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The Effect of the Deposition Parameters on the Microstructural and Optical Properties of Mn-Doped GeTe Chalcogenide Materials

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***Abstract*—**In this work, the effect of the magnetron sputtering system parameters on the optical properties of the Mn doped GeTe were investigated. The optical properties of the $Ge_{1-x}Mn_xTe$ thin films with different thicknesses are determined by analyzing the transmittance and reflectance data. The energy band gaps of the amorphous Mn-doped GeTe thin films with different thicknesses were calculated. The obtained results demonstrated that the energy band gap values of the amorphous films are quite different and they are dependent on the films thicknesses. The extinction coefficients of amorphous Mn-doped GeTe thin films as function of wavelength for different thicknesses were measured. The results showed that the extinction coefficients of all films are varying inversely with their optical transmission. Moreover, the results emphasis that, not only the microstructure, electrical and magnetic properties of Mn doped GeTe thin films vary with the films thicknesses but also the optical properties differ with the film thickness.

***Keywords*—**Phase change magnetic materials, transmittance, absorbance, extinction coefficients.

I. Introduction

An optical property of a material is defined as its interaction with electromagnetic radiation in the visible light [1], [2]. Interaction of light waves with the electronic or crystal structure of a material leads to a number of phenomena such as absorption, reflection and transmission [3], [4]. Therefore, the total intensity of the incident light striking a surface at any instance of light interaction with a material is equal to sum of the absorbed, reflected, and transmitted intensities.

In principle, the optical properties of thin films depend on various deposition parameters, such as thickness, substrate temperature, type of substrate, deposition rate etc. Therefore, the study of thickness dependence of optical constants of the transition metal doped chalcogenide semiconductor materials thin films are very important for understanding the mechanism of the optical processing [5].

Chalcogenide phase change semiconductor materials have significant optical properties such as high linear refractive index, broad IR transmitting optical windows and possibility to change its physical properties with composition [6]-[8]. The importance of these materials is due to their potential use in integrated optics, optical recording, optical data storage media and non-volatile phase change memories.

Over the past few years, the effect of the film thickness (from 10 to 30 nm) on the optical properties of the chalcogenide based-phase change materials, $Ge_1Sb_2Te_4$, was studied by [9]. They reported that the effect of the film thickness below 30 nm on the optical parameters of $Ge_1Sb_2Te_4$ are not constants, but depend on the films thicknesses.

Although much research has been done on optical properties of IV-VI diluted magnetic semiconductors (DMSs) compounds and PCM thin films, there are very few reports available on the effect of deposition parameters on the determination of optical constants and bandgap of the phase change magnetic materials (PCMM) thin film [10]-[12]. Therefore, in the present study, the optical properties of amorphous Mn-doped GeTe thin films with different thicknesses have been investigated. The experimental results demonstrate that, the average transmission spectra, the energy band gaps and the optical constants of Mn-doped GeTe thin films with different thicknesses clearly differ when the thickness changed.

II. Experimental Details

The Mn-doped GeTe thin films were deposited on glass substrates at room temperature by using RF magnetron sputtering technique. In order to investigate the influence of the film thickness into the optical properties of the amorphous $Ge_{1-x}Mn_xTe$ thin films, a set of four samples were prepared by changing the deposition time to yield films with different thicknesses. The thicknesses of the films were measured using step test system and field emission scanning electron microscopy (FESEM). The thicknesses of these samples were found to be 60 nm, 120 nm, 200 nm and 240 nm. The $Ge_{1-x}Mn_xTe$ thin film sputtering was carried out under argon gas with flow rate of 30 sccm. The background pressure maintained at 10^{-4} mTorr and the pressure during sputtering was 3 mTorr. The sputtering power for all films was fixed at 50 W. The microstructure of the films was analyzed by X-ray diffractometer (XRD). The thicknesses of the films were measured using step test system and field emission scanning electron microscopy (FESEM). The optical properties of Mn-doped GeTe thin films with different thicknesses were brought out using UV/VIS/NIR spectrophotometer (Perkin Elmer LAMBDA 950).

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III. Results and Discussions

The microstructure of Mn-doped GeTe thin films with different thicknesses of all samples were investigated [13]. The X-ray diffraction patterns demonstrate that the as-deposited Mn-doped GeTe thin films with difference thicknesses are amorphous.

