Journal of Modern Materials
ISSN: 2456-4834
Volume 1, Issue 1, pp. 17-23, 2016
DOI: https://doi.org/10.21467/jmm.1.1.17-23

RESEARCH ARTICLE

Laser Irradiation Effect on the Optical Band Gap of $Se_{96-x}Te_4Hg_x$ Thin Films

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Received: 19 April 2016 / Revised: 18 July 2016 / Accepted: 19 July 2016 / Published: 24 July 2016

ABSTRACT

The effect of laser irradiation on the optical band gap of $Se_{96-x}Te_4Hg_x$ thin films has been studied. Thin films of thickness 300 nm have been deposited on glass substrate by using thermal evaporation technique. The result shows that irradiation causes a red shift in the absorption edge and hence optical band gap decreases with increasing irradiation time. The results have been analyzed on the basis of laser irradiation-induced defects in the films. Furthermore, optical band gap has been found to decrease by increasing mercury concentration and optical band gap decreases rapidly with a higher concentration of mercury.

Keywords: Laser Irradiation; Chalcogen alloys; Thin films; Irradiation effect; optical band gap

1. Introduction

The optical band gap and extinction coefficient are the most significant optical parameters in amorphous semiconductors. Optical absorption measurements are widely used for studying disorder and defects as well as modifications of the density of states upon alloying and laser irradiation. It is well known that in amorphous semiconductors the optical band gap depends mostly on the film composition. The disorder and defects have a strong influence on the band structure of Chalcogenides and the detailed electronic structure of a given Chalcogenides are determined by the chemical bonding which occurs and therefore by its composition. Recently, there has been an increasing interest in the semiconductor thin films due to their exceptional properties, which are remarkably different from those of bulk materials [1-6]. The interest in these materials is principally due to low phonon energy, extended infrared transparency, high refractive index, high photosensitivity, in a reversible phase change optical recording etc. [1-3]. Se-Te based alloys have created extreme interest due to their greater hardness, higher photosensitivity, higher crystallization temperature, and lower aging effects as in comparison to pure amorphous Se [7, 8]. It has been pointed out that Se-Te based alloys have more advantages than a-Se from the technological point of view. The addition of Te into Se improves the corrosion resistance [9]. Therefore, Se-Te based alloys are thought to be promising media, which make use of, a phase change between an amorphous state and a crystalline state and used to extend the utility of a-Se. Selenium–Tellurium based semiconductors have been the focus of interest in thin film form because of properties suitable for device applications [10-12]. These materials are optically highly non-linear and sensitive to the laser irradiation [13-15]. Recently Laser irradiation effect on different materials has been studied [16

- 27]. The absorption of laser irradiation in chalcogenide thin film depends strongly on their electronic structure which in turn changes by the interaction with photons. The additional absorption of Te containing alloys is due to the increase in the number of thermally excited free carriers with Te content. The most important applications of chalcogenide are now in the field of optics [28 -30] and arising mainly from their exhibited infrared transmitting properties [31, 32]. Their potential uses have been reported in integrated optics, optical imaging, optical data storage and infrared optics. Se-Te based alloys have gained much attention and found to be useful in practical applications [33]. Among various applications of these chalcogenides, the optical recording of information has a leading importance. The principle underlying optical recording consists in the appearance of a large change of certain physical and chemical parameters of the chalcogenides under the action of light. The energy of the light quanta situated in the UV spectral range is expected to induce qualitatively new changes in the chalcogenides because this energy is equal or higher than the chemical bond energy. The aim of the present work is to synthesize $Se_{96-x}Te_4Hg_x$ alloy and investigate the laser irradiation effect on optical band gap of thin films of said chalcogenide alloys.

