Original scientific paper

https://doi.org/10.33180/InfMIDEM2026.105

Informacije MIDEM
Journal of Microelectronics, Electronic Components and Materials
Vol. 56, No. 1(2026), XX – XX

Role of Deep Defect States in Anomalous Photovoltage Relaxation of Polycrystalline Silver-Doped Cadmium Telluride Thin Films

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Abstract: The present work investigates the long-term relaxation behavior of anomalous photovoltage and photoelectret voltage in polycrystalline CdTe:Ag thin films prepared by oblique thermal evaporation. The relaxation kinetics were studied at room temperature under controlled photopolarization conditions. Experimental results reveal a pronounced two-stage relaxation behavior, consisting of a fast initial component with characteristic times of 2–4 min and a slow long-term component extending up to 25–45 min.

The short-term relaxation is attributed to shallow trapping centers located near the band edges, while the long-term relaxation is governed by deep trapping centers associated with impurity–defect complexes involving silver atoms and cadmium vacancies. The observed metastable photoelectric states are explained within a bulk trapping model combined with near-surface band bending effects, which lead to asymmetric photocarrier separation and long-lived charge retention.

The obtained results provide new insight into defect-controlled relaxation processes in CdTe:Ag thin films and demonstrate the crucial role of deep trapping centers in the formation and stability of photoinduced electric states. These findings are relevant for optimizing the photoelectric performance and long-term stability of CdTe-based photovoltaic and optoelectronic devices.

Keywords: Photovoltage; Photoelectret; Defects; Trapping; Relaxation; Thin films

Vloga globokih defektnih stanj pri nepravilnem sproščanju fotonapetosti v polikristalnih tankih filmih kadmijevega telurida dopiranega s srebrom

Izvleček: V raziskavi preučujemo dolgoročno relaksacijsko obnašanje anomalne fotonapetosti in napetosti fotoelektrine v polikristalnih tankih slojih CdTe:Ag, pripravljenih s poševnim termičnim izparevanjem. Relaksacijsko kinetiko smo preučevali pri sobni temperaturi v nadzorovanih pogojih fotopolarizacije. Eksperimentalni rezultati kažejo izrazito dvofazno relaksacijsko obnašanje, ki ga sestavljata hitra začetna komponenta s karakterističnimi časi 2–4 min in počasna dolgoročna komponenta, ki traja do 25–45 min.

Kratkoročno relaksacijo pripisujemo plitvim ujetnim centrom, ki se nahajajo blizu robov pasov, medtem ko dolgoročno relaksacijo uravnavajo globoki ujetni centri, povezani s kompleksi nečistot in napak, v katerih sodelujejo atomi srebra in praznine kadmija. Opazovana metastabilna fotoelektrična stanja pojasnjujemo z modelom ujetja v masi v kombinaciji z učinki upogibanja pasov blizu površine, ki vodijo do asimetrične ločitve fotonosilcev in dolgotrajnega zadrževanja naboja.

Pridobljeni rezultati ponujajo nov vpogled v defektno nadzorovane relaksacijske procese v tankih filmih CdTe:Ag in dokazujejo ključno vlogo globokih ujetniških centrov pri nastajanju in stabilnosti fotoinduciranih električnih stanj. Ti izsledki so pomembni za optimizacijo fotoelektrične zmogljivosti in dolgoročne stabilnosti fotovoltaičnih in optoelektronskih naprav na osnovi CdTe.

Ključne besede: fotonapetost, fotoelektrina, napake, ujetje, relaksacija, tanke plasti

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1 Introduction

In recent years, thin-film semiconductor materials based on cadmium chalcogenides have attracted significant scientific and technological interest due to their wide application in photovoltaics [1], [2], photodetectors, and ionizing radiation sensors [3]. Among these materials, CdTe (Cadmium Telluride) is considered one of the most promising materials for thin-film solar cells owing to its optimal band gap ($E_g \approx 1.45$ eV) [4], high optical absorption coefficient (10^4–10^5 cm^{-1}) [5], and relatively low-cost fabrication methods [6]. Although polycrystalline CdTe thin films are widely used in industrial photovoltaic devices, their photoelectric properties remain strongly dependent on structural disorder and grain boundary–related defect states [3], [4], which strongly affect carrier transport and recombination processes.

