

## Structural and photoelectronic properties of vacuum evaporated CdSe thin films

Pradip Kumar Kalita<sup>1\*</sup>, B K Barma and H L Das

<sup>1</sup>Department of Physics, Gauhati University, Guwahati-781 014, Assam, India

<sup>2</sup>Department of Physics, Guwahati College, Guwahati-781 021, Assam, India

E-mail : pradipkalita@rediffmail.com

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**Abstract** : Thermally evaporated CdSe thin films deposited at high substrate temperature  $T_s \geq 473$  K are found to be polycrystalline with hexagonal ZnS type structure. The grain size increases with  $T_s$ . Film crystallinity shows the most preferential orientation along [002] reflection. The sublinear behaviour of photocurrent – light intensity characteristics, measured at different experimental condition for the films deposited at different  $T_s$  and  $t$ , clearly confirms the defect controlled photoconductivity in CdSe films. Activation energies in dark and under illuminations are calculated from the temperature dependence of dc conductivity of the films. The reduction of activation energy under illumination is explained on the basis of the grain boundary effect. The photocurrent in the films increases whereas the photosensitivity decreases exponentially with the rise of ambient temperature within the range 303–403 K which is attributed to the effect of different types of scattering on carrier mobility. Spectral response shows a direct transition at 725 nm with an additional absorption around 975 nm which also confirms the existence of inherent defect states in CdSe films.

**Keywords** : CdSe thin film, X-ray diffraction, defects, grain boundaries, annealing.

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### 1. Introduction

Cadmium selenide is a promising material because of its wide range of successful applications in solar cells, photoconductors, light amplifiers, light emitting diodes, lasers, photoelectrochemical cells, transistors and others optoelectronic devices [1–3]. It has a direct band gap of 1.7 eV and shows advantage over the other II–VI class materials due to its high photosensitivity. In view of these aspects, researches have been carried out to yield stable, reproducible and more consistent structural and photoelectronic characteristics.

A survey of literature reveals that various thin film preparation techniques such as vacuum evaporation, sputtering, spray pyrolysis, solution growth and electro-deposition have been employed for the deposition of CdSe films. However, the thermal evaporation technique is

generally preferred because of its simplicity. In thermally evaporated CdSe thin films, native defects are created by excess Cd and Se vacancies. They are expected to play a dominant role in determining the major features of photoelectronic properties as revealed by several workers [4,5]. Thermally evaporated CdSe thin films may possess cubic zincblende, wurtzite or mixed structure that depend on sensitive deposition parameters such as the order of vacuum, the rate of deposition and substrate temperature [6]. Although a variety of works have been done so far in CdSe films in both single crystal and thin film form, it is felt that the works are still insufficient in thermally evaporated films to establish its structural and photoresponse properties. Hence, further works on its structural characterization is desirable. An attempt has been made in this paper to explore the structural and photoelectronic properties of thermally evaporated CdSe thin films.

\*Corresponding Author

## 2. Experimental

Thin films of CdSe were prepared by thermal evaporation technique (HINDHIVAC 12 A4) on to highly cleaned glass substrates held at temperatures 303 K to 623 K ( $T_s$ ) in a vacuum of the order of  $10^{-6}$  torr. Pure (99.999%) CdSe powder obtained from Koch Light Lab., UK was used for deposition. A gap-type film configuration with a gap of 3 mm between the Al electrodes was made to facilitate the photoresponse studies. The film thickness ( $t$ ) was measured by multiple beam interferometry with an accuracy of  $\pm 20$  Å. The current measurement was done with the help of a high impedance ( $\sim 10^{14}$  Ω) ECIL electrometer amplifier with an accuracy  $\pm 3\%$ . A 250 W quartz halogen bulb was used for white light illuminations. Monochromatic radiations (400–1050 nm) were obtained by using metal interference filters. The intensity of white and monochromatic lights were measured by using a sensitive Aplab luxmeter. X-ray diffraction patterns of the films were taken by using Philips X-ray diffractometer (PW 1830).

