is revealed between the absorption coefficient and the frequency of the photon energy in the form ($h\upsilon$ - E_g) m. The exponent m should be one of the four values: 2, 1/2, 3 and 3/2, where these values can define the type of the optical transition in the films under investigation. Theoretically, m is equal to 2, 1/2, 3 or 3/2 for the indirect allowed, direct allowed, indirect forbidden and direct forbidden transitions respectively (Qasrawi, 2005). On other way, usual method for determining the type of the optical transition includes plots of $(\alpha h\upsilon)^{1/m}$ versus the incident photon energy ($h\upsilon$). This proportionality gives a set of plots with four values of the exponent n: $(\alpha h\upsilon)^{1/2} - h\upsilon$; $(\alpha h\upsilon)^2 - h\upsilon$; $(\alpha h\upsilon)^{1/3} - h\upsilon$; and $(\alpha h\upsilon)^{2/3}$ - $h\upsilon$. One of these plots satisfies the widest linearity of data in high absorption region, and hence its exponent determines the type of the optical transition. In the cases under investigation ($Ge_{15}Sb_5Se_{80}$ and $Ge_{15}Sb_5Te_{80}$ films), the exponent m indicates that the dominant transition is direct allowed one. Consequently, $(\alpha h\upsilon)^2$ was plotted against ($h\upsilon$), and this graphs is illustrated in Figures 4a and 4b (also this Figure was plotted for all investigated temperatures in the photon energy ranges 1.14 to 2.7 and 1.22 to 2.87 eV for the $Ge_{15}Sb_5Se_{80}$ and $Ge_{15}Sb_5Te_{80}$ films respectively). In this work, the optical band gaps were calculated by linear fittings in the high absorption regions for the two films under investigation. This fitting intersects the $h\upsilon$ - axis at the value of the optical band gap width for the completely investigated temperatures. The estimated values of the gap widths were plotted against the values under investigation of temperature (see Figures 5a, 5b) for studying the temperature coefficient (dE_g /dT) of the optical gap in the $Ge_{15}Sb_5Se_{80}$ and $Ge_{15}Sb_5Te_{80}$ films. As the temperature increases (increase in the motion of the atoms broadens the energy levels), the lattice expands and the oscillations of the atoms around their equilibrium lattice points increase. Due to the electron-lattice interaction that depends strongly on temperature at temperatures much lower than the Debye temperature, the gap width varies proportionality to the square

