Influence of gamma radiation on optical constants and optical energy gap of ZnO thin fi<u>lms</u>



Abstract

We have studied the effect of gamma irradiation on the absorption spectra and the optical energy gap of ZnO thin films. The optical transmission (T%) in the wavelength range 300-900 nm of film deposited at 200 °C on glass substrate was measured. The dependence of the absorption coefficient (α) on photon energy was determined as a function of gamma irradiation. The films show direct allowed interband transition that influenced by the gamma irradiation. Optical constants like refractive index, extinction coefficient and dielectric constants (ϵ_r),(ϵ_i) increased after irradiation.

The results can be discussed on the basis of γ -irradiation-induced defects in the film.

Introduction:-

High-energy radiations, such as γ -rays, change the physical properties of the materials they penetrate. The changes are strongly dependent on the internal structure of the absorbed substances. Studies on the changes in optical properties of thin films irradiated with ionizing radiations yield valuable informations regarding the electronic processes in these materials. Ionization occurs and charged species, both ionic and free radical, are formed. It is believed that ionizing radiation causes structural defects (called color centers or oxygen vacancies in oxides) leading to their density change on the exposure to γ -rays.^[1] The study of radiation-induced defects is not only important in observing the changes in the physical properties degradation or efficiency improvement in its applicability in a radiation environment, but it is also critical in getting basic information on vacancies, interstitials and their interaction with impurities.

In the present contribution we measure the transmission, reflectance of ZnO films under the influence of gamma ray and determine the optical band gap (E_g) and optical constants(Extinction Coefficient (K_0), Absorption Coefficient(α), Refractive index(n) and dielectric constants (ϵ_r),(ϵ_i)

Expremantal Part:-

ZnO films were prepared by dc magnetron sputtering. Argon gas 99.9998 % is used as sputter gas in pressure $6*10^{-2}$ Torr. Substrate temperature equals to 200° C, and deposition time 1.5 hours. Target-anode distance (30 mm) .The film was deposited on (glass) with thickness (0.85µm). These films were exposed to gamma rays results from Co⁶⁰ with activity 38 µci, during 21 days. The optical transmittance of the unirradiated ZnO and Irradiated ZnO thin films was determined by the spectrophotometer within the wavelength range of 200-900nm.

Results and discussion:

Figure (1) shows the variation of Transmittance [%] with Wavelength for Unirradiated ZnO & Irradiated one. The behaviors of the curves in this figure can discuss easier if it is divided to two parts, before (λ =390 nm) and after it. In the first region < λ =390 nm, Transmittance for Irradiated ZnO is higher than that for Unirradiated one, but it decreases rabidly with increasing wavelength , in this region , the main radiation effect traps charges into the film which induce transmittance degradation {also called the Radiation-Induced-Attenuation (RIA)}^[2]. In the second region, Transmittance for Unirradiated film is higher than that for Irradiated one, this might be attributed to the one or all of the following reasons:-



Figure(1) Transmittance[%] as a function of Wavelength.

- 1- A decrease in light scattering losses ^[3]
- 2- The increased roughness of the irradiated thin films contributed to the drastic decrease of optical transmittance ^[4].
- 3- The increased scattering of photons by crystal defects, and the free carrier absorption of photons contributed to the reduction in optical transmittance.^[5]

It is observed that the transmittance (for both curves) decreases at the spectral region of fundamental absorption. In this region the incoming photons have sufficient energy to excite electrons from the valence band to the conduction band and thus these photons are absorbed within the material to decrease the transmittance.^[6]

The irradiated thin film shows a much softer absorption edge, possibly indicating the presence of sub-band gap levels associated with defects.

Figure (2) shows the absorption coefficient (α) of unirradiated & irradiated ZnO films vs.



Figure (2) Absorption coefficient as a function of Photon energy.

photon energy, from this figure α (irradiated ZnO) > α (unirradiated ZnO), this is attributed to increase the defect states which leads to increase absorption coefficient.

