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Studies on Phase Change $Se_{80}In_5Te_9Sn_6$ Chalcogenide Thin Films by Laser Irradiation

ARCHANA SRIVASTAVA
Department of Physics, St. Andrew's college, Gorakhpur, U.P., India

Abstract- ***In the present work, we have deposited $Se_{80}In_5Te_9Sn_6$ thin films using the thermal evaporation technique under a vacuum of 10^{-6} Torr for phase change studies. The films were then irradiated with a Transverse Electrical Excitation at Atmospheric Pressure (TEA) nitrogen laser for varying exposure times. X-ray structural characterization indicated that the films are amorphous, whereas the laser-irradiated thin films exhibit a crystalline nature. Structural changes were further investigated using FESEM. The results are discussed with respect to structural aspects and the amorphous-to-crystalline phase transition. The observed changes are attributed to the interaction between incident photons and lone-pair electrons, which influence the band gap. The optical constants of the thin films were determined from absorption spectra as a function of photonic energy in the wavelength range of 400–900 nm, revealing a decrease in the optical band gap and an increase in the absorption coefficient with increasing laser irradiation time. The reduction in the optical band gap is attributed to the change in the film structure from an amorphous to a crystalline phase with increasing exposure time.***

Keywords: Thin film, Laser-irradiation, Optical band gap, XRD

I. INTRODUCTION

In recent decades, extensive theoretical and experimental studies have demonstrated the remarkable properties of chalcogenide materials and their significant role in the advancement of emerging technologies for the benefit of mankind. Chalcogenide materials are chemical compounds containing at least one chalcogen element, S, Se and Te. Recent progress in multimedia technologies has been driven by increases in computer processing speed and data storage density, along with the growing demand for rewritable media. Among the various candidates, phase-change (PC) materials based on chalcogenides have emerged as one of the most promising materials for rewritable data storage applications [1], [2].The easier switching between amorphous-to-crystalline phase of these materials makes them compatible for information storage. Chalcogenide thin films have attracted considerable attention and have been systematically investigated over the past few decades. Owing to their intriguing physical and optical properties, they continue to be a focus of both experimental and theoretical research. These materials are well recognized as promising candidates for a wide range of photonic applications, including ultrafast optical switches, frequency converters, optical amplifiers, infrared lasers, phase-change memories, and infrared-transmitting optical fibers [3], [4]. Optical storage based on the amorphous–crystalline phase transition exploits the significant changes in optical reflectivity and absorption induced in certain semiconductor–semimetal thin films through heat treatment/laser irradiation. To study the possibilities of several applications and improvements, researchers are continuously worked on the investigations of optical, structural and thermal properties of ternary and quarternary chalcogenide glasses [5], [6], [7], [8]. These glassy materials are responsive to extraneous means, such as Gamma irradiation, laser irradiation, thermal annealing, electric field etc. because of their flexible structure. The optical properties of these materials altered due to irradiation. Irradiation excites electrons and induces ionization, which can result in atomic displacement within the material. The photo-generated electrons undergo dynamic motion and are subsequently trapped at defect or localized states, leading to new electronic configurations. These modifications significantly influence the optical properties of the chalcogenide thin films. As a result, the pronounced sensitivity of these glasses to gamma irradiation, reflected in changes to their optical behavior, renders them suitable for a wide range of technological applications [9], [10], [11], [12].

Many workers have studied the effect of laser irradiation on the optical properties of chalcogenide

glasses [13], [14], [15], [16]. Laser-induced optical properties change in Sb-S-Se chalcogenide thin films has been investigated through FTIR and XPS measurements by Naik et al. [17]. The variation in structural, optical and electrical properties of thin films of Pb-doped Ga-Se chalcogenide glasses before and after laser irradiation has been studied by Alvi [18]. The phase change studies of Ge15Se77Sb8 thin films by laser irradiation have been done by Srivastava et al. [19]. Parida et al. [20] has been studied the influence of time dependent laser-irradiation for tuning the linear–nonlinear optical response of quaternary Ag-In-S-Se films for optoelectronic applications. Deepika et al. [21] has been analyzed the effect of 532 nm Nd:YAG laser irradiation on the optical properties of Ge1Se2.5 glass film. Behara and Naik have studied the optical parameters due to the deposition and photo-induced diffusion of Te layer into the chalcogenide As_2Se_3 film [22]. Al-Hazmi [23] studied the optical constants of $Se_{75}S_{15}Ag_{10}$ chalcogenide thin films and observed an increase in optical band gap with increasing laser-irradiation time. Mao et al. [24] analyzed composition-dependent photostability of chalcogenide thin films within a germanium-sulfur binary system under femtosecond laser irradiation. The influence of 532 nm laser irradiation on the optical changes of $Ag_{10}Te_{10}As_{20}Se_{60}$ quaternary thin films has been studied by Das and his co-workers [25] This irradiation influenced the structural, linear and nonlinear optical parameters. Priyadarshini et al. [26] depicted the laser irradiation-induced effect on the optoelectrical and structural properties of thermally evaporated $Bi_{15}In_{20}Se_{65}$ thin films with different exposure durations. Jena and his coworkers [27] analyzed the Stability of Ag_2S/As_2Se_3 thin films under time-dependent laser irradiation and its impact on linear-nonlinear optical properties for optoelectronic applications. Sahoo et al. [28] reported the in-situ laser irradiated changes in the nonlinear/linear optical properties of $As_{50}Se_{40}Sb_{10}$ thin films for photonic applications.