One of the main challenges associated with polycrystalline CdTe thin films is the presence of deep trapping levels related to intrinsic defects and impurity atoms, as observed in recent studies [5], [6]. These deep defect states play a crucial role in carrier recombination and charge trapping processes [7], as well as in the formation of internal electric fields in polycrystalline CdTe films [10].

Deep trapping centers located within the energy range of 0.2–0.6 eV typically exhibit markedly different capture cross-sections for electrons and holes [22]. As a consequence, non-equilibrium photoinduced states may form, manifesting as anomalous photovoltage (APV), long-lived polarization effects, and metastable photoelectric behavior, which have been widely discussed in the context of defect-mediated photoresponse in CdTe-based structures [7]–[9]. One of the most intriguing manifestations of such non-equilibrium phenomena is the formation of a photoelectret state (PES) in CdTe thin films even in the absence of an external polarizing electric field, as demonstrated in experimental studies reported in the literature [11].

In this case, residual photovoltage arises due to spatial separation of photogenerated charge carriers under the action of built-in electrostatic fields in the near-surface space-charge region, followed by their asymmetric trapping by deep impurity–defect complexes located within crystallites and at grain boundaries, as explained within the framework of defect-assisted charge separation models developed for CdTe and analogous semiconductor systems [10], [26], [27]. After termination of illumination, the photovoltage may persist for extended periods and relax through several characteristic stages, reflecting the hierarchical nature of trapping centers, as revealed by time-resolved photoelectric and relaxation measurements [12].

Recent experimental and review studies published between 2020 and 2025 have highlighted the importance of long-term relaxation processes and metastable photoelectric states in CdTe-based thin films [31]. From this perspective, investigation of the relaxation kinetics of APV provides valuable insight into the energy spectrum, concentration, and physical nature of deep trapping centers, as well as their role in the generation and decay of photoinduced internal electric fields, as demonstrated in defect-sensitive photoelectric studies of CdTe thin films [11].

Previous studies have demonstrated that the relaxation of photoelectret voltage (PEV) in impurity-doped CdTe thin films may proceed via multiple exponential stages, indicating the coexistence of shallow and deep trapping levels with substantially different carrier lifetimes [12], [14]. Nevertheless, the specific mechanisms governing long-term relaxation, particularly the contribution of impurity–vacancy complexes formed by silver incorporation, remain insufficiently clarified for polycrystalline CdTe thin films deposited under inclined substrate geometry, despite several related investigations [28].

Silver was selected as a dopant for CdTe because it can form deep acceptor-like defect states, especially Ag–V_{Cd} complexes, inside the band gap. These deep centers can efficiently trap photogenerated carriers and therefore strongly affect the generation and long-term relaxation of photovoltage. In addition, low-level Ag incorporation can noticeably modify

the electrical and photoelectric properties of CdTe without causing significant structural degradation. For this reason, Ag is an appropriate dopant for studying defect-controlled photovoltage relaxation in polycrystalline CdTe thin films [16].

In the present work, the long-term relaxation of APV and PEV in polycrystalline CdTe:Ag thin films is systematically investigated, with particular emphasis on the role of Ag-related deep trapping centers. Special attention is devoted to identifying characteristic relaxation times, separating short-term and long-term relaxation components, and elucidating the role of deep trapping centers responsible for metastable photoelectric behavior. The obtained results contribute to a deeper understanding of defect-controlled relaxation processes in CdTe thin films and are relevant for improving the stability and performance of CdTe-based photovoltaic structures.

2 Materials and Methods

Polycrystalline CdTe thin films were prepared by thermal evaporation under high-vacuum conditions, following conventional procedures widely used for CdTe thin-film fabrication [15]. The deposition process was carried out at a residual pressure in the range of (5×10^{-5} – 1×10^{-4}) Torr, which is typical for modern thermal evaporation systems and ensures stable and reproducible growth of chalcogenide semiconductor films [15]. The CdTe material was evaporated from a resistively heated crucible, producing a vertical vapor flux, while the glass substrates were intentionally positioned at an inclination angle of approximately 40-50° with respect to the vapor direction. This oblique deposition geometry promotes structural anisotropy and enhances near-surface electric field formation, which is essential for the development of APV and photoelectret effects in CdTe thin films [15].

A schematic illustration of the evaporation geometry and experimental configuration is presented in Fig. 1.

