

EFFECT OF DEPOSITION PRESSURE ON THE OPTICAL AND STRUCTURAL PROPERTIES OF NANO GTC FILMS

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XRD of thermally evaporated $\text{Ge}_{0.15}\text{Te}_{0.80}\text{Cu}_{0.05}$ thin films at higher deposition pressure show amorphous state while the lower deposited films show poly crystalline state. Different preferred orientations were identified; (222), (411) for GeTe_4 , (101), (003) for Te and (311) for GeTe. The corresponding crystallites sizes are 5,10,11,10 and 9 nm respectively. The lower deposition pressure, LDP develops the optical properties; direct, indirect optical gap and phonon energy are increasing as a function of deposition pressure. Optical gap was affected by carrier concentration. Burstein-Moss effect, B-M effect was used to discuss the variation of the optical properties.

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1. Introduction

Chalcogenides, Ch are compounds or alloys have at least one of the following chalcogen elements; Se, Te and S. Ch. alloys have many advantages; large scale production, tuned properties, doped materials, phase change and semiconducting materials, memory medium, IR applications, high values of refractive index, covalent bond and easy prepared materials [1]. Pressure is an important parameter that may affect the properties of materials. It can be divided into low, normal and hydrostatic pressure, the scope of the current study is low pressure. GeTeCu alloys, GTC have many extensive work due to their modern technological applications; optical data storage [2], switching, semiconducting lasers [3], potential thermoelectric materials [4], infrared transmission, photoconductivity, photo resistors, photonic and electronic devices [5], thin film transistors [6], Bragg-reflector fibers [7], spherical and cylindrical photo-induced lenses [8], solar cells, solid-state battery, light transmitting media, optical integrated circuits [9] optical amplifier [10], random access memory, RAM [11], ion sensors [12]. Also, Ch. can be used as photo darkening, photo bleaching and oscillation phenomena materials [9]. Amorphous Ch was classified to elemental, binary, ternary Ch, etc. Also they divided alloys to stoichiometric and non-stoichiometric alloys [9].

It was mentioned that glass transition and crystallization depend on pressure [13-15]. System of GTC alloys can be introduced with 13-20 at.%, 73-85 at.% and 9-10 at.% of Ge, Te and Cu respectively. Homogeneity, the crystallization ability, the thermal stability and consequently the structural, optical and electrical properties of GTC alloys can be enhanced by the high ratios of Cu at the expense of Te atoms [16-20]. Deposition and growth conditions may affect the final characterization of produced materials; thin, thick, bulk and liquid. The effect of pressure on the optical properties of semiconductors was reported in 1958 for the first time and many researchers tried to discuss these conditions [21-25]. They found a great pressure effect on the structural, electrical and optical properties of materials and the color of material may be changed because of pressure effect [26]. No available information about the effect of deposition pressure on the

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properties of GTC alloys. According to the aforesaid, Ge:Te:Cu of 15:80:5 at.%, or $\text{Ge}_{0.15}\text{Te}_{0.80}\text{Cu}_{0.05}$ is considered for this study as a candidate of GTC films. So, we believe the study and the discussion of the structural and optical properties of $\text{Ge}_{0.15}\text{Te}_{0.80}\text{Cu}_{0.05}$ thin films as a function of deposition pressure will provide new insights and information to the data base.

2. Experimental technique

2.1. Materials

Ge, Te and Cu elements of 6N purity from Sigma Aldrich chemicals were used to prepare $\text{Ge}_{0.15}\text{Te}_{0.80}\text{Cu}_{0.05}$ alloy.

2.2. Preparation of $\text{Ge}_{0.15}\text{Te}_{0.80}\text{Cu}_{0.05}$ alloy

Appropriate weights of the mentioned elements were sealed in a quartz ampoule of 1.2 cm diameter and 15 cm length and pressure of 10^{-4} Torr. The constituents were heated up to 1373K for 24 hrs. The ampoule was rapid quenched in ice-water bath to obtain the bulk alloy of $\text{Ge}_{0.15}\text{Te}_{0.80}\text{Cu}_{0.05}$.