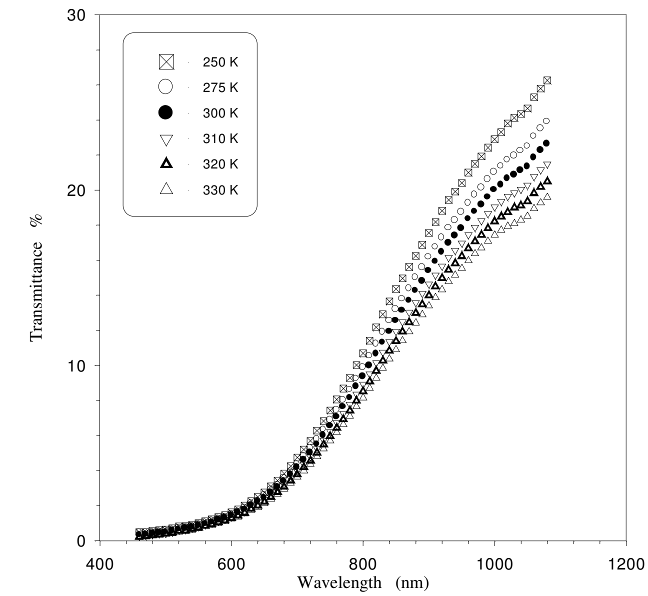

Figure 2a

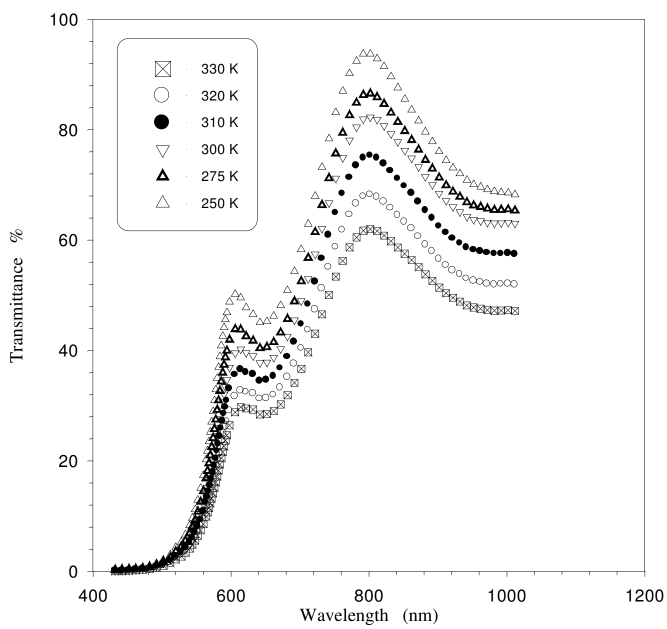

Figure 2b

Figure 2. The temperature dependence of reflectance spectra of the investigated films.

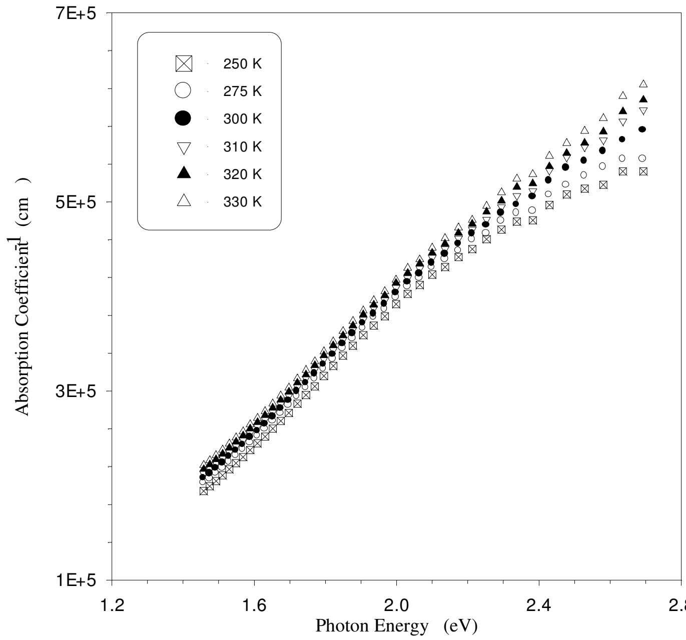

Figure 3a

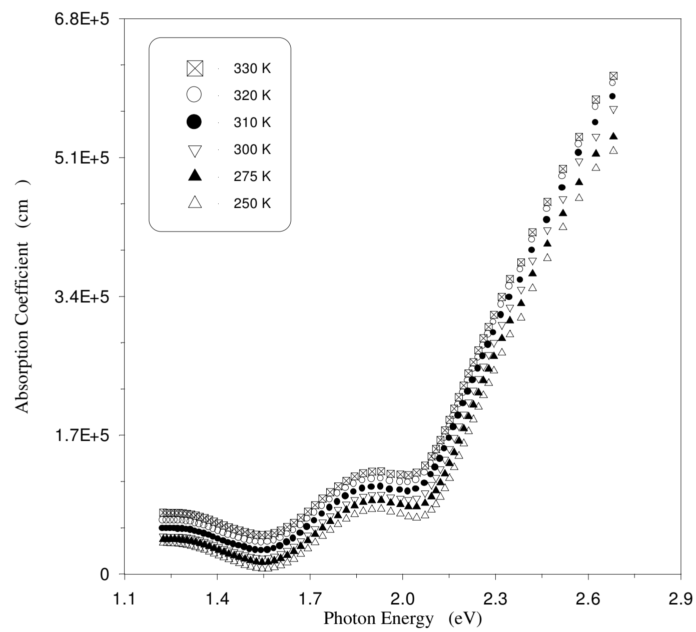

Figure 3b

Figure 3. The photon energy dependence of absorption coefficient in the investigated films.

of the temperature, whereas much above the Debye temperature the gap width varies linearly with the temperature (Pankove, 1971). Therefore, the following empirical relation has fit the temperature dependence of the band gap width for many semiconductors:

$$E_g\ (T) = E_g\ (0) - \left(\frac{\alpha T^2}{T+\beta}\right) \qquad (3)$$

Where, at 0°K, the optical gap E_g is denoted as E_g (0); and α, β are constants.

