

Impact of the Annealing Temperature on the Local Se Distribution and Optical Properties in $\text{CdSe}_x\text{Te}_{1-x}$ Solar Cells by Multimodal X-ray Microscopy

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The introduction of Se in CdTe solar-cell absorbers has led to multiple beneficial effects in the devices, including an increased short-circuit current and an extended charge-carrier lifetime. A critical manufacturing step of $\text{CdSe}_x\text{Te}_{1-x}$ solar cells is CdCl_2 annealing at high temperatures, during which Se diffuses through the absorber layer. Understanding the local Se distribution in the absorber and its impact on optical properties of the material is key for the further development of this thin-film solar cell technology. In a multimodal x-ray scanning microscopy approach, we investigated the impact of the annealing temperature of $\text{CdSe}_x\text{Te}_{1-x}$ solar cells on the Se distribution and on optical characteristics of the material. For this purpose, we exploited an upgraded x-ray excited optical luminescence (XEOL) detection unit that enables simultaneous spectrally and temporally resolved XEOL mapping at synchrotron facilities that are ideal for studying thin-film solar-cell materials with hard x-rays featuring high penetration depth and lateral resolution.

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I. INTRODUCTION

Polycrystalline cadmium telluride (CdTe) thin-film solar cells have benefited from several advances over the last years, such as the incorporation of a Se gradient into the absorber layer of the cells, which has recently led to conversion efficiencies of 23.1% [1]. The Se gradient can be achieved by sequentially depositing a $\text{CdSe}_x\text{Te}_{1-x}$ and a CdTe layer on top of each other, followed by a CdCl_2 annealing step at high temperatures, during which Se diffuses from the front to the back of the absorber [2], thereby creating a graded absorber. This has several desired effects on the solar cell, such as increased

short-circuit current [3], graded bandgap energy [4], and extended charge-carrier lifetime [5].

The effects of Se on the structure, elemental distribution, and optoelectronic performance of $\text{CdSe}_x\text{Te}_{1-x}$ solar cells have been widely studied. Çiriş *et al.* used scanning electron microscopy and x-ray diffraction to show that the open-circuit voltage, short-circuit current, and conversion efficiency are optimized at the lowest bandgap energy of $E_g \approx 1.4$ eV, which is found in $\text{CdSe}_x\text{Te}_{1-x}$ solar cells with $x \approx 0.3$ [6].

Munshi *et al.* applied scanning transmission electron microscopy (STEM) and energy-dispersive x-ray spectroscopy (EDX) to $\text{CdSe}_x\text{Te}_{1-x}$ solar cells and observed that the CdCl_2 treatment, which leads to a graded absorber layer, removes detrimental structural defects, such as stacking faults, and promotes recrystallization of the film and grain growth [7].

Fiducia *et al.* used cathodoluminescence (CL) and secondary-ion mass spectrometry (SIMS) to show that Se diffuses primarily along grain boundaries during the

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CdCl₂ treatment. A higher CL signal and longer charge-carrier diffusion length in Se-rich regions of the cell indicated an improvement in the charge-carrier lifetime with increasing Se concentration, and a graded bandgap energy from the front to the back of the absorber layer was shown by spectrally resolved CL measurements [4].

Poplawsky *et al.* applied electron-beam-induced current (EBIC), transmission electron microscopy, and atom probe tomography to study the dependence of the EBIC signal on the Se concentration and crystalline structure of the CdSe_xTe_{1-x} layer with varying initial layer thickness, showing that the CdSe_xTe_{1-x} alloy is zincblende-structured or contains wurtzite phases, depending on the initial layer thickness, which influences the local short-circuit current [8].

The influence of the CdCl₂ annealing temperature on the optoelectronic characteristics of CdSe_xTe_{1-x} solar cells was systematically investigated by Paul *et al.* through external quantum efficiency and current-voltage (*JV*) measurements, where an overall improvement in short-circuit current, open-circuit voltage, and conversion efficiency was observed with increasing CdCl₂ treatment temperature [9].

The beneficial effects of Se are currently limited to the front and bulk of the absorber layer due to the low Se concentration at the back of CdSe_xTe_{1-x} solar cells. Stronger Se diffusion to the back of the absorber could further increase the beneficial effects of Se, ultimately leading to higher conversion efficiencies. One of the decisive manufacturing steps to achieve stronger Se diffusion is the CdCl₂ treatment; however, the influence of the treatment temperature and the resulting Se distribution on local optical and electrical parameters is not widely studied and understood. That is addressed in this publication, in which x-ray scanning microscopy was used to investigate the impact of the annealing temperature on the optical characteristics of CdSe_xTe_{1-x} solar cells. Therefore, three CdSe_xTe_{1-x} solar cells, fabricated using identical processes except for the CdCl₂ treatment temperatures (480°C, 450°C, 420°C), were investigated: CdSeTe₄₈₀, CdSeTe₄₅₀, CdSeTe₄₂₀. A twofold multimodal scanning x-ray microscopy approach at the microprobe endstation of the beamline P06 at PETRA III, DESY [10,11], gave access to the local elemental distribution, effective bandgap variation, and charge-carrier lifetime via x-ray fluorescence (XRF) as well as spectrally and temporally resolved x-ray excited optical luminescence (SR- and TR-XEOL), respectively. This approach enabled a direct local correlation of the elemental distribution with the optical characteristics of the cells. Note that the solar cells were measured at the Se-poor backside of the devices that was exposed to the incident beam whereas the front side was covered by x-ray intransparent glass.

XEOL measurements at synchrotron beamlines present an experimental challenge, since the XEOL signal strength is limited by the x-ray beam flux available at the facility,

and the confined space available in the beamline hutches complicates the integration of an XEOL detection unit into the beamline environment. An XEOL instrument named AXOLOTL was developed at DESY, and its applicability was showcased in a multimodal approach analyzing triple-junction perovskite solar cells [12]. Here, the upgraded AXOLOTL 2.0 detection unit enabled simultaneous SR- and TR-XEOL measurements with an optimized signal-to-noise ratio. The setup is designed in a transportable and modular configuration, allowing it to adapt to different beamline environments and samples and contributing to the development of synchrotron instrumentation for future experiments, notably at the upgraded storage ring PETRA IV [13].

