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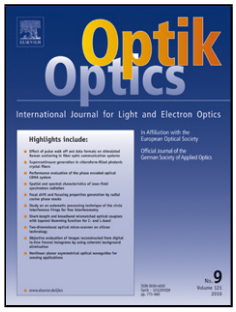

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Investigation of dispersion parameters, dielectric properties and opto–electrical parameters Of ZnO thin film grown by ALD

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Abstract

Highly transparent zinc oxide thin films with varied layer thicknesses have been prepared on microscopic glass substrates at 200 °C. Films thickness was measured by stylus profilometer. The films have been investigated for their structure using X-Ray diffraction; the patterns showed their amorphous nature. The dispersion parameters i.e. refractive index (n) and extinction coefficient (k) are computed in the wavelength range (350 -2500 nm). The Tauc model was used to determine the optical band gap and Urbach tail with direct allowed transitions. The real and imaginary parts of the high frequency dielectric constant were discussed. Other parameters such as penetration depth, cut-off wavelength, dissipation factor, volume and surface energy loss functions, reflection loss factor, optical, electrical and thermal conductivities have also been determined. A systematic study of a wide range of optical parameters of ALD prepared ZnO films can serve as a valuable data source and can enrich the knowledge of the studied material.

Keywords:

ZnO; ALD; Thin Films; Opto-electrical parameters; Direct optical band gap

1. Introduction

Zinc oxide (ZnO), a perspective transparent conductive oxide (TCO), has been a hot spot of research because it provides extremely valuable insight in recent device applications like solar cells owing to its excellent properties for obtaining high efficiency with safety and simultaneously maintaining low cost [1-3]. These TCOs have a combination of two coadunate properties that include high electrical conductivity and optical transparency. The thin films of these transparent conductive oxide materials having exceptional advantages such as chemical stability and electrochemical activity that are worthy for recent device applications like optical waveguides, thin film solar cells, light emitting diodes, gas sensors, thin film transistors, etc. [4-8]. ZnO also emits intrinsic luminescence with minor emission tail in the green spectra. The emission is on account of the presence of defects of oxygen and the zinc vacancies [9].

ZnO can be considered as a replacement for some other wide energy gap semiconductor, likegallium nitride (E_g = 3.4 eV at 300 K), for applications in optoelectronic components owing to its stability under high energy radiation. In addition,ZnO can be grown in several nanostructure morphologies owing to its relatively cheap production and simultaneously have dominant optical properties [2,10-14]. In addition, ZnO has also been investigated for UV compelledtunable light sources, green phosphors,white light generation, nano-lasing, and optical non-linear applications [8,15-18]. ZnO has shown exciting photoluminescence just about 380 nm [18]. Due to these pronounced physical properties, many attempts and various effective methods of preparation have been accounted for in the published work to prepare ZnO nanomaterial. A variety of techniques can be used to produce ZnO, such as mechano-chemical processing, precipitation, sol-gel method, hydrothermal processing, solvothermal method, spray pyrolysis, thermal decomposition of organic precursors, chemical vapor deposition (CVD), pulsed laser deposition (PLD), radio frequency (RF) sputtering, direct oxidation from Zn metal block and metal organic chemical vapor deposition (MOCVD)[19-30].

According to the literature[31-33], the detailed investigationof ZnO thin films prepared by ALD for the optical properties is still required to have more reliable optical and physical properties. In fact, that is why, in the present study, first we discuss the effect of the thickness of ZnO thin films prepared by the ALD technique on the optical and physical parameters. Finally, to the best of our knowledge, no attention has been paid in the literature to study the optical parameters that are investigated for ZnO thin films prepared by ALD technique. Therefore; we found difficulties in comparing the obtained physical parameters with similar results. Consequently, it is interesting to study the optical behaviour of this material which might be useful for industrial considerations, too, when decided which techniques is more beneficial for a specific application.

2. Experimental techniques

ZnO was prepared on glass substrates using a Beneq TFS-200 atomic layer deposition ALD unit in thermal mode. Diethylzinc (DEZ) and water precursors were used for the deposition. During the deposition, the base pressure of the reactor was kept around 1 mbar and the pressure in the main chamber was kept at 9 mbar. The temperature of the chamber was set at 200 0C. One complete ALD cycle consisted of the following four steps: (1) pulse of DEZ for 0.1 sec, (2) purge the reactor with nitrogen for 3 seconds, (3) pulse of water for 0.1 sec, and finally (4) purge the reactor again with nitrogen for 3 seconds. During the process, the thickness of the deposited ZnO layer was operated by the number of cycles and was determined by stylus profilometer (AMBIOS XP-1).

The optical transmittance (T) and reflectance (R) of four ALD grew ZnO films having varied thicknesses (130nm, 190nm, 260nm and 335 nm) were recorded by utilizing double beam spectrophotometer (UV-3101PC Shimadzu)in the wavelength range(350 to 2500) nm. The observation reported in this work were taken at room temperature (27 °C). The structure of the ALD grew ZnO films with varied thicknesses was analyzed by the X-ray diffraction (in the range of 2θ =10°-80°) method. The X-ray spectrum of the sample with 260 nm thickness, as a representative example, is shown in Fig. 1. The XRD spectra of various ALD grew ZnO thin films with varied thicknesses (130, 190, and 335 nm) resembled and proved to be amorphous too.

3. Results and discussion:

3.1. Dispersion parameters of ALD grew ZnO:

Transmission (T) and reflectance (R) of films depend on the thickness of the film and its crystal structure and the degree of substrate heating as well. Figure 2 depicts the T and R spectra (measured at room temperature) from 350 nm to 2500 nm of the four ZnO films prepared with varied thicknesses (130 nm, 190 nm, 260 nm and 335 nm). From figure 2 it has been found that the ALD grown thin films have a maximum transmittance in the visible region *i.e.* above 80 %. It is also observed that the T and R are affected by the film thickness. Moreover, the investigated films are transparent in this region. This emphasizes that the ZnO films grown by ALD have no absorbed or scattered light. The variation in T& R depends on the method of preparation, structure and surface homogeneity. T and R measured data were performed in order to determine the optical parameters, the refractive index n, the extinction coefficient k and absorption coefficient α, according to the equations given in details [34], in combination with a particular computer program given in [35, 36]. As a general feature, the obtained results of n & k are not depending on the film thickness since the variation in the values of the parameters lies within the experimental errors. Figure 3 shows the spectral distribution of the averages of n & k for ZnO thin films. As seen in figure 3, the values of both n & k decrease with the increase of λ. The decrease in n with λ shows a normal dispersion behaviour while the decrease in k with an increase

in λ shows that the proportion of light loss on account ofthe decrease in scattering and absorbance. Our results are in a good agreement with [37, 38] for ZnO and with [39] for other different oxides. The k values computed for ZnO thin films have been analyzed to estimate the optical band gap and absorption coefficient (α). The absorption coefficient has been determined using the relation [40]

$$\alpha = 4\pi k/\lambda \qquad (1)$$

Figure 4 shows an increase in the absorption coefficient (α) ofZnO thin films with an increase in hν.

