

## Synthesis and Effect of Thickness on the Structure and Optical Properties of ZnO Thin Films Prepared By Sol-Gel Spin Coating Method

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**Abstract:** In this work , (ZnO) films are prepared by (sol – gel) method . Zinc Oxide (ZnO) thin films was growth on to hot glass substrate at 60 oC temperature by spin coating. Then samples were annealed at temperature (450) °C with three thicknesses (58 ,69 and 77)nm for 1 hr. The experimental diffraction angle  $\Theta$  and d-spacing values are in agreement with the standard ASTM data for all thin film thicknesses .The structure parameters like lattice parameter (a); grain size (D) ; dislocation density ( $\sigma$ ) ; micro strain ( $\epsilon$ ) are calculated . The absorbance (A) , transmittance (T) and reflectance (R) are recorded in the range (300 – 1000) nm , and used to calculate the refractive index (n) , extinction coefficient (k) , band gap (Eg) , optical conductivity ( $\sigma_{opt}$ ) , complex dielectric constant ( $\epsilon_1, \epsilon_2, \epsilon_\infty$ ), relaxation time ( $\tau$ ) , average interband oscillator wave length ( $\lambda_0$ ) , average oscillator strength (So) , (N/m<sup>3</sup>), dissipation factor (tan $\delta$ ) and the optical dispersion parameters (E<sub>0</sub> , E<sub>d</sub>) were determined.

**Keywords:** Zinc Oxide ; Sol-Gel ; thin films ; X-ray diffraction , structure parameters ,Optical properties.

### 1-Interoduction

Zinc Oxide (ZnO) belongs to the family of II-VI compound wide – gap semiconductor with a room temperature direct band gap of (3.37) eV and a large exciton binding energy of about (60) mV, which makes it a very attractive material for the application to the advanced optoelectronic devices [1], and good efficiency of exciton recombination that with result in a high gain at room temperature. Also, it displays good piezoelectric, good catalysis and novel optical proportions, such as UV-light –emitting or lasing proportion. Specially, ZnO has a high break down voltage that is almost four times that of GaAs [2-4]. This is the reason that ZnO has recently received more and more attention from many researchers.

The structure of ZnO is a mixture of cubic and hexagonal structure depending on the manufacturing conditions. The electronic transport mechanism in polycrystalline thin films strongly depends on their structure (i.e . grain size , grain boundaries and structure defects). The X-ray diffraction technique was used to determine the crystalline structure and grain size Of ZnO thin films [5].Most prominent crystalline structure of ZnO is wurtzite type, although it also exists in the cubic Zinc blende and rock salt structure. In wurtzite type each Zn ion is surrounded by a tetragonal coordination. This give rise to polar symmetry along the hexagonal axes which is

responsible for a number of properties of ZnO [6]. Zinc Oxide is one of transparent conducting oxide (TCO) materials whose thin films attract much interest. (TCO's thin film is a wide direct band gap compound semiconductor such as In<sub>2</sub>O<sub>3</sub> , SnO<sub>2</sub> , ZnO , CdO , Cd<sub>2</sub>SnO<sub>4</sub> ,.....)[7].

It is important material due to its typical proportion such as high chemical and mechanical stability and high optical transparency in visible and near – infrared region (more than 80% depends on the deposition technique and thickness), It can be used as antireflection coating layer solar cells [8] and gas sensor [9] . Moreover, ZnO is promising material for short wave optoelectronic devices, especially for UV light – emitting diode and laser diode, due to its large exciton binding energy is much larger than the room temperature thermal energy [10].

Many techniques have been employed to prepare ZnO films such as pulsed laser deposition (PLD), magnetron sputtering (MOCVD), spray pyrolysis [11-14]. Thermal Vacuum Evaporation (TVE) and molecular beam epitaxy [15]. Sol-gel technique is widely adopted due to its comparatively simple procedure as there is no need of costly vacuum system and it has a wide-range advantage of large area deposition and uniformity of the films thickness. The sol-gel process also offers other advantages for thin film deposition including outstanding control of the stoichiometry and easy doping in film composition. The structure and physical properties of ZnO thin films prepared by sol –gel technique using various inorganic and organic precursors at different deposition conditions have been reported in literature [16, 17].

F.Leandro et al [18] found that the film thickness has a strong influence on the optical absorption of nanostructured  $\alpha$ - Fe<sub>2</sub>O<sub>3</sub> nano thin film. They report that, stress induced due to increased thickness, generator detects in the crystal lattice of the hermatite film, which intern increase the electron-hole recombination process [6, 19]. In the present work , thin films of ZnO have been fabrication by spin coating on to glass substrats .optical constant and oscillating parameter have been determined as an include the absorbance , transmittance , reflectance spectra , refractive index , extinction coefficient , optical conductivity , complex dielectric constant and thickness.

## Experimental procedure

ZnO thin films were deposited on glass substrates by using the sol-gel method according to reference [20]. Zinc acetate ( $Zn(C_2H_3O_2)_2$ , Aldrich-99.99%) was added in a mixture of isopropanol ( $(CH_3)_2CHOH$ ), and ethanolamine ( $H_2NCH_2CH_2OH$ , Aldrich-99%, MEA). The molar ratio of MEA to zinc-acetate was maintained at 1, where the amount of zinc acetate was adjusted to 1.0 mole. The mixture was stirred at 60 Co for 1 hr. and then aged at room temperature for 24 hrs. To yield a homogeneous solution. ZnO thin films were prepared by spin coating, the stored solution on a glass substrate at rotation speeds (1000, 2000, and 3000) rpm for thicknesses ZnO thin films (77, 69, and 58) nm respectively for 60 sec. The ZnO thin films were then dried on a hotplate at 60 oC for 30 min. The sample was heated in a furnace at a temperature of (450) oC for 1 hr. at a heating rate of 50 oC / min [20]. The transmittance (T) and absorbance (A) were measured by using a (6800 UV/VIS) Jenway Double Beam Spectrophotometer - England, in the wavelength range of 300 to 1000nm. The effect of thin film thickness on the structure and optical properties has also been studied. X-ray diffraction (XRD) spectra were recorded with an Philip Xpert PRO diffractometer using  $CuK\alpha$  radiation ( $\lambda=1.5406$  oA, 40 KV and 20 mA) for  $2\theta$  values over (20 - 80o)

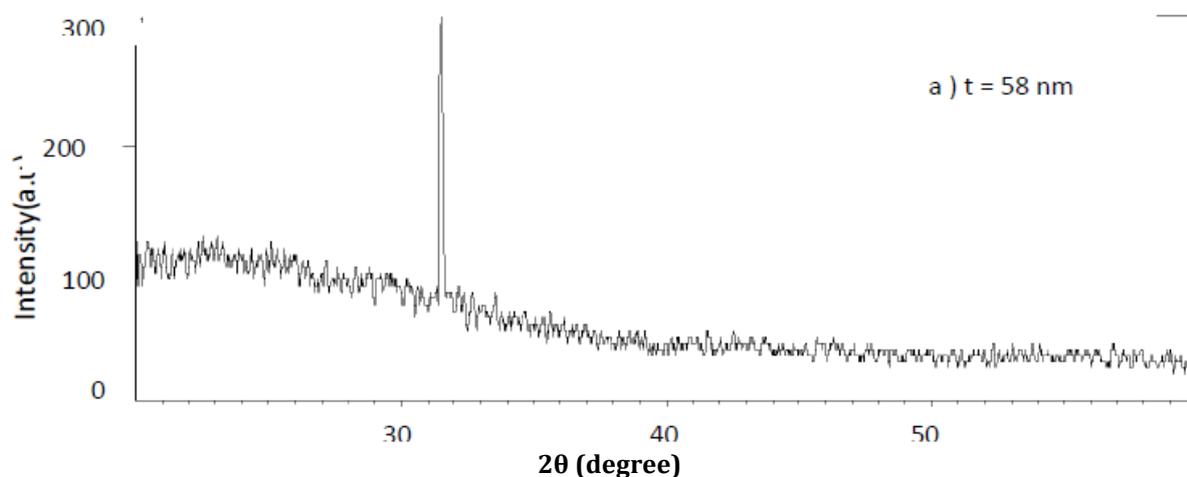
## Results and Discussion

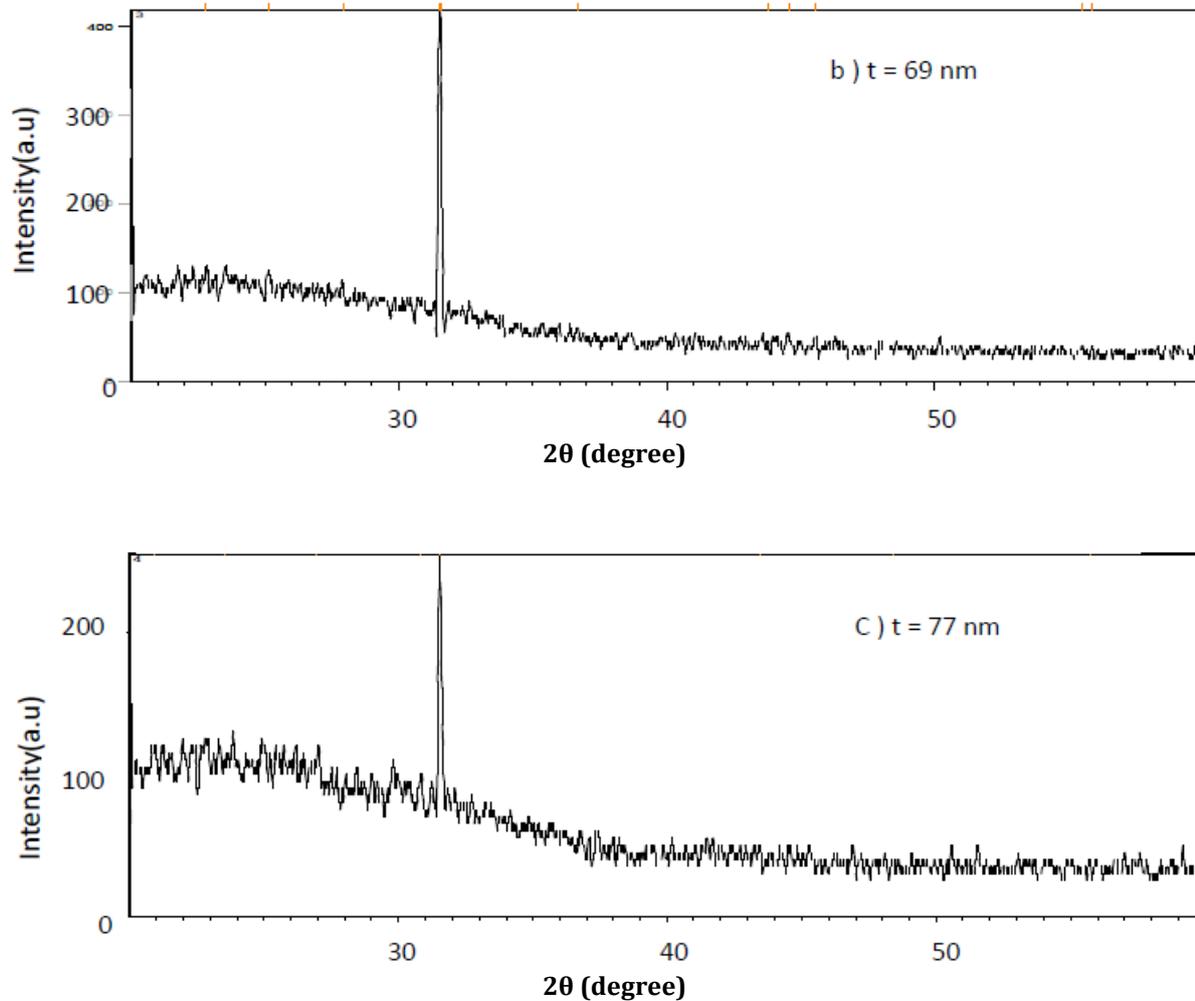
### Structural Analysis of ZnO Thin Films

Zinc Oxide (ZnO) films have been found to grow in cubic and hexagonal forms. The deposited (ZnO) thin films were analyzed by X-ray diffraction pattern for structural analysis by using Philips X-ray diffractometer (XRD), as shown in figures(1), which shows the X-ray for different thin films thicknesses (58, 69, 77) nm at annealing temperature (450) oC, which was obtained with  $2\theta$  (from 25o to 50o) glancing angle appear only one sharp peak.

