Supporting Information

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

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Experimental section

Materials.

Lead iodide (PbI₂, 99.9%), lead bromide (PbBr₂, 98%), lead chloride (PbCl₂, 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)₂PbX₄ (X=Cl, Br, I) nanosheets (NSs).

In a typical synthesis of (PEA)₂PbI₄ NSs, 0.1 mmol of PEAI and 0.05 mmol of PbI₂ 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)₂PbI₄ NSs were obtained after centrifugation at 7,000 rpm for 1 min. The (PEA)₂PbBr₄ and (PEA)₂PbCl₄ NSs were synthesized by using the same method. The perovskite precursors were changed to PEABr and PbBr₂ for (PEA)₂PbBr₄ NSs and PEACl and PbCl₂ for (PEA)₂PbCl₄ NSs, respectively.

Material characterization

The X-ray diffraction (XRD) patterns were recorded by a Bruker D8 X-ray diffractometer with Cu Ka 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 cm² 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. Subsequently, the semiconductor films were fabricated by spin-coating the composite solutions of P3HT and (PEA)$_2$PbX$_4$ NSs (X= Cl, Br, I) on the dielectric layer and annealing for 10 minutes at 110 °C. The composite solution formed by dispersing the obtained (PEA)$_2$PbX$_4$ 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 ($5 \times 10^{-6}$ 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)$_2$PbI$_4$ NSs. (b) SEM images and (e) the corresponding lateral size distribution of (PEA)$_2$PbBr$_4$ NSs. (c) SEM images and (f) the corresponding lateral size distribution of (PEA)$_2$PbCl$_4$ NSs.
Figure S2. (a) UV/Vis absorption and (b) photoluminescence (PL) spectra of (PEA)$_2$PbX$_4$ NSs (X= Cl, Br, I).
Figure S3. AFM height images and the corresponding thickness of the (a) (PEA)$_2$PbBr$_4$ NSs and (b) (PEA)$_2$PbCl$_4$ NSs.
Figure S4. Photographs of (PEA)$_2$PbX$_4$ 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-
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Stable Low-Bandgap Pb–Sn
Binary Perovskites for Tandem
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