

Variation of band edge, optical constant and dispersion parameters of CdS thin films due to disorder induced by fast neutron irradiation

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Abstract

CdS thin films were prepared by thermal evaporation method, optical constant of the deposited films were obtained from the analysis of the experimental recorded transmittance spectral data over a wavelength range of (200-900)nm. The value of refractive index of unirradiated and radiated samples are determined from these spectra using Swanepoel's model. It has been found that the dispersion data obey single-oscillator relation of the Wemple-DiDomenico model from which the dispersion parameters were determined the third order optical susceptibility $\chi^{(3)}$ of unirradiated and irradiated CdS thin film were also determined using these spectra. and the optical absorption at the fundamental absorption edge was discussed. Direct optical energy gap E_g^{opt} and Urbach (localized states) energy were also calculated for all unirradiated and irradiated CdS thin films.

Introduction

Thin films nowadays occupy a prominent space in basic research and solid state technology. The use of thin film semiconductors has attracted much interest in an expanding variety of applications in various electronic and optoelectronic devices due to their low production costs. Cadmium sulphide CdS is one of the most promising II-VI compound materials due to its wide range of applications in various optoelectronic [1], piezo-electronic [2] and semiconducting devices. Thin films of CdS are of considerable interest for their efficient use in fabrication of solar cells [3,4].

Cadmium sulphide which belongs to II-VI group is the most widely used material for CdS/CdTe and CdS/Cu₂S heterojunction solar cells, because of the fact that CdS has intermediate energy band gap, reasonable conversion efficiency, stability and low cost [5,6]. In semiconducting materials under irradiation, a chain of reactions, in which oxygen and moisture from environment may be incorporated, starts with rapid electronic phenomena, this is followed by the generation of reactive, short-lived intermediate compounds and complex mixture of chemical products. The main result of ionization is breaking of chemical bonds and the creation of new ones, which causes change in optical and electrical properties and lead to long-lived forms of physical breakdown. [7]. The importance of studying the optical properties of a material is offered by the ability of this technique to provide informations regarding the fundamental gap, electronic transition, trapping levels and localized states [8]. The aim of this work is to study the effect of fast

neutron radiation on the band edge, localized states, optical constant, and dispersion parameters of CdS thin films.

Experimental

The pure CdS thin films were prepared on glass substrates at room temperature by thermal evaporation method at vacuum 10^{-5} Torr.

The crystalline structure of the films were investigated by standard X-ray diffraction (XRD) technique, using $\text{CuK}\alpha$ radiation ($\lambda=1.54\text{\AA}$). More detailed information about the preparation procedure and XRD pattern for pure CdS thin film can be found in the work of H.S. Al-Selman [9].

CdS sample were irradiated with fast neutrons that emitted from Am-Be (in Contact). The yield of the neutron source was 1.1×10^7 n/s the neutron fluence of Am-Be source was found to be $(3.9 \pm 0.78) \times 10^9$ n/cm² for each day of a irradiation and the mean energy of its emitted neutrons was 4.5 MeV [10]. The source was supplied by Radio-chemical Ltd, Amersham, England.

The optical absorbance (A) and Transmittance (T) of the unirradiated and irradiated samples were measured at room temperature using (Helios- α spectronic

Results and discussion

Transmission spectrum is used to calculate the refractive index using the envelope method proposed by Swanepoel ,The refractive index (n) has been obtained using the following expressions [11].

$$n = [M + (M^2 - S^2)^{1/2}]^{1/2} \quad \text{----- (1)}$$

$$M = \frac{25}{T_m} = \frac{(S^2 + 1)}{2} \quad \text{----- (2) for transparent region}$$

$$M = 25 \frac{T_M - T_m}{T_M T_m} + \frac{(S^2 + 1)}{2} \quad \text{----- (3) for weak and medium absorption region.}$$

$$S = \frac{1}{T_s} + \sqrt{\frac{1}{T_s^2} - 1} \quad \text{----- (4)}$$

T_M and T_m are the values of maximum and minimum transmission values at a particular wavelength, "S" is the refractive index of the substrate, T_s is the transmission of the substrate. Refractive index can be estimated by extrapolating envelopes corresponding to T_M and T_m . As the thickness of the film is uniform, interference give rise to the spectrum as shown in Fig (1) .

