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Challenges and Opportunities with Highly Brilliant X-ray Sources for multi-Modal in-Situ and Operando Characterization of Solar Cells

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The generation of electricity is one of the great challenges of our time, and its success shall be measured by the sustainability from ecological, economical, and social points of view. The advent of photovoltaics has led to a dramatic decrease of the levelized cost of energy (LCOE) such that it has not only reached grid parity in many countries but is often the most competitive energy source.

Today's photovoltaics market is dominated by single-junction solar cells with a crystalline silicon (c-Si) absorber. Higher efficiencies can be achieved in tandem junction devices, with a thin top cell converting the high-energy part of the solar spectrum in a wide-bandgap absorber, and a bottom c-Si cell converting the low-energy part of the spectrum [1]. Alternatively, lower manufacturing costs can be achieved for polycrystalline solar cells based on direct bandgap absorbers such as $Cu(In,Ga)Se_2$. In both cases, the research focus lies on the development of cost-effective, high-efficiency thin-film (TF) solar cells with polycrystalline absorbers, including CdTe, $Cu(In,Ga)Se_2$, III/V semiconductors and metal-halide perovskites such as the archetypical methyl-ammonium lead iodide (MAPI).

Of critical importance for the large-scale development of TF solar cells is the characterization of their composition, structure, and local performance. TF solar cells are based on multi-element absorbers and consist of a multitude of nanometer-thick layers. Vertical gradients and horizontal inhomogeneities at all length scales from nanometer to centimeter, grain boundaries with complex passivation and diffusion chemistry, and trace elements that govern the device performance challenge characterization methods.

We have demonstrated that hard X-ray nanoprobe beamlines are ideally suited to address characterization challenges of TF solar cells [2-7], and the high penetration depth of hard X-rays combined with a long working distance enables the study of the relevant layers in encapsulated modules at nanometer scale resolution and unparalleled sensitivity. Particularly powerful is the multimodal mapping, as it allows the direct point-by-point correlation of performance, bandgap, composition, and structure by X-ray beam induced current (XBIC) and voltage (XBIV), X-ray excited optical luminescence (XEOL), X-ray fluorescence (XRF), X-ray diffraction (XRD), and ptychography based methods. Here, we demonstrate for the first time that ptychography unveils grain boundaries with high sensitivity (see Fig. 1 (c-d)).

We have successfully introduced lock-in amplification of the electrical solar cell response to X-rays [2,6] at the nanoprobe endstations 2-ID-D & 26-ID-C (APS), 10-ID-C (CLS), ID16B (ESRF) and P06

(DESY). Lock-in amplification boosts the field of XBIC applications: first, it improves the signal/noise ratio by orders of magnitude. Second, it decreases the sensitivity to measurement artifacts (see Fig. 1 (a-b)). Third, it truly expands the dimensionality by giving access to the full current—voltage ($I(V)$) relationship through the application of bias voltage during XBIC measurements. Forth, it enables XBIC measurements under light bias. The combination of these *operando* measurement modes with *in-situ* techniques—varying the gas environment or temperature—ultimately gives access to nanoscale variations of the temperature coefficients [6] and enables the study of solar cells under outdoor conditions (see Fig.1 (e)).

Today, the photon flux limits the measurement speed in most cases, particularly for photon-hungry techniques such as XEOL. However, other limitations exist already for more sensitive measurements: beam damage is particularly challenging for perovskite solar cells [4], soft- or hardware limitations rarely allow dwell times < 10 ms, and XRF detectors are regularly saturated by the signal from solar cells with strongly absorbing metals as main constituents. Overcoming these limitations is necessary to make optimum use of high-brilliance hard X-ray nanoprobe endstations that will be available at 4th generation synchrotrons and X-ray Free Electron Lasers. With scan rates > 1 kHz, scanning tomography can become a commodity, and 4D or even 5D measurements will be within reach [8].

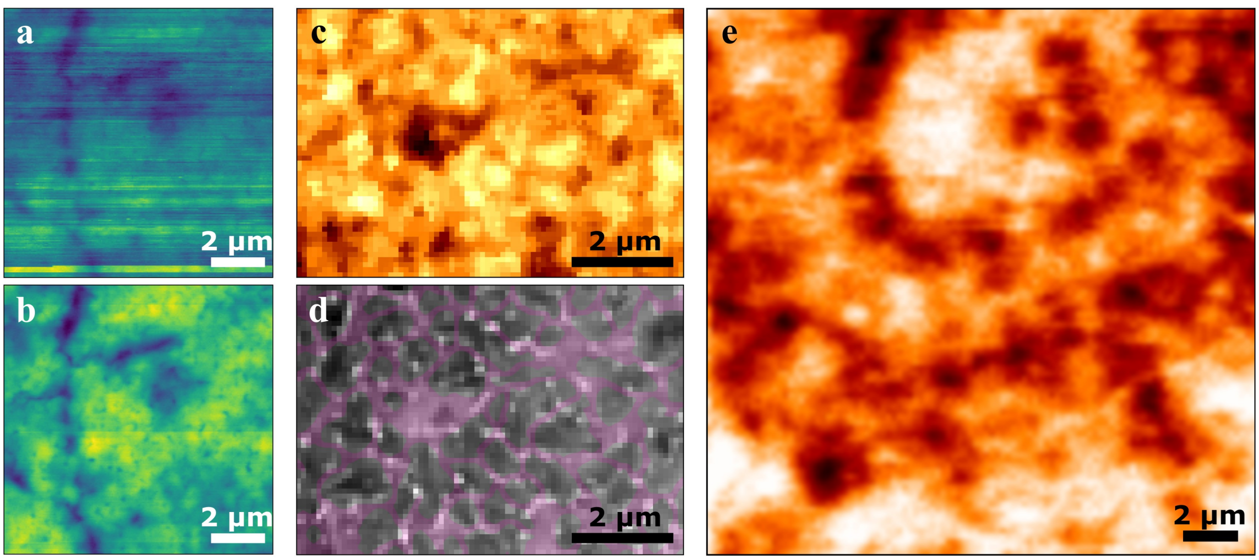

Figure. 1. X-ray microscopy of a Cu(In,Ga)Se_2 solar cell. Left: XBIC measured as Direct Current (a) and with lock-in amplification (b). Center: Copper distribution from XRF (c) and phase image from ptychography (d), overlaid with the grain topology extracted through watershed analysis. Right: XBIC measured at 60 ˚C, with bias light and bias voltage applied (e).

[1] Z Yu, M Leilaeioun and Z Holman, Nature Energy **1** (2016), 16137.
[2] M Stuckelberger *et al*, J. Mater. Res. **32** (2017), p. 1825.
[3] BM West *et al*, Nano Energy **32** (2017), p. 488.
[4] M Stuckelberger *et al*, IEEE J. Photovolt. **7** (2017), p. 590.
[5] BM West *et al*, J. Synchrotron Rad. **24** (2017), p. 288.
[6] M Stuckelberger *et al*, IEEE J. Photovolt. **8** (2018), p. 278.
[7] Y Luo *et al*, Adv. Func. Mat. (2018), 1706995.
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