II. RESULTS

A. Diffusion and segregation of Se

The counts for each of the elements obtained from the XRF analysis are used as a measure for the projected relative element density or concentration. The validity of this interpretation depends on the attenuation length of the respective fluorescence line relative to the absorber thickness ($\approx 4 \mu\text{m}$). The attenuation length of the Se $K\alpha$ emission line ($E_{\text{Se}_{K\alpha}} = 11.22 \text{ keV}$) is $16.8 \mu\text{m}$ in CdTe and $18.5 \mu\text{m}$ in CdSe_{0.3}Te_{0.7} [14], exceeding the absorber thickness by a factor of 4–5. Hence, self-absorption is negligible [15], and the Se XRF signal represents the projected Se content integrated along the full beam path through the absorber. Consequently, the lateral Se variations resolved in such plan-view maps of this work represent depth-projected contrasts, distinct from the front-to-back Se gradients commonly obtained when techniques such as SIMS or STEM-EDX are applied to cross-sectional specimens.

The resulting maps of the Se counts $I_{\text{XRF, Se}}$ from the scans of each of the cells are shown in Figs. 1(a)–1(c). Visually, the CdSeTe₄₈₀ cell shows the largest features, and different areas with high and low Se concentration are clearly resolved. As the annealing temperature decreases, the features become smaller and less pronounced, indicating a more uniform distribution of Se as a result of a weaker diffusion. This effect is also reflected in the relative standard deviation of the Se concentration within each cell, yielding $\sigma_{\text{Rel}} = 9.4\%$, 5.5% , and 3.8% for CdSeTe₄₈₀, CdSeTe₄₅₀ and CdSeTe₄₂₀, respectively.

The attenuation length of the Cd $L\alpha_1$ emission line ($E_{\text{Cd}_{L\alpha_1}} = 3.1 \text{ keV}$) is only $3.2 \mu\text{m}$ in CdTe and $3.0 \mu\text{m}$ in CdSe_{0.3}Te_{0.7} [14], comparable to or smaller than the absorber thickness. The Cd count maps ($I_{\text{XRF, Cd}}$), shown in Figs. 1(d)–1(f) ($\sigma_{\text{Rel}} = 1.9\%$, 1.4% , and 2.3% for CdSeTe₄₈₀, CdSeTe₄₅₀ and CdSeTe₄₂₀, respectively), are therefore susceptible to self-absorption and represent at best the back surface of the device. Since the front surface of the absorber was deposited on flat glass and can be assumed smooth at the scale of the x-ray

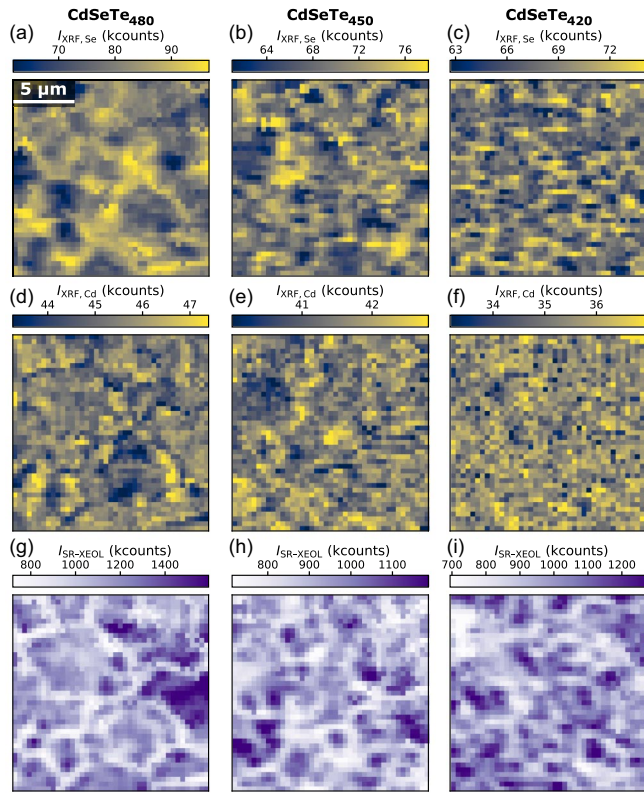


FIG. 1. (a)–(c) Maps of the Se counts acquired via XRF for the cells CdSeTe₄₈₀, CdSeTe₄₅₀, and CdSeTe₄₂₀, respectively. (d)–(f) Maps of the Cd counts acquired via XRF for CdSeTe₄₈₀, CdSeTe₄₅₀, and CdSeTe₄₂₀. (g)–(i) Maps of the panchromatic XEOL signal acquired via SR-XEOL for CdSeTe₄₈₀, CdSeTe₄₅₀, and CdSeTe₄₂₀.

microscopy measurements, topographic sensitivity is limited to the back surface. Atomic force microscopy (AFM) measurements of representative areas, conducted at the DESY NanoLab [16] (see Fig. S2 and Sec. II in Supplemental Material [17]), yield minima at grain boundaries with depths on the order of 30–100 nm, corresponding to 0.8%–2.5% of the absorber thickness. Indeed, the network-like, Cd-poor structures visible in the Cd maps—particularly pronounced for the large grains of the CdSeTe₄₈₀ cell—may therefore be assumed to be grain boundaries whose morphology is confirmed by these AFM images. Given that columnar grain growth perpendicular to the p - n junction is typical of CdTe [18,19], the back-surface grain structure probed by the Cd maps is representative of the absorber as a whole.

The Pearson correlation coefficient between Se and Cd counts yields $r_{\text{PCC}} = -0.2$ for the CdSeTe₄₈₀ cell, indicating that Se is located in higher concentrations at the Cd-poor regions. For the CdSeTe₄₂₀ and CdSeTe₄₅₀ cells, $r_{\text{PCC}} = 0.04$ and $r_{\text{PCC}} = 0.01$ indicate a negligible correlation. If topography dominated both Se and Cd XRF signals, a consistent positive correlation would be expected across all cells. Hence, the negative or negligible

correlation further supports that the Se distribution is not primarily governed by topographic effects. Together with the large Se $K\alpha$ attenuation length and the AFM data, this confirms that the spatial variation in the Se maps reflects genuine compositional differences.

Overall, the observed trends result from the interplay between annealing temperature, grain size, and Se diffusion. The grains formed during the recrystallization process during the annealing step increase in size with the annealing temperature [7,20,21]. Hence, the grains formed in the case of the cell annealed at 480°C are large enough to be resolved at the P06 microprobe endstation with the given beam size and chosen step size. This is less the case for the solar cells annealed at 450 and 420°C due to smaller and vertically overlapping grains. Furthermore, Se diffuses from the CdSe _{x} Te_{1- x} layer toward the back of the cell during the annealing step, and an increase in the annealing temperature leads to stronger Se diffusion. Se diffuses preferentially through the grain boundaries of the cell, leading to higher Se concentrations at the grain boundaries compared to the grain interiors [22]. Thus, a strong diffusion of Se throughout the absorber layer and segregation of Se at the grain boundaries are observed upon annealing at 480°C. This effect is weaker at lower temperatures, leading to a relatively homogeneous distribution of Se.