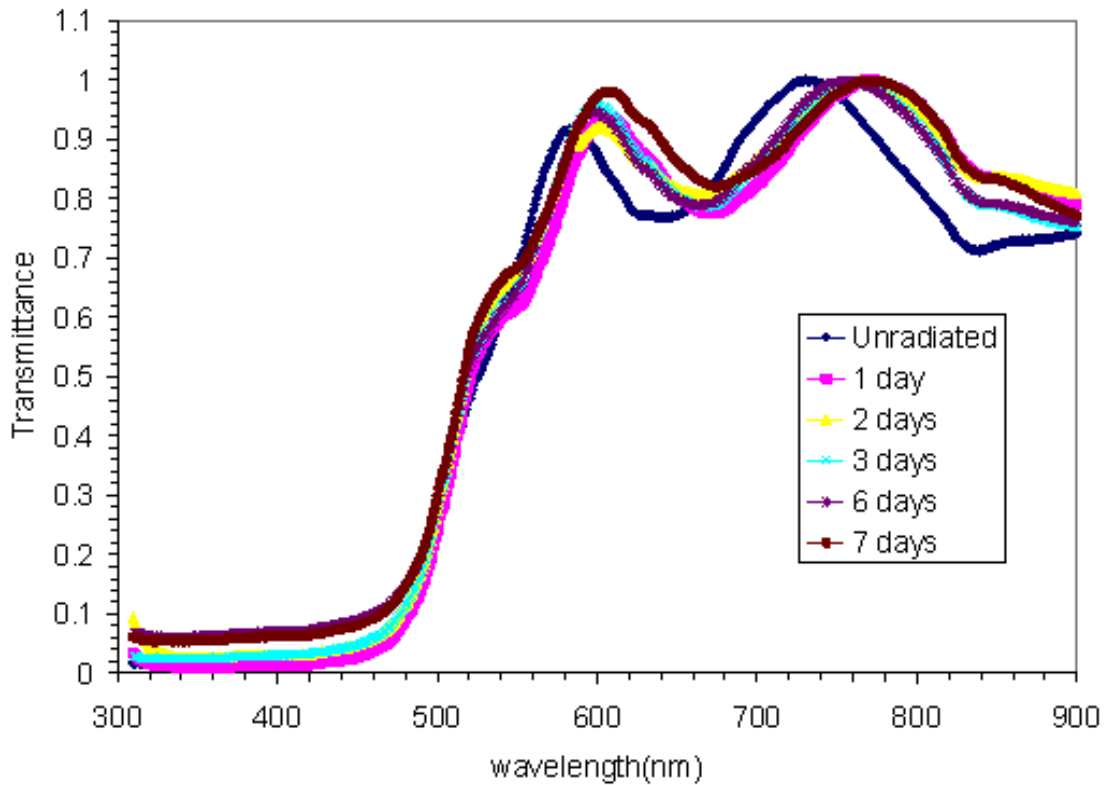


Fig1.Variation of transmittance with wavelength for unradiated and irradiated CdS thin films

The variation of refractive index with the wavelength is shown in Fig(2) for unradiated and irradiated CdS thin films.

We studied the spectral dispersion of the refractive index for unradiated and irradiated CdS samples. The dispersion spectrum of the refractive index was fitted using the Cauchy formula [12]

$$n = A + \frac{B}{\lambda^2} \quad \text{----- (5)}$$

where A and B are the Cauchy's parameters.

The calculated values for A and B are given in table I.

Refractive index found to decrease with increasing irradiation time. This may be due to the change of the lattice structure.

