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Nd:YAG Laser Annealing: Impact on the Photoconductive Properties of CdS Thin Films

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A R T I C L E I N F O

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A B S T R A C T

This study investigates the effects of pulsed Nd:YAG laser annealing at a wavelength of 532 nm on the photoconductivity properties of cadmium sulfide (CdS) thin films prepared by thermal evaporation. In addition, measurements at room temperature showed an ohmic behavior in the voltage-current characteristics of the CdS thin films. It was observed that after laser irradiation, the photosensitivity of the film increased due to the improved crystallinity and decreased defect density of the thin films, as shown by the increase in the ratio of light to dark current (I_{ph}/I_d) from 0.35 to 0.42. The photocurrent also follows the relationship ($I_{ph} \propto F_{\gamma} I$), with γ-values of 1.037 and 1.047 after annealing due to monomolecular recombination, reduced grain boundaries and enhanced recrystallization. The spectral response peaked at 585 nm, which corresponds to the optical band gap of the CdS thin film. The transient photoconductivity, which describes the time-dependent change in the electrical conductivity of the material when exposed to light, was measured and showed significantly increased decay rates. The differential lifetime (τ_d) decreased from (90.8 sec) to (39.2 sec) after Nd:YAG laser annealing, which can be attributed to a lower density of defect states and an improvement in film quality. The results highlight the ability of Nd:YAG laser annealing to maximize the photonic and electronic properties of CdS thin films through structural and carrier recombination dynamics, increasing their use in optoelectronic devices.

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1. Introduction

Metal chalcogenide semiconductors (MCS) are recognized with distinctive characteristics, such as high radiation sensitivity, tuneable band gaps, and thermal and chemical stability, which make them perfect for applications such as photodetectors, solar cells and memory storage. Therefore, they have attracted significant attention because of their further applications in electro-optics, electronics, and solar energy conversion [1, 2]. MCSs are widely popular and highly valued due to their exceptional electronic qualities and advanced manufacturing methods, which enable precise control over their adaptability and features for many uses. Such manufacturing processes include spray thermal decomposition, vacuum thermal evaporation, DC plasma, pulsed laser, and vapour deposition [2-7]. By optimizing the crystal structure, it is possible to customize and improve the electrical characteristics of chalcogenide semiconductors [7]. Understanding the consequences of laser irradiation on the

electronic processes in amorphous semiconductors and investigating charge carrier transport [8] depends on the interaction of a laser beam with such materials. Adding thermal or metal annealing, which changes their structure and optical and electrical characteristics, could transform amorphous to crystalline chalcogenide materials [9].

Because of its optical transparency, high quantum efficiency, direct band gap, good electrical conductivity, and enormous exciton energy, cadmium sulfide (CdS) is one of the significant II-VI compounds semiconductors used practically in many optoelectronic device applications, including solar cells, light-emitting diodes, photoconductors, and thin film transistors [10, 11]. Usually forming a hexagonal (wurtzite) structure, CdS could adopt a cubic form once doped with zinc [12]. Thermal annealing is a commonly used technique to improve semiconductor structural, electrical, and optical characteristics [13-15]. Nevertheless, the thermal annealing technique's long processing periods and high energy consumption encourage researchers to investigate alternative methods [16]. One alternative method is laser annealing, which was observed by researchers for its capacity for addressing such challenges through localized and rapid heating with minimal energy loss, enabling exact structural modifications and structural changes through adjusting parameters including power, wavelength, and number of pulses [17-20].

Previous work has shown laser annealing to enhance optical characteristics in CdS thin films, change crystal structure, and increase crystallite size. With the use of a Nd:YAG laser (532 nm), Khudiar *et al*. [20], for example, observed a crystallite size increase from 25.91 nm to 103.69 nm and a structural shift from cubic to hexagonal after annealing the CdS thin films. Their observations included a redshift in the absorption spectrum and a reduction in the optical band gap from 2.42 eV to 2.31 eV post-annealing; such redshift corresponds with the increase in crystallite size since larger crystals tend to lower the bandgap, so improving light absorption in the visible and near-infrared regions. Laser annealing or thermal heating techniques allow materials such as CdS to shift from cubic to hexagonal structure utilizing increased thermal energy, causing the atoms to reorganize into a more stable hexagonal structure. The change from cubic to hexagonal structure is crucial since it enhances optical and electrical characteristics, such as lowering the bandgap and raising charge carrier mobility, improving electronic device performance. This work examines the effects on the photoconductivity, intensity dependence of photocurrent, spectral response, and transient photoconductivity measurements of CdS thin films of Nd:YAG laser annealing with 5-minute irradiation durations.

