The Effect of Substrate Temperature on Optical Constants and Refractive Index Dispersion of DC Sputtered ZnO Thin Films

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Abstract

Zinc oxide (ZnO) thin films were prepared by DC- magnetron sputtering technique on glass at different substrates temperature from 350 to 550 ºC in 50 ºC step. The optical properties of the ZnO films were studied using UV-VIS-NIR spectrometer. The effects of substrate temperature on the absorption coefficient, optical band gap, refractive index, , extinction coefficient, single-oscillator energy, dispersion energy, high- frequency refractive index, high- frequency dielectric constant, moments of the optical spectra , average oscillator strength and average oscillator wavelength of the ZnO thin films were investigated . It was found that, the substrate temperature for these investigated films plays an important rule for changing an optical properties and dispersion parameter results of these films.

Keywords: ZnO, Thin films, DC sputtering, Optical constants, Dispersion parameter.

تأثير درجة حرارة القاعدة على الثوابت البصرية وتفريق معامل الانكسار بتقنية الترذيذ المغناطيسي لأغشية ZnO

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الخلاصة

تم تحضير اغشية اوكسيد الخارصين ZnO على قواعد من الزجاج وبدرجات حرارة مختلفة تتراوح من^{o}C 350 الى ^{o}C 550 وبمعدل زيادة ^{o}C 50 بواسطة تقنية الترذيذ المغناطيسي DC. درست الخصائص البصرية للغشاء باستخدام مطياف UV-VIS-NIR. ودرس تأثير درجة حرارة القاعدة على معامل الامتصاص فجوة الطاقة البصرية معامل الانكسار معامل الخمود طاقة التذبذب ألأحادي طاقة التفريق ، معامل الانكسار عند الترددات العالية، ثابت العزل عند الترددات العالية، معاملات الزخم للطيف البصري،معدل قوة التذبذب و معدل الطول الموجي للمتذبذب. لوحظ من خلال النتائج ان درجة حرارة القاعدة تلعب دورا مهما في تغير الخصائص البصرية ومعلمات التفريق للاغشية قيد الدراسة.

الكلمات المفتاحية: الاغشية الرقيقة ،ZnO ، تقنية الترذيذ المغناطيسي DC ، الثوابت ألبصرية معلمات التفريق.

Introduction

Semiconductor materials are important materials for optoelectronics device applications. The study of optical properties of semiconductors as a thin films is very important for many applications, including interference devices, such as antireflection coatings, laser mirrors and monochromatic filters, as well as optoelectronics, integrated optics, solar power engineering, microelectronics and optical sensor technology depending on the reflectance and transmittance properties of the films during their preparation. The determination optical parameters of the semiconductor materials are very important, both from a fundamental and a technological viewpoint[1-4]. Furthermore, the changes in refractive index are important for controlling optical properties of semiconductors, because optical properties are directly related to their structural and electronic properties. ZnO is a technologically one of important materials. It have been investigated because of their interesting optical, piezoelectric and electrical properties. ZnO is an II–VI semiconductor with a stable wurtzite structure at room temperature. It has a wide direct band gap (3.37 eV) at 300 K and a large exciton binding energy (60 meV)[5]. ZnO can be obtained by various techniques such as spray pyrolysis [5,6], chemical vapor deposition [7], pulsed laser deposition [8], sol-gel process [9] and sputtering [10-11] . Among these methods, sputtering has advantages such as good uniformity, high process controllability, and large-area deposition and has been widely used. Ellmer investigated the effect sputtering parameters on the properties of ZnO films [12]. Chung et al. investigated the effect of the TiO_2 content of the target, the working pressure, and the substrate temperature on the properties of TiO_2-doped ZnO films [13]. The aim of this study is to investigate the optical properties and determine optical constants of the semiconductor ZnO by optical characterization method.

