

## Supplementary Materials for **Enhanced mobility CsPbI<sub>3</sub> quantum dot arrays for record-efficiency, high-voltage photovoltaic cells**

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Published 27 October 2017, *Sci. Adv.* **3**, eao4204 (2017)  
DOI: 10.1126/sciadv.aao4204

### **This PDF file includes:**

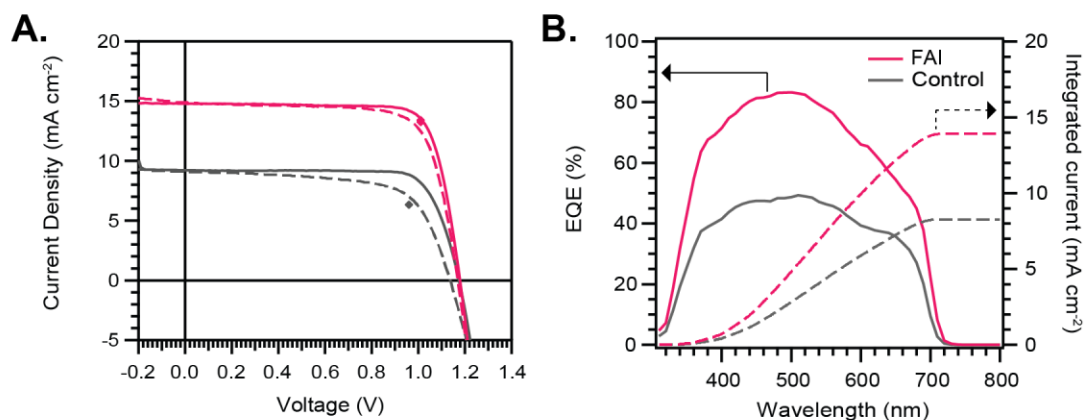
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## Supplementary Materials and Methods

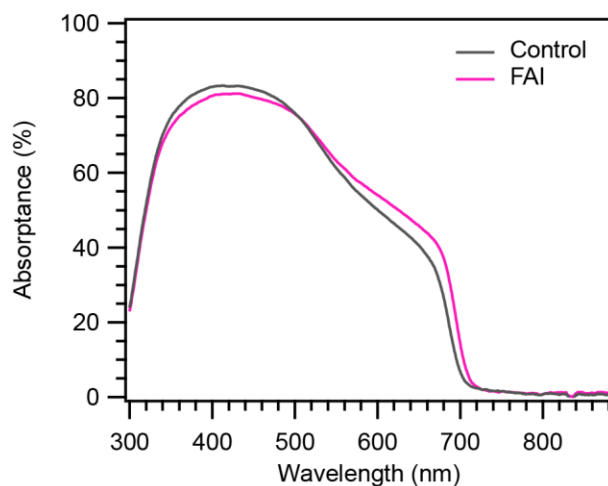
**X-ray photoemission spectroscopy.** XPS data was taken on a Kratos NOVA spectrometer calibrated to the Fermi edge and core level positions of sputter-cleaned metal (Au, Ag, Cu, Mo) surfaces. Spectra were acquired using monochromated Al K $\alpha$  radiation (1486.7 eV) at a resolution of 600 meV (pass energy 20 eV). The data was averaged from multiple spots on the sample while the X-ray intensity was held low (15 W anode power) to avoid sample degradation (34) and fit using Pseudo-Voigt profiles.

**Scanning electron microscopy.** Samples were mounted on aluminum stubs with double-sided carbon tape and sputter-coated with 3 nm of Iridium prior to imaging. Images were obtained with a FEI Quanta 400 FEG instrument (FEI, Hillsboro, OR). Imaging was performed with a beam accelerating voltages of 30 keV.