The Transmittance date of our films can also be used to determine the absorption coefficient α according to the following relation [41]

$$\alpha = (1/t)\ \ln(1/T) \qquad (2)$$

Where t is the thickness of the film.

For many materials in the week-absorption region, the values of α depend exponentially on hν and can be given as [42]

$$\ln(\alpha) = \ln(\alpha_o) + h\nu/E_u \qquad (3)$$

Whereα_0 is a constant and E_u is a slope parameter and this parameter is generally believed to constitute the irregularity of the glass structure. The above equation(3) was first suggested by Urbach [42] to report the absorption coefficient $\alpha<10^4$ cm^{-1} where the absorption is on account ofthe electronic transition between the tail states of the valence band and conduction band. The absorption coefficient has shown exponential dependence on photon energy. From equation (3), the absorption edge behavior has been determined and is plotted in Fig.(5). The value of E_u and α_o are listed in Table 1. As seen they are in agreement with the results of X-ray given in Fig.1 corresponding to the present work.

Extending with the investigation of the absorption edge for higher values of the absorption coefficient α that is $>10^4$ cm^{-1}, the optical band gap E_g can be defined from Tauc's relation [43]

$$\alpha h\nu = A(h\nu - E_g)^r \qquad (4)$$

where A is a constant and r is a power which represents the type of optical transition process. After following different values of r, it has been found that the ZnO films conform to Eq. (4) The value of r=0.5 was suggested by our wark which indicates the direct allowed transition. Considering equations (1&2) , $(\alpha h\nu)^2$ values can be plotted as a function of photon energy according to equation (4) (see Fig.6 a&b) . The E_g is estimated from the intersection of the straight line with the photon energy axis. In Table 1 the values obtained for E_g are listed. It's found that, the two obtained E_g values are in a good agreement with each other within the experimental error. Our average value of E_g is supported with previously reported results of other researches for ZnO films prepared by different methods [37, 44-50] as seen in Table 2.

The variation of the optical absorption (α) inside the thin film depends upon many parameters, one of the important parameters that relate the absorption of the photons with the texture of the thin film is skin depth or penetration depth (φ). The penetration depth is a measure of how deep light or any electromagnetic radiation can penetrate into a material; it is defined as the depth at which the intensity of the radiation inside the materials falls to about 37% of its original value. The absorption coefficient is the best parameter for determining penetration depth according to the following relation [51]

$$\varphi = 1/\alpha \qquad (5)$$

The dependence of the calculated skin depth (φ) on the photon energy is given in Fig. (7). As observed the values of φ decrease with the increase of the photon energy.It is clear from the above Figure , that the cut-off wavelength ($\lambda_{cut\text{-}off}$) the absorption effect vanishes (see Table 1).

3.2. Dielectric properties of ZnO films

The optical constants "n and k" computed for the ZnO thin films can be utilized to obtain dielectric parameters and that are used to explain the electronic excitation spectrum of thin films. The real (ε_1) and imaginary (ε_2) parts of dielectric constants are very important parameters because they provide information concerning the optical behaviour and the dispersion factor.

The real ε_1 and imaginary ε_2 parts of the dielectric constants can be derived using the following relations [52, 53]:

$$\varepsilon_1 = n^2 - k^2 \qquad (6)$$

$$\varepsilon_2 = 2nk \qquad (7)$$

The dependence of $\varepsilon_1$and $\varepsilon_2$upon the incident photon energy for ZnO film is shown in Fig. 8. As observed the obtained values of $\varepsilon_1$and $\varepsilon_2$are detected to enhance with the increase of photon energy. The value of ε_1 is found to be much higher than ε_2; this comparative result reveals the dependence of$\varepsilon_1$on n where n › k as seen in Fig. 3. The real part of the dielectric constant enhancesabruptly with the increase of hν, on the other hand, the imaginary part of the dielectric constant is nearly constant in the lower energy region and increases quickly starting from 2.8 eV.

The dielectric loss factor (tan (δ)) is a valuable factor in the investigation of the structure and defects in solids and also within the sample, it represents the phase difference owing to the loss of energy. The tan (δ) is dependent on the real and imaginary part as[54, 55]

$$\tan(\delta) = \varepsilon_2 / \varepsilon_1 \qquad (8)$$

Figure 9 depicts the graph of tan (δ) versus photon energy. Nearly constant dependence is observed at lower photon energy and a strong dependence is observed at incident energy above 2.8 eV; this because of the abrupt rise in the absorption coefficient. Accordingly, the

origin and nature of dielectric losses are interesting. The same observed behavior of the dissipation factor was also reported previously by others [56, 57]

When the energetic electron beam goes through the thin films, the passing electrons experience energy loss [58]. The probability of this energy lossis governed by the passage of electrons, if they pass inside the material then the loss is determined by the volume(VELF) energy loss function and if they pass over the surface of the material then the loss is determined by the surface(SELF) energy loss function. These two functions can be estimated by using the following relations [58, 59].

$$\mathrm{VELF} = \frac{\varepsilon_2}{\varepsilon_1^2 - \varepsilon_2^2} \qquad (9)$$

$$\mathrm{SELF} = \frac{\varepsilon_2}{(\varepsilon_1+1)^2 + \varepsilon_2^2} \qquad (10)$$

Figure10 represents the spectral dependence of VELF and SELF for ZnO film. As obvious both curves behave similarly, however, VELF is a bit prominent than SELF. Consequently, the energy loss of the free charge carriers suffers from collisions within the material that causes more loss of energy than when travelling on the surface [59].