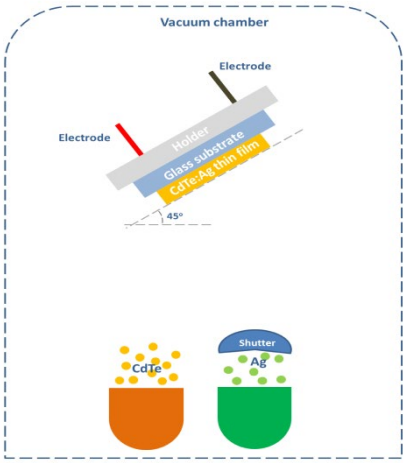

Fig. 1. Schematic diagram of oblique thermal evaporation and APV measurement configuration inside the vacuum chamber.

During deposition, the substrate temperature was maintained in the range of 260–320 °C, providing optimal conditions for polycrystalline CdTe film growth [15]. The total deposition time was adjusted to obtain film thicknesses of approximately 1.0–1.5 μm, as estimated from the deposition rate and growth duration.

The evaporation process was performed using a laboratory-scale high-vacuum thermal evaporation system that is functionally comparable to contemporary physical vapor deposition platforms employed in thin-film research in terms of achievable vacuum level and deposition stability [15].

Silver doping was introduced during the CdTe deposition process by thermal evaporation from a separate crucible, allowing independent control of the host material and dopant fluxes. Such a co-evaporation approach is widely used for controlled impurity incorporation in CdTe thin films and enables the formation of electrically active defect complexes [16]. The Ag source was activated 20–35 min after the onset of CdTe deposition, when a continuous CdTe layer had already formed on the substrate. After Ag incorporation, CdTe deposition was continued for an additional 30–50 min, ensuring that Ag atoms were embedded within the bulk of the film rather than localized near the substrate

interface. This deposition sequence is known to favor the formation of Ag–V_{Cd} impurity–defect complexes, which govern long-term photoelectric relaxation behavior in CdTe thin films [14], [24]. Based on the relative evaporation times and crucible temperatures, the nominal Ag concentration in the CdTe:Ag thin films is estimated to be below 1 at.%, which is sufficient for defect activation without inducing substantial modifications of the host crystal structure [16], [25].

In this study, APV refers to the photovoltage measured immediately after illumination, whereas PEV denotes the residual voltage remaining after short-circuiting the sample electrodes. APV and PEV measurements were performed at room temperature (290–300 K) under ambient conditions. Photopolarization was induced by continuous illumination using a mercury lamp with an intensity of 450–550 lux, similar to conditions employed in earlier photoelectret studies of CdTe thin films [17].

The experimental structure consisted of polycrystalline CdTe:Ag thin films deposited on glass substrates. For photovoltage measurements, electrical contacts were formed on the film surface using graphite in a planar configuration. Graphite contacts were selected due to their ability to provide stable and reproducible electrical contact with p-type CdTe without introducing significant additional interface states. The measurement geometry allowed direct illumination of the film surface under ambient conditions, and no transparent electrode was required.

The dark resistance of the samples prior to illumination was very high and temperature-dependent, decreasing from approximately 7×10^{13} Ω to 2×10^{12} Ω in the temperature range of 100–500 K. At room temperature, the resistance remained on the order of 10^{12}–10^{13} Ω, confirming the high-resistivity nature of the CdTe:Ag thin films.

The photovoltage was recorded using a high-input-resistance electrostatic voltmeter to ensure minimal charge leakage during long-term relaxation measurements [23]. After switching off the illumination, the decay of the photovoltage was monitored over time intervals ranging from several minutes to several hours. To separate the PEV from the APV component, the sample electrodes were briefly short-circuited for 2–3 s after photopolarization. Following reconnection to the measuring circuit, the residual PEV was recorded as a function of time.

Photovoltage relaxation curves were analyzed using semi-logarithmic representations, as commonly adopted for multi-exponential relaxation analysis in defect-dominated semiconductors [17]. From the slopes of the relaxation curves, two characteristic time constants were extracted: a short-term relaxation time $\tau_{init} \approx$ 2–4 min, corresponding to shallow trapping states, and a long-term relaxation time $\tau^* \approx$ 25–45 min, associated with deep trapping centers.

The presence of two distinct relaxation regimes reflects the coexistence of shallow and deep defect states governing the photoelectret behavior in CdTe:Ag thin films. The long-term relaxation component was further used to estimate the activation energies of deep trapping centers responsible for metastable photoelectric states.