2. Experimental

2.1 Materials

Cadmium sulfide powder was deposited on a glass substrate under thermal vacuum evaporation and at a vacuum pressure (2×10^{-5} mbar). The substrates were carefully cleaned prior to the deposition technique to ensure optimal adhesion and quality of the thin films. The cleaning process consisted of a 10-minute ultrasonic cleaning after a successive rinse with deionized water, acetone and isopropyl alcohol. After cleaning, the substrates were dried under nitrogen gas and treated with UV to remove final organic contaminants. The film was left in the deposition chamber for 24 hours to reach a quasi-stable equilibrium. This phase before further treatment was important to ensure the chemical and thermal stability of the film and to prevent contamination.

2.2 Nd; YAG Laser Annealing

The thin film was annealed by a pulsed Nd:YAG laser (532 nm wavelength, 2 mJ energy, 5 ns pulse duration) for 5 min irradiation. The size spot diameter of the laser was 5 mm, with a repetition rate of 1 Hz. The film was uniformly exposed to the laser beam, ensuring consistent annealing across the surface. The laser beam was precisely directed on the film surface.

2.3 Characterization

Photoconductivity experiments were carried out to assess the impact of laser annealing on the electronic and optical properties of CdS thin films, while light conductivity measurements were undertaken utilizing photocurrent intensity and spectral response. Before and during exposure to the Nd:YAG laser annealing process, measurements were made of the cadmium sulfide thin films. A Horiba Uvisel ellipsometer (Horiba, NJ, USA) with a wavelength range of 250–800 nm and a measurement accuracy of ±5 nm was used to quantify the films' thickness. With the use of a mask on the substrate glass, the thin film diameter was measured to be 5±1 mm, and the film thickness

was 200±5 nm. The thickness was verified at multiple points across the film to ensure uniformity. To study the changes in photocurrent as a function of incident light wavelengths on CdS thin film before and after Nd:YAG laser annealing. Four different filters (535, 585, 615, and 660 nm) were used in this study. Each measurement was repeated under the same conditions multiple times to ensure the reproducibility of the experiments.

3. Results and Discussion

3.1 Photoconductivity Measurements

The impact of Nd:YAG laser annealing on the photoconductivity of CdS thin films has been investigated in this study. **Fig. 1** displays the dark current and photocurrent before and after five minutes of Nd:YAG laser annealing. **Fig. 1** shows that the photocurrent value is less than the dark current value. The interplay between photoexcitation and thermal excitation mechanisms explains this photoconductivity phenomenon. Particularly, the degree of photoexcitation is less than that of thermal excitation, which results in a limited generation of free carriers.