3.3. Optical, electrical and thermal conductivities:

The optical conductivity(σ_{opt}) of a material which generally represents the optical response, is conveniently studied using optical absorption according to the relation [60]

$$\sigma_{opt} = \alpha nc/4\pi \qquad (11)$$

where c is the velocity of light. Fig. 11 (a) shows the increase of the optical conductivity in ZnO thin film with hv. The optical conductivity right away relies upon the absorption coefficient and the refractive index and is noticed to comply with the identical trend with the increase of hv. The high magnitude of the σ_{opt} answers the high photo-response of the ZnO film.

The high magnitude of the σ_{opt} could also be attributed to the increase of electron excitation with the increase of photon energy hv. The electrical conductivity of ZnO film can be correlated with the optical conductivity σ_{opt} and absorption coefficient α, from the relation [61, 62]

$$\sigma_{elec} = 2\,\lambda\,\sigma_{opt}/\,\alpha \qquad (12)$$

Fig. 11(b) shows the photon energy dependence of the electrical conductivity for ZnO film. One can observe that the σ_{elec} decreases as the hv increases.

Moreover, the thermal conductivity of ZnO thin films has also been computed from electrical conductivity σ_{elec} by the relation

$$\sigma_{therm} = L\,T_a\,\sigma_{elec} \qquad (13)$$

where L is the Lorentz's number and T_a is the absolute temperature. Fig.11(c) depicts the plot of the thermal conductivity with hv for the ALD grown ZnO thin films. The decrease of the electrical conductivity and the thermal conductivity with the increase in hv corroborates the semiconducting behavior of ZnO thin films.

The relationship between the refractive index n , and the reflection loss factor R_L can be described from the relation[63]

$$R_L = (n^2-1)/(n^2+2) \quad (14)$$

The dependence of R_L of ZnO upon the incident photon energy is shown in Fig 12 .The value of R_L can be determined and reported also in Table1. The reflection loss factor is related to the field of optics, therefore for manufacting any optical nonlinearity material we have to put this strong correlation into consideration.

4. Conclusions

Atomic layer deposition technique has been used to grow the ZnO thin films. The ALD grown thin films have been investigated for structural, optical and dielectric properties to enhance the knowledge of ZnO thin films. The X-ray diffraction of the as-deposited films shows their amorphous structure. The dispersion parameters n & k were used for the determination of all the studied parameters. Optical analysis indicated direct allowed transitions with optical band gap E_g seems to be in a good agreement with available published data. The variation of the real and imaginary parts of dielectric constants with photon energy indicates the interaction between the free electrons and the incident photons. The dielectric constants, the dissipation factor, the volume and surface energy loss functions, optical, electrical thermal conductivities, and the reflection loss factor were determined. The penetration depth ϕ and the value of the cut-off wavelength ($\lambda_{cut\text{-}off}$) have been calculated and discussed in terms of the absorption coefficient.. A systematic study of a wide range of optical parameters of ALD prepared ZnO films can serve as a valuable data source, owing to its superior optical properties as well as its relatively low-cost production. Unfortunately we could not get similar previous work for the penetration depth and the reflection loss to compare with our results.

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List of Figure captions:

Figure 1

The X-ray pattern of ZnO thin film with thickness of 260 nm.

Figure 2

The distribution of R and T versus the wavelength λ.

Figure 3

The spectral distributions of the mean values of n and k vs. wavelength.

Figure 4

The plot of (α)versus photon energy ($h\nu$).

Figure 5

The plot of ln (α) versus photon energy ($h\nu$).

Figure 6

The Tauc plot for $(\alpha h\nu)^2$ as a function of photon energy for ZnO films

(a) α according equation (1) (b) α according equation (2).

Figure 7

The plot of penetration depth)vs. photon energy

Figure 8

The real ε_1 and imaginary ε_2 parts of the complex dielectric constant.

Figure 9

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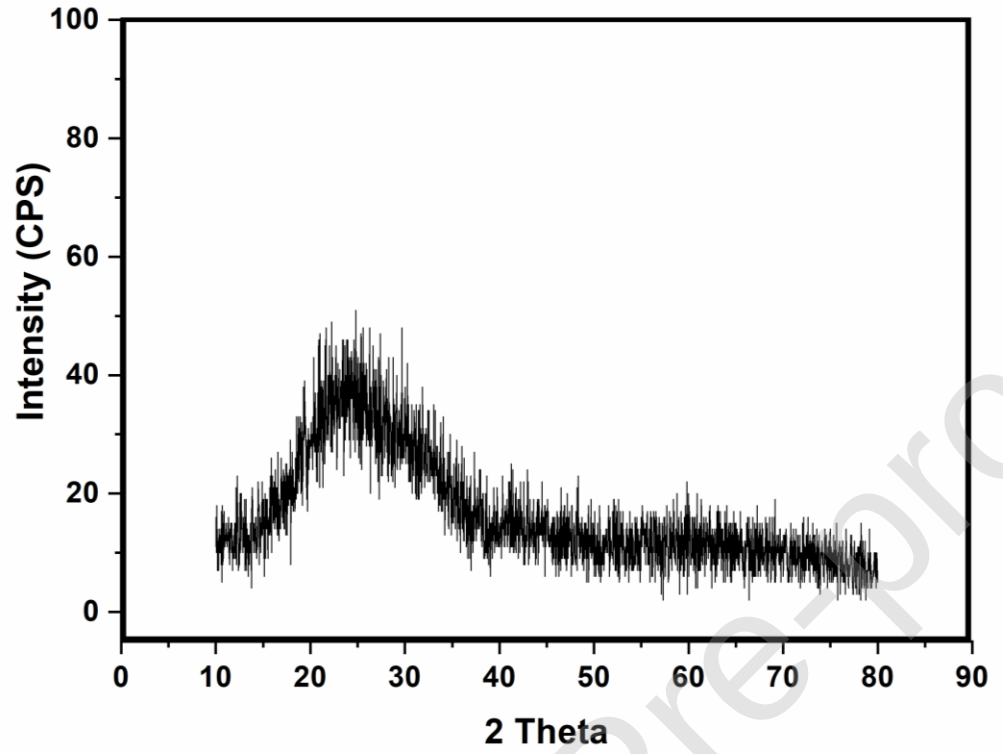


Figure 1

The X-ray pattern of ZnO thin film with thickness of 260 nm.

