Structural and Optical Properties of Different Composition of $Se_{90}Cd_{10-x}In_x$ thin Films by Vacuum Evaporation Technique

D.K. Dwivedi[1,*], Nitesh Shukla[1], H.P. Pathak[1], Kedar Singh[2]

[1]Amorphous Semiconductor Research Lab, Department of Physics, Madan Mohan Malaviya University of Technology, Gorakhpur
[2]Department of Physics, Banaras Hindu University, Varanasi
*Corresponding author: todkdwivedi@gmail.com

Abstract $Se_{90}Cd_{10-x}In_x$ (x=2,4,6 &8) thin films have been deposited onto a chemically cleaned glass substrate by thermal evaporation technique under vacuum. The effects of different doping concentration of In have been investigated X-ray diffraction technique has been employed to investigate the structural characterization of the films. X-ray diffraction measurement indicates that the $Se_{90}Cd_{10-x}In_x$ films possess polycrystalline structure. Absorption spectra measured at normal incidence in the wavelength range 400-1100 nm have been analyzed for the optical characterization of the thin films under consideration. The optical constants (absorption coefficient (α), extinction coefficient (k)) and optical band gap E_g have been calculated. The absorption coefficient (α) is found to increase with photon energy. It has been found that extinction coefficient (K) decreases with increase in wavelength (λ). Optical band gap (E_g) has also been evaluated for the $Se_{90}Cd_{10-x}In_x$ thin films with different compositions. It has been found that optical band gap (E_g) increases with In incorporation in $Se_{90}Cd_{10-x}In_x$ alloys.

Keywords: Chalcogenide glasses, amorphous semiconductors, thin films, optical properties, optical band gap

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1. Introduction

Among Chalcogenide glassy semiconductors the II-VI compound semiconductors are extremely vibrant because of their uses in the optoelectronic devices and solar cells [1-10]. These materials particularly Se glasses exhibit unique property of reversible transformation [11]. This property makes these glasses very useful in optical memory devices [12,13,14]. Se based Chalcogenide glasses have high transparency in the broad middle and far infrared region and have strong nonlinear properties [15]. Though amorphous Se has got various device applications which make it attractive but pure Se has disadvantage like short life time and low sensitivity. This problem can be overcome by alloying Se with some impurity atoms, which gives higher sensitivity, higher crystallization temperature and smaller aging effect [16,17,18]. In the present work In has been chosen as an additive element in Se-Cd matrix to prepare Se-Cd-In alloys, because it is recognized as one of the most efficient elements used to improve the opto-electronic properties of compounds [19]. The third element behaves as chemical modifier and creates compositional as well as configurational disorder in the material with respect to binary alloys, which will be useful in understanding the structural, electrical and optical properties of Se-Cd-In chalcogenide glasses

[20,21,22]. Se-Cd-In is a semiconductor ternary chalcogenide glass of the type $A^{VI}B^{II}C^{III}$, where A = Se, Te, S, B = Cd, Zn and C=In, Tl, Ga. The ternary chalcogenides have potential applications in solar energy conversion due to their interesting tailored properties [23-28], in optoelectronic devices [29] and in nonlinear optics [30,31]. A number of techniques have been employed to prepare semiconductor ternary chalcogenide glasses of the type $A^{VI}B^{II}C^{III}$ including sputtering, physical vapor deposition, thermal evaporation method, spray pyrolysis and electro deposition technique [32-37]. In the present work resistive thermal evaporation method has been used due to considerably large mean free path of the vapor atoms at low pressure and a sharp thin film is obtained. This technique provides a wide window for the selection of substrate with minimum impurity concentration in the film.

In the present work our aim is to study the effect of In incorporation on the structural and optical properties of Se-Cd matrix. X-ray diffraction technique has been employed to investigate the structural properties of the $Se_{90}Cd_{10-x}In_x$ alloys. The optical behavior of a material is utilized to determine its optical constants. Therefore, an accurate measurement of optical constants is extremely important. Optical absorption spectra of $Se_{90}Cd_{10-x}In_x$ (x = 2, 4, 6 & 8) thin films have been recorded in the wavelength range 400-1100 nm by double beam UV-Vis spectrophotometer. Optical parameters like absorption

coefficient (α), extinction coefficient (k) and optical band gap (E_g) have been calculated for $Se_{90}Cd_{10-x}In_x$ alloys.