The (XRD) pattern of the film shows that the films are polycrystalline, crystallized in the wurtzite phase and presents a preferential orientation along the c-axis. The results were in agreement with American Standard of Testing Materials (ASTM) as listed in table (1). The d-values were calculated by calculating ( $\Theta$ ) values from the peak of X-ray spectrum by using Bragg's relation ( $2d \sin\Theta = n\lambda$ ), ( $n=1$ ) in the present study and used K-Alpha wave length=1.5406 oA. These d-values were compared with standard (ASTM) data to confirm the Structure of ZnO [02]. The peak broadening at lower angle is more meaningful for the calculation of particle size; therefore size of crystals has been calculated using Deby-Scherre formula [21]. Deby-Scherre formula for grain size determination is given by [20] :-

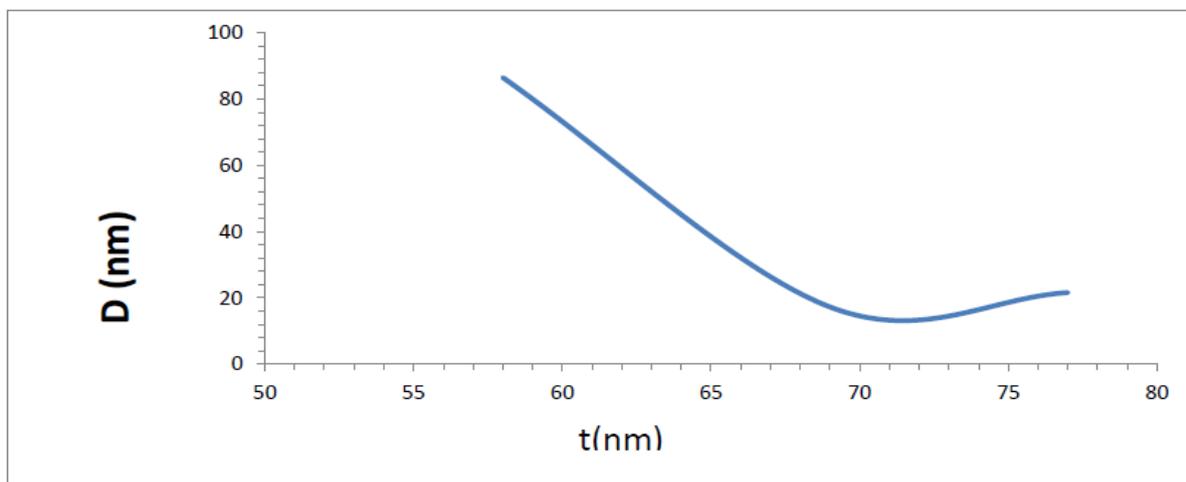
$$D = \frac{0.94\lambda}{\beta \cos\theta} \dots \dots \dots (1)$$





**Fig (1):** X-Ray diffraction pattern XRD of ZnO thin films annealed at (450 oC) for different thin film thicknesses [ a)  $t = 58$  nm ; b)  $t = 69$  nm ; c)  $t = 77$  nm ] between ( $2\theta$ ) and count/sec where  $D$ : is the grain size ;  $\lambda$  :is the wave length of X-ray ;  $\beta$ : is the Full Width at Half Maximum (FWHM) of the film diffraction peak at ( $2\theta$ ) ,where  $\Theta$ : is the Bragg diffraction angle . The mean crystallite size of the films

were about (86.46 , 17.28 , 21.6 )nm for samples deposited at room temperature and annealed at 450 oC with thickness (58 , 69 , 77) nm respectively. The value of grain size obtained from XRD for different film thickness are listed in table (1).It is clear that the grain size decreases when film thickness increase (as shown in figure (2)).



**Fig (2):** The grain size of ZnO thin film as function of thin film thickness.

The lattice constant "a" and "c" of the wurtzite structure of ZnO can be calculated using the relations (2) and (3) given below. [20]:

$$a = \sqrt{\frac{1}{3} \frac{\lambda}{\sin\theta}} \dots\dots\dots (2)$$

$$c = \frac{\lambda}{\sin\theta} \dots\dots\dots (3)$$

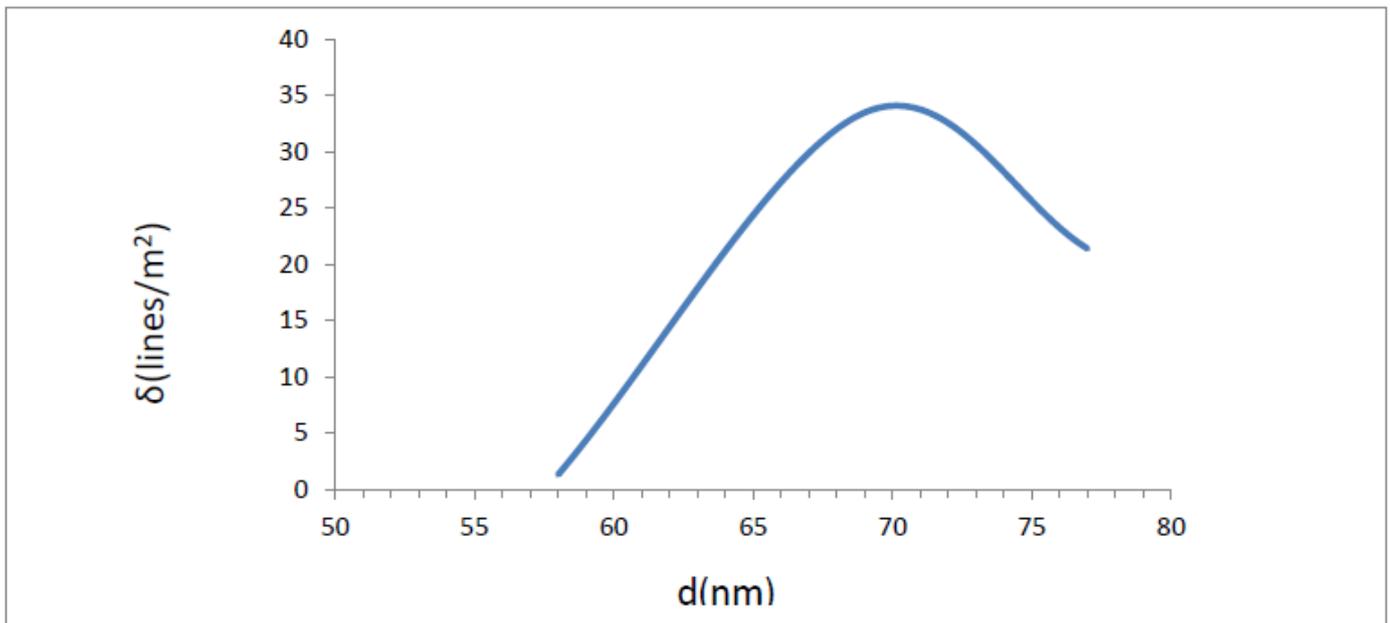
The calculated values of lattice parameter (a) and (c) are agreement with stander values [23], as shown from table (1).

The values of Observed and ASTM for displacing are showing in the table (1); which in agreement with each other.

A dislocation is a crystallographic defect, or irregularity, with a crystal structure. The presence of dislocation ( $\delta$ ) influences many of properties of materials. The dislocation density was also calculated from the relation [21].

$$\delta = \frac{1}{D^2} \dots\dots\dots (4)$$

The maximum value of dislocation density ( $\delta$ ) is for thin film of thickness (t=69 nm) as shown in figure (3)

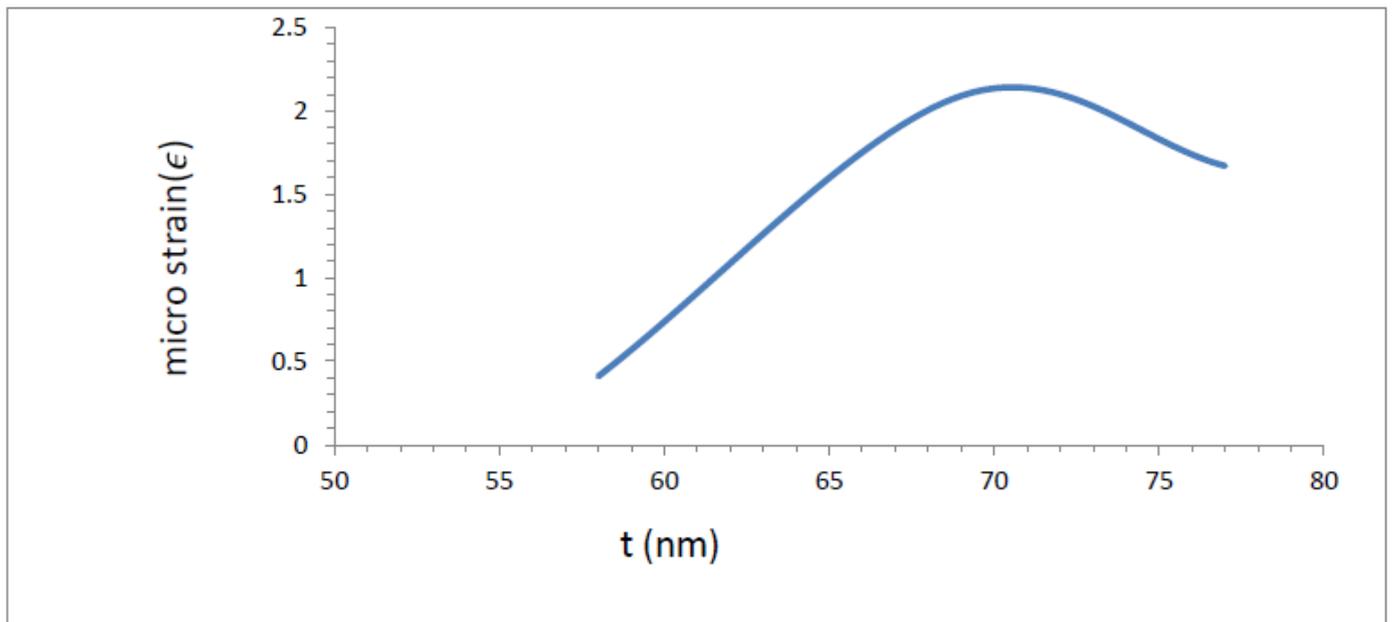


**Fig (3):** Dislocation density of ZnO thin films as a function of thin film thickness

The micro strain ( $\epsilon$ ) of ZnO thin films has estimated from the relation :[21]

$$\epsilon = \frac{\beta}{4} \cos\theta \dots\dots\dots (5)$$

Where ( $\beta$ ) is the Full Width at Half Maximum (FWHM) and ( $\theta$ ) is the observed 'Bragg' angle . The micro strain ( $\epsilon$ ) of ZnO thin films annealed at temperature (450 oC) is shown in figure (4) with maximum value of micro strain for (t= 69 nm) thin films was ( $\epsilon = 2.09$ )



**Fig (4):** Micro strain of ZnO thin films as a function of thin film thickness

The structure parameters of ZnO thin films are summarized in table (1). We can show from the table (1), that the best thin film thickness is (t=58 nm) because at this thin film thickness grain size is increase to (D=86.46

nm) and the dislocation density and micro strain are decreases to ( $\delta= 1.33 \times 10^{14}$  line /m<sup>2</sup>, =  $0.41 \times 10^{-3}$ ) respectively.

