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Applying Optical Swanepoel's Model to Assess the Effect of UV-Irradiated Time on Optical Properties of ZnSe Thick Film

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Abstract: Zinc selenide thick films were prepared by evaporation technique onto glass substrates. The nature of the films was investigated by XRD pattern. The optical properties of the as-deposited and UV-irradiated films with different exposure times (0, 30, 60, 90, and 120 min.) were reported. The polycrystalline nature of the films was detected by XRD measurements. XRD patterns for UV irradiated films show that the intensities of the peaks increase with increasing the UV-irradiated time, but the full width at half maximum (FWHM) decreases. The optical constants of the films were calculated by Swanepoel's method in terms of wedge shape model in order to obtain both thickness and refractive index with high precision. The energy gap of ZnSe films were determined by using the transmission in terms of transmission spectra in the strong absorption region. In terms of the obtained results, the increase of exposure time improved shrinkage of non-uniform transmission in both the medium and robust absorption region of the ZnSe film. The optimal improvement was at UV-irradiation time equal 120 min. Owing to tunability in the optical constant and energy gap of ZnSe thin films with UV-irradiation ZnSe serves as promising films in optoelectronic devices. Also, The dielectric constant and dissipation factor of ZnSe films and found to be decreased with increasing of the exposure time of UV-irradiation.

Keywords: ZnSe thin films; UV-irradiation; Optical constants; Wedge shape model; The dielectric constant.

1 Introduction

Zinc selenide (ZnSe) is an appealing component of II-VI Semiconductor compounds due to their unique uses for optoelectronic devices[1]. So it is crucial to study the optical properties of ZnSe over a wide range of wavelengths. Thin film technology is vital for the manufacture and development of integrated circuit industry needs[2]. The development of the smallest devices at a considerable speed, particularly in the new generation of integrated circuits, needs advanced materials and new processing routes appropriate for the future technology of giga scale integration(GSI) [3, 4]. Thus, physics and thin-films technology play an essential role in validating this target. In this sense, the thin films have several applications, especially, micro-electronics, communication, optical-electronic devices, catalyst, etc. It worth mention that the ZnSe thin-films have numerous applications in optical devices technology, for instance: solar cell, photodetector, blue light-emitting devices, etc.). Furthermore, a great portion of the mentioned devices can be fabricated in terms of thin-film technology these methods include thermal evaporation under vacuum, pulsed-laser deposition, molecular beam epitaxy, electrodeposition, and spray pyrolysis [5-8]. Moreover, the studying of the structural and optical properties of ZnSe thin film gives valuable information about the ZnSe properties. A lot of publications have been determined both the refractive index and thickness of thin films in terms of double beam spectrophotometer and optical spectroscopic ellipsometry (OSE). Both of the two techniques are very important to investigate the optical response of materials [9-13]. Radiation and its various sources improve many properties of crystalline materials. In particular, as it adjusts improves the structural properties, optical crystallization and improves the rate of electron transport between the energy levels of the amorphous materials, which makes them the transition to the crystalline phase, not to mention its ability to convert it to behave as approximates the mineral behavior in most cases. [14, 15]. Radiation also plays with its various sources the role of controlling local atomic changes for most of the substances subject to radiation [15-17]. Generally, significant changes in both linear and non-linear optical constants and parameters are

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mainly linked to radiation effect or on the other words related to irradiative stimulation if this expression is correct [18-19]. In this regard, absorption-edge transmits or its shifts as photo-darkening or photo-bleaching warrants the use of these materials in the main manufacture of a great number of optical tools [20-23]. Based on what has been mentioned, the precise identification of constants and optical parameters is very important, and this importance is not limited to knowledge of the basic mechanisms resulting from all these effects, but extends to the use of these materials in modern technology. As to the method which utilizes to determine the optical constants and parameters of thin films that linked to it, one must consider that, in case of the optical thickness of the thin film is enough to create sundry interference extremes, it is possible to compute both of the refractive index and film thickness with high precision [8-16].