2.3. Preparation of $\text{Ge}_{0.15}\text{Te}_{0.80}\text{Cu}_{0.05}$ films

Thermal evaporation technique is the most common preparation method of Ch films [13] so $\text{Ge}_{0.15}\text{Te}_{0.80}\text{Cu}_{0.05}$ thin films were deposited on quartz substrates by thermal evaporation technique at different values of deposition pressure [27] using Edwards coating unit 306A. Pressurized air is used as a supplied working gas within pressure range 4×10^{-3} to 4×10^{-5} Torr.

2.4. Measurements

The prepared films were used to study the structural and optical properties as a function of deposition pressure by using X-ray diffraction, XRD and spectrophotometer. Both transmittance, T and reflectance, R were measured to study the optical properties of GTC films. More experimental details are available at [17,28,29].

3. Results and discussions

3.1. Structural properties

The amorphous nature characterizes the as prepared films deposited at 4×10^{-3} Torr due to the familiar and the well known hump of amorphous materials. Fig. 1 shows the XRD of $\text{Ge}_{0.15}\text{Te}_{0.80}\text{Cu}_{0.05}$ thin films deposited at 4×10^{-5} Torr. LDP develops the structural properties of GTC films; many preferred orientations were identified; (222) and (411) at 2θ of 27.88° and 34.44° for GeTe_4 , (101) and (003) at 29.847° and 45.02° for Te and (311) at 2θ of 50.78° for GeTe. The size of the crystallites of the indicated preferred orientations is calculated by using Scherrer's equation [18,30-33]:

$$D = \frac{K\lambda}{\beta \cos \theta} \quad (1)$$

D , the mean crystallite dimension ($\text{\AA}$), K is a constant (typically assumed to be 1), β is the full width half maximum and λ is the wavelength of the X-ray used ($\text{CuK}\alpha$ radiation) equal to $1.54056 \text{ \AA}$. The obtained crystallites sizes of the mentioned preferred orientations are 5, 10, 11, 10 and 9 nm. Unidentified preferred orientation at 32.58° was also observed. It is concluded that LDP induces different phases and consequently different microstructures which led to increase the

crystallite size of the prepared films within nano size and to enhance the structural properties of GTC films[34].

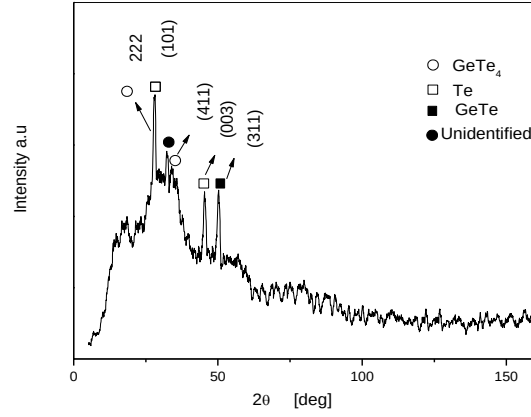


Fig. 1. XRD of nano $Ge_{0.15}Te_{0.80}Cu_{0.05}$ thin films deposited at 4×10^{-5} Torr.

3.2. Optical properties

3.2.1. Spectral distribution of T and R

Fig. 2 and 3 show the spectral distributions of T and R of $Ge_{0.15}Te_{0.80}Cu_{0.05}$. Two regions were observed; the first one is located at $\lambda \leq 1280$ nm and characterized by absorption processes. Many optical constants can be calculated through this region such as absorption coefficient, α , absorption index, k and optical gap, E_g^{opt} – direct, E_{gd}^{opt} or indirect, E_{gi}^{opt} . The second region is located at $\lambda > 1280$ nm and characterized by non absorption processes [35]. Dielectric constant, ϵ and its derivatives; real part ϵ_1 , imaginary part, ϵ_2 , loss tangent, $\tan(\delta)$ and other constants; N/m^* , lattice dielectric constant, ϵ_L infinite wavelength dielectric constant, ϵ_∞ , oscillation energy, E_o , dispersion energy, E_d , real part, σ_1 , imaginary part σ_2 of optical conductivity, σ and the two parts of ELF; surface energy loss function, SELF and volume energy loss function, VELF can be studied and calculated through this region. It could be noticed that, T increases with LDP and shifts to the lower wavelength (blue shift) while R is vice versa. Around $\lambda = 1200$ nm, one peak could be distinguished and attributed to the excitons process; excitons and excitations processes are involved in the absorption process as indicated in Figs. 2 and 3.