2. Experimental

Alloys of $Se_{90}Cd_{10-x}In_x$ (x = 2, 4, 6 & 8) were prepared by melt quenching technique. The exact proportions of high purity (99.999%) Se, Cd and In elements, in accordance with their atomic percentages, were weighed using an electronic balance (LIBROR, AEG-120) with the least count of 10^{-4} gm. The material was then sealed in evacuated (~10^{-5} Torr) quartz ampoule (length ~ 5 cm and internal diameter ~ 8 mm). The ampoule containing material was heated to 800 ^{0}C and was held at that temperature for 12 hours. The temperature of the furnace was raised slowly at a rate of 3 - 4°C / minute. During heating, the ampoule was constantly rocked, by rotating a ceramic rod to which the ampoule was tucked away in the furnace. This was done to obtain homogeneous alloy. After rocking for about 12 hours, the obtained melt was rapidly quenched in ice-cooled water. The quenched sample was then taken out by breaking the quartz ampoule. Thin films of $Se_{90}Cd_{10-x}In_x$ (x = 2, 4, 6 & 8) were prepared by vacuum evaporation technique, in which the substrate was kept at room temperature at a base pressure of 10^{-6} Torr using a molybdenum boat. The films were kept inside the deposition chamber for 24 hours to achieve the metastable equilibrium as suggested by Abkowitz [38]. The thickness of the film was measured using a single crystal thickness monitor. The XRD patterns of the films were recorded with the help of x-ray diffractometer (XPERT-PRO) using Cu-Kα radiation (λ=1.54 Å). The tube was operated at 45 kV and 35 mA. The normal incidence absorption spectra of $Se_{90}Cd_{10-x}In_x$ (x = 2, 4, 6 & 8) thin films have been taken by a double beam UV-Vis-NIR computer controlled spectrophotometer (ECIL-Hyderabad, India, Model No.5704 SS) in the wave length range 400-1100 nm.

3. Results and Discussion

3.1. Structural Property

Figure 1 shows the XRD pattern of $Se_{90}Cd_{10-x}In_x$ (x = 2, 4, 6 & 8) thin films. X-ray diffraction pattern reveal that the $Se_{90}Cd_{10-x}In_x$ thin films are polycrystalline in nature. For $Se_{90}Cd_{10-x}In_x$ thin films, the peaks at 2θ=23.6º, 2θ=25.5º,2θ=29.8º, 2θ=43.4º reveal the existence of (100),(002),(101) and (110) diffraction peaks which corresponds to hexagonal structure [22,39,40]. It has been observed that the intensity of all peaks increases significantly with the incorporation of In in Se-Cd alloys. A comparison of the observed and standard d values for (hkl) planes ensures that $Se_{90}Cd_{10-x}In_x$ (x=2, 4, 6&8) show hexagonal structure [41].

The lattice parameters of the hexagonal planes are calculated using the following relationship

$$1/d = (4/3)[(h^2 + k^2 + \ell^2)/a^2] + (\ell^2/c^2) \qquad (1)$$

The values of lattice parameters for $Se_{90}Cd_{10-x}In_x$ thin films are listed in Table1. The crystallite size D has been estimated by using the Debye-Scherer's formula [39]

$$D = 0.9\lambda / \beta Cos\theta \qquad (2)$$

Where β is full width at half maximum (FWHM) in radians, λ is the wavelength of X-rays used and θ is the Bragg's angle.

The strain (ϵ) has been evaluated using the formula

$$\epsilon = \beta Cos\theta / 4 \qquad (3)$$

The dislocation density (δ), defined as the length of dislocation lines per unit volume of the crystal is evaluated from the formula [38]

$$\delta = 15\beta Cos\theta / 4aD \qquad (4)$$

Where D is the crystallite size, β is the full width at half maximum,θ is the Bragg's angle and a is the lattice parameter.