In the present framework, we will mainly look at starting with the analysis of Swanepoel's method in terms of wedge shape model [23] in the study of optical properties for as-deposited and UV treated ZnSe thin films in the time range between 0 and 120 minutes. This method based on the extremes of the interference fringes of the transmission spectrum for determination, both film thickness, and refractive index. The absorption coefficient and the energy gap have been determined in terms of transmission spectra in the high absorption region. Also, the present framework was concentrated fundamentally on all these properties, which are surely those that demand a much more itemized computation method in the present explanation, which are the most rarely found in the literature.

2 Experimental

High purity ZnSe powder (99.999 %) from Aldrich Company was used for the preparation of ZnSe thin films with different thicknesses onto ultrasonically cleaned glass substrate kept at constant temperature (373 K) using a thermal evaporation unit (Denton Vacuum DV 502 A) under a vacuum of 10^{-6} Pa. In order to obtain the optimal conditions for a homogeneous thin film, a mechanical rotation of the substrate holder (≈30 rpm) during deposition was used. Both the film thickness and the deposition rate were controlled using a quartz crystal monitor DTM 100. The deposition rate was kept at 2 nm/s during the preparation of thin films. Both the structure of the powder and thin films were examined by X-ray diffraction (XRD) (Philips X-ray diffractometry (1710)) with Ni-filtered CuK_{α} radiation with (λ = 0.15418 nm). Both the morphology the elemental composition of the ZnSe thin films were analyzed by using energy dispersive X-ray spectrometer unit (EDXS) interfaced with a scanning electron microscope, SEM (JOEL XL) operating an accelerating voltage of 30 kV. The relative error of determining the indicated elements does not exceed 2 %. Illumination of the studied films was carried out at 300 K using a UV lamp (Minera-light, San Gabriel, CA) with 20 mW/cm^2 and infrared cut filter, in order to suppress any temperature effect on the absorption edge shift. The UV exposure time was taken as UV-irradiated films with different exposure times (0, 30, 60, 90, and 120 min.). The measurements of the transmittance, T was carried out in terms of a double-beam spectrophotometer (Jasco V670) at normal incidence of light in a wavelength (λ) range between 400 and 2500 nm. The measured transmittance spectrum was carried out without a glass substrate in the reference beam in order to calculate the refractive index and the film thickness of ZnSe thin films according to Swanepoel's method.

3 Theoretical Foresights

In terms of Swanipoel relationship, the transmission of thin films of uniform thickness, d and their complex refractive index, $\tilde{n}$ which equaled to $\tilde{n} = n - ik$ and α refers to on absorption coefficient can be expressed as follows [25, 27, 28]:

$$T(\lambda, s, n, d, k)\big|_{k=0} = \frac{Ax}{B - Cx\cos(\varphi) + Dx^2} \tag{1}$$

It should be noted that the interference is done through three media that have different refractive indexes, which are air, that has a refractive index equal to the correct one, namely $n_o = 1$ and also a refractive index for the glass substrate, s and finally a refractive index for the studied thin films, n.

Here, $A = 16n^2 s$, $B = (n+1)^3(n+s^2)$, $C = 2(n^2-1)(n^2-s^2)$, $D = (n-1)^3(n-s^2)$, $\varphi = 4\pi n d/\lambda$, $x = \exp(-\alpha d)$ and $k = \frac{\alpha\lambda}{4\pi}$.

