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CODEN: IAASCA **ORIGINAL ARTICLE**

Optical changes in amorphous $As_{40}Se_{60}$ thin films with Ge addition

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ABSTRACT
Amorphous thin films of glassy alloys of $Ge_xAs_{40}Se_{60-x}$ (x = 0, 15, 20) are prepared by thermal evaporation technique on clean glass substrates at room temperature. Structural and compositional analysis is done using XRD and EDAX measurements. Optical band gap, linear optical absorption coefficient, extinction coefficient and linear refractive index are evaluated using optical transmission spectra in wavelength range 300-900 nm. Explanation for variation in optical properties is based on structural changes and density of defect states.
Key words: *Amorphous Thin Film, Optical absorption, Optical band gap, Linear refractive index*

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INTRODUCTION

Amorphous chalcogenides, which are based on one or more chalcogen elements (S, Se, Te) are promising elements for various photonic and optical applications due to their well-known properties such as photosensitivity, high third order optical non-linearities, IR transparency, high refractive index etc [1]. $As_{40}Se_{60}$ is one of the widely studied materials of chalcogenide glassy system [2-4]. Addition of Ge in $As_{40}Se_{60}$ system transforms it from floppy mode to rigid mode [5]. Such change in structural configuration cause change in optical properties of the system, which makes possible to alter optical properties according to desired technological applications. Lot of study was made on Ge-As-Se system because of its broad glass formation region [5-8]. Ge-As-Se thin films are frequently used for various active and passive optical applications [1].

In present study, effect of Ge addition in amorphous $Ge_xAs_{40}Se_{60-x}$ system is observed on various optical properties using optical transmission spectra in wavelength range 300-900 nm.

EXPERIMENTAL

Glassy alloys of $Ge_xAs_{40}Se_{60-x}$ (where x = 0, 15, 20) were prepared by melt quenching technique. Materials (99.999% pure) were weighted according to their atomic percentages and be sealed in quartz ampoules in the vacuum of 10^{-5} Torr. The sealed ampoules were kept inside a furnace, where the temperature raises to 950°C at the rate of 3-4°C per minute. The ampoules were frequently rocked for 10 hours at a maximum temperature to make the melt homogenous, and the quenching was done in ice water. Thin films of glassy alloys of thickness 260 - 1225 nm were prepared by thermal evaporation technique using rotating substrate at room temperature. Thermal evaporation process was performed inside a coating system (HIND-HIVAC 12A 4D-T) at a pressure of ~ 10^{-6} Torr. The deposition rate was ~ 10 nm/sec. The target was rotated with a frequency of 5 Hz to produce uniform films. Substrates were chemically clean, circular microscope glass slides. Thickness of the films is measured by thickness profilometer (Tencore Instrument, Model Alpha Step 100). Glassy alloys as well as thin films were characterized using X-ray diffractometer (Thermo Electron Corporation, Model ARL X'TRA) at scan rate of 3 deg/min. No prominent peak was observed in XRD of bulk samples as well as thin films, which verifies their amorphous nature [6, 7]. The homogeneity and correct compositions of all the alloys and thin films were determined by means of scanning electron microscope (Zeiss, Model EVO 40). Film compositions were different from by bulk samples by up to ~ 3% [3, 6]. The optical transmission of $Ge_xAs_{40}Se_{60-x}$ thin films was measured by double

beam UV/Vis computerized spectrophotometer (Hitachi, Model U-3310) in wavelength range 300-900 nm.

RESULTS AND DISCUSSION

When a photon of known energy excites an electron from top of the valance band to bottom of the conduction band, then linear optical absorption phenomena takes place in the material. This phenomenon requires that energy of incident photon to be of the order of bandgap of the sample. Linear optical absorption of the material is represented by linear optical absorption coefficient (α), which can be evaluated from optical transmission spectra as [3]:

$$\alpha = (1/d) \ln(1/T) \quad (1)$$

where, T is optical transmittance and d is thickness of the film. In present study, thickness of $As_{40}Se_{60}$ thin film is 1225 nm and for other thin films are 260 nm.

The absorption spectra of amorphous semiconductors are different from crystalline semiconductors because of presence of localized states within band gap. Optical absorption coefficient (α) follows Urbach rule [9] over wide range of photon energy, according to which, absorption coefficient (α) increases exponentially with photon energy:

$$\alpha = \exp[C (h\nu - h\nu_0) / kT] \quad (2)$$

where, C is a small constant of the order of unity, ν_0 is a constant corresponding to lowest excitonic frequency.

In the high absorption region, α obeys Tauc's relation for indirect bandgap material [10], which is:

$$(\alpha h\nu)^{1/2} = B^{1/2} (h\nu - E_g) \quad (3)$$

where, B is band tailing parameter.