With the aids of the linear portions (high temperature regions) in graphs 5a and 5b, the temperature coefficients of the optical gap width were calculated for the $Ge_{15}Sb_5Se_{80}$ and $Ge_{15}Sb_5Te_{80}$ films to be ~ -4.8 x 10^{-3} and ~ -2.3 x 10^{-4} eV / K and similar errors respectively. These sug-

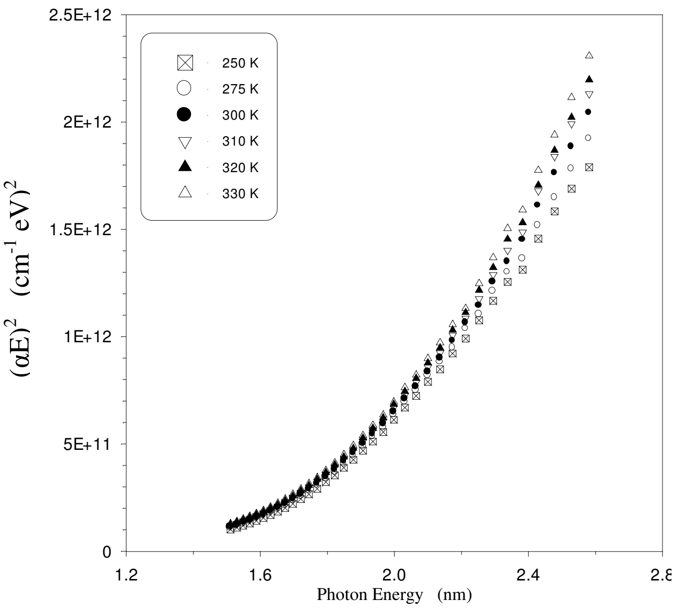

Figure 4a

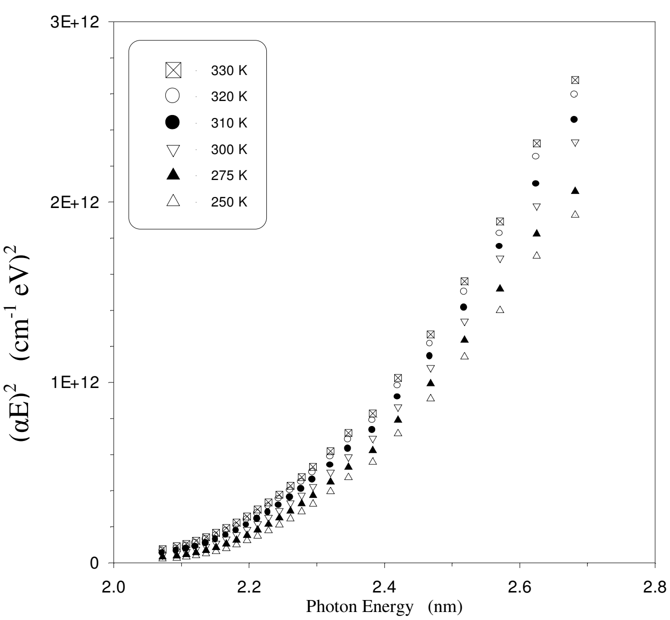

Figure 4b

Figure 4. Shows the quantity $(\alpha h\upsilon)^2$ as a function of $(h\upsilon)$ for the investigated films.

