RESEARCH PAPER

Study of the Effect of Laser Irradiation on Some Structural, Optical and Electrical Savor of $Se_{75}S_{25-x}Sn_x$ Thin Film Ready by Vacuum Thermal Evaporation Technique

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ABSTRACT

This research involves the preparation of (400 ± 20) nm thick $Se_{75}S_{25-x}Sn_x$ thin films by vacuum thermal evaporation method on glass Slides and studying the effect of laser irradiation on some structural, optical and electrical savor of the films. X-ray diffraction analysis reveals that the films have a random structure at (x = 0 and 5) while single crystal growth starts at (x = 10, 15). After laser irradiation, the diffraction pattern shows an improvement in crystal growth and all films are polycrystalline. FESEM examination also shows a clear effect on the surface morphology of the films exposed to the laser. By measuring the transmittance and absorbance spectra in the wavelength scope (400 - 1100 nm), it was found that the transmittance decreases and the absorbance increases as a function of wavelength with increasing tin content before and after irradiation. It is also shown that the absorbance of the films decreases after laser irradiation while the transmittance increases. The energy gap for the allowed direct transition was also calculated and it was found to decrease with increasing tin content before and after irradiation. It also decreases with irradiation, but the effect of irradiation decreases with increasing metal content. Hall effect analysis revealed that the films prepared before irradiation at (x = 0 and 5) are of the P type, while at (x = 10, 15) they transform to the N type, and the conductivity increases with increasing tin content and the resistivity decreases. After irradiation, all films are of the N type, and the conductivity decreases with increasing tin content and the resistivity increases.

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INTRODUCTION

The term "thin film" refers to one or more layers [1] of certain materials with a thickness between one nanometer and one micrometer. [2] The attention of researchers was drawn to the study of the characteristics and properties of materials in the form of thin films at the beginning of the second half of the seventeenth century, and many important studies were conducted in this regard [3,4]. At the beginning of the nineteenth century, the study of thin films developed and advanced from the practical side, with the emergence of many semiconductors, such as selenium and silicon, being used to prepare thin

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films. [5,6] The field of semiconductor physics has entered all areas of daily life and continues to advance, contributing significantly to scientific and technological development. One of the most important technologies that has greatly contributed to the development of the study of semiconductors is thin-film technology. Since ancient times, thin films have been of great importance for use in many different fields [7-9].

Chalcogenide glasses continue to attract the attention of researchers and engineers as a very large group of interesting solid materials, which reveal About unusual physical and chemical phenomena. One of the most famous classes of amorphous materials is the chalcogenides, which have been extensively studied for decades due to their unique properties that attract researchers. It can have applications in optical recording, imaging, microelectronics, amplifiers, optical communications, integrated optics, voltage stabilizers, fiber optics, switches, memories, modulators, display control circuits, converters, sensors, networks, and more. These glassy materials are formed by doping them with other elements such as Ge, S, Te, As, Zn, Sn, Cd, etc. [10-12]. Chalcogenides are extremely interesting in multiple sensing systems used to measure different ions simultaneously in solutions. Exposing glass to laser radiation causes crystallization and/or spatially selected structural modification. The changes induced in amorphous chalcogenides by laser radiation are the subject of systematic studies aimed at better understanding the mechanisms of the phenomena they exhibit, as well as their practical applications. To develop information technology, we need to develop optical recording materials, therefore, it is of great importance to know the linear optical properties of these materials. Extensive research [8-13] is currently being conducted on the effects of laser irradiation, annealing, UV radiation, gamma rays, etc., on the optical and electrical savor of chalcogenide thin films. This research aims to study the effect of laser irradiation on the physical properties of $Se_{75}S_{25-x}Sn_x$ chalcogenide thin films. We chose selenium because selenium-based ternary chalcogenide glass is of interest to us due to its diverse properties, such as high temperature resistance, high sensitivity, efficiency, less aging effect, and high hardness compared to pure selenium. We used sulfur as an additive to the selenium. Tin was placed in the Se-S system.