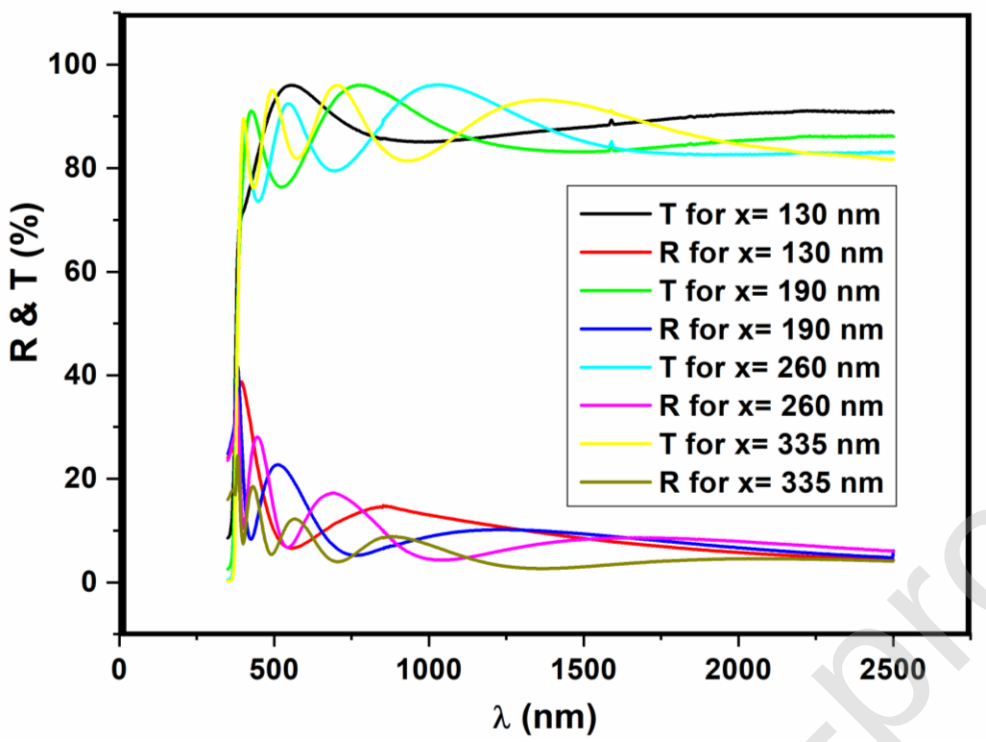


Figure 2

The distribution of R and T versus the wavelength λ.

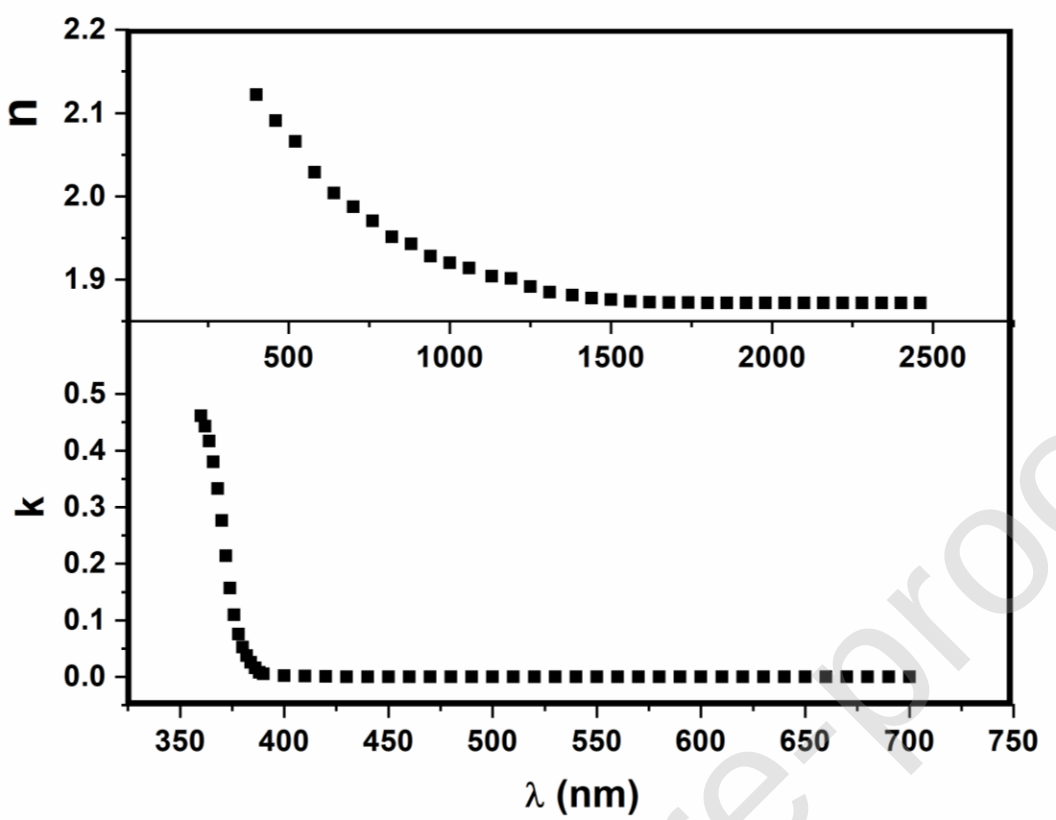


Figure 3

The spectral distributions of the mean values of n and k vs. wavelength.