All parameters in these equations are a function of wavelength. It is not complex to view that at $\cos(\varphi) = \pm 1$, that is, one can reformulate the first equation (Eq. 1) at the extremes of the interference points as follows:

$$T_{Mo} = \frac{Ax}{B - Cx + Dx^2} \quad \text{for (the interference maxima)} \tag{2}$$

And

$$T_{mo} = \frac{Ax}{B + Cx + Dx^2} \quad \text{for (the interference minima)} \tag{3}$$

Schematically, the expression about non-uniform thin films has been illustrated in Fig. 1. This figure is presumed that the thickness changes linearly through the illuminated area [29, 30]; thus, it can be illustrated by $d = \bar{d} \pm \Delta d$ Δd refers to the actual changes in thickness from the average thickness $\bar{d}$, as appeared in Fig. 1, and must not be

confounded with a standard deviation of computed values, RMSE. Deviation utilized by other workers [25]:

$$RMSE = \sqrt{\frac{\sum_{i=1}^{q}(d-(\bar{d}))}{q}} \quad (4)$$

Here, the interference style shrinks dramatically on account of the thickness is not uniform. Now, Eq. (1) must be integrated over both the thickness of the film Δd and x for it to more exactly depict the transmission spectrum $T_{\Delta d}(\lambda)$ [26]. The transmittance then becomes as follows:

$$T_{\Delta d} = \frac{1}{\varphi_2 - \varphi_1}\int_{\varphi_1}^{\varphi_2} \frac{A\bar{x}}{B - C\bar{x}\cos\varphi + D\bar{x}^2} d\varphi \quad (5)$$

Here $\varphi_1 = 4\pi n(\bar{d} - \Delta d)/\lambda$, $\varphi_2 = 4\pi n(\bar{d} + \Delta d)/\lambda$ and $\bar{x} = \exp(-\alpha\bar{d})$.

The integral yields:

$$T_{\Delta d} = \frac{\lambda}{4\pi n\Delta d}\frac{a}{(1-b^2)^{1/2}}\left[\tan^{-1}\left(\frac{1+b}{(1-b^2)^{1/2}}\tan\frac{\varphi_2}{2}\right) - \tan^{-1}\left(\frac{1+b}{(1-b^2)^{1/2}}\tan\frac{\varphi_1}{2}\right)\right] \quad (6)$$

Here,

$$a = \frac{A\bar{x}}{B + D\bar{x}^2} \quad \text{and} \quad b = \frac{C\bar{x}}{B + D\bar{x}^2} \quad (7)$$

As a result, one will have:

Maxima:

$$T_{M\bar{x}} = \frac{\lambda}{4\pi n\Delta d}\frac{a}{(1-b^2)^{1/2}}\left[\tan^{-1}\left(\frac{1+b}{(1-b^2)^{1/2}}\tan\frac{2\pi n\Delta d}{\lambda}\right)\right] \quad (8)$$

Minima:

$$T_{m\bar{x}} = \frac{\lambda}{4\pi n\Delta d}\frac{a}{(1-b^2)^{1/2}}\left[\tan^{-1}\left(\frac{1-b}{(1-b^2)^{1/2}}\tan\frac{2\pi n\Delta d}{\lambda}\right)\right] \quad (9)$$

Substituting: Eq. (7) in: Eqs. (8 & 9) and utilizing Eq. (5), the subsequence consolidated equations between the main experimental envelopes T_M and T_m of the non-uniform film and the envelopes T_{Mo} and T_{mo} of the uniform film, with a thickness equal to the average thickness of the non-uniform film, are obtained:

$$T_M = \frac{\sqrt{(T_{Mo}T_{mo})}}{\chi}.\tan^{-1}\left[\sqrt{\left(\frac{T_{Mo}}{T_{mo}}\right)}.\tan\chi\right] \quad (10)$$

$$T_m = \frac{\sqrt{(T_{Mo}T_{mo})}}{\chi}.\tan^{-1}\left[\sqrt{\left(\frac{T_{mo}}{T_{Mo}}\right)}.\tan\chi\right] \quad (11)$$

Where

$$\chi = \frac{2\pi n\Delta d}{\lambda} \quad (12)$$

and

$0 < \chi < \pi/2$ (or equivalently, $0 < \Delta d < \lambda/4n$) (13)

4 Results and Discussion

4.1 The Cystallographic Structure of ZnSe Thin Films

Fig. 2 displays XRD diffractogram of ZnSe powder according to JCPDS Data file no.: 05-0566; it can be indexed to ZnSe cubic phase with polycrystalline in nature. The XRD patterns of UV-irradiated ZnSe films with different exposure times (0, 30, 60, 90, and 120 min.) are shown in Fig. 2. The observed peaks of ZnSe thin films are compared with the standard JCPDS values, which showed that the films give a structure matching with (JCPDS file no.: 05-0566-cubic) zinc blend Cubic structure.