MODELING AND WORKING METHODS

Thin films of ($Se_{75}S_{25-x}Sn_x$) were prepared using the vacuum evaporation method. The ($Se_{75}S_{25-x}Sn_x$) compound was prepared by mixing proportions of each of its constituent elements with a purity of (99.99%) and according to the (x) values of (0, 5, 10, 15) %. After determining the mass of the mixture (6gm), the mass of each of the constituent elements was calculated. A sensitive electric balance with a sensitivity of four decimal places was used. The mixture was then placed in quartz tubes of a suitable length (15cm). These tubes were evacuated of air using a mechanical vacuum system to prevent oxidation of the compound during the heating process. The tubes were sealed using an oxyacetylene torch. The tubes were then placed in an electric furnace, where the components of the compound were gradually heated from room temperature to a temperature above the melting point of the components (450°C) for two hours since the melting point of selenium is 221°C, sulfur 113°C and tin 232°C, the tubes were placed at an angle of (45°) to ensure greater homogeneity. They were then left for 24 hours gradually cool to room temperature. The tubes were then removed from the furnace to be broken and the compound was extracted from them in the form of a mold. It was then crushed using a hammer and a special laboratory ceramic bowl for this purpose, thus obtaining a powder that was stored in clean, dry containers. Then, an appropriate amount of the compound powder ($Se_{75}S_{25-x}Sn_x$) was placed in a molybdenum boat to obtain the required thickness according to the equation $m= 4\pi p t r^2$[24]. After preparing the evaporation system, the boat was placed between the two electrodes and the substrates bases were fixed, after being thoroughly cleaned, on the sample holder at a distance from the evaporation source in order to get rid of the heat emitted from heating the tank and to ensure the largest possible deposition area. The gravimetric method was used to measure the thickness of the prepared film according to the relationship: $t = m/S.\rho$ [25]. The films thus prepared were irradiated (for approximately 10 seconds) with a continuous wave semiconductor laser operating at a wavelength of 532 nm, a power of 500 mW, and a distance of 36 cm.

RESULTS AND DISCUSSION

Laser irradiation is known to produce defects

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in chalcogenide glasses. Wagner et al. [21] demonstrated optically induced crystallization in amorphous Ag(Sb) This new electronic arrangement causes a change in the optical and electrical properties. [22–24]. In materials with amorphous structure, such as ChG glass, the relationship between electronic properties and disorder is worthy of attention. [25–28]. Parthasarathy et al. [29] Bulk $Se_{1-x}Te_x$ glass is reported to have electrical conductivity that is directly proportional to temperature (up to 77 K) and pressure (up to 8 GPa). Prakash et al. [30] Using the extended pair model, the DC conductivity of chalcogenide glass was obtained. Murphy et al. [31] The DC electrical conductivity and the effect of structure on it of selenium at field strengths less than 100 V/cm² over a temperature range of 300 to 600 K were studied by thermal analysis. [32] reported laser-induced crystallization and deformation of $Se_{80}Te_{20-x}Sb_x$ thin films. They noticed that the energy gap created by distortion and crystallization changed. They interpreted this change in the forbidden energy gap as resulting from a decrease in the disorder of the system and an increase in the size of the grains. Khan et al. [33] investigated the fabrication of as-prepared $Se_{75}S_{25-x}Cd_x$ chalcogenide thin films and thermally annealed them. They demonstrated the variation of optical constants (refractive, absorption, and extinction indices) in the wavelength range 400-1000 nm as a function of photon energy. An increase in the absorption coefficient and forbidden energy gap was observed with increasing annealing temperature. Also, the extinction coefficient (k) and refractive index (n) had the same trend with increasing annealing temperature. Baheshti et al. [34They demonstrated that the optical properties of $Se_{96-x}Te_4Ga_x$ thin films are affected by laser irradiation. They demonstrated that the forbidden energy gap increases in the prepared thin films and exhibits similar behavior with the addition of Ga. The forbidden energy gap decreases after laser irradiationAl-Hazmi [35] studied the effect of laser on the optical properties of $Se_{75}S_{15}Ag_{10}$ thin films. He demonstrated an increase in the forbidden energy gap and absorption coefficient, while, in contrast, with increasing laser irradiation time, there is a decrease in the extinction and refractive coefficients. He interpreted the results using the change in the concentration of localized states resulting from the shift of the Fermi level. When studying $Se_{88}Te_{12-x}Al_x$ films, Uddin et al. [36] observed a decrease in the forbidden energy gap value as the amount of aluminum in the non-radioactive sample increasedThe estimated forbidden energy gap values after laser irradiation of thin films show the same behavior. The increased defect density/particle size is believed to be the reason for the decreased forbidden energy gap due to the increase in metal (Al) ratio/laser exposure time. The forbidden energy gap value also decreased after laser exposure, indicating that the particles size increased after laser exposure. Based on the Mott and Davis model, this decrease in the forbidden energy gap value with the increase in (Al) content in the $Se_{88}Te_{12-x}Al_x$ nanoparticle thin film can be understood. [37]. In this a-$Se_{88}Te_{12-x}Al_x$ system, the decreased forbidden energy gap may be attributed either to the width of the localized states changing due to the presence of unsaturated bonds or crystal defects, or to the localized states having a high concentration in the forbidden energy gap. This may be attributed to the shift in the Fermi level position due to the increasing Al concentration, and the location of the Fermi level is determined by the distribution of electrons in the localized states. [38].