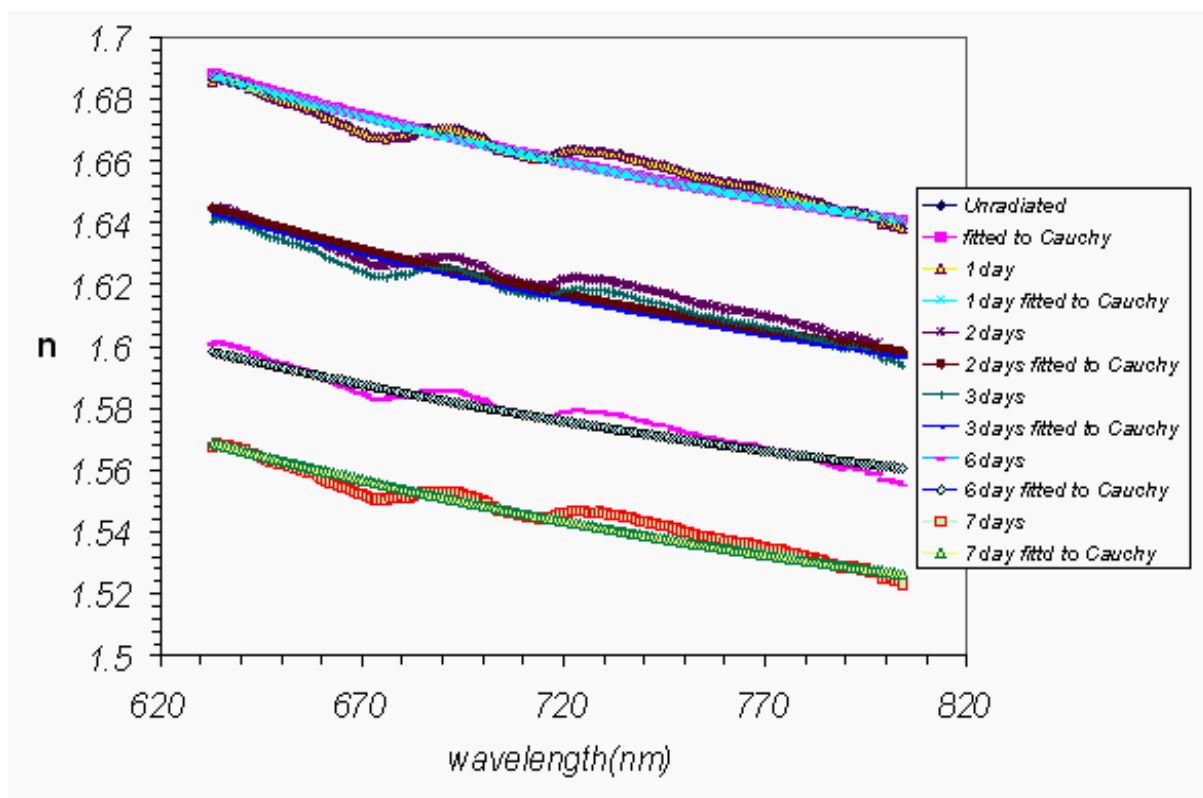


Fig.2 Variation of refractive index with wavelength for unirradiated and irradiated CdS thin films

Table I: Cauchy Parameters for unirradiated and irradiated pure CdS thin films

Irradiation		A	B (nm) ²
Dosage (day)	Fluence $\frac{\text{neutron}}{\text{cm}^2} \times 10^9$		
0	0	1.5643	49403
1	3.9	1.5630	50190
2	7.8	1.5225	39614
3	11.7	1.5209	48936
6	23.4	1.4994	48786
7	27.3	1.4571	44800

Based on single-oscillator model, the Wemple-DiDomenico is a semi-empirical dispersion relation for determining the refractive index at photon energies below the interband absorption edge as a variety of materials, it is given by[12].

$$n^2-1 = E_o E_d / \{E_o^2 - (hv)^2\} \quad \text{----- (6)}$$

Where hv is the photon energy, E_o is the energy of the effective dispersion oscillator also called average energy gap, E_d is the oscillator strength or the dispersion energy, which measures the average strength of the interband optical transitions. Plotting $(n^2-1)^{-1}$ against $(hv)^2$ allows us to determine the oscillator parameters by fitting a straight line to the points. Fig (3) shows the plot of $(n^2-1)^{-1}$ versus $(hv)^2$. The value of E_o and E_d can be directly determined from the slope $(E_o E_d)^{-1}$ and the intercept on vertical axis (E_o/E_d) . The calculated values of E_o and E_d are given in table II.

