

Supplementary Information

Processing solvent dependent electronic and structural properties of cesium lead iodide thin films

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Experimental Methods

CsPbI₃ films with a thickness of 150-200nm were formed through spin-coating from 0.5M solutions using the following solvents: dimethylformamide (DMF), dimethylsulfoxide (DMSO), DMF with 3.3% hydroiodic acid (HI) and DMF:DMSO mixture in a 2:1 ratio. All films were fabricated at 2000rpm in an inert atmosphere onto fluorine doped tin oxide (FTO) glass. The thin films were then annealed at the following times and temperatures: DMF, DMSO and DMF:DMSO were annealed at 310°C for 10 minutes and DMF w/HI was annealed at 100°C for 5 minutes as following previously reported experimental conditions required for complete crystallization of perovskite film into the “black” polymorph.^{6,7}

Scanning electron micrographs were obtained using an FEI Quantum 600 FEG scanning electron microscope. X-ray diffraction (XRD) measurements were carried out on using a PANalytical X'Pert Pro MPD diffractometer with monochromatic Cu K α_1 radiation. X-ray Photoelectron Spectroscopy (XPS) data were collected using a Thermo Scientific K-Alpha⁺ system. The samples were measured using a monochromated Al K α X-ray source. The measurements were conducted at room temperature and at a takeoff angle of 90° with respect to the surface parallel. The core level spectra were recorded using pass energy of 20 eV (resolution approximately 0.4 eV) from an analysis area of 300 μm \times 300 μm . The spectrometer work function and binding energy scale were calibrated using the Fermi edge and 3d_{5/2} peak recorded from a polycrystalline Ag sample prior to the commencement of the experiments.

Secondary ion mass spectrometry (SIMS) analyses were performed on an IONTOF ToF-SIMS V instrument. The primary ion beam was a 25keV Bi⁺ in BAM (bunched to 4 pulses to avoid detector saturation). The sputter beam was Cs⁺ at 1keV and a beam current of 75nA.

The analytical area was $100\mu\text{m}^2$ with a sputter crater of $300\mu\text{m}^2$. Low energy ion scattering measurements were performed on an IONTOF Qtac 100 using an 8keV Ar^+ primary ion beam with a current of $\sim 1\text{nA}$. An area of 1mm^2 was analysed. The scattered primary ions were collected at a fixed angle of 145° over the full azimuth. Peaks were observed for I, Cs and Pb at energies of 2435, 2579, and 3936eV respectively. Surface stoichiometries for all CsPbI_3 films were calculated from LEIS measurements using the relative ratios of the peak areas corrected for scattering cross-sections.

Secondary ion mass spectrometry (SIMS) mapping with associated scanning electron microscope (SEM) images