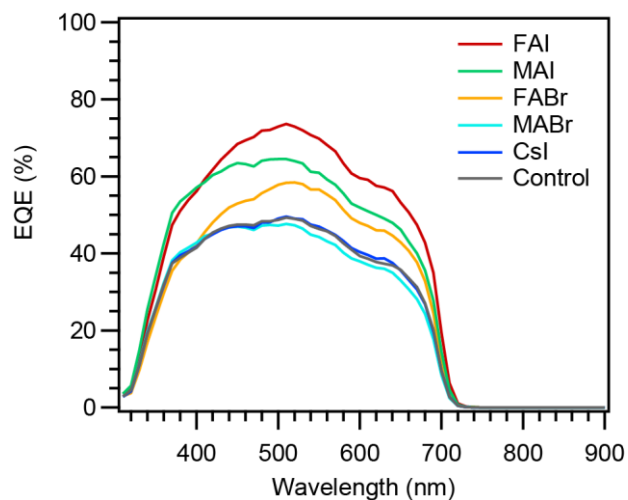
**Time-resolved photoluminescence.** Time resolved photoluminescence was measured using a Hamamatsu streak camera system (C10910-05), while the excitation source was a Fianium Supercontinuum high power broadband fiber laser (SC400-2-PP). The chosen excitation wavelength was 530 nm at  $\sim 25$   $\mu$ W power on a spot size of 0.02 mm<sup>2</sup>.



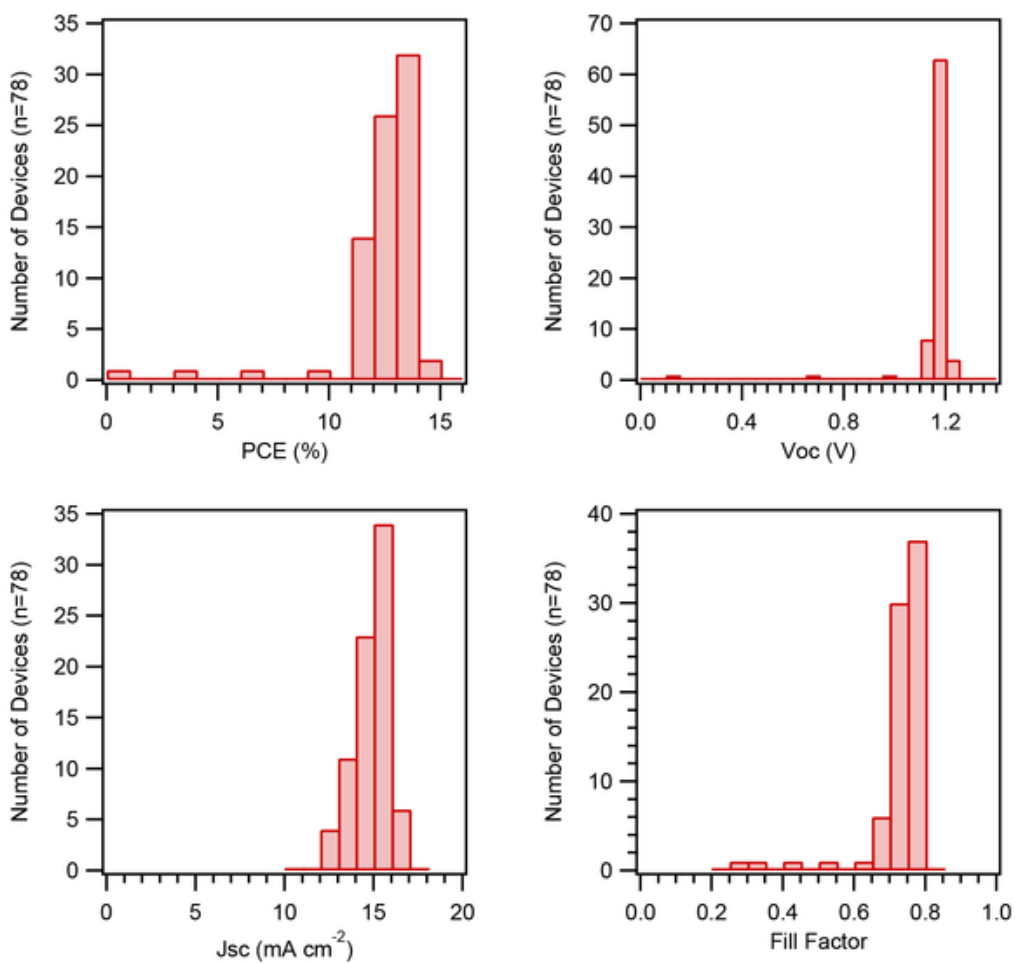
**fig. S1. Performance of FAI-coated and control devices.** (A) *J-V* scans in the forward (dotted) and reverse (solid) directions for FAI-coated (pink) and control (grey) devices. The SPO for each device is indicated by the diamond marker. (B) EQE (solid) and integrated current (dotted) for the FAI-coated (pink) and control (grey) devices.