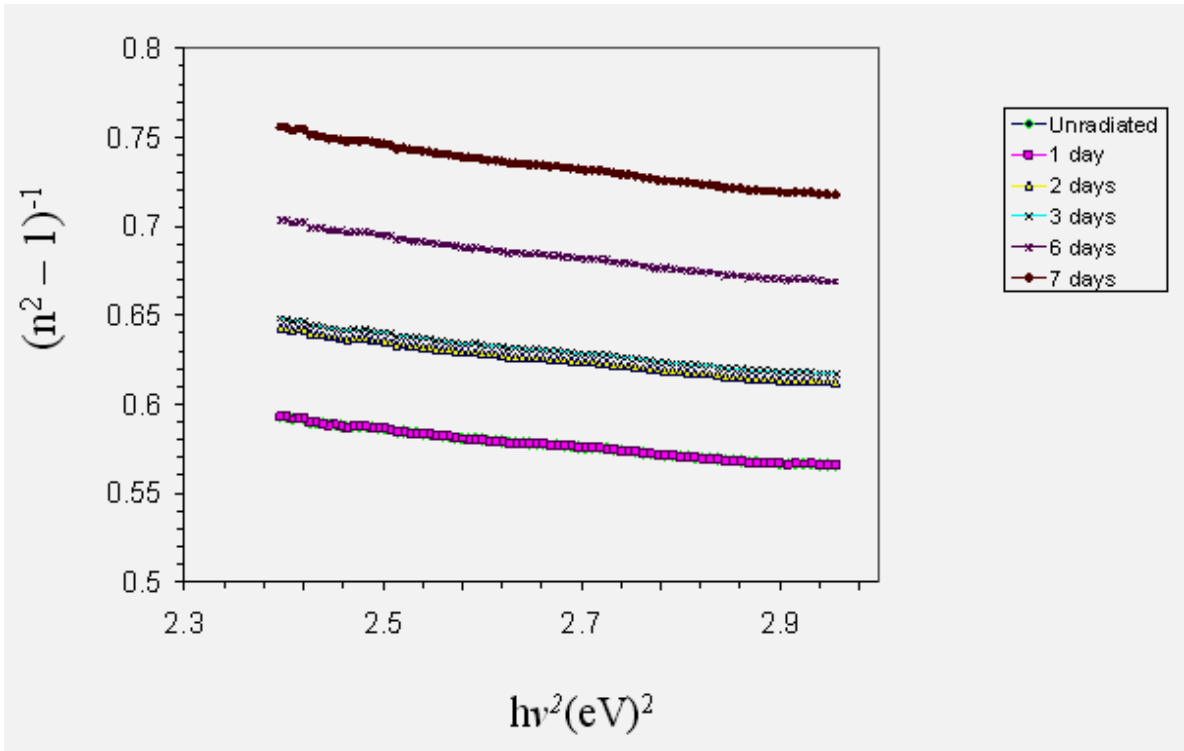


Fig.3 Variation of $(n^2-1)^{-1}$ with $(hv)^2$ for unradited and irradiated CdS thin films

The values of static refractive indices $n(0)$ for unradited and irradiated CdS thin films were calculated by extrapolating the Wemple-DiDomenico dispersion to $(hv \rightarrow 0)$

Table II : Single oscillator parameters calculated by WDD approximation for unradiated and irradiated pure CdS thin films

Irradiation		E ₀ (ev)	E _a (ev)	E _g ^{opt} = $\frac{E_0}{1.6}$ (ev)	n(o)	ε _∞
Time (day)	Fluence $\frac{\text{neutron}}{\text{cm}^2}$					
0	0	3.85	5.5	2.406	2.47	6.10
1	3.9	3.84	5.4	2.40	2.428	5.89
2	7.8	3.81	4.76	2.38	2.25	5.06
3	11.7	3.78	4.74	3.36	2.23	4.99
6	23.4	3.74	4.70	2.33	2.219	4.92
7	27.3	3.62	4.067	2.26	2.20	4.86

According to Wagner [13] the Miller rule is very convenient for visible and nonlinear and near infrared frequencies, which equalize the third order nonlinear polarizability parameter, $\chi^{(3)}$, the so-called nonlinear optical susceptibility, and the linear optical susceptibility, $\chi^{(1)}$, through the equation[13].

$$\chi^{(3)} = A (\chi^{(1)})^4 = A[E_0 E_d / 4\pi(E_0^2 - (hv)^2)]^4 = A/(4\pi)^4(n^2-1)^4 \text{ ----- (7)}$$