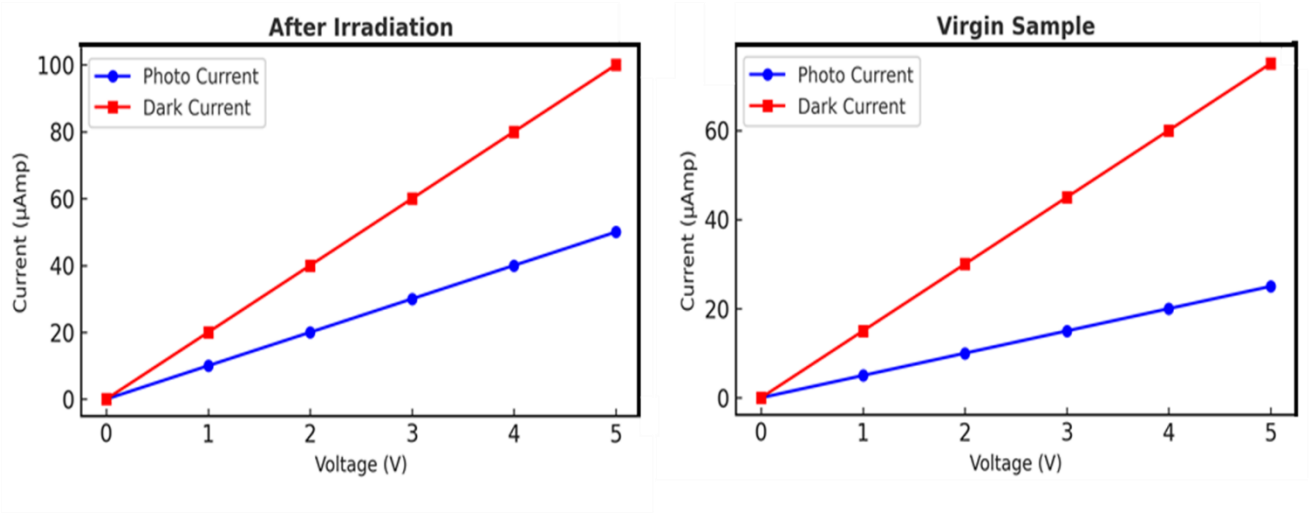

Figure 1: Comparison of dark current and photocurrent for CdS thin films before and after annealing with an Nd:YAG laser.

Furthermore, trap states inside the bandgap are essential in capturing and releasing charge carriers, affecting the general conductivity. By serving as temporary storage for photo-generated carriers, such trap states help to lower the photocurrent contribution. Moreover, carrier recombination dynamics, including radiative and non-radiative recombination events, influence photoconductivity. The observed result corresponds with a rise in photocurrent, so enhancing trap state density and reducing recombination pathways seems to improve photoconductive characteristics. Two separate energy regions comprise the energy gap in materials: one is close to the valence band, while the other is between the Fermi level and the conduction band. High electron and hole capture cross-sections correspond to the second case, in which electrons are captured from the conduction band and holes from the valence band. This results in a decrease in charge carriers in the conduction band and, consequently, a reduction in current when exposed to radiation [21, 22].

The absence of maxima in the photocurrent has been predicted in noncrystalline semiconductors if the photo-generated carrier densities are shorter than the dark current ($I_{ph} < I_d$) at room temperature. This result agrees with that reported by other researchers [23-25], the observation that the dark current is more than the photocurrent in glassy chalcogenides. Also, from **Fig. 1**, it was observed that the photocurrent increased twice, and the dark current decreased after laser annealing; this is due to a decrease in grain boundaries after an increase in the crystallite size, as confirmed by the XRD and SEM results in our previous study [13, 20]. Therefore, the photosensitivity is increased due to an increase in the photocurrent and a decrease in the dark current. Kushwaha *et al*. [26] have reported that cadmium selenide thin film's photosensitivity increases with grain size. By lowering the grain boundary potential barrier height, this procedure improves the films' overall photoconductivity. According to Khudiar *et al*. [16], this behaviour may be caused by the increased strength of the new bonds in the strand tails or by an expansion in the bandwidth of local states brought on by laser annealing. Increased carrier mobility due to

partial crystallization is linked to the increase in photocurrent. According to FTIR observations, pulsed laser annealing creates new chemical bonds between comparable atoms, such as Cd-Cd and S-S [1, 16]. One of the crucial factors in photo-conductivity studies is the photosensitivity (I_{ph}/I_d) of semiconductor thin films, which depends on the chalcogenide atoms' electrons and holes. Covalent and van der Waals bonds are formed by sulfur atoms in typical chalcogenide glasses. Because van der Waals bonds are polar and weaker than covalent bonds, they undergo more significant modifications when exposed to light [16-18].

3.2 Intensity Dependence of Photocurrent Measurements

The dependence of photocurrent (I_{ph}) on light intensity (F) has been studied for CdS thin films at room temperature, both before and after Nd:YAG laser annealing, as shown in **Fig. 2**. The resulting curves are nearly straight lines, indicating that I_{ph} follows a power-law relationship with intensity, expressed by Eq. (1) [20, 27]:

$$I_{ph} = F^{\gamma} \qquad (1)$$

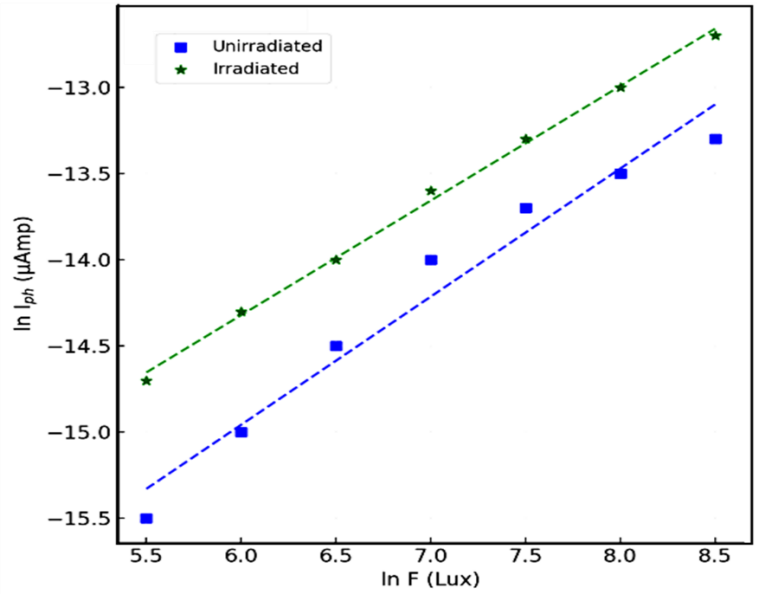

Figure 2: Logarithmic plots of photocurrent (Iph) as a function of light intensity (F) for CdS thin films, comparing results before and after Nd:YAG laser annealing.

The power exponent, γ, and the intensity, F, determine the recombination mechanism. The molecular recombination process is indicated by γ ~ 0.5, but the mechanism itself is marked by γ ~ 1.0 [27, 28]. According to Crandall [27], a continuous distribution of traps within the band gap is necessary to explain a value of γ between 0.5 and 1.0, which cannot be explained by the assumption of a set of discrete energy levels. The photocurrent in the current investigation was found to fluctuate in proportion to the square root of the visible light intensity [29]. **Table 1** provides the values of γ, which are derived from the slope of the curves in **Fig. 3**. At room temperature, the values of γ are around 1.037 and 1.047, respectively, which could be regarded as a monomolecular recombination for the CdS thin film before and during laser annealing.

Table 1: Parameters of photo-electrical before and after laser annealing for 5 min of CdS thin film.

Sample	I_{ph}/I_d	Γ	τ_d (sec)
Un annealed	0.35	1.037	90.8
Annealed	0.42	1.047	39.2

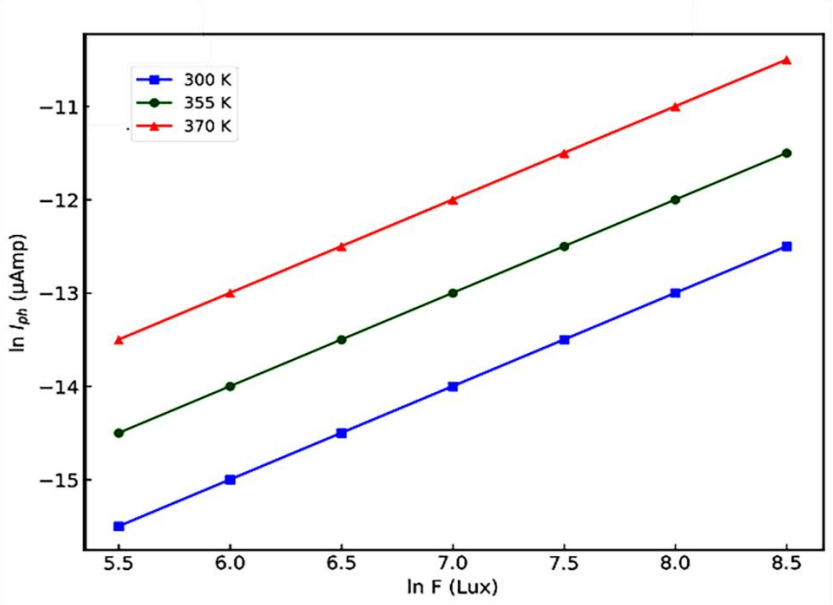

Figure 3: Logarithmic plots of photocurrent (I_{ph}) versus light intensity (F) for CdS thin films measured at various temperatures.

The value of γ remains nearly unchanged after laser annealing, confirming monomolecular recombination. Monomolecular recombination indicates that photo-generated carriers are lower than the dark current and recombine into a pool of thermally generated recombination centres, giving rise to monomolecular recombination [29, 30].