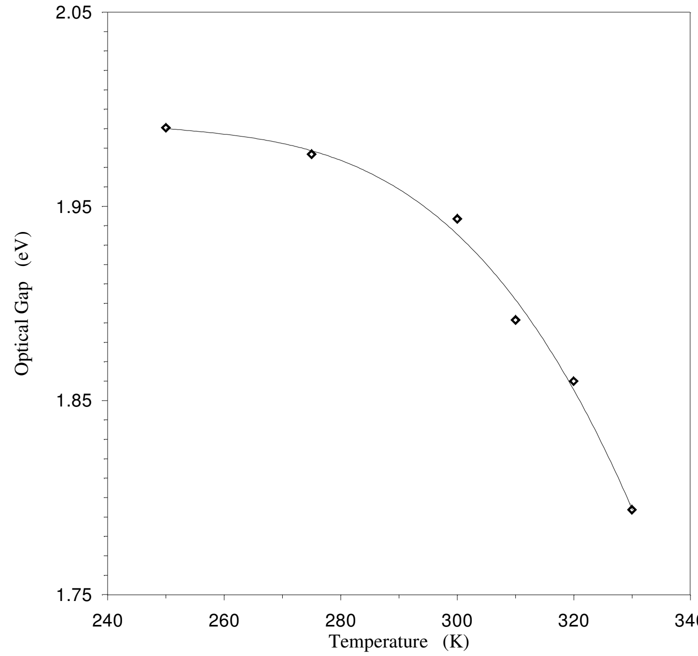

Figure 5a

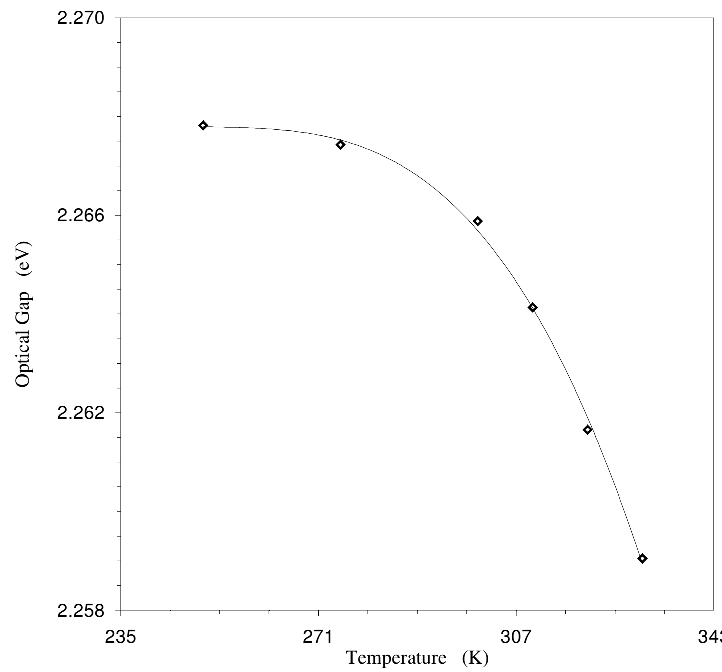

Figure 5b

Figure 5. Shows temperature dependence of the direct band gap in the investigated films.

gest that the optical bandwidth of the $Ge_{15}Sb_5Se_{80}$ films strongly depends on the variation in temperature than that of the $Ge_{15}Sb_5Te_{80}$ films. It should be emphasis here that different reasons could be responsible for the effect of temperature on the band-gap. Such explanation was suggested to elucidate the decreasing of the band-gap energy after illumination.