B. Low XEOL intensity at grain boundaries

The panchromatic SR-XEOL signal $I_{\text{SR-XEOL}}$ of the three cells is shown in Figs. 1(g)–1(i). The maps are representative of the optical performance at the back of the cells due to an interplay of several effects, the most important ones being (a) internal photon reflection due to the larger refractive index of the absorber in comparison to air, (b) the limited numerical aperture (0.42) and depth of focus ($\approx 3 \mu\text{m}$) of the photon-collecting objective, (c) the focus of the objective being optimized at the back surface of the absorber, (d) a decrease of the x-ray beam intensity while passing the absorber, thus generating fewer electron-hole pairs (smaller XEOL signal) along the beam path in the cell, and (e) higher reabsorption probability of photons generated deeper in the absorber.

Due to the complexity of these effects, a single effective XEOL probe depth cannot be rigorously defined. However, the bandgap gradient within the absorber provides a qualitative argument for an extended probe depth: since the Se concentration generally increases toward the front of the absorber (hence, the bandgap decreases), photons generated deeper in the absorber and traveling toward the back collection side pass through regions of progressively increasing bandgap, reducing their reabsorption probability along the collection path. This suggests that the long-wavelength XEOL signal is collected from a substantial portion of the absorber, albeit with a bias toward the back. We note, however, that the steepness and spatial extent of the bandgap gradient vary with annealing

temperature and exhibit local variations at the grain scale, such that this argument about the effective XEOL probe depth should be understood as qualitative rather than quantitative.

For the cells CdSeTe_{420} and CdSeTe_{450} , it is not evident that characteristic features in $I_{\text{SR-XEOL}}$ would correlate with the corresponding Se maps. This is contrary to the CdSeTe_{480} cell, where a weaker XEOL intensity is observed in the Se-rich (and Cd-poor) regions. Due to two competing effects, this observation is worth a short discussion. On the one hand, a stronger luminescence signal is expected in areas with a higher Se concentration [4,23]. On the other hand, grain boundaries in CdTe solar cells act as non-radiative recombination centers, leading to a weaker XEOL signal than that in grain interiors [4,24–26]. Apparently, the latter effect dominates the trend, and the results indicate that the observed features unveil the grain structure with a high Se concentration and low XEOL intensity at grain boundaries, which is consistent with the negative correlation coefficient of $I_{\text{SR-XEOL}}$ and $I_{\text{XRF, Se}}$ in the cell CdSeTe_{480} ($r_{\text{PCC}} = -0.19$).

C. Impact of Se on the effective bandgap wavelength

While the Se XRF signal is a depth-integrated projection through the full absorber, the XEOL signal is biased toward the back of the absorber. The depth integration of the Se XRF may therefore dilute the lateral Se contrast present at the back surface, potentially underestimating, but not reversing, the correlation between Se concentration and optical properties. Figure 2 shows the averaged XEOL spectrum of each cell. The spectrum of the cell CdSeTe_{420} shows a prominent peak at $\lambda_p = 825$ nm along with an adjacent shoulder toward longer wavelengths up to $\lambda \approx 875$ nm. The prominent peak is attributed to the bandgap of CdTe ($E_{\text{g,CdTe}} \approx 1.5$ eV and therefore $\lambda_{\text{g,CdTe}} \approx 825$ nm). The shoulder is the result of different $\text{CdSe}_x\text{Te}_{1-x}$ species with varying x value. The bandgap energy of $\text{CdSe}_x\text{Te}_{1-x}$ varies between $1.4 \text{ eV} \lesssim E_{\text{g}} \lesssim 1.5 \text{ eV}$ ($886 \text{ nm} \gtrsim \lambda_{\text{g}} \gtrsim 825 \text{ nm}$) with $0.3 \gtrsim x \gtrsim 0$ [27]. Hence, the stronger contribution of $\text{CdSe}_x\text{Te}_{1-x}$ species with higher x values (and longer wavelengths) is another indication of weak Se diffusion from the $\text{CdSe}_x\text{Te}_{1-x}$ front layer toward the back of the cell: the high luminescence at

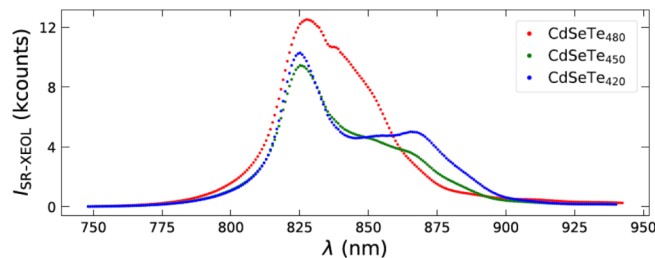


FIG. 2. Averaged SR-XEOL spectra of the three solar cells.

875 nm originates from as-deposited $\text{CdSe}_{0.3}\text{Te}_{0.7}$ that has barely been diluted by the CdCl_2 treatment.

The cell CdSeTe_{450} shows a similar behavior with a prominent peak at $\lambda_p = 825.8$ nm and a shoulder at longer wavelengths. The shoulder is less pronounced than for CdSeTe_{420} , indicating a stronger diffusion of Se into the absorber layer and the formation of $\text{CdSe}_x\text{Te}_{1-x}$ species with smaller x values.

The cell CdSeTe_{480} shows a prominent peak at $\lambda_p \approx 829$ nm along with a shoulder toward longer wavelengths closer to the main peak, indicating the strongest Se diffusion among the investigated cells. The overall red shift, including the prominent peak, is an indication that $\text{CdSe}_x\text{Te}_{1-x}$ is the dominant species in the cell and not CdTe as in the other two cells, which is a consequence of strong Se diffusion upon annealing at 480°C .

D. Spatial distribution of the charge-carrier lifetime

The TR-XEOL decays of each scan pixel were individually analyzed as introduced in Sec. V F, providing the monoexponential charge-carrier lifetime $\tau_{\text{TR-XEOL}}$ of each decay; the resulting maps are shown in Fig. 3. The charge-carrier lifetime maps appear noisy, and the features are not as pronounced as those in the XEOL intensity maps, which is understandable considering the rather poor signal-to-noise ratio of the acquired histograms at each pixel, especially in the region of interest of the decay used for the fitting procedure (as discussed in Sec. V F and Sec. IV in Supplemental Material) [17].