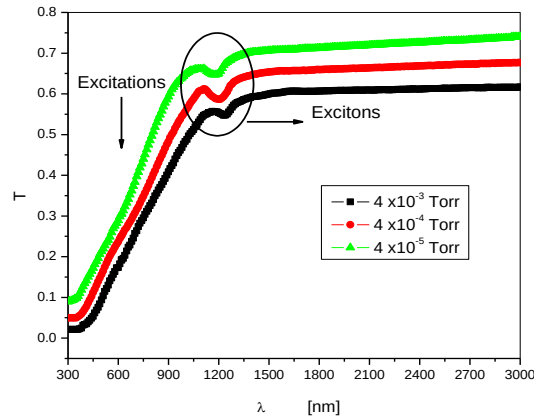


Fig.2. T of nano $Ge_{0.15}Te_{0.80}Cu_{0.05}$ thin films at different deposition pressures.

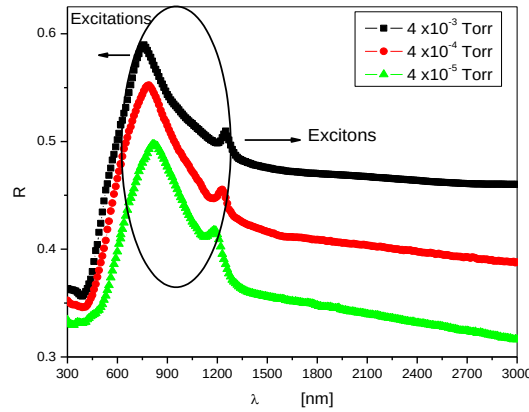


Fig.3.R of nano $Ge_{0.15}Te_{0.80}Cu_{0.05}$ thin films at different deposition pressures.

3.2.2. Optical gap calculations

α and k could be calculated using T , R and thickness, d according to the following equations: [18,35-37].

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

$$\alpha = \frac{1}{d} \ln \frac{(1 - R)^2}{T} \quad (3)$$

Values of α are included within high absorption region; $\alpha \geq 10^4 \text{ cm}^{-1}$ [35-40].

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

Optical gap, E_g^{opt} may be direct E_{gd}^{opt} or indirect E_{gi}^{opt} according to the following formula;

$$(ah\nu) = A(h\nu - E_g^{opt})^r \quad (5)$$

$h\nu$ is photon energy and r is the transition coefficient. The value of r identify the type of the electronic transition; direct or indirect. Its values are 1/2 and 2 for allowed direct and indirect transition respectively. It has also values of 3/2 and 3 for forbidden direct and indirect transition respectively. More details about conditions of r values are available at [35,36,41,42].

The quality of material – ordered or disordered - can be inferred from the values of constant A [18,35,43,44];

$$A = 4\pi\sigma_{\min} / nc\Delta E_{\text{tail}} \quad (6)$$

; $\sigma_{\min}$ is the minimum metallic conductivity, c is the velocity of light, and $\Delta E_{\text{tail}} = \Delta E_c - \Delta E_v$ represents the band tailing [35,43]. According to [35,36,45] our E_g^{opt} values are E_{gi}^{opt} and E_{gd}^{opt} and can be calculated using the well known equations;

$$(ah\nu)^{1/2} = A(h\nu - E_{gi}^{opt}) \quad (7)$$

$$(ah\nu)^2 = A(h\nu - E_{gd}^{opt}) \quad (8)$$

The slope of linear part of equations 7 and 8 and the intercept are used to calculate both E_{gi}^{opt} , E_{gd}^{opt} at $(\alpha h\nu)^{1/2}$ and $(\alpha h\nu)^2 = 0$ and the constant A respectively. As depicted in Fig's.4,5; both E_{gi}^{opt} , E_{gd}^{opt} are found. Due to the presence of excitons, two values of E_{gi}^{opt} ; E_{gi1}^{opt} and E_{gi2}^{opt} are calculated. Because of phonon energy that assisting the indirect transition [36,46], Eq.7 could be written as:

$$(\alpha h\nu)^{1/2} = A(h\nu - E_{gi}^{opt} \pm E_{phonon}) \quad (9)$$

E_{phonon} is the phonon energy assisting the indirect transition. The average indirect optical gap, E_{av-gi}^{opt} is calculated according to [35,36]

$$E_{av-gi}^{opt} = \frac{E_{gi1} + E_{gi2}}{2} \quad (10)$$

E_{av-gi}^{opt} , E_{gd}^{opt} and E_{phonon} are increasing with LDP and the values of constant A as well, such this increasing releases the enhancement of material's order; XRD data confirm this issue. E_{av-gi}^{opt} and E_{gd}^{opt} have different values, such this difference may attributed to spin orbit splitting [47]. The obtained data are summarized in Table 1. A widening of the optical gaps E_{av-gi}^{opt} , E_{gd}^{opt} and E_{phonon} energy were observed [48] due to the lower deposition pressure. The blue shift of the absorption edge was interpreted in the view point of the band-filling effect. The phenomenon of increasing values of E_{av-gi}^{opt} , E_{gd}^{opt} and E_{phonon} with LDP or the shift of the optical absorbing edge to the shorter wavelength is called B-M effect. More details about B-M effect are available at [49-51]. B-M effect attributes the optical gap broadening to the carrier concentration and consequently the lifting of the Fermi level through the conducting band of the degenerate semiconductor.

$$\Delta E_g^{opt} = \frac{\hbar^2}{2m^*} (3\pi^2 N)^{2/3} \quad (11)$$

; $\hbar$ is Plank's constant, m^* is the electron effective mass in the conduction band and N is the carrier concentration. The increase of the optical gap with the lower of deposition pressure can be attributed to the increase of carrier concentration, the values of N/m^* in Table 1 confirm the proposed discussion[52-54]. Also, the increasing of the optical gap may also attributed to the formation of different phases in the lower pressure deposited films which led to different microstructure and consequently different optical gaps as previously obtained from XRD and optical data [34].

Table 1. Optical constants of nano $Ge_{0.15}Te_{0.80}Cu_{0.05}$ films as a function of deposition pressure.

Deposition pressure [Torr]	$A \times 10^5$ ($cm^{-1}eV^{-1}$)	E_{gd}^{opt} [eV]	E_{gi1}^{opt} [eV]	E_{gi2}^{opt} [eV]	E_{gi-tot} [eV]	E_{ph} meV	$N/m^* \times 10^{56}$ ($kg^{-1}m^{-3}$)	ϵ_L	N	E_d [eV]	E_0 [eV]	ϵ_∞
4×10^{-3}	4.7	2.2	0.48	0.75	0.62	0.27	1.60	29.9	5.5	34.4	1.4	26
4×10^{-4}	5.3	2.3	0.59	0.89	1.48	0.30	1.85	22.1	4.7	31.5	1.89	17
4×10^{-5}	5.7	2.5	0.66	1.10	1.76	0.44	1.88	16.6	4.1	27.6	2.3	13

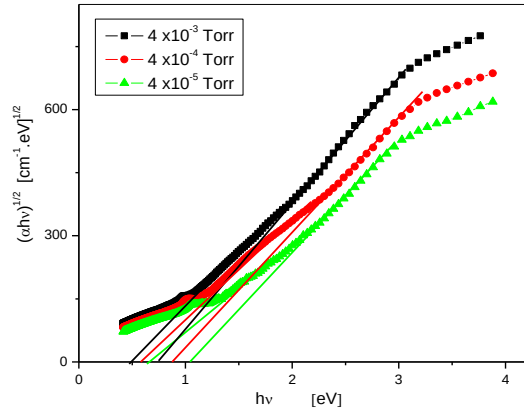


Fig.4. $(\alpha h\nu)^{1/2}$ Vs $(h\nu)$ of nano $Ge_{0.15}Te_{0.80}Cu_{0.05}$ thin films at different deposition pressures.