Experimental

The ZnO thin films were prepared by using DC-magnetron sputtering source Edwards 306 pumping system . The sputtering conditions are: Target – anode distance (30mm), substrate type (glass), magnetic field 370G, DC power 50W, gas pressure $6x10^{-2}$Torr and at atmosphere of argon 100% . ZnO thin films were sputtered from target's on the substrate at different temperature 350 °C, 400 °C, 450 °C 500°C and 550°C. The target materials are in the form of plates with 60mm diameter and 2mm thickness made from ZnO powder (99.98% purity). Optical transmission and reflection of the prepared films were recorded over the wavelength range from 350 - 1100 nm using Shimadzu double-beam spectrophotometer.

Results and discussion

Optical transmittance and energy band gap

Optical transmittance of the films deposited at various substrate temperatures are shown in Fig.(1). The optical transmittance spectrum was recorded from 350 to 1100 nm wavelength regions. The average transmittance increases to approximately 95% as the substrate temperature is increased from 350–450 °C, due to improvement in the crystallinity of the films. However, at a higher temperature of 450 °C, the transmittance reduces to around 65% this is probably due to the structural disorders in the samples. The optical band gap of the film was evaluated from the relation[14,15]:

$$\alpha h\nu = B(h\nu - E_g)^r \quad \ldots\ldots\ldots 1$$

$$\alpha = \frac{1}{t}\ln(T) \quad \ldots\ldots\ldots 2$$

Where r is equal to 1/2 and 2 for direct and indirect transitions respectively, A is a proportionality constant, E_g is optical energy band gap and α is absorption coefficient, t is film thickness.

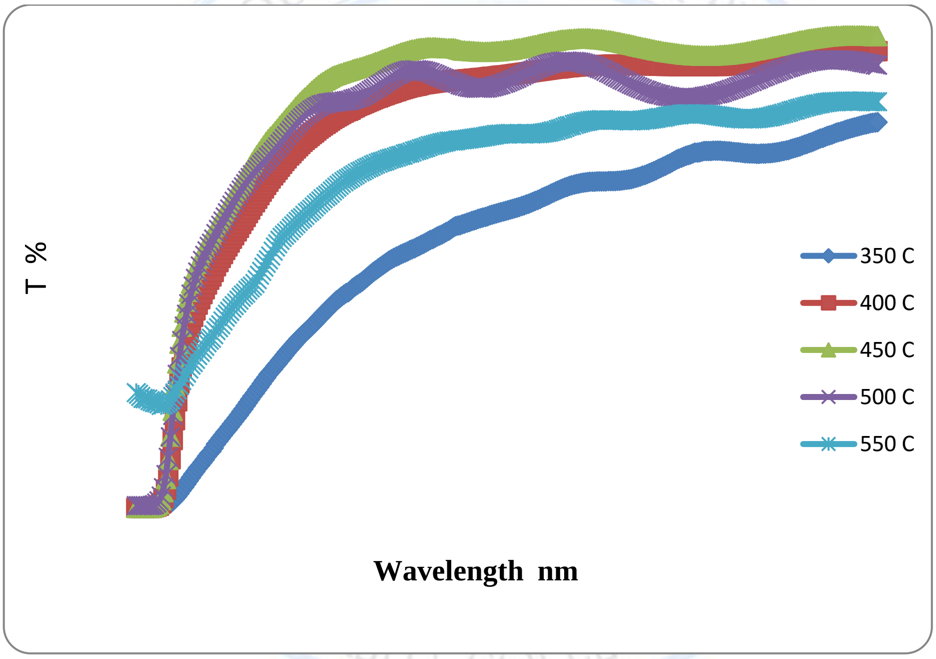


Fig (1) Transmittance Spectra of ZnO thin films

Fig. (2) show a plot of $(\alpha h\nu)^2$ vs. photon energy in the wavelength range 350–1100 nm. The linear nature of the plot indicates the existence of direct transition. The optical band gap E_g, was determined by extrapolating the straight portion to the energy axis at $(\alpha h\nu)^2 = 0$, and are given in Table (1). It is clear that the energy gap has the same behavior of transmittance due to crystallinity of the films .