The average lifetime $\langle \tau_{\text{TR-XEOL}} \rangle$ was extracted for each of the cells, resulting in $\langle \tau_{\text{TR-XEOL}} \rangle_{\text{CdSeTe}_{420}} \approx 0.81$ ns, $\langle \tau_{\text{TR-XEOL}} \rangle_{\text{CdSeTe}_{450}} \approx 0.76$ ns, and $\langle \tau_{\text{TR-XEOL}} \rangle_{\text{CdSeTe}_{480}} \approx 1.02$ ns; comparable lifetimes have been observed in similar solar cells in other studies [23,28–31]. As discussed in Sec. IV in Supplemental Material [17], the monoexponential lifetime is governed by a fast decay component and represents a convolution of multiple recombination mechanisms rather than the bulk lifetime alone. Cells CdSeTe_{420} and CdSeTe_{450} yield comparable average lifetimes, whereas cell CdSeTe_{480} exhibits a

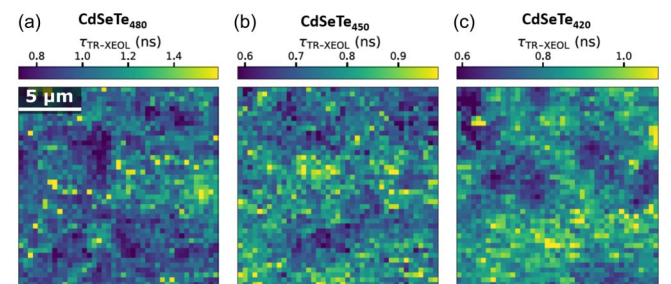


FIG. 3. (a)–(c) Charge-carrier lifetime maps acquired from the analysis of the individual decays measured at each pixel. Each decay was fitted according to the monoexponential function given by Eq. (1).

notably longer lifetime. This nonmonotonic trend is independently confirmed by TR-PL measurements (Table SI in Supplemental Material) [17,32]. A biexponential analysis of cumulated histograms is presented in Sec. II E 2 with improved counting statistics.

E. Differentiation between Se-rich/Se-poor and GB/GI regions

The differentiation between grain boundary (GB) and grain interior (GI) as well as Se-rich and Se-poor regions is key for the interpretation of the results.

GBs and the GIs were defined only for the CdSeTe₄₈₀ cell using the watershed tool provided in ImageJ [33,34]. Therefore, the panchromatic XEOL signal shown in Fig. 1(g) was used to define GBs as regions with low XEOL counts and GIs as regions with high XEOL counts. The resulting GB and GI regions are shown in Figs. 4(a)–4(b).

Se-rich regions of the cells were defined as the pixels in which the Se counts are larger than the 90th percentile of the Se counts, and the Se-poor regions were defined as the pixels in which the Se counts are smaller than the 10th percentile of the Se counts. The Se-rich and Se-poor regions are exemplary shown in Figs. 4(c)–4(d) for the CdSeTe₄₈₀ cell. The Se counts are plotted on top of the panchromatic XEOL signal, signaling that the Se-rich areas are not homogeneously distributed across all GBs but rather that Se segregates at some GBs, partially diffusing into the surrounding GIs; occasionally, GIs close to GBs show high Se concentration.

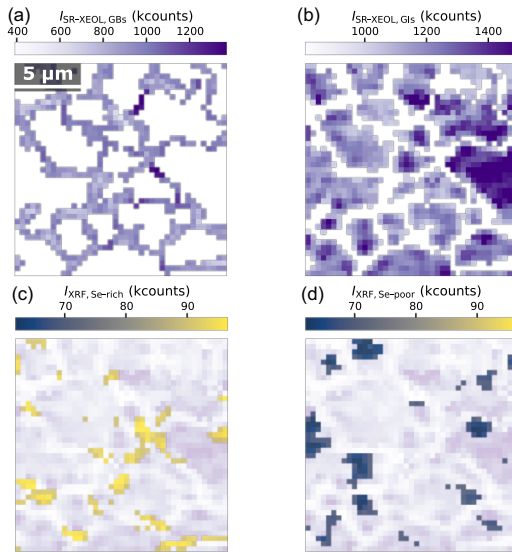


FIG. 4. (a) GBs and (b) GIs of the CdSeTe₄₈₀ solar cell defined via the panchromatic SR-XEOL signal of the scans using the watershed tool provided in ImageJ. GBs are defined as the regions with low SR-XEOL counts and GIs as the regions with high SR-XEOL counts. (c) Se-rich and (d) Se-poor regions of the CdSeTe₄₈₀ solar cell plotted on top of the panchromatic SR-XEOL maps for easier orientation in the cell.

1. Effective bandgap wavelength

The averaged spectra of the Se-rich and Se-poor regions are shown in Figs. 5(a), 5(c), and 5(e) for the three cells, and the ones of the GBs and GIs of CdSeTe₄₈₀ are shown in Fig. 5(g). The spectra of the CdSeTe₄₂₀ and CdSeTe₄₅₀ cells show a noticeable difference in the wavelength region $840 \text{ nm} < \lambda < 870 \text{ nm}$ corresponding to CdSe_xTe_{1-x} with large x .

The prominent peak wavelength for the Se-rich and Se-poor regions of the CdSeTe₄₂₀ and CdSeTe₄₅₀ cells were determined to be $825 \text{ nm} < \lambda_p < 826 \text{ nm}$ corresponding to the bandgap wavelength of CdTe at $\lambda_g \approx 825 \text{ nm}$. Thus, the Se diffusion into the absorber was not strong enough to impact the effective bandgap of the material that is governed by CdTe.

In contrast, a significant discrepancy can be observed between the Se-rich and Se-poor spectra for the CdSeTe₄₈₀

