Electronic Supplementary Information

A universal top-down approach toward thickness-controllable perovskite single-crystalline thin films

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S1. Thickness limit of the perovskite single-crystalline film

prepared by top-down method

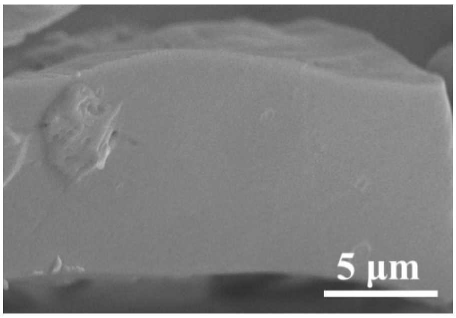

Fig. S1 Cross-sectional view of the thinnest $CH_3NH_3PbI_3$ single-crystalline thin film fabricated by top-down method.

Figure S1 shows the thinnest $CH_3NH_3PbI_3$ single-crystalline film obtained by top-down method. The thickness was less than 10 μm but its lateral dimension was less than 500 square microns and was difficult to be controllably fabricated. The reason lies in that when the film was thin to a certain extent, lateral etching to the film was dominant and the area of the thin film would quickly shrink. Therefore, 15 μm was considered as the thickness limit of the perovskite single-crystalline film prepared by such a top-down method.

S2. Optical transmission spectrum of $CH_3NH_3PbI_3$ single-

crystalline film

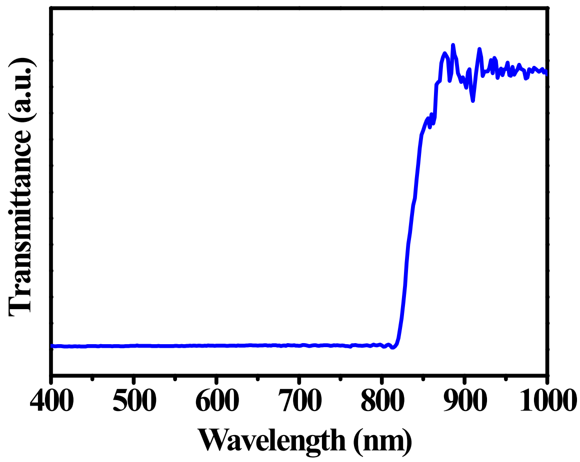

Fig. S2 Optical transmission spectrum of as-prepared $CH_3NH_3PbI_3$ single-crystalline film.

S3. Schematic illustration of the dissolution-crystallization equilibrium

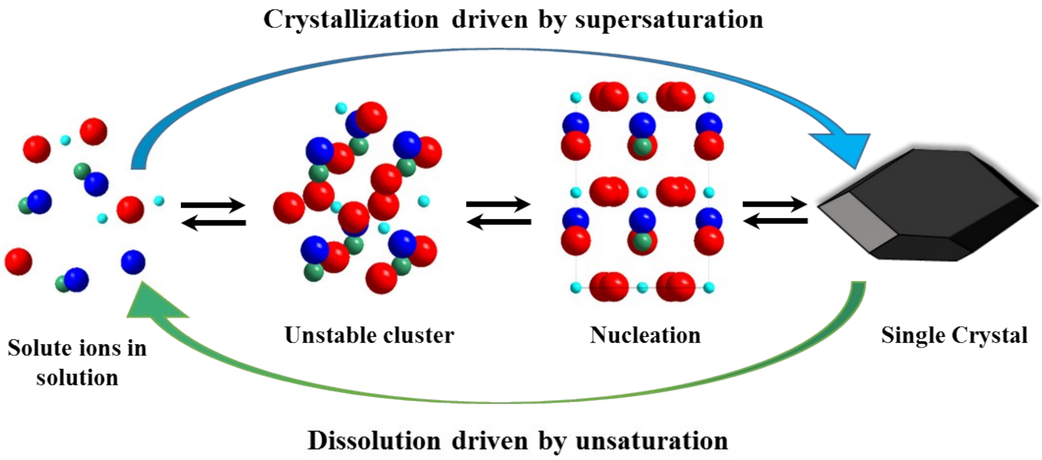

Fig. S3 Reversible dissolution-crystallization process of $CH_3NH_3PbI_3$ in aqueous solution system.

S4. Surface morphology of $CH_3NH_3PbI_3$ single crystal in

thinning process

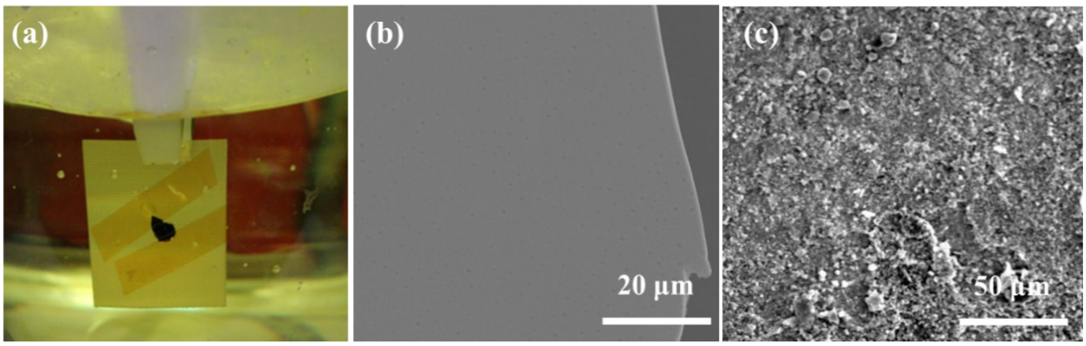

Fig. S4 (a) Photograph of a $CH_3NH_3PbI_3$ wafer immersed in etching solution for thinning. (b) The surface of the single crystal before thinning. (c) The surface of the single-crystalline film when residual etching solution was not timely removed.

