Research Article

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Investigation on Thickness Dependent Structural, Optical and Electrical Properties of Cadmium Selenide Thin Films Deposited *via* Thermal Process

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ABSTRACT

Nanocrystalline CdSe thin films of various thicknesses have been deposited on cleaned glass substrates using thermal vacuum evaporation technique at substrate temperature of 333K. The effect of thickness on the structural, optical, dielectric, and electrical properties of CdSe thin films has been studied. Structural studies by X-Rady Diffraction (XRD) reveal that the deposited samples are polycrystalline. Optical investigation using ultraviolet visible (UV-Vis) spectroscopy shows that the films exhibit direct allowed electronic transitions, and the optical absorptions are found to shift towards lower energies with increasing thickness. The optical energy band gap (E_g) decreases from 2.36 eV to 2.12 eV as the sample thickness increases, attributed to the rise in the grain size of the CdSe nanoparticles. The fundamental dielectric constant decreases with an increase in film thickness, and the imaginary dielectric constant increases with increase in film thickness. The increase of photonic energy increases both real and imaginary dielectric constants. External illuminating radiation generates photocarriers that interact with the grain boundary state and reduce potential barrier.

Keywords: CdSe thin films; Band gap energy; Photo carriers; Dielectric constant; Potential barrier; Photoconductivity.

1. INTRODUCTION

Binary CdSe semiconductor belongs to the II-VI group of semiconductors, and in recent years it has attracted enormous interest amongst researchers because of its size-dependent electrical, structural, and optical properties (Hodes, 2003; Woggon, 1997). Nanocrystalline CdSe semiconductor has been able to draw more and more attention because of its unique fundamental (Wang and Zunger, 1996), experimental, and applied potentialities. The fundamental properties of nanocrystalline CdSe compound semiconductors reflect the size dependent optoelectronic and optical absorption, such as red shift absorption spectra, distinct wurtzite or zinc blende crystal structure, surface properties, quantum confinement, etc. CdSe possesses a direct intrinsic optical band gap of 1.70 eV and often has n-type conductivity in bulk as well as thin film form (Shreekanthan *et al.* 2003). Semiconducting CdSe thin films developed at high temperatures are usually polycrystalline (nanocrystal form) and such nanocrystals find usefulness in the manufacture of devices such as PV cells (Li *et al.* 2021), electronic devices (Olusola *et al.* 2015), light-emitting diodes (Park *et al.* 2004), and other nanoscale devices (Cai *et al.* 2006). CdSe nanocrystalline films have been synthesized by various techniques such as vacuum evaporation (Ramaiah *et al.* 2001), chemical deposition technique (Perez *et al.* 2008; Nemec *et al.* 2000), sputtering, spray pyrolysis, physical vapour deposition, electrodeposition, etc. (Rouleau and Lowndes, 1998; Cachet *et al.* 2000), substrate vibration assisted drop casting method (Sachdeva *et al.* 2020), inert gas condensation (Sharma *et al.* 2018), etc. CdSe is widely preferred in the manufacture of various optoelectronic devices because of its high photosensitive character (Mahato and Kar, 2015). A shift in the band gap is found to affect different optoelectronic properties of the films, and the optical behavior of semiconducting CdSe thin film is usually dependent on structure. The structure of CdSe thin films relies on several key factors, including the rate of deposition, substrate temperature, film thickness, and the order of vacuum. Optical absorption arises due to interactions of the excited electrons with the lattice perturbed by imperfections or vibrations. The thermal vacuum evaporation deposition technique is suitable because, through this method, it is possible to change the deposition parameters and develop uniform films of desired material structures and characteristics (Baban *et al.* 2005). The analysis of dielectric properties of nanocrystalline CdSe compound semiconductor enables in characterizing the capability of storing electric charges in it so that the thickness and optical absorption depend on dielectric constant of the semiconductor which is important for the purpose of fundamental as well as technological consideration. Illumination has a direct impact on the optoelectronic properties of compound semiconductors, particularly CdSe. Illuminating the CdSe

thin films with various white light intensities at different temperatures yields interesting results, including trapping. Influence of illumination on CdSe films can cause a vast change in mobility activation energy and trap depth length at various ambient conditions. In the present work, the preparation of CdSe thin films adopting the thermal vacuum evaporation method and a study on the thickness dependent structural, optical, dielectric, and electrical behaviour of CdSe films have been reported.

