Supporting Information

High performance photodetectors based on solution-processed epitaxial grown hybrid halide perovskites

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Materials and Methods

Perovskite precursor preparation

Methylamine iodide (MAI) was prepared by reacting methylamine, 33wt% in ethanol (Sigama-Aldrich), with hydroiodic acid (HI) 57wt% in water (Sigma-Aldrich) at room temperature. After drying at 100 ^{o}C, white powder was formed, followed by overnight drying in a vacuum oven. To form the precursor solution, MAI and PbI2 (Sigama-Aldrich) were dissolved in anhydrous N,N-Dimethylformamide (DMF) at a 1:1 molar ratio.

Perovskite deposition

To form the perovskite layer, the prepared precursor was spin-coated on KCl(001) substrate at 2000rpm in a Ar-filled golvebox. After the spin-coating, the films were annealed at 100 ^{o}C for 20 mins.

Photodetectors fabrication

Au metal contacts were deposited directly onto perovskite thin film by e-beam evaporation through a stainless steel shadow mask. The temperature was monitored and kept under 40 ^{o}C during deposition.

Characterizations

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Scanning electron microscopy (SEM)

A field emission scanning electron microscopy system (Zeiss) was used to acquire SEM figures. The beam voltage is 2 kV, to minimize electron induced damage.

Photoluminescence measurements and fits

Steady-state and time-resolved PL measurements were acquired using a time correlated single photon counting setup. Film samples were photoexcited using a laser head pulsed, providing < 200ps pulses with the fluence of ~ 30 nJ/cm^2.

Raman Spectroscopy

The Raman measurements were performed with excitation lines of 532 nm. Laser power density was 1 mW.

X-ray diffraction (XRD)

XRD were obtained using a double-axis high resolution X-ray diffractometer, using CuKα(λ = 1.5405 Å) radiation source. The x-ray generator was set to 40 kV and 45 mA.

Electron back scattered diffraction (EBSD)

The EBSD patterns were obtained in a SEM system (Zeiss Neon 40 FE-SEM) equipped with an EDAX (Mahwah, NJ) APEX 2 integrated EDS and EBSD system, by focusing the electron beam onto the sample surface at a tilt angle of 70^o with respect to the horizontal. The scanned data were post analyzed using the EDAX TSL OIM^{TM} 5.0 software.

SEM characterizations

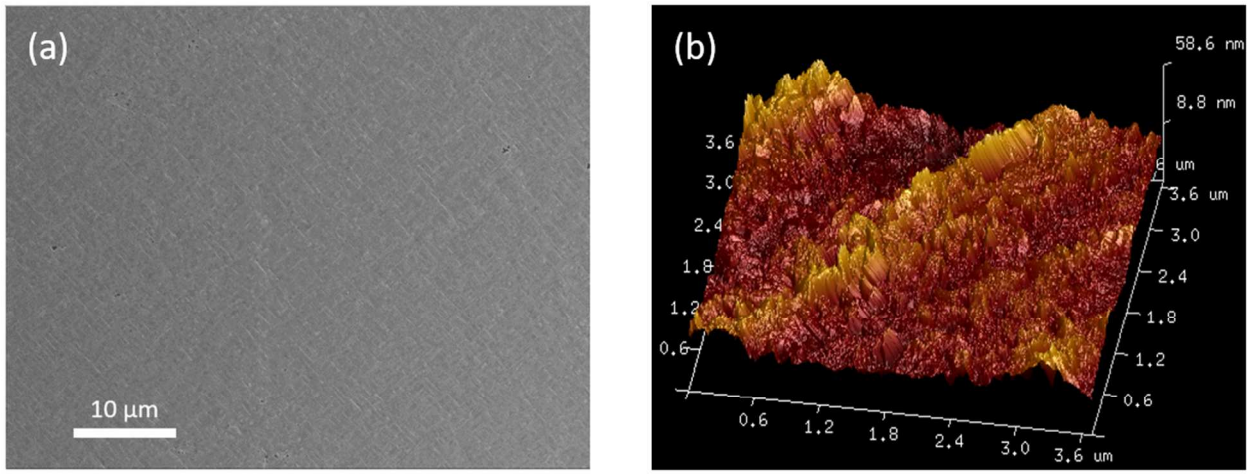

Fig. S1. (a) SEM and (b) AFM characterization of epitaxial MAPbI3 films

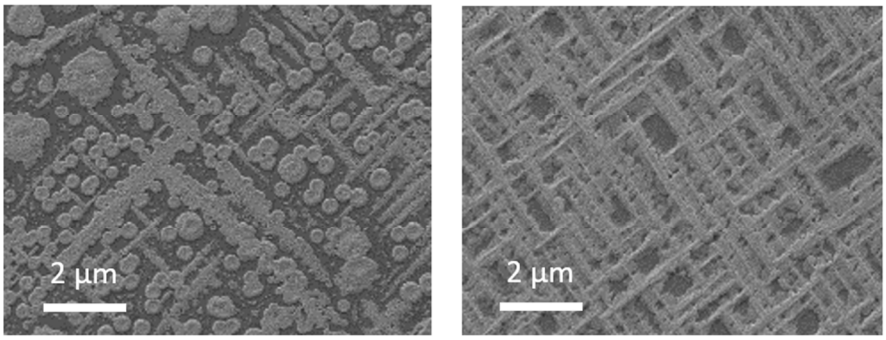

Fig. S2. Un-optimized processing condition, resulting in large pin-holes and big clusters.

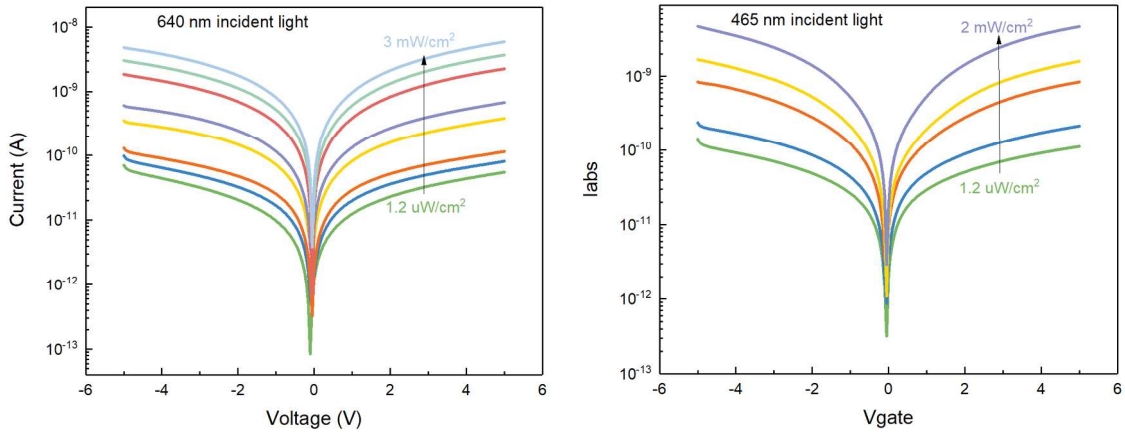

Fig. S3. I-V curves of the photodetector measured under dark and various light illumination (650 nm and 465 nm), respectively.