

## Study of Structure, Optical and Dielectric Properties of Nano Sb<sub>2</sub>Te<sub>3</sub> and Sb<sub>2</sub>Se<sub>3</sub> Thin Films as a New Optical Recording Material

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**Abstract:** Sb<sub>2</sub>Te<sub>3</sub>, Sb<sub>2</sub>Se<sub>3</sub> films of thicknesses 650 nm, 720 nm respectively were prepared by thermal evaporation. To estimate the sticheometry for the prepared powder for these compounds Energy Dispersive X-ray (EDX) was used. The optical parameters such as (refractive index, extension coefficient, absorption coefficient, optical energy gap) for these films were determined. The as-deposited Sb<sub>2</sub>Te<sub>3</sub>, Sb<sub>2</sub>Se<sub>3</sub> has an amorphous structure using X-ray diffraction.

**Key words:** Sb<sub>2</sub>Te<sub>3</sub> and Sb<sub>2</sub>Se<sub>3</sub> thin films, structure, optical properties and dielectric results

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### INTRODUCTION

Most of the technological electronic and optoelectronic applications utilize materials in thin film form. Recently, there has been growing interest in the dielectric-like behaviour in the amorphous state (Mitsuyu T *et al.*, 1981, Easwaran N, *et al.*, 1992). Binary compounds of the form A<sub>2</sub><sup>v</sup>B<sub>3</sub><sup>vi</sup> have been studied extensively in the past (Abd El Salam F *et al.*, 1993, Gilbert LR, *et al.*, 1974, Fadel M, *et al.*, 1999, Segura A, *et al.*, 1977, Sobolev V.V. *et al.*, 1968). Antimony telluride is a member of this group. It is a narrow band gap semiconductor, and belongs to the rhombohedral system. The structure comprises alternate layers of three-fold axis. This results in anisotropy in many properties. It has been observed that the conductivity of Sb<sub>2</sub>Te<sub>3</sub> is not susceptible to change in doping or to appreciable alterations in the proportion of antimony and tellurium (Damodara V *et al.*, 1989). These materials can be reversibly switched between the amorphous and crystalline state and find applications in rewritable optical recording (Gravesteyn D.J. *et al.*, 1989, Yamada N, 1996, Borg H.J. *et al.*, 1999) and in electrically programmable non-volatile memories (Ovshinsky S. 1998). Phase change optical recording has evolved to a mature technology that is applied in the re-writable versions of optical data storage systems of CD (compact disc) and DVD (digital versatile disc).

In phase-change optical recording, the recording of information is based on writing and erasing amorphous marks in a crystalline layer of a phase change material. Amorphous marks are written by a pulsed focused laser beam which makes the temperature of the phase-change layer locally higher than the melting point for a short period of time. After the pulse, the molten area is quenched to the amorphous state. Since the optical properties of the amorphous phase are different from those of the crystalline phase, the written mark can be read out as a contrast in the reflectance. The recorded information can be erased by heating the material with the same focused laser beam above the crystallization temperature but below the melting temperature. Due to the increased mobility of the atoms at increased temperature, the amorphous state rapidly returns to the thermodynamically more stable crystalline state. Besides sufficient optical contrast between the crystalline and amorphous state and a sufficiently low melting point attainable with moderate laser powers, the crystallization behavior at various temperatures is one of the most important issues in developing phase-change materials.

At elevated temperatures the crystallization rate should be large enough to enable a high data rate. That is, in phase-change recording the maximum rate at which previous data can be overwritten with new data is limited by the speed at which the amorphous marks of the previous data can be erased. On the other hand, at low temperature the crystallization rate has to be practically zero in order to ensure that recorded amorphous marks are stable against spontaneous re-crystallization for at least several years (archival life). These two conflicting requirements complicate the development of new phase-change materials with high crystallization rates for future products like for instance the Digital Video Recording system (DVR) (Dekker M.J., *et al.*, 2000, Tieke B. *et al.*, 2000). In addition, a recording material should also have the following characteristics: a sufficient contrast between crystalline and amorphous phase, i.e. difference in optical constants, leading to