2. EXPERIMENTAL DETAILS

CdSe thin films have been developed onto thoroughly cleaned glass substrate, maintaining a vacuum of 10^{-6} Torr (base pressure). The vacuum coating unit HINDHIVAC was used to deposit the thin films. High purity CdSe powder (about 99.99%) was obtained from Koch Light Lab, U.K. To prepare thin films of CdSe, the sublimation process was adopted. In sublimation process, CdSe powder was directly evaporated (without melting) for the preparation of thin film. In this process high growth rate of CdSe thin films within confined glass chamber in high vacuum was obtained. Tantalum was used as a boat material to evaporate the samples because of its low partial pressure, which goes up to the material's evaporation temperature. The material's sublimation temperature was attained by adjusting the heating current within the 40-60 A range at a 10 V filament voltage. The distance between the source and the substrate was maintained at 6.6 cm. The interferometric method (Fizeau's method) (Tolansky, 1960) was used to measure the thickness of a deposited film. The deposited films with thickness 1759 Å, 2946 Å and 3869 Å exhibited a corresponding deposition rate of 4.89 Å/sec, 8.18Å/sec and 11.72 Å/sec, respectively. Absorption coefficient was estimated from the transmitted and reflected monochromatic radiation, received from the CZ metal interference filter (range 333 to 1050 nm). Transmitted and reflected monochromatic radiation from the sample was measured by using an Aplab luxmeter (model 5011S). Absorbance and transmittance spectra were measured using a UV-VIS spectrophotometer (SHIMADU 1800) in the wavelength range 300-1100 nm. For measuring photocurrent, an ECIL electrometer amplifier with an input impedance of $10^{14}\Omega$ (and more) was employed. To provide bias, a series of dry cells having 9 V emf each was used. A continuously evacuating glass jacket was designed to suspend the samples in it by using thin enameled copper wire. To avoid pick-up noise, a Faraday case was suitably fabricated inside. A tungsten halogen projector lamp (250 watts, operated at 24 V) was employed for white light illumination. A luxmeter (APLAB) was used to measure illuminating intensities. An electrical heater was used to heat the samples. A copper constant thermocouple thermometer was used to measure the corresponding sample temperature.

3. RESULTS AND DISCUSSIONS

The structural properties of CdSe were investigated using XRD spectra. The XRD patterns of nanocrystalline CdSe thin films prepared at a substrate temperature of 333K are shown in Fig. 1. The well-defined diffraction peaks correspond to (002), (101), (110), and (112) planes for both the samples, having thicknesses of 2946Å and 5164Å, respectively. A smaller number of XRD diffraction peaks are observed for the sample of lower thickness. The XRD spectra showed that the CdSe samples are polycrystalline, having wurtzite (hexagonal) ZnS structure, and the grain sizes of the films are strongly affected by the grain boundaries (Inbanathan *et al.* 2021). For the polycrystalline material, grain boundary is the interface between two grains. The enhancement in grain size of the samples is attributed to the increase in film thickness, which is very distinctly shown by the intense peaks in the diffraction spectra. With the rise in the film thickness, the diffraction intensity increases because of the development of the materials incorporated in the process of diffraction. The film, having more thickness, has a larger grain size, and it provides sharp, intense diffraction peaks (Nair *et al.* 1993).

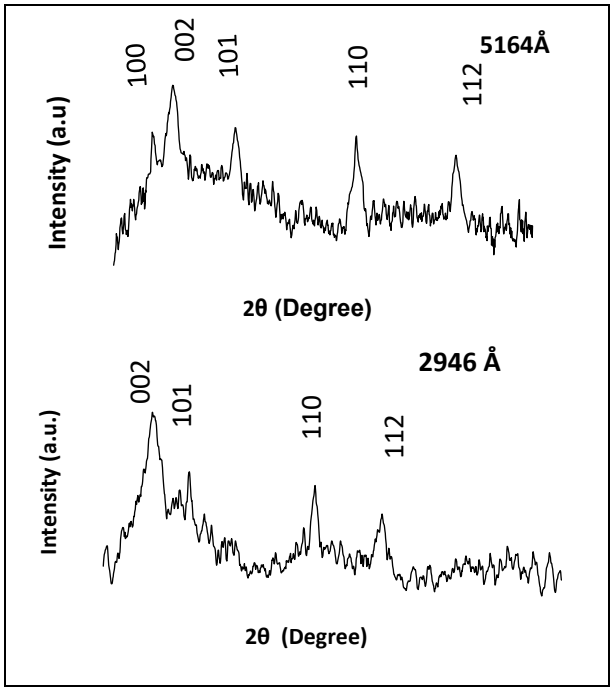


Fig. 1: XRD pattern of CdSe films deposited at 333K, having thicknesses of 2946Å and 5164Å

