

# **Supporting Information**

# Ultrasensitive Photodetectors Based on Island-Structured $\text{CH}_3\text{NH}_3\text{PbI}_3$ Thin Films

*Yin Zhang<sup>†,⊥,‡</sup>, Juan Du<sup>†,‡</sup>, Xiaohan Wu<sup>†</sup>, Guoqian Zhang<sup>†</sup>, Yingli Chu<sup>†</sup>, Dapeng Liu<sup>†</sup>, Yixin Zhao<sup>\*,§</sup>, Ziqi Liang<sup>\*,<sup>‡</sup></sup> and Jia Huang<sup>\*,†,⊥</sup>*

<sup>†</sup> School of Materials and Engineering, Tongji University, Shanghai, 201804, P. R. China.

<sup>⊥</sup> Key Laboratory of Advanced Civil Engineering Materials, Tongji University, Ministry of Education, Shanghai, 201804, P. R. China

<sup>§</sup> School of Environmental Science and Engineering, Shanghai Jiao Tong University, Shanghai 200240, China

<sup>‡</sup> Department of Materials Science, Fudan University, Shanghai 200433, China

\* Address correspondence to [huangjia@tongji.edu.cn](mailto:huangjia@tongji.edu.cn), [yixin.zhao@sjtu.edu.cn](mailto:yixin.zhao@sjtu.edu.cn), and [zqliang@fudan.edu.cn](mailto:zqliang@fudan.edu.cn)

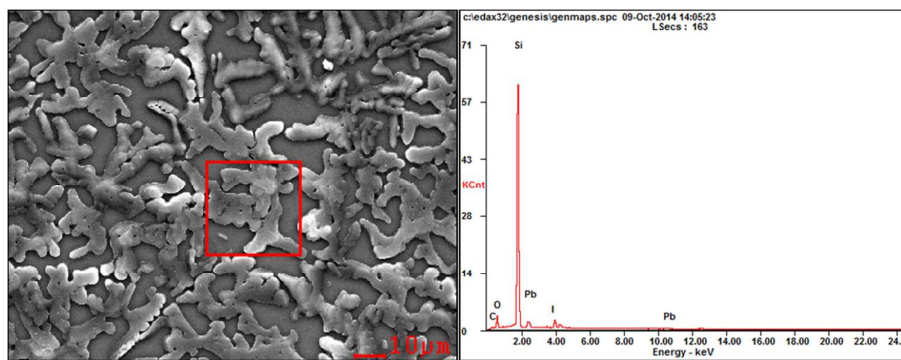
<sup>‡</sup>These authors contributed equally to this work

## Materials and Methods:

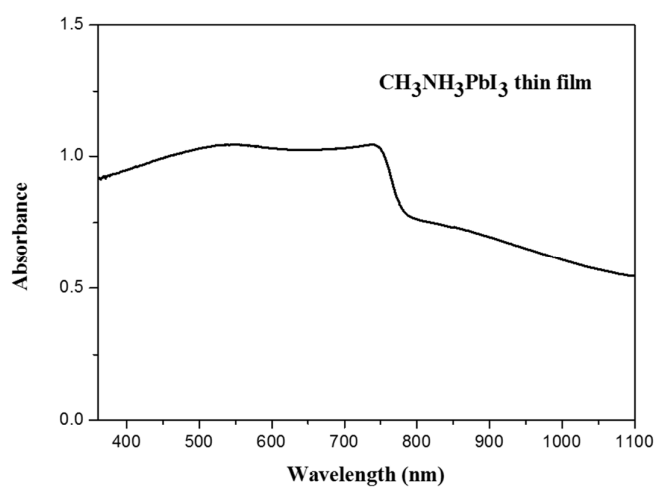
$\text{CH}_3\text{NH}_3\text{I}$  was synthesized by mixing 33 wt% methylamine (purchased from Sigma-Aldrich) in ethanol and 57 wt% hydroiodic acid (purchased from Acros) in water, in an ice bath for 2 h with stirring. After removing the solvent by rotary evaporator, the obtained solids were washed with ethyl acetate for several times. Similarly,  $\text{CH}_3\text{NH}_3\text{Cl}$  was synthesized by reacting 33 wt% methylamine and 33 wt% hydrochloric acid in an ice bath for 2h with stirring followed by vacuum drying and cleaning with acetonitrile.

