

Mapping the Local Photo-Electronic Properties of Polycrystalline Solar Cells through High Resolution Laser-Beam-Induced Current Microscopy

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Abstract—To boost the efficiency of thin film polycrystalline solar cells that are microscopically inhomogeneous, it is imperative to understand how the grain interiors (GIs) and grain boundaries (GBs) within these materials affect its overall electronic properties. By using an apertured near-field scanning optical microscope in an illumination mode we determined the local photocurrent generated within the GIs and at the GBs with nanoscale resolution, and correlate the results with surface morphology and composition.

Index Terms—Cadmium compounds, Current measurement, Grain boundaries, Photovoltaic cells, Scanning probe microscopy, Thin film devices, Wavelength measurement.

I. INTRODUCTION

THE understanding of how the grain interiors (GIs) and grain boundaries (GBs) affect and favor the overall performance of polycrystalline thin film solar cells is still an open question [1-3]. The presence of grains with different crystallographic orientations (Fig. 1(a)) induces microscopic dislocations, vacancies, and distorted bonds at the interfaces, among other imperfections in the crystal lattice. In CdTe/CdS solar cells these defects introduce extra electronic states and, consequently, significant band bending, as represented in Figs. 1(b) and (c). In the case of a *p*-type material, these gap states,

also called ‘trap states’, are spatially localized and trap holes, giving rise to an accumulation of positive charges p at the boundaries. The presence of GBs affects, therefore, the transport properties of both majority and minority carriers, and the overall performance of the polycrystalline device, in particular the open circuit voltage (V_{oc}) [1]. Historically, the somewhat beneficial role of the GBs in CdTe/CdS devices has been mainly attributed to built-in electric fields [4, 5], while the composition variations within the polycrystalline grains [6, 7] were also discussed. In the latter case, S from the CdS layer was found to preferentially inter-diffuse into the GBs as a result of grain-boundary-assisted diffusion mechanism, leading to a CdTe_{1-x}S_x ternary phase.

Presently, there is a great effort to engineer and increase the V_{oc} of polycrystalline CdTe/CdS devices. Although the bandgap of CdTe is 1.44 eV, the V_{oc} of world record cells is

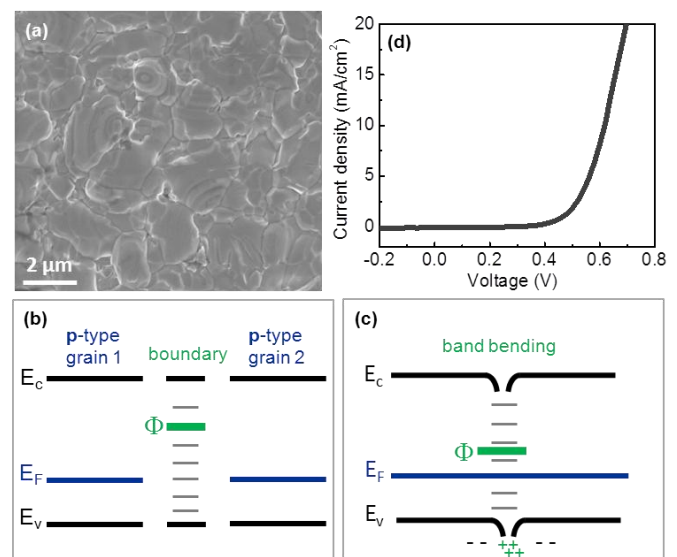


Fig. 1. (a) SEM image of thin-film CdTe layer showing microscale grains and interfaces with structural defects. Illustrations of (b) band diagram for two grains and (c) the band bending caused by the interface states. The extra states trap holes, resulting in electric field on both sides of the grain boundary. E_c , E_F , E_v , and Φ refer to the conduction, Fermi and valence energies, and the neutrality level, respectively. (d) Dark J-V curve showing diode behavior of CdTe/CdS solar cell device.