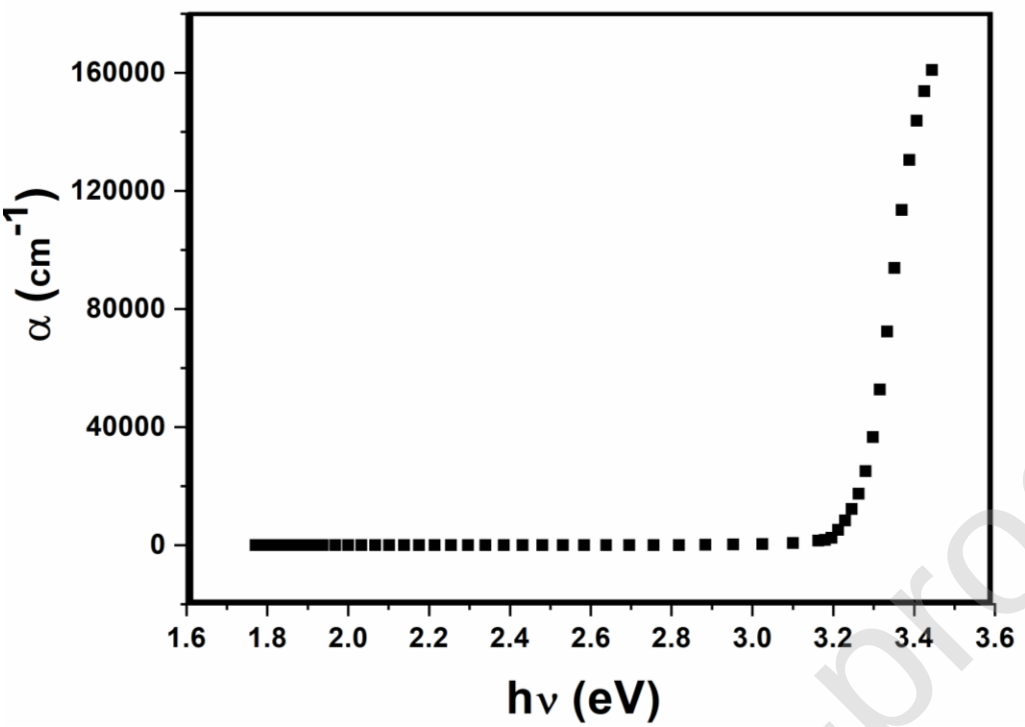


Figure 4

The plot of (α)versus photon energy (hν).

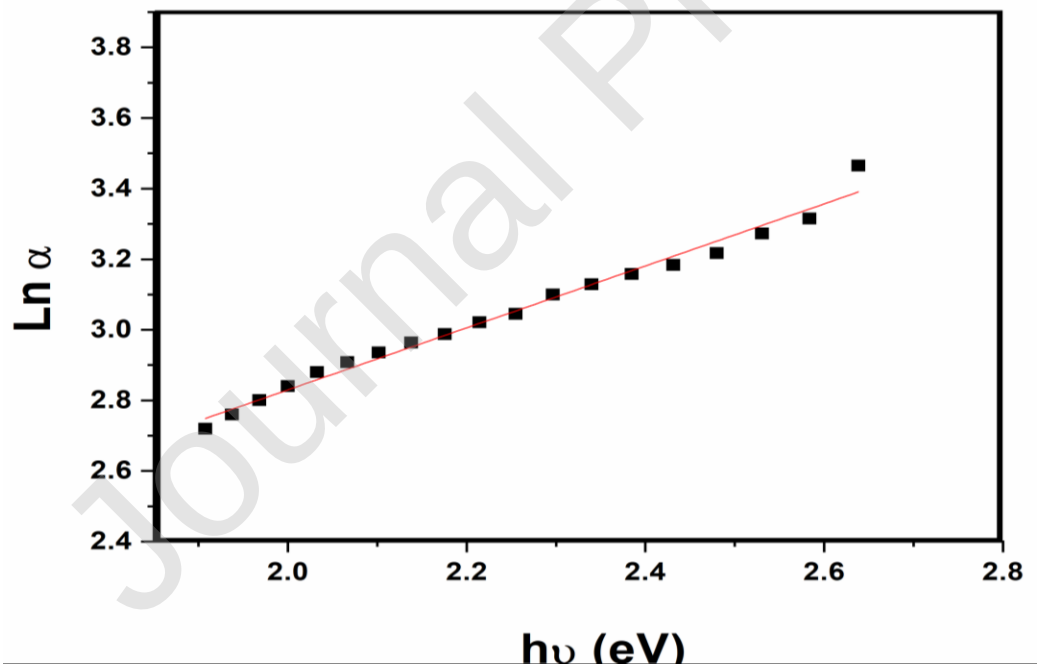
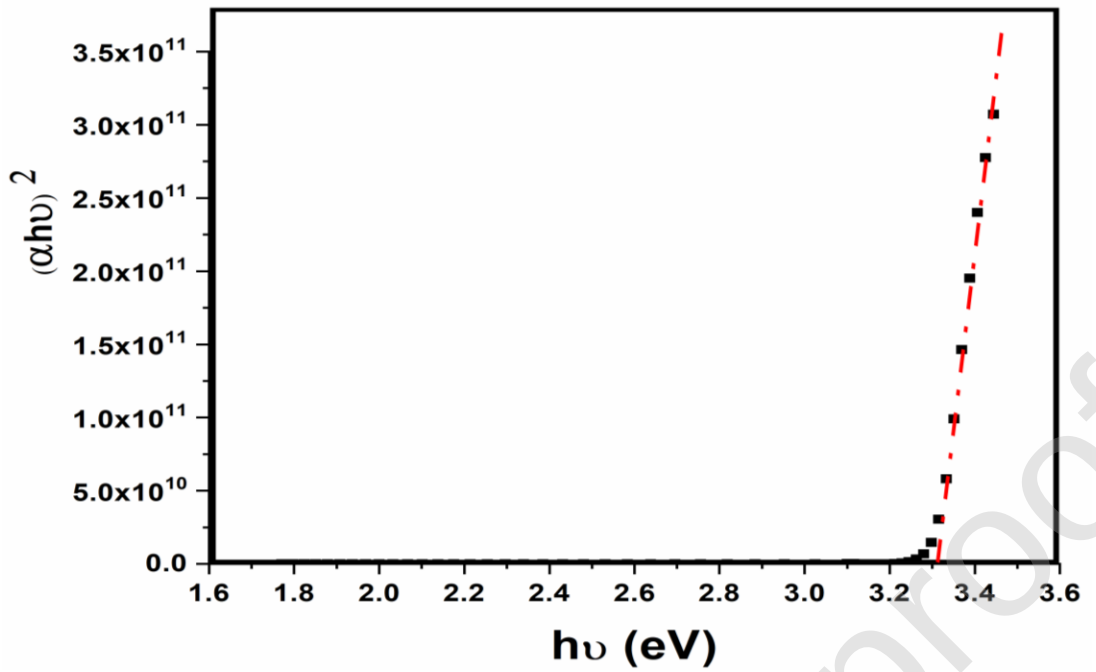


Figure 5

The plot of ln (α) versus photon energy (hν).