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amplitude and/or phase modulation of the reflected laser beam during readout; an excellent over writability; a high crystallization temperature, preferably above 150 °C (thermally stable) and a sufficiently low melting point (see also [Borg H.J., Ullmann's Encyclopedia, Vol. A14, Information Storage Materials). Research for a rewritable optical recording disk involves mainly a search for recording materials. The conventional Phase-change optical disc is generally fabricated by the sputtering process, which has a drawback of requiring an initialization process to change the as-deposited recording layer in the disk from amorphous to crystalline phases. In order to minimize the cost, many researches have been carried out to skip this initialization process (Miao X.S *et al.*, 2000, Miao X.S. *et al.*, 2002, Voutsas G.P *et al.*, 1985). The  $Sb_2Te_3$  film could be used as an additional layer to enhance the crystallization of recording layer during low temperature sputtering process (Miao X.S *et al.*, 2000, Miao X. S. *et al.*, 2002), which is called 'Initialization-free' process. Sb layer could also enhance the crystallization of the recording material in the disk (Miao X.S. *et al.*, 2002). Although effect of enhanced crystallization with additional  $Sb_2Te_3$  layer was reported in the literature, the exact kinetic mechanism has not been explored satisfactorily. Furthermore, molecular modeling beside experimental work provides valuable information about structural properties of matter (Elhaes, H. *et al.* 2009; Ibrahim, M. and Abdel Aziz, A-A 2009).

In this recent paper we investigate the structure of  $Sb_2Te_3$  and  $Sb_2Se_3$  materials, the optical parameters for them were calculated and finally the dielectric properties were studied.

## **2. Experimental Work:**

$Sb_2Te_3$  and  $Sb_2Se_3$  powder were prepared by addition Sb element with Te element with chemical ratio, into very clean silica tubes, these tubes were closed under vacuum  $1.5 \cdot 10^{-3}$  torr. The first silica tube which contain Sb, Te and the second silica tube which contain Sb and Se was heated up to 1100 °C and 1200 °C respectively for 28 hours with checking to be sure that there is complete mixing and homogeneity for each compound. The  $Sb_2Te_3$  and  $Sb_2Se_3$  films of thicknesses 650 nm, 720 nm respectively were prepared using thermal evaporation under vacuum  $2 \cdot 10^{-5}$  Torr using coating unit of type Edwards E 306 onto well-cleaned glass substrates at 27 °C. The thickness of the films and rate of evaporation was controlled by a quartz crystal thickness monitor (Edwards FTM5) attached to the evaporation system, the film thickness and deposition rate were kept constant at (film thickness  $\pm$  20 nm) and 1 nm/sec consequently.

$Sb_2Te_3$  and  $Sb_2Se_3$  thin films formed by a thermal evaporation under a vacuum of  $\sim 1.3 \cdot 10^{-5}$  Torr. The stichometry of these two compounds were measured using EDX of type (Oxford ISI3 attached to Scanning Electron microscope SEM model Joel -5400). The structure of these powders and thin films were carried out using X-Ray Diffraction (XRD) of type Shimadzu XRD-6000, Japan, with wavelength 0.15425 nm of  $CuK\alpha_1$ . The XRD measurements were in the angle range from  $2\theta=4^\circ$  to  $2\theta=90^\circ$ . The optical transmittance and reflectance for these films were measured using spectrophotometer Jasco (V-570) in the range of wave length (190 – 2500 nm).

## **Results:**

### **a. Structure:**

The EDX measurements for the powder and thin films for the prepared samples shows that for the powder of  $Sb_2Te_3$  material have a nearly stochometry ratio because the ratio of elements of  $Sb_2Te_3$  powder was 41% : 49% respectively, secondly for  $Sb_2Se_3$  powder the elements ratio was 38% : 52% respectively.

