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Effect of thickness on the optical properties of ZnO thin films prepared by pulsed laser deposition technique (PLD)

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Abstract
Zinc Oxide (ZnO) thin films of different thickness were prepared on ultrasonically cleaned corning glass substrate, by pulsed laser deposition technique (PLD) at room temperature, since most application of ZnO thin film are certainly related to its optical properties, so the optical properties of ZnO thin film in the wavelength range (300-1100) nm were studied, it was observed that all ZnO films have high transmittance (> 80%) in the wavelength region (400-1100)nm and it increase as the film thickness increase, using the optical transmittance to calculate optical energy gap (E_g^{opt}) show that (E_g^{opt}) of a direct allowed transition (~3.2ev) and it increase slightly as the film thickness increase, so Zn0thin films were used as a transparent conducting oxide (TCO) in various optoelectronic application such as a window in a thin film solar cells.

Keywords: ZnO, thin films, PLD, optical properties

1. Introduction
Zinc Oxide (ZnO) a (II – IV) semiconductor compound is one of the transparent conducting material (TCO), it has a direct band gap of (3.37 ev) at room temperature. that is suitable for short wavelength optoelectronic application [1–5] undoped ZnO is usually n- type semiconductor, which may be due to the presence of nature point defects such as oxygen vacancies and interstitial Zinc [6, 7], a polycrystalline ZnO thin films attract much interest because of their unique optical and electrical properties, such as high optical transparency in visible and infrared wavelength, high electron mobility, high chemical and mechanical stability in hydrogen plasma, in addition to its low cost and nontoxicity [8, 9], for those properties various applications in electronic and optoelectronic device such as a window in thin film solar cells [anti reflecting coating and transparing conducting material], transparent electrode in liquid crystal display (LCD),photodetector, short wavelength light emitting diode [LED], heat protecting window, magnetic memories, and surface acoustic wave device, in addition to the traditional application of ZnO film could be used in integrated optics and gas sensors [10 – 13]
The most frequently used ZnO thin film growth techniques are pulsed laser deposition (PLD), magnetron sputtering, metal oxide chemical vapor deposition (MOCVD), molecular beam epitaxial (MBE), spray pyrolysis, sol- gel method etc…., [2-5].
pulsed laser deposition technique (PLD) was chose for ZnO thin film deposition, since it has several advantages compared to the others, the composition of the target used and the prepared thin films are quite close, PLD film were crystalline at lower substrate temperature with respect to other physical Vapor deposition (PVD) asa result of high kinetic energies of the ejected atoms and the ionized species in the plasma, so this method was used to grow ZnO films on a flexible panel display in addition to ZnO thin film is an extremely smooth surface [13].
In designing modern optoelectronic and optical device, it is very important to know film thickness and their optical properties such as transmittance (T), absorption coefficient (α) and refractive index (n) as function of wavelength (λ) to Predict the photoelectric behavior of the device, Unfortunately there are great discrepancies among various studies on the optical Properties of ZnO thin film, Reliable determination of the optical properties of ZnOthin film is still Issue [14].

Researcher have found that the optical properties strongly depend on the thickness of ZnO film, In this work ZnO thin film of different thickness were prepared by pulsed laser deposition technique (PLD) on ultrasonically cleaned corning glass substrate at room temperature and the effect of film thickness on their optical properties were studied.

2: Experimental Details

Zinc Oxide (ZnO) thin films were prepared by pulsed laser deposition technique (PLD) on an ultrasonically cleaned corning glass substrate, Target were prepared from pure Zinc oxide powder (99,99%) using cold pressing (5 Ton on 12mm diameter pellet), the substrate. was placed on a rotating holder, at a distance of 1.5 cm above the target, the target (ZnOpellet) were ablated using a-ND – YAG (λ =1064 nm), the fluence of 5 mJ / cm ^{2} were kept constant, the ablation were carried when the substrate at room temperature and an ambient Oxygen pressure of 0.02 mbar for a period of 3 to 6min at a pulse repetition of 6HZ. all the prepared ZnO thin films were annealed to 450 c° in air for 3hr, to increase Oxygen absorption which will predically Improve stoichiometry through elimination of Oxygen vacancies [13].