## 3. Results and discussion

### 3.1. Structural characterization :

Thin films of CdSe grown at room temperature are amorphous and those grown at elevated substrate temperature  $T_s \geq 473$  K are polycrystalline in nature. Figure 1 represents the XRD patterns of CdSe films of thickness 1400 Å deposited at elevated temperatures 473 K to 623 K. All polycrystalline CdSe films have hexagonal ZnS type structure as confirmed by JCPDS X-ray Powder file data (card no. 8-459). The lattice parameters  $a$  and  $c$  are calculated which showed good agreement with the results of other workers. It is observed that the lattice constants of CdSe thin films deviate from the bulk values which clearly indicates that the film grains are

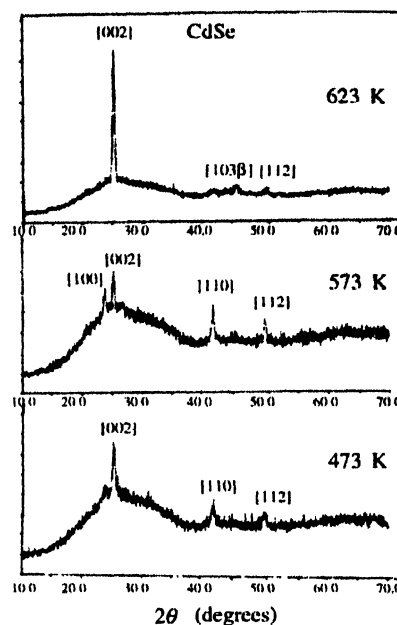


Figure 1. XRD patterns of representative CdSe films (1400 Å) deposited at 473 K, 573 K and 623 K

strained. The microstrain ( $\epsilon$ ) developed in the film grains are determined using X-ray line broadening [7]. It is seen that the microstrain decreases on increasing substrate temperature which in turn, reduces the defects in the films. The strained grains correspond to the change in concentration of vacancies and interstitials created in the films. Thus, the variation in lattice parameters owing to the imperfections may change the density of the film in accordance with  $T_s$  [8]. The grain sizes ( $D$ ) of the films are calculated by using Scherrer formula which is found to increase with  $T_s$ . A relatively higher thickness may also enhance the grain size. All structural parameters are systematically represented in Table 1. It is seen that the most dominant orientation

Table 1. Structural parameters of thermally evaporated CdSe thin films having thickness 1400 Å.

$T_s$ (K) ( $t = 1400$ Å)	$2\theta$ (degrees)	$[hkl]$ from JCPDS Data	$d$ (Å)	$a$ (Å)	$c$ (Å)	$D$ (Å)	$\epsilon \times 10^{-3}$
473	25.2600	[002]	3.5229	4.3146	7.0458	180	2.03
	41.8750	[110]	2.1556	4.3111	7.0401		
	49.4925	[112]	1.8402	4.3156	7.0473		
523	25.1775	[002]	3.4933	4.3285	7.0685	191	1.91
	23.7675	[100]	3.7407	4.3193	7.0534	241	1.69
	25.2500	[002]	3.5243	4.3163	7.0485	216	
573	41.8200	[110]	2.1583	4.3166	7.0490	348	
	49.4625	[112]	1.8412	4.3180	7.0513	323	
	25.3050	[002]	3.5168	4.3071	7.0335	295	1.23
623	45.7100	[103]	1.9833	4.3034	7.0275		
	49.5800	[112]	1.8371	4.3084	7.0357		
	25.2625	[002]	3.5226	4.3142	7.0451	244	1.49
473 ( $t = 2000$ Å)	41.9025	[110]	2.1512	4.3084	7.0357		
	49.3300	[112]	1.8459	4.3289	7.0691		