It is clear from all of the above that the presence of radiation-induced defects and irregularities in amorphous materials significantly affects the characteristic features of their physical and chemical properties. Here we study some of the physical properties of unirradiated and irradiated samples of $Se_{75}S_{25-x}Sn_x$ glass.

Structural Properties

X-ray Diffraction

Fig. 1a and b show the X-ray diffraction patterns of $Se_{75}S_{25-x}Sn_x$ films before and after irradiation, respectively. It is clear that at a tin content of 0.5, there are no clear peaks, confirming the random nature of these films. As the tin content increases (10.15), a single peak begins to appear, which becomes more pronounced with increasing tin content. This indicates that increasing the tin content improves the crystallization process [39].

After exposing the thin films to laser radiation, the diffraction pattern indicates polycrystalline growth for all ratios. Crystallinity increases in thin films exposed to laser radiation as the metal content increases. (Sn), but there is a slight creep in the peak positions with increasing tin content. Furthermore, the clarity of some peaks increases, while the intensity of others decreases with

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increasing tin content.

FESEM Analysis

All $Se_{75}S_{25-x}Sn_x$ films were examined using a scanning electron microscope (FESEM) at 500 nm to determine the surface texture of the thin films and observe the effect of tin doping ratios before and after laser irradiation. It was found that varying tin ratios had a clear effect on the surface structure of these films. This is consistent with most random state electron microscopy studies [40]. Fig. 2 shows that the films exhibit a random appearance. However, for the films doped with 15% tin, we observe a granular appearance consisting of small, uniform, and almost homogeneous spherical grains.

Fig. 3 shows that laser irradiation had a clear effect on the surface morphology of the film. At x = 0, we observe the formation of large agglomerates, but the surface remains random. The effect is also clear at x = 5, with smaller, more homogeneous, and less rough agglomerates, with some large

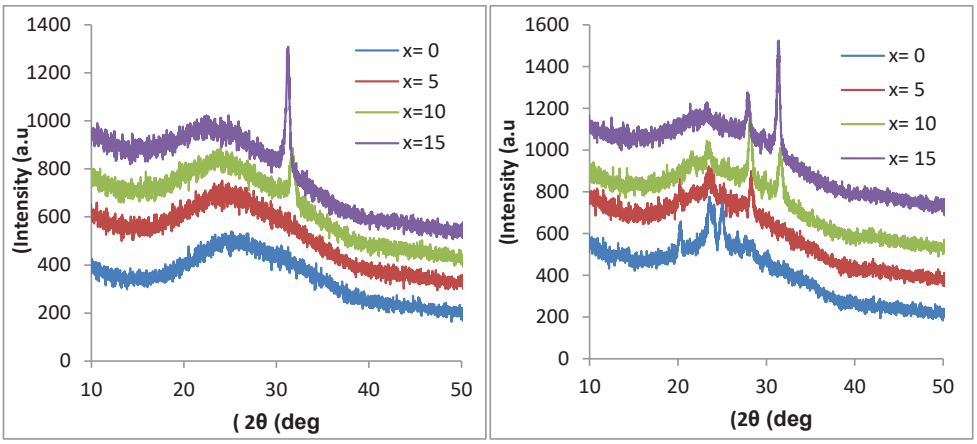

Fig. 1. a,b) XRD patterns of $Se_{75}S_{25-x}Sn_x$ thin films with different tin concentrations films before and after irradiation.