Studies on the photocurrent's intensity (F) dependence in CdS thin films at varying temperatures are also being conducted. The results in **Fig. 3** indicate an increase in intensity with increasing temperature in the full range (300-370 K). The curves of ln I_{ph} versus ln F appear to form almost straight lines, indicating that the photocurrent closely follows the power law. The potential γ is equal to unity for all thin films, indicating a monomolecular type of recombination. Depending on the Simons and Taylor model [31], these results can be explained by the fact that at high temperatures or low illumination levels, the quasi-Fermi levels of trapped electrons and holes are fixed in position at their equilibrium values, causing the thermal carrier concentration to exceed the photo-carrier concentration in High temperatures, which leads to an increase in the photocurrent with increasing temperature and is proportional to F and I_{ph} [32, 33]. When the temperature rises, the demarcation levels of holes and electrons are expected to increase towards the band tails [34].

3.3 Spectral Response of the CdS Thin Films

Fig. 4 illustrates the variation in photocurrent with incident light wavelengths for CdS thin films before and after Nd:YAG laser annealing. Four different filters (535, 585, 615, and 660 nm) were used in this study. Each of these curves shows a broad peak, with the peak-response wavelength for the CdS thin film being about 585 nm, corresponding to the optical energy band gap of the CdS thin film. Also, the results show the increase in photocurrent after laser annealing of the thin film, which will be reflected in increased photosensitivity due to enhanced recrystallization and a decrease in grain boundaries of the thin film. The sharp reduction in photocurrent at the shorter wavelength is due to the higher recombination rate of unbalanced carriers on the surface of the virgin sample [35-38]. Reduced grain boundaries and enhanced crystallization improve photosensitivity by: a- Fewer Grain Boundaries: Reduces defect sites that trap carriers and act as recombination centres, allowing better charge carrier mobility. b- Enhanced Crystallization: Creates a more ordered lattice with fewer defects, improving carrier mobility and reducing recombination. These effects lead to longer carrier lifetimes, higher mobility, and greater quantum efficiency, resulting in improved photoconductive performance [1, 16, 20].

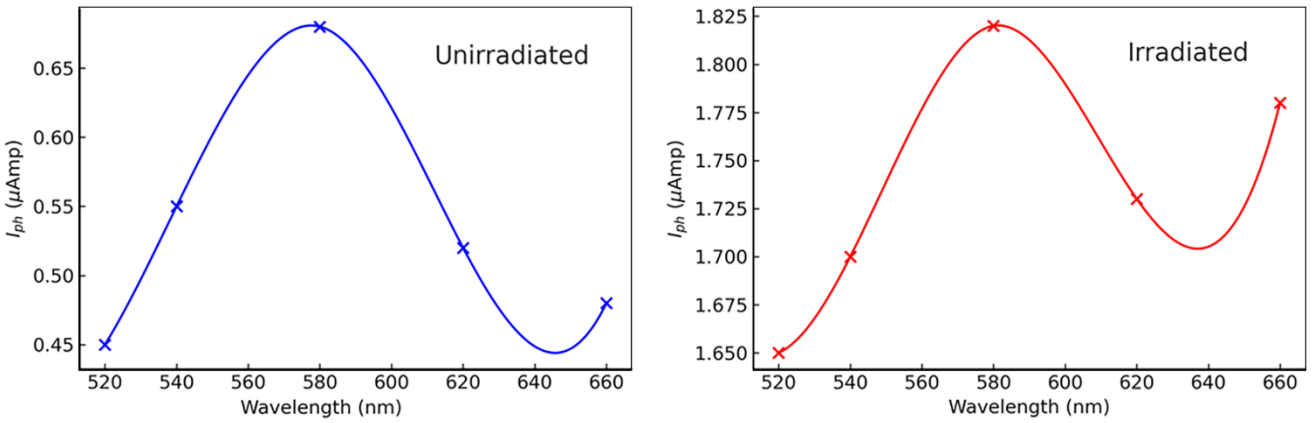

Figure 4: Dependence of photocurrent (I_{ph}) on wavelength for CdS thin films, comparing results before and after Nd:YAG laser annealing.