3 Results

The time relaxation of anomalous photovoltage V_{APV} in polycrystalline CdTe thin films is governed by the trapping and release of photogenerated charge carriers, as widely reported for defect-dominated semiconductor systems [17]. As shown in Fig. 2, the relaxation behavior strongly depends on the film composition. For the undoped CdTe thin film, the decay of V_{APV} after switching off the illumination can be reasonably described by a single-exponential law, which is typical for relaxation controlled by a dominant shallow trapping level [12].

$$V(t) = V_0 exp\left(\frac{-t}{\tau}\right), \qquad (1)$$

where V_0 is the initial photovoltage and τ is the characteristic relaxation time.

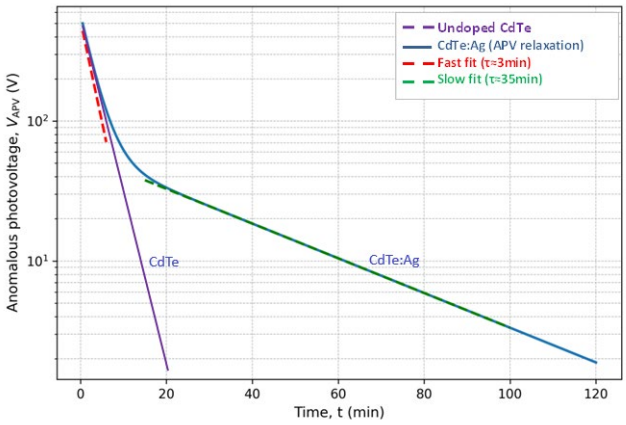

Fig. 2. Semi-logarithmic plot of the time relaxation of the anomalous photovoltage V_{APV} in CdTe and CdTe:Ag thin films measured at T=293 K after switching off illumination. The undoped CdTe film (purple dashed line) exhibits a fast single-stage decay, whereas the CdTe:Ag film demonstrates a pronounced two-stage relaxation consisting of a fast component (τ≈3 min) and a slow long-term component (τ≈35 min). The relatively large initial amplitude of V_{APV} reflects its origin associated with photoinduced charge separation and internal electric fields. The voltage axis is presented on a semi-logarithmic scale.

In contrast, the semi-logarithmic plots for CdTe:Ag thin films clearly deviate from single-exponential behavior, revealing the presence of at least two relaxation mechanisms. Therefore, the experimental data for CdTe:Ag thin films are more accurately described by a two-component model, indicating the coexistence of shallow and deep trapping centers with different relaxation times [12], [14].

$$V(t) = V_1 exp\left(\frac{-t}{\tau_{init.}}\right) + V_2 exp\left(\frac{-t}{\tau^*}\right), \quad (2)$$

where τ_{init} and τ^* denote the fast and slow relaxation time constants, respectively, and V_1 and V_2 are the corresponding voltage amplitudes associated with shallow and deep trapping processes.

Each photovoltage relaxation curve was obtained by averaging the results of at least three independent measurements performed on the same sample under identical illumination conditions. The extracted relaxation times were reproducible within experimental uncertainty.

The error bars shown in Figures 2–4 represent the standard deviation of the relaxation times determined from repeated measurements and reflect minor variations in initial photovoltage and illumination stability.

Analysis of the relaxation curves (Figures 2 and 3) shows that the short-term relaxation time τ_{init} lies in the range of 2–4 min and remains nearly independent of the photopolarization duration. This behavior suggests that the initial decay is controlled by shallow trapping centers with low activation energies and fast carrier emission rates, as commonly observed in CdTe thin films and related semiconductor materials [17]. In contrast, the long-term relaxation time τ^* increases markedly with increasing photopolarization duration and reaches values of 25–45 min. This behavior indicates progressive filling of deep trapping centers during illumination, which dominate the metastable photoelectret state and control the long-term relaxation of photovoltage [37], [38].

