**Supplementary Information** for

# Grain Boundary Dominated Current Hysteresis and Ion Migration in Polycrystalline Perovskite Solar Cells

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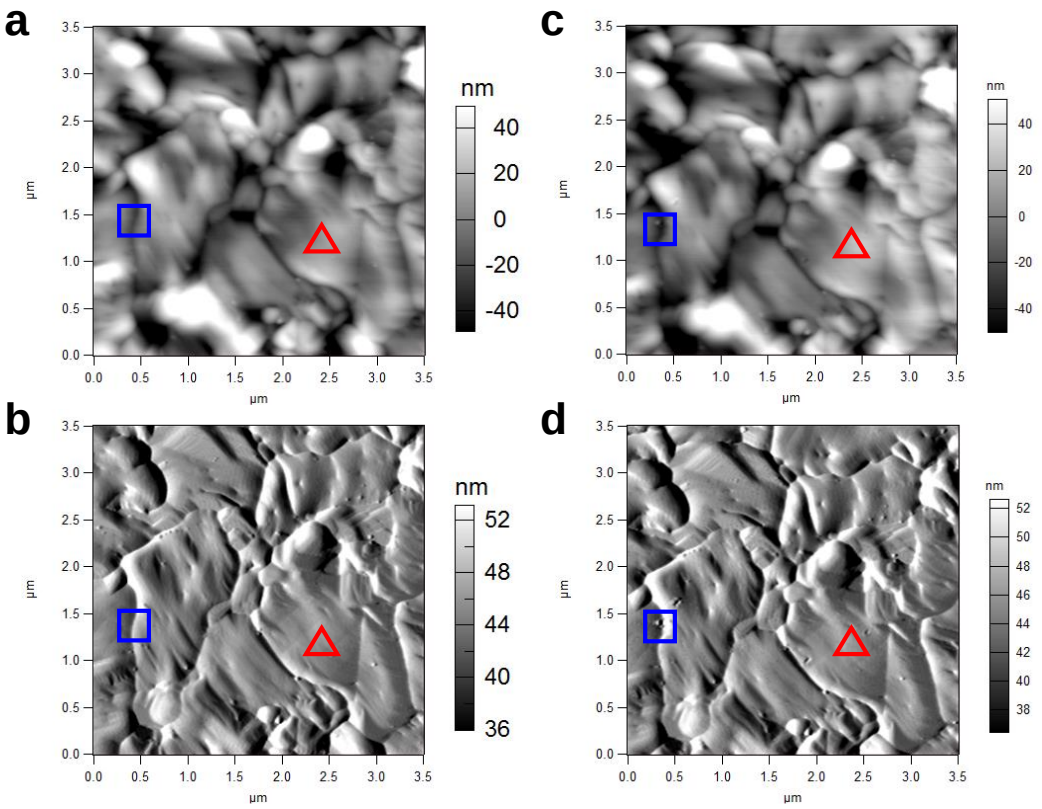
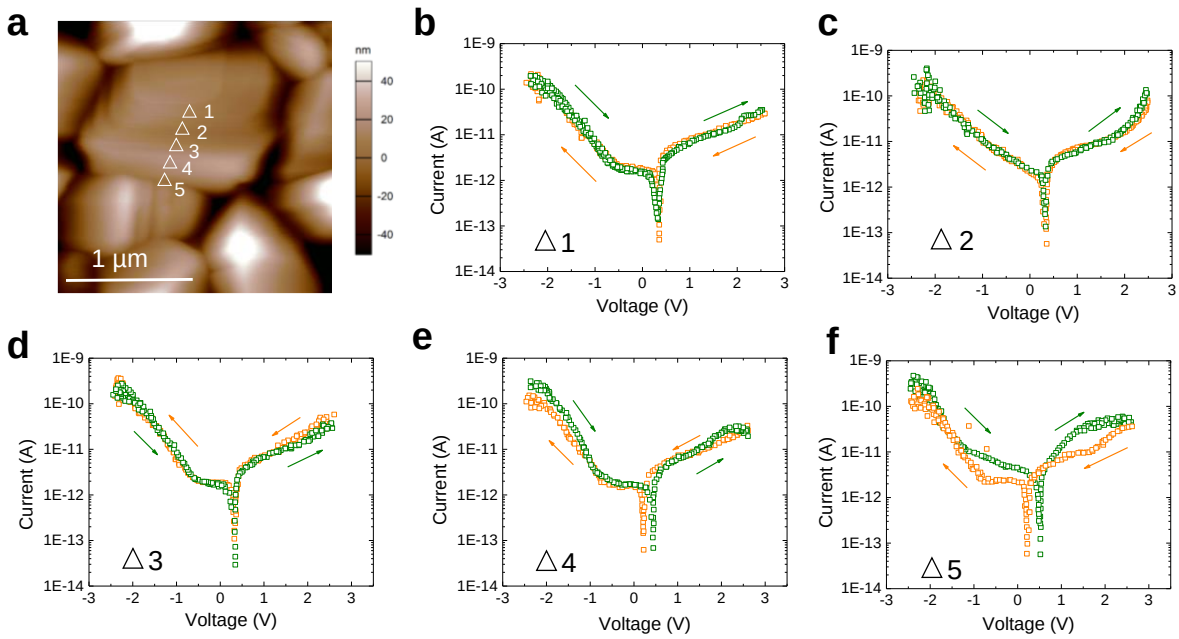
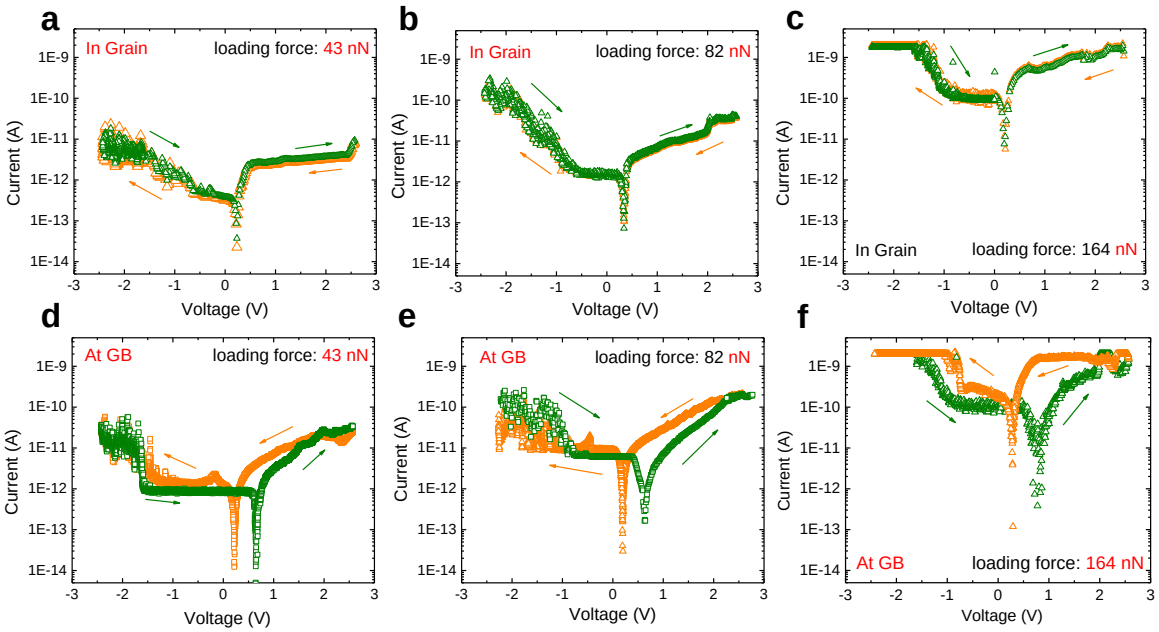