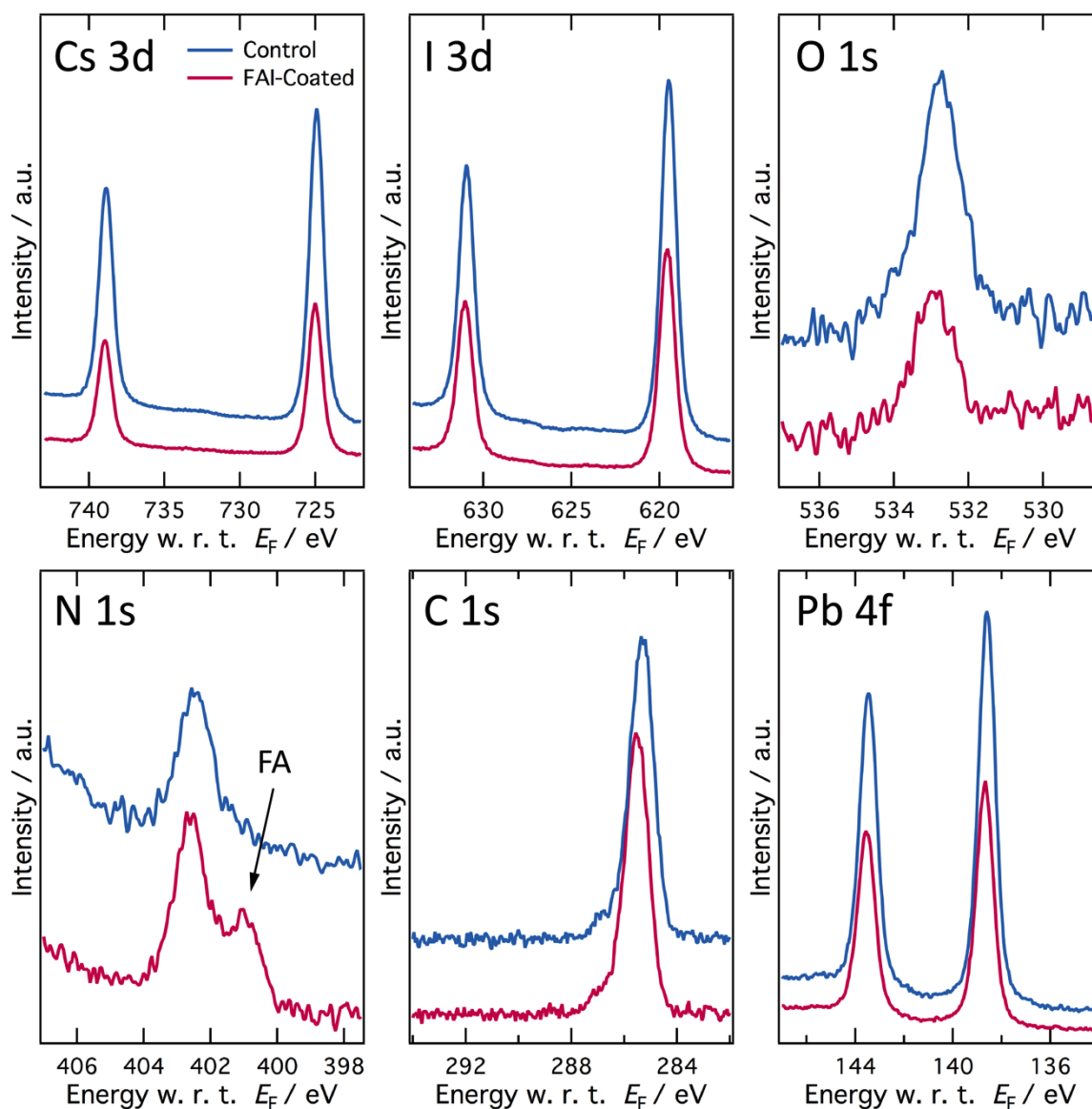
**fig. S2. Light absorption following FAI posttreatment.** Absorbance spectra of CsPbI<sub>3</sub> QD films on glass with (pink) and without (grey) the FAI post-treatment.