The thickness-dependent grain size is shown in Fig. 2. The figure clearly shows a rapid increase in crystallite size with the rise in thickness of the sample (Nasir *et al.* 2019). This occurs because an increase in film thickness allows more grains to form on the surface of the smaller grains. It is also observed that with further enhancement in film thickness, the thickness dependence of crystallite size becomes less.

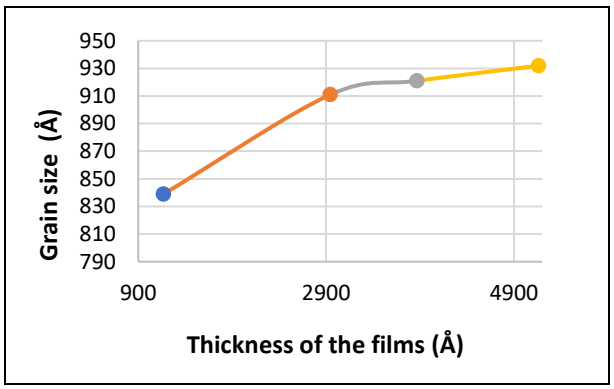


Fig. 2: Plot of average grain size vs thickness of pristine CdSe having thicknesses 1759Å, 2946Å, 3869Å, and 5164Å

Fig. 3 shows the plot of grain size obtained for the (002) hexagonal phase for annealed CdSe samples of thickness 1759Å, 2946Å, and 3869Å. From the plot, it can be concluded that the parameters undergo tensile stress arising from the internal stress of the crystallites. The phenomenon is attributed to the lattice mismatch and variation in thermal expansion coefficients (Chopra, 1969).

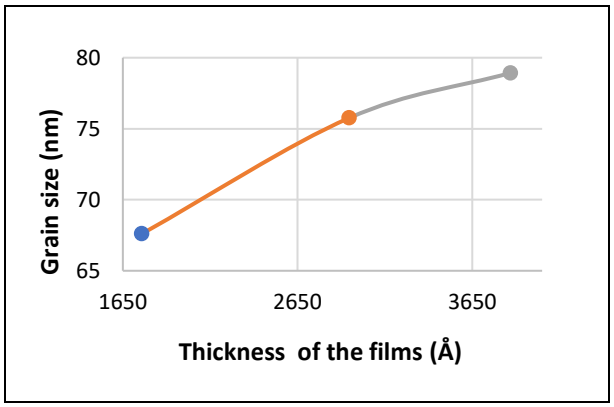


Fig. 3: Thickness dependence of grain size of (002) hexagonal annealed CdSe having thickness 1759Å, 2946Å, and 3869Å

Optical absorbance of the sample was estimated from the transmission spectra using Swanepoel's method. Absorbance (A) and the absorption coefficient (α) are related by the relation (Pankove, 1971)

$$\alpha = 2.303\frac{A}{t} \quad \ldots (1),$$

where t represents the thickness of thin films. The higher values of the absorption coefficient are related to the transitions of electrons among extended states (both in conduction and valence bands). Absorption coefficient (α) is a function of photon energy, $h\nu$, and in general can be written as (Tauc, 1972)

$$(\alpha h\nu)^r = B(h\nu - E_{gd}) \quad \ldots (2),$$

where E_{gd} is the optical band gap energy, B is a characteristic parameter that doesn't depend on photonic energy. r is a constant dependent on the transition nature occurring between valence and conduction bands (Kazmerski, 1980), for direct and allowed transition r=2. The plots of $(\alpha h\nu)^2$ versus $h\nu$ are observed to be linear over a wide range of photon energies, which suggests transitions for CdSe nanocrystalline thin films are direct transition type (Sadekar *et al.* 2015). The extrapolations (intercepts) of $(\alpha h\nu)^2$ versus $h\nu$ plots on the energy axis provide the optical energy band gaps (shown in Fig. 4).

