Supporting Information

Synthesis of Ultrathin Two-Dimensional Organic-Inorganic Hybrid Perovskite Nanosheets for Polymer Field-Effect Transistors

Lijie Zhu[a], Huaye Zhang[a], Qipeng Lu[b], Yue Wang[a], Zhenbo Deng[*a], Yufeng Hu[*a], Zhidong Lou[a], Qiuhong Cui[a], Yanbing Hou[a] and Feng Teng[a]

[a]*Key laboratory of Luminescence and Optical Information, Ministry of Education, Beijing JiaoTong University, Beijing 100044, China*

[b]*School of Materials Science and Engineering, Nanyang Technological University, 50 Nanyang Avenue, Singapore 639798, Singapore.*

**Corresponding authors. Tel: +86-10-51684860; Fax: +86-10-51688018.*

zbdeng@bjtu.edu.cn (Z. Deng) and yfhu@bjtu.edu.cn (Y. Hu)

Experimental section

Materials.

Lead iodide (PbI_2, 99.9%), lead bromide ($PbBr_2$, 98%), lead chloride ($PbCl_2$, 98%), N, N-dimethylmethanamide (DMF, 99.8%), toluene (99.8%), triethylamine (TEA) were purchased from Alfa Aesar. Phenylethylammonium iodide (PEAI, 99%), phenylethylammonium bromide (PEABr, 99%) and phenylethylammonium chloride (PEACl, 99%) were purchased from Xi'an Polymer Light Technology Corp. PVP powder (Mw~20000), PVA powder (Mw~30000-70000), 4, 4′-(hexafluoroisopropylidene) diphthalic anhydride (HDA, 99%) and propylene glycol monomethyl ether acetate (PGMEA) were purchased from Sigma Aldrich. All the chemicals were used as received without further purification.

Synthesis of 2D $(PEA)_2PbX_4$ (X=Cl, Br, I) nanosheets (NSs).

In a typical synthesis of $(PEA)_2PbI_4$ NSs, 0.1 mmol of PEAI and 0.05 mmol of PbI_2 were dissolved in 1 mL of DMF to form a perovskite precursor solution. Then, 15 μL of the perovskite precursor solution were quickly dropped into 10 mL of toluene under vigorous stirring. The $(PEA)_2PbI_4$ NSs were obtained after centrifugation at 7,000 rpm for 1 min. The $(PEA)_2PbBr_4$ and $(PEA)_2PbCl_4$ NSs were synthesized by using the same method. The perovskite precursors were changed to PEABr and $PbBr_2$ for $(PEA)_2PbBr_4$ NSs and PEACl and $PbCl_2$ for $(PEA)_2PbCl_4$ NSs, respectively.

Material characterization

The X-ray diffraction (XRD) patterns were recorded by a Bruker D8 X-ray diffractometer with Cu Kα radiation (λ=1.5406Å, 40 kV, 40 mA). Samples used for

TEM, SEM and AFM characterizations were prepared by dropping colloidal dispersion in toluene onto the amorphous carbon-coated copper grids, Si, and Si, respectively, and then naturally dried. Transmission electron microscopy (TEM) images were recorded on a JEM-1400. SEM and EDS mapping images were obtained by a Hitachi (S-4800) field emission scanning electron microscope. AFM (Cypher S, Asylum Research) was used to characterize the 2D perovskite NSs in tapping mode under ambient conditions. Fluorescence spectra were recorded on a Cary Eclipse spectrophotometer at the excitation wavelength of 365 nm. The UV-vis absorption spectra were taken on a Shimadzu UV-3101 PC spectrophotometer. All the liquid samples were tested using 1×1 cm^2 path length quartz cuvettes

Device fabrication

The FETs were fabricated with a structure of bottom gate and bottom contact. The patterned indium tin oxide (ITO) coated glass substrates which act as the gate electrode were pre-cleaned with deionized water, acetone, and isopropanol in an ultrasonic bath for 15 min each. Subsequently, Polyvinyl alcohol (PVA) film and the surface modification layer of crosslinking poly (4-vinylphenol) (CL-PVP) were fabricated according to the literature.[1, 2] Subsequently, the semiconductor films were fabricated by spin-coating the composite solutions of P3HT and $(PEA)_2PbX_4$ NSs (X= Cl, Br, I) on the dielectric layer and annealing for 10 minutes at 110 ^oC. The composite solution formed by dispersing the obtained $(PEA)_2PbX_4$ NSs (X= Cl, Br, I) into the P3HT toluene solution which had pre-dissolved in toluene with a concentration of 2 mg/ml. Finally, the gold (Au) film was thermally evaporated on top of the films through a

shadow mask to make the top contact source-drain electrode (50 nm) under the high vacuum (5×10^{-6} torr).