**Table (1):** The values of structure parameters for different thickness of ZnO thin films

Parameters	Thickness		
	d = 58nm	d = 69 nm	d = 77nm
a (A°)	3.2	3.2	3.2
a stander 3.2	3.15	3.15	3.15
c (A°)	5.6	5.6	5.6
c stander	5.29	5.29	5.29
D (nm)	86.46	17.28	21.6
ASTM (2θ) (degree)	31.514	31.508	31.50288
Observed (2θ) (degree)	31.54	31.498	31.5028
ASTM d-spacing(A°)	2.8342	2.83824	2.8382
Observed d-spacing(Ao)		2.8378	2.83757
Dislocation density $\delta(\text{line/m}^2) \times 10^{14}$	1.33	33.48	21.4
Micro strain ( $\epsilon \times 10^{-3}$ )	0.41	2.09	1.67

**Optical Analysis**

The optical properties of sol-gel deposited ZnO thin films on the glass substrates with different thickness(58 , 69 and 77) nm with annealing temperature (450 oC) were determined from the transmittance and absorbance measurements in the rang (300-1000) nm. The

thicknesses of thin films prepared were measured by weight method according to the following relation [24].

$$t = \frac{\Delta m}{S\rho} \dots \dots \dots (6)$$

Where, the mass difference after and before deposition; S: the area of the deposited film; the density of ZnO.

The Transmission, Absorption and Reflectance Spectra

Figure (5): Shows That the Optical Transmittance of ZnO Films For Different Thickness.

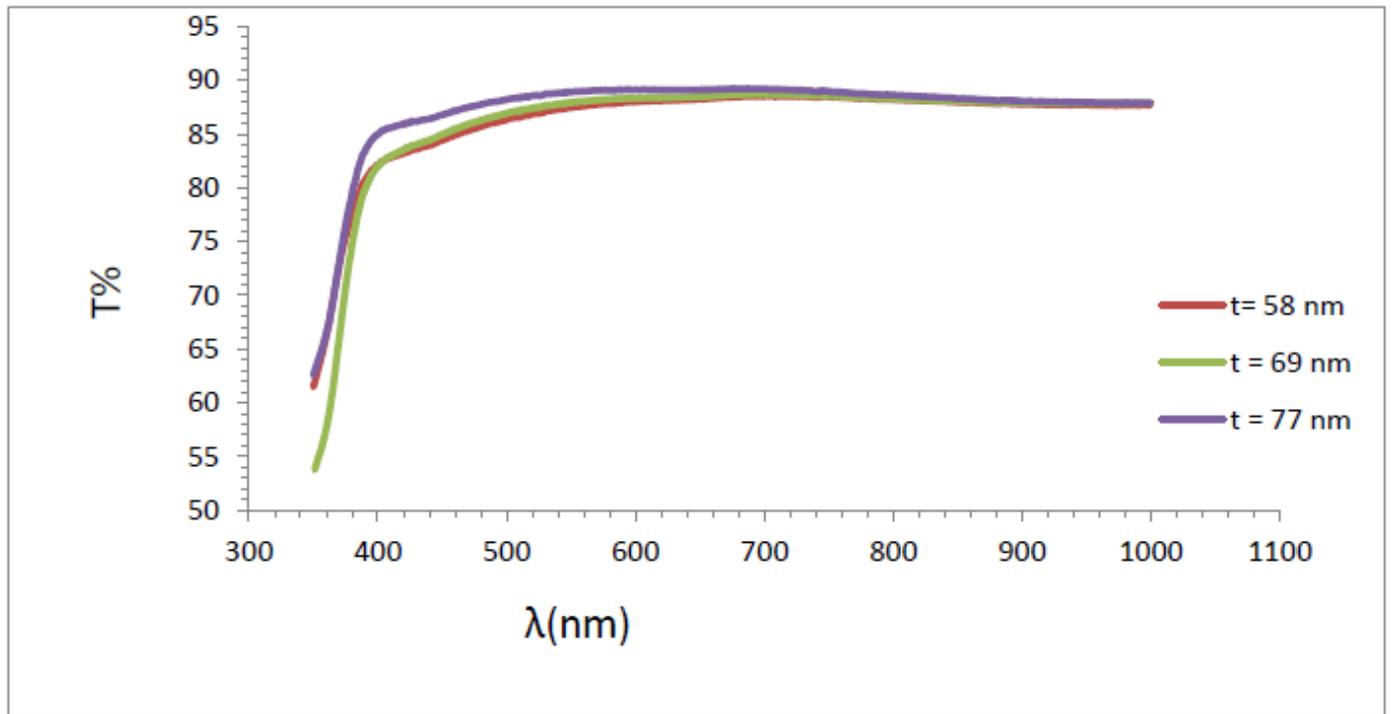


Fig (5): Transmission as function of wavelength for ZnO thin films with different thicknesses.

All films have high transmission (more than 85%) in the visible and UV region. Figure (6) shows that ZnO thin films have a good absorption at short wave length region for

different thicknesses and the absorption decrease with increasing wave length.

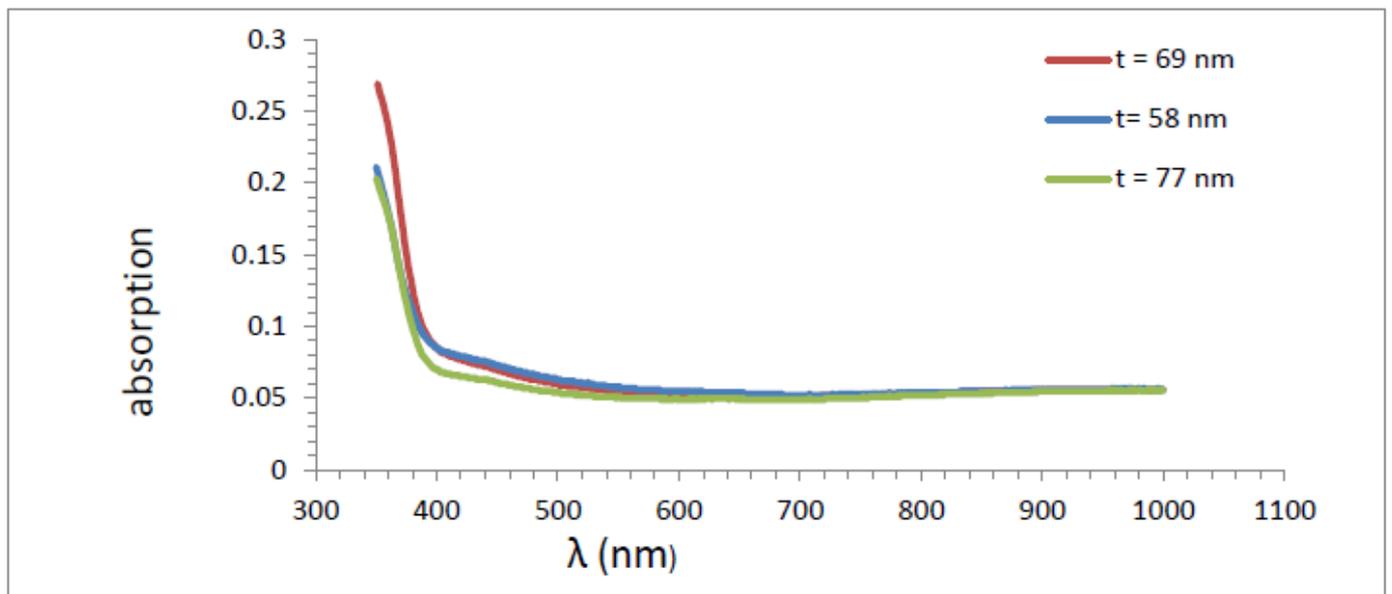


Fig (6): Absorption as Function of Wavelength for ZnO Thin Films with Different Thicknesses

The reflectance (R) of ZnO film can be calculated from the relation [26, 27].

$$R+T+A=1 \dots\dots\dots (7)$$

Figure (7) show the reflectance of ZnO thin films as a function of wave length for different film thickness at annealing temperature (450 °C).

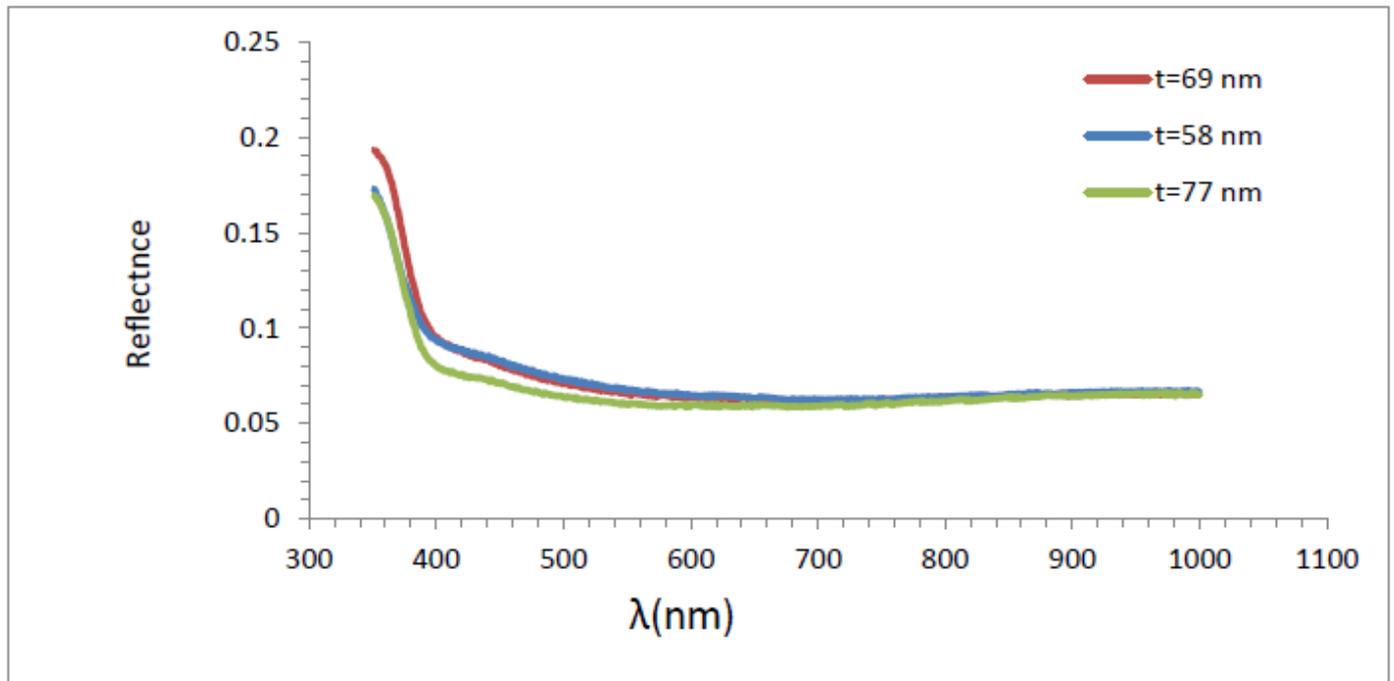


Fig (7): Reflection as Function of Wavelength for ZnO Thin Films with Different Thicknesses

**The Absorption Coefficient (α) and Extinction Coefficient (k)**

The absorption coefficient (α) of ZnO thin films was determined from the absorbance measurement by using the following equation [28].

$$\alpha = \frac{2.303}{t} \dots \dots \dots (8)$$

Where: A: is the absorbance and t: is the thickness of the film. Figure (8) shows the relation between the absorption coefficient (α) and photon energy (hv) for ZnO thin films, as shown in this figure, (α) is very small at low photon energy (hv < 2.5 eV) because the probability of the electron transfer between valance band and conduction band is very rare but it will be increase in the edge of the absorbance toward the high energy (hv > 2.5 eV).