corresponding to hexagonal phase is along [002] in all polycrystalline CdSe thin films. Dominance of [002] plane indicates that the preferential growth of crystallite is in this direction. Similar results in vacuum evaporated CdSe thin films are also reported by others workers [6,9]. It is to be noted that when substrate temperature is increased, the crystallinity of the film is found to be improved. It shows other planes [100] and [103] at  $23.76^\circ$  and  $45.7^\circ$  ( $2\theta$ ) respectively when films are deposited at 573 K and 623 K. Also the grain size corresponding to [100], [110] and [112] reflections of the CdSe film deposited at 573 K, are quite higher than those oriented along [002] plane. This shows the abundance of more numbers of planes oriented in [100], [110] and [112] direction for this film. During growth of film at higher  $T_s$ , the ad-atoms possess greater mobility along the direction parallel to the substrate surface that favours grain growth, recrystallization and increasing order of microstructure. Thus, the film crystallinity has been enhanced due to the predominant recrystallization along with the recovery process. The structural defects originating from lattice misfit in CdSe thin film, therefore can be minimized by depositing the films at a higher substrate temperature.

### 3.2. Photocurrent versus light intensity characteristics :

The I-V characteristics in dark and under illumination are found to be ohmic in all films. The photocurrent ( $I_{ph}$ ) is defined as  $I_{ph} = (I_L - I_D)$ , where  $I_L$  and  $I_D$  are the currents under illumination and in dark respectively. The photocurrent varies with light intensity  $\phi$  as [10],

$$I_{ph} \propto \phi^\gamma \quad (1)$$

The exponent  $\gamma$  is a number which determines the recombination process in the films.

In the present work, the exponent  $\gamma$  was obtained from the slope of the  $\log_{10} I_{ph}$  vs  $\log_{10} \phi$  plot which is nearly 0.5 in fresh and annealed CdSe films deposited at different  $T_s$  (Figure 2). However for the film deposited at 573 K,

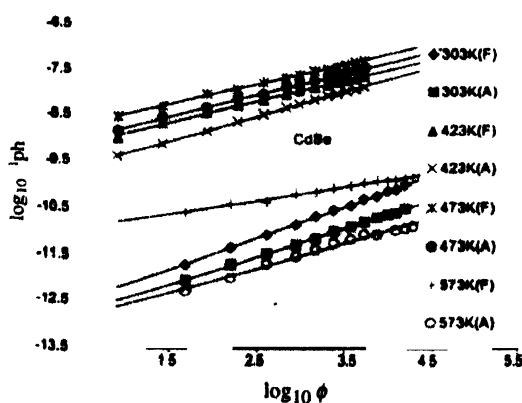


Figure 2. Variation of  $\log_{10}(I_{ph})$  vs  $\log_{10}(\phi)$  for fresh (F) and annealed (A) CdSe films deposited at 303 K to 573 K;  $I_{ph}$  being in amp and  $\phi$  in lx.

$\gamma$  value is found to increase from 0.3 to 0.5 on thermal annealing. Hence, the predominance of bimolecular recombination in these films, clearly indicates that the photoconductivity processes are essentially defect-controlled. In bimolecular recombination regime as in the present work, the density of free carriers for conduction thus exceeds the density of trapped carriers under illuminations. The lifetime of free carriers varies inversely as the photocurrent. In pure CdSe film, defects are created by the excess Cd or Se vacancies which become electron donor sites and effectively act as electron trap centres. As the light intensity is increased, most of the trapping centres get converted to recombination centres. These recombination centres shorten the lifetime of the free carriers and thus result in a sublinear behaviour [11]. Similar sublinear behaviour having  $\gamma$  value of nearly 0.5 is also found for the CdSe films when different bias voltages and ambient temperatures are applied. Such type of independence of exponent  $\gamma$  on applied bias and temperatures is in conformity with the observation of others [12]. Figure 3 depicts the same characteristics under monochromatic illuminations which also suggests predominance of bimolecular recombination process in the films. A slightly higher value of  $\gamma = 0.92$  for  $\lambda = 725$  nm is due to the maximum generation rate of photogenerated carriers. Relatively small values (0.65 and 0.78 for 400 nm and 600 nm wavelengths respectively) are due to the resultant small carrier generation because of rapid surface recombination. However, a higher value 0.87 for 975 nm wavelength compared to those for 400 nm and 600 nm is due to the additional carrier density generated from the defect levels.