Device characterization

Transistor electrical characterization was performed with a Keithley 4200SCS semiconductor parameter analyzer connected to a standard probe station setup at room temperature in the air. Capacitance measurements were carried out using a sandwich electrode configuration with an Agilent E4990A Impedance Analyzer for frequencies ranging between 100 Hz and 1MHz.

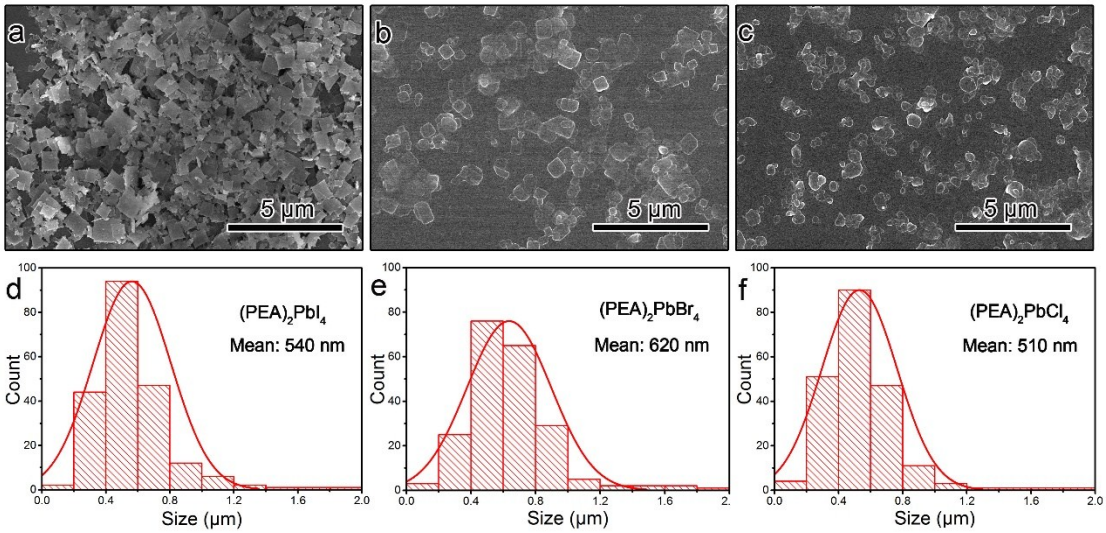

Figure S1. (a) SEM images and (d) the corresponding lateral size distribution of $(PEA)_2PbI_4$ NSs. (b) SEM images and (e) the corresponding lateral size distribution of $(PEA)_2PbBr_4$ NSs. (c) SEM images and (f) the corresponding lateral size distribution of $(PEA)_2PbCl_4$ NSs.

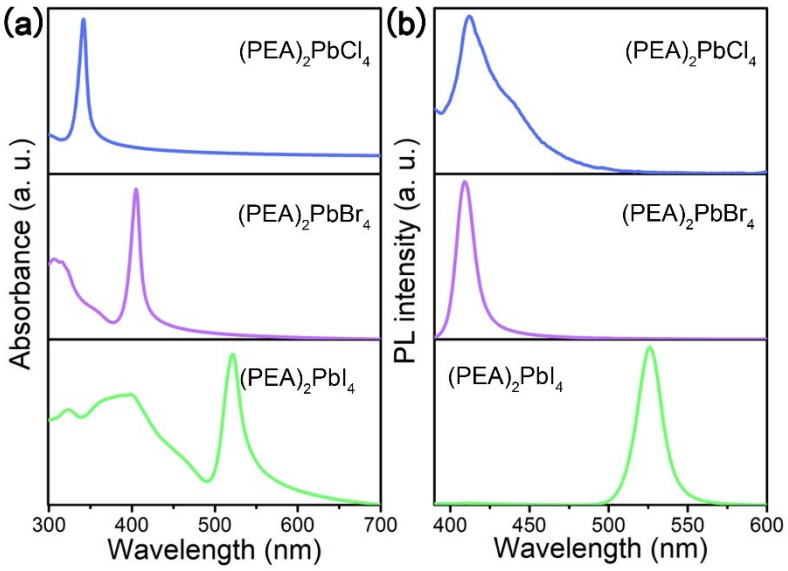

Figure S2. (a) UV/Vis absorption and (b) photoluminescence (PL) spectra of $(PEA)_2PbX_4$ NSs (X= Cl, Br, I).

