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Influence of Thickness on Optical Properties of a: As_2Se_3 Thin Films

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Abstract

This paper reports optical properties of amorphous chalcogenide thin films of As_2Se_3 of different thicknesses 2000–6800 Å. The transmittance and reflectance of thin films were measured in the wavelength range 500–1000 nm. It was found that the optical band gap increases with film thickness. Variation of refractive index n and extinction coefficient k with thickness have been studied to analyze optical efficiency of the As_2Se_3 thin films. The dielectric behavior of the films has also been studied and high frequency dielectric constants ϵ_∞ has been estimated as a function of thickness of films.

Key Words: Chalcogenide, Thin films, Optical Constants.

1. Introduction

Selenium based chalcogenide glasses have extensive applications as electronic and optoelectronic device materials [1], due to their transparency in the IR region, good thermal, mechanical and chemical properties [2]. The high refractive index makes them suitable to be used as core materials for optical fibers for light transition in short wave length region. Se is promising material for large number of applications in xerography, photocells, switching and memory devices. However, in pure Se the life-time is short and the sensitivity is low. To achieve high sensitivity, high crystallization temperature and smaller ageing effect, Se is alloyed with impurity atoms such as Ge, Sb and As [3–6]. As_2Se_3 is a predominantly covalent solid, and one of the most extensively studied binary system [7].

Knowledge of optical constants of a material such as optical band gap, refractive index and extinction coefficient is quite essential to examine material's potential opto-electronic applications [8]. Further, the optical properties may also be closely related to the material's atomic structure, electronic band structure and electrical properties. An accurate measurement of the optical constant can be easily performed on thin film specimens.

The optical behavior of materials is important to determine it's usage in optoelectronic devices [9]. The electrical properties of semiconductors are strongly dependent upon the band gap [10, 11]. In the present work, we report the effect of thickness on optical properties of As_2Se_3 thin films. The optical band gap, refractive index, extinction coefficient and dielectric properties are significantly dependent on the film thickness. Dispersive nature of As_2Se_3 based chalcogenide glasses have also been analyzed by studying the variation of refractive index with wavelength.

2. Experimental Details

Thin films of As_2Se_3 with different thicknesses were prepared by vacuum evaporation technique at 10^{-6} Torr using a Hind Hivac Vacuum Coating Unit, Model No. 12-A4. Thin films were deposited on ultrasonically cleaned glass substrates (corning glass 7059) in the shape of squares, using appropriate masks, for optical measurements. The substrate was maintained at room temperature (300 K). The film thicknesses were controlled using a quartz crystal monitor (CTM-200 Vactech, India). The films were kept inside the vacuum chamber for 24 hours to attain metastable equilibrium. [12]. Transmission and reflection of the thin films were measured using a Shimadzu SolidSpec 3700 computerized spectrometer in the wavelength range of 500–1000 nm. The amorphous nature was investigated by the XRD patterns of the films using a Bruker Advance D8 x-ray diffractometer using CuK ($= 1.54056 \text{ \AA}$) radiation.

3. Result and Discussion

The XRD spectra shown in Figure 1 reveal the amorphous nature of As_2Se_3 films. Since no peaks were observed in the XRD pattern.

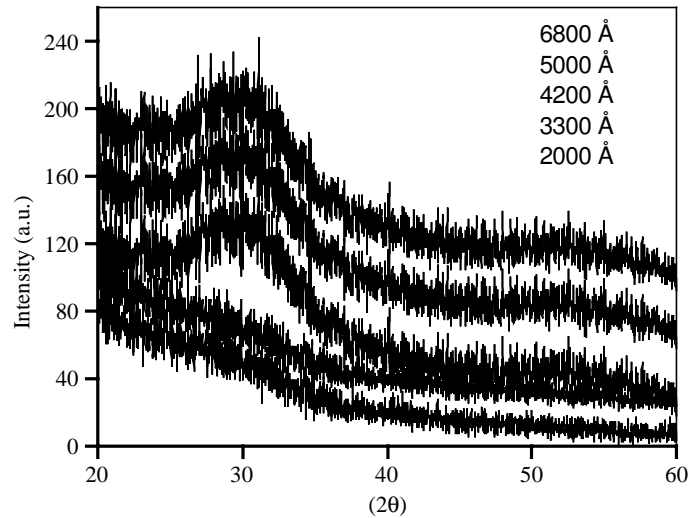


Figure 1. XRD spectra of As_2Se_3 thin films of different thickness.