It is observed that with the increasing film thickness, the absorption edge shifts towards the lower photonic energies, which is attributed to stoichiometry and a change in the grain size. The estimated band gap energies of prepared CdSe nanocrystalline thin films, which exhibit a dependence of band gap on the thickness of the films, are shown in Table 1. Fig. 4 clearly shows that the band gap decreases with an increase in the thickness of the film, and it matches the investigation by earlier workers (Dhanam *et al.* 2008; Choudhary and Chouhan, 2019). The observed shifting of the band with the film thickness may be due to the presence of the quantum confinement effect. The increase in grain size, decrease in strain and dislocation density, reduction of trapping centres, and improvement in film microstructure are responsible for the decrease in optical band gap energy of the samples prepared (Bube, 1960).

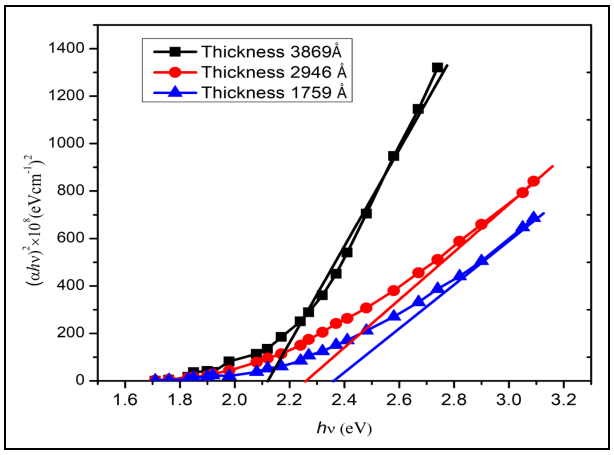


Fig. 4: Plots of $(\alpha h\nu)^2$ vs. $h\nu$ of CdSe films having thicknesses 1759 Å, 2946 Å & 3869 Å, deposited at 333K

Table 1. Band gap energy (E_{gd}), average grain sizes of CdSe films of different thicknesses generated at 333K and annealed at 400K

Film thickness (Å)	Deposition Temperature (K)	E_{gd} (eV)	Grain Size (nm)	Absorption Coefficient (α) (cm^{-1}) x $10^{\wedge 5}$
1759	333	2.36	70.6	0.54
2946	333	2.26	77.7	0.45
3869	333	2.12	79.9	0.36

The dielectric constants (ε_r) and (ε_i) are important optical parameters, and they are related to the refractive index and absorption coefficient (Lucas, 2004)

$$\varepsilon_r = n^2 - K^2 \quad \ldots (3)$$

$$\text{and } \varepsilon_i = 2nK \quad \ldots (4)$$

Plots of real and imaginary dielectric constants with photon energy, hv, as a function of the thickness of the film of the prepared CdSe films are shown in Fig. 5 and Fig. 6, respectively. Fig. 5 exhibits that the fundamental part of the dielectric constant (ε_r) rises slowly up to 2.12 eV and then increases steeply up to 2.58 eV. From there, it again rises slowly with further increase of photon energy. The direct relation between the thickness of CdSe films and their fundamental dielectric constant is such that the effective real dielectric of a thin film, as simulated by Lorentz's local field theory, decreases as the film's thickness decreases. It is due to a reduction in the local fundamental dielectric constant on the film surface because of the absence of an enforcing local electric field induced by the nearest dipoles in the adjacent layers, for which the effective real dielectric content decreases (Zubair *et al.* 2019; Natori *et al.* 1998).

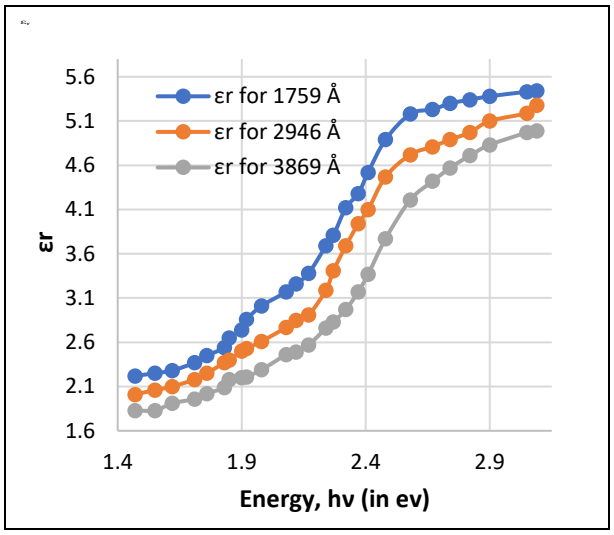


Fig. 5: Photon energy vs. real dielectric constant plot of CdSe films having thickness 1759Å, 2946Å, and 3869Å