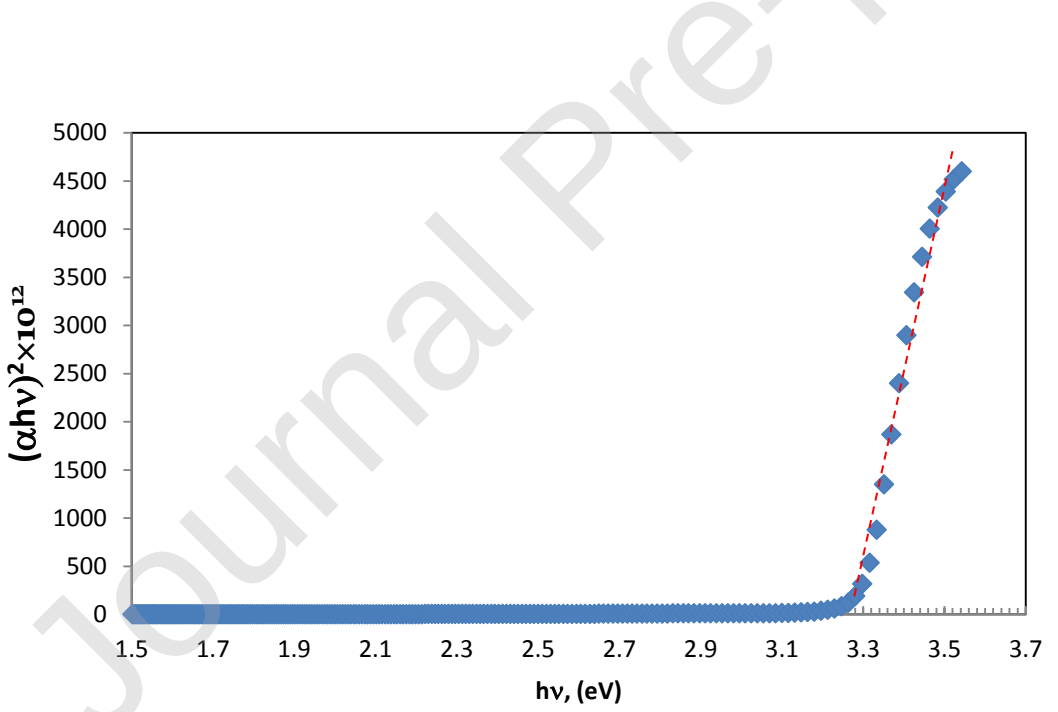


Figure 6

The Tauc plot for $(\alpha h\nu)^2$ as a function of photon energy for ZnO films (a) α according equation (1) (b) α according equation (2).