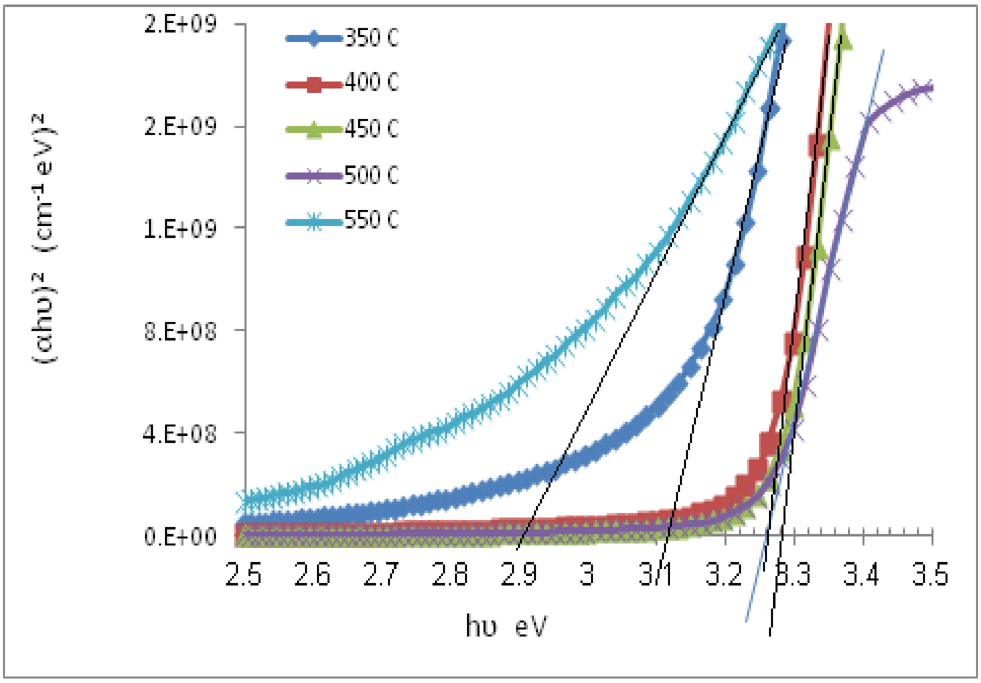


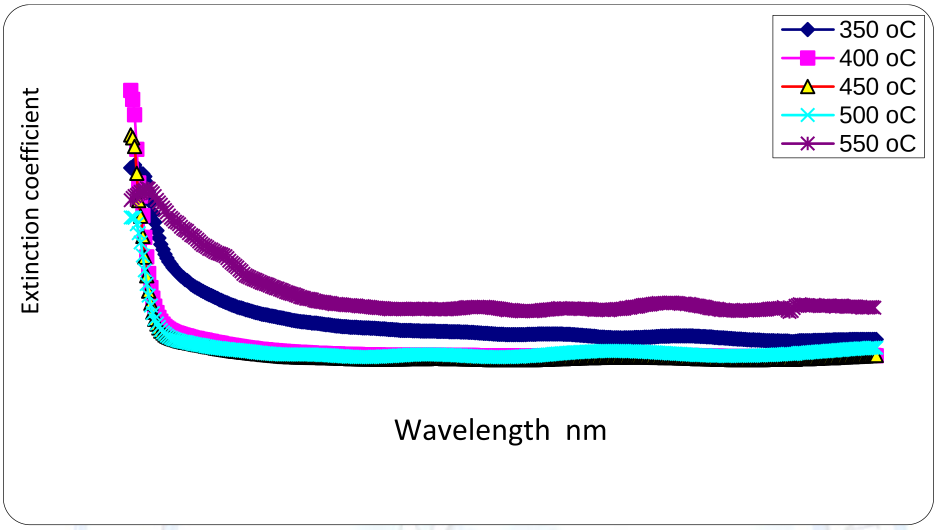
Fig (2) Plot between (αhυ)2 and hυ for different substrates temperature

Optical constants

The extinction coefficient is a optical parameter that measures the fraction of light that is lost per unit distance of the penetration medium due to both absorption and scattering and is estimated using the relation [14,15] :

$$k = \frac{\alpha\lambda}{4\pi} \qquad \text{………… 3}$$

Where λ is incident wavelength . Variation of k with wavelength λ of the incident beam for the ZnO films has been shown in fig. (3). Extinction coefficient has been observed to be decreasing with the rise in substrate temperature indicating decrease in scattering and absorption with the increase substrate temperature indicating more ordered structure but at higher substrate temperature extinction coefficient increases due to the structural disorders in the films.