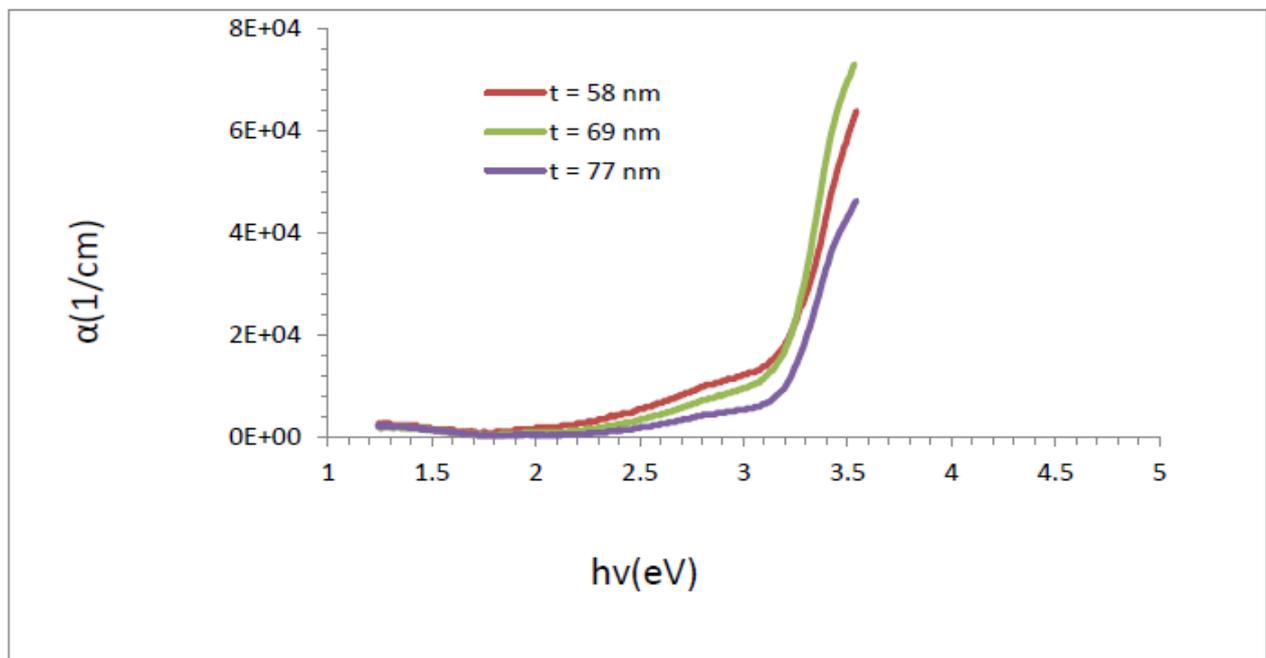
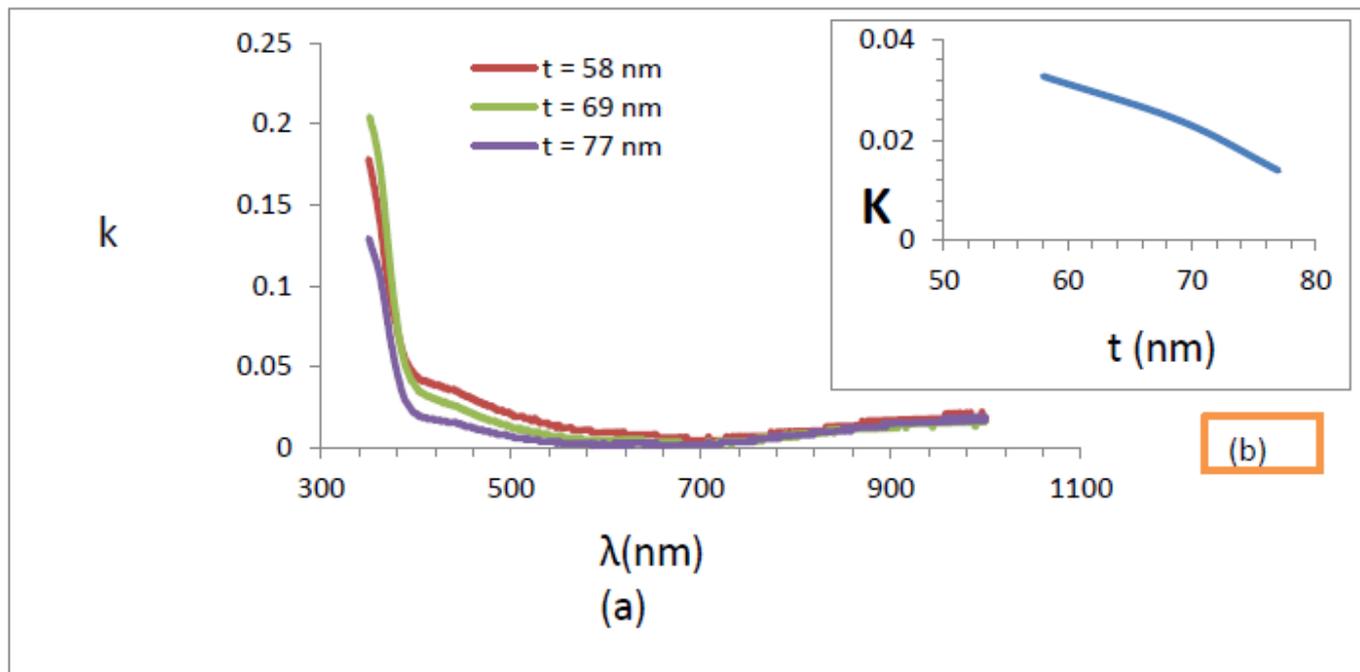


Fig (8): The Absorption Coefficient Vs. Photon Energy for ZnO Thin Films with Different Thicknesses

The extinction coefficient (k) determined from absorbance spectrum as function of photon energy at wave length within the range (300 – 1000) nm, it can be determined from the relation.

$$k = \frac{\alpha\lambda}{4\pi} \dots\dots\dots (8)$$

The relation between (k) and ( $\lambda$ ) spectra is shown in figure (9) for different ZnO thin film thicknesses. The extinction coefficient (k) decrease with increasing thin film thickness (t) in visible region due to reduce in absorption coefficient ( $\alpha$ ) with thickness (t). This result is agreement with equation (8).



**Fig (9): a)** Dependence of the extinction coefficient (k) on the wavelength for ZnO thin films with different thicknesses. **b)** Dependence of extinction coefficient (k) in thin film thickness (t) at visible region

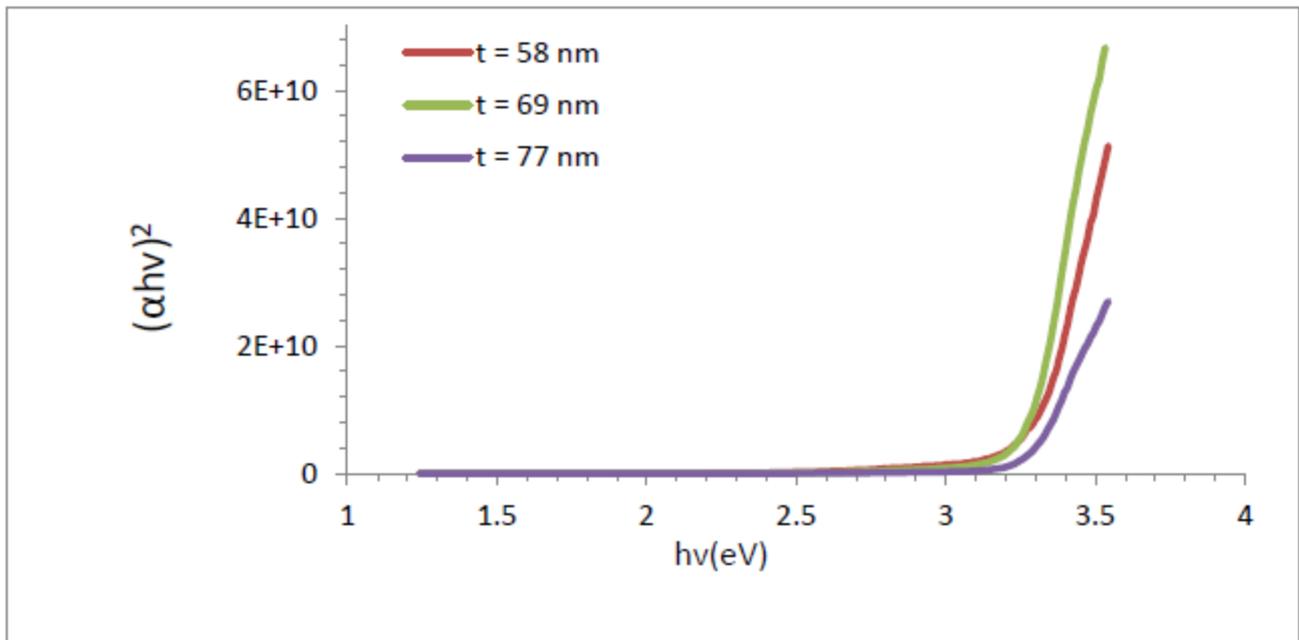
**The optical band gap ( $E_g^{opt}$ )**

The band gap  $E_g$  depends on the type of transition. The values of absorption coefficient ( $\alpha$ ) plays an important role to limitation the type of transition [29].

