



Contribution ID: 112

Type: Poster

Probing molecular and crystalline orientation in solution-processed perovskite solar cells

Thursday, 26 November 2015 13:30 (45 minutes)

We investigate the microstructure of solution-processed organometallic lead halide perovskite thin films using a combination of synchrotron based techniques. Using a combination of GIWAXS and NEXAFS spectroscopy we separately probe the orientational alignment of $\text{CH}_3\text{NH}_3\text{PbI}_3$ crystallites and CH_3NH_3^+ cations. The GIWAXS results reveal that the orientation of $\text{CH}_3\text{NH}_3\text{PbI}_3$ crystallites is sensitive to film thickness, solvent evaporation rate, and the underlying TiO_2 morphology. In perovskite films prepared by a gas-assisted method, oriented perovskite crystallites are detected in thin films ($\sim 60\text{nm}$) deposited on a dense TiO_2 blocking layer. As the thickness of the perovskite layer is increased to $\sim 250\text{ nm}$, however, this preferential orientation of perovskite crystals disappears. In contrast, for both thin and thick perovskite films deposited on an underlying mesoporous TiO_2 layer randomly orientated crystallites are observed. NEXAFS measurements on all samples prepared by the gas-assisted method found that CH_3NH_3^+ cations exhibit a random molecular orientation with respect to the substrate, independent of the TiO_2 architecture and the perovskite film thickness. The lack of any NEXAFS dichroism for the thin $\text{CH}_3\text{NH}_3\text{PbI}_3$ layer deposited on planar TiO_2 in particular indicates the absence of any preferential orientation of CH_3NH_3^+ cations within the $\text{CH}_3\text{NH}_3\text{PbI}_3$ unit cell for as-prepared layers (that is, without any poling). Solar cells based on the thicker ($\sim 250\text{ nm}$) perovskite films were also prepared to enable correlation with microstructural results, with solar cells based on planar TiO_2 achieving an efficiency of 14.3% compared to 12% for cells fabricated with mesoporous TiO_2 layers.

Keywords

Perovskite solar cells, GIWAXS, NEXAFS

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Session Classification: Poster Session 1

Track Classification: Energy Materials