# **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 °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 °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 °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  $\sim 30 \text{ nJ/cm}^2$ .

#### 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  $CuK\alpha(\lambda = 1.5405 \text{ Å})$  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° with respect to the horizontal. The scanned data were post analyzed using the EDAX TSL OIM<sup>TM</sup> 5.0 software.

## **SEM** characterizations

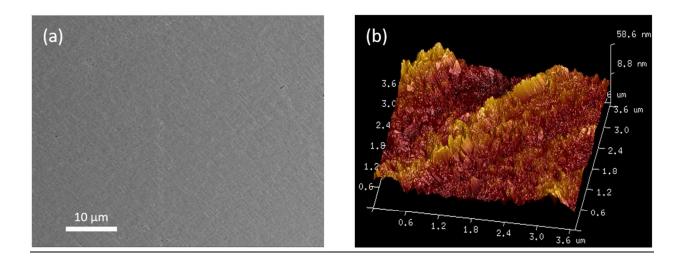


Fig. S1. (a) SEM and (b) AFM characterization of epitaxial MAPbI3 films

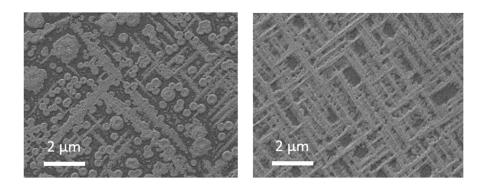


Fig. S2. Un-optimized processing condition, resulting in large pin-holes and big clusters.

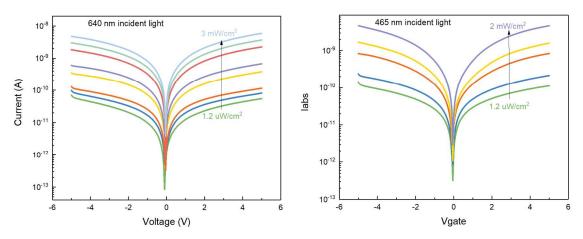


Fig. S3. I-V curves of the photodetector measured under dark and various light illumination (650 nm and 465 nm), respectively.