3.4 Transient Photoconductivity Measurements

Fig. 5 shows transient photocurrent measurements of the CdS thin film before and after laser annealing of the thin film. The thin film is initially in a dark chamber to avoid background interference. A rise in photocurrent was observed using light shined at the specific wavelength of 585 nm for 2 minutes. Additionally, this photocurrent was shown to increase with illumination, attributed to the rapid recombination of electrons and hole pairs due to the absorption of photons of light. The photocurrent reached its peak value and then gradually stabilized. After the light illumination is turned off, photocurrent decay is relatively slow, as the number of traps in the material gap exceeds the free carrier density [39, 40]. **Fig. 5** shows that the persistence of photocurrent in the sample remains even after a long time when the light is turned off. Traps play an essential role in photoconductivity, which involves capturing and releasing the trap with electrons. After annealing, the value of τ_d decreases, indicating increased photocurrent decay. Higher dissolution indicates decreased density of defect states after laser annealing. In non-exponential decay, the differential lifetime rises with time, unlike exponential decay, which remains constant with time. It is observed that the values of τd increase with time from the slope of **Fig. 5** (in the decay region) to I_{ph}, supporting the idea of non-exponential decay. This behaviour is because of the recombination coefficient, which depends on the distance of the crystallographic planes within a semiconductor. The slow decay can be explained as the trapping of charge carriers in more profound levels and the effect of local recombination [40, 41]. The requirement of τ_d on time was studied by plotting the relationship between τ_d and time, as shown in **Fig. 6** for CdS thin films before and after annealing. It is observed that the increase in the value of (τ_d) over time is due to the dependence of the recombination coefficient on the specific energy levels involved [42]. After laser annealing, the decay time (τ_d) decreases, reflecting improvements in recombination dynamics, as the decay becomes faster due to a reduction in trap states. Laser annealing reduces the defect density, especially at grain boundaries, improving the structural and electronic properties of the thin film. Enhanced crystallinity and reduced grain boundaries improve carrier mobility, leading to a more efficient photoconductive response. This excess degeneracy shows a decrease in the density of defect states after laser annealing. For non-exponential long-range I_{ph} decay, single-molecular interactions are assumed to dominate the recombination processes. In chalcogenide materials, when one pair of orbitals overlap the valence band, the combined valence band broadens dramatically, reflecting the charge carriers' effective mass inversely with the corresponding bandwidth. Thus, holes move faster than electrons in those materials, making holes the dominant carriers in chalcogenides [42]. Yamçiçier *et al*. [43] reported that when impurities are added to the chalcogenide's semiconductor material, the photocurrent will decrease as the impurities form charged states. Charged defects control the drift motion of charge carriers in the CdS thin film. Activation by impurities doping in the chalcogenide semiconductor material will change these local states' concentration to a change in the drift motion. The drift motion can be controlled through specific energy levels in local states. Its value can be expressed by Eq. (2) [28, 40]:

$$\mu = \mu_0 \, (N_c/N_t) e^{(-\varepsilon/kT)} \qquad (2)$$

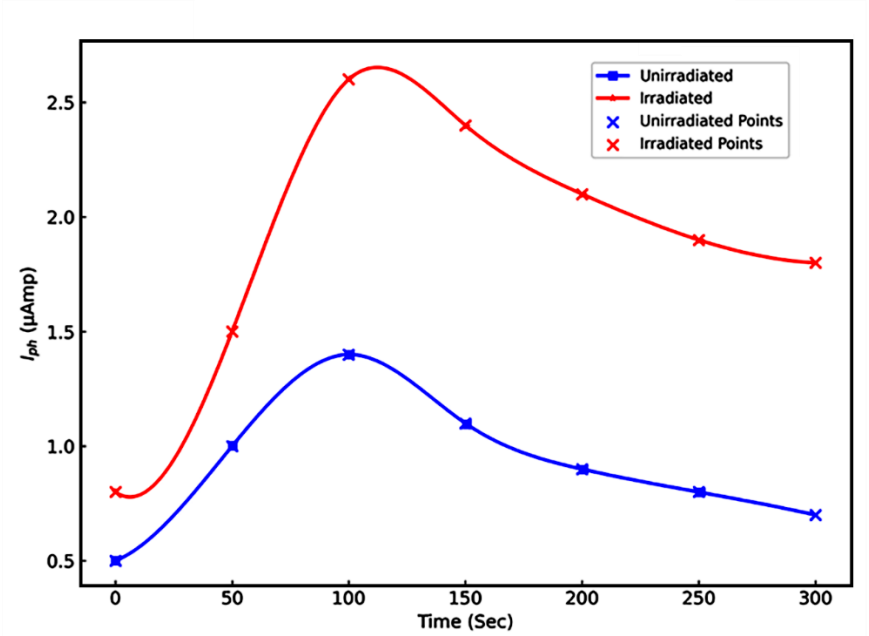

Figure 5: Transient photocurrent (I_{ph}) as a function of time for CdS thin films, comparing results before and after Nd:YAG laser annealing.