The microstructural parameters such as crystallite size (D), strain (ϵ), dislocation density (δ) and the lattice parameter values have been evaluated using above equations and the results are listed in Table 1. As the In dopant concentration increases the crystallite size decreases. This happens because In doping causes broadening of (100) peak, corresponding to a decrease of crystallite size of the films. The dislocation density estimated using Eq.(4), increases with increase in the In doping concentration which may be related to the increase in lattice dislocation due to the increase in doner sites. These observations match with the earlier results reported on Se-Cd-In thin films [38].
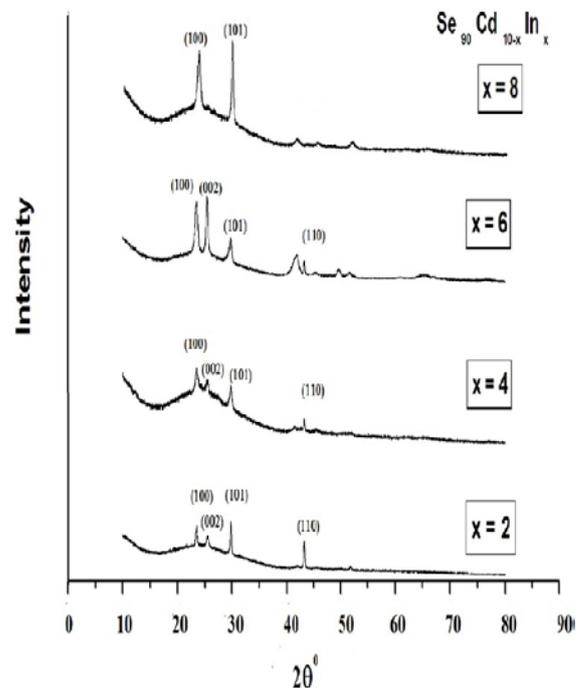

Figure 1. XRD pattern of $Se_{90}Cd_{10-x}In_x$ (x=2, 4, 6&8) thin films

Table 1. Microstructural parameters of $Se_{90}Cd_{10-x}In_x$ thin films

In doping concentration	Crystallite size D (nm)	Dislocation density δ×10^{15}(lines m^{-2})	Strain ϵ×10^{-3}	Lattice parameters (nm)	
				a	c
X=2	26.22	1.70	1.33	4.34	3.76
X=4	17.40	3.97	1.99	4.34	--
X=6	14.40	5.80	2.40	4.34	--
X=8	13.92	6.20	2.49	4.34	--

3.2. Optical Properties

3.2.1. Absorption Coefficient (α) and Optical Band Gap (E$_g$)

The absorption coefficient (α) has been obtained directly from the absorbance against wavelength curves using the relation [42-50].

$$\alpha = OD / t \qquad (5)$$

Where OD is the optical density measured at a given layer thickness (t).

A plot of absorption coefficient (α) as a function of photon energy (hv) is given in Figure 2 and values are given in Table 2. It has been observed that absorption coefficient (α) increases with increase in photon energy (hv). Absorption coefficient (α) decreases with increase in concentration of In in Se-Cd-In matrix.
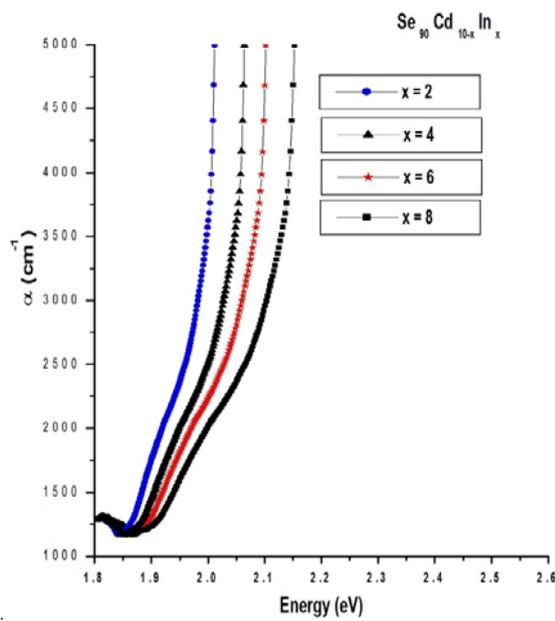
Figure 2. Variation of absorption coefficient (α) with photon energy (hv) in Se$_{90}$Cd$_{10-x}$In$_x$ (x=2, 4, 6&8) thin films