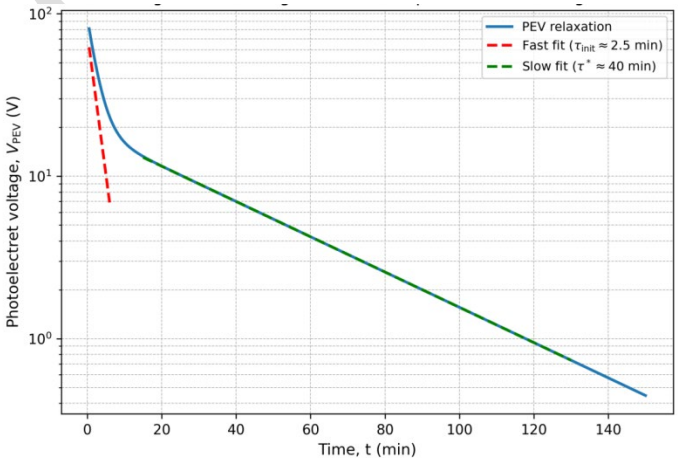

Fig. 3. Semi-logarithmic plot of the photoelectret voltage V_{PEV} relaxation in CdTe:Ag thin films at room temperature. The relaxation curve reveals two distinct components: a fast initial decay (τ_{init}≈2.5 min) and a slow component (τ≈40 min), associated with shallow and deep trapping states, respectively. Compared to the anomalous photovoltage, the lower initial amplitude of V_{PEV} reflects its different physical origin related to trapped charge polarization (photoelectret state). The voltage axis is presented on a semi-logarithmic scale.

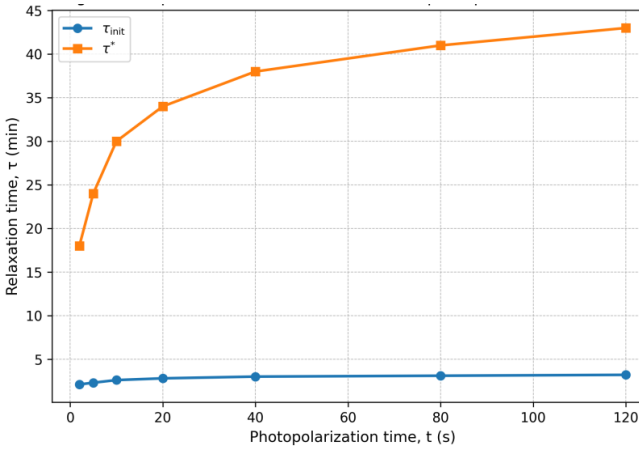

Fig. 4. Dependence of the short-term (τ_{init}) and long-term (τ^*) relaxation times on the photopolarization duration for polycrystalline CdTe:Ag thin films measured at room temperature. The weak variation of τ_{init} and the pronounced increase of τ^* indicate different physical origins of shallow and deep trapping centers.

The temperature-activated release of carriers from deep traps can be qualitatively described by an Arrhenius-type relation, which is widely used to analyze deep-level emission processes in semiconductors [19].

$$\tau^* = \tau_0 exp\left(\frac{E_t}{kT}\right), \quad (3)$$

where τ_0 is the pre-exponential factor, E_t denotes the activation energy of the deep trap state governing the relaxation process, k is the Boltzmann constant, and T is the absolute temperature.

The experimentally observed long relaxation times at room temperature indicate that the dominant deep traps are located relatively far from the band edges, consistent with impurity–defect complexes involving Ag atoms and cadmium vacancies, which are known to introduce deep acceptor-like states in CdTe [15], [24]. Although temperature-dependent measurements were not performed in the present study, the long relaxation times observed at room temperature ($\tau^* \approx 25$–45 min) indicate that the dominant deep trapping centers are likely characterized by activation energies on the order of 0.3-0.5 eV, in agreement with previously reported Ag–V_{Cd}-related deep acceptor levels in CdTe-based materials [24], [25].

To interpret the experimentally observed two-stage relaxation behavior of anomalous photovoltage, a bulk trapping model previously proposed for defect-rich CdTe thin films is employed and schematically illustrated in Fig. 5 [20]. The diagram reflects the presence of energetically distinct trapping centers within the band gap of CdTe:Ag thin films, which govern the kinetics of photovoltage relaxation.

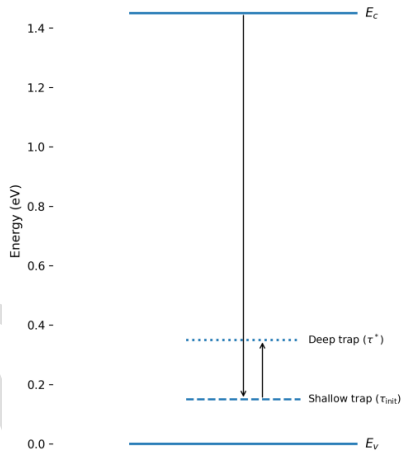

Fig. 5. Schematic energy band diagram of CdTe:Ag thin film showing shallow and deep trapping levels associated with two-stage APV relaxation.