2. Materials and Methods

Alloys of a-$Se_{96-x}Te_4Hg_x$ (x= 4, 8, 12) have been prepared by melt quenching technique. The high vapour pressure of chalcogenide melts and the tendency, especially at higher temperatures, to react with Oxygen, requires the work in a closed system under the vacuum condition. Hence, highly pure materials (99.999%) having the desired compositional ratio of elements have been sealed in quartz ampoules under a vacuum of 10^{-5} torr. The sealed ampoules are kept inside a programmable furnace where the temperature is raised up to 900 K at the rate of 4 K / minute for 10 hours with frequent rocking to ensure the homogenization of the melt. The quenching has been done in ice-cold water. As-quenched alloys have been grounded and the resulting fine powder has been used to prepare the thin films by PVD method. Thin films of Se-Te-Hg alloy of thickness 300 nm have been deposited on a well-cleaned glass substrate in the shape of squares by vacuum evaporation technique at room temperature and in a vacuum of ~10^{-5} torr. Films have been kept inside the deposition chamber for 24 h to achieve metastable equilibrium. The thickness of the film has been measured under a single-crystal thickness monitor. The prepared thin films have been irradiated with a pulsed Transverse Electrical Excitation at Atmospheric pressure (TEA) Nitrogen laser for 5, 10, 15 & 20 minutes. Thin film has been placed in a specially designed sample holder as shown in figure 1, which kept at a distance of 15 cm from output laser head. For laser irradiation, a spot of 6 mm diameter has been adjusted which irradiates the thin film with a peak average energy density of ~3.5 x 10^5 W/cm^2. For optical measurements of the thin films, a double beam UV/VIS/NIR Scanning Spectrophotometer (Camspec M 550) has been used. The optical spectrum has been measured as a function of wavelength (190-1100 nm) of incident light.

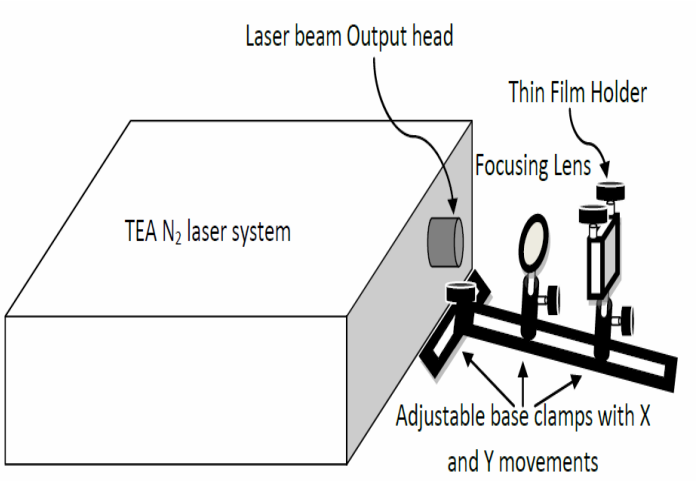

Figure 1: *Layout of N_2 laser irradiation system*

3. Theory/Calculation

Optical absorption measurements are very useful for studying the modifications of the density of states upon alloying. Measurement of the absorption coefficient (α) as a function of frequency (ν) provides a mean to determine the optical band gap (E_g) of thin film. The absorption

coefficient has been calculated directly from the following well known relation [34]-

$$\alpha = \frac{1}{t}\ln\left(I/I_O\right) \quad (1)$$

where "t" is the film thickness and $\ln\left(I/I_O\right)$ corresponds to absorbance, neglecting the reflection coefficient, which is negligible and insignificant near the absorption edge.

In a-semiconductors, due to the presence of band tails and a wide distribution of defect states in the gap, the absorption coefficient does not show any sharp drop at any energy which would correspond to the band edge separation. An "optical gap" (Eg) is usually defined by extrapolation of the observed absorption coefficient behavior in the high energy range where the absorption can be assigned to transitions from the extended states in the valence band to the extended states in the conduction band. In this region, the absorption coefficient α is $>10^4$ cm^{-1} involving optical transitions between conduction and valence band states and the spectrum shows a square root dependence on photon energy $(h\nu)$ given by Tauc et al [35] and discussed in more general terms by Devis and Mott [36, 37], whose equation is in the form of-

$$\alpha h\nu = \beta\left(h\nu - E_g\right)^n \quad (2)$$

where β is a constant, E_g is the optical energy gap of the material. The phonon absorption in many amorphous materials is found to obey the Tauc's relation. The index n is a number which characterizes the transition process involved. n has discrete values like 1/2, 3/2, 2 or more depending on whether the transition is direct or indirect and allowed or forbidden, respectively. In the direct and allowed cases, the index n is 1/2 whereas for the direct but forbidden cases it is 3/2. But for the indirect and allowed cases n=2 and for the forbidden cases it will be 3 or more. According to Tauc [38] the absorption tail related to localized states into the pseudo-gap, which localized states can be arised from the existence of vacancy defects and/or impurities. The optical band gap (E_g) can be measured from the plot $(\alpha h\nu)^{1/n}$ versus $h\nu$ by extrapolating the curves to $h\nu$ axis at $(\alpha h\nu)^{1/n} = 0$.