where $A = 1.7 \times 10^{-10}$. Fig(4) shows the plot of $\chi^{(3)}$ with hv

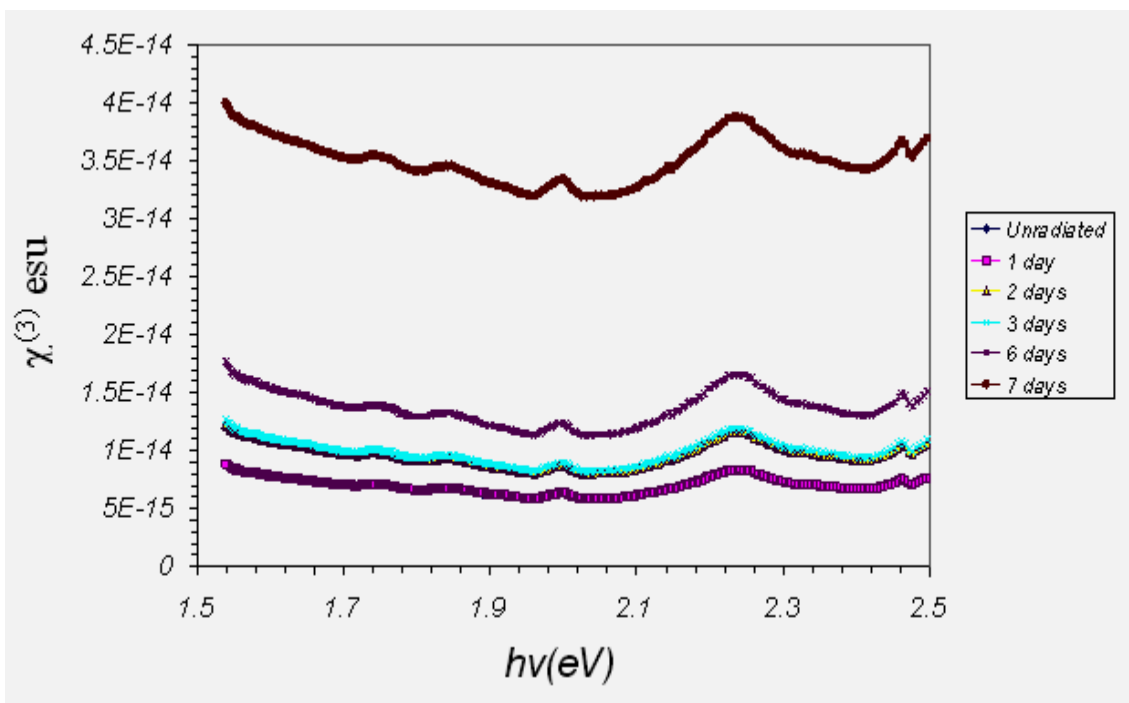


Fig.4 Variation of third order nonlinear optical susceptibility with photon energy For unradiated and irradiated CdS thin films

At the absorption edge, the absorption coefficient can be calculated using the expression [14].

$$\alpha = \frac{1}{d} \ln\left(\frac{1}{T}\right) \quad \text{----- (8)}$$

where d is the thickness of the sample

In the high absorption region from which the optical band gap is determined, the absorption is characterized by Tauc's relation [1].

$$\alpha hv = \beta (hv - E_g^{opt})^r \quad \text{----- (9)}$$

Where hv , E_g^{opt} and β denotes the photon energy, the optical gap and band tailing parameter respectively. In the above equation; $r=1/2$ were used for direct allowed transition. Figs (5) shows the plot between $(\alpha hv)^2$ vs (hv) for calculation of optical band gap E_g^{opt} . The optical gap determined by the intercept of the extrapolations to zero absorption with the photon energy axis [$(\alpha hv)^2 \rightarrow 0$] Tauc extrapolation [14].