**Supplementary Figure 1**: (a) Height and (b) deflection images of the perovskite sample before the *I-V* measurement. (c) Height and (d) deflection images of the same area after the *I-V* measurement. The blue squares and red triangles indicate where the *I-V* measurements were performed at grain boundary and grain interior, respectively.

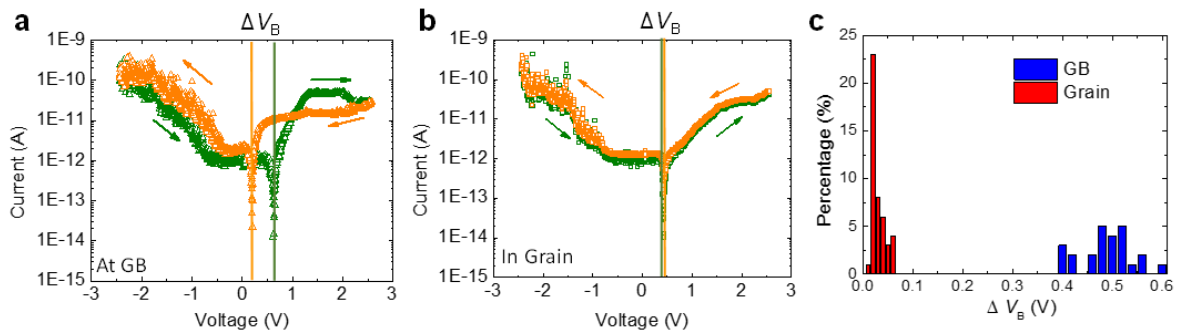



**Supplementary Figure 2:** (a) Topography AFM image of the perovskite thin film. Five locations where c-AFM tip was placed at to measure dark current are labeled with white triangles. (b)-(f) Dark curves measured at various points with an interval of ~100 nm in the direction from the center of a grain to the grain boundary (Δ1- Δ5 points as shown in the Supplementary Figure 2 (a)).

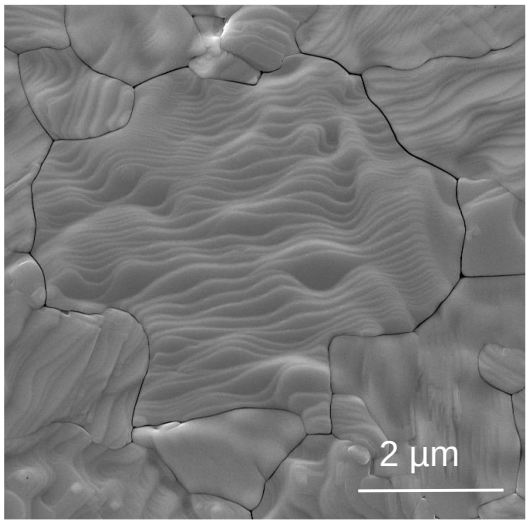
The results show that the dark-current hysteresis only appeared when the c-AFM tip placed with a range about 100 nm (Δ4 point) from the grain boundary for the 500-nm-thick perovskite film. This support our scenario that grain boundaries dominates the ion migration.



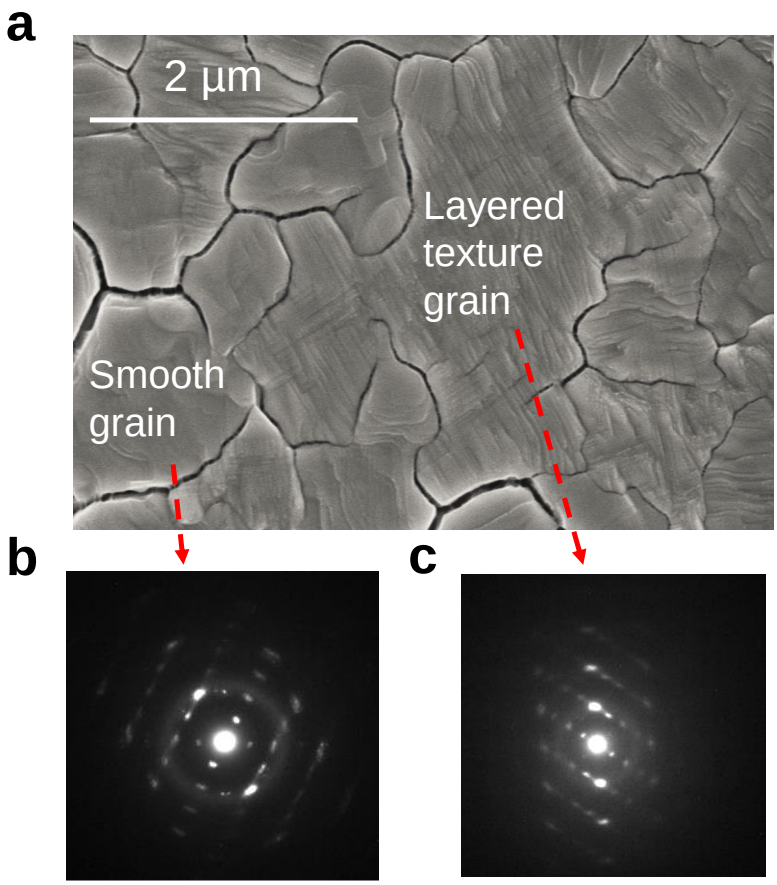

**Supplementary Figure 3:** Local dark-current measured on a grain with applying 43 nN (a), 82 nN (b) and 164 nN (c) loading force on the c-AFM tip, respectively. Local dark-current measured at a grain boundary (GB) with applying 43 nN (d), 82 nN (e) and 164 nN (f) loading force on the c-AFM tip, respectively.