The extinction coefficient (k), which indicates the amount of absorption loss when the electromagnetic wave propagates through the material, has been calculated using well-known relation [39, 40]

$$k = \frac{\alpha\lambda}{4\pi} \quad (3)$$

Where, α is absorption coefficient and λ is the corresponding wavelength.

4. Results

The optical absorption coefficient *a* has been calculated from the optical data using equation 1 and obtained value for all pristine and laser irradiated samples has been listed in table 1. The variation of absorption coefficient (α) as a function wavelength (λ) for all pristine thin films shows that α decreases with increasing λ for all samples as shown in figure 2.

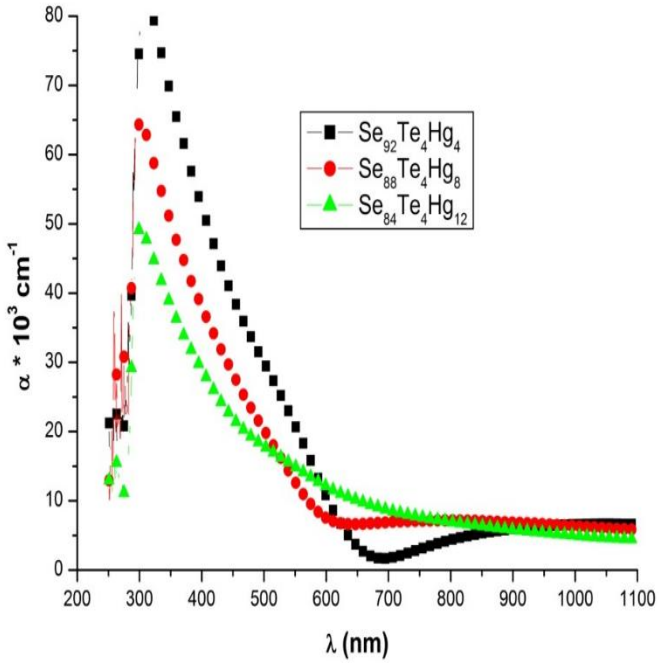

Figure 2: *The variation of absorption coefficient a as a function of wavelength λ for different Hg concentration*

For the pristine thin films, increase in Hg concentration results in a reduction of the absorption coefficient (α) in the range 290-580 nm and then it increases with increasing Hg concentration in the range of 580-800 nm. Furthermore, α increases with increasing irradiation time.

The plots of Tauc, Equation 2, for all the pristine and laser irradiated films are shown in Figures 3-5. In Se-Te-Hg system, best fit value of n is found to be 2 which shows indirect transition and the optical band gap(E_g) have been measured from the plot $(\alpha h\nu)^{1/2}$versus $h\nu$ by extrapolating the curves to $h\nu$ axis at $(\alpha h\nu)^{1/2} = 0$. Estimated optical band gap E_g for all samples is given in Table 1. During optical parameters estimation from absorption data the systematic error of instrument has been considered and respective uncertainties are shown in figure also given in table 1. The estimated optical energy gap decreases with increasing laser irradiation time as well as Hg concentration in Se-Te-Hg alloy. The amount of absorption loss, when the electromagnetic wave propagates through the material i.e. extinction coefficient, is frequency dependent.

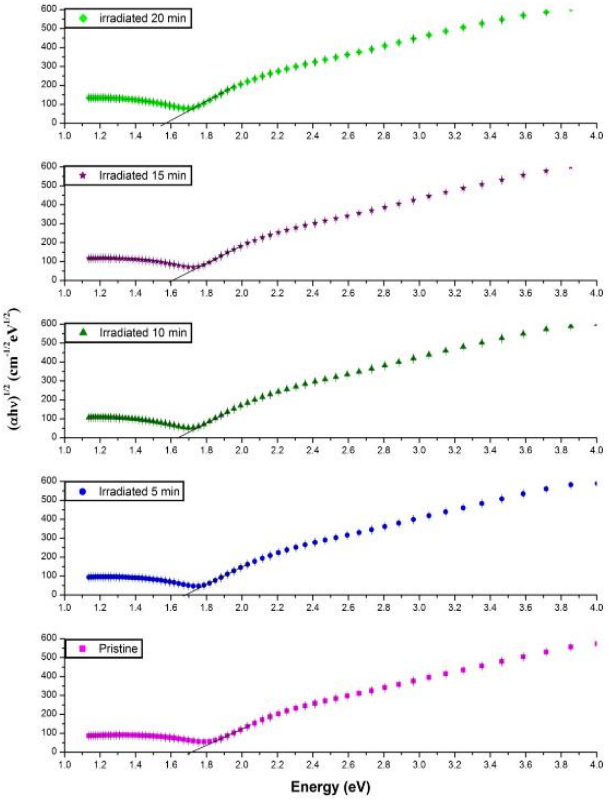

Figure 3: *The variation of $(\alpha h\nu)^{1/2}$ with photon energy (hv) with and without laser irradiation for $Se_{92}Te_4Hg_4$ thin film*