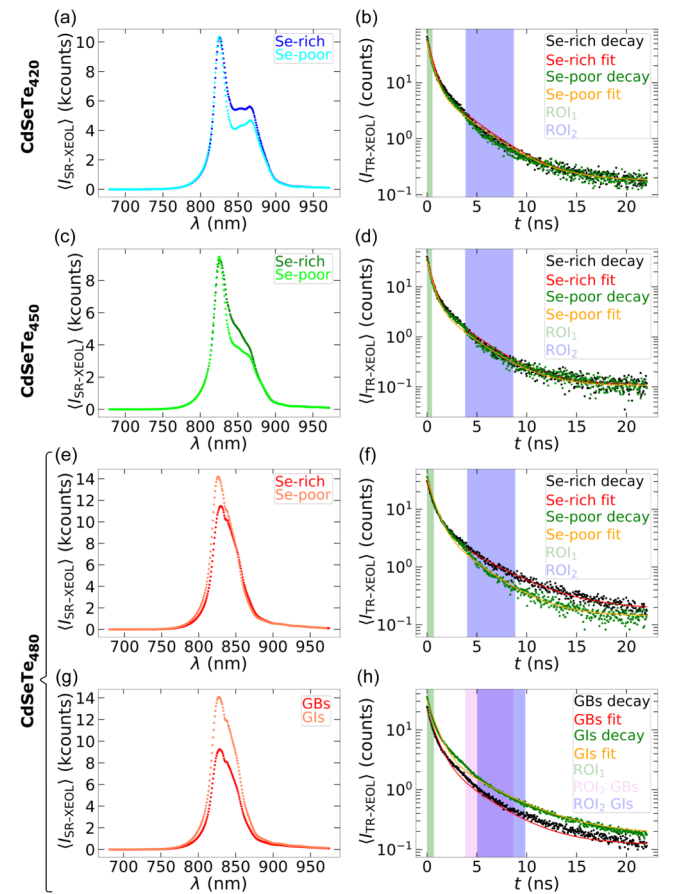


FIG. 5. (a),(b) Comparison of the spectra and decays of the Se-rich and Se-poor regions of cell CdSeTe₄₂₀, (c),(d) of cell CdSeTe₄₅₀, and (e),(f) of CdSeTe₄₈₀. (g) and (h) Comparison of the spectra and decays of the GB and GI regions of cell CdSeTe₄₈₀. The decays are shown along with their respective bi-exponential fits following Eq. (2) and the ROIs defined for the fitting procedure as explained in Sec. IV in Supplemental Material [17].

cell: the high Se concentration in the Se-rich regions leads to the above-mentioned overall red shift of the spectrum and, most importantly, to a red shift of the prominent peak wavelength to $\lambda_p \approx 830$ nm. In contrast, the Se-poor regions show a prominent peak wavelength at $\lambda_p \approx 826$ nm similar to the other two cells. Thus, the Se diffusion through the absorber at high annealing temperature is strong enough to impact the effective bandgap wavelength of the material at the back of the cell, especially in regions where Se segregates.

Considering GBs vs GIs, both GB and GI regions of the cell CdSeTe₄₈₀ exhibit a red shift of the prominent peak wavelength, yielding $\lambda_p \approx 829$ nm and $\lambda_p \approx 827$ nm, respectively. Note that GBs show a slightly weaker red shift than Se-rich regions, and GIs show a stronger red shift than Se-poor regions, suggesting that Se concentration is not only localized at GBs but also diffused into GIs. This supports the need to differentiate between Se-rich, Se-poor, GB, and GI regions. Moreover, the XEOL intensity at GBs and in Se-rich regions is significantly lower than at GIs and in Se-poor regions. These observations are attributed to a higher concentration of recombination centers at GBs enhancing Shockley-Read-Hall recombination, which is expected to decrease the charge-carrier lifetime. However, it is acknowledged that the statistics is too low for such a fourfold segmentation.

2. Charge-carrier lifetime

The averaged decays of the Se-rich and Se-poor regions for the three cells are presented in Figs. 5(b), 5(d), 5(f), along with their fits and the ROIs used for the definition of τ_1 and τ_2 . The averaged decays were analyzed with the procedure described in Sec. V F and Sec. IV in Supplemental Material [17] and fitted with a biexponential decay following Eq. (2). The values of τ_1 and τ_2 obtained from the fits are summarized in Table I, along with the intensity-weighted average charge-carrier lifetime $\langle\tau\rangle_{\text{Int}}$.

The decays of the Se-poor and Se-rich regions in the cells CdSeTe₄₂₀ and CdSeTe₄₅₀ overlap strongly, indicating similar charge-carrier kinetics in the Se-rich and Se-poor regions. Correspondingly, no significant differences are observed between the lifetimes in Se-rich and

Se-poor regions. In contrast, the CdSeTe₄₈₀ cell shows an overall improvement in all charge-carrier lifetimes compared to the other two cells; the improvement is stronger in the Se-rich regions than in the Se-poor regions. Note that τ_2 , which gives the best estimate of the bulk charge-carrier lifetime, exhibits about a 32% longer lifetime in the Se-rich regions of the CdSeTe₄₈₀ cell compared to the Se-poor regions of the cell. The observations are consistent with those from the XEOL spectra: Se diffusion is strong enough to dominate the material properties only at high annealing temperatures.

This is further corroborated by the observation that the Se-poor regions of the CdSeTe₄₈₀ cell yield $\tau_2 \approx 3.09$ ns, comparable to the values obtained in both regions of cells CdSeTe₄₂₀ and CdSeTe₄₅₀ ($\tau_2 \approx 2.79$ ns to 3.07 ns).

The averaged decays of the GB and GI regions of the CdSeTe₄₈₀ cell are shown in Fig. 5(h). The GI regions exhibit a stronger XEOL signal than the GB regions, in accordance with the intensities of the XEOL spectra shown in Fig. 5(g). The extracted charge-carrier lifetimes τ_1 , τ_2 , and $\langle\tau\rangle_{\text{Int}}$ are summarized in Table I, where considerably larger lifetimes are observed in the GI regions compared to the GB regions. This is expected as enhanced charge-carrier recombination from nonradiative defects at GBs aligns with the observed weakening of the XEOL signal at those sites. A more aggressive CdCl₂ treatment increases the charge-carrier lifetime not only at GBs but also in GIs.

III. DISCUSSION

A. Role of Se in CdSe_xTe_{1-x}

The multimodal x-ray microscopy approach combining XRF and XEOL allowed us to relate the spatial distribution of Se in solar cells annealed at different temperatures to its impact on the optical properties of the absorber. We found that the Se diffusion from the front to the back of the absorber layer during the annealing step at lower temperatures was weak, leading to a rather homogeneous lateral distribution of Se and to the formation of small grains, which could not be clearly resolved with the given measurement parameters. In those cells, no impact of Se on the effective bandgap wavelength and charge-carrier lifetime of the material could be observed. In contrast,

TABLE I. Prominent peak wavelength λ_p , charge-carrier lifetimes τ_1 , τ_2 , and $\langle\tau\rangle_{\text{Int}}$ extracted from the spectra and decays of the Se-rich and Se-poor regions of the three investigated solar cells and of the GB and GI regions of cell CdSeTe₄₈₀.