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currently limited at 0.86 V, leading to an efficiency of 18.7 % [8, 9]. For total area modules, the current V_{oc} record is 0.90 V, with efficiency record of 16.1 % under global AM1.5 illumination [10]. The significant difference between the material bandgap and the best qV_{oc} achieved is limited by a consistently high forward dark current (see Fig. 1(d)) and indicates that proper material engineering and device processing can lead to higher V_{oc} (≈ 1.1 V), comparable to III-V compound semiconductor solar cells, such as GaAs. The open circuit voltage in CdTe solar cells can be increased through the optimization of the device structure, including the microstructure of the absorber layer, and by a better control of the dopant distribution within the p -layer. Moreover, segregation and defect passivation can limit the recombination and leakage current within the polycrystals.

Here, we use laser-beam-induced current (LBIC) microscopy to measure the photo-electronic properties of the GIs and GBs. LBIC microscopy is used to spatially resolve and quantify the current distribution within the CdTe polycrystalline layer of the solar cell. This technique allows us to assess composition variations at the GBs, which can give us valuable information about the recombination sources at the grains' interfaces.

II. LASER-BEAM-INDUCED CURRENT MICROSCOPY

Several scanning probe microscopy techniques, including conductive atomic force, Kelvin probe and capacitance microscopies, have been employed to characterize the role of GBs on the performance of CdTe/CdS and CIGS solar cells [3, 11-18]. Nevertheless, often scanning probe measurements do not reproduce the operation conditions of the solar cells. Here, we use LBIC microscopy [6, 19-21] to locally probe the photoconductivity of the polycrystals constituting CdTe/CdS solar cells with nanoscale resolution. By using a sub-wavelength apertured near field scanning optical microscope (NSOM) in an illumination mode we determine the local photocurrent generated within the GIs and at the GBs through the electrical signal detection of the *in operando* device under different illumination conditions. Measurements are performed

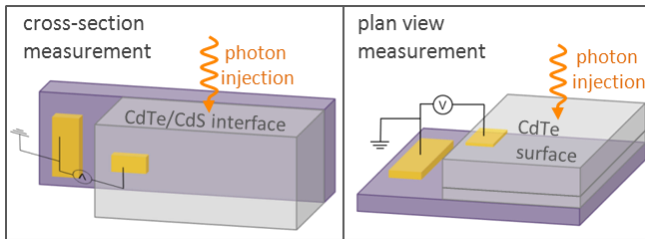


Fig. 2. Illustration of LBIC microscopy setup, which allows for both cross-section and plan-view measurements. In both cases, photons are injected through an apertured NSOM probe. The wavelength of the photons can be selected by using various laser sources, permitting the investigation of bandgap variations and/or sub-gap absorption within the polycrystalline material.

at multiple wavelengths exciting CdTe above, near and below the bandgap with the aim to differentiate between the effects of the built-in electric fields and the local variations of optical absorption related to compositional inhomogeneity. While other techniques such as electron beam induced current microscopy (EBIC) [17],[22] can detect local variation of collection due to built-in electric fields, the additional advantage of LBIC lies on the fact that the photocurrent signal can be also be sensitive to local absorption and the compositional variation depending on the wavelength of the excitation source.

Fig. 2 shows a schematic of the LBIC microscopy setup, which allows for both cross-section and plan-view measurements on the device. Topography and photocurrent were acquired simultaneously using a XYZ-*piezo* feedback-controlled setup. Therefore, the surface topography and electrical signal can be directly correlated. Incident light wavelength was varied either using an optical parametric oscillator (OPO) laser or a supercontinuum laser with the appropriate combination of band-pass and neutral density filters. The photons are injected through an apertured NSOM probe. Nanoscale spatial resolution is achieved by placing the tapered fiber probes (with 50 nm to 500 nm in diameter) extremely close to the surface of the grains (10 nm), therefore providing a local source of excitation. Thus, the resolution is primarily affected by the probe diameter and the materials' absorption coefficient α . The transmittance of the optical fibers