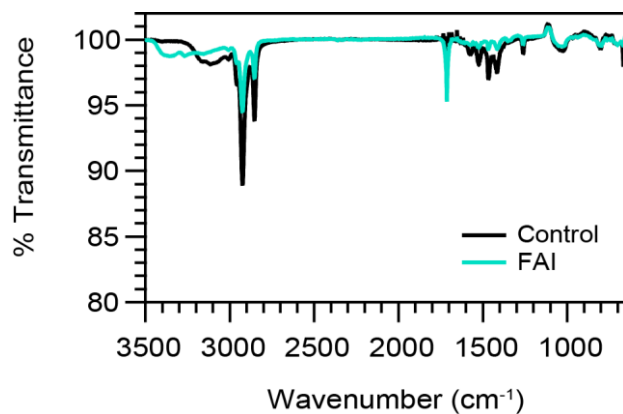
**fig. S3. Comparison of EQE with AX posttreatment.** EQE of devices treated with different AX salts observe similar EQE onset at  $\sim 700$  nm.



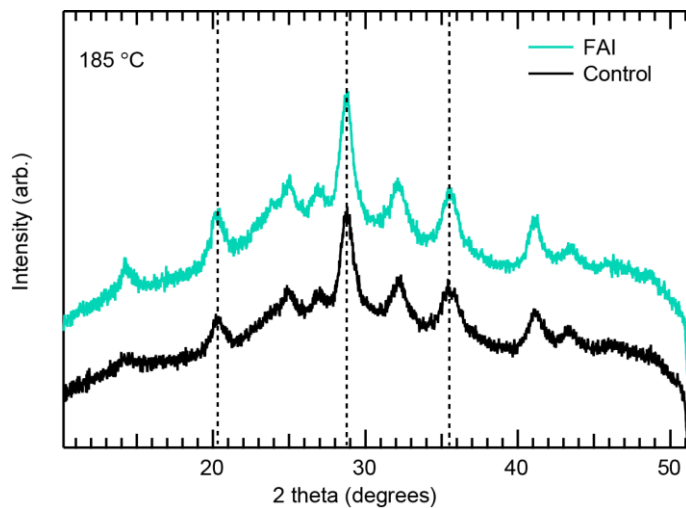
**fig. S4. Reproducibility of FAI-coated CsPbI<sub>3</sub> QD device performance.** Histograms of the PCE,  $V_{OC}$ ,  $J_{SC}$  and FF of FAI-coated CsPbI<sub>3</sub> QD devices ( $n=78$ ).



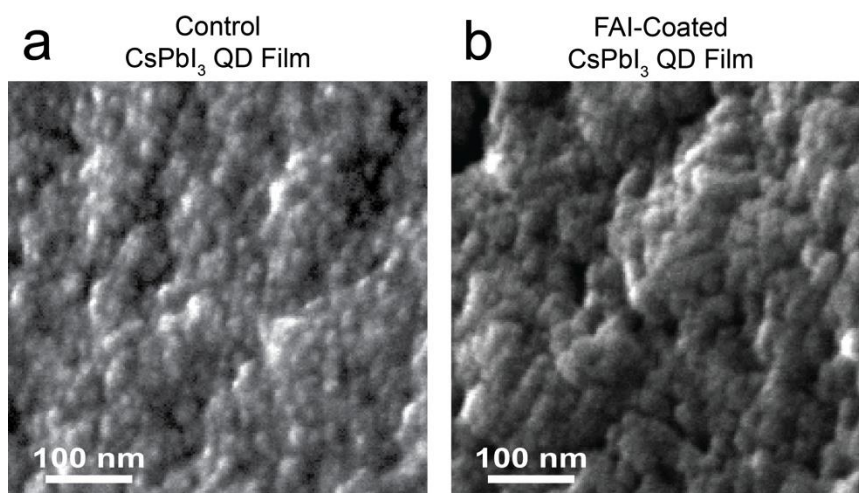
**fig. S5. XPS spectroscopy.** XPS core level spectra of CsPbI<sub>3</sub> QD films on TiO<sub>2</sub>/FTO/glass. The presence of FA<sup>+</sup> species is observed by an additional peak centered at 401 eV binding energy in the N 1s core level region. Furthermore, the surface composition changes upon FAI treatment. The Cs<sup>+</sup>:Pb<sup>2+</sup> ratio drops from 0.91 to 0.74 indicating a decrease in Cs<sup>+</sup> content at the surface. At the same time, the I:Pb<sup>2+</sup> ratio rises from 2.73 to 2.83 corroborating the results of the ToF-SIMS measurements. A decrease in the oxygen content upon FAI treatment indicates the further removal of residual oleate groups from the QD surface.