$\text{CH}_3\text{NH}_3\text{PbI}_3$  perovskite precursor solutions were synthesized by adding 1.5 mmol  $\text{PbI}_2$  (purchased from Sigma Aldrich), 1.5 mmol  $\text{CH}_3\text{NH}_3\text{I}$  and  $\text{CH}_3\text{NH}_3\text{Cl}$  in N,N-Dimethylformamide (DMF), followed by a gentle sonication until a homogeneous solution was formed. For the islands-structured  $\text{CH}_3\text{NH}_3\text{PbI}_3$  films, 0.75 mmol  $\text{CH}_3\text{NH}_3\text{Cl}$  was used. For the compact film, 1.5 mmol  $\text{CH}_3\text{NH}_3\text{Cl}$  was used. Note that the amount of  $\text{CH}_3\text{NH}_3\text{Cl}$  used is essential to the resulted thin-film morphology. ( J. Phy. Chem. C 2014, 118 (18), 9412-9418) Si/SiO<sub>2</sub> substrates were cleaned by ultra-sonication in acetone, isopropanol and ultrapure water for 30 min respectively followed by N<sub>2</sub> drying, and then treated by plasma cleaning for 5 min (Plasma Cleaner PDC-32G).

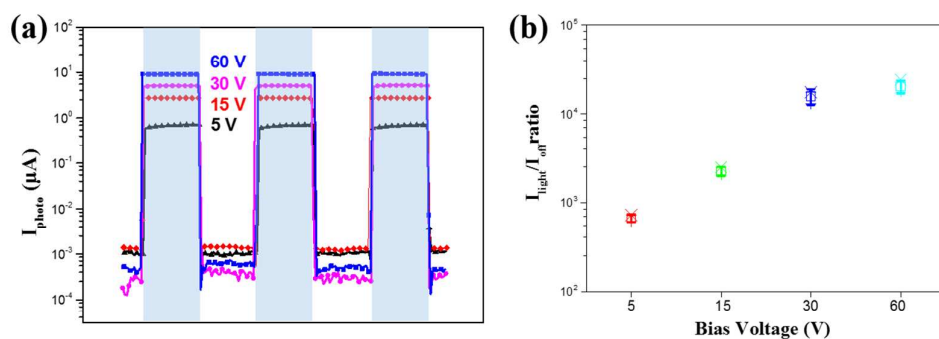
The photodetector devices were fabricated by spin-coating the precursor solutions onto the cleaned substrates at 2500 rpm for 30 s, followed by thermal annealing at 100 °C for 25 min. During the fabrication process, the color of the  $\text{CH}_3\text{NH}_3\text{PbI}_3$  perovskite film changed from light yellow to black after thermal annealing, indicating that the  $\text{ABO}_3$  structure was gradually formed at high temperature during the evaporation of the DMF solvent. 80 nm gold electrodes were deposited by thermal evaporation. A shadow mask with channel width of 600 $\mu\text{m}$  and channel length of 200 $\mu\text{m}$  was used to pattern electrode pads on the film. To protect the device from the moisture in air, PMMA thin films were spin-coated onto the surface of devices used for electrical measurement. All device fabrications were performed in a glove box filled with Ar. The Electrical characterizations were performed using a Keithley 4200 Semiconductor Characterization System connected to a vacuum probe station.



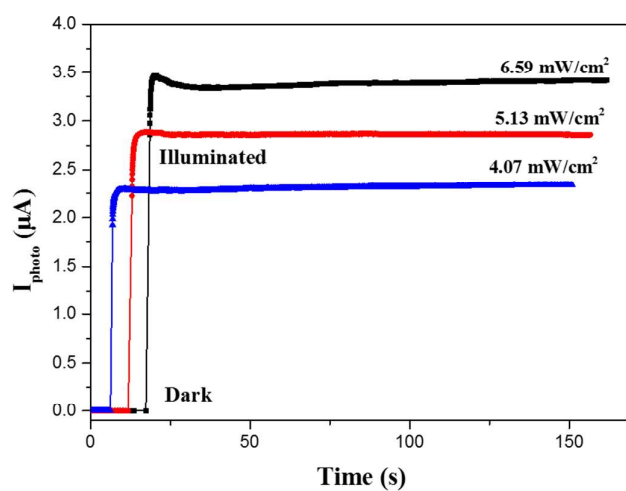
**Figure S1.** Energy Dispersive Analysis System of X ray (EDX) result of perovskite thin film.