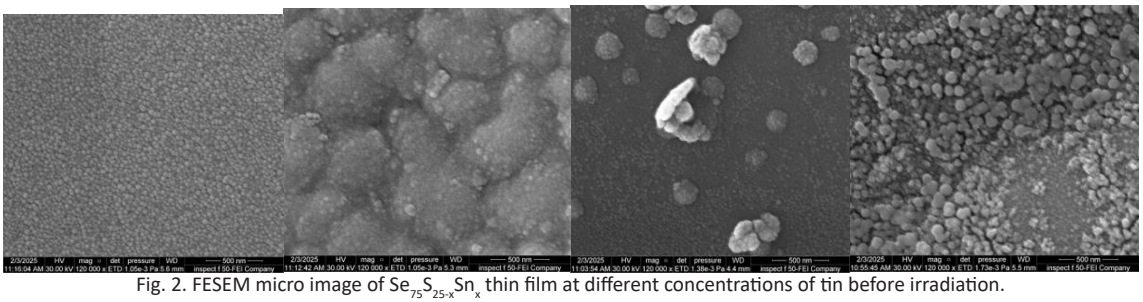

Fig. 2. FESEM micro image of $Se_{75}S_{25-x}Sn_x$ thin film at different concentrations of tin before irradiation.

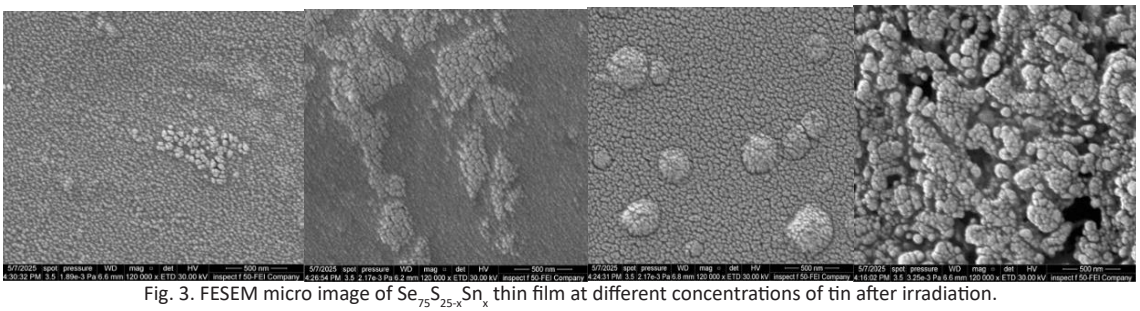

Fig. 3. FESEM micro image of $Se_{75}S_{25-x}Sn_x$ thin film at different concentrations of tin after irradiation.

agglomerates present before and after irradiation. Also, at x=10, the effect is clear and we notice that the surface has become smoother. However, at x=15, the effect is small and hardly noticeable, and the surface consists of small, almost homogeneous spherical grains.

Optical Properties

Absorption

The optical properties of $Se_{75}S_{25-x}Sn_x$ films (x=0, 5, 10, 15) deposited on glass slides by vacuum evaporation were studied. Fig. 4 (a and b) show the change in absorbance (A) of $Se_{75}S_{25-x}Sn_x$ films before and after irradiation as a function of wavelength (λ). We observe a decrease in absorbance values after laser irradiation, and all films have the highest absorbance at wavelengths below 500 nm, reaching (0.45-0.35) for the unirradiated films and (0.24-0.40) for the laser irradiated films. This means that absorption is highest at the beginning of the visible region, where the energy is greater than the energy gaps for direct transitions allowed by the films, which have values of (2.42-2.16 eV) before irradiation and (2.35-2.15 eV) after irradiation, which will be discussed later. This means that the absorbed photons are able to excite electrons in the valence band and move them to the conduction band.