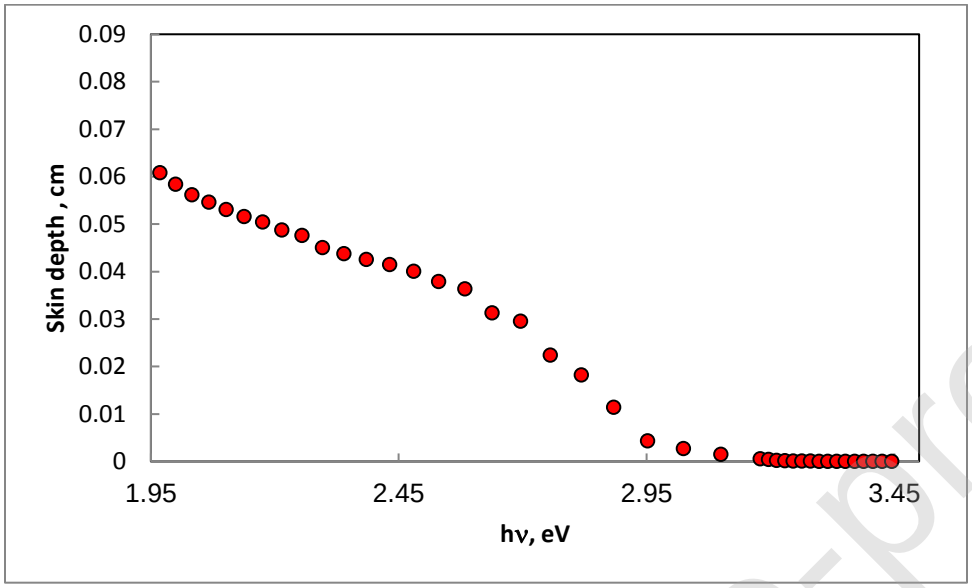


Figure 7

The plot of skin depth)vs. photon energy

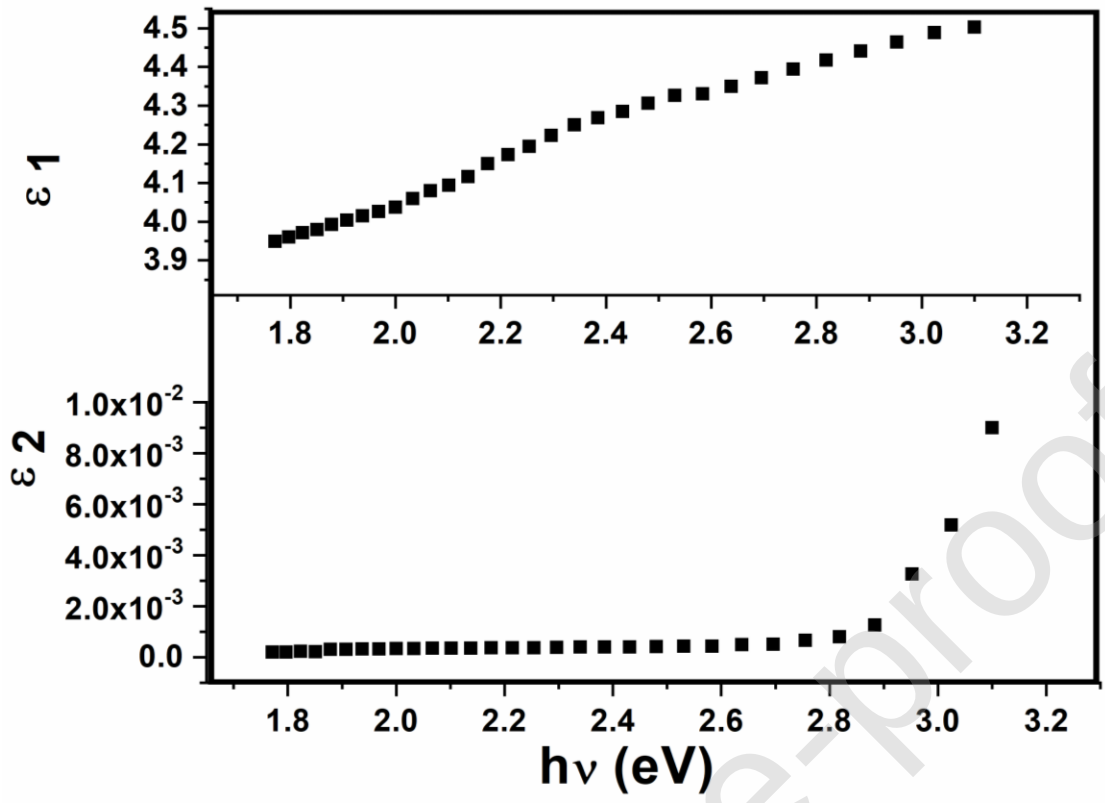


Figure 8

The real ε_1 and imaginary ε_2 parts of the complex dielectric constant

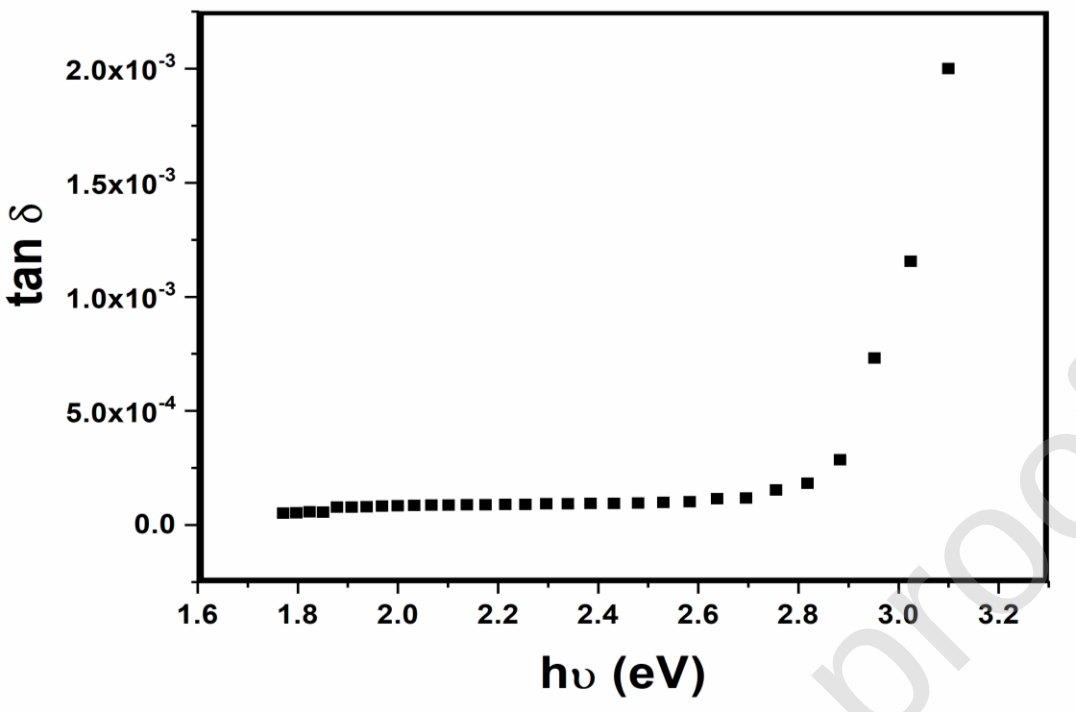


Figure 9 The photon energy dependence on the tan δ.