Shallow trapping states located close to the band edges are responsible for the fast initial relaxation component characterized by the relaxation time τ_{init}. These states exhibit relatively small activation energies and short carrier lifetimes, allowing rapid release of trapped carriers after illumination is switched off. As a result, they contribute primarily to the early-stage decay of anomalous photovoltage.

In contrast, deep trapping levels situated further from the band edges play a dominant role in the long-term relaxation process characterized by the relaxation time τ^*. These deep states efficiently capture photogenerated carriers and retain them for extended periods of time, leading to the formation of metastable photoelectret states. In CdTe:Ag thin films, such deep levels are most plausibly associated with impurity–defect complexes involving Ag atoms and cadmium vacancies, which are known to introduce deep acceptor-like states within the band gap.

The coexistence of shallow and deep trapping levels, schematically illustrated in Fig. 5, provides a consistent explanation for the two-stage exponential relaxation behavior observed experimentally.

While bulk deep trapping centers determine the long-term stability of the photoelectret state, the formation of high anomalous photovoltage values requires the presence of an internal electric field. The near-surface band bending model shown schematically in Fig. 6 illustrates the physical mechanism responsible for anomalous photovoltage generation in polycrystalline CdTe thin films.

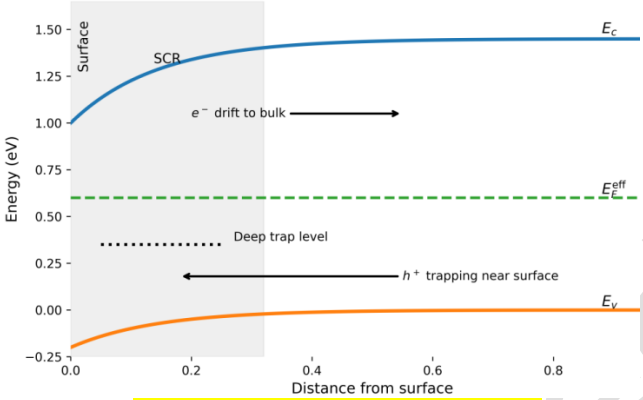

Fig. 6. Schematic band diagram illustrating near-surface band bending and asymmetric photocarrier separation responsible for anomalous photovoltage formation in polycrystalline CdTe thin films. The horizontal axis represents the relative distance from the surface, highlighting the near-surface space-charge region and carrier trapping processes.

Surface states and grain boundary defects give rise to band bending in the near-surface region, leading to the formation of a space-charge region accompanied by a built-in electrostatic field, as widely discussed for CdTe and related semiconductor thin films [26], [27]. Under illumination, this internal field induces an asymmetric spatial separation of photogenerated charge carriers: electrons are driven toward the bulk of the film, whereas holes drift toward the surface and grain boundaries.

The preferential trapping of holes at deep surface-related defect states suppresses recombination and results in the accumulation of spatially separated charge. This process leads to the buildup of a macroscopic electric field and the appearance of APV even in the absence of an external polarizing electric field [10]. The persistence of this photovoltage after illumination is switched off is ensured by the slow release of carriers from deep trapping states, as discussed in the previous subsection.

Thus, the combined action of near-surface band bending and bulk deep trapping centers, illustrated in Figures 5 and 6, provides a unified physical picture explaining both the generation and the long-term relaxation of APV in polycrystalline CdTe thin films.

The analysis of anomalous photovoltage and photoelectret relaxation kinetics reveals two well-separated characteristic time constants in CdTe:Ag thin films. The short-term relaxation time τinit, lying in the range of 2–4 min, is attributed to shallow trapping states located near the band edges. In contrast, the long-term relaxation time τ*, varying between 25 and 45 min, reflects the contribution of deep trapping centers associated with Ag-related impurity–defect complexes.

At room temperature, the anomalous photovoltage reaches values as high as 600 V, indicating the formation of a stable photoelectret state. These parameters summarize the dominant features of the photoelectric response of CdTe:Ag thin films and are consistent with the defect-controlled relaxation mechanisms discussed above.