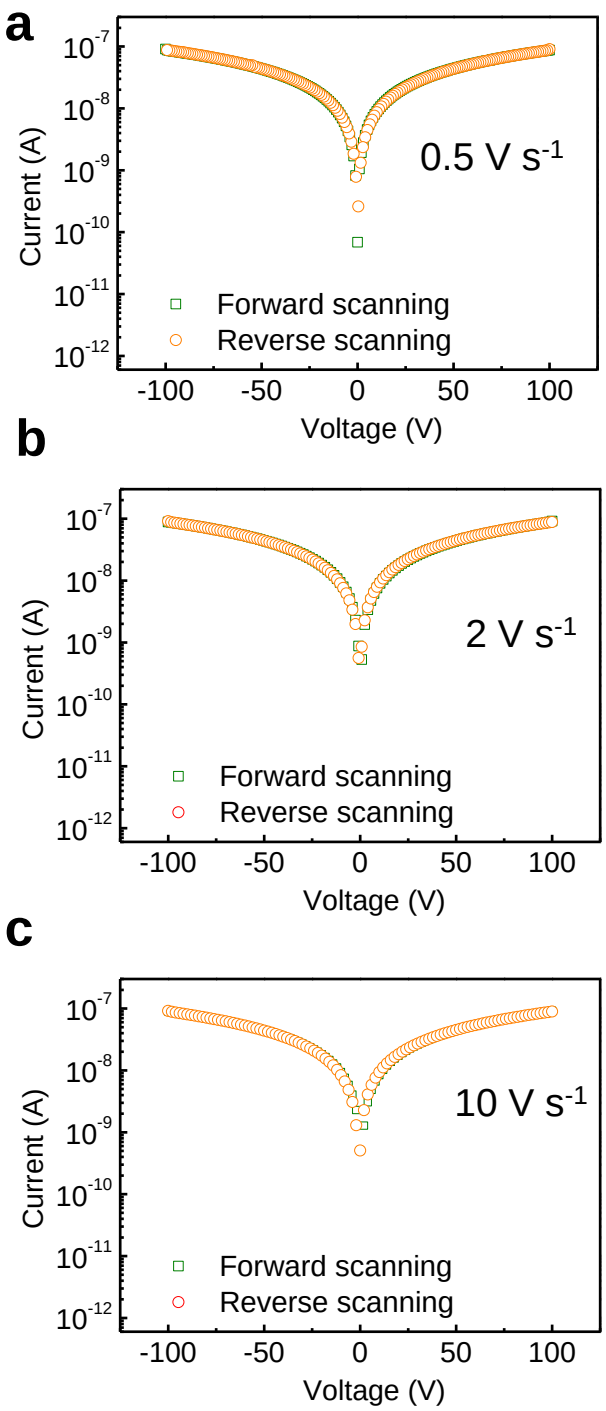

**Supplementary Figure 4:** (a) Dark-current measured at GB showed a large built-in potential difference ($\Delta V_B$) with reverse and forward scanning, as a result of fast ion migration. (b) Dark-current measured in a grain showed negligible built-in potential difference ($\Delta V_B$) with reverse and forward scanning, as a result of slow ion migration. (c) Statistical results demonstrate that the $\Delta V_B$ difference between GBs and grains is well reproducible.




**Supplementary Figure 5: Grains with textured structure in $CH_3NH_3PbI_3$ thin film**.




**Supplementary Figure 6:** (a) A SEM image shows grains with different topography. (b) Diffraction pattern of a smooth grain. (c) Diffraction pattern of a textured grain.




**Supplementary Figure 7: No dark current hysteresis was observed in the $CH_3NH_3PbBr_3$ single-crystal device regardless of the scanning rate:** (a) 0.5 V $s^{-1}$, (b) 2 V $s^{-1}$, (c) 10 V $s^{-1}$.