In the present work, we have taken Selenium (Se) due to its appropriateness for PCM devices based on reversible phase change property. However, these materials suffer from aging effects, poor stability, and high sensitivity [29]. These limitations can be mitigated by incorporating an appropriate metallic additive, such as indium, into selenium. Indium-doped selenium has applications in gas sensors, thermoelectric materials, and solar cells [30], [31]. Furthermore, tellurium has been added to the Se–In alloy to enhance its crystallinity and improve corrosion resistance. Tellurium-based glasses are widely used as active layers in memory devices because of their low melting point and are considered among the most promising materials for phase-change memory applications [32], [33]. Metallic doping in chalcogenides alters the average coordination number and induces structural transformations in the glassy matrix, leading to flexible, intermediate, and rigid phases. In the present work, Sn has been incorporated into the ternary Se–In–Te glassy alloy, which enhances the glass-forming region and introduces configurational and compositional disorder in the glassy system [34], [35] . The aim of present work is to study the influence of laser irradiation on structural and optical properties of $Se_{80}In_5Te_9Sn_6$ chalcogenide thin films for phase change memory device application.

II. EXPERIMENTAL

The melt-quenching technique was adopted for the synthesization of $Se_{80}In_5Te_9Sn_6$ chalcogenide glass using 99.999 % pure elements. The materials were weighted according to their atomic percentages and sealed off in quartz ampoule under a high vacuum. Further, the evacuated and sealed ampoule was placed in a Temperature Controlled Furnace, where the temperature of the furnace was increases in four steps. Initially, at 673K for 2 h; 873 K for 2 h; 973 K for 3 h and finally 1023 for 4 h. In the furnace, the ampoule was shaken frequently for the homogeneity. Very fast quenching in ice-water bath has been done to obtain the glassy material. The glassy as well and the amorphous nature of the sample was confirmed by DSC measurements. Thin films of thickness 300 nm of $Se_{80}In_5Te_9Sn_6$ glass were prepared on glass/Si wafer substrate by using Edward Coating Unit E-306 operated at 10^{-6} Torr, at a rate of 4 nm/s. To investigate the phase-change study in $Se_{80}In_5Te_9Sn_6$ thin films, films were induced by TEA Nitrogen Laser having pulse width: 1 ns, peak power energy density 3.5 X 10^5 W/cm^2, size of laser spot: 6 mm with peak power 100 kW and wavelength: 337.1 nm for 15, 25 and 35 min

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at room temperature. A Regaku X-ray diffractometer Ultima IV was used for structural analysis of as-deposited and irradiated thin films. The morphological analysis of all films was done by field emission scanning electron microscope (FESEM) (QUANT FEG 450, Amsterdam, Netherlands). A UV–Vis–NIR spectrophotometer (A JASCO, V-500) has been used for optical absorption measurement of as-prepared and irradiated thin films in the wavelength range 400–900 nm.

III. RESULT AND DISCUSSION

The glassy and amorphous nature of the synthesized $Se_{80}In_5Te_9Sn_6$ material was confirmed by a non-isothermal Differential Scanning Calorimetric (DSC) scan performed at a heating rate of 25 K/min (shown in Fig. 1). The endothermic peak corresponds to the glass transition of the sample, as glass transition is a relaxation process that requires heat absorption. Further, the exothermic peak corresponds to the crystallization of the material. The presence of well-defined glass transition (T_g) and crystallization (T_c) peaks verifies the glassy state of the material. The values of T_g and T_c were determined to be 354 K and 398 K, respectively.