Table 2. Optical band gap (E$_g$), absorption coefficient (α) and extinction coefficient (k) for Se$_{90}$Cd$_{10-x}$In$_x$ thin films

S.No.	Sample	Optical Band Gap (E$_g$) in eV	Absorption Coefficient (α) (cm^{-1})×10^3	Extinction Coefficient (k) 10^{-2}
1.	Se$_{90}$Cd$_8$In$_2$	1.67	5.7050	0.3950
2.	Se$_{90}$Cd$_6$In$_4$	1.95	5.1820	0.3380
3.	Se$_{90}$Cd$_4$In$_6$	2.50	4.8625	0.3093
4.	Se$_{90}$Cd$_2$In$_8$	2.80	4.6250	0.3089

Large amount of dopants induces a large free carrier density in the bands and a high density of ionized dopants ions which cause the formation of band tails and variation of density of states, which strongly influence the optical properties. In defect free crystalline semiconductors, an energy gap is clearly defined because the conductor and valence band distribution of states terminate abruptly at their respective band edges. But presence of tail states in doped semiconductor makes it difficult to give a realistic energy gap value for these materials. One of the standard empirical method to obtain the optical gap values is the Tauc method. The fundamental absorption, which corresponds to electron excitation from the valence band to conduction band, can be used to determine the nature

and the value of optical band gap. The relation between absorption coefficient (α) and the incident photon energy (hv) be written as [51].

$$(\alpha h\nu)^{1/n} = A(h\nu - E_g) \qquad (6)$$

where, A is the edge width parameter representing the film quality, which is calculated from the linear part of this relation and E$_g$ is the optical band gap of the material. The exponent n depends on the type of transition. For direct allowed n=1/2, indirect allowed transition n=2 and for direct forbidden n=3/2. To determine the possible transition (αhv)$^{1/n}$ vs hv is plotted and corresponding E$_g$ values are obtained by extrapolating the straight line portion of the graph on hv axis as shown in figure 3.
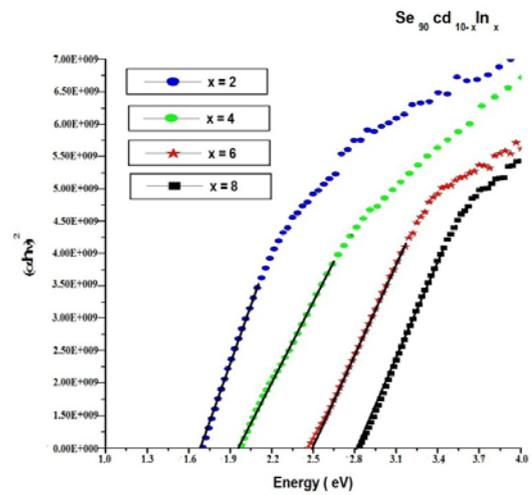
Figure 3. Variation of (αhv)^{2}with (hυ) in Se$_{90}$Cd$_{10-x}$ In$_x$ (x = 2,4,6 & 8) thin films

The direct optical band gap values calculated from (αhv)^2 vs hv are listed in Table 2. It is evident from Table 2 that the values of optical band gap(E$_g$) increases with increasing In concentration. It has been observed from XRD that crystallite size decreases with increases in In doping concentration, therefore the increase in band gap values with the increasing doping concentration may be attributed to size quantization. Variation of optical band gap (E$_g$) with In concentration in Se$_{90}$Cd$_{10-x}$In$_x$ (x=2,4,6&8) alloys is shown in Figure 4.