Optical bandgap (E_g) can be obtained by extrapolating the Tauc's plot for indirect band gap material, which is shown in Figure 1. Table 1 shows observed values of optical band gap for $Ge_xAs_{40}Se_{60-x}$ thin films and compared with the values given in the literature. There is a good agreement in both. Results show that optical band gap (E_g) increases upon addition of Ge content, which can be explained on the basis of Davis and Mott model [11]. According to this model, addition of Ge creates additional defect states inside the valance band and localized states within the band gap and change in width of localized states. As the width of localized states increases, value of optical band gap decreases or vice-versa. Calculation for width of localized states can be done according to L. Tichy et. al. [12]. Density of defect states and width of localized states are maximum for $As_{40}Se_{60}$ thin films, hence optical band gap is minimum for this sample. Extinction coefficient (k) represents attenuation of light in the sample due to both absorption and scattering process. It can be calculated from linear optical absorption coefficient using the relation:

$$k = \alpha\lambda/4\pi \quad (4)$$

Variation of extinction coefficient is shown in Figure 2. It is clear from this figure that extinction coefficient reduces with wavelength i.e. optical losses reduce with wavelength and with Ge addition. Hence, these thin films are good optical materials for long-wavelength region. Determination of linear refractive index (n_0) can be done using the relation [13, 14]:

$$[(n_0^2 - 1)/(n_0^2 + 2)] = 1 - (E_g/20)^{1/2} \quad (5)$$

Calculated values of linear refractive index are shown in Table 1. From this table, it is clear that the value of linear refractive index is reduced upon Ge addition from 2.85 to 2.79. The reason of reduction in linear refractive index is the consequence of increment in wrong bonds (homopolar bonds) in the system with Ge addition.

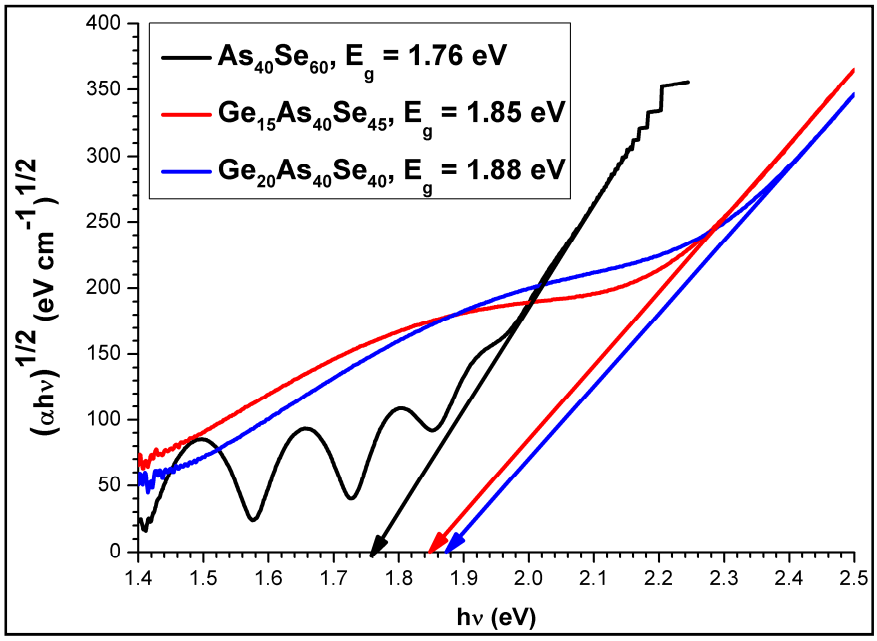

Fig.1. Tauc's plots for $Ge_xAs_{40}Se_{60-x}$ thin films

Table 1: Optical band gap and linear refractive index for $Ge_xAs_{40}Se_{60-x}$ thin films

Sample	Optical band gap (present study)(eV)	Optical band gap (literature)(eV)	Linear Refractive Index
$As_{40}Se_{60}$	1.76	1.76 [3]	2.85
$Ge_{15}As_{40}Se_{45}$	1.85		2.80
$Ge_{20}As_{40}Se_{40}$	1.88		2.79

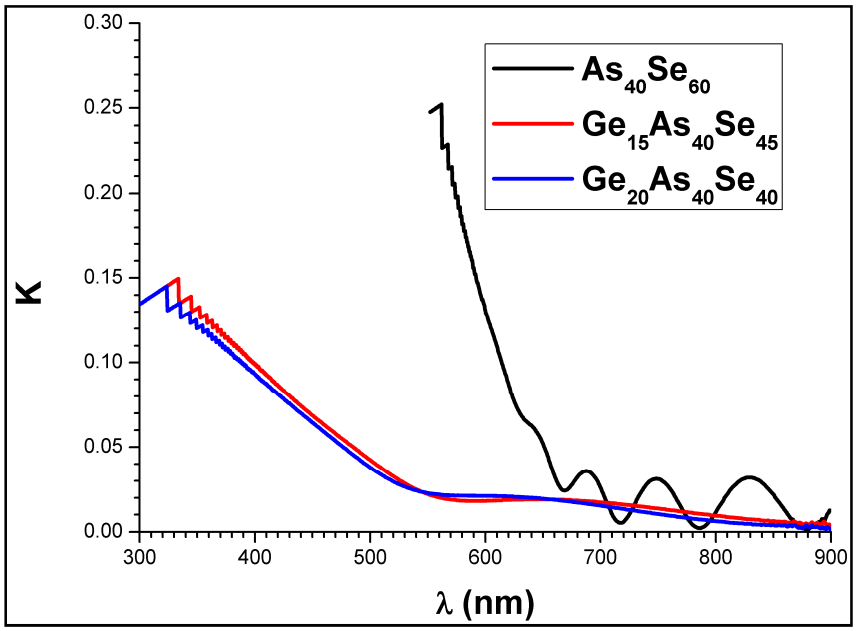

Fig.2. Plots of extinction coefficient for $Ge_xAs_{40}Se_{60-x}$ thin films

CONCLUSION

The optical band gap of $Ge_xAs_{40}Se_{60-x}$ thin films increases from 1.76 to 1.88 eV with Ge addition. It is also observed that linear refractive index reduces from 2.85 to 2.79, and extinction coefficient reduces with

wavelength and Ge addition. These changes in optical properties can be explained on the basis of structural changes and density of defect states.

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