Fig(3) The variation of extinction Coefficient ZnO thin films wavelength

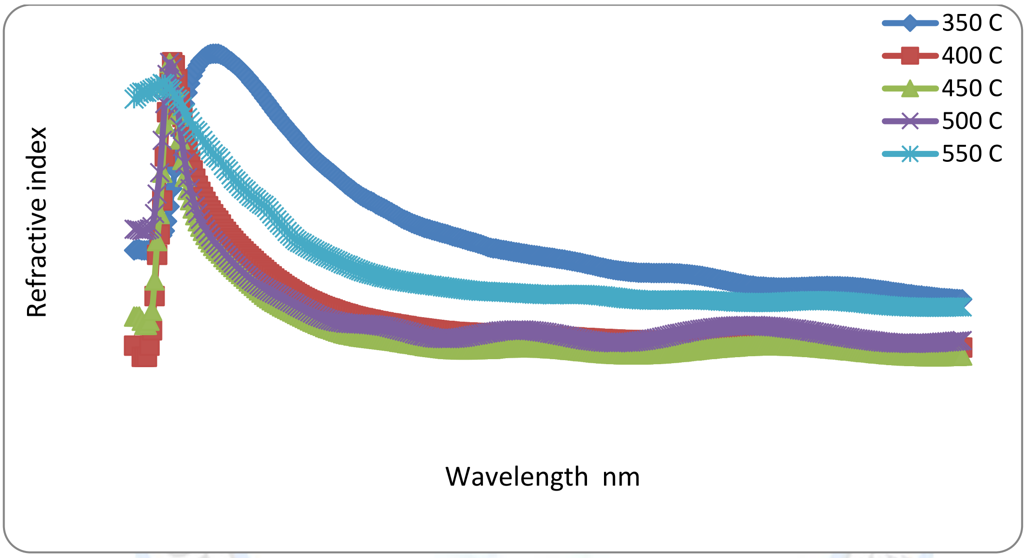
Refractive index is an important parameter for optical materials design and it includes valuable information for higher efficiency optical materials, because it is closely related to the electronic polarization of ions and the local field inside materials. Optical constants define the interaction of light traveling through a material and are expressed as complex number and can be separated in to real and imaginary components. The most common representation are complex refractive index, n* related by [14,15]:

$$n^* = n + ik \qquad \ldots\ldots\ldots\ldots 4$$

Here, the real part of the refractive index (n) can be calculated using the following relation [15,16]

$$n = \left[\left(\frac{1+R}{1-R}\right)^2 - \left(k^2+1\right)\right]^{\frac{1}{2}} + \frac{1+R}{1-R} \qquad \ldots\ldots\ldots\ldots 5$$

Where, R is the reflectance of the film. Fig. (4) show refractive indices as a function of wavelength at different substrate temperature. The decrease in refractive index with the incident wavelength has been observed indicating the normal dispersion behavior of the ZnO films .Also variation in refractive index with increasing substrate temperature can be attributed to optical absorption of the ZnO films with increase in substrate temperature.