Here transitions are direct because ( $\alpha$ ) is more than (104 cm<sup>-1</sup>), (when  $\alpha < 104$  cm<sup>-1</sup> the transition is indirect transition) [29]. These transitions are described by the relation [30].

$$(\alpha hv) = B(hv - E_g^{opt})^r \dots\dots\dots (9)$$

Where  $B$  : is constant which is proportion inversely with amorphosity,  $\alpha$ : is the absorption coefficient,  $hv$ : is the incident photon energy, and (r) takes the values ( $\frac{1}{2}, \frac{3}{2}, 2$ ) depending on the type of transition (r = 1/2 for direct allowed transition, r = 3/2 for direct forbidden transition and r = 2 for an indirect allowed transition) [25]. figure (10) shows the relation between  $(\alpha hv)^2$  vs ( $hv$ ).



**Fig (10):** Plot of  $(\alpha hv)^2$  vs. Photon Energy ( $h\nu$ ) Near the Absorption Edge of ZnO Thin Film with Different Thicknesses

The values of optical band gap ( $E_g^{opt}$ ) are calculated from the intercept of the extrapolation the straight line portion of the curves. Accordingly, values of band gaps are ( $E_g = 3.225, 3.222, 3.223$ ) eV for ( $t = 58, 69, 77$ ) nm respectively (see table (3)). The average value of energy gap ( $E_g = 3.22$  eV) is agreement with values obtained before as shown in table (2).

**Refractive index (n):**

The refractive index ( $n$ ) of thin films was determined from a transmittance spectrum (or reflectance spectrum) as a function of the photon energy within the wave length in the range (300-1000) nm for different ZnO thin films thicknesses (58 , 69 ,and 77) nm by using Swanepoel's method [31].

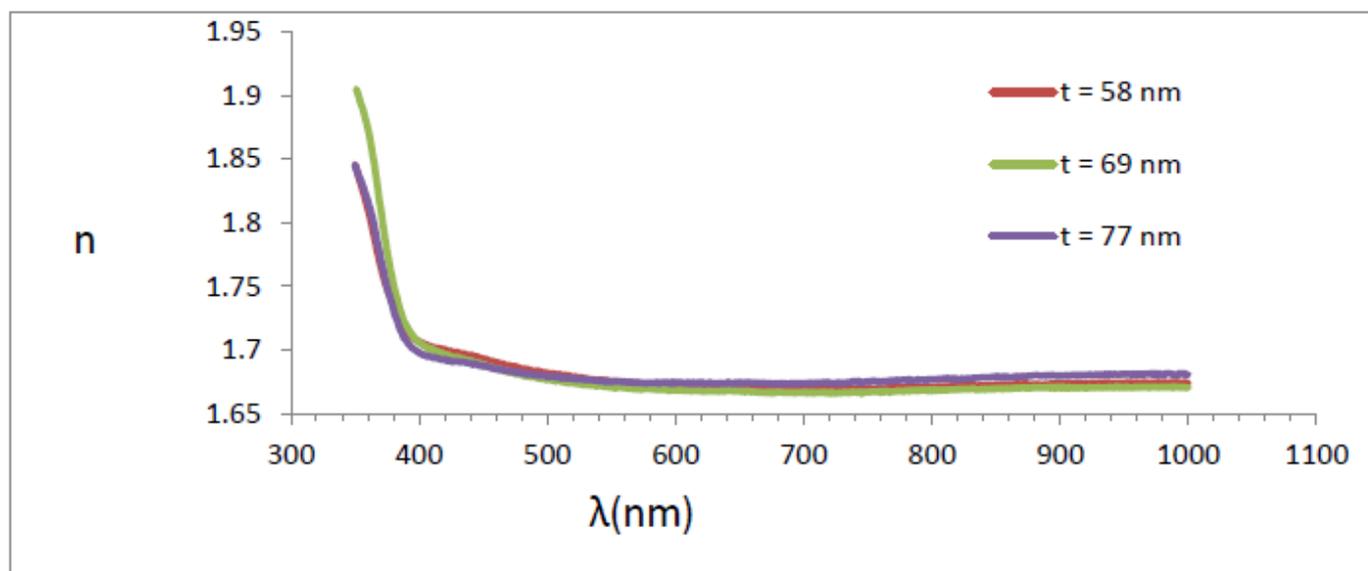
$$n = \frac{1 + \sqrt{R}}{1 - \sqrt{R}} \dots \dots \dots (10)$$

Where R: is the reflectance of ZnO thin film.

**Table (2):** comparison of optical band gap energy of ZnO thin film

Reference	Energy gap ( $E_g$ )(eV)
Nanda Shakti, 2010 [44]	3.21
S.M.H.Al-Jawad, 2011 [45]	3.27
S.S. Shariffudin, 2012 [46]	3.26
In this work	3.22

As observed from figure (11), there is a sharp decreasing in the refractive index for all thicknesses used for ZnO thin films in the wave length range



**Fig (11):** Refractive Index (n) as a Function of the Wavelength for ZnO Thin Films with Different Thicknesses

(350 - 400) nm. Also it is observe that the refractive index decreasing slightly in visible region (400 -700) nm, but in the UV region (700 – 1000) nm the refractive index fixed at a constant value almost.

**The Dispersion Energy Parameter (E<sub>o</sub>, E<sub>d</sub>) and Dielectric Constant (ε<sub>r</sub>, ε<sub>i</sub>, ε<sub>r</sub>)**

The analysis of the refractive index (n) with help of the extinction coefficient (k) has been carried out to obtain the real and imaginary part of the complex dielectric constant(ε<sub>r</sub>, ε<sub>i</sub>), dissipation factor (tan δ), relaxation time (T) and the optical conductivity (σ<sub>opt</sub>)

Wimple and Didomenico [32, 33], use a single oscillator description of the frequency dependent dielectric constant to define the dispersion energy parameter E<sub>d</sub> and E<sub>o</sub>. The relation between the refractive index (n) and single oscillator strength below the band gap is given by the expression [32, 33].

$$n^2 = 1 + \frac{E_o E_d}{E_o^2 - (hv)^2} \dots\dots\dots (11)$$

Where , E<sub>d</sub> :is the single oscillator constant (dispertion energy ), E<sub>o</sub> is the energy of the effective dispersion oscillator .The oscillator energy E<sub>o</sub> is an average of the

optical band gap E<sub>g</sub> and can be obtained from the Wemple – Didomenico model .

Experimental verification of equation (11) can be obtained by plotting (n<sup>2</sup>-1)<sup>-1</sup> against E<sup>2</sup> as shown in figure (12) for ZnO thin films at annealing temperate (450 °C) and thin films thicknesses (58, 69, and 77) nm respectively, which gives oscillator parameters by fitting a straight line for normal behavior having the slope (E<sub>o</sub>E<sub>d</sub>)<sup>-1</sup> and the intercept with the vertical axis equal to (E<sub>o</sub>/E<sub>d</sub>). The values of the parameters E<sub>o</sub> and E<sub>d</sub> are listed in table (3). The first procedure describes the contribution of the free carriers and the lattice vibration modes of the dispersion. The second procedure, however, is based upon the dispersion arising from the bound carriers in an empty lattice. To obtain a reliable value for the high frequency dielectric constant ε<sub>∞</sub>we employed both procedures.

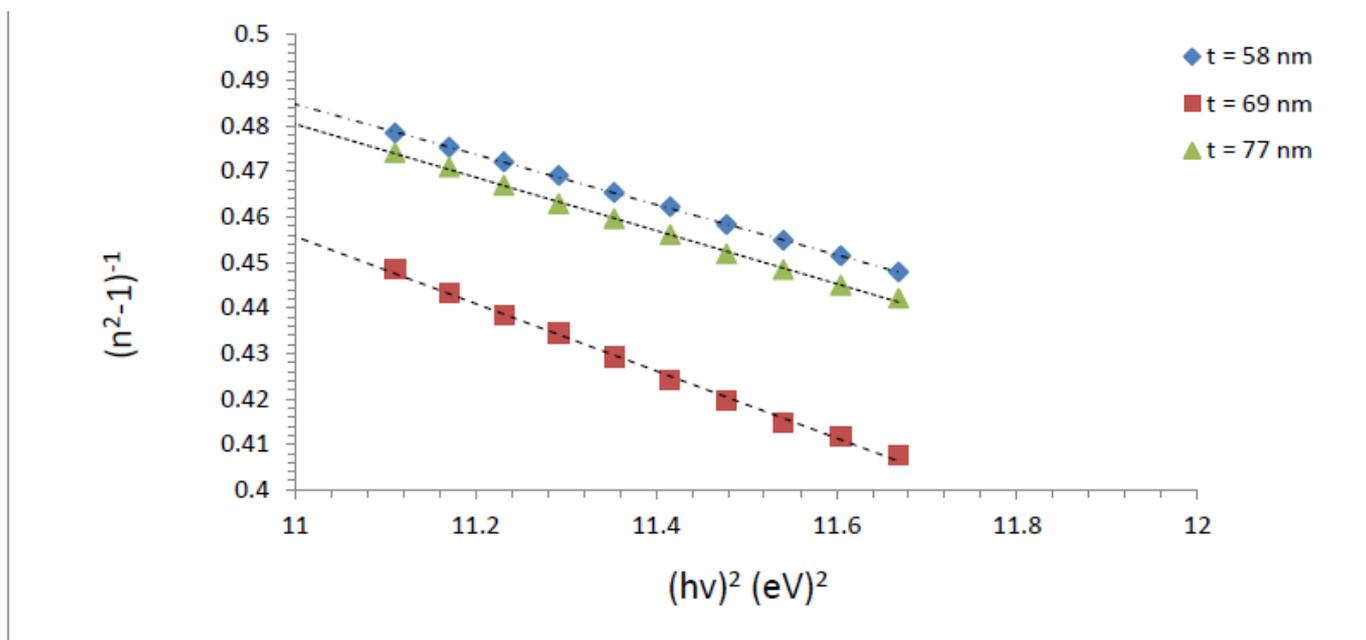
**The First Procedure**

The following equation can be used to obtain the high frequency dielectric constant [34]:

$$\epsilon_1 = \epsilon_{\infty(1)} - B\lambda^2 \dots\dots\dots (12)$$

Where

$$B = \frac{e^2 N}{4\pi^2 c^2 \epsilon_0 m^2} \dots\dots\dots (13)$$



**Fig (12):** Plot of  $(hv)^2$  against  $(n^2-1)^{-1}$  of ZnO thin films for different thickness (58 , 69 , 77 ) nm.

where  $\epsilon_1$  is the real part of dielectric constant,  $\epsilon_{\infty(1)}$  the lattice dielectric constant or (the high frequency dielectric constant) according to first procedure,  $\lambda$  the wavelength,  $N$  the free charge carrier concentration,  $\epsilon_0$  the permittivity of free space ( $8.854 \times 10^{-12}$  F/m),  $m^*$  the effective mass of the charge carrier and  $(c)$  the velocity of light. The real part of dielectric constants  $\epsilon_r = n^2$  was calculated at different values of  $(\lambda)$ . Then, the obtained values of  $\epsilon_r$  are plotted as a function of  $\lambda^2$  as shown in figure (13).

It is observed that the dependence of  $\epsilon_r$  on  $\lambda^2$  is linear at longer wave lengths. Extrapolating the linear part of this dependence to zero wavelength gives the value of  $\epsilon_{\infty(1)}$  and from the slopes of these lines, values of  $N/m^*$  for the investigated oxides were calculated according to the equation(13), of the constant  $B$ . The obtained of ZnO values of  $\epsilon_{\infty(2)}$  ,  $N/m^*$  are given in table(3).