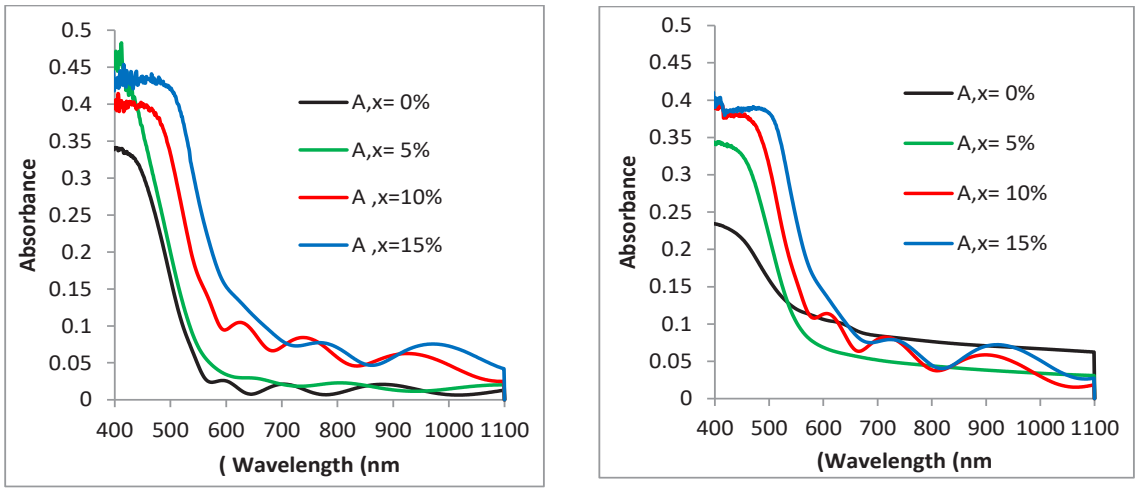

Fig. 4. a,b) Absorption spectrum as a function of wavelength of $Se_{75}S_{25-x}Sn_x$ thin film at different concentrations of tin before and after irradiation.

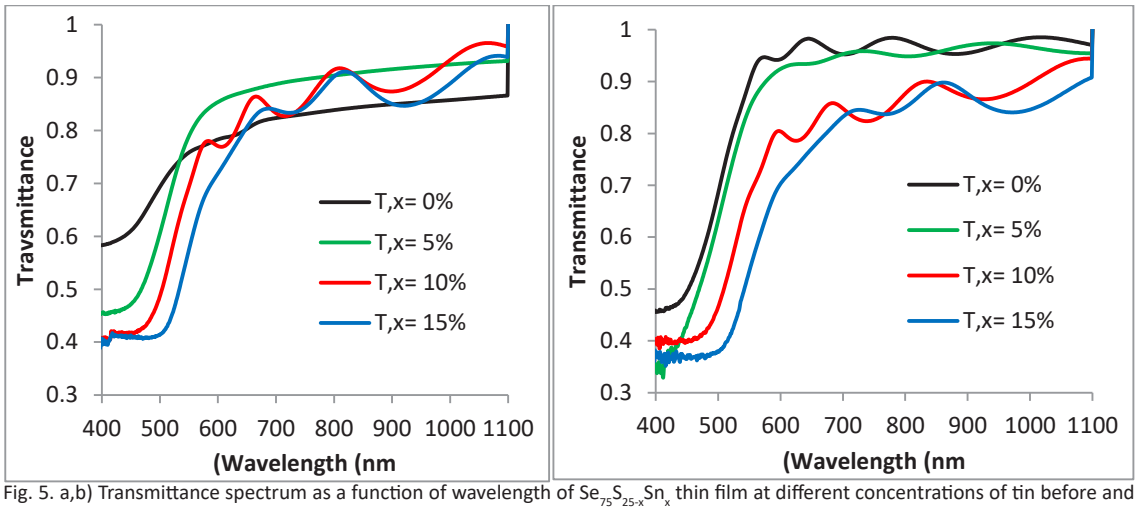

Fig. 5. a,b) Transmittance spectrum as a function of wavelength of $Se_{75}S_{25-x}Sn_x$ thin film at different concentrations of tin before and after irradiation.

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We then observe a sharp decrease in absorption at long wavelengths. We also note that the absorbance increases with increasing tin content before and after laser irradiation as a result of absorption processes resulting from tin doping levels. In other words, the tin levels formed local levels between the valence and conduction bands, acting as auxiliary levels that enabled electrons that did not absorb enough energy to overcome the energy gap to move from the valence band to the conduction band. The decrease in absorbance after irradiation is explained by the increased density of defect states. The sharp edges also shifted towards longer wavelengths [41], with the largest shift at 15%.