The imaginary part of the dielectric constant (ε_i) signifies the radiation absorption by free carriers. The characteristics of the imaginary part of the dielectric constant (ε_i) with hv as a function of the thickness of the film are shown in Fig. 6. From the figure, it is observed that ε_i rises smoothly up to 1.7 eV, and then it again starts to fall slowly. It is observed that ε_i depends on the thickness of the film and it increases with the increase of the film thickness.

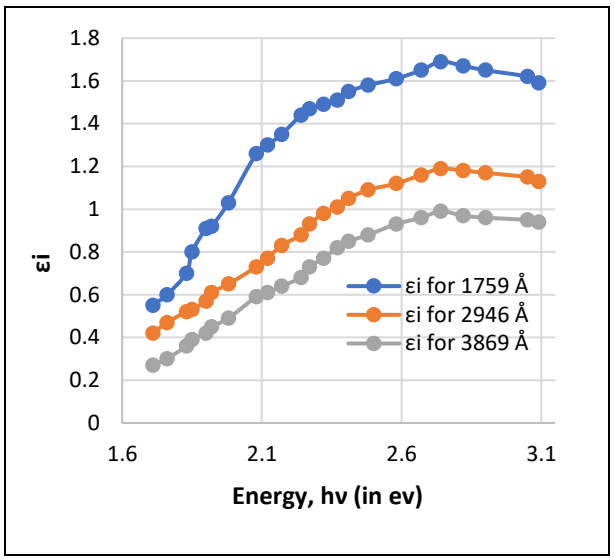


Fig. 6: Photon energy versus imaginary dielectric constant plot of CdSe having thicknesses 1759Å, 2946Å, and 3869Å developed at 333K

Excess carriers (photo generated) of photosensitive CdSe samples under the illumination of external radiation in a given interval of time are

$$\frac{d}{dt}(\Delta n) = \alpha\eta\psi - (\Delta n/\tau) \quad \ldots (5),$$

where α is the light absorption coefficient, η, ψ, and τ denote quantum yield, light intensity, and photocurrent decay time, respectively. The samples generate potential barriers (Φ_b) at grain boundaries of the samples, given as (Sarma and Das, 1991)

$$\Phi_b = \frac{(Q_t - \Delta P_t)}{8\varepsilon\varepsilon_0\Delta n} \quad \ldots (6),$$

where Δn is the photo-generated carriers, ε and ε_0 denote respectively the dielectric constants of the sample and free space, ΔP_t represents the density of trapped minority under illumination.

The plots of Fig. 7, clearly show that σ_{ph} is proportional to ψ (irrespective of temperature) and justifies the equation (5). Under illumination, excess carriers (photo-generated) undergo interaction with the grain boundary and decrease the potential barriers. The decrease of potential barrier (Φ_b) increases photoconductivity (σ_{ph}) as (Petriz, 1956)

$$\sigma_{ph} = \text{Cexp}\left(\frac{-q\Phi_b}{KT}\right) \quad \ldots (7),$$

where C is a constant, by correlating the relations of σ_{ph} and N (where $N = \sigma_{ph}/e_\mu$), it is found that Φ_b is inversely proportional to ψ, as per equation (7).