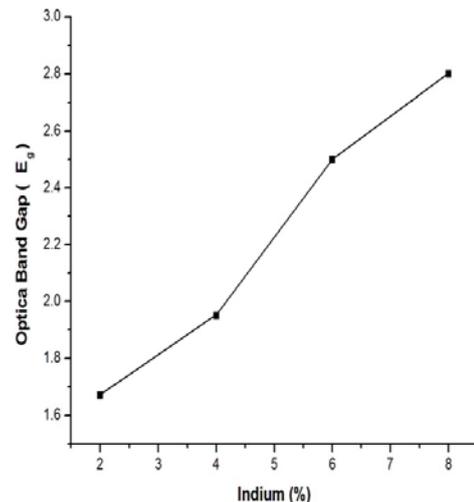**Figure 4.** Variation of optical band gap (Eg) with In concentration in Se$_{90}$Cd$_{10-x}$ In$_x$ (x = 2, 4, 6 & 8) thin films

3.2.2. Extinction Coefficient (k)

The optical behavior of the material has been utilized to determine its extinction coefficient (k). The extinction coefficient has been calculated using the relation

$$k = \alpha\lambda / 4\pi \qquad (7)$$

Where α is optical density/film thickness

Figure 5 shows the spectral dependence of k for $Se_{90}Cd_{10-x}In_x$ (x = 2, 4, 6 & 8) thin films. It is clear from the figure that k decreases linearly with an increase in λ for all the samples. This behavior is due to decrease in absorption coefficient with increase in λ. It is also evident from Table 2 that k decreases with In concentration in $Se_{90}Cd_{10-x}In_x$ thin films.
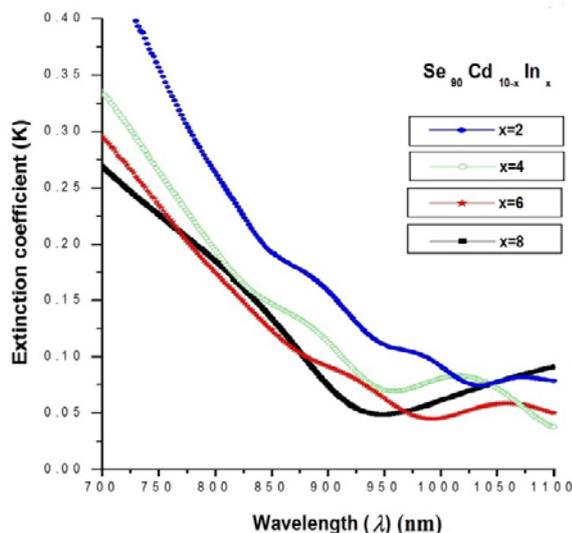

Figure 5. Variation of extinction coefficient (k) with wavelength (λ) in $Se_{90}Cd_{10-x}In_x$ (x = 2, 4, 6 & 8) thin films

4. Conclusion

Thin films of $Se_{90}Cd_{10-x}In_x$ (x = 2, 4, 6 & 8) alloys have been deposited onto a chemically cleaned glass substrate by thermal evaporation technique. An attempt has been made to understand the effect of Indium doping concentrations on the structural and optical properties of Se-Cd-In thin films. XRD measurement indicates that $Se_{90}Cd_{10-x}In_x$ thin films are polycrystalline in nature and possess hexagonal structure. Crystallinity increases with the increase of In incorporation in Se-Cd alloys. The optical absorption spectra of $Se_{90}Cd_{10-x}In_x$ alloys have been studied in the wavelength range of 400-1100 nm by spectrophotometer. The optical parameters like absorption coefficient (α), extinction coefficient (k) and optical band gap (E_g) have been calculated for $Se_{90}Cd_{10-x}In_x$ alloys. It is observed that extinction coefficient (k) decreases with wavelength (λ) for all the samples. Absorption coefficient (α) is found to increase linearly with incident photon energy (hν) for all the samples. It has further been observed that both k and α decreases with increasing In concentration in Se-Cd matrix. Absorption measurement show the presence of direct band gap for $Se_{90}Cd_{10-x}In_x$ thin films and the band gap increases as the In dopant concentration increases. The increase in band gap values with increasing In doping concentration may be due to the effect of size quantization.

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