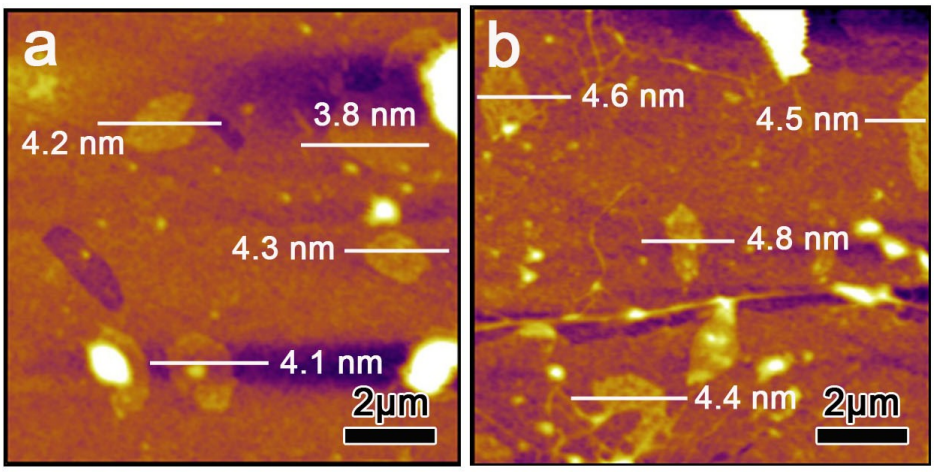

Figure S3. AFM height images and the corresponding thickness of the (a) $(PEA)_2PbBr_4$ NSs and (b) $(PEA)_2PbCl_4$ NSs.

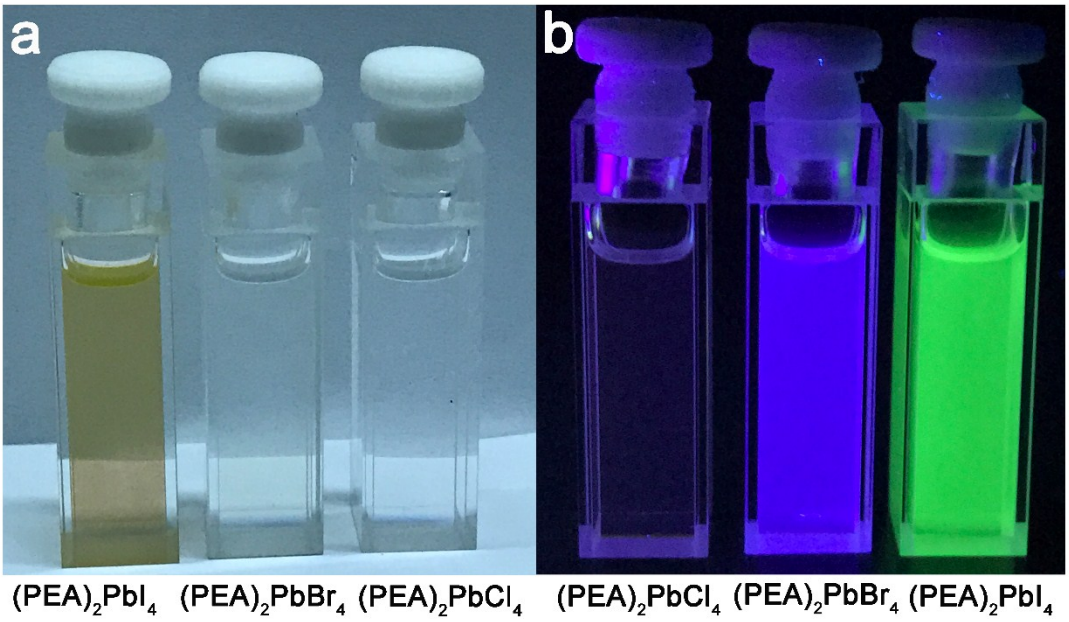

Figure S4. Photographs of $(PEA)_2PbX_4$ NSs solutions (X= Cl, Br, I) (a) under the day light and (b) under the UV irradiation at 365 nm.

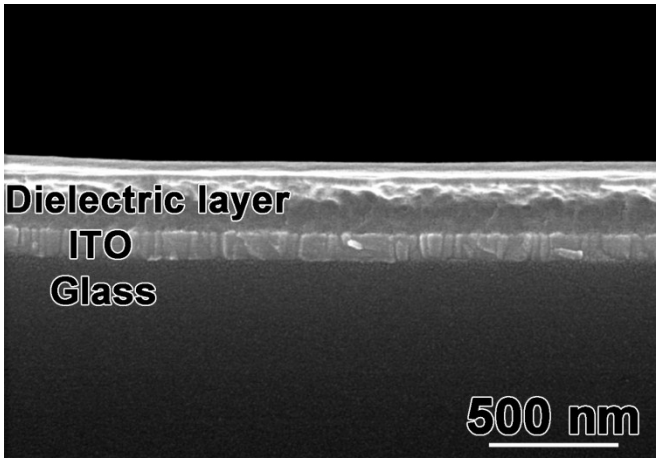

Figure S5**.** The cross-sectional SEM image of dielectric layer on ITO substrate.