Transmittance

Transmittance behaves exactly opposite to absorbance. Fig. 5a and b show the transmittance spectrum as a function of wavelength in the spectrum region (40FF0-1100 nm). We observe an increase in the transmittance of the thin films exposed to laser radiation, and that the transmittance of all films prepared before and after irradiation increases with increasing wavelength, up to the near-infrared region, where

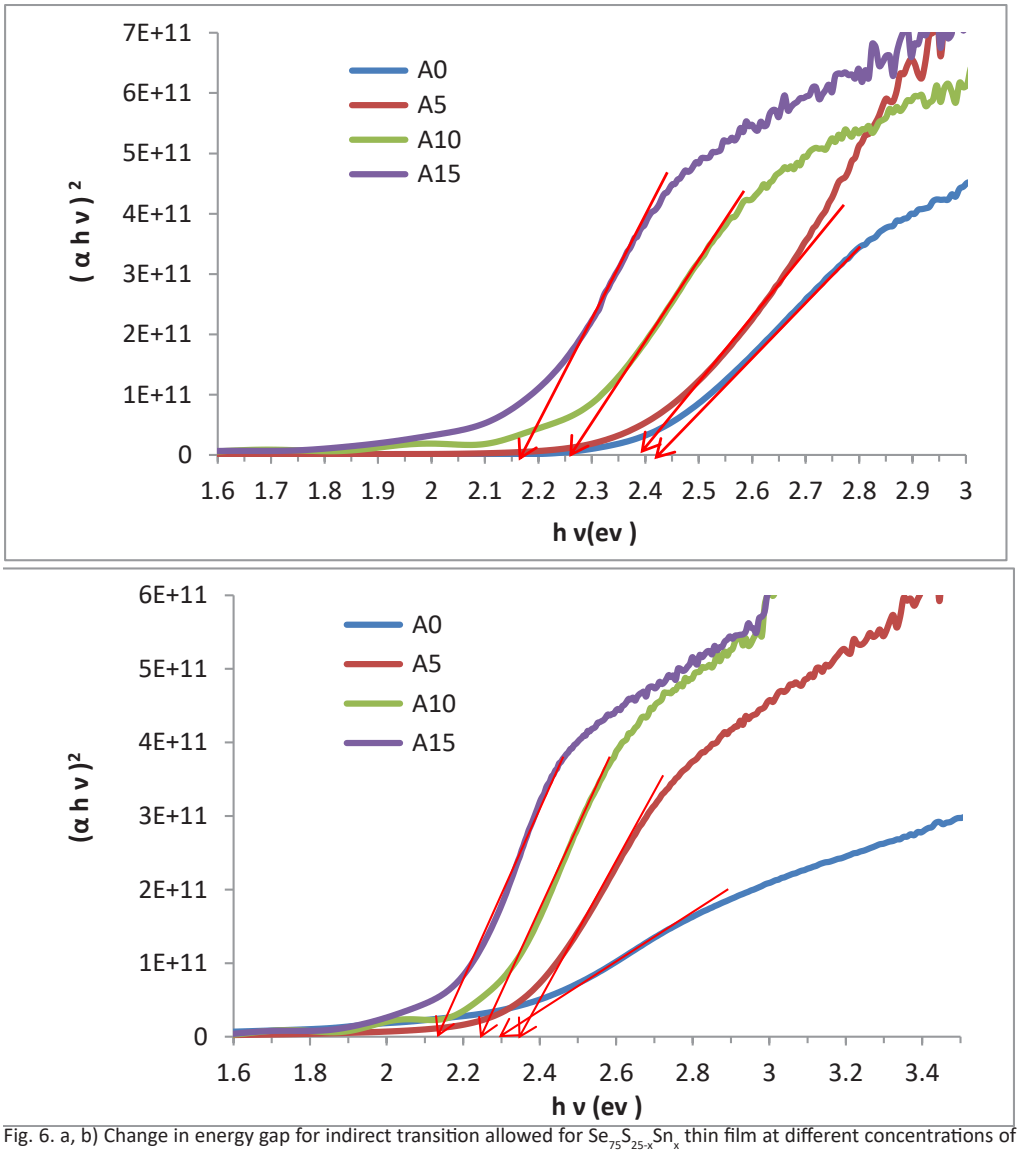

Fig. 6. a, b) Change in energy gap for indirect transition allowed for $Se_{75}S_{25-x}Sn_x$ thin film at different concentrations of tin before and after irradiation.

it extends from 0.36 to 0.46 before irradiation and from 0.42 to 0.58 after irradiation at wavelengths (400-500 nm). This means that the transmittance decreases when the material is doped with tin [42-43]. The decrease in the transmittance spectrum is attributed to the increased light absorption by the localized levels formed by tin within the prepared energy gap. The transmittance increases after laser irradiation due to the increased roughness of the irradiated thin films.

