

Electronic Supplementary Information

for

Factors determining the vertical orientation of two-dimensional perovskites

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1. Materials

Hydroiodic acid (HI), *N,N*-dimethylformamide (DMF), and *N,N*-dimethylacetamide (DMAc) were purchased from the FUJIFILM Wako Pure Chemical, Ltd. All other chemicals were obtained from the Tokyo Chemical Industry Co., Ltd. HOOC(CH₂)₆NH₃I (7-AHAI) and CH₃(CH₂)₆NH₃I (HeAI) were synthesized by neutralizing HOOC(CH₂)₆NH₂ and CH₃(CH₂)₆NH₂ with stoichiometric quantities of hydroiodic acid, respectively.

2. Sample preparation

7-AHAI and PbI₂ were dissolved in DMF or DMAc at r.t. for 1 h to obtain the precursor solution for the preparation of 7-AHAPbI. The concentrations of these solutions were 40 wt.% or 10 wt.%. Similarly, the precursor solution of HeAPbI was obtained. Films for XRD, and *In-plane* XRD measurements were fabricated on preheated hydrophilic substrates by spin-coating at 2000 rpm using a MIKASA SPINCOATER 1H-D7. In a glove box, films were fabricated by spin-coating at 1000, 2000, or 5000 rpm using an EHC heat-type spin-coater (SC-158HS). The heating temperature of substrates was 100°C for DMF solution (b.p. 153 ° C), and 120°C for DMAc solutions (b.p. 165°C) under consideration of the b.p. of the solvents. The substrates were heated at ca. 120°C during the spin-coating process to obtain high quality films when the EHC Heat-type spin coater was used.

3. Characterization

X-ray diffraction (XRD) patterns were obtained with an X-ray diffractometer

(SmartLab, Rigaku), in conjunction with a Ni-filtered copper $K\alpha$ target and operating at 45 kV and 200 mA. Scanning electron microscopy (SEM) images were obtained with an SU8000 instrument (HITACHI). Contact angles were obtained with an automatic contact angle meter (Drop Meter 300, manufactured by Kyowa Interface Science Co., Ltd.).