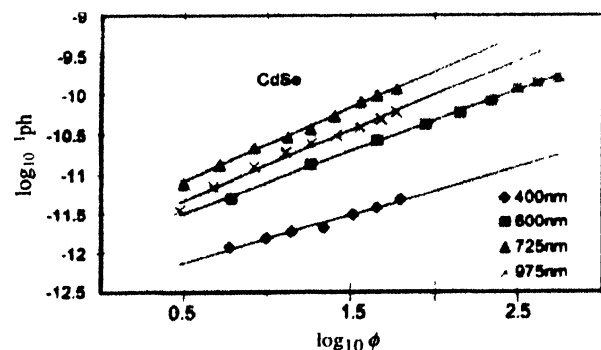


Figure 3.  $\log I_{ph}$  vs  $\log \phi$  plot under monochromatic illuminations for a CdSe film having thickness 2000 Å and deposited at 473 K;  $I_{ph}$  being in amp and  $\phi$  in lx.

### 3.3. Temperature dependence of dc photoconductivity :

A constant 24 V stable bias voltage was applied across the film when the currents in dark and under illuminations were measured during temperature variation from 303 K to 403 K. The temperature dependence of the dark and photoconductivities for a representative CdSe film deposited

at 423 K is depicted in Figure 4. The activation energies in dark ( $\Delta E_D$ ) and under illuminations ( $\Delta E_L$ ) are evaluated from the respective slopes of  $10^3/T$  versus  $\ln \sigma$  plots. It is seen that the  $\Delta E_L$  decreases over  $\Delta E_D$  when the intensity of illumination is increased. This reduction in  $\Delta E_L$  can be explained by grain boundary effect in the semiconductor thin film suggested by Petritz [13] and other workers [14,15].

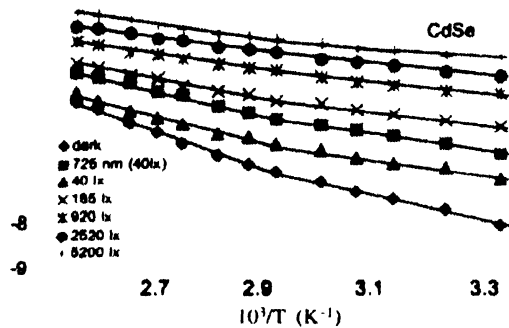


Figure 4.  $\ln \sigma$  vs  $10^3/T$  plot for CdSe film (1400 Å) deposited at 423 K,  $\sigma$  in  $(\Omega^{-1}m^{-1})$  and  $T$  in (K)

Grain boundaries in a polycrystalline film contain large number of defects arising mainly from incomplete atomic bonding which result in the formation of energy states within the band gap. These states act as effective carrier traps, and after trapping the carriers, these states become charged and thereby give rise to potential barriers. The potential barriers localized at the grain boundary, modulate the conductivity in polycrystalline films. The current carriers must pass across numerous such grain boundaries potential barriers. In the simplest case of uniform grain boundary structure with identical barriers, the conductivity can be expressed as

$$\sigma = ne\mu^* \quad (2)$$

where  $n$  is the majority carrier concentration and  $\mu^*$  is the effective mobility of the carrier modified by the grain boundary barrier as [13],

$$\mu^* = \mu_0 \exp(-e\Phi/KT). \quad (3)$$

Here,  $\mu_0$  is the carrier mobility without grain boundary effect and  $\Phi$  is the grain boundary barrier height. The term  $\exp(-e\Phi/KT)$  provides the essential characterization of a barrier. Considering one type of carrier predominating the conductivity process, preferably the electrons [3,4] in CdSe films, the dark conductivity ( $\sigma_D$ ) can be expressed as [16]

$$\sigma_D = 2e\mu_0(2\pi m^*KT/h^2)^{3/2} \exp(-\Delta E/KT) \exp(-e\Phi_D/KT). \quad (4)$$