Material parameter	CdSeTe ₄₂₀		CdSeTe ₄₅₀		CdSeTe ₄₈₀			
	Se-rich	Se-poor	Se-rich	Se-poor	Se-rich	Se-poor	GBs	GIs
λ_p (nm)	825	825	826	825	830	826	829	827
τ_1 (ns)	0.52	0.50	0.46	0.48	0.71	0.65	0.76	0.79
τ_2 (ns)	3.06	3.07	2.81	2.79	4.08	3.09	3.42	4.30
$\langle\tau\rangle_{\text{Int}}$ (ns)	1.81	1.83	1.64	1.58	2.52	1.78	1.99	2.37

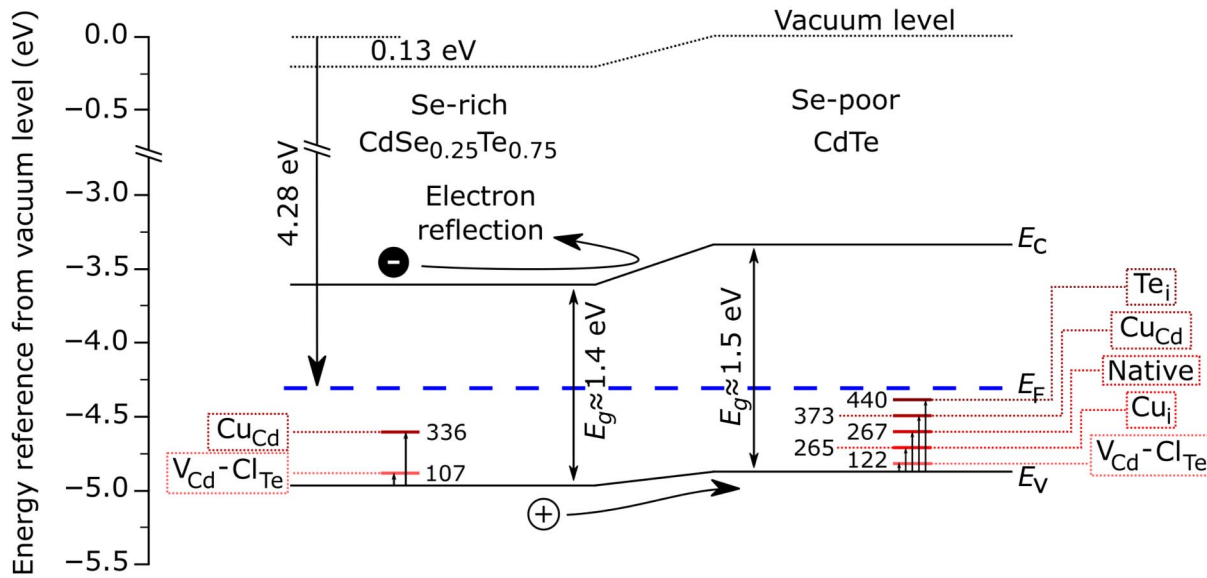


FIG. 6. Hypothetical effect of electron reflection at the interface between a Se-rich $\text{CdSe}_x\text{Te}_{1-x}$ (left, $x \approx 0.25$) and Se-poor $\text{CdSe}_y\text{Te}_{1-y}$ (right, $y \approx 0$) region in CdSeTe solar cells. Based on energy-band values from Ref. [45], the energy barrier formed in the conduction band at the interface acts as an electron reflector, hindering the diffusion of electrons from the Se-rich to the Se-poor region, hence reducing recombination. The band-bending effects should show the same trends for all interfaces as long as $0 \leq y < x \leq 0.3$. The red lines in the bandgap represent possible trap states that enhance the Shockley-Read-Hall recombination rate and decrease the charge-carrier lifetime. Their respective activation energies are shown in meV above the valence band edge E_V and were taken from measurements performed by Artegiani *et al.* [35,36].

after a high annealing temperature of 480°C , the presence of Se toward the back of the absorber led to a red-shift of the effective bandgap wavelength, indicating that Se diffusion into the absorber was strong enough to alter the optical properties of the material at the back of the cell. Moreover, segregation of Se at GBs and the surrounding GIs was observed for this cell, accompanied by a prolongation of the charge-carrier lifetime, especially at Se-rich regions. The longer charge-carrier lifetimes in the Se-rich regions can be explained by a combination of two main effects, as schematically shown in Fig. 6.

1. Chemical passivation effects due to Se

Deep-level defects in the bandgap of the absorber layer act as detrimental recombination centers, leading to shorter charge-carrier lifetimes. Exemplary trap states are shown in Fig. 6, as measured by Artegiani *et al.* [35,36]. The Se-poor region, shown on the right side of the figure, exhibits defect centers that are both more numerous and located deeper in the bandgap than those in the Se-rich region shown on the left side [37]. The trap states expected in the Se-poor region originate from $\text{V}_{\text{Cd}}\text{-Cl}_{\text{Te}}$ complexes probably formed during the CdCl_2 treatment, interstitial copper atoms (Cu_i) that have been characterized as electron traps, native defects (such as Cd interstitials (Cd_i), Te vacancies (V_{Te}), and Cd antisite (Cd_{Te}) [38]),

copper impurities at cadmium sites (Cu_{Cd}) that improve the p -type conductivity [39], and Te interstitials (Te_i) that are deep-level defects and could limit the charge-carrier lifetime [40]. The native defects can be suppressed by adding copper [41], however, possibly leading to a higher concentration of Cu_i and Cu_{Cd} defect centers.

Se increases the solubility of Cu in the lattice site, effectively suppressing the Cu_i and the native defect centers, both of which are not significant in the Se-rich regions. In addition, the presence of Se appears to reduce the activation energy of the Cu_{Cd} defect centers [35,42,43]. Deep-level Te_i defect centers could act as the main recombination centers in CdTe solar cells and be most detrimental to the charge-carrier lifetime. The absence of these recombination centers in the Se-rich region can be attributed to a change in the stoichiometry due to the introduction of Se, leading to a lower concentration of Te in the Se-rich regions than in the Se-poor regions and could substantially account for the prolonged charge-carrier lifetime [36]. The mitigation of defect centers in the Se-rich region strongly suggests that effective chemical passivation effects are introduced by the addition of Se and its diffusion during the CdCl_2 annealing step, reducing the recombination rate and prolonging the charge-carrier lifetime. The exact passivation mechanism and the strength of the passivation effects depend on the local crystal structure [44] and are the subject of current research.

2. Band bending at the interface between the Se-rich and Se-poor regions

$\text{CdSe}_x\text{Te}_{1-x}$ with $x < 0.3$ has lower conduction- and valence-band-edge energies than CdTe [45]. Hence, an energy barrier is formed in the conduction band at the interface between the two materials that can act as an electron reflector, keeping the minority carriers in the Se-rich and better passivated regions and preventing them from diffusing to the Se-poor regions with a higher recombination probability.

