Supporting Information for

Highly Oriented Two-Dimensional Formamidinium Lead Iodide Perovskite with a Small Bandgap of 1.51 eV

Jielin Yan[†a], Weifei Fu[†a], Xinqian Zhang [a], Jiehuan Chen [a], Weitao Yang [a], Weiming Qiu [c], Gang Wu [a]*, Feng Liu [b]*, Paul Heremans[c], Hongzheng Chen [a]*

a…State Key Laboratory of Silicon Materials, MOE Key Laboratory of Macromolecular Synthesis and Functionalization, Department of Polymer Science and Engineering, Zhejiang University, Hangzhou 310027, P. R. China

b…Department of Physics and Astronomy and Collaborative Innovation Center of IFSA (CICIFSA), Shanghai Jiaotong University, Shanghai 200240, China

c…. IMEC, Kapeldreef 75, Heverlee, B-3001, Belgium.
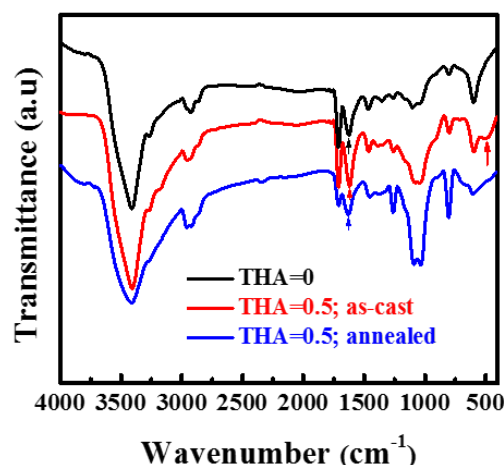
† These authors contributed equally to this work.

Figure S1. Fourier transform infrared (FTIR) spectra of $(BA)_2(FA)_2Pb_3I_{10}$ films fabricated with or without thiourea in precursors.

As shown in Figure S1, a weak absorption peak assigned to thiourea appears at around 500 cm[-1] for the as-casted film with 0.5M thiourea.[1] The N-H deformation of

FA group at 1625 cm^{-1} shifts to 1620 cm^{-1} when thiourea is added, indicating an interaction between thiourea and FA cation probably due to the H-bond formed between $-NH_2$ groups in FA and thiourea.[2] After annealing, the peak at 500 cm^{-1} disappears and the peak at 1620 cm^{-1} shifts back to 1625 cm^{-1}. Thus, we can speculate that thiourea is evaporated after thermal annealing.
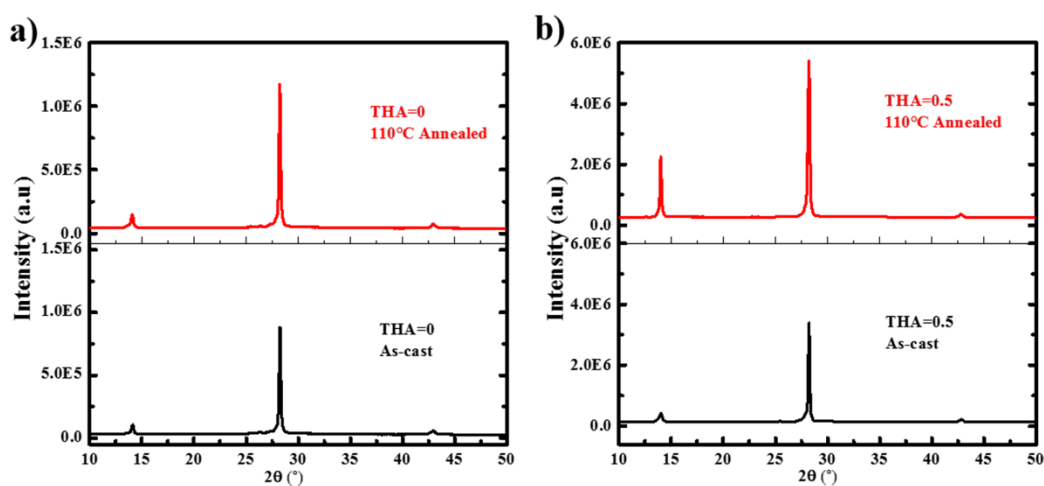