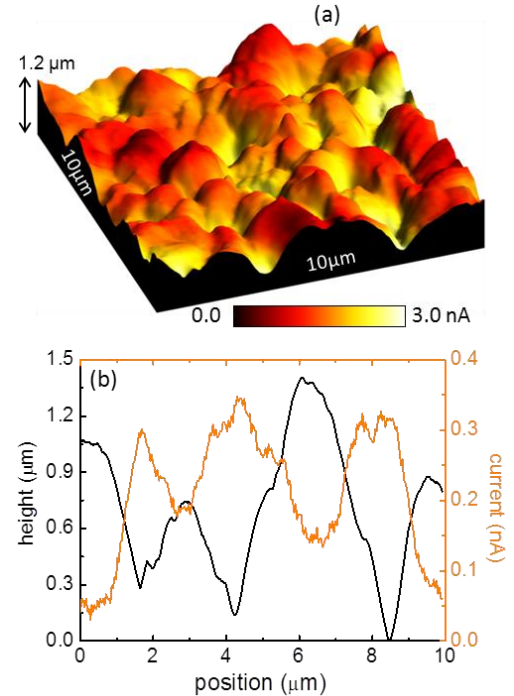


Fig. 3. (a) 3D topography overlaid with LBIC microscopy measurement of p -CdTe layer showing. The color scale refers to the photocurrent generated by the solar cell. (b) Line profile showing consistent better current transport at grain boundaries. Illumination source: 532 nm laser, power = 14 mW, NSOM probe = 300 nm.

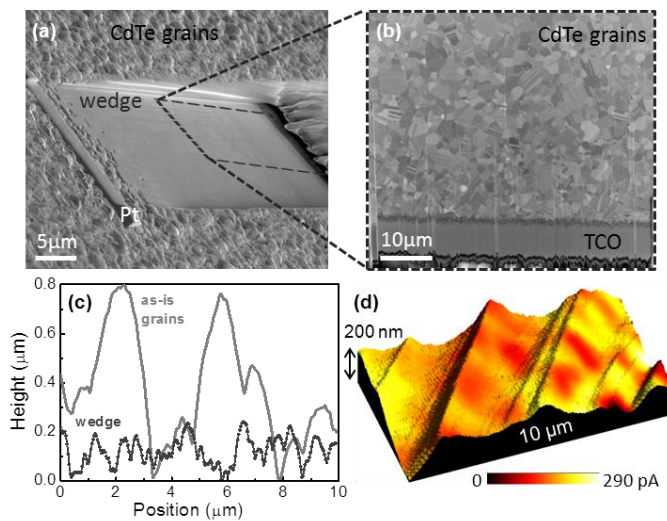


Fig. 4. (a)-(b) SEM images of wedge milled by Ga focused ion beam. (c) Surface topography of CdTe grains before and after milling process. (d) 3D topography overlaid with photocurrent measurement. Illumination source: 532 nm laser, power = 2 mW, NSOM probe = 200 nm.

used vary from 10^{-5} to 10^{-1} , depending on the diameter of the fiber. For 300 nm probes we measured transmittance of $\sim 10^{-4}$.

The samples used in this work are commercially available CdTe solar cells formed by the following layers (from top to bottom): 4.0 mm of glass substrate, 550 nm of $\text{In}_2\text{O}_3/\text{SnO}_2$ (bilayer transparent conductive oxide - TCO), 50 nm of n -type CdS, and 3.5 μm of p -type CdTe. The LBIC microscopy measurements were performed on the exposed p -doped CdTe grains of the back side of the cell injecting light in a region without back contact. 100 nm thick platinum contact pads were evaporated through stencil masks, and the measurements were performed with the tip positioned off the contact edge. As a result, the tip can be placed in a close proximity to CdTe surface to analyze variations of photocurrent within the CdTe grain, as well as at the GIs and GBs' surface. In this particular geometry, holes (p) and electrons (n) are majority and minority carriers, respectively.

Most of the LBIC measurements were performed on 'as-is grains' injecting light at different wavelengths through 200 to 400 nm NSOM probes. The plan view LBIC microscopy measurements consistently showed higher photocurrent generation at the GBs than at the GIs, as shown by the representative image in Fig. 3(a). The overlay between the 3D topography with the photocurrent measurement shows the spatial distribution of photocurrent and its correlation with the surface topography (valleys at GBs), as highlighted by the line profiles shown in Fig 3(b).