Figure 1.a. shows the X-ray Diffraction patterns (XRDP) of the  $Sb_2Te_3$  and  $Sb_2Se_3$  respectively, from this fig it is clear that powder diffraction for these two materials have polycrystalline structure as matched with (JCSd using POWD +12) (Voutsas G.P. *et al.*, 1985), While Figure 1. b shows the x-ray diffraction pattern for as deposited  $Sb_2Te_3$  and  $Sb_2Se_3$  films, it is clear that from this fig that both of two samples have amorphous structure which agree with (Wei Hsiang Wang *et al.*, 2004).

### **Optical Results:**

Figures 2.a,b show the transmission and reflection spectra for as deposited  $Sb_2Te_3$  and  $Sb_2Se_3$  films which measured at room temperature, for as deposited  $Sb_2Se_3$  films exhibit a good extreme for reflection and a considerable extreme for reflection comparing with as deposited  $Sb_2Te_3$  films, this can be attributed to the density of  $Sb_2Te_3$  greater than the density of  $Sb_2Se_3$  which effected on the photon to penetrate through the film.

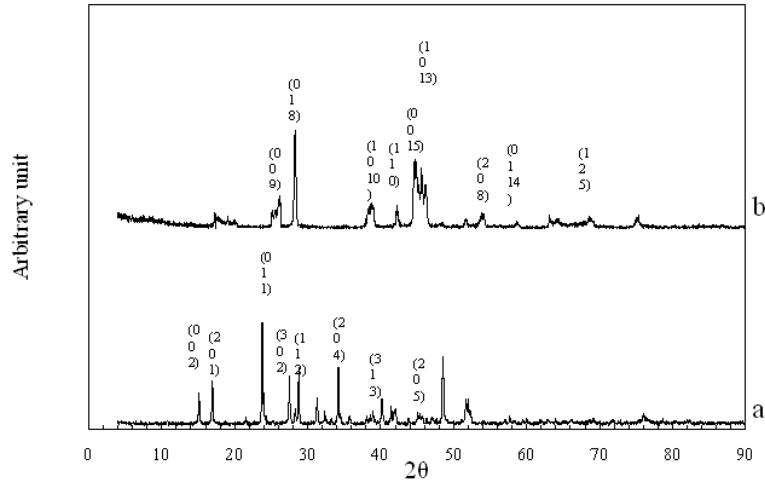
### **Absorption Coefficient, Energy Gap and Refractive Index:**

The study of the optical absorption is a useful method for investigating optically the induced transition and the interaction of dipoles with the incident light. Therefore, some optical parameters have been investigated

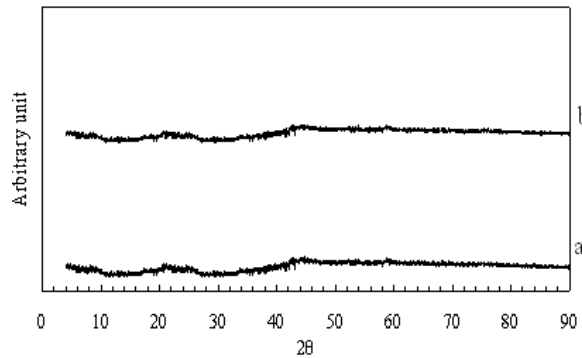
to explore the optical behaviour of solid materials. According to the band theory of solids, when a material is exposed to light, the absorption coefficient ( $\alpha$ ) can be calculated from the Urbach rule (Urbach F., 1953):

$$I = I_0 e^{-\alpha x} \tag{1}$$

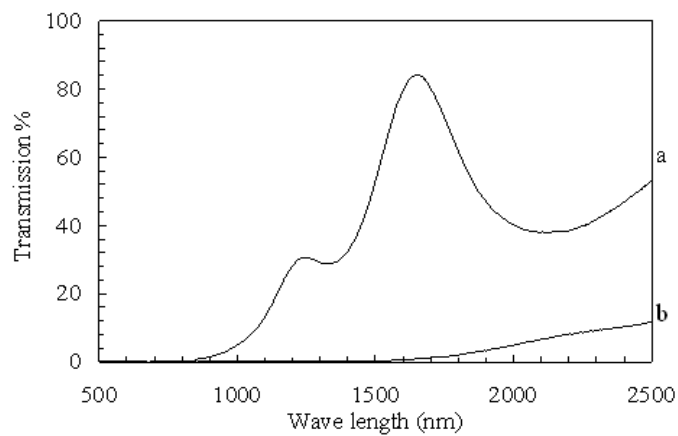
where  $I$  and  $I_0$  are the intensities of the incident and transmitted light, respectively, and  $x$  is the material thickness.