Weighting method were used to estimate the amount of ZnO needed to prepare given thickness (t), according to the following formula (15)

$$t= \frac{m}{2\pi R^2 p} \ldots\ldots\ldots\ldots\ldots (1)$$

Where m : is the mass of the material to be evaporate (gm), R: is the source (target) to the substrate distance(cm), p : is the density of the material to be evaporated, (gm/cm^3) the error percentage of this method ~ 30%, while the precious thin film thickness were measured to the following formula Using [16].

$$t = \frac{\lambda}{2}\frac{\Delta x}{x} \ldots\ldots\ldots\ldots\ldots (2)$$

Where x is the fringe width,
Δχ: the fringe shift,
λ: is the laser wavelength,

3. Results and Discussion

The optical properties of ZnO thin films of different thickness (150, 180, 210, and 240) nm in the wavelength range (300-1100) nm were studied, the transmittance spectra for all ZnO thin films were measured Using Shimadzu UV1650PC spectrophotometer.

Fig. (1) show the transmittance spectra of ZnO thin film of different thickness, In general all ZnO thin films show good optical transmittance (> 80%), their high transmittance in visible range make these films an excellant Candidates for transparent window in thin film solar cells, and transparent transistor the transmittance (T), In general decrease as the energy of the Incident radiation increase (λ decrease) in (400-1100) nm till the energy corresponding to (λ- 380 nm) drastic decrease in the transmittance (absorption edge) which result due to the increase in the absorption of photons because its energy is sufficient to cause an electron transition from the valence band to the conduction band [3], which will be shown in strong absorption at the related wavelengths, while as the film thickness increase, the transmittance (T) in (400-1100) nm increase slightly, which subsequently result in a decrease in the absorption (A) this may be due to the increase in the grain size as thickness increase, which in turn cause the grain boundaries to decrease, so photon scattering decrease resulting in an increase in the transmittance [15].

The absorbance (A) of the incident radiation for ZnO thin film were calculated according to the following relation [17].

$$A = \log \frac{1}{T} \ldots\ldots\ldots\ldots\ldots (3)$$

Fig. (2) Show the absorbance spectra for ZnO thin films of different thickness, the absorbance (A) in general, decrease as the film thickness increase, this may be due the decrease in grain boundaries of the film structure [1] the absorption coefficient (α) were calculated according to the following relation [18]

$$\alpha = 2.303 \frac{A}{t} \ldots\ldots\ldots\ldots\ldots (4)$$

Fig. (3) show the absorption coefficient (α) as a function of wavelength which certainly show the same behavior of the absorbance (A)

The extinction coefficient (K) which related to the exponential decay of the electro. Magnetic wave in the medium, were calculated according to the following relation [19]

$$K = \frac{\alpha\lambda}{4\pi} \ldots\ldots\ldots\ldots\ldots (5)$$

Fig (4) show the extinction coefficient (K) as function of wavelength (λ) for different thickness, the same behavior of the related absorption coefficient (α)