To evaluate the possible effects of rough topography on local light absorption, smooth surfaces were obtained by milling wedges at grazing angles using a Ga focused ion beam (FIB), as shown in Fig. 4(a). A sequence of low-current milling steps at grazing incidence ($5-7^\circ$) was used during the wedge milling process in order to preserve the CdTe polycrystals (Fig. 4(b)). This unique geometry allowed us to deconvolute topography effects from the photocurrent

variations at the material surface. The line profiles in Fig. 4(c) display the surface of the same CdTe sample before (as-is grains, in light grey) and after milling the wedge (dotted line, dark grey). As expected, the wedge fabrication dramatically modified the topography of the CdTe grains, making them significantly smoother. The presence of a randomly oriented texture (with root mean square of 0.1 μm in height) is a direct result of the originally rough surface. This well-known 'curtain effect' does not affect the structural properties of the grains, as confirmed by electron back-scattering diffraction measurements. The overlay of a representative LBIC map with the corresponding wedge surface showed that the higher photocurrent was indeed generated in the vicinity of GBs. This result demonstrates that variations in photocurrent are in fact due to inhomogeneities within the GIs and GBs of the device that can be related to the band bending efficiently separating electrons and holes (Fig. 4(d)).

III. PHOTOCURRENT DISTRIBUTION IN CdTe SOLAR CELLS

To assess the bandgap and, therefore, stoichiometry variations within the GBs, separating these from the effect of built-in electric field, we measured the same grains in plan-view (Fig. 2) exciting CdTe at different wavelengths (λ) from well above the bandgap, to close and below the band edge (Fig. 5). Optical filters were used to adjust the intensity of the focused laser illumination.

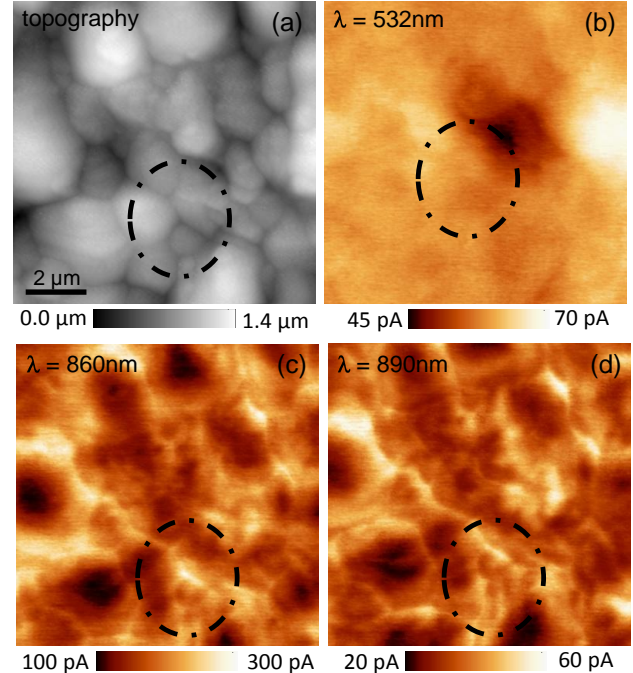


Fig. 5. (a) Topography and (b)-(d) LBIC microscopy measurements of p -CdTe layer under different illumination wavelengths (λ). The current scale was adjusted to clearly show the contrast between GIs and GBs in all images. The highlighted area shows variations in contrast due to composition variations within the grain boundaries. Illumination source: OPO laser, power ≈ 1 mW, probe diameter = 300 nm.

