## Supporting Information for

## Unveiling the role of linear alkyl organic cations in 2D layered tin halide

## perovskite field-effect transistors

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Figure S1. RMS of the five perovskite films based on different processing parameters.



Figure S2. Top-view SEM images of (a) (PA)<sub>2</sub>SnI<sub>4</sub>; (b) (BA)<sub>2</sub>SnI<sub>4</sub>; (c) (PenA)<sub>2</sub>SnI<sub>4</sub>; (d) (HA)<sub>2</sub>SnI<sub>4</sub>; and (e) (HepA)<sub>2</sub>SnI<sub>4</sub> thin films on silicon substrates prepared at the perovskite precursor concentration of 0.2 M in pure DMF solvent, respectively.



Figure S3. (a) PL and (b) time-resolved PL decay curves of the optimized five thin films with the concentration of 0.2 M and pure DMF solvent.



Figure S4. Optical image of (HepA)<sub>2</sub>SnI<sub>4</sub> film cast from 0.2 M DMF:50%DMSO.



Figure S5. The intensity GIWAXS profiles along the  $q_z$  axis for perovskite films obtained by different processing parameters of (a) 0.1 M DMF; (b) 0.2 M DMF; and (c) 0.2 M DMF:DMSO.



Figure S6. The intensity GIWAXS profiles along the  $q_{xy}$  axis for perovskite films obtained by different processing parameters of (a) 0.1 M DMF; (b) 0.2 M DMF; and (c) 0.2 M DMF:DMSO.



Figure S7. Transfer characteristics of (a) (PA)<sub>2</sub>SnI<sub>4</sub>; (b) (BA)<sub>2</sub>SnI<sub>4</sub>; (c) (PenA)<sub>2</sub>SnI<sub>4</sub>; (d) (HA)<sub>2</sub>SnI<sub>4</sub>; and (e) (HepA)<sub>2</sub>SnI<sub>4</sub> at 295 K based on processing parameters of 0.1 M DMF.



Figure S8. Ultraviolet photoelectron spectroscopy (UPS) of (a)  $(PA)_2SnI_4$ , (b)  $(BA)_2SnI_4$ , (c)  $(PenA)_2SnI_4$ , (d)  $(HA)_2SnI_4$ , and (e)  $(HA)_2SnI_4$  perovskite thin films. (f-j) The corresponding cutoff region of the spectrum of these five perovskite materials.



Figure S9. The energy level diagrams of the perovskite thin films and the work function of the Au electrode.



Figure S10. Transfer curve of 2D  $(HA)_2SnI_4$  FET measured in linear and saturation region at room temperature.



Figure S11. Bias stress stability of  $(HA)_2SnI_4$  FET under a constant bias of  $V_g = V_{ds} = -60$  V.

Table S1. Comparison of charge carrier mobilities at room temperature of reported 2D layered Snperovskite field-effect transistors.

Perovskite layer	Optimization	Device structure	Dielectric layer	Channel length/width µm	Mobility $\mu_h$ (cm <sup>2</sup> V <sup>-1</sup> s <sup>-1</sup> )	Ref.
$(4TM)_2SnI_4$	-	BG-TC	SiO <sub>2</sub>	40/2880	2.32	1
(FA/PEA)SnI <sub>x</sub>	2D/3D	TG-BC	Al <sub>2</sub> O <sub>3</sub> /PMMA	20/10000	0.21	2
(PEA) <sub>2</sub> SnI <sub>4</sub>	PEO	BG-TC	PVA/CL-PVP	50/1000	1.31×10 <sup>-2</sup>	3
(TEA) <sub>2</sub> SnI <sub>4</sub>	Hot-casting	BG-TC	SiO <sub>2</sub>	80/1000	0.34	4
(TEA) <sub>2</sub> SnI <sub>4</sub>	Additive	BG-TC	SiO <sub>2</sub>	80/1000	0.7	5
(POE) <sub>2</sub> SnI <sub>4</sub>	NMP	BG-BC	CL-PVP/SiO <sub>2</sub>	100/1000	0.3	6
(HA) <sub>2</sub> SnI <sub>4</sub>	-	BG-TC	SiO <sub>2</sub>	80/1000	9×10 <sup>-3</sup>	This work

Note: All above summarized 2D perovskite thin-films were prepared by solution-processing unless otherwise noted.

Supplementary references:

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