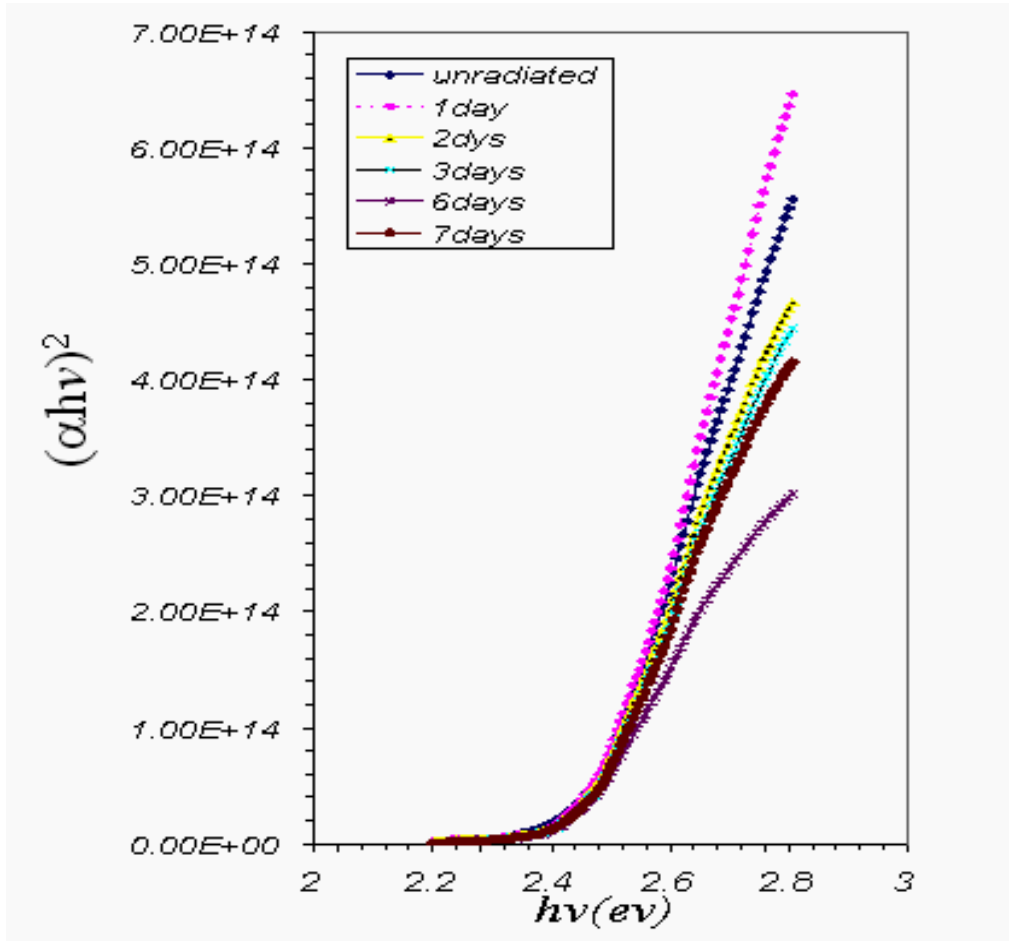


Fig.5 Variation of $(\alpha hv)^2$ with hv for unradiated and irradiated CdS thin

Urbach [14] proposed an empirical rule between the absorption coefficient $\alpha(hv)$ and the photon energy (hv) known as Urbach tail:-

$$\alpha(h\nu) = \alpha_o \exp[h\nu / E_u] \quad \text{-----} \quad (10)$$

where E_u represents the width of the band tails (Urbach tail states).