The observed peaks of ZnSe films are polycrystalline with peaks detected at 2θ = 28.56°, 47.52° and 56.29° corresponding to C(1 1 1), C(2 2 0) and C(3 1 1) orientations, respectively. Fig. 3 shows that the intensities of the peaks increase with increasing the UV-irradiated time, but the full width at half maximum (FWHM) decreases with increasing the film thickness.

4.2 Optical Analysis

Figure 4 exhibited the transmission T(λ) of ZnSe thin film in a spectral range from 400 to 2500 nm as the function exposure time of the UV- irradiation. Obviously, at the transmission spectrum, particularly in the transparent region, there are distinct interference fringes with high intensity of around≈ 86 %. As we transfer to the shorter wavelength since the absorption starts to occur within the film, the interference fringes intensity starts to decrease gradually till it near zero at the edge of the optical bandgap of the deposited films.

Besides, shrinkage appears in the transmission spectra in both the weak and the medium absorption regions, which reflect non-uniformity of the films. This shrinkage obviously has enhanced with the increase in the exposure time of UV-irradiation, as demonstrated in Fig. 5. Fig. 6 shows the blue shift of the transmission spectra with the increase in UV-irradiation time ZnSe thin films.

The two relations (10 & 11) are independent transcendental in T_{Mo}, T_{mo} and χ. $T_{Mo} \equiv T_s$ in the transparent region as illustrated in Fig. 7. In order to compute T_{mo} and χ in the mentioned spectral region, one can solve Eqs. (10, 11) utilizing an approximate solution to a system of nonlinear relations by minerr function in Mathcad 2000 program.

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The fundamental relation for the interference fringe points starting from the long-wavelength will be written in accordance with previous papers [10, 15, 16] as follows:

$$\frac{l}{2} = \frac{2n\bar{d}}{\lambda} - m_1 \qquad l = 0,1,2,3,4.... \tag{14}$$

Here: m_1 is the order number of the first at (l = 0) extreme considered, an integer for a maximum of half-integer for a minimum. Substituting Eq. (12) in Eq. (14) yields:

$$\frac{l}{2} = \left(\frac{\bar{d}}{\pi \Delta d}\right)\chi - m_1 \tag{15}$$

In case of plotted $\frac{l}{2}$ as a function of χ for the transparent region, the slope and m_1, of Eq. (15) can be obtained, as shown in Fig. 8 (for the first studied thin film). Eq. (15) can now be utilized to compute χ for the extrema in the absorption region, and T_{Mo} and T_{mo} can be computed from Eqs. (10 &11), as illustrated below.

Now T_{mo} and χ are computed from Eqs. (10 &11), utilizing the values of T_M and T_m at the extrema of each spectrum summarized in Table 1 and putting $T_{Mo}(\lambda) = T_s(\lambda)$. The values of χ are shown as χ_1, in Table 1, and $\frac{l}{2}$ is plotted as a function of χ, according to Eq. (15) in Fig. 8. Next, the best straight lines through the points of the transparent region are drawn. The deviation of the points for larger χ, from these straight lines indicates the onset of absorption, and these points must be rejected. From Fig. 8 (for the first studied film, as an example), and Eq. (15) can be represented by the following expressions for the last four studied films, respectively:

$$l/2 = 7.339\chi - 4.425 \tag{16}$$

$$l/2 = 9.701\chi - 5.617 \tag{17}$$

$$l/2 = 10.481\chi - 5.505 \tag{18}$$

$$l/2 = 10.713\chi - 4.902 \tag{19}$$

$$l/2 = 10.713\chi - 4.545 \tag{20}$$

The value of χ at each extreme is now computed from the expressions which result from modifying these last four equations. The new values χ are shown in Table 1. Using these values of χ, together with the values of T_M and T_m, the T_{Mo} and T_{mo} are computed from Eqs. (10 & 11). These values T_{Mo} and T_{mo} are shown in Table 1.