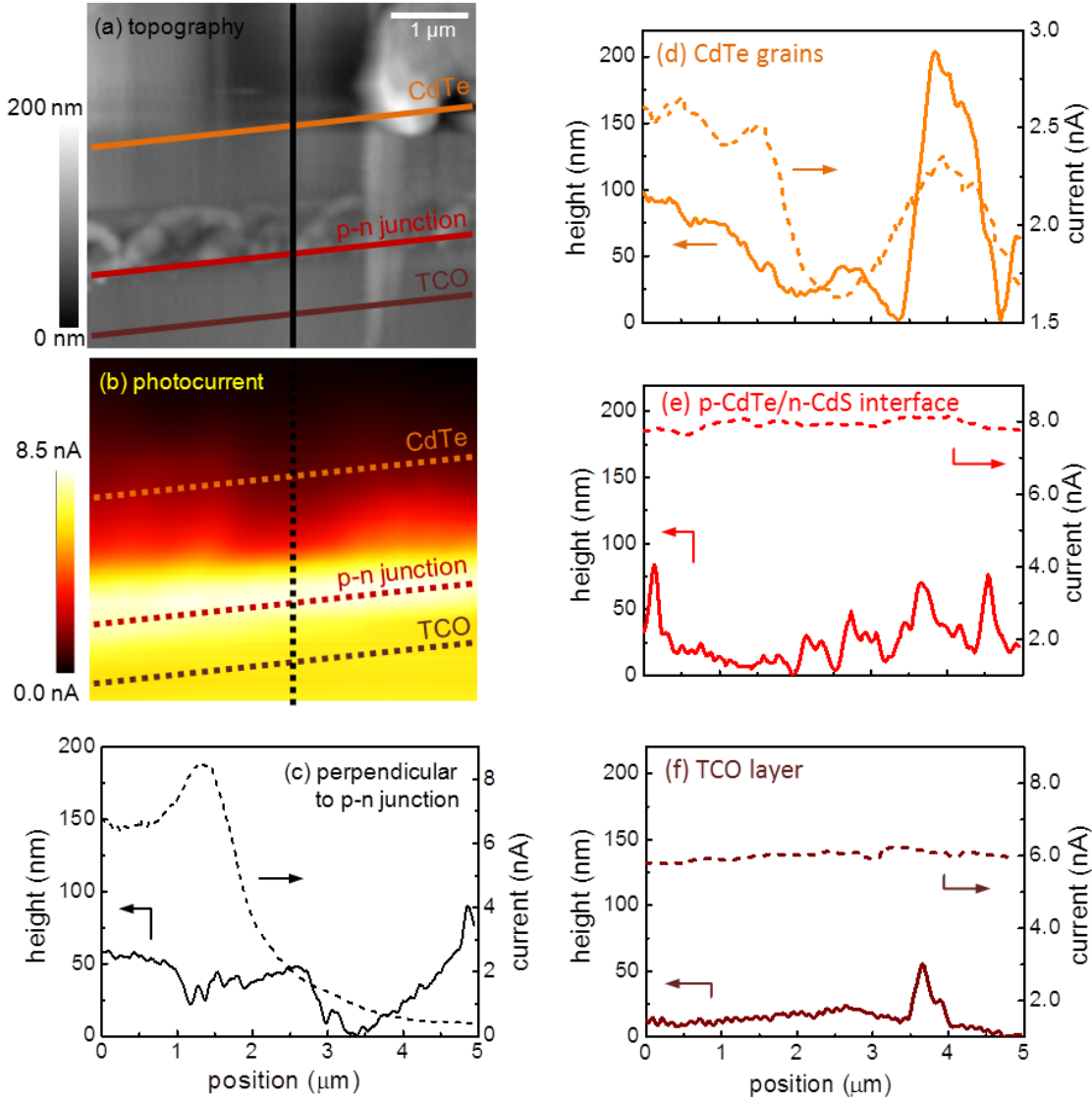


Fig. 6. Cross-section (a) topography and (b) LBIC measurements. (c) Line profiles perpendicular to the p-n junction; from the TCO layer ($x = 0$) to the CdTe layer ($x = 5 \mu\text{m}$). (d) – (f) Line profiles parallel to the p-n junction. In all cases the solid and dashed lines refer to topography and current, respectively. Illumination source: 532 nm laser at 14 mW, probe = 500 nm in diameter.

For $\lambda = 532 \text{ nm}$, the absorption coefficient α is 10^7 m^{-1} , and the incoming light is absorbed at 100 nm from the material top surface, far away from the p - n junction. As a result, the generated photocurrent is very small. The contrast between the GIs and the GBs is rather poor (Fig. 5(b)), and it can be additionally affected by a high recombination at the exposed top surface of CdTe.