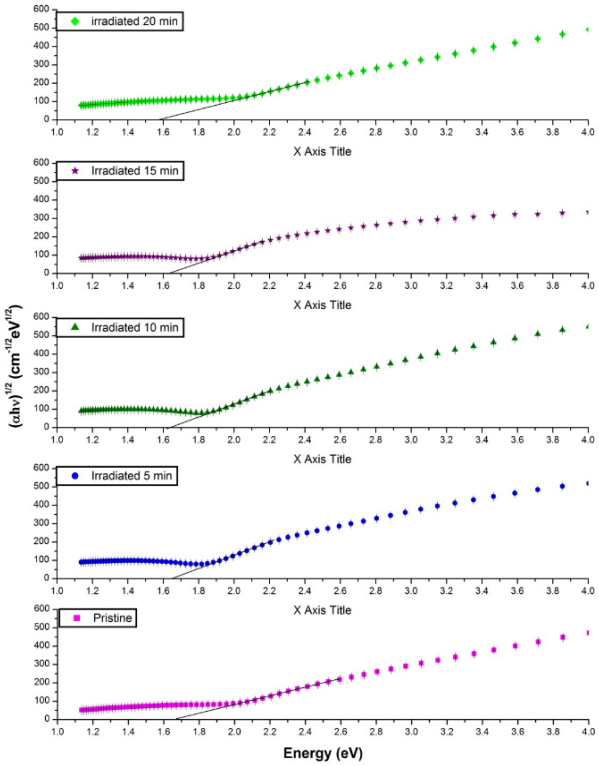

Figure 4: *The variation of $(\alpha h\nu)^{1/2}$ with photon energy (hv) with and without laser irradiation for $Se_{88}Te_4Hg_8$ thin film.*

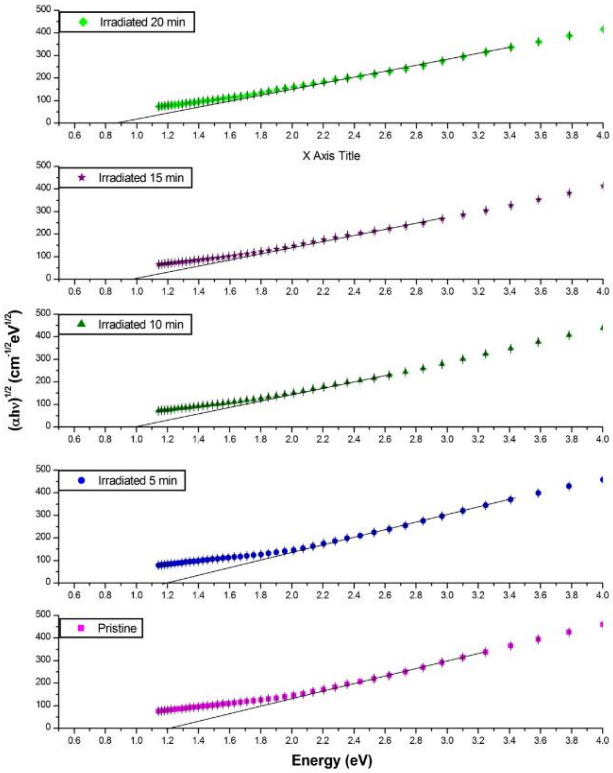

Figure 5: *The variation of $(\alpha h\nu)^{1/2}$ with photon energy (hv) with and without laser irradiation for $Se_{84}Te_4Hg_{12}$ thin film*