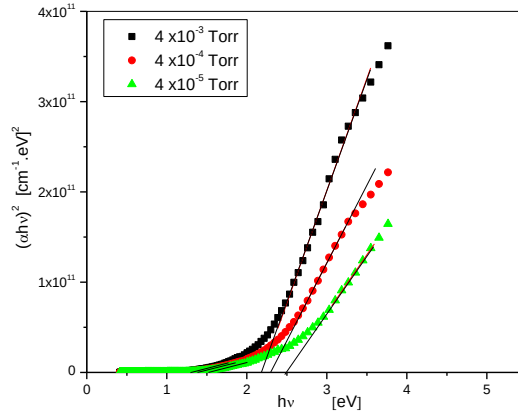


Fig.5. $(\alpha h\nu)^2$ Vs $(h\nu)$ of nano $Ge_{0.15}Te_{0.80}Cu_{0.05}$ thin films at different deposition pressures.

3.2.3. Refractive index and dielectric constant

Refractive index and extinction coefficient n and k can be calculated using the following equation[35,36,55]:

$$n = \frac{1+R}{1-R} + \sqrt{\frac{4R}{(1-R)^2} - k^2} \quad (12)$$

The values of E_o , E_d and ϵ_∞ were calculated by using Wemple–DiDomenico (WDD) Model [18,41,56] according to the following equation;

$$\left(n^2(h\nu) - 1\right)^{-1} = -\frac{1}{E_o E_d} (h\nu)^2 + \frac{E_o}{E_d} \quad (13)$$

from the slope of the linear part and the extrapolation of $\left(n^2(h\nu) - 1\right)^{-1}$ Vs $(h\nu)^2$ relation at $(h\nu)^2 = 0$ as depicted in Fig.6.

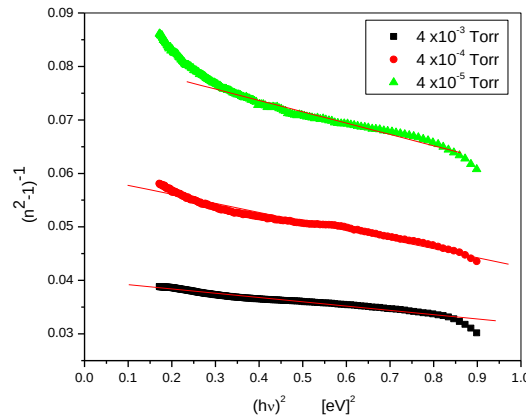


Fig.6. $(n^2-1)^{-1}$ Vs $(hv)^2$ of nano $Ge_{0.15}Te_{0.80}Cu_{0.05}$ thin films at different deposition pressures.

Additional details are available at [41,56]. ϵ_L and N/m^* could be calculated from the extrapolation at $\lambda^2 = 0$; ($\epsilon_L = n^2$) and the slope of the plot of n^2 Vs λ^2 as depicted in Fig.7 according to the following relation [41,56]:

$$n^2 = \epsilon_L - \left(\frac{e^2}{4\pi^2 \epsilon_0 c^2} \right) \left(\frac{N}{m^*} \right) \lambda^2 \quad (14)$$

e and ϵ_0 are the electronic charge and the permittivity of free space respectively. Fig. 7 shows that n has two types of dispersions; anomalous dispersion at $\lambda \leq 1280$ nm (absorption processes) where multi oscillator model can be used and the normal dispersion at $\lambda > 1280$ nm where single oscillator model can be used. The obtained values of ϵ_L and ϵ_∞ are summarized in Table I. their values are decreasing with LDP and had different values. The disagreement of their values may attributed to the contribution of free charge carriers to the polarization process[43,57].