The bandgap energy of $\text{CdSe}_x\text{Te}_{1-x}$ has a minimum around $x \approx 0.3$ [27]. Thus, the strength of the band bending at internal interfaces in $\text{CdSe}_x\text{Te}_{1-x}$ depends on the local Se concentration and varies with x [46].

Furthermore, the strength of these band-bending effects depends on the local crystal orientation [45,47,48].

B. Ramifications of injection level

The x-ray excitation differs from the intended 1-sun operation of the solar cells both in the pulsed time structure and in the highly focused injection of charge carriers by x-rays. Each pulse generates a locally quasi-instantaneous excess carrier concentration $\Delta n \approx 2 \times 10^{17} \text{ cm}^{-3}$, far above the doping ($\approx 10^{14} \text{ cm}^{-3}$) and the steady-state 1-sun level ($\approx 10^{12} \text{ cm}^{-3}$), as detailed in Sec. III in Supplemental Material [17,49]. This high initial injection could partially saturate shallow traps and enhance Auger recombination shortly after the pulse, but the bulk lifetime τ_2 is extracted from ROI_2 several nanoseconds later, when the carrier concentration has decayed by orders of magnitude. We note that carriers slowly released from trap states may still contribute to this part of the decay, so the low injection reached at later times is not strictly equivalent to a measurement performed at low injection from the outset.

The time-averaged excitation rate within the probed volume is $\bar{G}_X = \Delta n / \Delta t_{\text{pulse}} \approx 10^{24} \text{ cm}^{-3} \text{ s}^{-1}$, about 3 orders of magnitude above the 1-sun value $\bar{G}_{1\text{-sun}} \approx 5 \times 10^{20} \text{ cm}^{-3} \text{ s}^{-1}$. In the $\text{CdSe}_x\text{Te}_{1-x}$ system, deep point defects have detrapping times far exceeding the 192 ns pulse separation [50]. Hence, trap populations do not reset between pulses but may accumulate across pulse cycles up to an upper limit set by $\bar{G}_X$. The deep-trap occupation may thereby be driven closer to saturation than under 1-sun operation, though the complex spatiotemporal relation between pulsed local charge-carrier injection in a scanning scheme, Fermi-level splitting, and trap occupation is beyond the scope of this study.

The absolute charge-carrier lifetimes reported here are thus representative of the x-ray excitation conditions and are suitable for a qualitative comparison of the charge-carrier lifetime within individual cells and across a series

of cells, rather than for a quantitative evaluation under solar irradiation.

IV. CONCLUSIONS

Based on the results presented here, the following can be concluded:

- (i) A high annealing temperature of 480°C leads to an overall improvement in the charge-carrier lifetime in $\text{CdSe}_x\text{Te}_{1-x}$ solar cells compared to cells annealed at lower temperatures. Especially in Se-rich regions, a significantly longer charge-carrier lifetime was observed, along with a red shift of the effective bandgap wavelength, indicating positive consequences arising from the chemical passivation and band-bending effects of Se. In the case of lower annealing temperatures of 420 and 450°C, these positive effects could not be observed, suggesting that the diffusion of Se in the absorber layer is too weak to effectively passivate the defect centers in these cells and induce band bending.
- (ii) The local lifetime improvement in Se-rich regions of cell CdSeTe_{480} does not translate into superior device performance: the 480°C cell exhibits the lowest conversion efficiency ($\eta = 14.0\%$) and open-circuit voltage ($V_{\text{OC}} = 720 \text{ mV}$) of the series (Table SI in Supplemental Material). This apparent contradiction is explained by the pronounced lateral nonuniformity of the CdSeTe_{480} cell ($\sigma_{\text{Rel}} = 9.4\%$ for the Se count map): while Se-rich regions benefit from chemical passivation and band bending resulting in $\tau_2 \approx 4.08 \text{ ns}$, the Se-poor regions yield $\tau_2 \approx 3.09 \text{ ns}$, comparable to those of cells annealed at lower temperatures. In a parallel-diode picture, these Se-poor regions with higher recombination rates limit V_{OC} , outweighing the gains in the Se-rich regions. The cells annealed at 420 and 450°C, while lacking locally enhanced lifetime, benefit from a more spatially uniform absorber quality.
- (iii) GBs exhibit a shorter charge-carrier lifetime than GIs. Notably, the lifetime enhancement associated with Se-rich regions exceeds the lifetime reduction attributable to GB, indicating that Se content is the more dominant factor governing local charge-carrier lifetime. This is consistent with Se passivation playing a significant role even at structurally defective GB regions, though we note that GBs are not uniformly Se-rich and regions at GBs could be observed, where Se segregated and partially diffused into the adjacent GIs. Hence, the overall improvement in the charge-carrier lifetime of the device in comparison to the cells annealed at lower temperatures is attributed not only to the passivation effects at the GBs but also to the passivation effects in the GIs.

V. EXPERIMENTAL PROCEDURES

A. Sample preparation

Three Cu-doped $\text{CdSe}_x\text{Te}_{1-x}$ solar cells were manufactured in the typical superstrate configuration [51] at the National Laboratory of the Rockies (NLR), Colorado, USA. NSG TECTM12D glass was cleaned and loaded into an Angstrom Engineering evaporator for absorber deposition. $\text{CdSe}_{0.3}\text{Te}_{0.7}$ and CdTe were sequentially deposited to thicknesses of 500 nm and 3.5 μm , respectively, using 99.999% purity source material from 5N Plus. The samples were then CdCl_2 treated using a vapor-based close-space sublimation process in a He ambient for 10 min at 480, 450, or 420°C; the solar cells are correspondingly denoted CdSeTe_{480} , CdSeTe_{450} , and CdSeTe_{420} . During this CdCl_2 -treatment/annealing step, Se diffused from the $\text{CdSe}_{0.3}\text{Te}_{0.7}$ regions toward the CdTe region, forming a Se-graded absorber layer. After the CdCl_2 treatment, the samples were immersed in a solution of 0.1 mM CuCl_2 in deionized (DI) water for 3 min, rinsed in DI water, and then annealed in air for 25 min at 210°C. The back contacts that defined the actual solar-cell area were finally formed by 100 nm of evaporated gold (Au). The thick glass prohibited x-ray beam measurements from the front side. Hence, the measurements had to be performed from the back of the solar cells. There, the part next to the Au-covered cell had the absorber exposed, enabling optical measurements.