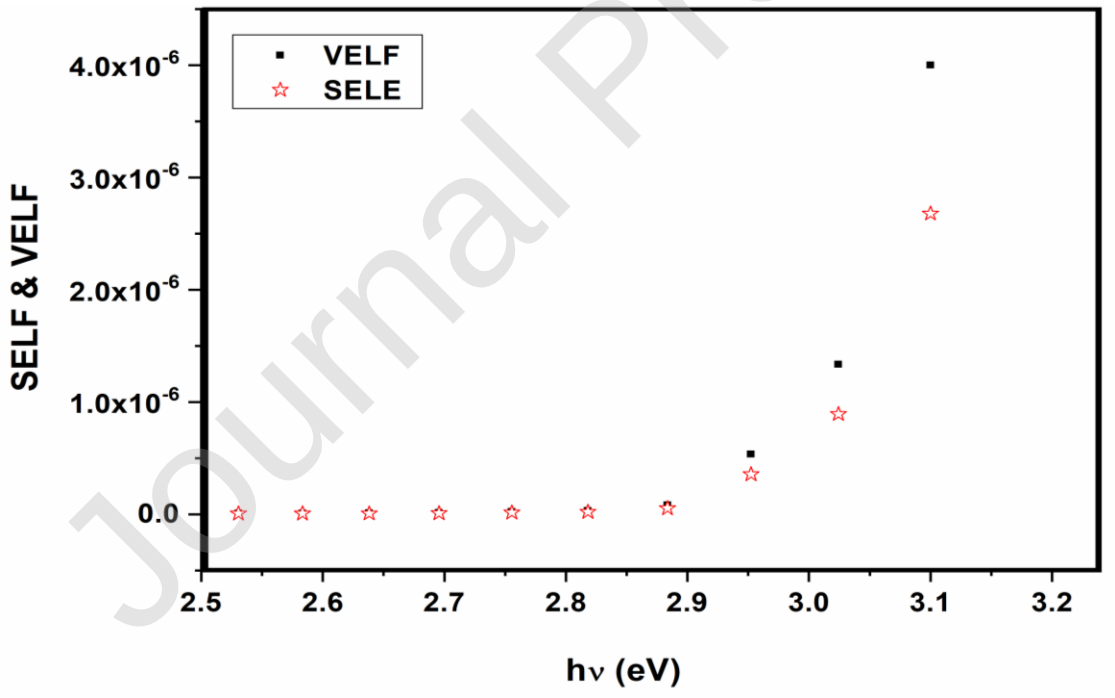
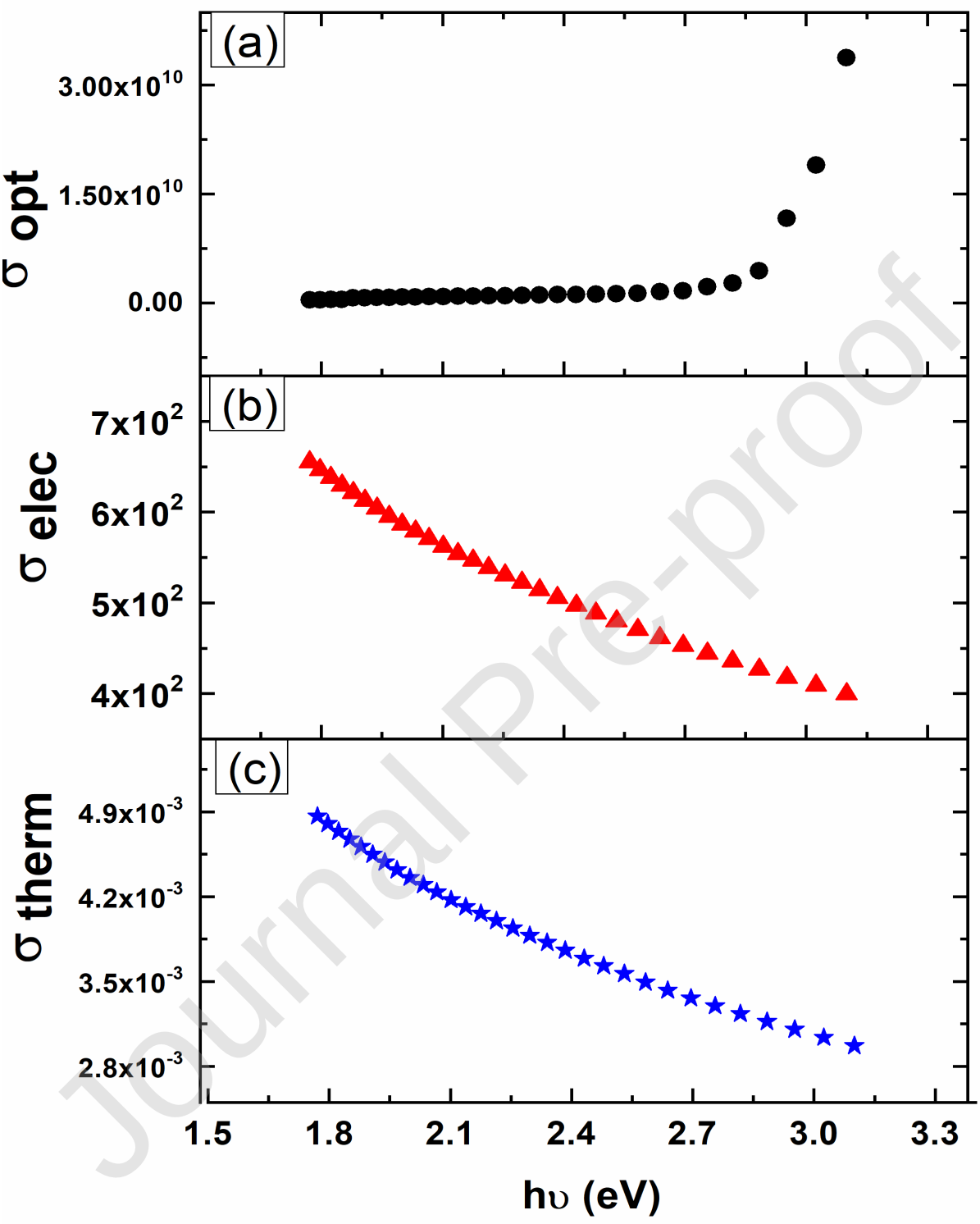


Figure 10

The spectral distributions of VELF and SELF for the film samples.



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The photon energy dependence of

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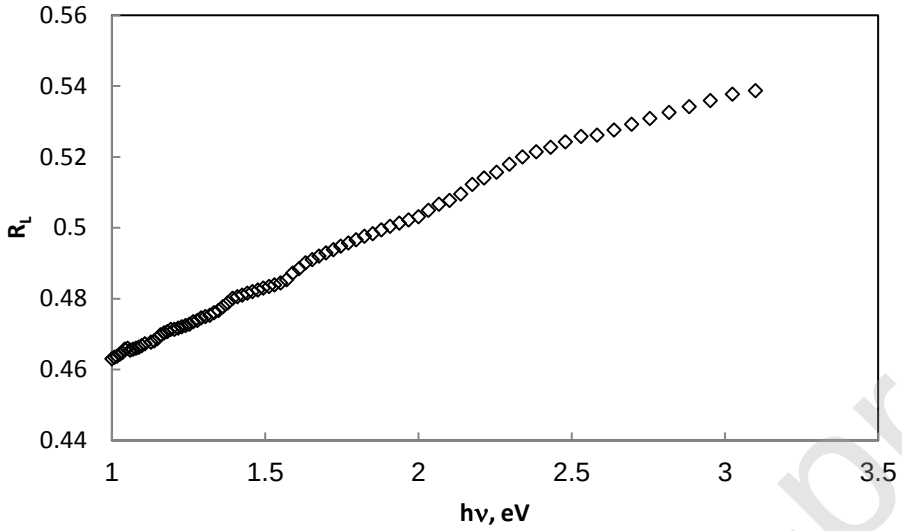

Figure 12

The plot of reflection loss factor as a function of photon energy

List of Tables:

Table 1:

The direct optical energy gap E_g, Urbach energy E_U, reflection loss R_L and cut-off wavelength $\lambda_{cut\text{-}off}$ for the studied ZnO films prepared by the ALD technique.

Parameter	Value
E_u, meV	95
$\lambda_{cut\text{-}off}$, nm	384
R_L,cm^{-3}	0.463
E_g(T), eV	3.276
E_g(T&R), eV	3.272

Table 2:

Optical energy gap E_g for the studied ZnO films and some previously prepared results of ZnO thin films prepared by different methods

ZnO In thin film form	**E_g, eV**	**Preparation method**	**Transitions type**
Our results (average value)	3.274	Atomic Layer Deposition	Direct
Ref. [38]	3.21	RF. magnetron sputtering	Direct
Ref. [39]	3.31	Spray pyrolysis	Direct
Ref. [40]	3.31-3.25	Sol-gel spin coating technique	Direct
Ref. [41]	3.24	Sol-gel spin coating technique	Direct
Ref. [42]	3.2 -3.225	Wet-processing techniques	Direct
Ref. [43]	3.31-3.17	RF. Sputtering power	Direct
Ref. [44]	3.25	Sol-gel technique	Direct
Ref. [45]	3.25	Ultrasonic spray method	Direct