The obtained values of T_{Mo} and T_{mo} can now be utilized to derive: $\bar{d}$, Δd and $n(\lambda)$ via the method for uniform films, discussed in detail in our previous works [29, 30]. We shall now briefly discuss this method.

The value of Δd can be calculated in terms of comparison of the slope of Eq. 15 and the slopes of Eqs. (16, 17, 18, 19 and 20). Fig. 9 shows the variation of Δd as a function of UV-irradiated. This Figure shows that the value of Δd decreases with increasing exposure time, i.e the shrinkage of transmission has improved.

In addition, the values of n_1 can be used to derive d_2 from the basic equation for the interference fringes $2n\bar{d} = m\lambda$ (m is the order number of the interference extrema) and also improved values for n, shown as n_2, as described in detail previously [32-34]. Fig. 10 illustrates the dependence of n on wavelength for different exposure times (0, 30, 60, 90, and 120 min.) of UV-irradiated of ZnSe films. This figure shows that the refractive index, n_2, decreases with increasing exposure times over the entire spectral range studied.

In accordance with Swanepoel [25] recommends, in the case of uniform films, that the T_{Mo} curve be utilized over all spectrum range, namely, strong, medium, and weak absorption regions. The corresponding expression to compute the absorbance, x is as follows:

$$x = \frac{E_{Mo} - [E_{Mo}^2 - (n^2-1)^3(n^2-s^4)]^{1/2}}{(n-1)^3(n-s^2)},$$

$$\alpha = -\frac{1}{d} \times \ln(x) \tag{21}$$

Where $E_{Mo} = \frac{8n^2 s}{T_{Mo}} + (n^2-1)(n^2-s^2)$. $\alpha(\lambda)$ is then computed, utilizing the main expression $\alpha = -\frac{1}{d} \times \ln(x)$.

The absorption coefficients, α of the studied thin films depending on the wavelength, λ in the regions of their transparency $\lambda > 500(nm)$ were also computed utilizing the subsequence relation [35-37]:

$$\alpha(\lambda) = \frac{1}{d}\left[\ln(1-R_1) + \ln(1-R_2) + \ln(1-R_{12}) - \ln(T(\lambda))\right] \tag{22}$$

On the other word, based on [35], the absorption in the ranging from 300 to 500 nm is almost characterized by the

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all met conditions: strong absorption in the film, transparency of substrate, and satisfied inequality $n^2 >> k^2$ and thus according to [25, 37], one can express about absorption coefficient in the mentioned range by the following equation:

$$\alpha(\lambda) = \frac{1}{d}\left[\ln(1-R_1) + \ln(1-R_2) + \ln(1-R_{12}) - \ln(T(\lambda))\right] \quad (23)$$

Here: $T(\lambda)$ is the transmittance of the film at the selected λ and parameters R_1, R_{12}, R_2 reflection coefficients from the following boundaries:

Air_film:

$$R_1 = \left(\frac{(n(\lambda)-1)}{(n(\lambda)+1)}\right)2 \quad (24)$$

Substrate_air:

$$R_{12} = \left(\frac{(1-s(\lambda))}{(1+s(\lambda))}\right)2 \quad (25)$$

Film_substrate:

$$R_2 = \frac{(s(\lambda)-n(\lambda))}{(s(\lambda)+n(\lambda))})2 \quad (26)$$

Fig. 11 shows the dependence of the absorption coefficient α on the incident photon energy ($h\nu$) for the different exposure times (0, 30, 60, 90, and 120 min.) of UV-irradiated of ZnSe films.