Absorption of photons creates electron-hole pairs. In turn, the field of such pairs may modify the electronic structure and hence optical properties of ZnO film.^[7]

Generally the equation (written here in a simplified form) used to determine the band gap nature and the value of E_g (the gap energy) is ^[8] $\alpha = (c/hv) (hv-E_g)^n$

Where c is a constant nearly independent on photon energy and known as the disorder parameter, hv is the photon energy. The value of the optical energy gap E_g is obtained by plotting $(\alpha hv)^{1/n}$ in the high

absorption range followed by extrapolating the linear region of the plot $(\alpha hv)^{1/n} = 0$. This extrapolated value is used to define the so called optical gap. The plots of $(\alpha hv)^{1/n}$ against hv (n=1/2, 3/2, 2 and 3) give linear relation. For ZnO films the most fit for above equation gave the value of n=1/2. This indicates that allowed direct transitions are responsible for absorption in ZnO films. For direct transition, the absorption coefficient α is given by the following relation obtained by Mott and Davis ^[9]:

 $(\alpha hv)^2 = B (hv - E_g)$

Where B is a constant independent of the photon energy. Figure (3) shows the variation of $(\alpha hv)^2$ with hv for unirradiated & irradiated ZnO films. The optical energy gap, $\mathbf{E}_{\mathbf{g}}$, for direct allowed transitions, can be obtained by extrapolating the linear portions of the curves to α hv = 0.



Figure(3) Band gap $E_{\rm g}$ estimation for Unirradiated & irradiated Zn0 films.

Figure(3) shows the evolution of permanent changes with γ irradiation and indicates that the absorption edge for the allowed direct transition shifts to low photon energies when the sample irradiated by γ ray [E_g(Unirradiated ZnO) = 3.0545eV., E_g(Irradiated ZnO)=2.787eV]. Gamma doses cause the breaking of bonds, leading in turn to the increase of dangling bonds and of defects, as well as the trapping of the generated carriers. This may be the cause for the increase in band tail width, and then decrease energy gap.

Figure (4) shows the variation of refractive index with photon energy. Figure (5) shows the extinction coefficient (k_0) as a function of photon energy, the behavior of (k_0) is corresponding to that for (α). (K_0) for Unirradiated ZnO is smaller than that for irradiated one,

because it has smaller absorption coefficient and due to increase the structure defects with gamma ray.



Figure(4) Refractive Index vs. coefficient vs. Photon energy.

Figure(5) Extinction Photon energy.

The dielectric constants consists of real part (ε_r) & imaginary part (ε_i) , the variations of them with photon energy were determined and shown in figure (6)&(7).



Figure(6) ε_r vs.Photon energy. Figure(7) ε_i vs.Photon energy. We can observe that the variation of ε_r has similar trend to the variation of n_0 because of smaller values of $(k_0)^2$ in comparison with $(n_0)^2$ where

 $\epsilon_r = (n_0)^2 - (k_0)^2$, while the variation of ϵ_i mainly depends on the variation of k_0 values which are related to the variation of α .

For most cases of optical absorption, the energy absorbed is proportional to the thickness of the specimen. The variation of optical

energy inside the absorptive medium is given by the following relationship:

 $I(x) = I(0). \exp(-\alpha \cdot x)$

and α is related to the optical constants by ^[10]:

 $\alpha = 4 \pi k_0 / \lambda$

Here we note that α (measured in cm⁻¹) describes the attenuation of the radiation intensity rather than that of the electric field .In spectral regions of intense absorption, all the energy that enters the medium is absorbed. The only part of the incident energy that remains is that which is reflected at the surface. In such a case, it is useful to define a characteristic "skin" thickness that is subject to an appreciable density of optical energy. A convenient form used widely is simply the inverse of α , i.e. 1 / α . This skin depth is usually denoted by χ : $\chi = 1 / \alpha$

Figure (8) shows skin depth as a function of wavelength.



Figure (8) Skin Depth vs. Wavelength.

It is clear from fig (8), the action of irradiation on the value of skin depth. In spectral region (380-900 nm) the probability of absorption – inside Unirradiated ZnO thin film - highly increase and the amplitude of the incident photons will be reduced by a factor 'e' through the short distance within the film thickness , then the skin depth was small .

Conclusions

The actions of irradiation by gamma ray on ZnO thin film are: decreasing the energy gap, increasing the absorption coefficient. Optical constants like refractive index, extinction coefficient and dielectric constants (ε_r), (ε i) increased after irradiation. Skin depth also decreases after irradiation.

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تاثير اشعة كاما على الثوابت البصرية وفجوة الطاقة لاغشية ZnO

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درسنا تأثير التشعيع باشعة كاما على طيف الامتصاص وفجوة الطاقة البصرية لاغشية اوكسيد الزنك . ($^{\circ}$ 200) لاغشية مرسبة على قواعد زجاجية بدرجة بدرجة مع طاقة الفوتونات الساقطة كدالة التشعيع باشعة كاما. وكان للاغشية (α) وقد لوحظ تغير معامل الامتصاص فجوة طاقة مباشرة متأثرة بالتشعيع .الثوابت البصرية مثل معامل الانكسار، معامل الخمود ، وثوابت العزل زادت بعد التشيع.($_{(3)}$),($_{(3)}$)