Fig. 1 shows the average transmission spectra of the $Ge_{1-x}Mn_xTe$ thin film with thickness 60, 120, 200 and 240 nm. The optical transmission data of as-deposited films were measured in the wavelength range of 300-1100 nm. The obtained results showed that the amorphous films exhibited more than 45% transmittance. Moreover, the increment of the film thickness leads to the decrease in the transmittance. The transmittance decrease with increasing the thickness through the overall wavelengths was attributed to the induced increase of optical absorption as the film thickness increased. Similar behavior for the film thickness dependence of transmittance and absorbance has been previously reported for the chalcogenide semiconductor thin films [14], [15]. Furthermore, in the high wavelength, the transmittance curves show interference pattern of all films except that for the film with thickness 240 nm. This indicated that the studied thicknesses were enough to induce the envelope of the interference maxima and minima, which are very important to determine the optical constants (refractive index (n), extinction coefficient (κ), and absorption coefficient (α).

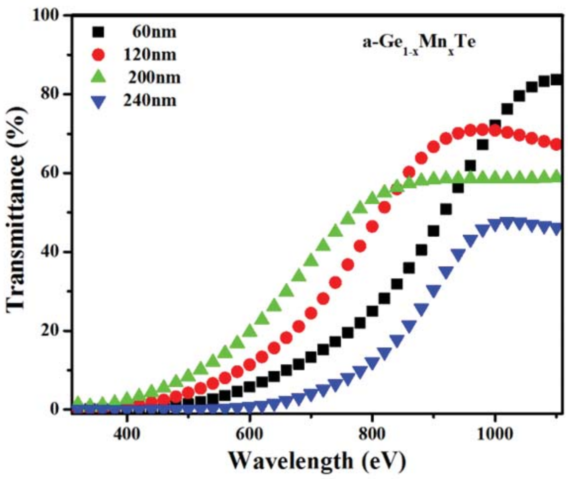

Fig. 1 The average transmission spectra of the Mn- doped GeTe thin films with various thicknesses

For further investigation of optical properties, the absorption spectra of the amorphous $Ge_{1-x}Mn_xTe$ thin film with different thickness are shown in Fig. 2. It can be observed that, the absorption edge of the film with thickness 60, 120 and 200 nm were centered at 340 nm while the absorption edge of the film with a thickness of 240 nm was centered at the 400 nm. Furthermore, the absorbance was high in the short wavelength region and decreased sharply beyond the edge to become approximately zero in the long-wavelength region. This could be related to the point defects.

In principle the mechanism of the absorption is a process in which the energy of a Photon was taken when the electron was excited from the valence band to the conduction band. Actually, there are two types of optical transitions that can occur at the fundamental edge of the semiconductor, direct and indirect. Both of them require the interaction of an electromagnetic wave with an electron in the valence band. It is well known that GeTe is a p-type degenerate semiconductor with a high carrier concentration, a narrow band gap and a direct optical absorption edge [10], [16]. Thus, we suppose that the secondary phase of Mn doped GeTe like MnTe is also a p-type direct band gap semiconductor. Since both GeTe and MnTe are direct bandgap semiconductors, the $Ge_{1-x}Mn_xTe$ is a direct bandgap material. Therefore, the optical band gap of $Ge_{1-x}Mn_xTe$ thin film can be estimated from absorption coefficient data as a function of wavelength by using Tauc Relation as shown in the following [15].

$$\alpha(h\nu) = A(\mathrm{h\nu} - \mathrm{E_0})^2 \quad (1)$$

where A is a constant, hν is the photon energy of the light, E_0 is the ground state energy.

The absorption coefficient can be calculated by using the transmittance (T) and the thickness (d) of the thin film.

$$\alpha = -\frac{1}{d} ln \frac{1}{T} \quad (2)$$

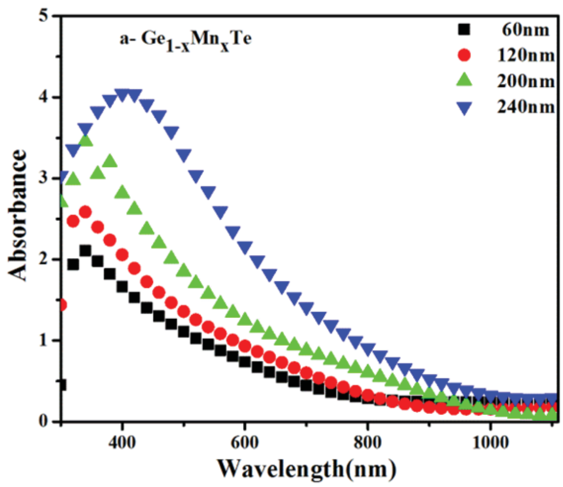

Fig. 2 The absorption spectra of the amorphous Mn-doped GeTe thin films with different thickness

