Effect of Beta Particles on Optical Properties of Zinc Oxide Thin Films

Ramla D. Al-Alawy
Physics Department, Education College
Al-Mustansiriyah University

Abstract:

Zinc oxide ZnO thin films were deposited by DC magnetron sputtering technique on preheated glass substrates at 500°C , and subsequently the effect of β- particles produced from ^{90}Sr source on the optical properties of the films was investigated. The optical properties of the films have been characterized using spectrophotometric measurements of transmittance and absorbance at the normal incidence of light in the wavelengths range over 380-900 nm. The absorption coefficient, optical energy gap, Urbach energy, optical constants (refractive index, extinction coefficient, dielectric constant) and dispersion parameters, such as single-oscillator energy, dispersive energy for un-irradiated and the irradiated ZnO thin films were determined and analyzed.

Keywords. Beta particles ; optical properties; ZnO thin films; DC magnetron sputtering.

Introduction

ZnO thin film is one of the II-VI compound semiconductors and is composed of hexagonal wurtzite crystal structure[1,2]. ZnO thin film presents investigating optical, acoustical and electrical properties which meet extent applications in the fields of electronics, optoelectronics and sensors[3-8]. ZnO thin film is applied to the transparent conductive film and the solar cell window because of the high optical transmittance in the visible region. The ZnO thin film is prepared using various methods such as spray pyrolysis, sputtering, sol-gel spin coating, chemical vapor deposition (CVD) [9-12]. The stability of these films, however, at high temperature and high radiation environment is still a matter of research[13-15]. Physical properties of ZnO are strongly influenced by the growth parameters and the post deposition treatments [16]. Irradiation of thin films with high energy radiations like γ-rays, X-ray, β- particles etc. is expected to affect their physical properties. The study of irradiated samples enhances the efficiency improvement in its applicability in a radiation environment and is also important in obtaining basic information on vacancies, defects and their interaction with impurities [17,18].

The purpose of this study is to prepare ZnO thin films using dc magnetron sputtering technique and to investigate the influence of β-irradiation on their optical properties.

Experimental

The ZnO thin films were deposited on glass substrates using the DC magnetron sputtering system. The sputtering system was pumped down to $3x10^{-6}$ Torr using a turbo molecular pump at atmosphere of argon 100% and magnetic field 570 gauss. ZnO films were sputtered from target on glass substrate at temperature 500 ^{o}C , the target materials are in the form of plates with 60mm diameter and 2mm thickness made from ZnO powder (99.98% purity). Films having thickness of 0.45 μm were chosen for the absorption spectra studies. ZnO thin films were exposed to β- particles produced from ^{90}Sr source with active radiation (0.011μci) and β emission energy is(0.54-0.55MeV) for 21 day at room temperature.

The Optical properties of films have been characterized using spectrophotometric measurements of transmittance and absorbance at the normal incidence of light in the wavelengths range over 380–900 nm. The optical absorption measurements were carried before and after irradiation of the sample by β - particles.

Results and discussion

Fig(1a,b)shows the spectral distribution of absorbance(A) and transmittance(T) for un-irradiated and the irradiated ZnO film. In this spectral region , transmittance of irradiated ZnO film is higher than that for un-irradiated one, this is possibly due to the increase in grain size and the decrease in the number of defects [19], but the difference is small may be due to low dose of β- particles. Also, it is observed that the transmittance (for both curve)decreases at the spectral region of fundamental absorption, where the incoming photons have sufficient energy to excite electrons from the valance band to the conduction band and thus these photons are absorbed within the material and cause the decreasing of the transmittance[20].

Fig(2) shows the absorption coefficient (α) of un-irradiated and irradiated films vs. photon energy *(hν)*, from this figure α(un-irradiated ZnO)> α (irradiated ZnO) ,this is may be due to increase the defect state which leads to increase a absorption coefficient. The absorption coefficient α is related to the energy band gap *Eg* and the photon energy *hν*:

$$\alpha = \alpha_o(h\nu - Eg)^r \qquad (1)$$

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

Where α_o is constant and *r* is 1/2, 3/2, 2 or 3 for direct allowed, direct forbidden, indirect allowed and indirect forbidden transitions, respectively and t is film thickness.