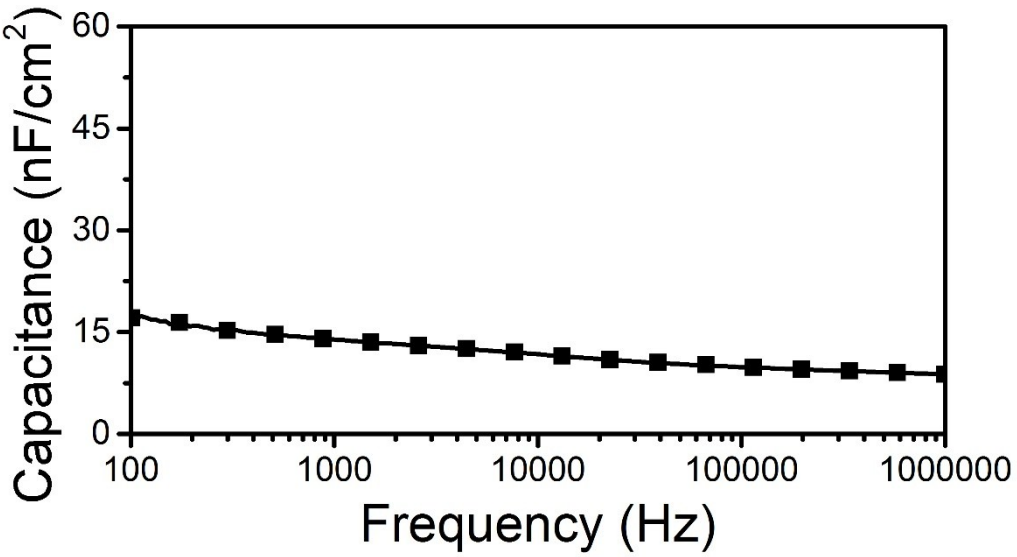

Figure S6. Capacitance *vs* frequency for characteristic with a capacitor area of 0.00125 cm^2

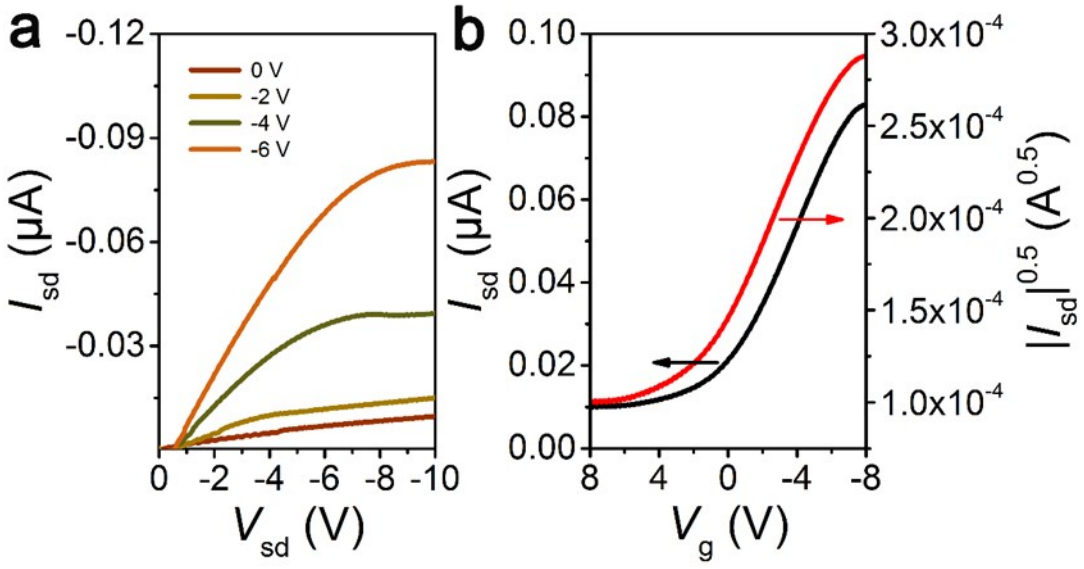

Figure S7. (a) Output and (b) transfer characteristics of a representative *p*-channel FET with P3HT.

Reference

Stable Low-Bandgap Pb–Sn

Binary Perovskites for Tandem

Solar Cells Stable Low-

Bandgap Pb–Sn Binary Perovsk

ites for Tandem Solar Cells

Stable Low-Bandgap Pb–Sn

Binary Perovskites for Tandem

Solar Cells

1. T. B. Singh, F. Meghdadi, S. Günes, N. Marjanovic, G. Horowitz, P. Lang, S. Bauer and N. S. Sariciftci, *Adv. Mater.*, 2005, 17, 2315-2320.
2. M. E. Roberts, N. Queraltó, S. C. Mannsfeld, B. N. Reinecke, W. Knoll and Z. Bao, *Chem. Mater.*, 2009, 21, 2292-2299.