Figure 2 shows the variation of transmission T with wavelength λ for different thicknesses of As_2Se_3 thin films. The observed fringe pattern of the transmission spectrum indicates that the films are homogeneous in nature. The interference fringe pattern can be analysed using Swanepoel's method [13], which is based on the envelope of the interference maxima and minima, appearing in the spectrum. The method provides basis for the calculation of optical constants of the material.

Figure 3 shows reflectance spectra of As_2Se_3 films with different thickness. It is observed that the reflectance decreases with increases of thickness.

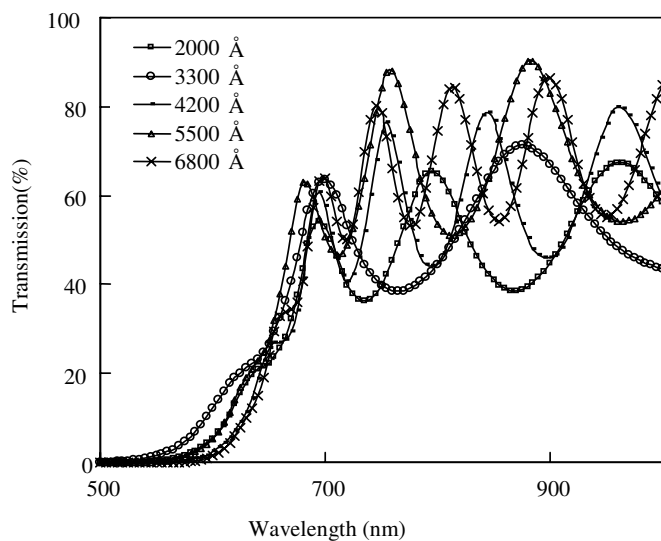


Figure 2. Variation of transmission spectra ($T\%$) of a- As_2Se_3 thin films of different thicknesses.

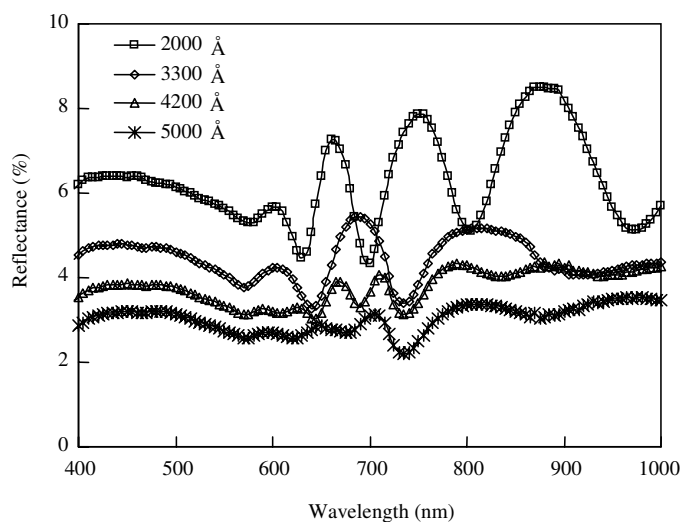


Figure 3. Variation of reflection spectra ($R\%$) of a- As_2Se_3 thin films of different thicknesses.

3.1. Determination of optical constants

The measurement of the absorption coefficient α as a function of frequency ν of the incident beam provides a mean to determine the band gap E_g of a material. The optical band gap in most of the amorphous semiconductors can be determined using the Tauc relation [14], which is expressed as

$$(\alpha h\nu) = B(h\nu - E_g)^r \tag{1}$$

where B is a constant and r is an index which depends on the nature of electronic transition responsible for the optical absorption. Values of r for allowed direct and non-direct transitions [14] are $1/2$ and 2 , respectively.

