Supporting Information

Capillary-Written Single-Crystalline All-Inorganic Perovskite Microribbon Arrays for Highly-Sensitive and Thermal-Stable Photodetectors

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Experimental Section

Materials: CsBr and PbBr₂ with purity of 99.99% was purchased from Sigma-Aldrich. *n*-Octadecyltrimethoxysilane (OTS) was purchased from Sigma Aldrich. DMF, DMSO and other solvents were purchased from Sigma Aldrich. All materials were used without further purification.

Wafer modification: Highly doped silicon substrates (1 cm^2) with 300 nm SiO₂ were used for PDs substrates. Before crystallization, the wafers were modified by OTS monolayer following previous report.¹

Preparation of CsPbBr₃ MRAs: Single-crystalline CsPbBr₃ MRAs was preparaed by a direct-written method using a hollow rectangular capillary. The experiment set-up was shown in **Figure 1a**. A 1 mL precursor solution with an equimolar ratio of 1mM CsBr:PbBr₂ (1:1) in dimethyl sulfoxide (DMSO) is held in the capillary by capillary forces. Here, DMSO was selected as a solvent instead of N,N-dimethylformamide (DMF) due to its overall higher solubility limit for the precursors. The capillary is bent at 1 cm from the end to form an "L" shape, so that a gravity feed at constant pressure corresponding to the height of the vertical part is maintained. A substrate is mounted on a computer-controlled linear translation stage equipped with a thermoelectric module for temperature control. Film deposition is accomplished by allowing the microdroplet of solution on the end of the capillary to contact the surface and then laterally translating the substrate at a controlled rate of 0.02 mm s⁻¹. After the solvent evaporation on the hotplate for 15 min at 80 °C and annealed at 120 °C for 3 min, highly-aligned single-crystal microribbon arrays were formed.

Characterization and measurement: A NT-MDT Ntegra atomic force microscope in semi-contact mode was used to characterize surface morphology of the singlecrystalline MRAs. OM images were taken by Zeiss Axios Scope in polarized operation mode. SEM images were obtained by XL30 FEG scanning electron microscope. TEM observations were performed with an ED configuration on a JEOL JEM-2010 transmission electron microscope with an accelerating voltage of 200 kV. XRD was performed by Siemens D5000 X-Ray Powder diffratometer. The UV-vis spectra were carried out on a SHIMADZU UV-2600 spectrometer with an integrating sphere over the spectral range of 400-700 nm. A FLS-920 fluorescence spectroscopy had been used to collect photoluminescence (PL) spectra, and the excitation wavelength was 405 nm. The time-resolved measurement was carried out on an Edinburgh Instruments FLS980 with a nanosecond fluorescence spectrometer under 443 nm excitation wavelength.

Photodetector Fabrication: Photodetectors were fabricated by growing CsPbBr₃ MRAs on OTS-modified silicon wafers with a 300 nm thick thermal oxide layer. The electrical contacts to CsPbBr₃ MRAs were defined by copper grid as shadow mask with the typical gap of 20 µm, and subsequently 50 nm Au was evaporated. The light absorption area (active area) was calculated according to the overlapping area between the mask area and the illumination area.

Photoresponse Measurement: Photoresponse measurements were performed on the Lake Shore model PS-100 tabletop cryogenic probe tation by two-terminal mode with varied applied voltage. The parameters were analyzed using a Keithley 4200 semiconductor characterization system. Power-adjustable semiconductor laser units

with different wavelength were used to illuminate the devices to initiate the photocurrent.



Figure S1 (a) Nanoscale AFM image and (b) associated height analysis of $CsPbBr_3$ microribbon.



Figure S2 Wavelength-dependent responsivity of as-fabricated photodetectors.

Reference

1 Y. Ito, A. A. Virkar, S. Mannsfeld, H. O. Joon, M. Toney, J. Locklin and Z. Bao, *J. Am. Chem. Soc.*, 2009, **131**, 9396–9404.