The absorption coefficient can be described in terms of the Eq. 27 based on the allowed direct band to band transition in the near of the absorption region.

$$\alpha(h\upsilon) = \frac{K(h\upsilon - E_g^{opt})^m}{h\upsilon} \quad (27)$$

where k independent parameter edge width of the photon energy for the respective transitions [38], E_g^{opt} is the optical band gap, and m is the number that characterizes the process of transition. Various studies [29-31] have suggested several values of m for different glasses, m = 2 for the most amorphous semi-conductors (indirect-transition); however, m= ½ for the crystalline semi-conductor (direct-transition). Meanwhile, the ZnSe thin films are polycrystalline, so the direct transition was vailed. Figure 12 displays the plots of $(\alpha h\nu)^2$ against photon energy (hν) for an exposure time of UV-irradiation for ZnSe thin films. The direct optical energy gap is determined via using the extrapolation in the linear part in the $(\alpha h\nu)^2$ Vs. (hν) at $(\alpha h\nu)^2 = 0$. The increase in the exposure time of UV-irradiation for ZnSe thin films lead to enhance the energy gap from 2.55 to 2.70eV. For further calculation of the rest optical constants, the extinction coefficient (*k*) is computed based on the values of α and λ utilizing the known formula k = αλ/4π. Figure 13 demonstrates the dependence of k vs. λ for numerous of an exposure time of UV-irradiation for ZnSe thin films.

4. 3 the Dielectric Constants and Loss Factor

We shed light on the dielectric constant, which helps in understanding the basic crystal physics, particularly the bonding in the crystals**.** In the theoretical studies of the lattice dynamics, the dielectric constant is considered one of the input parameters. The real part of the dielectric constant relates to dispersion, while the imaginary part of dielectric constants is directly linked to the density of states within the forbidden gap of semiconductors materials [42]. The real ($\varepsilon_r = n^2 - k^2$) and imaginary ($\varepsilon_i = 2nk$) parts of the dielectric constant [42, 43] of ZnSe films have been observed. They reveal a similar trend with photon energy. The variation of these two parameters with photon energy has appeared in Fig. 14. The obtained dielectric constants decrease with increasing photon energy, but it decreases with increasing of the exposure time of UV-irradiation. The values of ε_r is higher than the values of ε_i as shown in Fig. 14.

There is quantity frequently used to characterize the optical properties of thin films is called the dissipation factor, $\tan(\delta)$, which can be evaluated by using the following relation [44]:

$$\tan(\delta) = \frac{\varepsilon_i}{\varepsilon_r} \quad (28)$$

The variation of the dissipation factor as a function of photon energy is shown in Fig. 15. This figure appears that the dissipation factor increases with increasing photon energy.

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5 Conclusions

Polycrystalline ZnSe thin films with thickness 720 *nm* were prepared by thermal evaporation under ultra-vacuum onto glass substrates. XRD patterns for UV irradiated films show that the intensities of the peaks increase with increasing the UV-irradiated time, but the full width at half maximum (FWHM) decreases. The effect of increasing *UV* –irradiation time on optical constants and energy gap of the ZnSe films were investigated. The optical constants of the films were calculated by Swanepoel's method in terms of wedge shape model in order to obtain both thickness and refractive index with high precision. The energy gap of ZnSe films were determined by using the transmission in the strong absorption region that increases with increasing UV-irradiation time. In terms of the obtained results, the increase of exposure time improved shrinkage of non-uniform transmission in both strong and medium absorption region and converted it to uniform transmission at optimal UV-irradiation time equal 120 min. Accordingly, the

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possibility of asserting that such tunability in optical refractive index and energy gap of ZnSe thin films with UV-irradiation serves as promising films in optoelectronic devices.

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