Figure 4 shows a Tauc's plot of the optical absorption spectrum measured at room temperature for as-deposited films of different thickness of As_2Se_3 thin films. The indirect optical energy gap can be obtained from the intercept of the resulting straight lines with the energy axis at $(\alpha h\nu)^{1/2} = 0$.

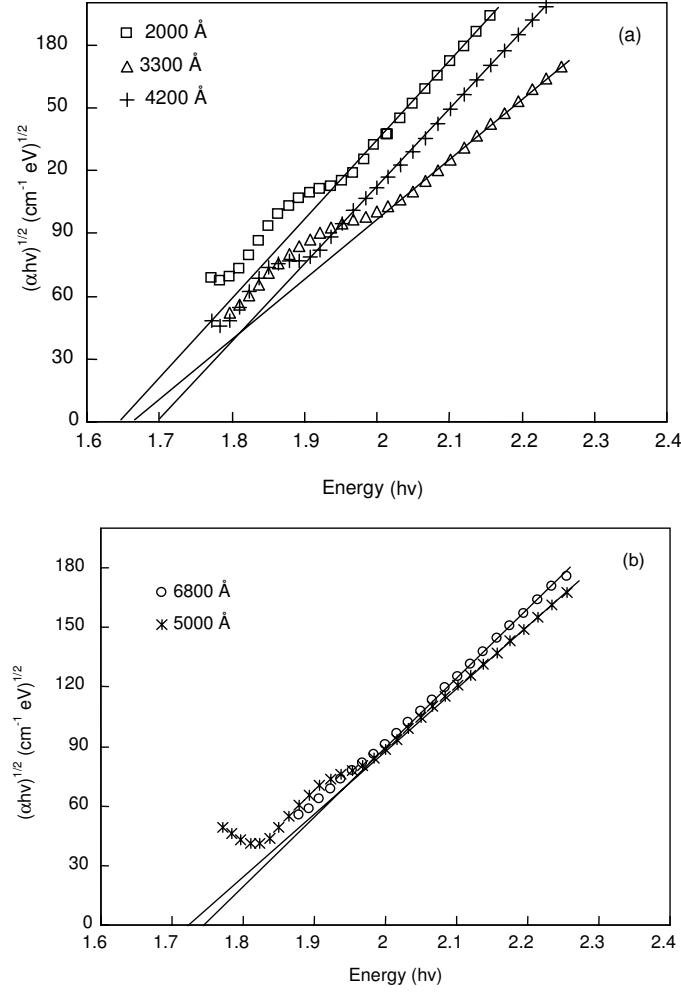


Figure 4. Variation of $(\alpha h\nu)^{1/2}$ versus photon energy ($h\nu$) for different thicknesses of a-As₂Se₃ thin films.

The values of optical band gap E_g are given in Table 1 for different thickness. It is clearly observed that the value of E_g increases with increasing thickness. The estimated values of the band gaps are similar to the value reported by Ponnampalam et al. [15].

It may be mentioned that, in amorphous chalcogenide thin films, the number of defects are higher due to the existence of unsaturated bonds [16]. The increase in the thickness of the films results in a homogeneous network with low density of defects [17] thereby, increasing the optical band gap.

The Refractive index n is deduced from the fringe patterns in the transmittance spectrum. In the transparent region, where the absorption coefficient $\alpha \approx 0$, and the refractive index n of the thin films is given by [13]

$$n = \left[M + (M^2 - S^2)^{\frac{1}{2}} \right]^{\frac{1}{2}}, \tag{2}$$

where M is given by

$$M = \frac{2S}{T_m} - \frac{S^2 + 1}{2}, \tag{3}$$

and S is the refractive index of the corning glass substrate and T_m is the minima in the transmission spectra. In the weak and medium absorption regions, where $\alpha \neq 0$, the refractive index n is given by

$$n = \left[N + (N^2 - S^2)^{\frac{1}{2}} \right]^{\frac{1}{2}}, \tag{4}$$

where

$$N = 2s \frac{T_M - T_m}{T_M T_m} + \frac{S^2 + 1}{2} \quad (5)$$

and T_M and T_m are the values of the envelope function at the wavelengths in which the transmittance is maxima and minima, respectively.