**The Second Procedure**

The dielectric constant of a material could be calculated using the dispersion relation of incident photon. The refractive index was also fitted using a function for extrapolation towards shorter wavelengths. The model of Moss [35] which stated that the free carriers. Contribution to dispersion are relatively small. This means that data corresponding to the wavelength range lying below the

absorption edge of the material are to be used. In such a case, so, one can apply the next relation.

The properties of the investigated ZnO could be treated as a single oscillator at wavelength  $\lambda_0$  at high frequency. The high frequency dielectric constant can be calculated by applying the following simple classical dispersion relation [34]:

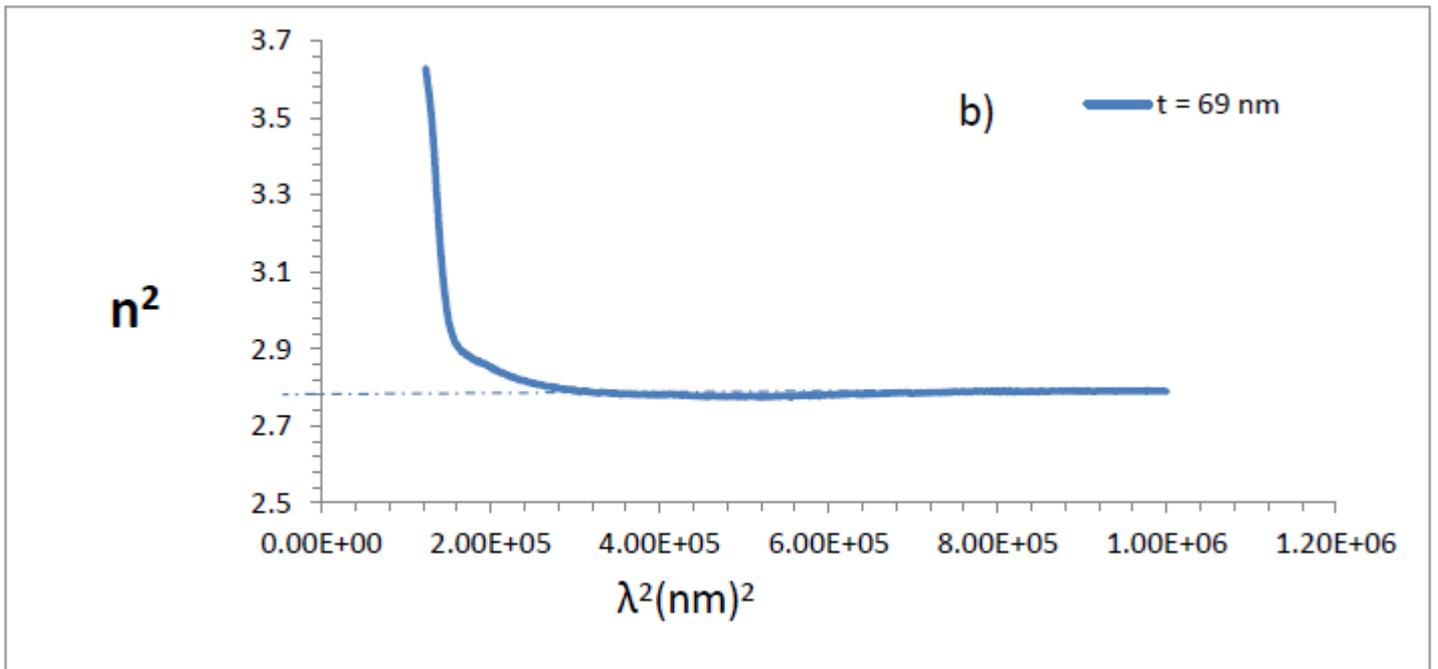
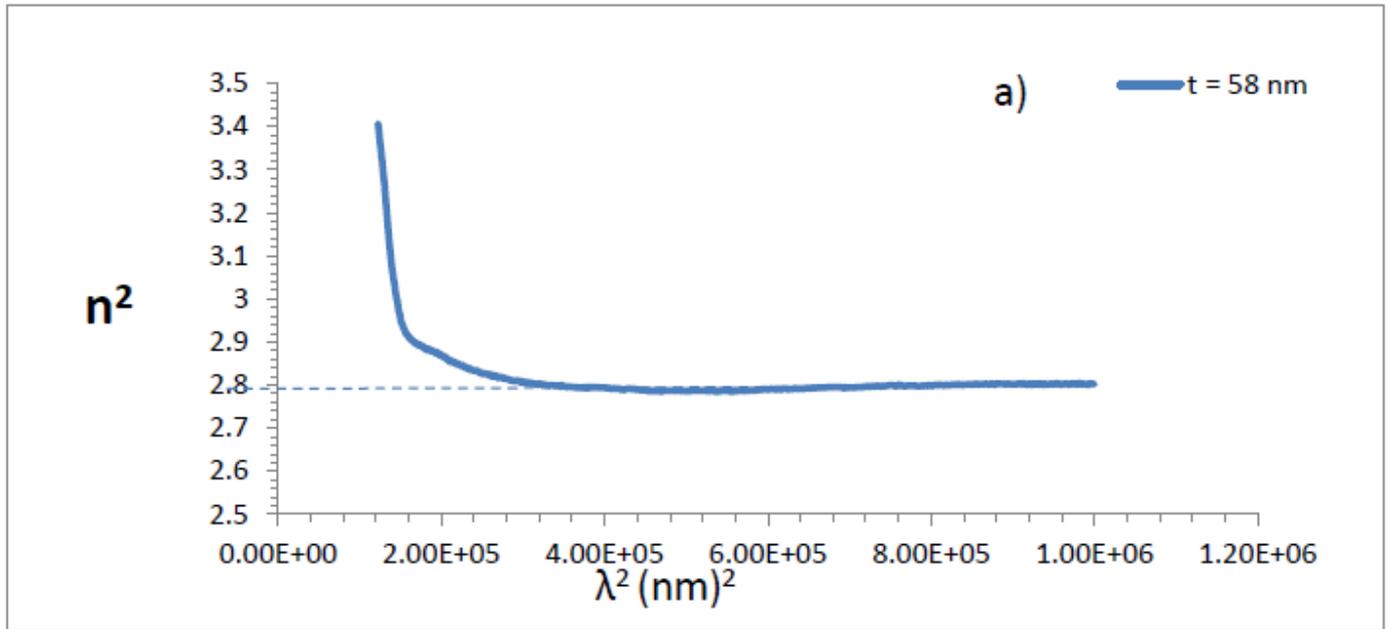
$$\frac{n_0^2-1}{n^2-1} = 1 - \left(\frac{\lambda_0}{\lambda}\right)^2 \dots\dots\dots (14)$$

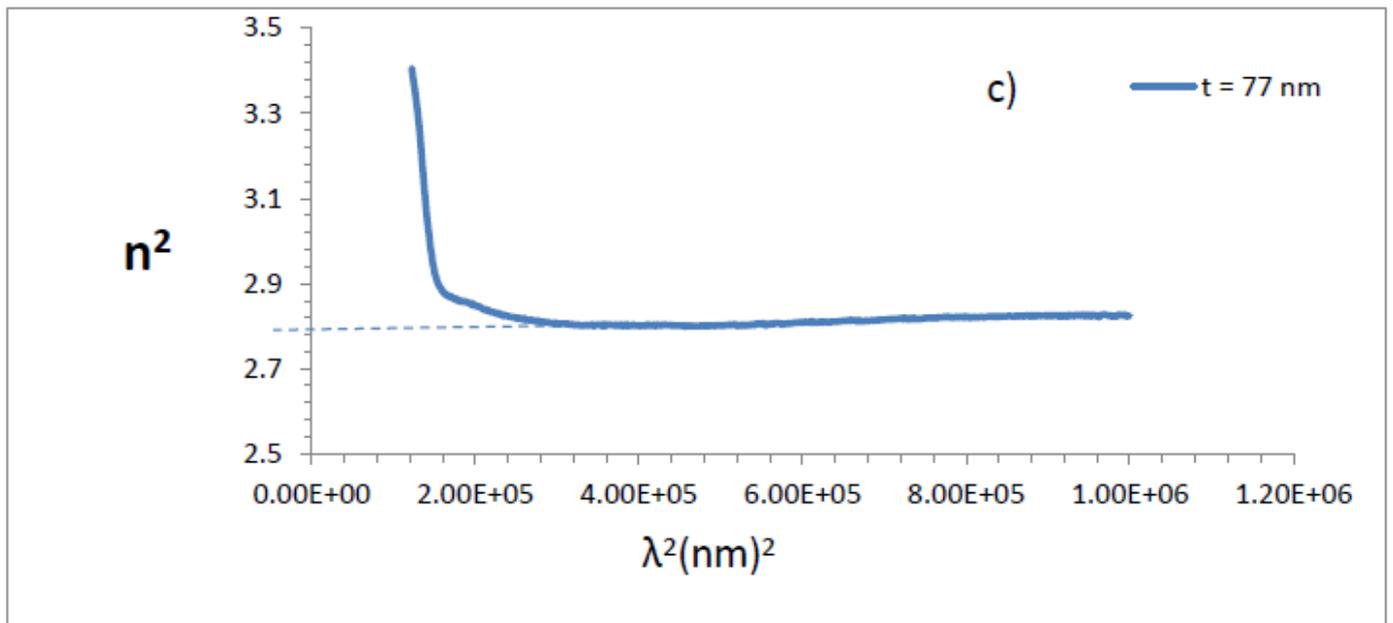
Where  $(n_0)$  is the refractive index at infinite wavelength  $\lambda_0$  (average interband oscillator wavelength),  $n$ : the refractive index and  $\lambda$ : the wavelength of the incident photon. Plotting  $(n^2 - 1)^{-1}$  against  $\lambda^{-2}$  which showed linear part, was below the absorption edge as shown in figure (14). The intersection with  $(n^2 - 1)^{-1}$  axis is  $(n_0^2 - 1)^{-1}$  and hence, at  $\lambda_0$  equal to  $\epsilon_{\infty(2)}$  (high frequency dielectric constant). Equation (14), can also be written as [36]:

$$n^2 - 1 = \left(\frac{S_0 \lambda_0^2}{1 - \left(\frac{\lambda_0}{\lambda}\right)^2}\right), \dots\dots\dots (15)$$