The estimating optical energy band gaps of a-$Ge_{1-x}Mn_xTe$ thin films have been done by using the linear extrapolation of the absorption coefficients. Fig. 3 shows the photon energy (hν) versus absorption coefficient $(\alpha h\nu)^2$ plotting of amorphous $Ge_{1-x}Mn_xTe$ films with thicknesses of 60, 120, 200 and 240 nm. The calculated energy band gap values of these films are found to be 2.6, 2.4, 2.15 and 1.8 eV respectively. The results showed that the energy band gap values of a- $Ge_{1-x}Mn_xTe$ thin films are similar to the nonmagnetic semiconductor counterpart. The straight line was the linear fitting to the experimental data. The obtained results of the energy band gap values are in the manner reverse proportional with the thickness, when the film thickness increases the values of energies band gap were decreases. This may be resulting from the deposition method where in order to increase the thickness of the film, much more atoms were placed on the film leading to unsaturated bonds which are responsible for the formation of some defects and localized states, thereby decreasing the optical energy band gap.

The optical constants such as extinction coefficient (κ) and refractive index (n) of thin films are very important physical parameters, which characterize how a material responds to an electromagnetic field excitation at a given frequency. The extinction coefficient κ can be calculated from the following formula [3].

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

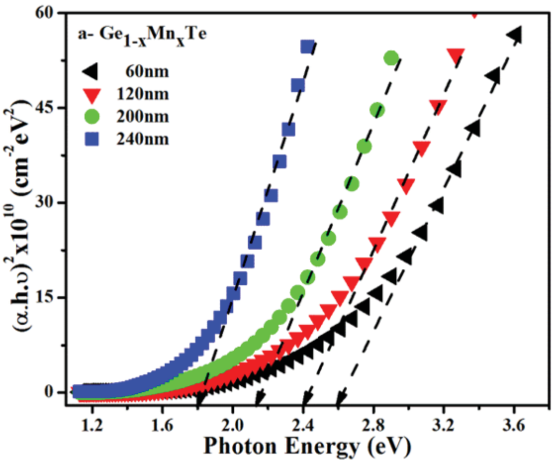

Fig. 3 The absorption coefficient $(\alpha h\nu)^2$ as a function of the phonon energy (hν) of Mn-doped GeTe thin films with different thicknesses

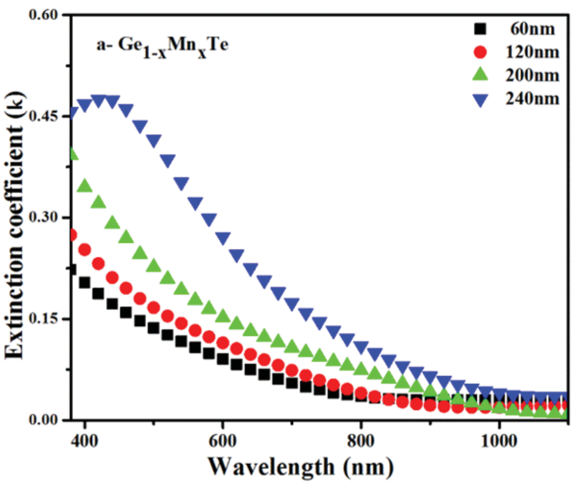

Fig. 4 The extinction coefficient a function of wavelength of Mn-doped GeTe thin films with different thicknesses

Fig. 4 shows the extinction coefficient of amorphous $Ge_{1-x}Mn_xTe$ thin films as function of wavelength for different thicknesses. It also observed that the extinction coefficients of amorphous $Ge_{1-x}Mn_xTe$ thin films decrease with the thickness increased. The obtaining results demonstrated that the optical properties (extinction coefficients) of all films are inversely various with the optical transmission. In other words, a film with high transmittance has low optical constants. These results indicate that the film thickness had great influence on optical constants.

IV. Conclusion

It can be concluded that the optical transmission, absorption coefficient, the energies band gaps corresponding to absorption coefficients and the extinction coefficient of the amorphous $Ge_{1-x}Mn_xTe$ thin films with different thickness were performed. The obtained results for the optical properties of the amorphous films indicated that there is a significant difference between the films when the thickness of the films differs. Moreover, the results also demonstrated that the optical properties of Mn-doped GeTe thin films are dependent not only on composition and the film structure, but also on the film thickness.

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