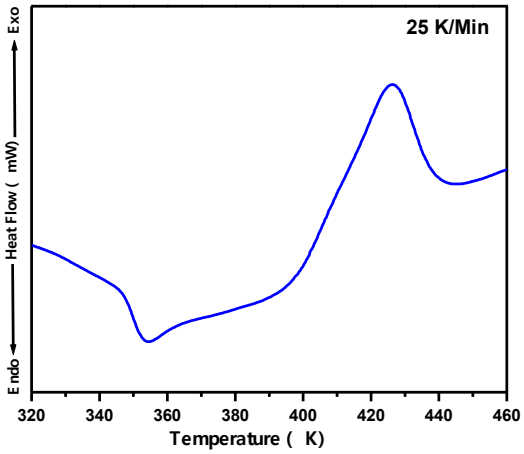

Fig. 1: DSC Thermogram of $Se_{80}In_5Te_9Sn_6$ at heating rate 25 K/min

Structural investigations of the as-prepared and laser-irradiated films were carried out using High Resolution X-ray diffraction (HRXRD) with a copper target (Cu $K\alpha_1$ radiation, λ = 1.5406 Å). The measurements were performed under operating conditions of 40 kV and 30 mA, with a step size of 0.05°, a counting time of 2.5 s per step, and a 2θ scan range from 10° to 70°. Fig. 2 represents the X-Ray diffraction pattern of as-deposited and irradiated thin films. The absence of sharp diffraction peaks in the pattern of as-prepared film confirms its amorphous nature, whereas the appearance of distinct sharp peaks in the irradiated films indicates the development of polycrystalline nature.

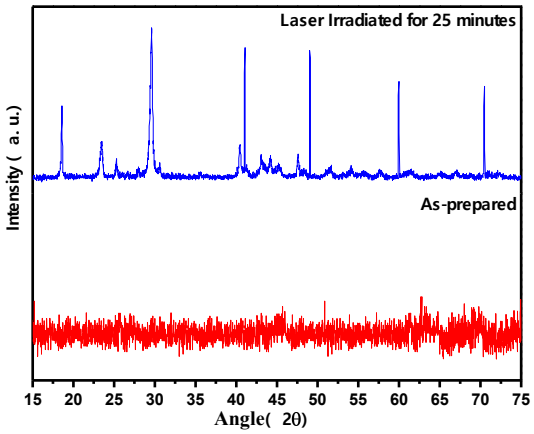

Fig. 2: X-ray pattern for as-prepared and laser irradiated $Se_{80}In_5Te_9Sn_6$ chalcogenide thin films.

The surface morphology of the as-prepared and laser-irradiated $Se_{80}In_5Te_9Sn_6$ chalcogenide glass thin films deposited on Si (100) wafers were examined using field-emission scanning electron microscopy (FESEM). The FESEM measurements were carried out at an accelerating voltage of 20 kV with a working distance of 1 cm. Fig. 3 presents the FESEM micrographs of the as-prepared and laser-irradiated thin films.

The FESEM micrographs indicate noticeable structural changes accompanied by crystal growth as a result of laser irradiation.

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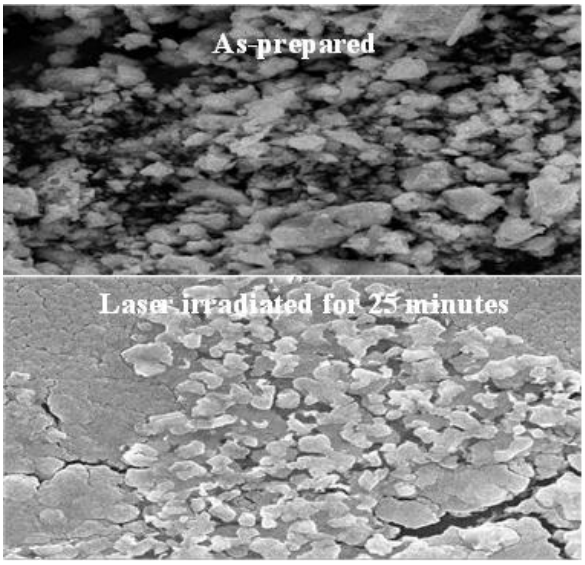

Fig. 3: FESEM Micrographs of as-prepared and Laser irradiated $Se_{80}In_5Te_9Sn_6$ Thin Films

We have recorded the optical absorbance data for as-prepared and laser crystallized thin films of $Se_{80}In_5Te_9Sn_6$ in the wavelength span of 400–900 nm. Using this data, we have been determined various optical parameters such as absorption coefficient (α) and optical band gap (E_g).

The absorbance is the measurement of light absorbed by the sample under explicit conditions. Absorption coefficient (α) has been calculated by using following equation [36] :

α = Optical Absorbance/Thickness of the film (1)

The variations of α with incident radiation energy (hν) for as-deposited and irradiated $Se_{80}In_5Te_9Sn_6$ thin films are shown in Fig. 4.