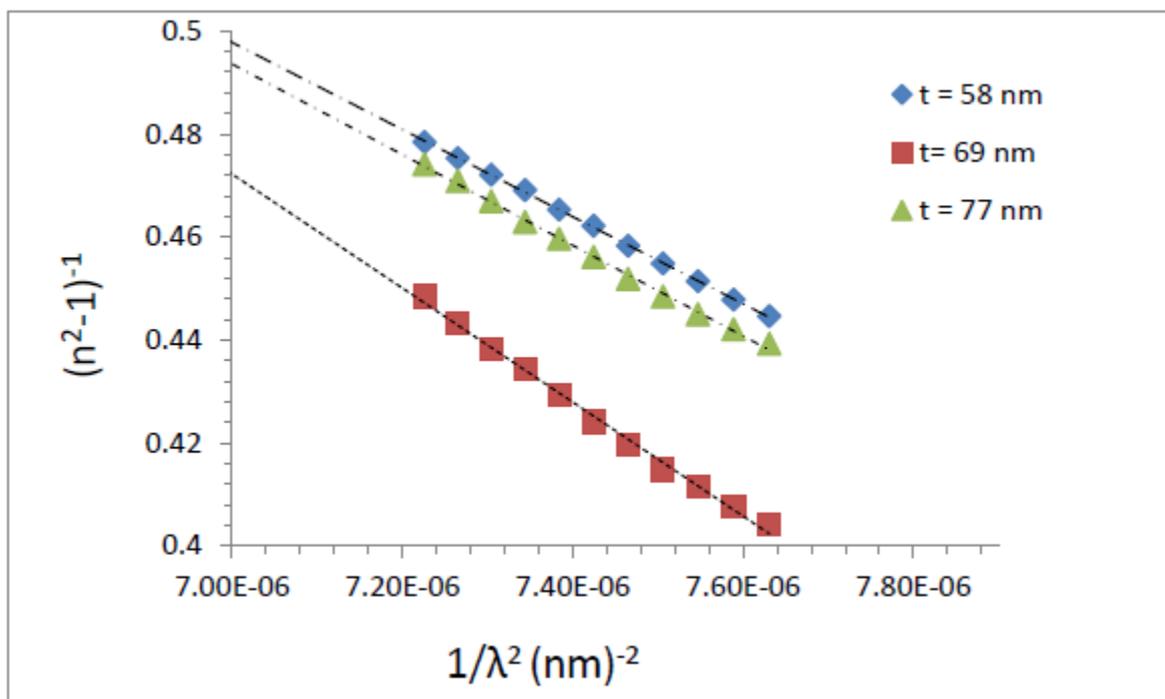
Where  $S_0$  is the average oscillator strength which equals to:

$$S_0 = \frac{n_0^2-1}{\lambda_0^2} \dots\dots\dots (16)$$





**Fig (13):** Plot of  $\epsilon_r = n^2$  as Function of  $\lambda^2$  for ZnO thin films thicknesses [a)  $t = 58$  nm , b)  $t = 69$  nm , c)  $t = 77$  nm]



**Fig (14):** Plot of  $\lambda^2$  against  $(n^2-1)^{-1}$  of ZnO thin films with thicknesses (58, 69, 77) nm.

The obtained values of  $S_0$  and  $\lambda_0$  are given in table (3).

**Table (3)** the value of optical parameter for ZnO thin films for different thickness

Parameters	Thickness		
	$t = 58$ nm	$t = 69$ nm	$t = 77$ nm
$E_g$ (eV)	3.225	3.222	3.222
$E_o$ (eV)	9.36	9.09	10.76
$E_d$ (eV)	15.93	15.27	18.58
$E_u$ (eV)	0.2340	0.2712	0.1839
$\epsilon_\infty = n^2$	2.716	2.680	2.759

$N/m^3 (m^{-3}.Kg^{-1})$	$2.454 \times 10^{56}$	$2.454 \times 10^{56}$	$1.227 \times 10^{56}$
$\lambda_0 (nm)$	123.77	134.39	109.14
$\epsilon_{\infty(2)} = n_0^2$	2.71	2.68	2.73
$S_0 (m^{-2})$	$1.12 \times 10^{13}$	$9.31 \times 10^{13}$	$1.45 \times 10^{13}$
$\epsilon_{\infty(1)}$	2.901	2.9377	2.754

It is clear from table (3) that the values of  $\epsilon_{\infty(1)}$  and  $\epsilon_{\infty(2)}$  obtained from the two procedures approximately agreed with each other, small differences may be attributed to the lattice vibrations and bounded carriers in an empty lattice which are in the transparent region [37].

**Determination of Complex Dielectric Constant**

The complex refractive index  $\bar{n} = n + ik$  and dielectric function  $\epsilon = \epsilon_r + i\epsilon_i$  characterize the optical properties of any solid material. The imaginary and real parts of dielectric constant of ZnO thin films were also determined by the following relations [38,39]:

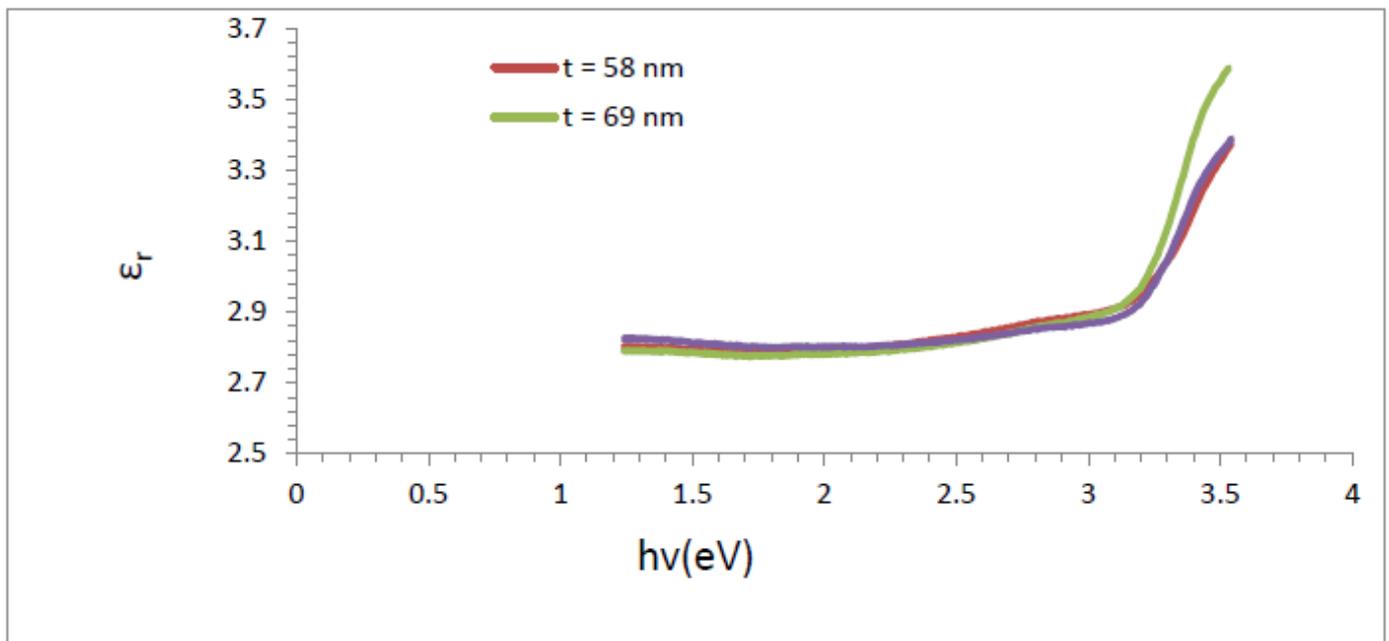
$$\epsilon_r = n^2 - k^2 = \epsilon_{\infty} - \left(\frac{e^2 N}{4\pi^2 c^3 \tau}\right) \lambda^2 \dots\dots\dots (17)$$

And

$$\epsilon_i = 2nk = \left(\frac{\epsilon_{\infty} \omega_p^2}{8\pi^2 c^3 \tau}\right) \lambda^3 \dots\dots\dots (18)$$

Where  $\epsilon_r$  is the real part,  $\epsilon_i$  the imaginary part of the dielectric constant,  $\epsilon_{\infty}$  is the high frequency dielectric constant,  $\omega_p$  is the plasma frequency,  $\tau$  the optical relaxation time where (k) extinction coefficient (given in equation(8)).

The imaginary and real parts of the dielectric constant can be calculated as it is directly related to the density of states within the forbidden gap of the investigated ZnO [38, 39]. The real and imaginary parts of the dielectric constant of the films are shown in figure. (15) And (16) For ZnO thin films. It is seen that both ( $\epsilon_r$ ) and ( $\epsilon_i$ ) increases with increasing photon energy. The real and imaginary parts follow the same pattern and it is seen that the values of real part are higher than the imaginary parts.



**Fig(15):**Plot of  $\epsilon_r$  as a Function of (hv) of Different Thin Films Thickness for ZnO Thin Films

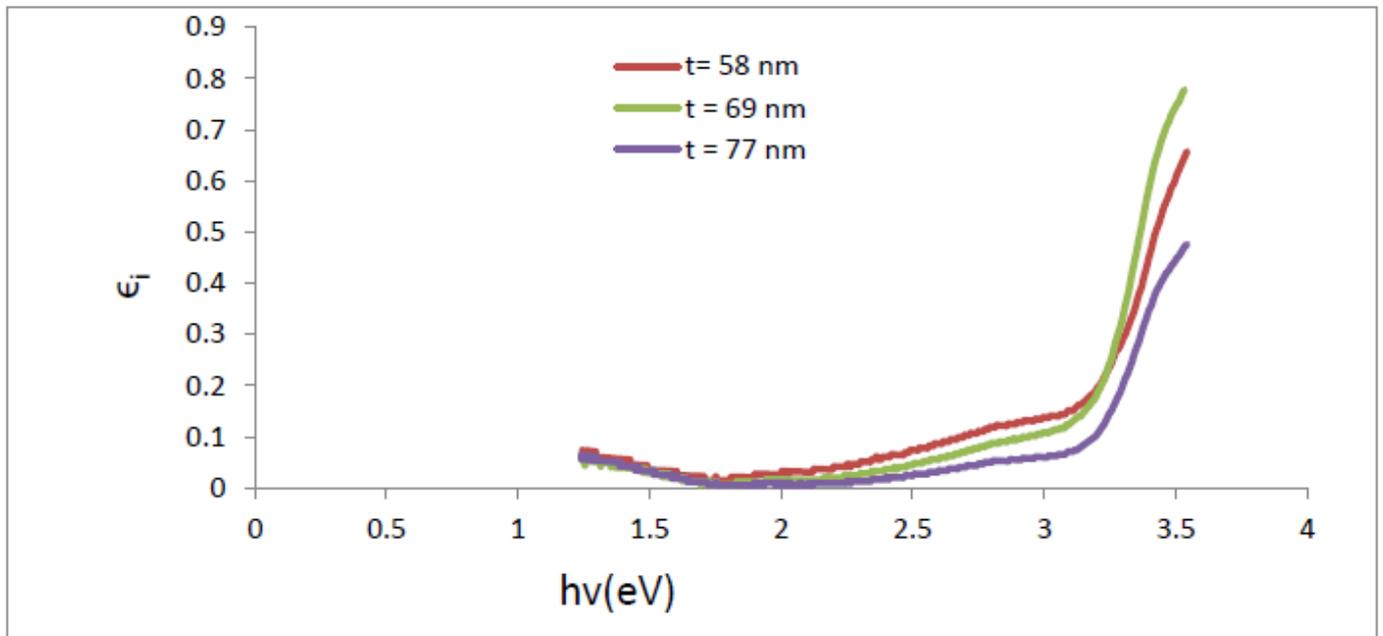
**Relaxation Time ( $\tau$ ), Dissipation factor ( $\tan \delta$ ) and optical conductivity**

The dielectric relaxation time  $\tau$  can be evaluated by using the relation [40,41].

$$\tau = \frac{\epsilon_{\infty} - \epsilon_r}{\omega \epsilon_i} \dots\dots\dots (19)$$

Figure (17) shows the dielectric relaxation time (T) as a function of photon energy ( $E=hv$ ) for ZnO thin films.

The curve between relaxation time and photon energy (hv) are applies for different values of thickness es(58, 69, 77) nm.



