Correlative transmission Kikuchi diffraction and atom probe tomography analysis of grain boundaries in $CuInS_2$ thin film solar cells

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The compound semiconductor Cu(In,Ga)(S,Se)2 (CIGSSe) is used as absorber in thin-film solar cells. Currently, a record efficiency of 22.9% is achieved for CIGSSe based devices. However, the efficiency of such single-junction based solar cells are reaching their practical limits. Therefore, multi-junction cells are a promising way to further increase the efficiency beyond 30%. One suitable material as top cell in such a tandem structure is Cu(In,Ga)S2, which has a tunable band gap of 1.55-2.4 eV. So far, a record efficiency of 15.5% has been achieved.

For further enhancement of the efficiency of this polycrystalline material it is important to understand how the microstructure affects the electrical properties of the absorber films and, hence, the device performance. Therefore, one needs to study the relationship between structural and chemical properties of grain boundaries (GB). Recently, we have shown by correlated transmission Kikuchi diffraction (TKD) and atom probe tomography (APT) that, for CIGSe based films, ∑3 twin boundaries (TB) show no compositional changes, whereas random high-angle GBs (RHAGB) show chemical fluctuations as well as impurity segregation, which passivate the GBs [1]. However, no such study exist for Cu(In,Ga)S2 films.

Here, we investigate the correlation between crystallographic and chemical information of GBs in two CuInS2 samples by using TKD and APT. While both samples were grown at high temperature (600 °C), one of them is Cu-rich ([Cu]/[In]>1) and the other one Cu-poor ([Cu]/[In]<1). We address practical issues related to specimen preparation for TKD. For the Cu-rich grown film, we detect Cu enrichment and In & S depletion at RHAGBs, which is accompanied by co-segregation of Na & C as shown in Fig. 1. In contrast, for the ∑3 TBs (see Fig. 1) we detect no change neither in composition nor in atomic density. While the TBs will most likely not affect the electrical properties, the Cu enrichment at the RHAGB will not lead to an electrical passivation, whereas the Na atoms may diminish detrimental point defects at the RHAGB. We will also present our latest results on the Cu-poor sample and will discuss possible effects of the observed phenomena on the cell performance.

References:

[1] T. Schwarz, et al., Correlative transmission Kikuchi diffraction and atom probe tomography study of Cu(In,Ga)Se2 grain boundaries, Prog Photovolt Res Appl. 26 (2017), 196-204.

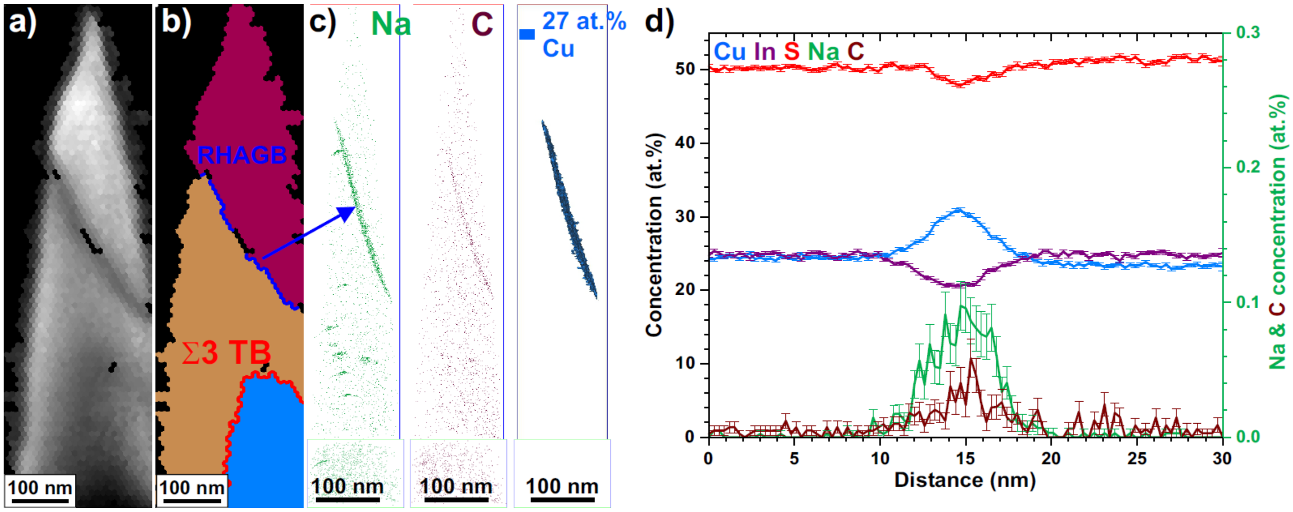

Fig. 1 (a) Image quality maps from a TKD measurement of an APT tip. (b), Corresponding unique color map to (a) showing a RHAGB (blue) and ∑3 TB (red). (c) Na & C co-segregation as well as Cu enrichment (blue iso-concentration surface) at the RHAGB shown in (b). (d) Concentration profile across the RHAGB revealing an atomic redistribution.