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Impact of film thickness on optical and electrical transport properties of noncrystalline $GeSe_{1.4}Sn_{0.6}$ films

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A R T I C L E I N F O

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A B S T R A C T

The optical and electrical properties for $GeSe_{1.4}Sn_{0.6}$ with a different film thickness of 145, 230 and 343 nm were studied. Structural properties affirm the amorphous nature of these films at different thickness. Transmittance (*T*) and Reflectance (*R*) of these films are measured with different film thickness and then the other optical properties are calculated. The calculated linear optical properties illustrated that the optical band gap, phonon energy, refractive index and dielectric constants of these films are increasing with increasing film thickness. On the other hand, the calculated third-order nonlinear optical susceptibility is found to be a function of the film thickness of $GeSe_{1.4}Sn_{0.6}$ films. Regarding the electrical properties, both dc electrical conductivity, and current-voltage dependence are studied for $GeSe_{1.4}Sn_{0.6}$ with different film thickness. The dc electrical conductivity as a function of the temperature is verified the Mott and Davis model. The experimental and calculated results have shown that the present film has two regions on the current-voltage relation. In the ohmic district, the conduction has no traps of electron in the forbidden gap of present films. In the range of higher voltage, the current is expected a limited space charge which controlled by a single trap level.

1. Introduction

Chalcogenide glasses are still interesting materials for different photonic applications, for example, ultrafast optical switches, frequency converters, and optical amplifiers. The enthusiasm for these materials appears from their low phonon energy, long infrared transparency and extraordinary second-/third-order optical nonlinearity [1–3]. The GeSe alloy has an optical band gap (1.1–1.2) eV [4,5]. This range of band gap makes GeSe an effective absorber of incident sunlight. The crystal structure of GeSe is an orthorhombic layered [6,7]. GeSe has a weak van der Waals interaction between the adjacent layers. This structural anisotropy of GeSe leads to the growth tendency of forming a two-dimensional nanostructure [8].

The additional of third element to the chalcogenide component extends the glass shaping zone and makes disorder in composition and configuration in the chalcogenide system. It is expected that this third element gives cross-linked structure consequently changing thermal properties of the binary alloys [9]. The Ge-Se-Sn alloy is one of the ternary systems as the base of GeSe. Numerous researchers have done works on Ge-Se-Sn compound to enhance the thermal and optical properties [10–13]. Petkov *et al.* were studied the optical constants of $GeSe_2$-Sb_2Se_3-AgI films [14]. They found that the estimations of refractive index change from 2.38 for $GeSe_2$ films up to 3.48 for Sb_2Se_3 films while the optical gap decreased from 1.92 to 1.29 eV, respectively. Tang *et al.* have studied the $GeSe_2$-Ga_2Se_3-KBr glasses. They presented the glass-forming region located at Ga_2Se_3/KBr = 1 [15]. As reported in [16] it is expected that the addition of As_2Se_3 to $Ge_{20}Te_{80}$ will increase the glass forming ability and the electrical resistivity. In addition, It has been shown recently [17,18] that adding of an amount of Se improves these qualities of optical properties. Based on a measurement of the glass transition, crystallization and melting the Se adding is the most suitable thermal characteristics [19]. The different film thickness of Ge-Se-Sn is found to adjust the structure and the optical and electrical properties. In this paper, the different film thickness of interest, as-deposited $GeSe_{1.4}Sn_{0.6}$ films with a thickness of 145, 230 and 343 nm are chosen. The thicker films have an interference pattern in the transmission curve. Therefore, the transmission spectrum has an oscillatory curve containing interference maxima and minima. The film thickness range of our $GeSe_{1.4}Sn_{0.6}$ film is below this thickness.

Structural investigations of as-deposited $GeSe_{1.4}Sn_{0.6}$ films with different thickness have been studied. The linear and nonlinear studies have been studied for these films. The third order nonlinear optical

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susceptibility of the present system is calculated. The electrical properties of these films are studied with different film thickness.

2. Experimental details

The $GeSe_{1.4}Sn_{0.6}$ glasses bulk materials were synthetic by the melt-quench method. The materials (Ge, Se, and Sn of 4 N purity) were weighted according to their atomic percentages. These materials were put in an emptied silica tube. This tube was heated at 1273 K for 24 h. The tube was shaken to make the melt homogeneous. The quenching procedure was performed in an ice bath. The choice of this composition renders to the fact that adding Sn to $GeSe_2$ with a percent more than the percentage used would affect the amorphous nature of the studied composition.

The thermal evaporation method was utilizing to fabricate $GeSe_{1.4}Sn_{0.6}$ films with different thickness at a vacuum of 1.34×10^{-5} mbar using a coating unit (HHV Auto 306). The films were grown onto optically and ultrasonically cleaned glass substrates. The substrate was settled onto a rotated holder to form better smooth fabricated films at a separation of 25 cm from the source. The temperature of the substrates was kept at room temperature. The quartz crystal monitor was utilized to control the rate of deposition at 2.7 nm/s and the film thickness, d, to be 145, 230, and 343 nm. Then the interferometric method was used to check the film thickness.