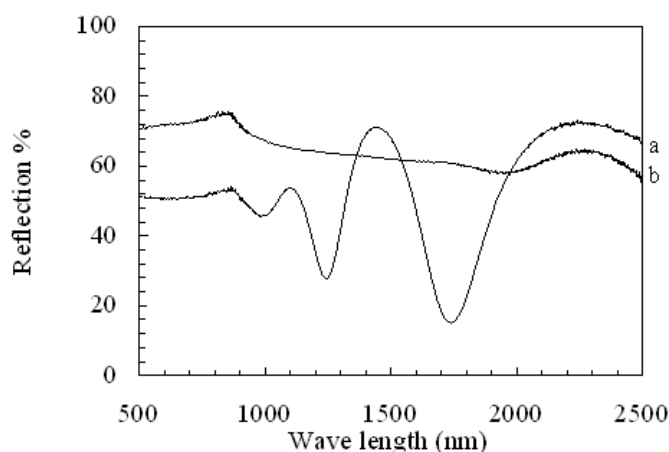
**Fig. 1.a:** X-ray diagram (a)  $Sb_2Se_3$  powder, (b)  $Sb_2Te_3$  powder



**Fig. 1.b:** X-ray diagram (a)  $Sb_2Se_3$  thin film, (b)  $Sb_2Te_3$  thin film

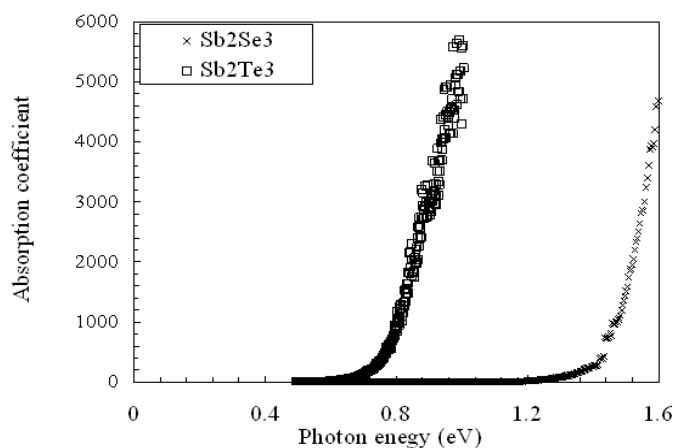


**Fig. 2.a:** Relation between transmission and wavelength for (a)  $Sb_2Se_3$  thin film, (b)  $Sb_2Te_3$  thin film



**Fig. 2.b:** Relation between reflection and wavelength for (a)  $Sb_2Se_3$  thin film, (b)  $Sb_2Te_3$  thin film

As shown in Figure 3 that the absorption coefficient behavior for  $Sb_2Te_3$  and  $Sb_2Se_3$  films, it seen that the absorption edge value  $Sb_2Se_3$  films is much smaller than that he absorption edge of  $Sb_2Te_3$  films it could be attributed to the energy gap values. The optical energy gap represents the most common optical parameter explains the optical transitions. It could be defined as the difference between the bottom of the conduction band and the top of the valence band. The general equation for the energy gap can be written as (Aranda J., *et al.*, 1984)



**Fig. 3:** Absorption coefficient dependance photon energy and wave length for  $Sb_2Se_3$  and  $Sb_2Te_3$  thin film

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

where, 'A' is a constant, 'E<sub>g</sub>' is the energy band gap, 'ν' is the frequency of the incident radiation and 'h' is Planck's constant. The magnitude of the exponent P is characteristic of the type of transition, and takes the value 1/2, 3/2, 2 and 3 for direct, allowed forbidden, indirect allowed and indirect forbidden transitions respectively. These as deposited films had indirect energy gap which change by annealing into direct energy gap, in order to determine both direct and indirect energy gap, for direct band gap must got the relation between  $(\alpha h\nu)^2$  and  $(h\nu)$  where  $h\nu$  is the energy of the incident light, on the other hand to get indirect band gap the relation between  $(\alpha h\nu)^{1/2}$  and  $(h\nu)$  is studied.