A satisfactory linear fit is obtained for $(\alpha\, h\nu)^2$ vs. *hν*, indicating the presence of direct allowed transition for ZnO thin films. The intercept on the energy axis, as shown in Fig(3), gives the band gap *Eg* of ZnO thin films. It was found to be 3.19 eV for un-irradiated ZnO films and shows 'blue shifts' to 3.3 eV after irradiation by β-irradiation. This increase in the optical bandgap is basically due to the decrease in the width of the band tails of localized states.

The width of localized states in the low energy range (near the band edge) shows an exponential dependence on the photon energy and obeys the Urbach's empirical formula (Urbach 1953) [21]:

$$\alpha(\nu) = \alpha_o(\nu)\exp\left(\frac{h\nu}{E_u}\right) \qquad (3)$$

Where α_μ is a constant and E_U is an empirical parameter or Urbach energy which corresponds to the width of the band tail and could be determined as the width of the localized states. Usually, E_U depends on temperature and describes the width of the localized states in the bandgap and could be function of the structural disorder [21,22]. The values of E_U were calculated from the slopes of the straight lines of $ln(\alpha)$ vs. $h\nu$ plots as in Fig(4). Clearly, the width of localized states decrease from 0·7 to 0·5 eV for irradiated ZnO thin films, this is due to that the defects are annihilated within the thin film during beta irradiation. The variation of the optical energy gap and width of localized with beta irradiation energies can be explained as the change in the degree of disorder [16].

Extinction coefficient (k) represent the imaginary part of complex refractive index and it can be defined as the amount of energy losing as a result of interaction between the light and the charge of medium, it can be calculated by the following equation[23]:

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

The relation between (k) and wavelength (λ) is illustrated in Fig(5),it is clear that the (k) value for irradiated is smaller than un-irradiated films, because it has smaller absorption coefficient and may be due to decrease the structure defects with irradiation. The refraction index (n), real part (ε_r) and imaginary part (ε_i) of dielectric constants were calculated from the following equations [23]:

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

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

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

Where *R is* the reflectance. It can be observed from Fig (6) to Fig (8), the values of n, ε_r and ε_i dependence on the wavelength, and irradiated films values is smaller than that un-irradiated films this is due to the change in crystallite size.

The refractive index dispersion of the thin film is expressed as Wemple and Di Domenico [24]:

$$n^2 - 1 = \frac{E_d E_o}{E_o^2 - h\nu} \qquad (8)$$

Where E_o is the average electronic energy gap for transition (average excitation energy) known as the oscillator energy, and E_d is the dispersion energy (average strength of iterband optical transition) called the oscillator strength. By plotting $(n^2-1)^{-1}$ vs. $(h\nu)^2$ as in Fig. (9) and fitting a straight line, the values of the parameters E_o and E_d were calculated from (E_o/ E_d) represents the intercept on the vertical axis and $(E_o\ E_d)^{-1}$ is the slope of the plot and listed in table(1). We found that average energy gap E_o =1.5Eg, E_o=1.3Eg for un-irradiated and irradiated ZnO films respectively. This is good agreement with the result of other worker, where they found the relation between E_o and Eg is E_o = 1.4Eg [24] and E_o = 1.2 Eg [25].

Conclusions

According to optical characterizations, un-irradiated and irradiated ZnO films exhibit a high transparency in the visible range . From the transmission spectra curves

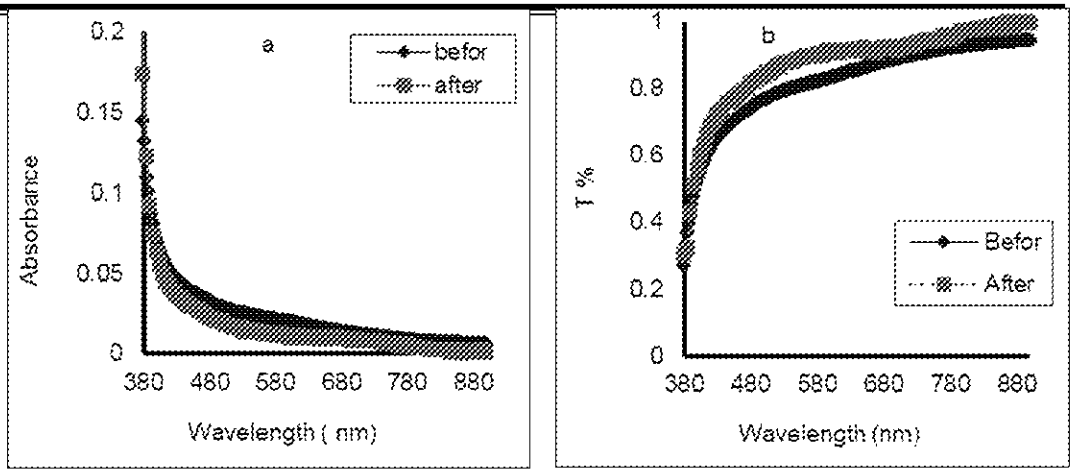
corresponding to the un-irradiated and irradiated samples, one can concluded that in the irradiated samples, an increase of the optical transmittance intensity (T > 90%) in the visible region by ~10% is obtained as compared with un-irradiated ZnO thin films. The optical transmission spectra in the transparent region were improved and the absorption edge shifted toward a shorter wavelength side by β- radiation, attributed to the decrease in structural disorder of irradiated ZnO thin film.