The extinction coefficient k can be calculated from the relation

$$k = \frac{\alpha \lambda}{4\pi}. \quad (6)$$

The spectral dependence of n and k for the different thickness of As_2Se_3 thin films are shown in Figures 5 and 6. It is observed that refractive index n and extinction coefficient k decreases with increasing thickness in the given wavelength range. Refractive index also decreases with the increase of wavelength. Variation in refractive index follows the normal dispersion law. The estimated value of refractive index is close to the reported value [15].

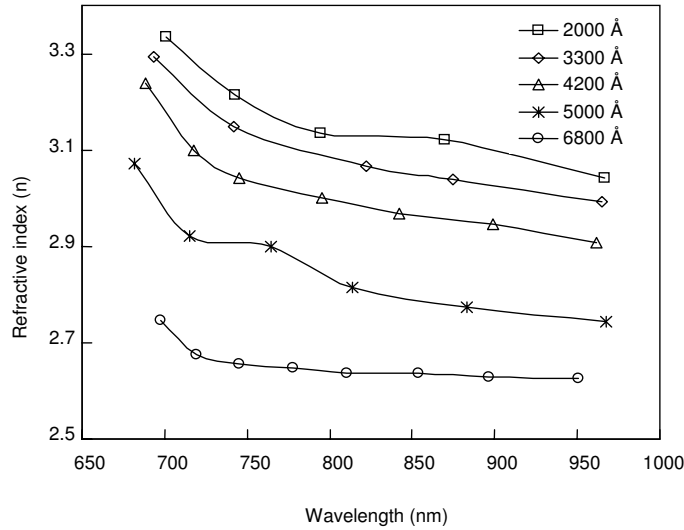


Figure 5. Variation of the refractive index (n) with wavelength of different thickness films of a- As_2Se_3 .

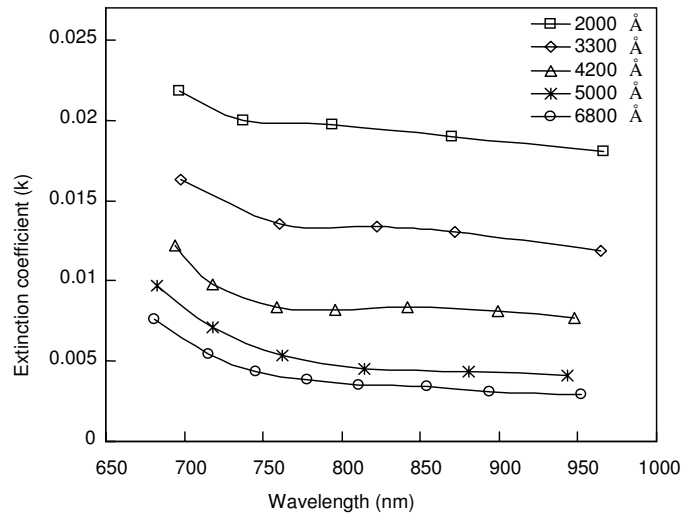


Figure 6. Variation of the extinction coefficient (k) with wavelength in different thickness films of a- As_2Se_3 .

The values of n and k for different thickness are given in Table 1.

Table 1. Thickness dependent optical band gap (E_g), refractive index (n), and extinction coefficient (k) at 900 nm.

| Thickness of the thin film | E_g (eV) | n | k |
|----------------------------|------------|------|---------|
| 2000 Å | 1.65 | 3.09 | 0.01875 |
| 3300 Å | 1.67 | 3.03 | 0.01259 |
| 4200 Å | 1.70 | 2.94 | 0.00810 |
| 5000 Å | 1.73 | 2.76 | 0.00420 |
| 6800 Å | 1.75 | 2.62 | 0.00310 |

The spectral transmittance data for thin film has also been used to derive the complex dielectric function of the thin film. The real and imaginary parts of the dielectric function, ϵ' and ϵ'' , are related to n and k [18]:

$$\epsilon' = n^2 - k^2 \text{ and } \epsilon'' = 2nk \tag{7}$$

The variation of ϵ' and ϵ'' with wavelength are shown in Figures 7 and 8. It is observed that the values of ϵ' and ϵ'' decrease with the increase in film thickness. The dissipation factor, $\tan \delta$, is expressed as

$$\tan \delta = \frac{\epsilon''}{\epsilon'}. \tag{8}$$

Variation of dissipation factor with thickness is shown in Figure 9. It is clearly observed that dissipation factor decreases with the increase in the thickness of films.

