1 Supplementary Information

- 2 Solution-processed visible-blind UV-A photodetectors based on
- 3 CH₃NH₃PbCl₃ perovskite thin films

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7 One-step spin coating

Equal mole CH_3NH_3Cl and $PbCl_2$ were dissolved in DMSO-DMF (1:1 by volume) to get a 1 9 M $CH_3NH_3PbCl_3$ precursor solution. The precursor was stirred for 12 h at 70 °C and filtered by a 10 0.2 µm PTFE filter. 100 µL of precursor was added onto a 15 mm × 15 mm substrate and spin 11 coated at 4000 rpm for 60 s. For thermal annealing, substrates were heated on a hotplate at 70 °C 12 for 60 min. For DMSO-vapor-assisted thermal annealing, substrates were first heated on a hotplate 13 at 70 °C for 60 min. Then the substrates were covered with a glass petri dish on the hotplate, 10 14 µL of DMSO was added from the edge of the petri dish and heated at 70 °C for 60 min.

15 Nano-pinning

16 Precursor solution was prepared as described above. 100 μ L of precursor solution was added onto a 15 mm \times 15 mm substrate. Nano-pinning method was performed as reported previously 17 with slight modification.¹ Two spin speeds were applied to perform nano-pinning: 500 rpm for 10 18 s and then 3000 rpm for 90 s. When 70 s passed, 100 µL of toluene or chloroform was added on 19 the substrate in less than 2 s. For thermal annealing, substrates were heated on a hotplate at 70 °C 20 for 10 min. For DMSO-vapor-assisted thermal annealing, substrates were first heated on a hotplate 21 at 70 °C for 10 min. Then the substrates were covered with a glass petri dish on the hotplate, 10 22 µL of DMSO was added from the edge of the petri dish and heated at 70 °C for 60 min. 23





Figure S1. SEM images of CH₃NH₃PbCl₃ thin film fabricated via (A) one-step spin coating with thermal annealing, (B) one-step spin coating with DMSO-vapor-assisted thermal annealing, (C) nano-pinning with chloroform followed by thermal annealing, (D) nano-pinning with toluene followed by thermal annealing, (E) nano-pinning with chloroform followed by DMSO-vapor-assisted thermal annealing and (F) nano-pinning with toluene followed by DMSO-vapor-assisted thermal annealing. Scale bar: 10 µm.



Figure S2. SEM images of CH₃NH₃PbCl₃ thin films made by two-step spin coating under different annealing conditions for each layer. PbCl₂ layer was annealed at X °C for 10 min. After spin coating CH₃NH₃Cl, the substrates were annealed at Y °C for 30 min. For DMSO-vapor-assisted thermal annealing, the substrates were annealed at Z °C for 1 h. The values of X, Y, and Z were (A, B) 70, 70 and 70; (C, D) 70, 100 and 100; and (E, F) 100, 100 and 100. Scale bars are 20 µm for A, C, E, and 5 µm for B, D, F.



Figure S3. SEM images of PbCl₂ thin film annealed at 100 °C for 10 min. Scale bars are 20 μm and 5 μm for A
and B, respectively.

40 Bulk crystal growth

Bulk crystal was grown by inverse temperature crystallization method as reported previously with slight modification.^{2, 3} A 1 M CH₃NH₃PbCl₃ precursor solution was prepared by dissolving equal mole CH₃NH₃Cl and PbCl₂ in DMSO-DMF (1:1 by volume). The solution was then filtered using a PTFE filter with 0.2 μ m pore size. The solution was sealed in a 10 mL vial and kept undisturbed for 1 h under 85 °C. The small crystals were kept as seeds. The same procedure was repeated by adding seeds to grow large crystals.



⁴⁸ Figure S4. Photograph of CH₃NH₃PbCl₃ bulk crystal grown via modified inverse temperature crystallization

⁴⁹ method.



51 Figure S5. SEM image of the CH₃NH₃PbCl₃ thin film fabricated with DMSO-vapor-assisted thermal annealing





58 Figure S6. Steady-state photoluminescence spectra of CH₃NH₃PbCl₃ thin film, pure PTAA thin film, and the



61 Figure S7. Specific detectivity of the device versus wavelength under different reverse biases.

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64 Figure S8. Photograph of devices exposed in ambient air (From left to right: 0 min, 1 min, 2 min, 3 min)

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