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Supporting information

## Understanding Temperature-Dependent Charge Transport, Structural Variation and Photoluminescent Properties in Methylammonium Halide Perovskite Single Crystals

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Figure S1. Evolution of the growth of  $MAPbI_3$  single crystals. The pictures in the bottom show the crystalizing samples soaked in fresh precursor solutions which are changed every 12 h for totally three times. Also shown in the right is the photo of finished MSC with a size up to 6 mm in diagonal and 3 mm in thickness.



**Figure S2.** (a) Snapshot of powder XRD patterns for MSC measured at different temperature (T). (b) Zoomed-in XRD pattern at 300 K.



**Figure S3.** *I-V* characteristics of MSC HO devices scanned in both upward and downward directions at (a) 295K, and (b) 110K.



**Figure S4.** (a) *I-V* characteristics (double-logarithm scale) of MSC HO devices scanned in both upward and downward directions at different *T*. (b) Compared *I-V* characteristics of initial scan at RT and re-scan after *T*-cycing at 300 K.



**Figure S5.** *I-V* characteristics (upward scan) of MSC HO devices in scanned with low biases (up to 10 V) in double-logarithm scale. Also compared is the *I-V* curve at 295 K after the cooling to hearing cycle (solid purple line) which overlaps with the initial measurement, indicating a *T*- reproducibility. Lines in red and pink are guidance to the eye with a slope of 1 and 2, respectively.



**Figure S6.** (a) *I-V* characteristics (double-logarithm scale) of electron-only (EO) devices based on MSCs with hole blocking interlayers scanned in both upward and downward directions. (b) Comparison of *I-V* curves scanned initially at 300 K and re-scanned at the same temperature after the cooling-to-heating cycle.



**Figure S7.** High resolution X-ray photoelectron spectra of (a) I3d and (b) Pb 4f measured on the surface of MSCs.



**Figure S8.** *I-V* characteristics in double-logarithm scale of MSC planar diodes in both upward and downward scan directions.