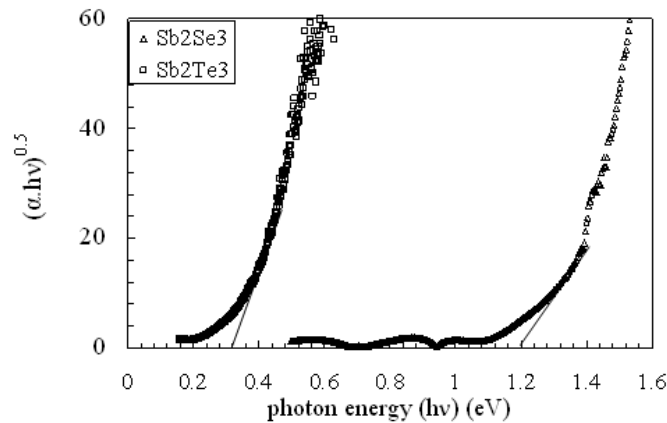
Figure 4. shows the relation between  $(\alpha \cdot h\nu)^{0.5}$ , photon energy and relation between in order to determine the indirect optical energy gap the tangent of the curve must be drawn, from this fig it was found that the energy gap for  $Sb_2Te_3$  and  $Sb_2Se_3$  films were 0.32 and 1.2 eV which agree with (Sehr R., *et al.*, 1962).

The Urbach tail for these samples were calculated using the following Equation [Urbach F., 1953]:

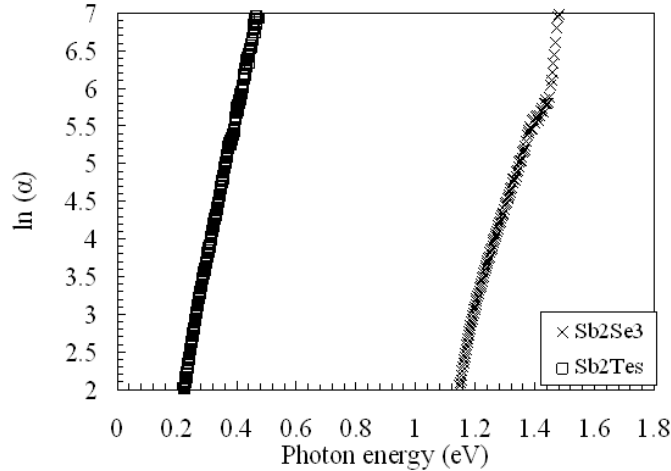
$$\alpha = \alpha_0 e^{h\nu/E_0} \tag{3}$$

Where  $\alpha_0$  is the material constant,  $E_0$  is the Urbach constant (tail). In order to determine the Urbach tail.

The relation between  $\ln(\alpha)$  and photon energy is shown in Figure 5. for both two used materials, the Urbach constant for  $Sb_2Te_3$  and  $Sb_2Se_3$  films were calculated from this Figure 5. using equation (1), and was found that the Urbach constant for  $Sb_2Te_3$  and  $Sb_2Se_3$  film were 0.13 eV and 0.55 eV respectively.



**Fig. 4:** Relation between  $(\alpha, hv)^{0.5}$  and photon energy for  $Sb_2Se_3$  and  $Sb_2Te_3$  thin film



**Fig. 5:** Relation between  $\ln$  absorption coefficient and photon energy for  $Sb_2Se_3$  and  $Sb_2Te_3$  thin film

Refractive index for these films were calculated using the following equation (Nayak A., *et al.*, 1991)

$$n = \left[ N + (N^2 - n_s^2)^{1/2} \right]^{1/2} \tag{4}$$

where

$$N = \left[ \frac{(1 + n_s^2)}{2} \right] + 2n_s \left[ \frac{(T^+ - T^-)}{(T^+ \cdot T^-)} \right] \tag{5}$$

$n_s$  is the refractive index of the used substrate.