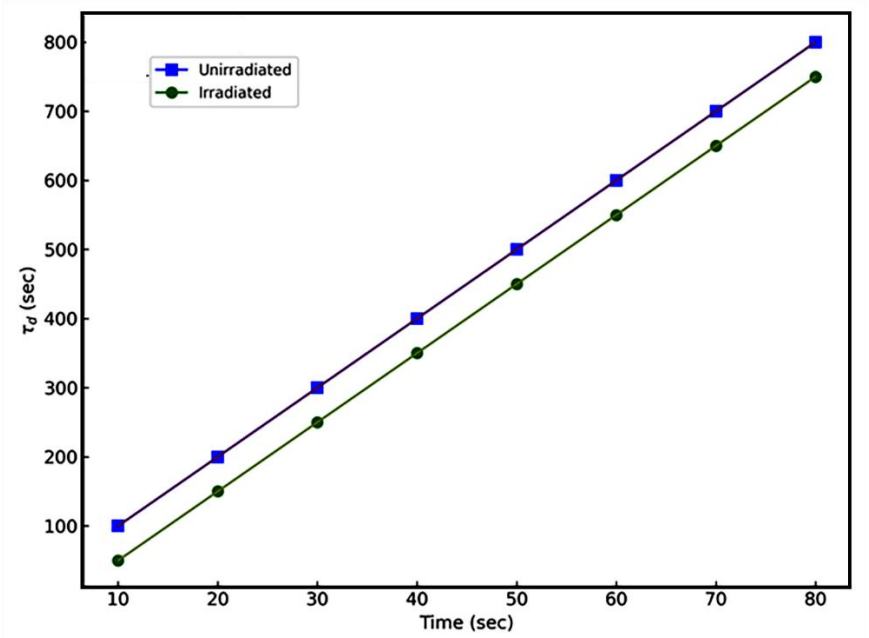

Figure 6: Variation of decay time (τ_d) with time for CdS thin films before and after Nd:YAG laser annealing.

Here, ε represents the energy difference between the level of the localized states and the band edge of the delocalized states, Nt and Nc stands for the effective densities of the delocalized and localized states, respectively, and μ_0 is the charge-carrier mobility in the band of delocalized states. While hole mobility is inversely proportional to the D^- centre concentration, electron drift mobility is inversely proportional to the D^+ centre concentration. This relationship arises because the nature of the localized states is linked to charged defects resembling D^+ and D^- centers [44]. What D^+ and D^- Centres stand for: When an atom loses one electron, it creates a positively charged defect called a D^- centre, which traps electrons and reduces their mobility. The mobility of holes (positive charges) is decreased by D^- centres, which are negatively charged defects that arise when an atom gains an additional

electron. By trapping electrons or holes and limiting their movement, both kinds impact the material's charge transport [45].

Finally, it can be concluded that Laser annealing improves the crystallinity and thus reduces grain boundaries in CdS thin films through: 1. Defect healing: Laser energy promotes the movement of atoms and repairs voids and disordered bonds. 2. Grain growth: Grain growth occurs, and smaller grains merge, reducing the density of grain boundaries and thus reducing charge trapping. 3. Bond reorganization: High temperatures due to pulsed laser annealing restructure bonds and reduce defect states. 4. Electronic enhancement: The reduction of defects and improvement of the crystallinity of a film leads to reduced trapping and recombination centres, improved carrier mobility, and optical conductivity. These mechanisms combined enhance the films' structural, photoconductivity, and electronic properties. One key factor affecting thin films' optoelectronic properties is their composition. Single-molecular recombination in CdS thin films refers to the effect of shallow traps that capture individual carriers, which can be reduced by laser annealing to improve the lifetime and mobility of carriers. Reducing this recombination enhances the charge collection efficiency in applications such as solar cells, making CdS films more effective for optoelectronic devices.

5. Conclusions

The Nd:YAG (532 nm) laser was used for laser crystallization, which effectively enhances the electronic and photonic properties of thin films deposited on glass substrates by increasing the crystallite size and reducing the grain boundary density. The observed improvements in photocurrent and photosensitivity, along with the maintenance of a monomolecular recombination mechanism, highlight the potential of this technique to advance the performance of thin film devices.

Conflict of Interest

The authors declare that they have no conflict of interest.

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