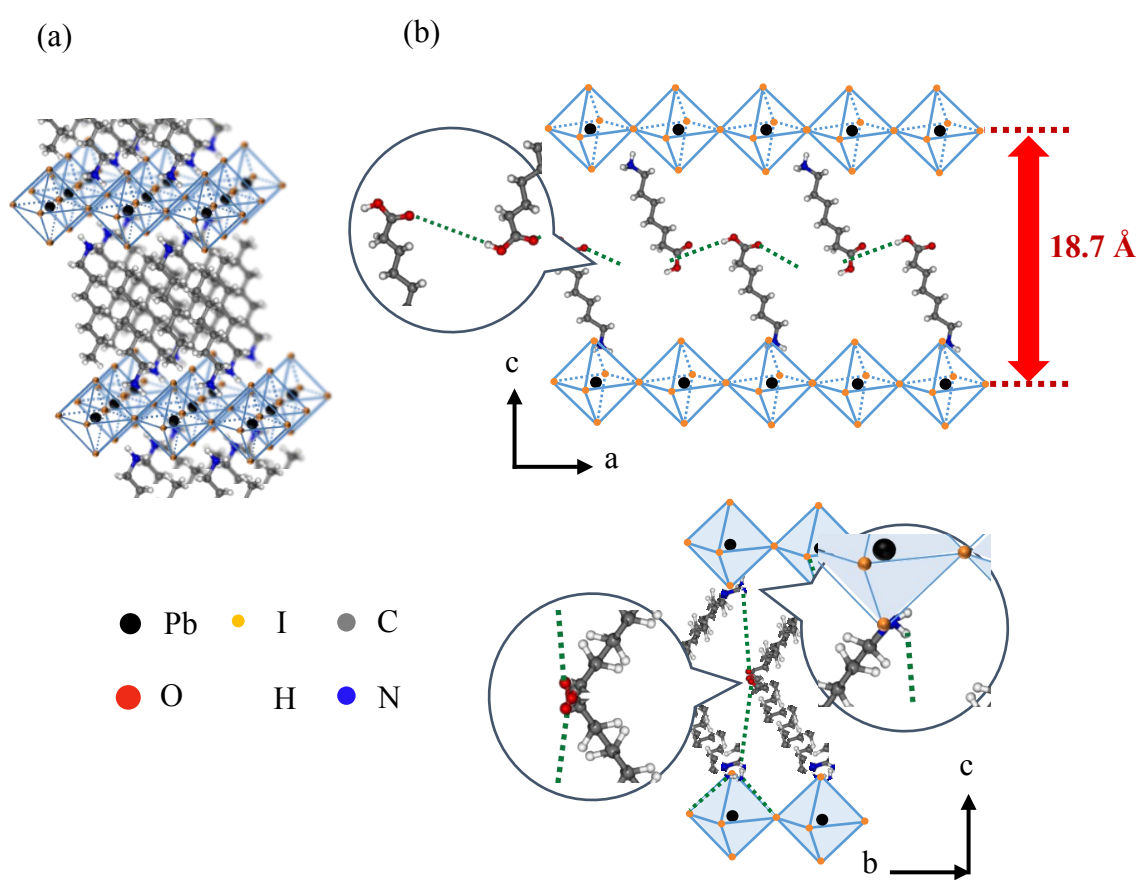


Fig. S1 Structures of (a) HeAPbI, and (b) 7-AHAPbI.

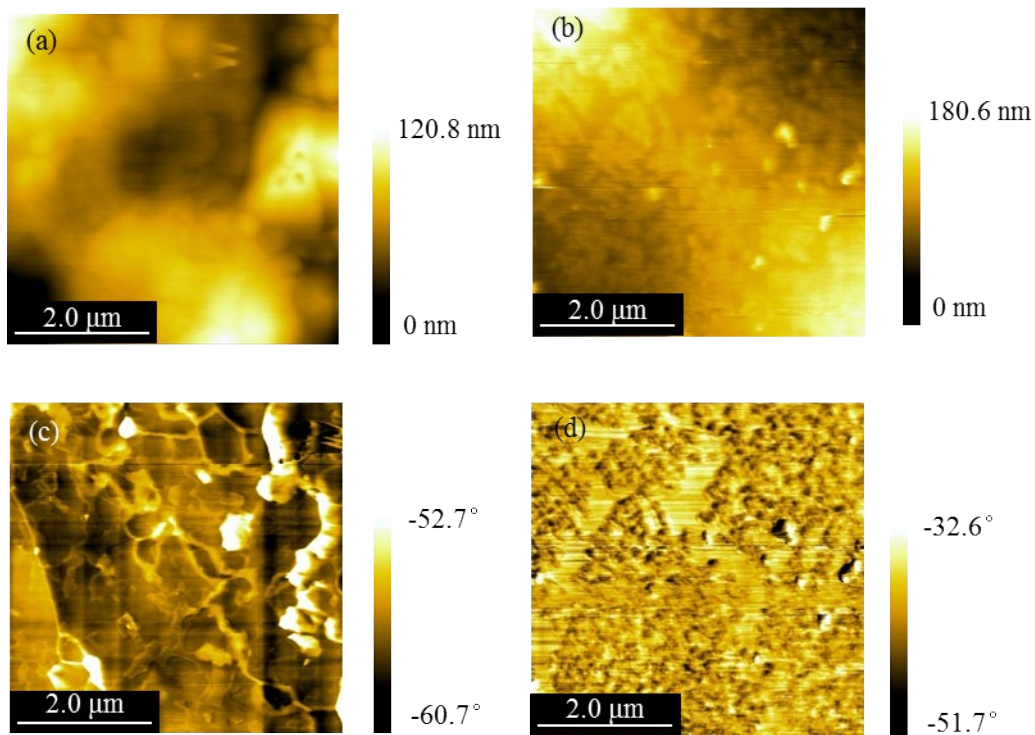


Fig. S2 Height (a, b) and phase (c, d) AFM images of 7-AHAPbI (a, c) and HePbI films (b, d) on mesoporous TiO₂ substrates.