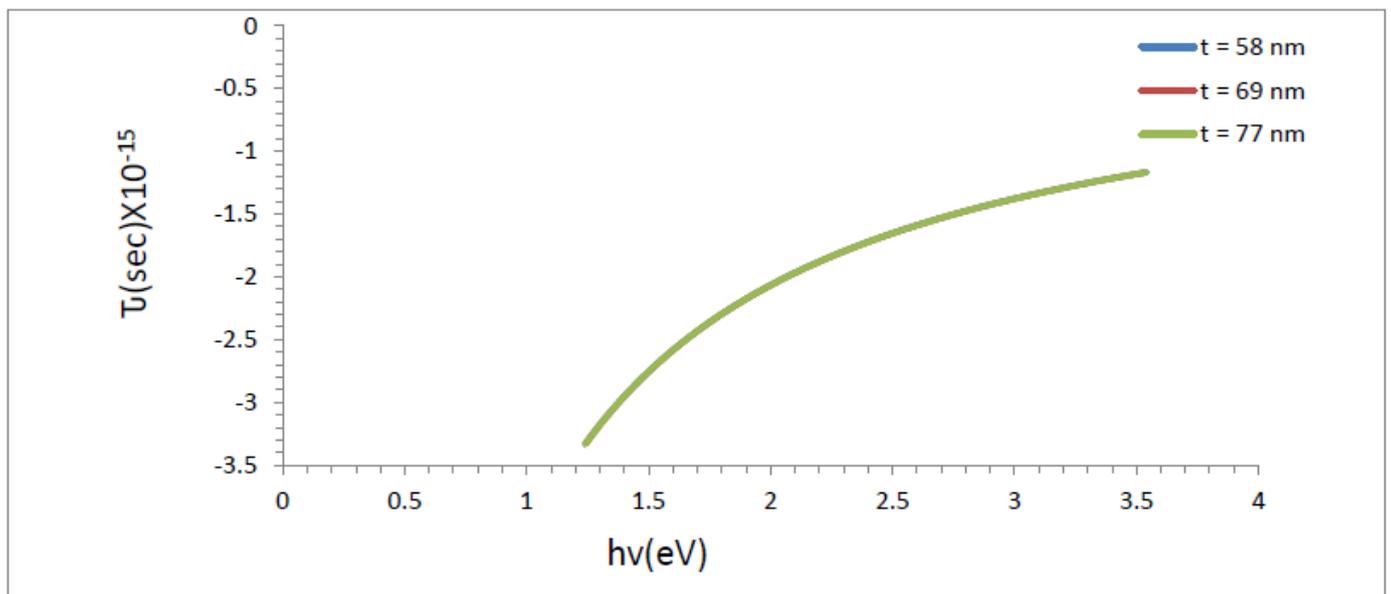
**Fig (16):** Plot of  $\epsilon_i$  as a Function of  $(hv)$  of Different Thin Films Thickness for ZnO Thin Films

The dissipation factor ( $\tan \delta$ ) is a measure of loss-rate of power of a mechanical mode, such as an oscillation, in a dissipative system. For example, electric power is lost in all dielectric materials, usually in the form of heat. The dissipation factor  $\tan \delta$  can be calculated according to the following equation [42]:

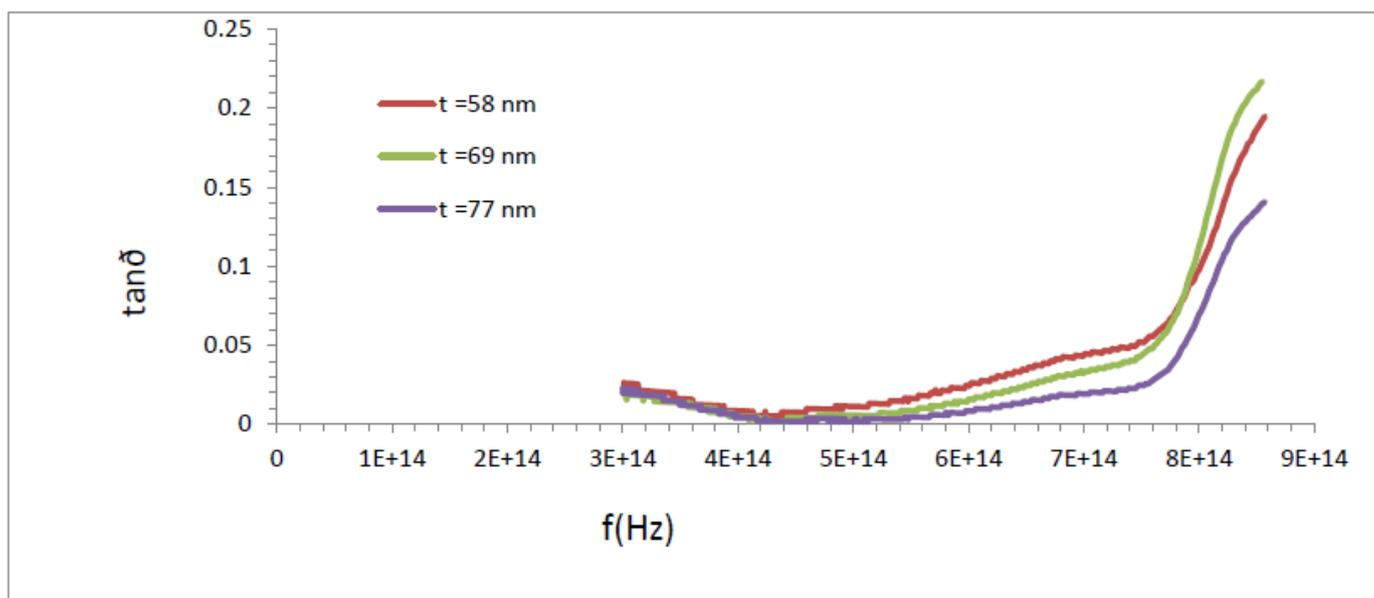
$$\tan \delta = \frac{\epsilon_i}{\epsilon_r} \dots (20)$$

The variation of dissipation factor of the investigated films with frequency ( $f$ ) is shown in figure. (18).

It is found that the dissipation factor increases with increasing photon energy in the absorption region.



**Fig (17):** Dependence of Relaxation Time on the Photon Energy With Different Thicknesses of ZnO Thin Films

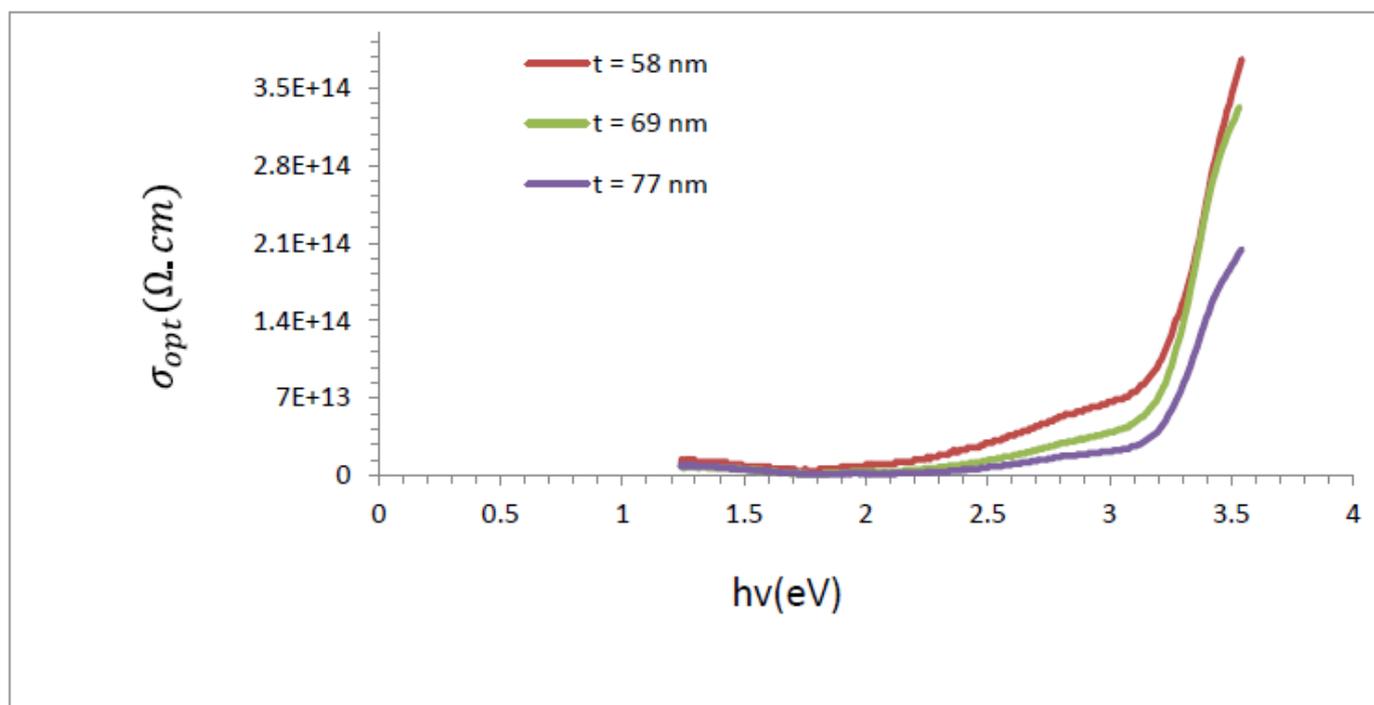


**Fig (18):** Dependence of Dissipation Factor  $\tan\delta$  on the Frequency at Different Thicknesses of ZnO Thin Films

The absorption coefficient  $\alpha$  can be used to calculate the optical conductivity  $\sigma_{opt}$  as follow [43]:

$$\sigma_{opt} = \frac{anc}{4\pi} \dots\dots\dots (21)$$

Figure (19) shows the variation of optical conductivity  $\sigma_{opt}$  as a function of photon energy  $h\nu$



**Fig.(19):** Dependence of Optical Conductivity on the Photon Energy  $h\nu$  at Different Thicknesses of ZnO Thin Films

**Conclusions**

ZnO thin films were successfully deposited on to glass substrate at room temperature by using Sol – Gel technique .The effect of thickness of thin film on the

structure and optical properties of ZnO thin films has been studied.

The XRD spectrum shows that thin films are polycrystalline, crystallized in the wurtzite phase. Thermal annealing of ZnO thin films can show that the best

temperature of annealing is at ( $T= 450\text{ }^{\circ}\text{C}$ ), that occurs in this temperature increase in grain size ( $D$ ) and decrease the dislocation density ( $\delta$ ) and micro-strain ( $\epsilon$ ), also mean values of lattice parameters have been as ( $a=3.2\text{ }A_o$ ,  $c= 5.6\text{ }A_o$ ) that agree with standard values. However no change in the orientation of the films is observed. The films show high transparency ( $>85\%$ ) in the visible region and UV region (as shown in figure 5), which makes it a good material for optoelectronic devices. High transparency may be due to diffusion of impurity ions from the sodalime glass.

The refractive index  $n$  and absorption index  $k$  were computed from the obtain transmittance  $T(y)$  using Swenepoel's method. On the basis of the optical investigations of the films, the following results were obtained. The type of optical transition responsible for optical absorption was indirect transition.

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