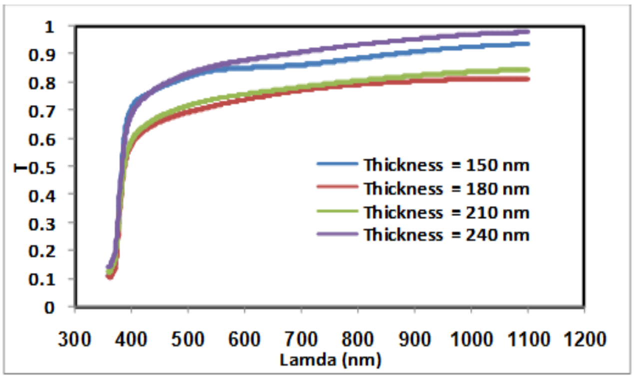

Fig 1: The transmittance spectra of for ZnO thin films of different thickness

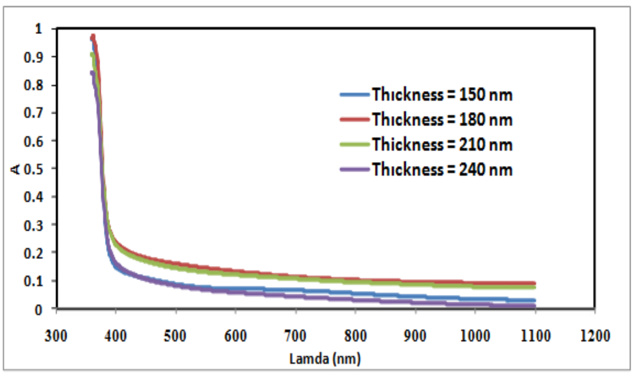

Fig 2: The absorbance spectra of for ZnO thin films of different thickness

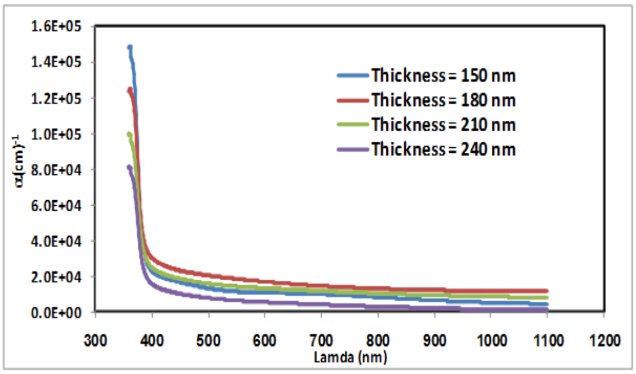

Fig 3: The absorption coefficient of for ZnO thin films of different thickness

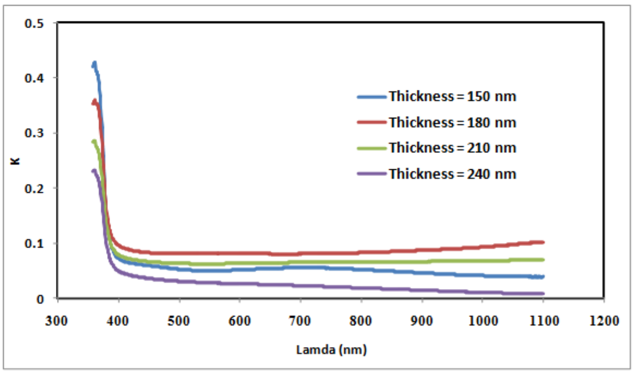

Fig 4: The extinction coefficient of for ZnO thin films of different thickness

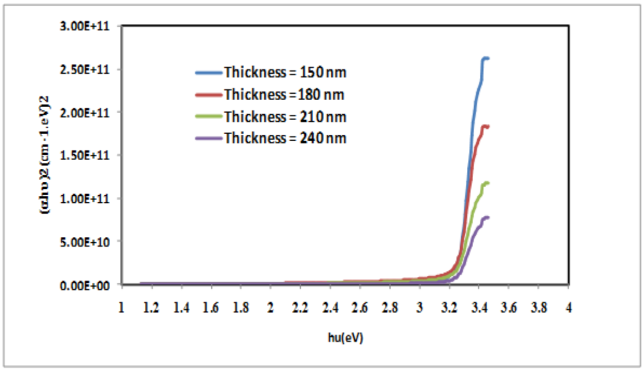

Fig 5: the variation of (αhɣ) as a function of photon energy.

The optical energy gap [E_g^{opt}] were determined using Taue equation [20]

$$\alpha h\gamma = B[h\gamma - E_g]^r \ldots\ldots\ldots\ldots\ldots\ldots..(6)$$

Where B : is Taue constant, hγ : is the photon energy, α is the absorption coefficient, for r = ½ a linear relation dependence, which describe the direct allowed transition, the optical energy gap where calculated by plotting $(\alpha h\gamma)^2$ versus (hγ) and extrapolating the straight Line portion of this plot to photon energy axis (hγ) (i.e αhγ =0).