Table (1) Optical parameters of un-irradiated and irradiated ZnO thin film.

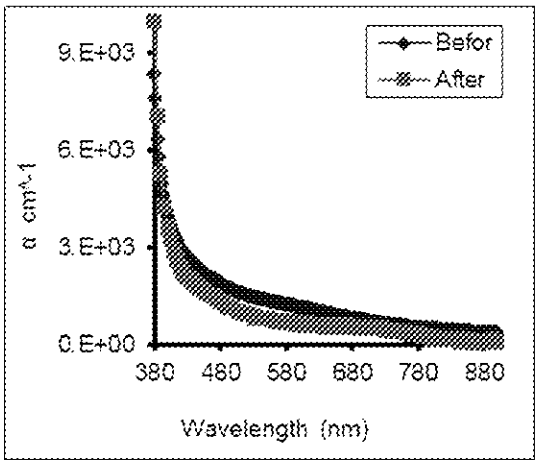
sample	E_o eV	E_d eV	Eg eV	E_o/Eg
un-irradiated	3.36	9.34	3.19	1.50
irradiated	4.36	5.31	3.30	1.30

References

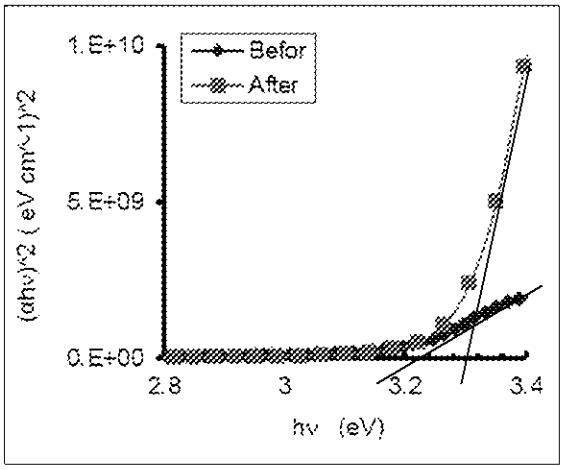
[1] H.E. Brown, Zinc Oxide Rediscovered, The New Jersey Zinc Company, New York,(1957).
[2] H.E. Brown, Zinc Oxide Properties and Applications, Pergamon, New York, (1976).
[3] T. Pauporte´ , D. Lincot, B. Viana, F. Pelle´ , Appl. Phys. Lett., 89 (2006) 233112.
[4] G. Chai, O. Lupan, L. Chow, H. Heinrich, Sens. Actuators A: Phys., 150 (2009) 184.
[5] O. Lupan, S. Shishianu, L. Chow, T. Shishianu, Thin Solid Films, 516 (2008) 3338.
[6] O. Lupan, G. Chai, L. Chow, Microelectron. Eng., 85 (2008) 2220;
[7] O. Lupan, L. Chow, G. Chai, Sens. Actuators B: Chem., 141 (2009) 511.
[18] O Lupan, S. Shishiyanu, V. Ursaki, H. Khallaf, L. Chow, T. Shishiyanu, V. Sontea , E.Monaico, S. Railean, Solar Energy Mater. Solar Cell, 93 (2009) 1417.
[9] F. Yakuphanoglu, S. Ilican, Y. Caglar, M. Caglar, J. Opto. Electro. And Advanced Mater., 9 (7) (2007) 2180.
[10] S.K. Hazaa, A. Budianto, Sudjatmoku, Atom Indonesia, 28 (2) (2002) 75.
[11] S. Ilican, Y. Caglar, M. Caglar, J. Opto. Electro. And Advanced Mater., 10 (10) (2008) 2578.
[12] T. Saeed, P. O'Brie, Thin Films Solid, 35 (1995) 271.
[13] Y. Jin, J. Wang, B. Sun, J.C. Blakesley and N.C. Greenham, Nano Lett., 8 (2008) 1649.
[14] Y.J. Lee, D.S. Ruby, D.W. Peters, B.B. Mckenzie and J.W.P. Hsu, Nano Lett., 8 (2008) 1501.
[15] J.J. Schneider, R.C. Hoffmann, J. Engstler, O. Soffke, W.Jaegermann, A. Issanin and A. Klyszcz, Advan. Mater., 20 (2008) 3383.
[16] S. Ambily, C.S.Menon, Thin Sold Films, 347(1999) 284.
[17] K.Arshak, O. Korostynska, F. Fahim, Sensors, 3 (2003) 176.
[18] A. A. EL-Fadl, E. M. EL-Maghraby, G. A.Mohamad, Cryst. Res. Technol., 39 (2004) 143.
[19] A. I. Khudiar, Siddhartha, M. Zulfequar, Z. H. Khan , Chalcogenide Letters,7(5) April (2010) 291.
[20] A.N.Banerjeel, C.K.Ghosh, S.Das, K.K.Chattopadhyay, Physica, B370 (2005) 264.
[21] F. Urbach, Phys. Rev., 92 (1953)1324.
[22] V. Strikant, D.R. Clarke, J. Appl. Phys., 81 (1997) 6357.
[23] N.A. Hassan, J. College of Basic Education, 16 (63) (2010) 49.
[24] S.H. Wemple, M. DiDomenico, Phys. Rev., B, 3 (1971) 1338.
[25] S. Ilican, Y. Caglar, M. Caglar, Physica Macedonica ,56 (2006) 43.