Optical Energy Gap

The value of the optical energy gap for allowed direct transmission was calculated from the equation $\alpha h\upsilon = \beta(h\upsilon - E_g^{opt})^r$ [44] by setting (r = 1/2). We then pFlot the relationship between $(\alpha h\upsilon)^2$ on the y-axis and (hυ) the photon energy on the x-axis. The energy gap is determined from the intersection of the straight line of the curve with the x-axis, which is represented by the photon energy at (α = 0). This point represents the energy gap value, which changes from 2.42 eV to 2.16 eV before irradiation and from 2.35 eV to 2.15 eV after irradiation (Table 1), decreasing with increasing doping ratio. The decrease in the energy gap value with doping with metals is consistent with researchers [45], who used indium, and researchers [46], who used silver. The explanation for this is that tin leads to an increase in the density of localized levels formed by tin atoms within the energy gap. It also decreases with laser irradiation at x=0, 5, while it is slightly affected at x=10, 15, i.e. the effect of irradiation decreases with increasing metal percentage.

Electrical

Hall Effect

A Hall effect experiment was conducted at room temperature on ($Se_{75}S_{25-x}Sn_x$) thin films at x=0.5 and 10.15 to determine their electrical properties, including the concentration, type, and mobility of the dominant charge carriers, as well as their conductivity and resistance. The results in Table 2 show that the conductivity before irradiation is P-type at (x=0.5), where the Hall coefficient is positive, meaning that holes are the dominant charge carriers while electrons represent the minority. However, the conductivity shifts to N-type at (x=10.15), where the Hall coefficient becomes negative, meaning that electrons are the dominant charge carriers, while holes represent the minority. After irradiation, the conductivity becomes N-type at all ratios. We note that as the tin content increases, the conductivity increases, reaching its highest value (31.9*10-6) at (x=15). The resistance decreases as tin content increases.

Table 1 shows the energy gap values of $Se_{75}S_{25-x}Sn_x$ thin film at different concentrations of tin before and after irradiation.

Concentration x	Eg (ev) before irradiation	Eg (ev) after irradiation
0	2.42	2.30
5	2.39	2.35
10	2.26	2.25
15	2.16	2.15

Table 2. Hall effect parameters of of $Se_{75}S_{25-x}Sn_x$ thin film at room temperature before and after irradiation at different concentrations.

variables	before irradiation				after irradiation			
Concentration x	0	5	10	15	0	5	10	15
RH (cm^3/c) *10^7	1.992	1.610	-0.119	-1.616	-0.126	-17.47	-4.099	-0.3714
N(cm^{-3}) *10^{11}	3.133	3.877	-52.26	-3.433	-49.54	-0.3573	-1.523	-16.81
μ (cm^2/v.s)	41.84	56.36	17.87	580	7.54	790.69	9.391	0.01601
ρ (Ω.cm) *10^5	4.762	2.857	0.6682	0.3135	1.671	2.192	43.65	2320
σ $(\Omega.cm)^{-1}$ *10^{-6}	2.1	3.5	14.97	31.90	5.984	4.562	0.2291	0.00431

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After irradiation, conductivity decreases as tin content increases, but it is greater than before irradiation at x=0.5 and significantly lower at x=10.15. Resistivity increases as tin content increases, but it is less than before irradiation at x=0.5 and significantly higher at x=10.15.

CONCLUSION

1. X-ray diffraction (XRD) spectra show the random phase of the thin films, with improved crystallinity as the tin content increases. The laser-irradiated films exhibit a polycrystalline phase.

2. Absorbance decreases and transmittance increases upon laser irradiation. Transmittance decreases and absorbance increases as the tin content increases before and after irradiation.

3. The energy gap narrows with increasing tin content, and laser irradiation also reduces the energy gap.

4. The Hall effect shows that the film transforms from P-type to N-type with increasing tin content, leading to increased conductivity and decreased resistance. Laser irradiation converts all films to N-type, increasing resistivity and decreasing conductivity.

CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this manuscript.

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