Here,  $m^*$  is the effective mass of electron for CdSe films,  $K$  is the Boltzmann constant and  $h$  is the Planck's constant,  $\Phi_D$  is the barrier height in dark. The dark conductivity increases primarily due to increase in thermally generated

carriers and this activation process is dominated by the first exponential term  $\exp(-\Delta E/KT)$  of eq. (4).  $\Delta E$  is the activation energy due to thermally generated carriers. Since the majority carriers maintain a constant thermal equilibrium density at any temperature, the effective barrier modulation is small in dark.

When the film is illuminated by white or monochromatic light, the conductivity increases. The total conductivity under external illuminations can be expressed as [16]

$$\sigma_L = 2e\mu_0(2\pi m^*KT/h^2)^{3/2} \exp(-\Delta E'/KT) \exp(-e\Phi_L/KT), \quad (5)$$

where  $\Phi_L$  is the barrier height under illumination. At any temperature, the photoconductivity is higher than the dark mainly due to two contributing factors (i) contribution from the excess photo-generated carriers and (ii) barrier modulation by the photo-generated carriers.  $\Delta E'$  is the total carrier activation energy which includes contribution from the photo-generated carriers. It may be noted that the carrier densities are not much affected by the presence of grain boundaries but the mobilities are altered [13,14]. Therefore,  $\Delta E'$  is nearly equal to  $\Delta E$ . The excess carriers effectively interact with the grain boundary states and reduce the potential barrier under illumination given by [17].

$$\Phi_L = (Q_t - \Delta P_t)/8\epsilon\epsilon_0\Delta n. \quad (6)$$

The photo-generated carriers ( $\Delta n$ ) is greater than the density of majority carrier traps ( $Q_t$ ) localized at grain boundaries.  $\epsilon$  and  $\epsilon_0$  are the dielectric constants of the film and the medium respectively.  $\Delta P_t$  is the density of trapped minority carriers under illumination in the depletion region. The grain are expected to be partially depleted under illumination and thereby decrease the barrier height. Since  $\Phi_L < \Phi_D$ , the effective mobility of the carriers will increase under illumination ( $\mu_L^* > \mu_D^*$ ). Therefore, the total observed activation energy in dark  $\Delta E_D$  and under illumination  $\Delta E_L$  include the contribution from carrier and barrier modulation and can be expressed as

$$\Delta E_D = \Delta E + e\Phi_D \quad \text{and} \quad \Delta E_L = \Delta E' + e\Phi_L.$$

Since  $\Delta E = \Delta E'$ , the photoconductivity will mainly be governed by the change in barrier height. The reduction of  $\Delta E_L$  over dark may be attributed to the mobility activation process of the carriers that can be written as

$$\Delta E_D - \Delta E_L = \Delta E_\mu = e(\Phi_D - \Phi_L). \quad (7)$$

In fact, the mobility activation energy is equal to the reduction of barrier height. The calculated mobility activation energies ( $\Delta E_\mu$ ) for three representative CdS films deposited at 303 K, 423 K and 473 K are shown in Table 2.  $\Delta E_\mu$  increases with the increase in intensity of

Table 2. Observed activation energies in dark ( $\Delta E_D$ ), under illuminations ( $\Delta E_L$ ) and mobility activation energies ( $\Delta E_\mu$ ) of CdSe films having thickness 1400 Å