The first explanation could be due to increasing the number of homopolar bonds (Pfeiffer et al., G 1991), essentially Se-Se and Te–Te, at the expenses of heteropolar Ge–Se, Ge–Te, Sb–Se and Sb–Te ones. The second explanation based on the increasing the randomness with increasing the temperature. Since the increasing of randomness in the medium-range structural order accompanied by distortion of the bond, it is assumed (Pfeiffer et al., 1991; Nagels et al., 1998; Wong et al., 1987) that the distorted bonds give shallow localized states at the top of

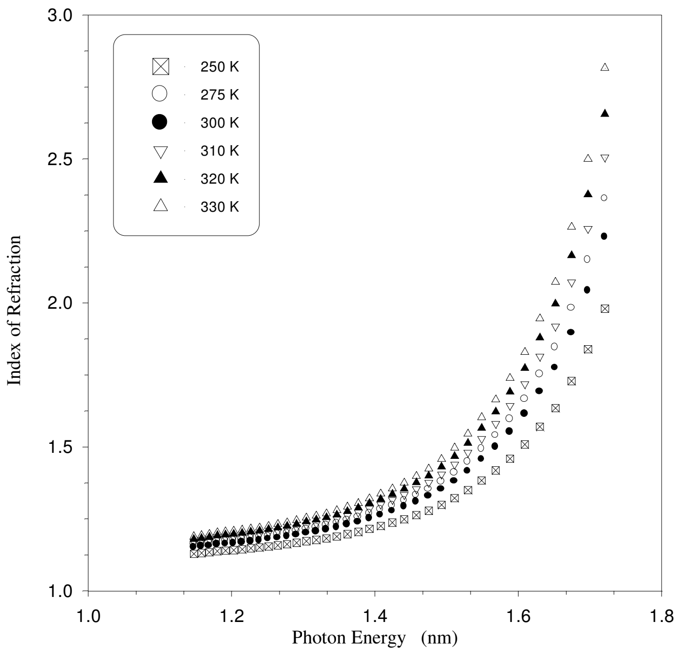

Figure 6a

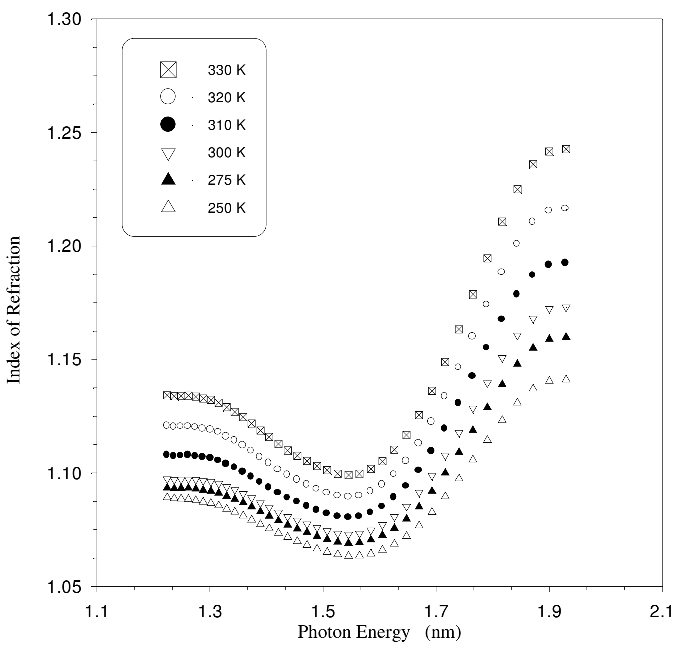

Figure 6b

Figure 6. Shows dependence of refractive index (n) on the incident photon energy.

valence band and the bottom of conduction band and, consequently, narrowing the forbidden gap width. Another explanation for the reduction of E_g value could be the generation of charged defects in the band-tail regions (Shimakawa et al., 1993), which leads to narrowing of the bandgap.

Due to the transparency of the films under investigation, the reflectance spectra can used for determining the refractive index n of these films over the whole investigated range of temperature with the aids of the following relation:

$$R = [(n-1)/(n+1)]^2 \qquad (4)$$

Using above calculated values of the refractive index were employed for plotting the n – E relation of the above mentioned amorphous thin films over the whole investigated temperatures in the whole investigated range of the incident photon energy (see Figures 6a and 6b). It is obvious from these figures that: in the long wavelength region of the investigated photon energy range, the dispersion of the refractive index is normal for the whole investigated temperatures under investigation and can well be illustrated using a single oscillator model (Baban et al., 2005):

$$n^2 = 1 + \frac{E_o E_d}{E_o^2 - (h\upsilon)^2} \Rightarrow (n^2-1)^{-1} = \frac{E_o}{E_d} - \left(\frac{1}{E_o E_d}\right)(h\upsilon)^2 \qquad (5)$$