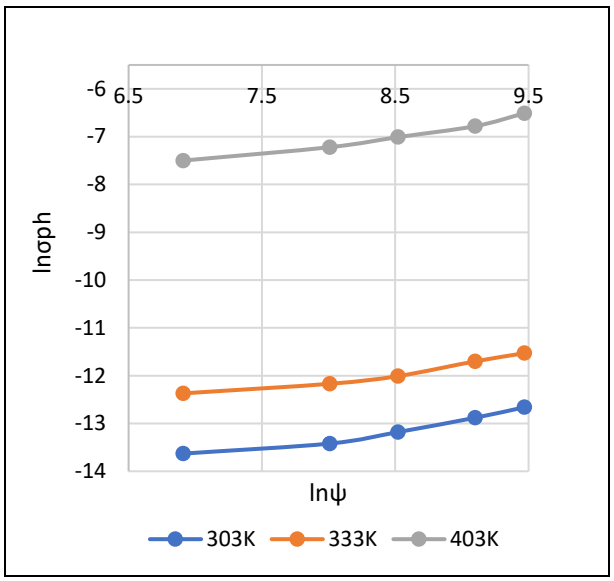


Fig. 7: Plots lnψ vs. lnσ_{ph} at ambient temperatures 303K, 333K, and 403K

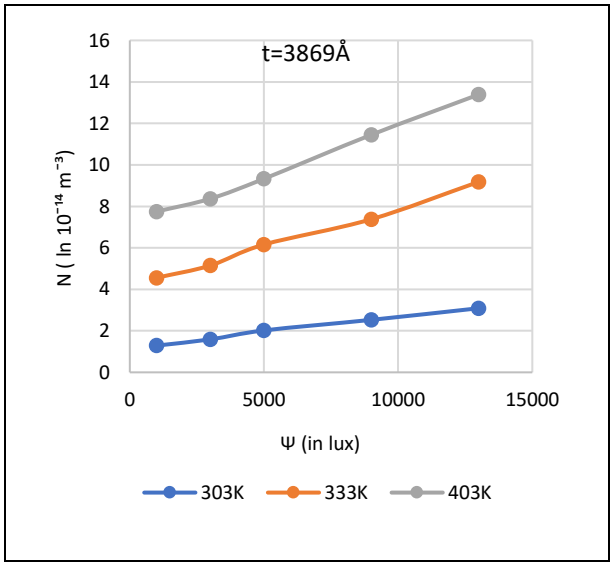


Fig. 8: Plot of intensity of illumination (ψ) and density of state (N) of CdSe film of thickness 3869Å at various temperatures

Electrons and holes are generated (photo-generated) because of the absorption of illuminating radiation. One portion of the generated carriers recombines with their opposite charges (which are localized at the grain boundary deflection layer). As a result, the grain boundary potential barriers, Φ_b, decrease (Das *et al.* 2018), and the remaining part of the carriers contributes to the photoconduction process. The reduction in grain boundary potential barrier enhances effective mobility (μ*) of carriers. Moreover, the enhancement in illumination intensity produces more carriers and, as a consequence, the effective mobility of the carriers is increased. The effective mobility of the sample is given as (Mort and Pai, 1976).

$$\mu * = \mu_0 \exp\left(-\frac{q\Phi_b}{kT_0}\right) \quad \ldots (8).$$

Here, T_0 denotes the characteristic temperature of the sample, μ_0 represents the carrier mobility in the absence of barrier potential effect.

Under illumination, at comparatively low temperature, the number of occupied energy levels (n) in the conduction band is identical to that of the effective density of state (N). The photoconductivity depends on the effective density of state ($\sigma_{ph} = Nq\mu^*$). But photoconductivity rises with the rise in intensity of illumination. This shows that the effective density of state (N) is dependent on the intensity of lighting, Ψ. Fig. 8 shows the linear increase of N with intensity of illumination, Ψ, which justifies the relation between N and ψ.

4. CONCLUSION

The XRD study done on CdSe thin film deposited *via* thermal evaporation technique reveals that the film samples are polycrystalline with hexagonal (wurtzite) zinc blende structure. The grain size of the films are strongly affected by the thickness of the films such as thicker films bear higher grain size. From the optical absorption spectra, an allowed direct transition is found for CdSe films and decreases with the increase of thickness. It allows strong optical absorption which is the essential property for fabrication of solar cells and other optoelectronic devices. The dielectric studies of the films lead to a decrease in real dielectric constant with the increase of the film thickness. This is a very important property of the dielectric for fabrication of ferroelectric and photonic devices. Both the real and the imaginary dielectric constants are found to increase with the enhancement of photonic energy. The annealed CdSe film samples of various film thickness undergo tensile stress along the plane parallel to the substrate surface, which is due to lattice mismatch, internal stress on crystallites, and thermal expansion coefficient differences. Photoconductivity of CdSe films (thermally deposited) occurs because of the increase in photo-generated carriers and an increase in effective mobility. The trapping centers of CdSe films control the photocurrent. Trapping centres have a direct impact on the decay time of photocurrent, and each multi-depth trap corresponds to a respective decay time of photocurrent.

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CONFLICTS OF INTEREST

The authors declare that there is no conflict of interest.

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