Fig (5) show $(\alpha h\gamma)^2$ versus (hγ) for ZnO thin film of different thickness

Table 1

Film thrcknes (t) nm	Optical energy gap (E_g^{opt})ev
150	3.2
180	3.205
210	3.21
240	3.22

Table (1) show that the optical energy gap (E_g^{opt}) increase slightly as the film thickness increase, this may be due to the decrease in the localized stale density near the bands edges [21, 22], whlle the low value of the optical energy gap (E_g^{opt}) may be due to the localized stales due to defects and impurities [2].

The reflectance spectra result due to the interference between the reflected rays from the upper and lower surface of the thin film, the reflectance were calculated using the following relation [21].

$$R = \frac{(n-1)^2+k^2}{(n-1)^2+k^2} \ldots\ldots\ldots\ldots\ldots (7)$$

Fig. (6) show the reflectance spectra for Zn0 thin film of different thickness, the Reflectance(R) in general decrease as the energy of incident radial decrease (λ increase) in the wavelength range (400-1100) nm, while it decrease as thin film thickness (t) increase Refractive index (n) were calculated according to the following relation [22].

$$n = \left[\frac{4R}{(R-1)^2} - K^2\right]^{1/2} - \frac{(R+1)}{(R-1)} \ldots\ldots\ldots\ldots\ldots\ldots..(8)$$

Fig (7) show the variation Refractive index (n) for ZnO thin films of different thickness (t) which is similar to the behavior of the Reflectance (R),the refractive index(n) increase as the energy of the incident radiation increase (λ decrease), while it decrease as the film thickness increase in the wavelength range of (400- 1100) nm [23].

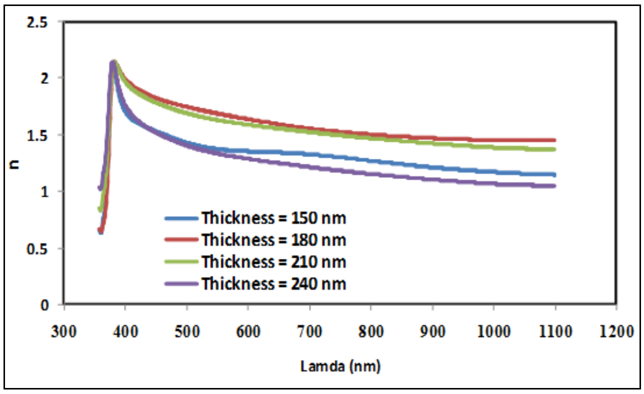

Fig 6: Reflectance spectra for ZnO thin films of different thicknesses

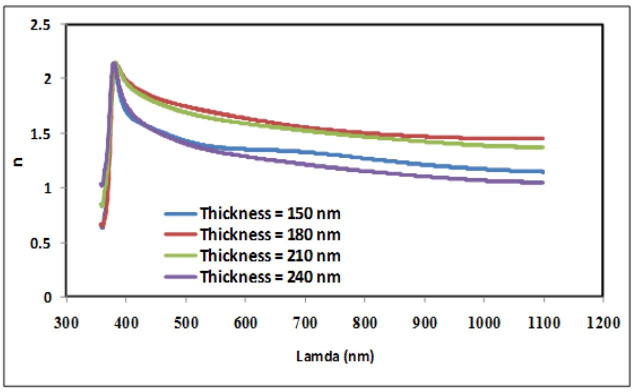

Fig 7: refractive index (n) for ZnO thin films of different thicknesses

4. Conclusion
Zine oxide (ZnO) thin film of different thickness (150, 180, 210 and 240) nm were grown on ultrasonically cleaned corning glass substrate by pulsed laser deposition (PLD) technique, all ZnO thin films show high transmittance (80%- 85% T) for thickness (180-240) nm in the wavelength range (400-800) nm. so the increase in the film thickness improve the quality of the etherthin film toward the higher transmittance, also the optical calculations show that ZnO thin film have a direct transition energy gap (Eg0pt= 3.2ev) and it increase as the thin film thickness increase, this mean that the increase in the thickness improve the film structure by lowering the defect states density in the band gap.

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