4 DISCUSSION

The experimentally observed relaxation behavior of APV and PEV in polycrystalline CdTe:Ag thin films clearly indicates the coexistence of trapping centers with different energetic depths, which is a characteristic feature of defect-dominated relaxation processes in CdTe-based materials [21]. In contrast to undoped CdTe films exhibiting predominantly single-stage relaxation, Ag-doped films demonstrate a pronounced two-stage relaxation behavior, reflecting the activation of additional deep trapping centers.

The fast initial relaxation component, characterized by relaxation times of 2–4 min,

exhibits only a weak dependence on the photopolarization duration. This behavior is consistent with shallow trapping states located near the band edges and associated with intrinsic point defects, which possess relatively small activation energies and enable rapid capture and release of photogenerated carriers [22].

In contrast, the long-term relaxation component displays significantly longer characteristic times in the range of 25–45 min and shows a pronounced dependence on the photopolarization duration. This behavior provides strong evidence for the involvement of deep trapping centers, which progressively accumulate charge during illumination and release it slowly after the removal of light. Long-lived photoinduced states of this type are widely associated with metastable photoelectret behavior in CdTe-based thin films and other defect-rich semiconductor systems [37], [38].

Although temperature-dependent measurements were not performed in the present study, the long relaxation times observed at room temperature suggest that the dominant deep trapping centers are characterized by relatively large activation energies. On the basis of comparison with previously reported spectroscopic and photoelectric studies, these deep states can be plausibly attributed to Ag–V_{Cd} impurity–defect complexes, which are known to introduce deep acceptor-like levels in CdTe [24], [25]. A quantitative determination of activation energies and trap concentrations, however, requires dedicated temperature-dependent relaxation experiments and remains a subject for future investigation.

The formation of APV in the absence of an external electric field can be consistently explained by near-surface band bending and internal electric fields arising from surface states and grain boundary defects. Under illumination, these internal fields induce asymmetric spatial separation of photogenerated carriers, leading to preferential trapping of one carrier type at deep defect states and suppression of recombination [26]. The oblique deposition geometry employed in this work is expected to enhance structural anisotropy, defect redistribution, and internal electric field formation, thereby amplifying the observed APV and photoelectret effects [28], [29].

Previous studies have shown that non-normal incidence deposition and related growth anisotropy can significantly influence defect distribution and photoelectric response in polycrystalline CdTe films and analogous systems. Nevertheless, a direct quantitative comparison with normally deposited films is required to unambiguously isolate the contribution of deposition geometry.

It should be emphasized that the unusually high APV values observed in this study reflect macroscopic charge separation across the film thickness under high-input-impedance measurement conditions and do not correspond to power-generating voltages [37]. Similar macroscopic charge separation effects resulting in large measurable photovoltages have been reported in CdTe-based systems exhibiting pronounced photoelectret behavior [29], [40].

Overall, the present results demonstrate that the combined action of oblique deposition geometry and Ag-induced deep trapping centers significantly enhances both the magnitude and long-term stability of photoinduced electric states in polycrystalline CdTe thin films. The proposed interpretation is consistent with previously reported bulk trapping and band bending models developed for CdTe and related semiconductor materials [39]–[41].

5 CONCLUSIONS

In this work, the relaxation kinetics of anomalous photovoltage and photoelectret voltage in polycrystalline CdTe:Ag thin films prepared by oblique thermal evaporation were systematically investigated. The experimental results reveal a two-stage relaxation process governed by shallow and deep trapping centers with distinctly different characteristic times.

The fast relaxation component, with characteristic times of 2–4 min, is attributed to shallow trapping states located near the band edges. In contrast, the slow long-term

relaxation component, extending over 25–45 min, is associated with deep trapping centers related to Ag-induced impurity–defect complexes. The formation of anomalous photovoltage is explained by the combined action of near-surface band bending, internal electric fields, and defect-assisted asymmetric separation of photogenerated charge carriers.

Although the present study was limited to room-temperature measurements, the obtained results provide clear qualitative insight into defect-controlled relaxation processes and the stabilization of long-lived photoinduced electric states in CdTe:Ag thin films. These findings are relevant for the design and optimization of CdTe-based photovoltaic and photoelectret devices, where long-term charge retention and metastable electric states play a crucial role.

6 Acknowledgments

The authors gratefully acknowledge Prof. Nosirjon Yuldashev for his valuable scientific guidance and constructive discussions.

7 Conflict of Interest

The authors declare no conflict of interest.

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Arrived: 04.10.2025
Accepted: 22.12.2025