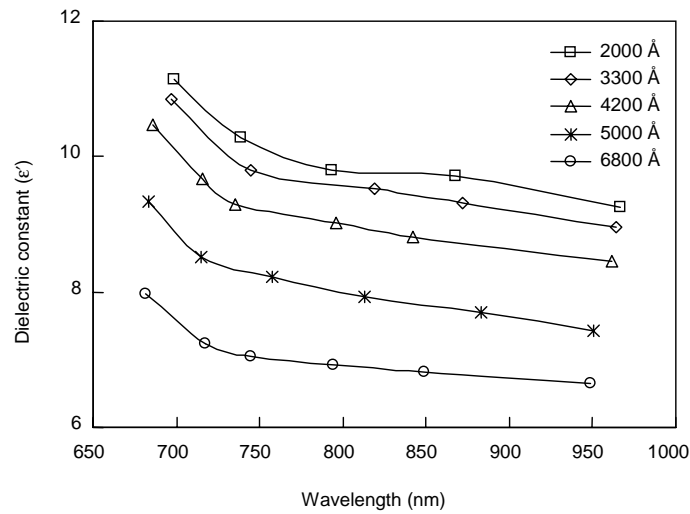


Figure 7. Variation of dielectric constants real (ϵ') with wavelength in different thickness a-As₂Se₃ thin films.

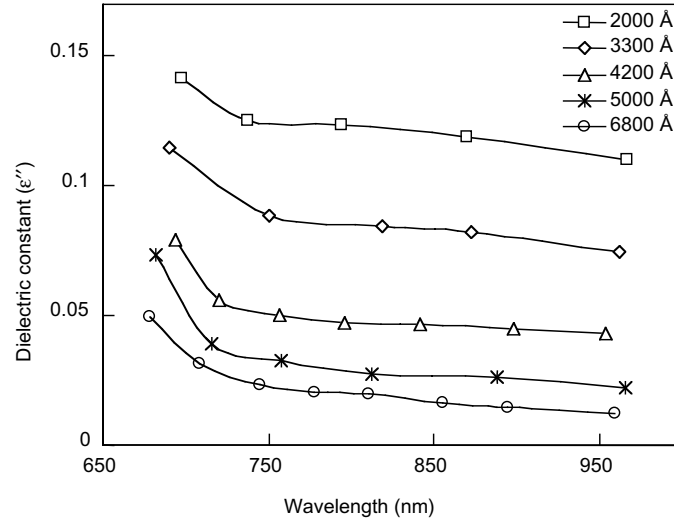


Figure 8. Variation of dielectric constants imaginary (ϵ'') with wavelength in different thicknesses a-As₂Se₃ thin films.

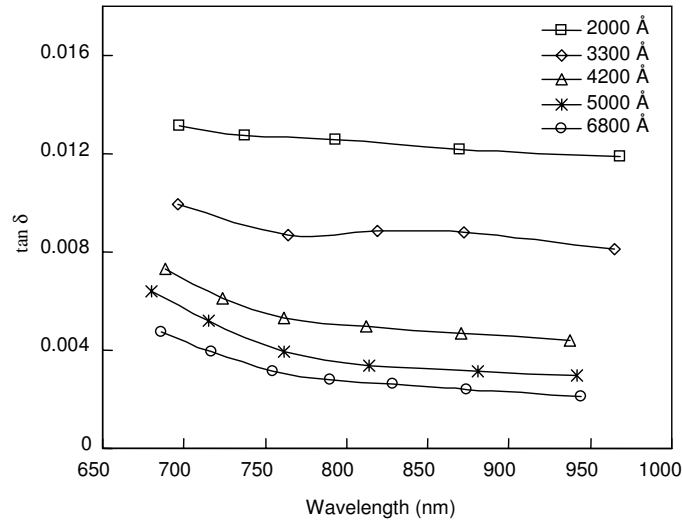


Figure 9. Variation of dissipation factor ($\tan \delta$) with wavelength in different thickness a-As₂Se₃ thin films.