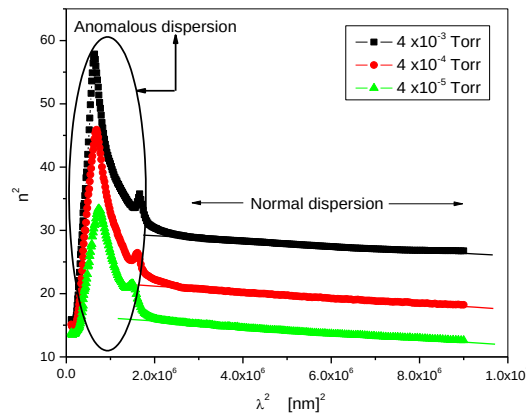


Fig.7. n^2 Vs λ^2 of nano $Ge_{0.15}Te_{0.80}Cu_{0.05}$ thin films at different deposition pressures.

It is known that the dielectric material will be more effective in case of the lower values of its dielectric loss, so the electrical applications of materials depend on its dielectric properties. Materials may be act as an insulator at the lower energy where the free carriers are limited or zero. The two parts of dielectric constant, ϵ ; the real, ϵ_1 and imaginary parts, ϵ_2 can be derived from n and k according to[35-37]:

$$\epsilon = \epsilon_1 - i \epsilon_2 \quad (15)$$

$$\varepsilon_1 = n^2 - k^2 \quad (16)$$

$$\varepsilon_2 = 2nk \quad (17)$$

The ratio of $(\tan \delta = \varepsilon_2/\varepsilon_1)$ represent the loss tangent or loss factor[35,58-60].

The effect of deposition pressure on ε_1 , ε_2 and $\tan \delta$ is shown in Figs.8 and 9. One can observe that LDP decreases ε_1 , ε_2 and $\tan \delta$ so the material turns to have good dielectric properties.

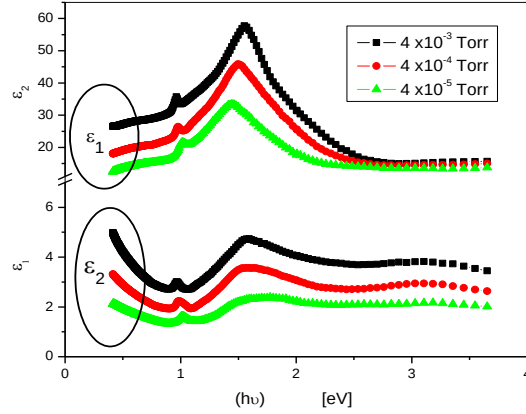


Fig.8. ε_1 and ε_2 Vs $(h\nu)$ of nano $Ge_{0.15}Te_{0.80}Cu_{0.05}$ thin films at different deposition pressures.

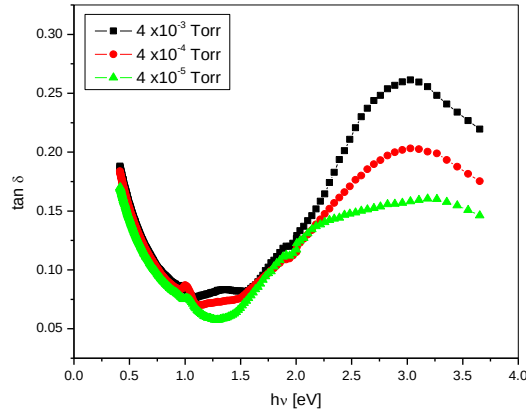


Fig.9. $\tan(\delta)$ Vs $(h\nu)$ of nano $Ge_{0.15}Te_{0.80}Cu_{0.05}$ thin films at different deposition pressures.

3.2.4. Optical conductivity

Electronic states (charge carriers) may be affected by deposition pressure and consequently the real, σ_1 and imaginary, σ_2 parts of optical conductivity, σ .

ε_1 and ε_2 are two major factors that may affect σ according to the following formula [35,55];

$$\left\{ \begin{array}{l} \sigma = \sigma_1 + i\sigma_2 \\ \sigma_1 = \omega\varepsilon_2\varepsilon_0 \\ \sigma_2 = \omega\varepsilon_1\varepsilon_0 \\ \varepsilon = \varepsilon_1 - i\varepsilon_2 \end{array} \right\} \quad (18)$$

ω in the mentioned equation identified as the angular frequency. Fig. 10 shows the dependence of σ_1 and σ_2 on deposition pressure. The higher deposition pressure and photon energy increase σ_1 and σ_2 . An absorption edge could be clearly seen near the optical gap value which can be attributed to electron excitations as a result of photon energy absorption [30,61].