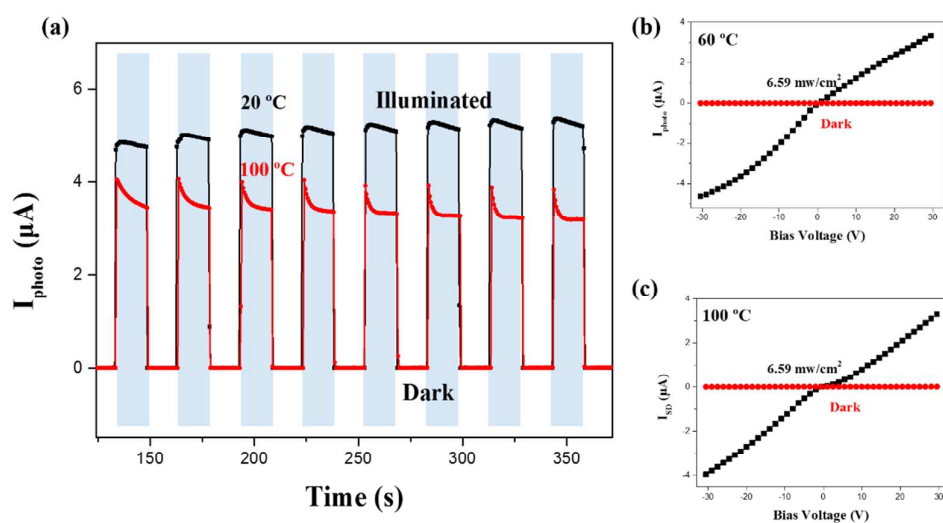
**Figure S2.** UV-Visible absorbance spectrum of hybrid perovskite thin film.



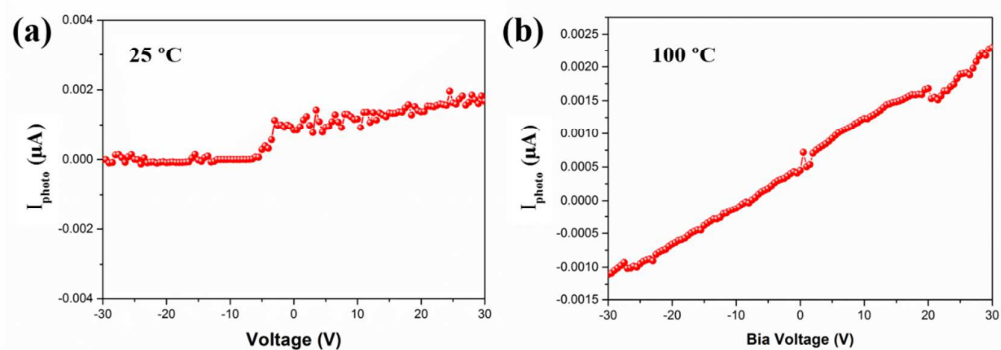
**Figure S3.** (a) Transient photoresponse properties with bias voltage of 5, 15, 30 and 60 V by log scale. (b) The dependency of light photocurrent to dark ratio on bias voltage. The light power density is 6.58 mW/cm<sup>2</sup>.



**Figure S4.** Working stability test of perovskite photodetector at 6.59, 5.13 and 4.07  $\text{mW}/\text{cm}^2$  power density, respectively.



**Figure S5.** Transient photoresponse (a) and I-V character (b) and (c) of  $\text{CH}_3\text{NH}_3\text{PbI}_3$  perovskite photodetector at various temperature.



**Figure S6.** The dark I-V curve of island-structured  $\text{CH}_3\text{NH}_3\text{PbI}_3$  photodetector at (a) room and (b) high temperature.