Table 1: *Calculated optical parameters @ 600 nm for* $Se_{96-x}Te_4Hg_x$

S.N.	Exposure time	α ($10^4 cm^{-1}$)	k	E_g (eV)
1.	$Se_{92}Te_4Hg_4$			
(a)	Pristine film	0.752±0.3	0 .0359	1.71±0.001
(b)	**Exposure time**			
(i)	5 min	0.782±0.3	0.0374	1.66±0.001
(ii)	10 min	1.001±0.3	0.0478	1.64±0.001
(iii)	15 min	1.077±0.3	0.0515	1.60±0.001
(iv)	20 min	1.079±0.3	0.0515	1.56±0.001
2.	$Se_{88}Te_4Hg_8$			
(a)	Pristine film	1.067±0.3	0 .0509	1.69±0.001
(b)	**Exposure time**			
(i)	5 min	1.297±0.3	0.0620	1.67±0.001
(ii)	10 min	1.413±0.3	0.0675	1.64±0.001
iii)	15 min	1.542±0.3	0.0736	1.62±0.001
iv)	20 min	1.599±0.3	0.0764	1.60±0.001
3.	$Se_{84}Te_4Hg_{12}$			
(a)	Pristine film	1.109±0.3	0.0530	1.24±0.001
(b)	**Exposure time**			
(i)	5 min	1.124 ±0.3	0.0537	1.21±0.001
(ii)	10 min	1.164±0.3	0.0556	1.02±0.001
(iii)	15 min	1.208±0.3	0 .5773	1.00±0.001
(iv)	20 min	1.339±0.3	0.0639	0.92±0.001

In the region of strong absorption, the interference fringes disappear and near the absorption edge reflection coefficient are negligible and insignificant. Hence, we choose a wavelength near the absorption edge to analyse the effect of laser irradiation and Hg content on absorption coefficient and extinction coefficient. The estimated values of the extinction coefficient before and after laser irradiation are given in table 1 and found to be increasing with increasing the laser irradiation time.

5. Discussion

The effect of Hg concentration on the absorption coefficient α is understandable in the light of the structural network. The clusters in Se-Te-Hg films are almost covered with the metallic additive Hg, which results in high reflectance and decrease the observed absorption coefficient (α). The obtained optical band gap E_g shows that the energy band gap decreases rapidly with higher Hg concentration. It indicates that the increase in mercury concentration results in rapid increase in the band broadening. The decrease of energy gap with radiation attributed to the variation of disorder and defects present in amorphous materials [41, 42]. The unsaturated bonds are responsible for producing localized defect states in the band structure of the amorphous solids. The presence of such localized states in the band structure is responsible for decreasing optical energy gap. The increase in the number of unsaturated defects increases the density of localized states in the band structure [43] and consequently decreases the optical energy gap E_g [44]. Further, increasing in Hg concentration is also responsible for producing defect states and hence, availability of more localized defect state as found in an earlier study of laser irradiation effect favored in Chalcogenide materials due to their structural flexibility and their high-lying lone-pair p states in their valence bands [4]. The decrease in the optical gap of Se-Te-Hg thin films by laser irradiation may be explained on the basis of bonds distribution model suggested by Golovchak et al. [45,46] that the bonds within the chains may be broken and change takes place in between chains. In the view of bond distribution model decrease in energy band gap with the addition of Hg content and laser irradiation can be expected. The increase in Hg concentration favored the availability of more Hg-Se bonds in the alloy. Since Hg-Se bond is weaker in comparison of other bonds present in the alloy and so easy to be broken by laser irradiation. With increasing Hg concentration, weaker bonds would be available to be broken and hence the chance of the formation of defects increases. Due to this, availability of more localized states increases with increasing Hg content, which might be a reason to show a strong effect of irradiation with higher Hg concentration in comparison of composition with less Hg concentration. It confirms that the Se-Te based alloy with a higher content of Hg is more sensitive to Laser irradiation.

6. Conclusion

Optical absorption measurements of thin films show indirect allowed transition in the Se-Te-Hg system. The result shows that the laser irradiation produces disorder in the material, causing an increase in the number of localized states in the band gap consequently optical band gap (E_g)

decreases with increasing irradiation. Furthermore, optical energy gap decreases rapidly with the increasing in Hg concentration. It suggests that addition of Hg makes an increment in the available localized density of the states. In addition, laser irradiation shows a strong effect at higher Hg concentration (12%). It might be due to the availability of more Hg-Se bond as Se content decreasing with increasing Hg content in the alloy. It concludes that alloy with rich Hg content is more sensitive to laser irradiation. It is also observed that absorption coefficient (α) and extinction coefficient (k) increases with laser irradiation time.

How to Cite this Article:

A. Bahishti, I. Uddin, M. Zulfequar, and T. Alharbi, "Laser Irradiation Effect on the Optical Band Gap of Se96-xTe4Hgx Thin Films", *J. Mod. Mater.*, vol. 1, no. 1, pp. 17-23, Jul. 2016. doi:10.21467/jmm.1.1.17-23

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