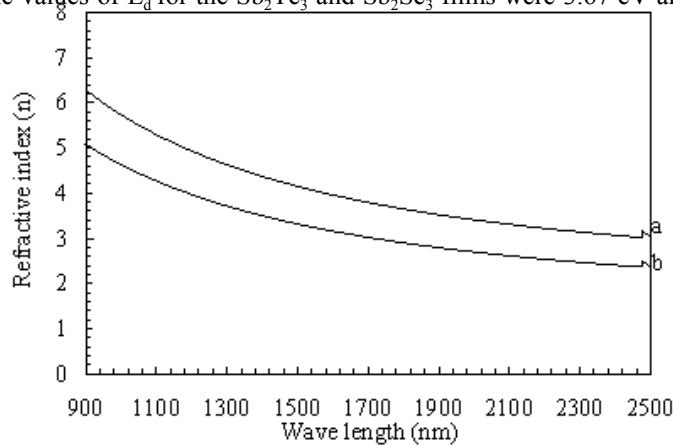
Figure 6. shows the refractive index dependence on wave length for both  $Sb_2Te_3$  and  $Sb_2Se_3$  films, the behaviors of these curves for both  $Sb_2Te_3$  and  $Sb_2Se_3$  films is in agree with (Richter W. *et al.*, 1982). From

this curve it is clear that the refractive index value of  $Sb_2Te_3$ , strongly increases with wave length in wave length range (500-800 nm) and rapid decrease in wave length range (850-1000 nm), this means that it is possible to control the values of  $n$  depending on the propose in which for use this material.

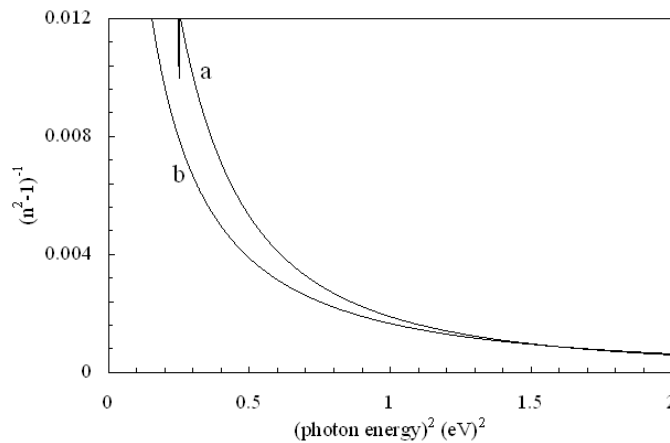
The dispersion of refractive index in  $Sb_2Te_3$  and  $Sb_2Se_3$  films was analyzed using the concept of the single oscillator and can be expressed by the Wemple–DiDomenico relationship (Torris J *et al.*, 1996) as

$$n^2(E) - 1 = \frac{E_o - E_d}{E_o^2 - E^2} \tag{6}$$

where  $E$  is the photon energy,  $E_o$  is the oscillator energy and  $E_d$  is the dispersion energy. The parameter  $E$ , which is a measure of the intensity of the inter-band optical transition, does not depend significantly on the band gap. The values of  $E_o$  and  $E_d$  are obtained from the intercept and the slope resulting from the extrapolation of the curve of Figure 7. for the values of  $E_o$  for  $Sb_2Te_3$  and  $Sb_2Se_3$  films were as 0.13 and 0.7 eV respectively, but for the values of  $E_d$  for the  $Sb_2Te_3$  and  $Sb_2Se_3$  films were 3.67 eV and 4.8 eV respectively.