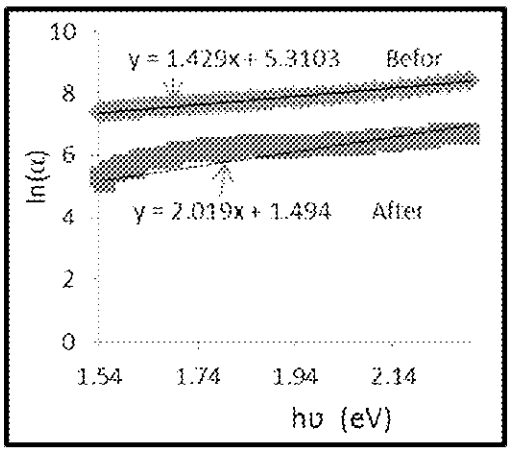
Fig(1) Spectral distribution of a- absorbance b- transmittance with wavelength for un-irradiated and the irradiated ZnO film.



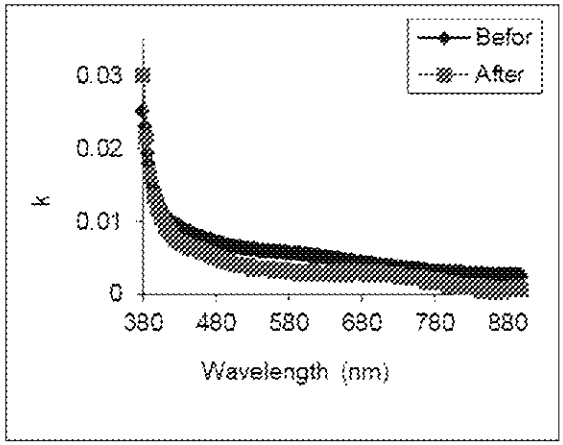
Fig(2) The variation of absorption coefficient vs. wavelength of un-irradiated and irradiated films.



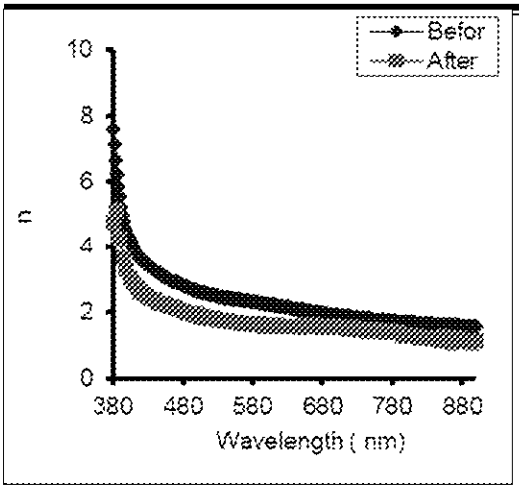
Fig(3) The variation of $(\alpha h\nu)^2$ vs. $h\nu$ of un-irradiated and irradiated films.