Close to the band gap ($E_g = 1.44 \text{ eV}$, $\lambda = 860 \text{ nm}$) the current generated at the GBs is consistently larger (3x) than the current generated by the GIs (Fig. 5(c)), confirming that the GBs are efficient local collectors for the minority carriers (electrons). At this wavelength, $\alpha = 6 \times 10^3 \text{ m}^{-1}$ for CdTe, and the penetration depth is 200 μm. The exciting photons can fully penetrate the CdTe layer, assessing the p - n junction. The resulting photocurrent signal is, therefore, generated by the volume of the CdTe layer corresponding to the area that is being scanned. At energies below the band gap the absorption quickly vanishes resulting in a significant reduction of the

generated photocurrent (Fig. 5(d)). At $\lambda = 890 \text{ nm}$, additional current contrast that was not evident at $\lambda = 860 \text{ nm}$ could be resolved (see highlighted region in Fig. 5, for example). These sharp features could be caused by stoichiometry variations at the GBs or by additional absorption due to impurities.

The precise identification of the chemical composition of the alloy forming the GBs requires the use of high resolution destructive techniques, such as atom probe tomography [23]. Another possible origin of the spatially sharp contrast can still be related to the wavelength-dependent light out-coupling in the near-field affected by the topographic features. Currently, 3D finite-difference time-domain simulations mimicking the measurement conditions are in progress.

IV. ASSESSING THE P-N JUNCTION

We performed cross sectional LBIC microscopy measurements (as represented in Fig. 2) under constant

illumination ($\lambda = 532$ nm, 14 mW) to access the device p - n junction. A cross-section of the sample was milled using a Ga focused ion beam source to provide a fairly smooth surface, as shown by the topography measurement in Fig. 6(a). The white region in the image corresponds to a piece of CdTe that was not fully 'polished away' during the final milling steps. Here, this topographic feature is used to corroborate the results of our plan-view measurements.

The CdTe grains' photocurrent (Fig. 6(b)) varied as a function of distance to the p - n junction. Fig. 6(c) shows the line scans for the topography and photocurrent perpendicular to the p - n junction. As expected, the maximum current is generated at the p - n junction region. The line profiles parallel to the p - n junction for height and photocurrent taken at the same region of the CdTe grains are displayed in Fig. 6(d)-(f) and showed no correlation between topography and photocurrent, consistent with our plan-view and wedge measurements at $\lambda = 532$ nm. The CdTe/CdS interface, i.e., the p - n junction (line profiles in Fig. 6(d)) was found to generate most of the photocurrent (≈ 8.0 nA), with very small variations, as displayed by the very bright region of Fig. 6(b). However, photocurrent was also generated when the probe was positioned on top of the TCO layer (Fig. 6(e)). This can be related to multiple scattering effects at the contact and substrate interfaces leading to light injection into the device, and the effect is likely non-local. A detailed analysis of the light-semiconductor interaction at the near field involving 3D finite-difference time-domain simulations will be conducted in a future work.

V. CONCLUSION

In summary, we mapped and quantified the photocurrent generated at CdTe/CdS solar cells GBs under different illumination conditions. We found that the GBs are very efficient current collectors and that the enhanced collection at GBs can be reliably resolved by LBIC microscopy. Modified CdTe surfaces were investigated to exclude possible artifacts caused by surface topography on coupling of the near-field localized light source. Plan-view measurements were performed as a function of different illumination wavelengths to investigate material's bandgap variations or absorption by the states in the gap within the GBs. Cross-section LBIC measurements were used to access the p - n junction of the device, showing a clear dependence of the GBs transport properties with distance from the junction.

Furthermore, LBIC microscopy can be used to probe the photoelectronic properties of devices under forward bias (at V_{oc} operation) and can be expanded to other polycrystalline materials, such as CIGS and mc-Si. The local measurement of the photoelectronic properties of GBs and GIs in thin film CdTe solar cells by LBIC microscopy represents an important step towards the understanding of how the GBs impacts the device performance, and can be used to map the diffusion length and recombination rates of the GBs and GIs[24], which

can be further used to engineer a device with enhanced V_{oc} .

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