Figure S2. XRD patterns of the as-cast and annealed $(BA)_2(FA)_2Pb_3I_{10}$ films fabricated with THA=0 (a) and THA=0.5 (b).
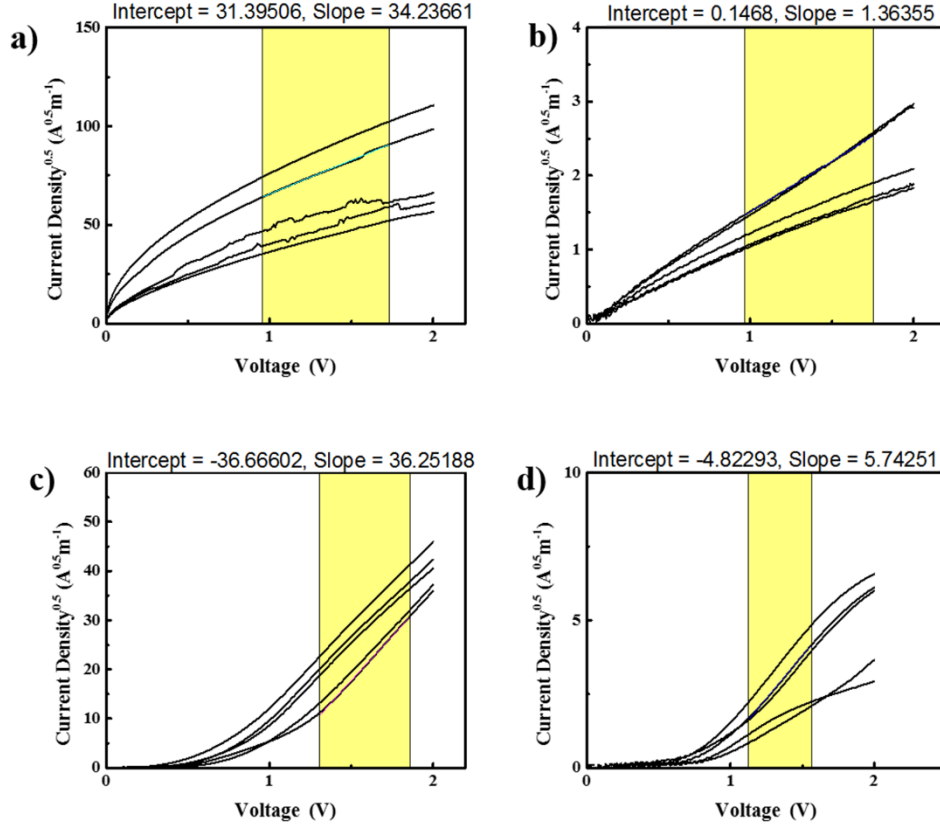
Figure S3. $J_D^{0.5}$-V curves (each contains five devices) for hole only (a, b) and electron only (c, d) devices of 2D perovskite films with THA=0.5 (a, c) and THA=0 (b, d). The devices were measured under dark state according to space charge limited current (SCLC) method.

The charge mobility was calculated by Mott-Gurney law:

$$J_D = \frac{9}{8}\varepsilon_0\varepsilon\mu\frac{V^2}{L^3}$$

Where J_D is current density, ε is the static permittivity (here $\varepsilon \approx 6$ for perovskite), ε_0 is the permittivity of free space, μ is the mobility, V is the applied voltage, and L is the film thickness (here L=300 nm). By fitting linear area of the $J_D^{0.5}$-V curve, the mobility can be obtained. The final mobility was calculated from five devices on average.
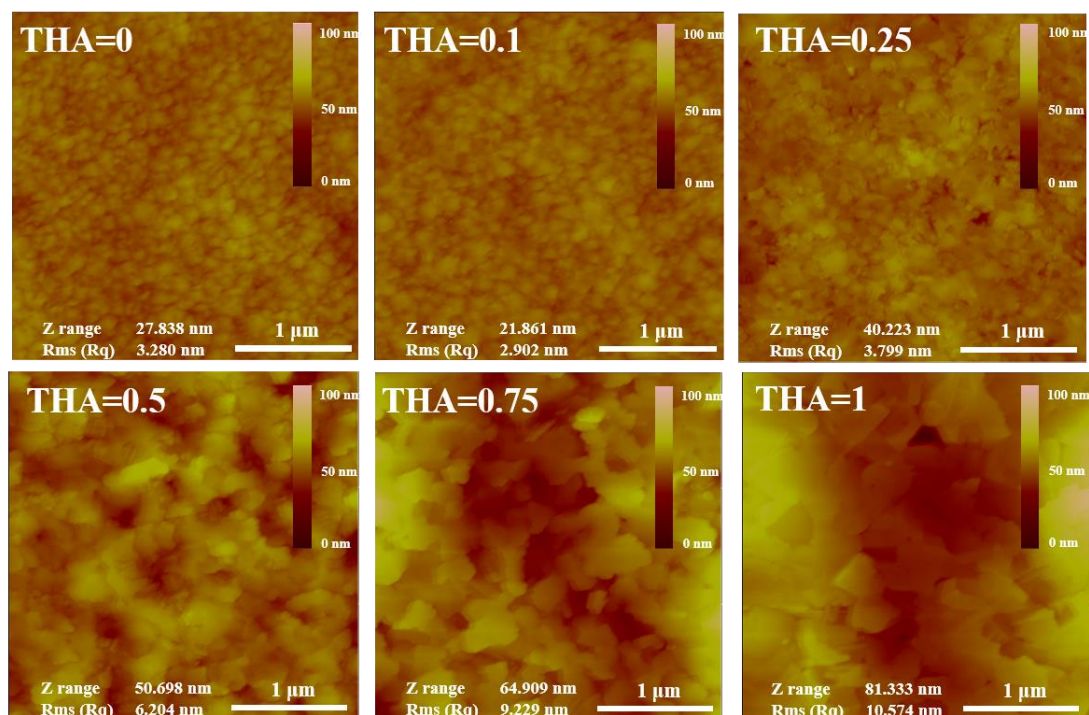
Figure S4. AFM images of the annealed $(BA)_2(FA)_2Pb_3I_{10}$ films fabricated with different precursors containing various amounts of thiourea.

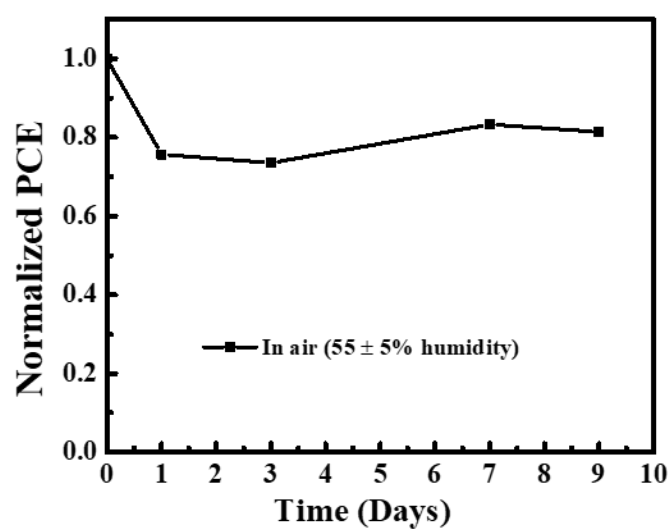**Figure S5**. Evolution of PCE under relative humidity ($55 \pm 5\%$) for unencapsulated device based on $(BA)_2(FA)_2Pb_3I_{10}$.

References

1. G. Madhurambal, M. Mariappan and S. C. Mojumdar, *J. Therm. Anal.*

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2.   J. W. Lee, H. S. Kim and N. G. Park, *Acc. Chem. Res.*, 2016, **49**, 311-319