It is seen from Table 1, the value of n is much greater than that of k . Assuming $\epsilon' = n^2$, the dependence of ϵ' on λ can be examined using the relation [19–21]

$$\epsilon' = n^2 = \epsilon_\infty - \left(\frac{e^2}{\pi c^2} \right) \cdot \left(\frac{N_c}{m^*} \right) \lambda^2, \quad (9)$$

where ϵ_∞ is high frequency dielectric constant, e is the electronic charge, N_c is the carrier density and m^* is the effective mass of the carrier. From a linear re-plot equation (9), ϵ' as a function of λ^2 , it is possible to estimate ϵ_∞ and $\frac{N_c}{m^*}$; Figure 10 shows just such a plot. The values of ϵ_∞ , determined from the extrapolation of the linear part of the curve, is listed in Table 2. The intersection at $\lambda^2 = 0$ for the linear part of the curve at higher wavelength gives the high frequency dielectric constant [22]. It is also observed from the Table 2 that ϵ' and ϵ_∞ decrease with increase of film thickness.

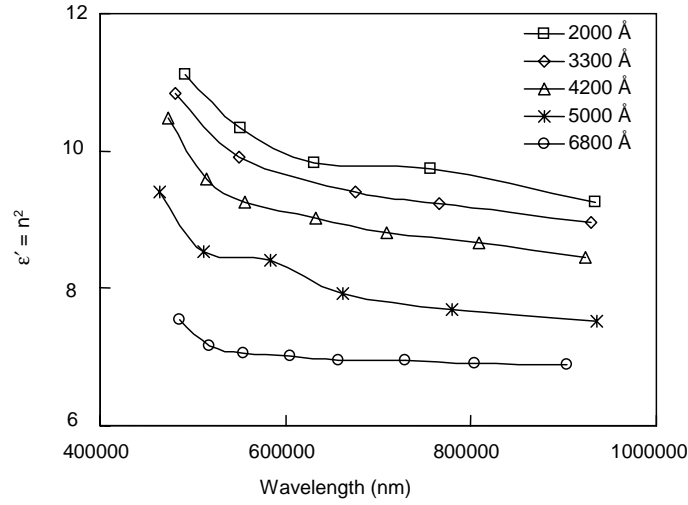


Figure 10. Variation of relative permittivity (ϵ') with wavelength in different thickness a-As₂Se₃ thin films.

Table 2. Thickness dependent real (ϵ') and imaginary (ϵ'') dielectric constant, dissipation factor ($\tan\delta$) at wavelength 900 nm, and high frequency dielectric constant (ϵ_∞).

| Thickness of the thin film | ϵ' | ϵ'' | ϵ_∞ | $\tan \delta$ |
|----------------------------|-------------|--------------|-------------------|---------------|
| 2000 Å | 9.55 | 0.115 | 11.27 | 0.0120 |
| 3300 Å | 9.19 | 0.079 | 10.70 | 0.0086 |
| 4200 Å | 8.64 | 0.044 | 10.24 | 0.0045 |
| 5000 Å | 7.63 | 0.024 | 8.79 | 0.0030 |
| 6800 Å | 6.72 | 0.014 | 7.22 | 0.0022 |

4. Conclusion

Various parameters related to optical properties were calculated for different thickness thin film of a-As₂Se₃. Optical constants of a-As₂Se₃films are strongly dependent on the thickness of the films. Increases in band gap of the films with the increase of thickness have been attributed to the decrease of localized states in the band gap. The corresponding decrease in optical constant such as refractive index, extinction coefficient, dielectric constant and the high frequency dielectric constant also decrease with increasing film thickness.

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