Substrate temperature (K)	Temperature range (K)	Observed activation energies in dark, under illuminations and mobility activation energies ( $\Delta E_\mu$ ) of CdSe films having thickness 1400 Å												
		Dark $\Delta E_D$	40 lx		185 lx		920 lx		2350 lx		5200 lx		725 nm (40 lx)	
			$\Delta E_L$	$\Delta E_\mu$	$\Delta E_L$	$\Delta E_\mu$	$\Delta E_L$	$\Delta E_\mu$	$\Delta E_L$	$\Delta E_\mu$	$\Delta E_L$	$\Delta E_\mu$	$\Delta E_L$	$\Delta E_\mu$
(303-404)	0.64	0.62	0.02	0.61	0.03	0.58	0.06	0.55	0.09	0.535	0.105	0.62	0.02	
(303-355)	0.23	0.16	0.07	0.13	0.10	0.12	0.11	0.10	0.13	0.092	0.138	0.13	0.10	
(355-393)	0.45	0.42	0.03	0.36	0.09	0.22	0.23	0.20	0.25	0.18	0.27	0.40	0.05	
(303-355)	0.174	0.115	0.059	0.105	0.069	0.09	0.084	0.07	0.104	0.04	0.134	0.11	0.064	
(355-393)	0.35	0.29	0.06	0.24	0.11	0.15	0.20	0.125	0.225	0.11	0.24	0.27	0.08	

illumination. However, no appreciable change is found due to 725 nm light keeping its intensity at minimum (40 lx). Buragohain and Barua [18] calculated mobility activation energies under different illuminations in thermally evaporated CdSe films which agree with the present results. Several other workers [18,19] also reported the reduction of activation energies under illuminations. The reduction mechanism is initiated by the photo-generated minority carriers which act as fast recombination centres and effectively recombine with the grain boundary states and thereby reduce the potential barrier heights.

In the present work, the entire region of temperature variation from 303 K to 403 K does not activate the intrinsic band to band transition process. The dark activation energies ( $\Delta E_D$ ) therefore, clearly signify the extrinsic region of temperature dependence of dc conductivity. In fact, the observed dark activation energies are due to the thermal activation process associated with the donor levels. Donor levels are likely to arise from the inherent defect Se vacancies in CdSe thin films [3,20]. A decrease in  $\Delta E_D$  at high  $T$ , results because of reduction in concentration of these defect levels. At higher  $T$ , CdSe film improve crystallinities with higher grain size which is confirmed by XRD and thereby reduces the grain boundary defect states.

#### 4.4. Photosensitivity :

The photosensitivity is an important parameter in characterization of photoresponse properties of thin films. A study of the dependence of photosensitivity on temperature as well as on the intensity of illumination thus may be used to explore the feasibility of application of CdSe in fabrication of photoconductive devices [21,22]. In the present work, this parameter is defined as the ratio of increase in conductivity (photoconductivity  $\sigma_{ph}$ ) in CdSe films under illumination to dark conductivity ( $\sigma_D$ ) by the relation :

Photosensitivity

$$S = \sigma_{ph} / \sigma_D = (\sigma_L - \sigma_D) / \sigma_D.$$

Using relations (4) and (5), photosensitivity can be expressed as

$$S = \sigma_{ph} / \sigma_D = [\exp\{(\phi_D - \phi_L) / KT\} - 1]. \quad (8)$$

Hence, the photoconductivity is

$$\sigma_{ph} = \sigma_D [\exp\{(\phi_D - \phi_L) / KT\} - 1]. \quad (9)$$

The photoconductivity increases rapidly in a similar fashion as dark conductivity within temperature range of 303-403 K. The magnitude of photoconductivity is modified by the photosensitivity term  $[\exp\{(\phi_D - \phi_L) / KT\} - 1]$ . Such a rapid variation of photoconductivity with temperature, clearly suggests the exponential distribution of traps in the films. Thus, the photoexcited carrier density trapped above the Fermi level is greater than the photoexcited electron density below the Fermi level which is also confirmed by the sublinear behaviour of photocurrent - light intensity characteristics found within the corresponding temperature range 303-403 K.

