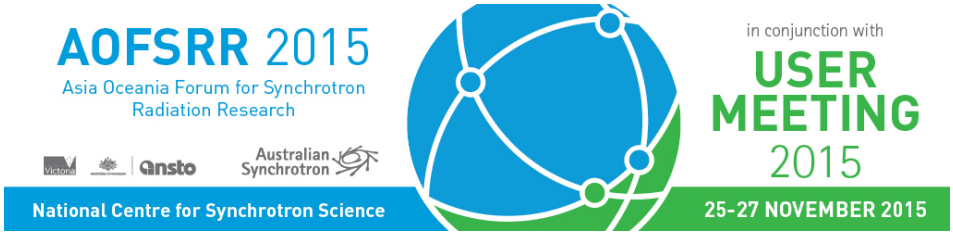

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Probing molecular and crystalline orientation in solution-processed perovskite solar cells

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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 CH3NH3PbI3 crystallites and CH3NH3+ cations. The GIWAXS results reveal that the orientation of CH3NH3PbI3 crystallites is sensitive to film thickness, solvent evaporation rate, and the underlying TiO2 morphology. In perovskite films prepared by a gas-assisted method, oriented perovskite crystallites are detected in thin films (~60nm) deposited on a dense TiO2 blocking layer. As the thickness of the perovskite layer is increased to ~250 nm, however, this preferential orientation of perovskite crystals disappears. In contrast, for both thin and thick perovskite films deposited on an underlying mesoporous TiO2 layer randomly orientated crystallites are observed. NEXAFS measurements on all samples prepared by the gas-assisted method found that CH3NH3+ cations exhibit a random molecular orientation with respect to the substrate, independent of the TiO2 architecture and the perovskite film thickness. The lack of any NEXAFS dichroism for the thin CH3NH3PbI3 layer deposited on planar TiO2 in particular indicates the absence of any preferential orientation of CH3NH3+ cations within the CH3NH3PbI3 unit cell for as-prepared layers (that is, without any poling). Solar cells based on the thicker (~ 250 nm) perovskite films were also prepared to enable correlation with microstructural results, with solar cells based on planar TiO2 achieving an efficiency of 14.3% compared to 12% for cells fabricated with mesoporous TiO2 layers.

Keywords

Perovskite solar cells, GIWAXS, NEXAFS

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