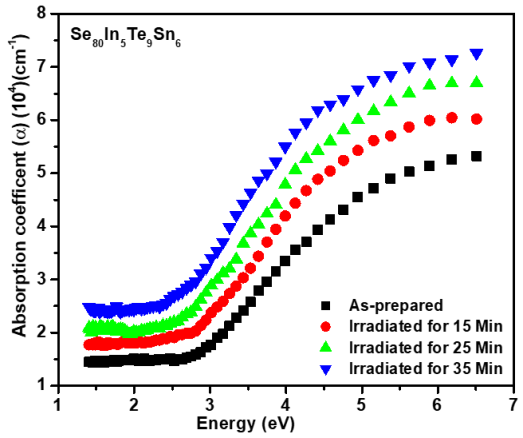

Fig. 4: The plot of α versus incident radiation energy (hν)

From Fig. 4, it has been found that α increases with the incident photon energy as well as the irradiation time. The values of absorption coefficients at 650 nm for radiation time are shown in Table 1.

The dependence of optical band gap on the incident radiation energy and absorption coefficient can be described by the by Tauc's relation [37]:

$$(\alpha h\nu)^k = A' (h\nu - E_g) \quad (2)$$

Where A' is a constant, E_g is optical band gap and k is an exponent which is may be 2, 1/2, 2/3 or 1/3, depending on the transition nature [38]. For $Se_{80}In_5Te_9Sn_6$ thin films, we have obtained the best fit plot for k = 2 which is representing direct transition. The plots of $(\alpha h\nu)^2$ versus incident energy (hν) for as-prepared and laser irradiated $Se_{80}In_5Te_9Sn_6$ thin films are shown in Fig. 5.

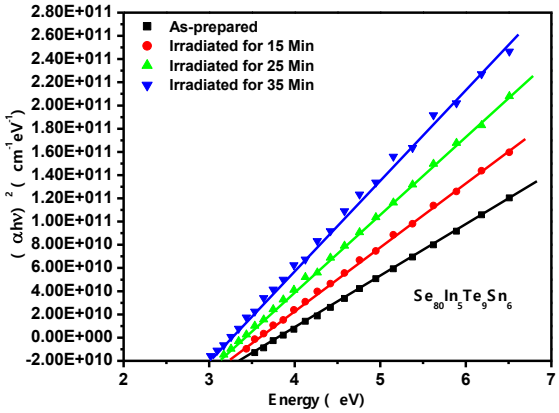

Fig. 5: Variation of $(\alpha h\nu)^2$ with incident radiation energy hν

The values of optical band gap for as-deposited and laser irradiated $Se_{80}In_5Te_9Sn_6$ thin films have been extrapolated by using the intersections on X-axis of the straight line plots of Fig. 5 (Shown in Table 1). Results are indicating that the band gap E_g decreases with the irradiation time which may be due to disorder and defect variation in the present amorphous system [39], [40]. Unsaturated bonds contribute to the formation of localized defect states in the band structure of amorphous solids. These localized states play a crucial role in reducing the optical energy gap. As the number of unsaturated defects increases, the density of localized states in the band structure also increases [41], which consequently leads to a decrease in the optical energy gap E_g [42], [43].

Table-1 Optical parameters of as-prepared and laser-irradiated thin films of $Se_{80}In_5Te_9Sn_6$ chalcogenide glass.

Optical constants	*As-Prepared thin films*	*15 min. laser-irradiated thin films*	*25 min. laser-irradiated thin films*	*35 min. laser-irradiated thin films*
Absorption coefficient $(\alpha)(10^4)$ (cm^{-1}) at 650 nm	1.46	1.87	2.06	2.37
Optical Band Gap $(E_g)(eV)$	3.34	3.23	3.16	3.05

IV. CONCLUSION

In the present research work, we have studied the laser-irradiation effects on the crystal structure and optical constants of $Se_{80}In_5Te_9Sn_6$ thin films. The laser irradiation causes the phase transformation from amorphous to crystalline state. The absorption mechanism is found to be direct transition. The crystallization of $Se_{80}In_5Te_9Sn_6$ thin films by laser irradiation is accompanied by a decrease in the optical band gap with irradiation time. This behavior is attributed to the production of surface dangling bonds around the formed crystallites during the process of crystallization. These transformations were studied using X-ray diffraction and FESEM. The values of absorption coefficient and extinction coefficient are found to increase with increasing the laser-irradiation time. All these parameters are very important in characterizing a material for its applications in various optoelectronic devices. Due to large absorption coefficients and change in structural and optical properties by laser–irradiation, there is a possibility of using them as erasable phase change optical recording material.

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