Where, E_o is the single oscillator energy, E_d is the dispersion energy, and $(h\upsilon)$ is the incident photon energy. It is clear from Figures 6a and 6b that the refractive index linearly increases with the increase in the incident photon energy in the low photon energy region 1.14 to 1.72 eV for the $Ge_{15}Sb_5Se_{80}$ films and 1.22 to 1.93 eV for the $Ge_{15}Sb_5Te_{80}$ films. This behavior was normally repeated over the completely investigated temperatures. In addition, there is a considerable change in the refractive index through the variation of temperature for the two investigated films. Both the single oscillator energy (E_o) and the dispersion energy (E_d) can be obtained for the investigated temperatures by plotting (n2 – 1)-1 as functions of the photon energy $(h\upsilon)$ in the long wavelength region of the investigated range of the photon energy. The above-mentioned relation is depicted for the tow-studied films in Figures 7a and 7b. By using these figures, the E_d and E_0 were calculated from the slope $(E_0E_d)^{-1}$ and the intercept (E_0 / E_d). The calculated values of oscillator energies (E_o), dispersion energies (E_d), and optical band widths (E_g) for the $Ge_{15}Sb_5Se_{80}$ and $Ge_{15}Sb_5Te_{80}$ amorphous films in the temperature range 77 to 300K are summarized in Table 1. The static refractive index $[n(0) = (1 + E_d / E_0)^{1/2}]$ and the static dielectric constant $[\varepsilon_s = n^2(0)]$ of the $Ge_{15}Sb_5Se_{80}$ films were also calculated using the above reported relations and found to be ~1.103 and ~1.216 respectively. The above-mentioned parameters were calculated for $Ge_{15}Sb_5Te_{80}$ films to be ~1.024 and ~1.049 respectively. The Data recorded in Table 1 suggests that with the increase in temperature, there is overall decrease in both the optical band gap and dispersion energy of the $Ge_{15}Sb_5Se_{80}$ films, where there is overall increase in the oscillation energy. However, with the increase in temperature, there is overall decrease in both the optical band gap and oscillation energy of the $Ge_{15}Sb_5Se_{80}$ films, where there is overall increase in the dispersion energy

Table 1. Optical parameters of the investigated amorphous films.

Temperature (K)	Optical gap (EV)		Oscillation energy (EV)		Dispersion energy (EV)	
	$Ge_{15}Sb_5Se_{80}$	$Ge_{15}Sb_5Te_{80}$	$Ge_{15}Sb_5Se_{80}$	$Ge_{15}Sb_5Te_{80}$	$Ge_{15}Sb_5Se_{80}$	$Ge_{15}Sb_5Te_{80}$
250	1.99	2.268	1.656	2.008	0.359	0.099
275	1.977	2.267	1.659	1.995	0.341	0.107
300	1.944	2.266	1.663	1.985	0.323	0.111
310	1.891	2.264	1.669	1.981	0.286	0.122
320	1.859	2.262	1.666	1.976	0.304	0.135
330	1.794	2.259	1.675	1.972	0.249	0.149