However, the photosensitivity  $S$  decreases exponentially with increasing the temperature in the range 303-403 K as shown in Figures 5 and 6 for two representative CdSe films which confirms the validity of eq. (8). Similar dependence

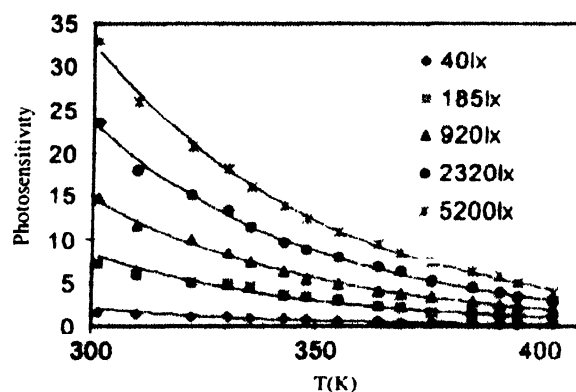


Figure 5. Variation of photosensitivity vs temperature for CdSe film (1400 Å) deposited 423 K and illuminated with white lights

of photosensitivity on temperature is also reported elsewhere in evaporated II-VI thin films [23,24].

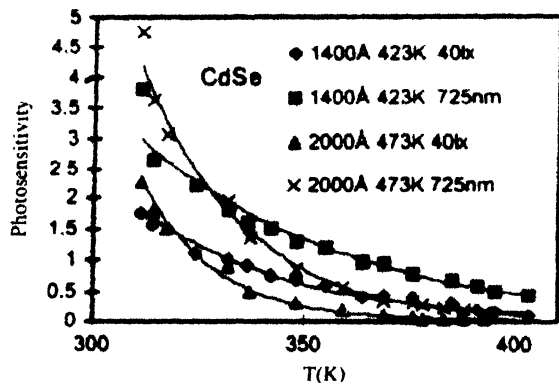


Figure 6. Variation of photosensitivity with ambient temperature when the film is illuminated with normalized 725 nm and white light 40 lx.

The photosensitivity is dependent on the effect of different types of scattering on carrier mobility. At low temperature, the grain boundary scattering is prominent. Therefore, the lowering of grain boundary potential barrier height is more at room temperature as in the present case which results maximum photosensitivity. As temperature increases, other types of scatterings arise and mainly the lattice scattering dominate over the grain boundary scattering and thereby greatly influences the carrier mobility. At high temperature region, therefore the effect of lowering of barrier height under illumination is considerably reduced causing poor photosensitivity.

It is observed that photosensitivity is more at all ambient temperatures when the film is illuminated with the threshold light of 725 nm compared to white light illumination of same intensity (Figure 6). This increase of photosensitivity is due to maximum absorption around 725 nm as found from the spectral response studies measured at room temperature.

### 3.5. Spectral response characteristics :

The spectral response characteristics of CdSe thin films deposited at 423 K, 473 K and 573 K are shown in Figure 7. All the films show a maximum photocurrent peak at 725 nm light. Similar spectral response are also found for relatively low resistivity films (Figure 8). The photocurrent peak has a tendency to increase with higher  $T_d$  and  $t$  that may be attributed to the improved crystallinity of the films. The optical band gap corresponding to this threshold wavelength 725 nm is 1.709 eV which results due to the direct transition of electrons from valence band to conduction band in these films. For shorter wavelength range  $\lambda < 725$  nm, the lights are strongly absorbed near the surface. Due to rapid surface recombination process, the photoexcitation is limited to the surface [23]. Thus, the

observed photocurrent are small when the films are illuminated with the lights of photon energies greater than