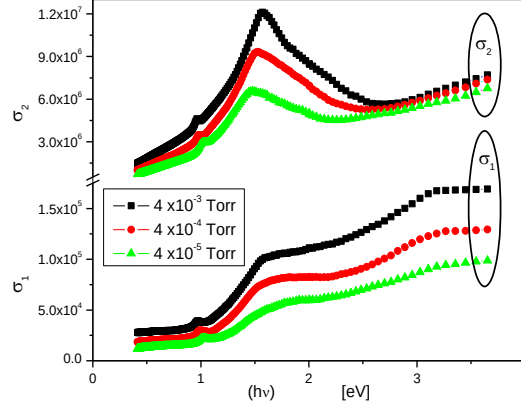


Fig.10. σ_1 and σ_2 Vs $(h\nu)$ of nano $Ge_{0.15}Te_{0.80}Cu_{0.05}$ thin films at different deposition pressures.

3.3.5. Energy loss function

Energy loss function, ELF depends on ϵ_1 and ϵ_2 . ELF has two parts; surface, SELF and volume energy loss functions, VELF, it can be calculated as [43,62]:

$$SELF = \text{Im}[-1/\epsilon(E) + 1] = \epsilon_2 / ((\epsilon_1^2 + 1) + \epsilon_2^2) \quad (19)$$

$$VELF = \text{Im}[-1/\epsilon(\omega)] = \epsilon_2 / (\epsilon_1^2 + \epsilon_2^2) \quad (20)$$

VELF/SELF ratio increases as a function of deposition pressure as depicted in Fig.11. Sharp edge could be seen near the value of the optical gap followed by steady state. Absorption processes and consequently the electron transitions are responsible for this behavior [55,62].

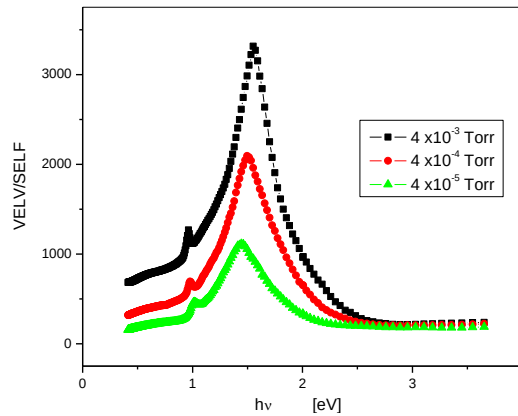


Fig.11. The ratio of (VELF/SELF) Vs $h\nu$ of nano $Ge_{0.15}Te_{0.80}Cu_{0.05}$ thin films at different deposition pressures.

4. Conclusions

$\text{Ge}_{0.15}\text{Te}_{0.80}\text{Cu}_{0.05}$ thin films were deposited by thermal evaporation technique on quartz substrates. XRD shows the amorphous state of the as prepared films of higher deposition pressure while the lower deposited one has poly crystalline nature with preferred orientations; [(222) and (411)] for GeTe_4 , [(101) and (003)] for Te and (311) for GeTe. The calculated crystallites sizes are 5, 10, 11, 10 and 9 nm which included in nano scale.

In addition, unidentified preferred orientation at 32.58° was observed. Direct, indirect optical gap and phonon energy were found and calculated. All are increasing with LDP because of ordering effect. The obtained data were discussed in terms of band-filling or B-M effect. The obtained data were confirmed by XRD, the values of constant A and carrier concentration, N in the ratio of carrier concentration to the effective mass, N/m^* . Other optical constants; n , ϵ_1 , ϵ_2 , $\tan(\delta)$, σ_1 , σ_2 , VELF/SELF ratio were also affected by LDP.

Finally, LDP has positive effects on the structural and optical properties of $\text{Ge}_{0.15}\text{Te}_{0.80}\text{Cu}_{0.05}$ and can be used to modify the structural and optical constants for optical data storage media and solar cell applications so, it is recommended to be done.

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