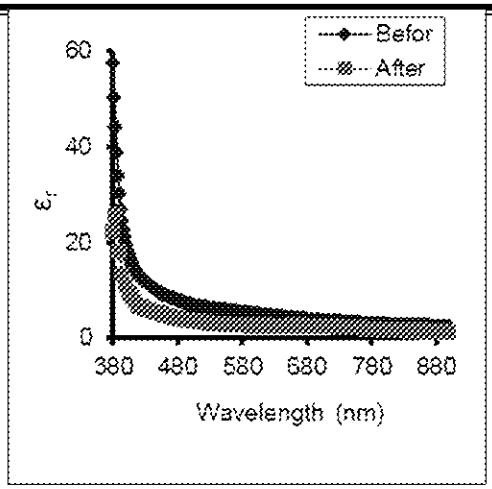
Fig(4) The variation of $ln(\alpha)$ vs. $h\nu$ of un-irradiated film.



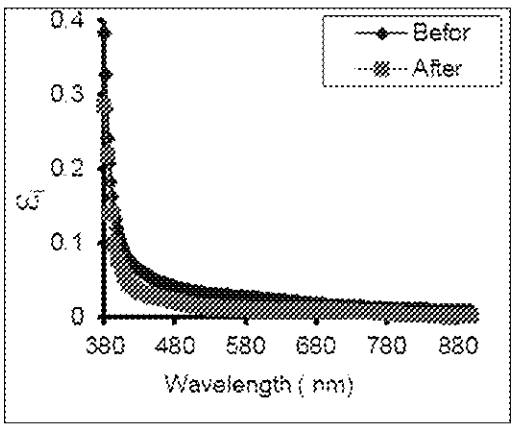
Fig(5) The variation of extinction coefficient **vs.**. wavelength of un-irradiated and irradiated films



Fig(6) The variation of refraction index vs. wavelength for un- irradiated and the irradiated film.



Fig(7) The variation of real part of dielectric constant vs. wavelength for un-irradiated and the irradiated film.



Fig(8) The variation of imaginary part of dielectric constants vs. wavelength for un-irradiated and the irradiated film

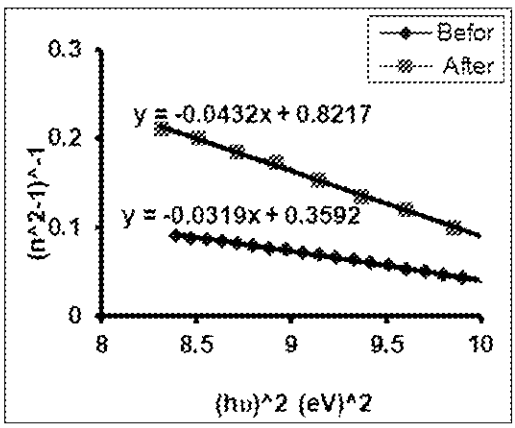


Fig (9) The variation of $1/(n^2 -1)$ with $(h\upsilon)^2$ of un-irradiated and the irradiated film

الخلاصة

حضرت اغشية ZnO الرقيقة المرسبة على قواعد زجاجية عند درجة حرارة $500^{\circ}C$ باستخدام تقنية الترذيذ المغناطيسي (DC- sputtering),من ثم دراسة تأثير التشعيع بجسيمات بيتا (β- irradiation) الناتجة من مصدرسترونتيوم (^{90}Sr strontium)على الخصائص البصرية للأغشية. تم دراسة الخصائص البصرية من طيفي الامتصاصية والنفاذية باستخدام المطياف الضوئي ضمن الطول الموجي (380-900 nm) للأغشية قبل وبعد التشعيع. حيث تم حساب معامل الامتصاص ,فجوة الطاقة البصرية ,طاقة اورباغ وبعض الثوابت البصرية (معامل الانكسار,معامل الخمود, ثابت العزل) اضافة الى معلمات التفريق المتمثلة بمتذبذب الطاقة الاحادي وطاقة التشتت للأغشية قبل وبعد التشعيع وتحليل النتائج.