B. Measurement settings

The x-ray measurements were conducted at the hard X-ray Micro/Nano-Probe Beamline P06 (microprobe end-station) at PETRA III (DESY, Germany) using a beam energy of 13.6 keV and Kirkpatrick-Baez mirrors (KB-mirrors) to focus the beam on the sample. The beam size was measured as $v \times h \approx 305 \text{ nm} \times 355 \text{ nm}$. The photon flux at the sample position was approximately 10^9 photons/s. PETRA III was operated in 40-bunch mode, resulting in x-ray pulses separated by $\approx 192 \text{ ns}$. The sample was fixed to a PCB [52] and mounted on a sample-stage tower consisting of a hexapod (for coarse scans and alignment in x , y , and z), a rotation stage, two linear centering stages, and a 3D piezo stage for precise scanning in x , y , and z . The measurements were performed by raster scanning the sample in the plane of the x-ray focus [53].

High-resolution step scans of an area of $16 \times 16 \mu\text{m}^2$ of each cell were performed with 41×41 pixels, resulting in a step size of 400 nm, and with a pixel dwell time of 2 s. The long pixel dwell time was chosen to enhance the signal across the modalities, which was particularly important for TR-XEOL. XRF, SR-XEOL, and TR-XEOL data were simultaneously collected at each measurement pixel of the scans.

C. Experimental setup

The current version of the XEOL detection unit, the AXOLOTL 2.0, presents an improved version of the AXOLOTL 1.0 setup introduced by Ossig *et al.* [12] and was rebuilt from scratch. The XEOL signal was collected via an optical mirror with a Mitutoyo MY50X-825 objective, which directed the signal as an infinity-corrected beam to the XEOL detection unit. A single-element Vortex fluorescence detector was placed at 45° between the sample and the incoming x-ray beam for elemental mapping via XRF.

A SuperKExtreme EXU6 laser unit, together with a SuperK VARIA filter system, was used for the alignment of the XEOL unit with the optical mirror and the x-ray focus. TR-XEOL measurements were performed using a Si-based PD-050-CTE APD from Micro Photon Devices paired with a Picoquant Hydraharp 400 TCSPC unit. SR-XEOL measurements were performed using an Andor Kymera 328i spectrograph paired with a Si-based CCD, back-illuminated Andor Newton camera, covering the VIS and NIR-IR range of the XEOL signal. The measurements shown here were taken with a 100 lines/mm grating blazed at 780 nm.

D. XRF analysis

The XRF spectra collected at each pixel were fitted with PyMca [54]. At $E_{\text{x-ray}} = 13.6 \text{ keV}$ incident photon energy, the XRF measurements were sensitive to the signal from the K edges of the main element of interest, Se, where the absorption edge is close to the beam energy and the energy of the emitted photons is high enough that they do not suffer substantially from self-absorption ($E_{\text{Se}_{K\alpha}} = 11.22 \text{ keV}$, $E_{\text{Se}_{K\beta}} = 12.50 \text{ keV}$).

The mapping of Cd and Te had to rely on the XRF L-emission lines, which impaired the quantification due to self-absorption artifacts.

E. SR-XEOL analysis

The locally measured SR-XEOL spectrum depends on the amount of various $\text{CdSe}_x\text{Te}_{1-x}$ species present in the beam-sample interaction volume. Since we were interested in the shift of the prominent peak of the spectrum due to the presence of Se, a semiquantitative analysis of the spectra was chosen, where the wavelength at which the SR-XEOL intensity is maximal is determined and used to describe the prominent peak wavelength of the spectra.

F. TR-XEOL analysis

The signal amplitude of the acquired TR-XEOL data scales linearly with the integration time chosen for each pixel of a scan. The large-area scans (presented in Sec. V in Supplemental Material [17]) resulted in very few counts per pixel, excluding any reasonable fitting of the decays. In contrast, the increased pixel dwell time during the

small-area scans (presented in the main article) allowed fits with a monoexponential function at each measurement point.

Throughout the analysis of the TR-XEOL data, pixels of selected areas of interest in the scans were averaged, resulting in high-statistics decays that were fitted with a biexponential function. Hence, two approaches were chosen to analyze the TR-XEOL decays:

Decays following a monoexponential form were fitted using:

$$I_{\text{XEOL}}(t) = A_{\text{TR-XEOL}} \cdot \exp\left[-\frac{t}{\tau_{\text{TR-XEOL}}}\right] + C, \quad (1)$$

where $A_{\text{TR-XEOL}}$ is the amplitude of the signal, $\tau_{\text{TR-XEOL}}$ is the charge-carrier lifetime, and C is an offset originating from the measurement environment and dark counts of the detector.

Decays following a biexponential form were fitted using:

$$I_{\text{XEOL}}(t) = A_0 \cdot A_1 \cdot \exp\left[-\frac{t}{\tau_1}\right] + A_0 \cdot A_2 \cdot \exp\left[-\frac{t}{\tau_2}\right] + C, \quad (2)$$

where A_0 is the intensity at the start of the decay, τ_1 and τ_2 are the decay constants corresponding to charge-carrier lifetimes, A_1 and A_2 are the fractional amplitudes of the first and second ROIs, respectively, and C is an offset originating from the measurement environment and dark counts of the detector.

Usually, the first and fastest part of the decay τ_1 is attributed to charge-separation effects and the second and slower part of the decay τ_2 is dominated by recombination and gives the best estimate of the bulk lifetime [23,30,47,55–61].

The acquired parameters from the biexponential decay analysis allowed the calculation of the intensity-weighted average charge-carrier lifetime $\langle\tau\rangle_{\text{Int}}$ as

$$\langle\tau\rangle_{\text{Int}} = \frac{\sum_{i=1}^n A_i \cdot \tau_i^2}{\sum_{i=1}^n A_i \cdot \tau_i}, \quad (3)$$

where τ_i is the lifetime of exponential i , and A_i the corresponding fractional intensity [62–64].

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J. L. Jr. B., Gero F., E. C., and M. E. S. designed the study. Gi. F., J. L. Jr. B., S. P., J. G., Gero F., C. S., T. K., and M. E. S. designed the instrument; J. L. Jr. B., Gero F., Gi. F., S. P., N. P., S. H., J. G., Gera F., C. S., T. K., and M. E. S. conducted the measurements; E. C. and A. M. prepared and precharacterized the samples; J. L. Jr. B. prepared the manuscript. N. P., E. C., and M. E. S. revised the manuscript; All authors were involved with feedback and discussions about the manuscript and data analysis.

The authors declare no competing interests.

DATA AVAILABILITY

The raw data of this study are published under Ref. [65].

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