Figure S4a shows a photograph of a $CH_3NH_3PbI_3$ wafer immersed in etching solution for thinning. Before thinning, the single crystal surface was very smooth (Fig. S4b). After etching, the surface of the wafer generally became rough because of two main reasons. Firstly, mechanical damage was easily induced during wire cutting and mechanical polishing. In the subsequent thinning process, $CH_3NH_3PbI_3$ at the locations where mechanical damage focused were more likely to be captured by etchant, resulting in nonuniform dissolution and uneven surface. If etching took place without stirring or at a high etching rate, this effect was more obvious. Secondly, after thinning, the timely removal of the residual solution on the single-crystalline film surface was necessary. If not promptly removed, it would cause microcrystalline precipitation on the crystal surface (Fig. S4c).

S5. Photographs of perovskite single-crystalline wafers

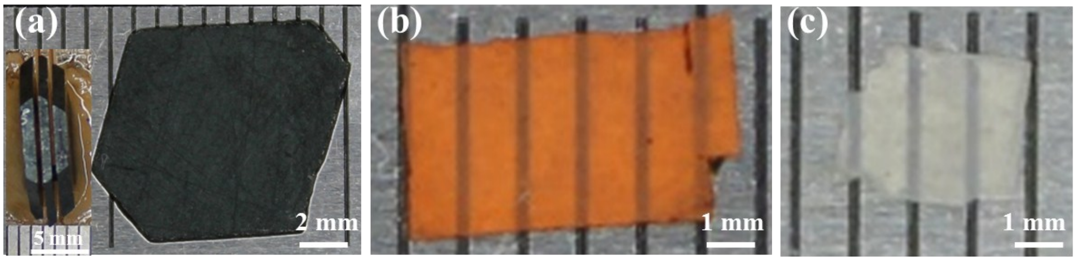

Fig. S5 Photographs of (a) $CH_3NH_3PbI_3$, (b) $CH_3NH_3PbBr_3$, (c) $CH_3NH_3PbCl_3$ single crystal wafers before wet thinning. The inset in (a) shows one bulk $CH_3NH_3PbI_3$ single crystal after wire cutting.

Figure S5 shows photographs of different types of perovskite single crystal wafers. They were fabricated by wire cutting and mechanical polishing from corresponding bulk single crystals. They were used as raw materials for sequent wet etching.

S6. Thickness evolution and crystalline quality evaluation of

$CH_3NH_3PbBr_3$ single-crystalline film during wet etching

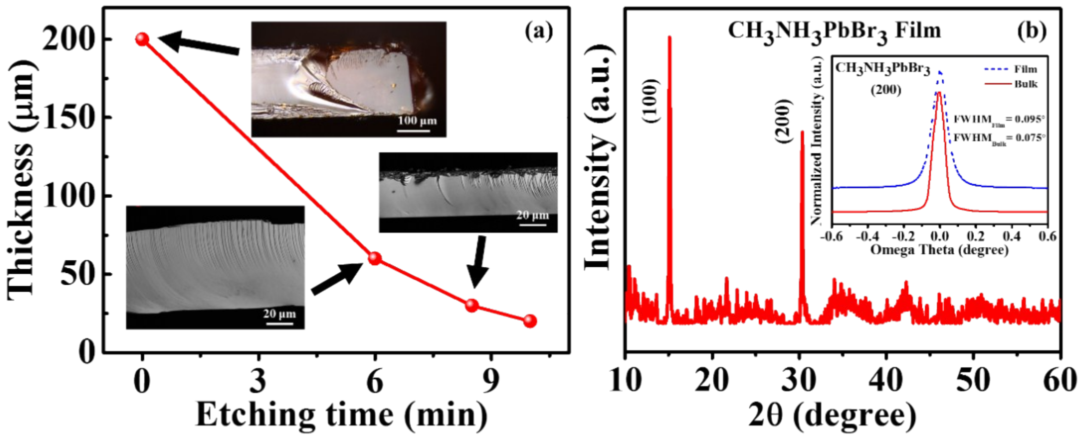

Fig. S6 (a) The relationship between $CH_3NH_3PbBr_3$ single crystal thickness and etching time. The inset is the corresponding CLSM observation of thickness evolution of $CH_3NH_3PbBr_3$ single-crystalline film. The point at 10 min etching time corresponds to Fig. 4c in the main text. (b) XRD pattern of a 20 um-thick (100)-oriented $CH_3NH_3PbBr_3$ single-crystalline thin film. The inset shows rocking curve of the (200) diffraction before and after thinning to evaluate its crystalline quality.