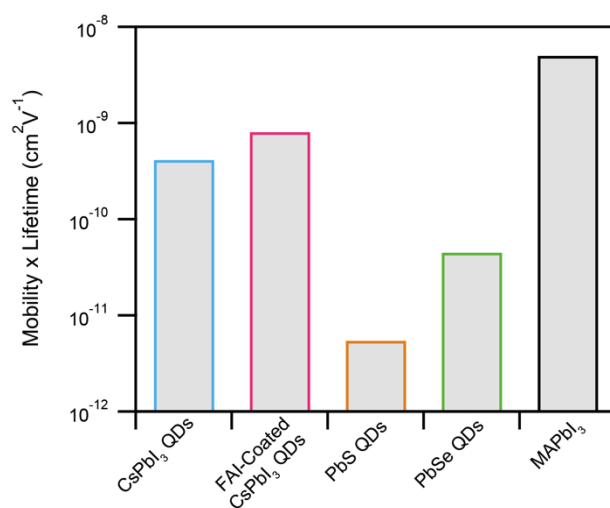
**fig. S6. FTIR spectra of CsPbI<sub>3</sub> QDs.** Fourier-transformed infrared (FTIR) spectra of CsPbI<sub>3</sub> QD films with (green) and without (black) FAI post-treatment. The emergence of the peak at 1712 cm<sup>-1</sup> is indicative of the presence of FA.



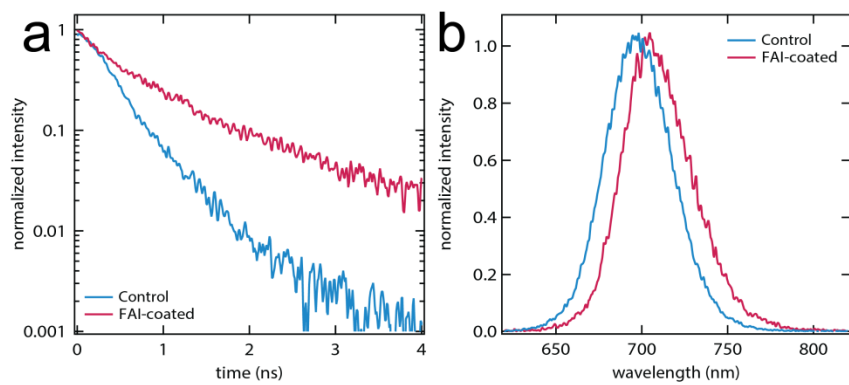
**fig. S7. Crystal structure of CsPbI<sub>3</sub> QDs.** X-ray diffraction pattern for CsPbI<sub>3</sub> QD films with (green) and without (black) FAI post-treatment.



**fig. S8. CsPbI<sub>3</sub> QD film morphology.** SEM micrographs of the surface of the control and FAI-coated QD films show closely-packed assemblies of discrete nanoparticles in both cases.



**fig. S9. Comparison of terahertz  $\mu_s \times \tau$  product.** Comparison of the  $\mu_s \times \tau$  product calculated from THz spectroscopy for the control CsPbI<sub>3</sub> QD and FAI-coated CsPbI<sub>3</sub> QD films compared to a PbS QD film, a PbSe QD film, and a MAPbI<sub>3</sub> thin film.



**fig. S10. PL lifetime of CsPbI<sub>3</sub>.** Time resolved photoluminescence decay plots for CsPbI<sub>3</sub> QD films with (pink) and without (blue) FAI post-treatment.