The Structural properties of $GeSe_{1.4}Sn_{0.6}$ films were checked utilizing Philips diffractometer 1710 with Ni sifted CuKα source (λ = 0.154 nm). The transmittance (T) and reflectance (R) were recorded at room temperature utilizing a double-beam spectrophotometer (JASCO, V-570 UV-VIS-NIR). The optical constants (optical absorption coefficient, α, and refractive index, n,) at each wavelength were computed from the rectification values of T and R utilizing a private computer program, which is described previously [20,21].

For the electrical measurements, the films were in the planar configuration for dc conductivity and sandwich configuration for space charge limited conduction (SCLC) measurements. The Ohmic contacts were made by evaporating Au with high purity through masks on the films. The Ohmic contact was checked by $I-V$ measurements at room temperature. A high impedance electrometer (Keithley, Model 610) measures the electrical resistance of $GeSe_{1.4}Sn_{0.6}$ films by the two-probe method. The measurements had been taken at various temperatures going from 300 to 450 K utilizing an electric heater and the temperature was measured by NiCr-NiAl thermocouple checked by a micro-voltmeter.

3. Results and discussion

3.1. Structural investigations

The X-ray diffraction (XRD) measurements are performed in order to define the total phase nature of the films with different thickness. Fig. 1 illustrates the chart of XRD of $GeSe_{1.4}Sn_{0.6}$ films with a thickness of 145, 230 and 343 nm. As illustrated by this figure, amorphous nature of these films is confirmed by absences of sharp diffraction peaks and the existence of hump in the patterns.

3.2. Optical investigations

3.2.1. Linear optical investigations

The investigation of optical constants of a material is an easy technique to clarifying some features concerning the band structure. In the spectral range of 300 to 2500 nm, T and R of $GeSe_{1.4}Sn_{0.6}$ films at different thicknesses of 145, 230 and 343 nm were measured. These measured values are used for calculation the optical constants (k and n) at each wavelength. These values are computed using a private computer program set up by the following equations [20]:

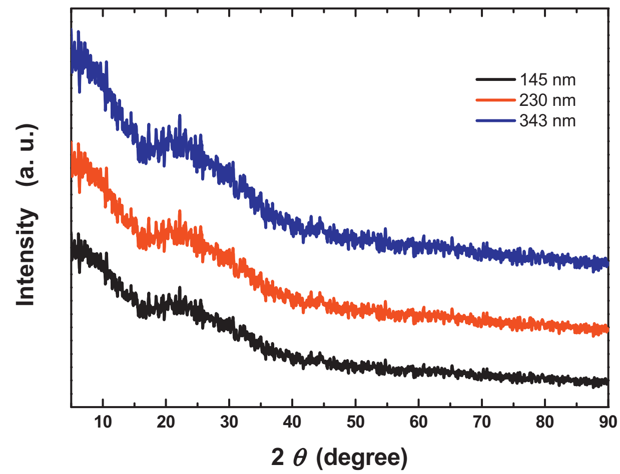

Fig. 1. XRD pattern of $GeSe_{1.4}Sn_{0.6}$ films as a function of the film thickness.

$$T = \frac{(1-R)^2 e^{-\alpha d}}{1 - R^2 e^{-2\alpha d}} \quad (1)$$

and

$$R = \frac{(n-1)^2 + k^2}{(n+1)^2 + k^2} \quad (2)$$

where α is computed from the following equation [22]

$$\alpha = \frac{1}{d} \ln \left[\frac{1-R^2}{2T} + \sqrt{\frac{1-R^4}{4T^2} + R^2} \right] \quad (3)$$

Then extinction coefficient k is given from [23]

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

Using Eq. (4), we can calculate n by the equation [23]:

$$n = \left(\frac{1+R}{1-R} \right) + \sqrt{\frac{4R}{(1-R)^2} - k^2} \quad (5)$$

Considered that the experimental error for d estimation was ± 2.5% and that for T and R computation was ± 1%. Then, the experimental error for n was ± 2.3% and for k was ± 2.1.

The spectral behavior of the dispersion curves of as-deposited $GeSe_{1.4}Sn_{0.6}$ films with a thickness of 145, 230 and 343 nm are illustrated in Fig. 2a. It is observed that the value of n has a maximum peak around λ ~ 750 nm. The decrease of n with λ after λ ~ 750 nm demonstrates the normal dispersion behavior of this system. On the other side, the increase in n with the increase of film thickness 145, 230 and 343 nm means an increase in the current carriers [24]. This behavior of the refractive index is due to the increase in the optical band gap energy which will be observed later.

The same behavior is clearly depicted in some previous works [25–28]. In [25], the authors studied the effect of the film thickness for $Ge_{20}Se_{70}Ag_{10}$ thin film in the normal dispersion range. The authors attributed an increase of the refracive index with film thickness to the discontinuity of the film in the initial stage of the deposition and for the thicker film, the discontinuity of the film achieves later than those of the thinner films.

Additionally, the refractive index of ZnO films increases with increasing thickness due to the change of the layer structure which include porosity, roughness and grain boundaries [26]. Furtherance, For the as-deposited and annealed In_2S_3 thin films, the refractive index of is found in the range 3.88–3.92 and 3.88–4.06 respectively. This increasing could be attributed to the change in the packing density, strain and improvement in crystallinity [27,28]. The dependence of the spectra of k on λ for as-deposited $GeSe_{1.4}Sn_{0.6}$ films with a thickness of