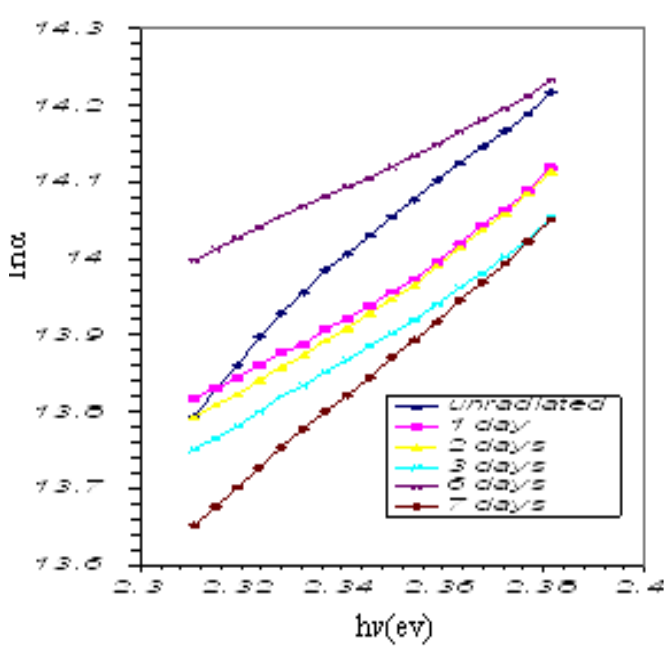


Fig 6. $\ln\alpha$ vis $h\nu$ for unradiated and irradiated CdS thin films

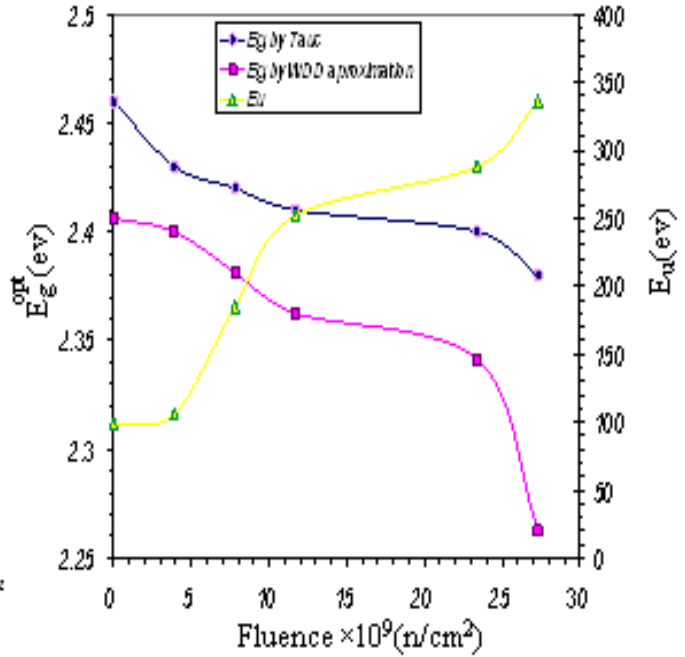


Fig7. E_g^{opt} of Tauc and E_g^{opt} of WDD approximation and width of the tail E_u as a function of fluence

A large variety of insulators and crystalline semiconductors exhibits similar tails in the optical-absorption coefficients. It generally believed that disorder plays a significant role in developing Urbach band tails.[14]

Irradiation		E_g^{opt} (ev)	E_u (mev)
Dosage (day)	Fluence $\frac{neutron}{cm^2} \times 10^9$		
0	0	2.46	99
1	39	2.43	106
2	78	2.42	185
3	117	2.41	252
6	234	2.40	288
7	273	2.38	336

Table III : Optical gap E_g^{opt} and the width of the band tail for unradiated and irradiated pure CdS thin films

Some theoretical and experimental studies have demonstrated that an optical band gap energy E_g in semiconductors can be expressed as [15]

$$E_g = E_x - \beta E_u \quad \text{----- (11)}$$

Some theoretical studies assert that both Tauc gap and Urbach energy are influenced from structural disorder, and that E_x represents the band gap in disorder-free materials.

Defects and a large density of broken bonds forming grain boundaries and leading to a discontinuous structure which does not influence the detailed shape of the band structure. It causes a considerable blurring of the edges of the conduction and the valance bands and gives rise to distribution of deep localized levels (tail states) in the forbidden gap [16,17]. The presence of high concentration of localized states in the band structure is responsible for low value of the optical energy gap E_g^{opt} [18,19].

Conclusions

Pure CdS thin films were fabricated using thermal evaporation method.,optical properties of unirradiated and fast neutron irradiated CdS samples were studied using transmittance spectra,refractive indices were calculated using Swanepoel's model of transmittance spectra, refractive index decreases with an increasing in neutron irradiation time (fluence),dispersion of refractive index was successfully fitted with Cauchy, and good agreement between the models is observed,dispersion energy and oscillation strength values were determined using Wemple-DiDomenico approximation, E_o , E_d $n(o)$ and ϵ_∞ falls with an increasing in neutron irradiation time (fluence),3rd order optical susceptibility $\chi^{(3)}$ was calculated for unirradiated and irradiated Cds samples, $\chi^{(3)}$ falls with an increasing fluence,optical energy gap E_g^{opt} falls while E_u rises with an increasing in neutron irradiation time (fluence),the change of the optical parameter values is an indication that the effect of fast neutron irradiation are able to systematically modify the optical properties of the pure CdS thin film.

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المخلص

حضرت أغشيه رقيقه من كبريتيد الكاديوم بطريقة التبخير الحراري وتم حساب الثوابت الضوئية من قيم النفاذيه المقاسه في مدى الأطوال الموجيه (٢٠٠-٩٠٠) نانوميتر وتم حساب قيم معامل الأنتكسار للنماذج المشععه وغير المشععه بأستخدام نموذج سوانبويل ووجد أن تشتت معامل الأنتكسار يخضع لنموذج وامبل ديدومينكو ومنه تم حساب معاملات التشتت كما وتم إيجاد الحد الثالث اللاخطي للتأثيريه الضوئيه للعينات كما وتم مناقشة الأمتصاص الضوئي عند حافة الأمتصاص الأساسيه وتم إيجاد قيم فجوة الطاقة وطاقة المستويات المقيدة للعينات المشععه وغير المشععه