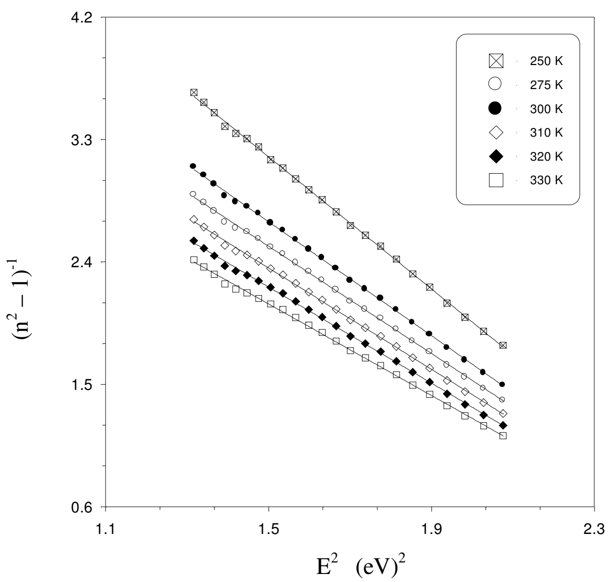

Figure 7a

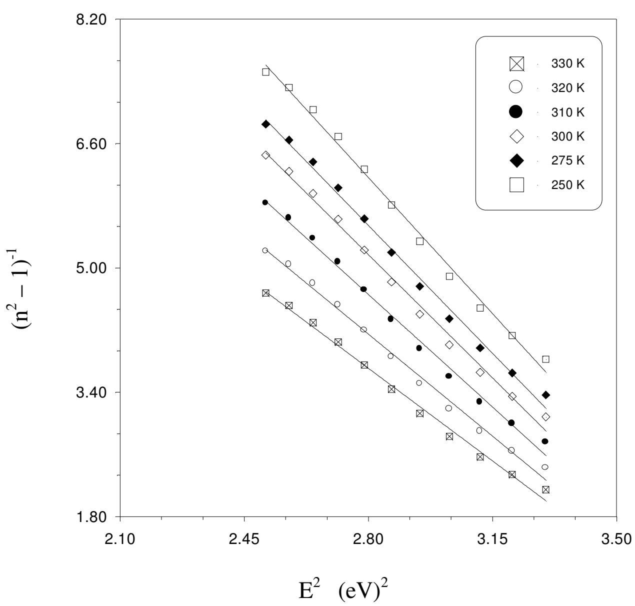

Figure 7b

Figure 7. Shows quantity (n¬2 - 1)-1 as a function of (hυ) 2 for the investigated films.

Conclusion

the photoelectrical transports and optical parameters were investigated with the aids of the transmittance and reflectance spectra of the $Ge_{15}Sb_5Se_{80}$ and $Ge_{15}Sb_5Te_{80}$ thin films over the temperature range 250 to 330 K in the incident photon energy ranges 1.14 to 2.7 and 1.22 to 2.87 eV for the $Ge_{15}Sb_5Se_{80}$ and $Ge_{15}Sb_5Te_{80}$ films respectively. In accordance with the α – E relation, the Ge_{15} Sb_5Te_{80} thin films exhibit a long band tail at the low energy region, which is most probably originated from defects and impurity states within the band gap.

Tailing of the band states into the gap width may be induced from a large concentration of free carriers resulting from screened Coulomb interaction between carriers that perturbs the band edges. But in the short wavelength region, the absorption coefficient linearly increases for the $Ge_{15}Sb_5Se_{80}$ and $Ge_{15}Sb_5Te_{80}$ films with increasing the incident photon energy. Direct allowed transitions were observed for the two investigated films. The direct allowed gaps of the $Ge_{15}Sb_5Se_{80}$ and $Ge_{15}Sb_5Te_{80}$ films were estimated for the whole investigated temperatures and was found to be ~ 1.944 and 2.266 eV at room temperature for the above mentioned films respectively. In addition, the temperature coefficients of the optical gap were determined for the $Ge_{15}Sb_5Se_{80}$ and $Ge_{15}Sb_5Te_{80}$ films to be ~ -4.8 x 10-3 and -2.3 x 10-4 eV / K and similar errors respectively, which suggests that the optical bandwidth of the $Ge_{15}Sb_5Se_{80}$ films strongly depends on the variation in temperature than that of the $Ge_{15}Sb_5Te_{80}$ films. As results of the linear proportionality of the refractive index for the two investigated films to the long wavelength region of the incident energy, all of the oscillator E0, dispersion Ed energies of the refractive index, static refractive index and static dielectric constant were calculated in the investigated range of temperature. These showed that there are overall increase and decrease in Eo and Ed respectively with the increase in temperature for the Ge_{15} Sb_5Se_{80} films, and the opposite was observed for the $Ge_{15}Sb_5Te_{80}$ films.

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