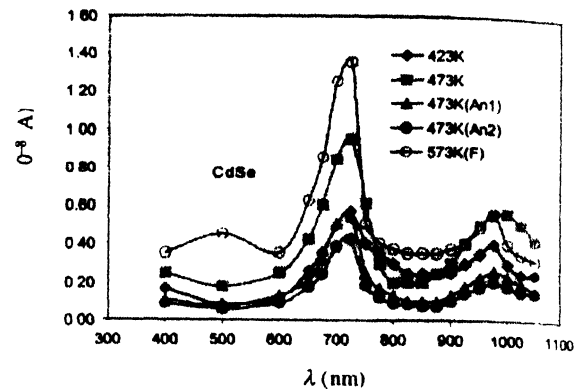


Figure 7. Spectral response of fresh (F) CdSe films deposited at 423 K, 473 K having thickness 1400 Å and at 573 K having 3000 Å. The films deposited at 473 K represent the effect of thermal annealing at 373 K for 1 hr (An 1) and 2 hr (An 2).

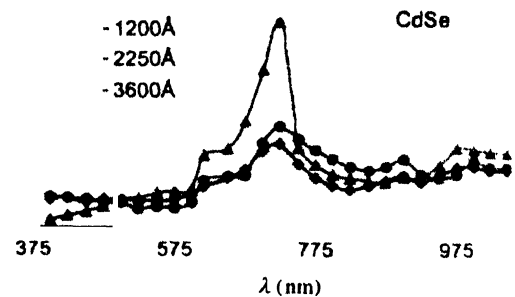


Figure 8. Spectral response characteristics of three low resistivity CdSe films deposited at 473 K having thicknesses of 1200 Å, 2250 Å and 3600 Å.

absorption edge. It is observed that a shoulder around 975 nm light is quite prominent in all the films. This shoulder may be due to the additional absorption at the Se vacancy [20] which is estimated nearly as 0.45 eV. In the present work, the existence of this level is in agreement with the dark activation energy found from the measurement of temperature-dependence of dark conductivity.

The magnitude of photocurrent in fresh CdSe film is found to decrease after thermal annealing in vacuum as shown for the representative films in Figures 2 and 7. This decrease may be due to the reduction of shallow states mainly arising from interstitial cadmium atoms and voids formed in the films [3]. The interstitial cadmium is singly ionized at room temperature and can contribute to the photocurrent. However in the thermal annealing process,  $Cd^+$  ions are expected to diffuse along the grain boundaries that may lead to the reduction of carrier density [25]. The concentration and size of voids also decrease on annealing. These cause a decrease in absorption coefficient [26] and thereby reduce the photocurrent.

#### 4. Conclusions

Thermally evaporated CdSe thin films deposited at 473 K to 623 K are polycrystalline having hexagonal ZnS type structure. All polycrystalline films show the preferential orientation of crystallites along [002] direction. The film crystallinity increases with  $T_s$ , which is attributed to the enhancement of grain size and a decrease of microstrain developed in the film.

The photocurrent versus light intensity characteristics of CdSe films show a sublinear behaviour that is independent of  $T_s$ , applied bias and temperature and thickness. Sublinear nature clearly indicates the defect control photoconductivity in CdSe thin films governed by the exponential distribution of traps which agrees well with the exponential rise of photoconductivity with temperature in the range 303–403 K. The photosensitivity is maximum around 303 K which is due to prominent grain boundary scattering whereas it decreases exponentially on increasing temperature because of onset of other types of scattering (mainly the lattice scattering). The total observed activation energies include the contributions both from carriers and barrier modulation process. Activation energy under illumination, decreases over dark on increasing the intensity of illumination. This decrease is attributed to the mobility activation process governed by the lowering of grain boundary potential height. The spectral response studies reveal that the CdSe films show the direct optical transition at 725 nm and an additional excitation due to the impurity around 975 nm. A decrease in photocurrent on thermal annealing is due to the diffusion of Cd<sup>+</sup> ions along the grain boundaries as well as the reduction of concentration and size of voids formed in the films.

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