**Fig. 6:** Relation between refractive index and wave length for (a)  $Sb_2Se_3$  thin film, (b)  $Sb_2Te_3$  thin film



**Fig. 7:** Relation between  $(n^2-1)^{-1}$  and  $(\text{photon energy})^2$  for (a)  $Sb_2Se_3$  thin film, (b)  $Sb_2Te_3$  thin film

**Dielectric results:**

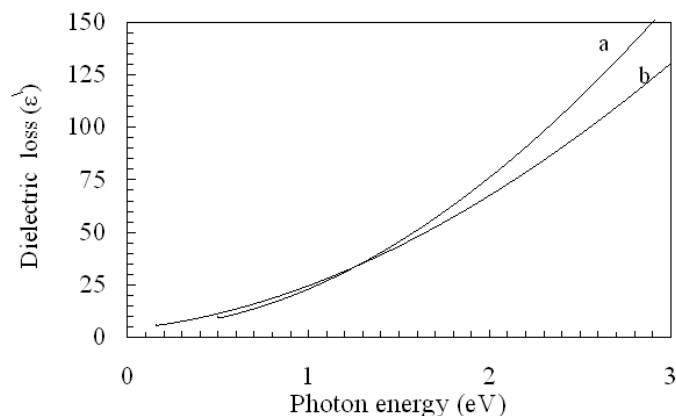
The dielectric constant ( $\epsilon'$ ), the dielectric loss ( $\epsilon''$ ) for this studied materials were calculated using the following equations (Aleksandra B. *et al.*, 1989):-

$$\epsilon' = (n^2 + k^2) \tag{7}$$

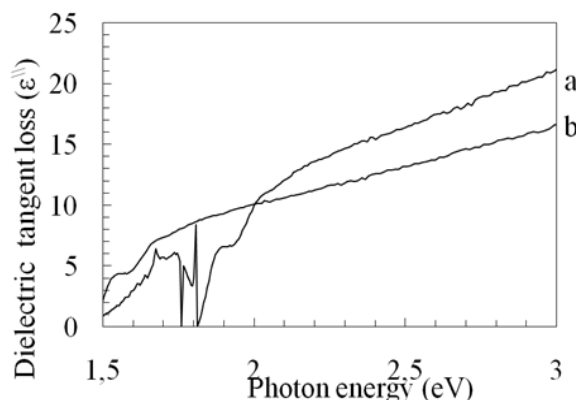
$$\epsilon'' = [(n^2 + k^2)^2 - (n^2 - k^2)^{0.5}]$$

Figures 8.a,b. show the dependence of The dielectric constant ( $\epsilon'$ ), the dielectric loss ( $\epsilon''$ ) on the photon energy for both  $Sb_2Te_3$  and  $Sb_2Se_3$  films.

From Figure 8.a it is clear that the behavior of the two curves is similar and the dielectric loss behavior has an exponential trend and the dielectric loss decrease with photon energy for both  $Sb_2Te_3$  and  $Sb_2Se_3$  films.



**Fig. 8a:** Relation between dielectric loss and photon energy for (a)  $Sb_2Se_3$  thin film, (b)  $Sb_2Te_3$  thin film



**Fig. 8b:** Relation between dielectric tangent loss and photon energy for (a)  $Sb_2Se_3$  thin film,

**Conclusion:**

$Sb_2Te_3$  and  $Sb_2Se_3$  materials were used widely as new optical recording materials, the optical and dielectric properties for these films were studied. The as deposited films of  $Sb_2Te_3$  and  $Sb_2Se_3$  were amorphous. The energy gap were calculated, which is indirect energy gap and was (0.3, 1.2 eV for  $Sb_2Te_3$  and  $Sb_2Se_3$  films), Urbach constant for these films were calculated (0.13, and 0.55 eV for  $Sb_2Te_3$  and  $Sb_2Se_3$  respectively). The oscillator energy and dispersion energy were calculated optically for these materials. The dielectric loss and tangent loss of these films were calculated optically the behavior of dielectric loss for  $Sb_2Se_3$  is nearly constant with wave length, while for  $Sb_2Te_3$ , the dielectric loss increase with wave length.

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