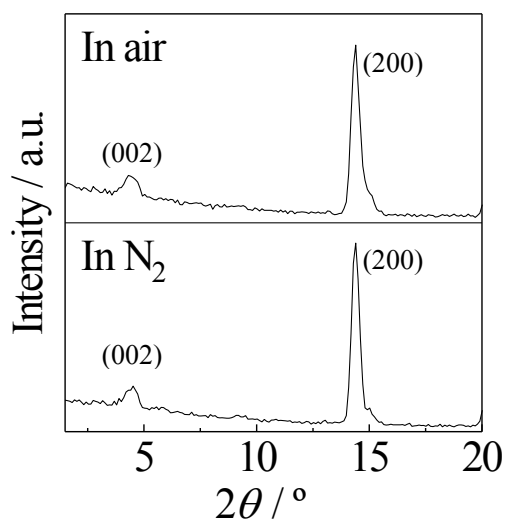


Fig. S3 *In-plane* X-ray diffraction patterns of 7-AHAPbI films fabricated from DMF precursor solution in air or N₂ atmosphere.

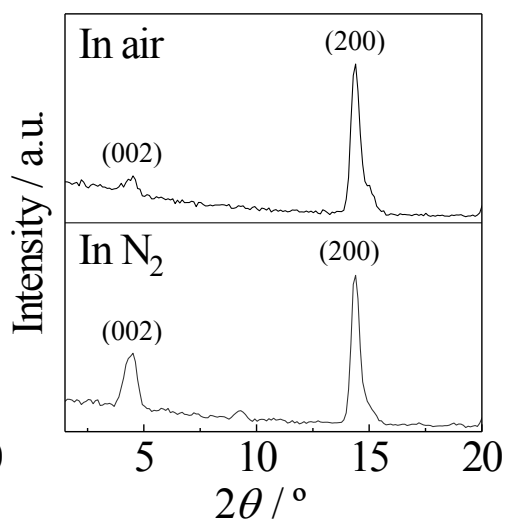


Fig. S4 *In-plane* X-ray diffraction patterns of 7-AHAPbI films fabricated from DMAc precursor solution in air or N₂ atmosphere.

Table S1 Ratio of intensities of (002) and (200) diffractions

Precursor solvent	Atmosphere	(002) : (200)
DMF	Air	11 : 89
	N ₂ (Glove box)	10 : 90
DMAc	Air	10 : 90
	N ₂ (Glove box)	28 : 72

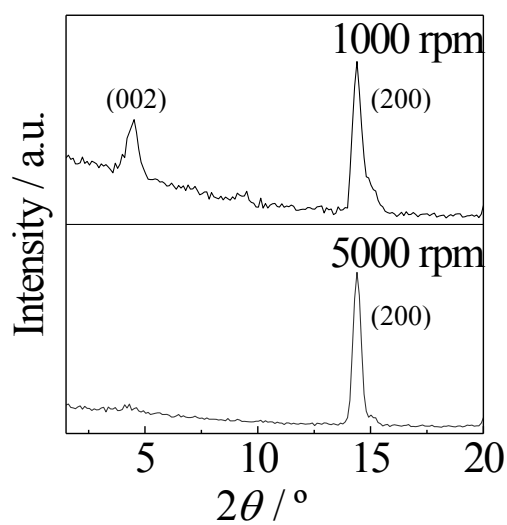


Fig. S5 *In-plane* X-ray diffraction patterns of 7-AHAPbI films fabricated by spinning at 1000rpm or 5000 rpm.

Table S2 Ratio of diffraction intensities (002) and (200)

Spin speed	(002) : (200)
1000 rpm	25 : 75
5000 rpm	×

× : (002) was not observed.

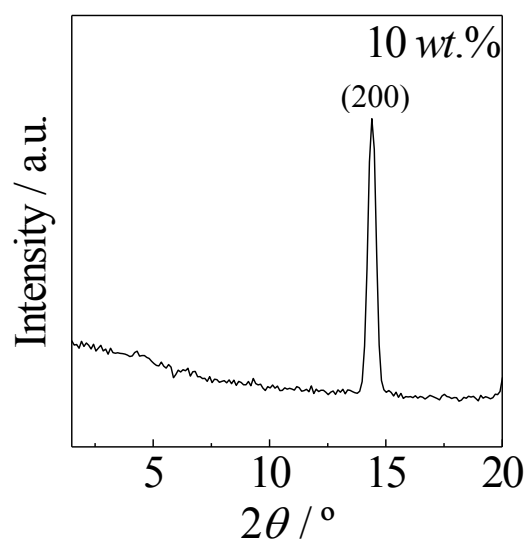


Fig. S6 *In-plane* X-ray diffraction pattern of 7-AHAPbI spin-coat film fabricated from 10 wt.% DMAc solution in N₂.