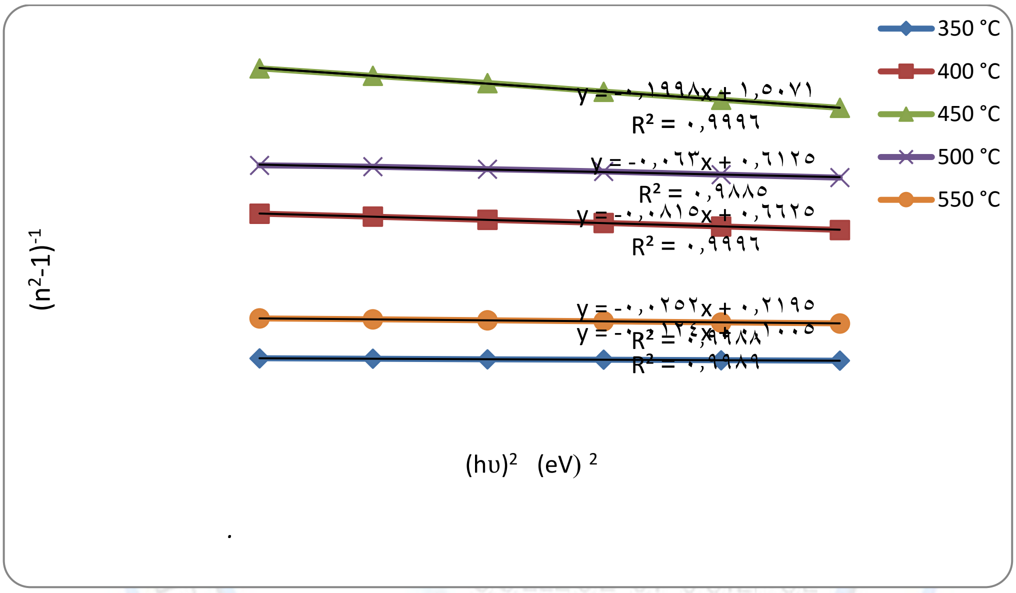
Fig(4) The variation of refractive index ZnO thin films with wavelength

According to the single-effective oscillator model proposed by Wemple and DiDomenico [17], the optical data can be described to an excellent approximation by the relation

$$n^2 - 1 = \frac{E_d E_o}{E_o^2 - h\upsilon} \qquad \ldots\ldots\ldots\ldots\ 6$$

Where $h\upsilon$ is the photon energy, n is the refractive index, E_0 is the single-effective oscillator energy and E_d is the dispersion energy, which is a measure of the average strength of the interband optical transitions. Plotting $(n^2 - 1)^{-1}$ against $h\upsilon^2$ for the films deposited at different substrate temperatures. Extrapolation of the lower energy part of the spectral range where the films are transparent displays a linear trend as shown in Fig.(5). The values of E_o and E_d can then be calculated from the slope $(E_oE_d)^{-1}$ and the intercept on the vertical axis (E_o/E_d). The values of the static refractive index (n_o) can be calculated by extrapolating the Wemple-DiDomenico dispersion equation (6) to $h\upsilon \rightarrow 0$[18].

$$n_o = 1+E_d/E_o \qquad \text{...... ....7}$$



Fig(5) Plots of $(n^2-1)^{-1}$ vs. $(h\upsilon)^2$ of ZnO thin films

Then the value of static dielectric constant and M-1 and M-3 moments of the optical spectrum which were expresses as using the relations:

$$\varepsilon_s = (n_o)^2 \qquad \text{..... .... 8}$$

$$E_o^2 = \frac{M_{-1}}{M_{-3}} \qquad \text{............... 9}$$

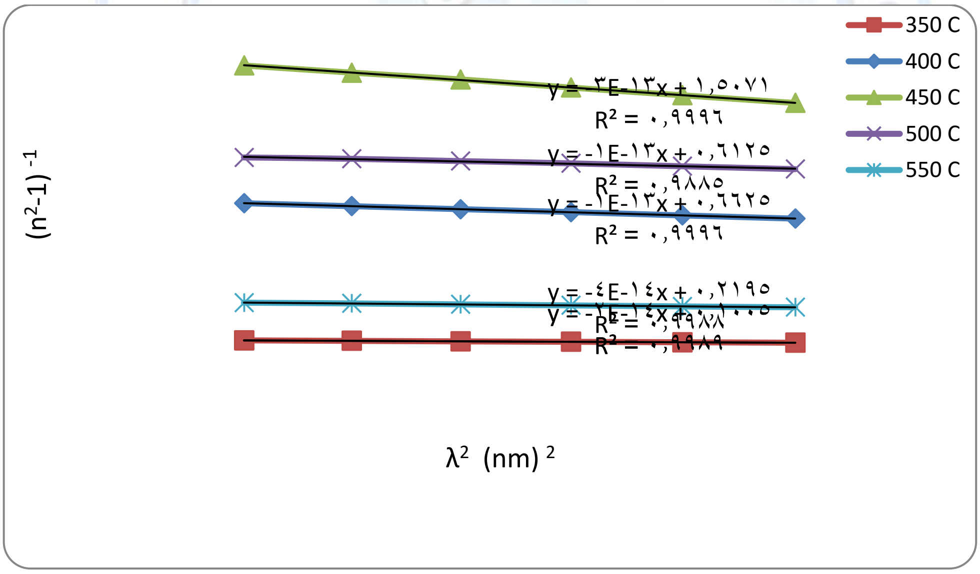
$$E_d^2 = \frac{M_{-1}^3}{M_{-3}} \qquad \text{............. 10}$$

The values of E_d, E_o, n_o and ε_s are listed in Table (1). The values of E_d, M_{-1}, M_{-3}, n_o, ε_s, M_{-1} and M_{-3} were found to be decreased with increasing the substrate temperature up to to 450°C and increase at a higher temperature of 450 °C, while optical energy gap and dispersion energy E_o has opposite behavior.

Another model used in the refractive index dispersion study is the single Sellmeier oscillator model at low energies can be deduced using the determined (n) values from relation [19-21]:

$$n^2 - 1 = \frac{S_o \lambda_0^2}{1-(\frac{\lambda_o}{\lambda})^2} \qquad \text{........... 11}$$

Where S_o oscillator strength and λ_o oscillator wavelength. The values of S_o and λ_o calculated from $(n^2-1)^{-1}$ versus λ^{-2} plot as shown in fig(6). The S_o^{-1} can be obtained from the slope while $(S_o\lambda_o)^{-1}$ could be obtained from the intercept of y-axis , and listed in table (1). The obtained values of oscillator strength and oscillator wavelength are of the same order as those obtained for ZnO and doped ZnO thin films[19-21].



Fig(6) Plots of $(n^2-1)^{-1}$ vs. λ^2 of ZnO thin films

Table(1) Optical and dispersion parameters of ZnO thin films at different substrate temperature

T °C	E_g eV	E_d eV	E_o eV	M_{-1}	M_{-3} $(eV)^2$	n(o)	$\varepsilon_\infty=n^2(o)$	S_o m^{-2}	λ_o m
350	3.12	17.2	4.37	3.92	0.21	2.22	4.92	2.5E+13	4.0E-07
400	3.26	15.6	4.46	3.49	0.17	2.12	4.49	1.0E+13	4.4E-07
450	3.28	13.6	4.60	2.96	0.14	1.99	3.96	3.3E+12	4.5E-07
500	3.26	16.2	4.48	3.62	0.18	2.15	4.62	1.0E+13	4.3E-07
550	2.90	20.6	4.12	4.99	0.29	2.45	5.99	5.0E+13	4.3E-07

Conclusion

Zinc oxide thin film was prepared by DC- magnetron sputtering technique on glass at various substrate temperatures. Transmittance spectra were used to determine the optical constants and refractive index dispersion of the films. The optical transmittance increases, consequently the optical band gap energy of ZnO film increases from 3.12 eV to 3.28 eV with increase in substrate temperature from 350 ^oC to 450 ^oC then the transmittance and optical band gap energy decrease as temperature increase. The refractive index dispersion data were analyzed using the Wemple- DiDomenico single –effective oscillator model. The oscillator energy, dispersion energy, oscillator strength , zero- frequency refractive index, zero- frequency dielectric constant, moments of the optical spectra , average oscillator strength and average oscillator wavelength were determined. In conclusion, we can state that the influence of the substrate temperatures on the optical properties of ZnO thin films is noticeable.

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