Electronic Supplementary Material (ESI) for Nanoscale.

Artificial Intelligence Atomic Force Microscope Enabled by Machine Learning

Supplementary Materials

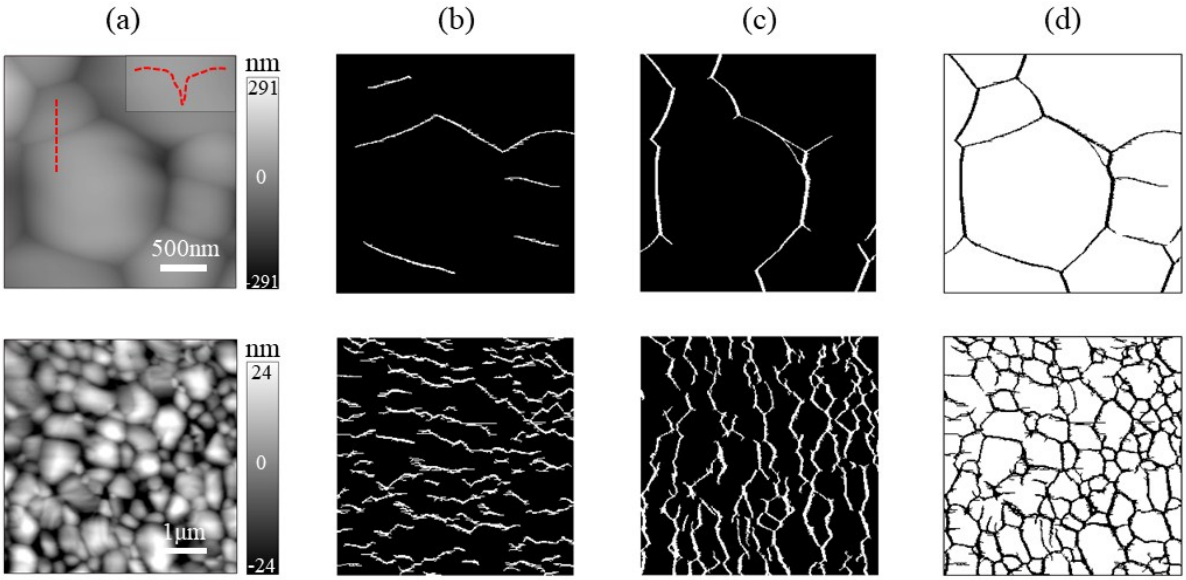

Fig. S1 GBs of Ceria (1st row) and $CH_3NH_3PbI_3$ (2nd row) identified by the second-order derivative method. (a) Topography and a height profile as an upright inset; (b) second-order derivative along y direction; (c) second-order derivative along x direction; (d) GBs generated from the union of (b-c).

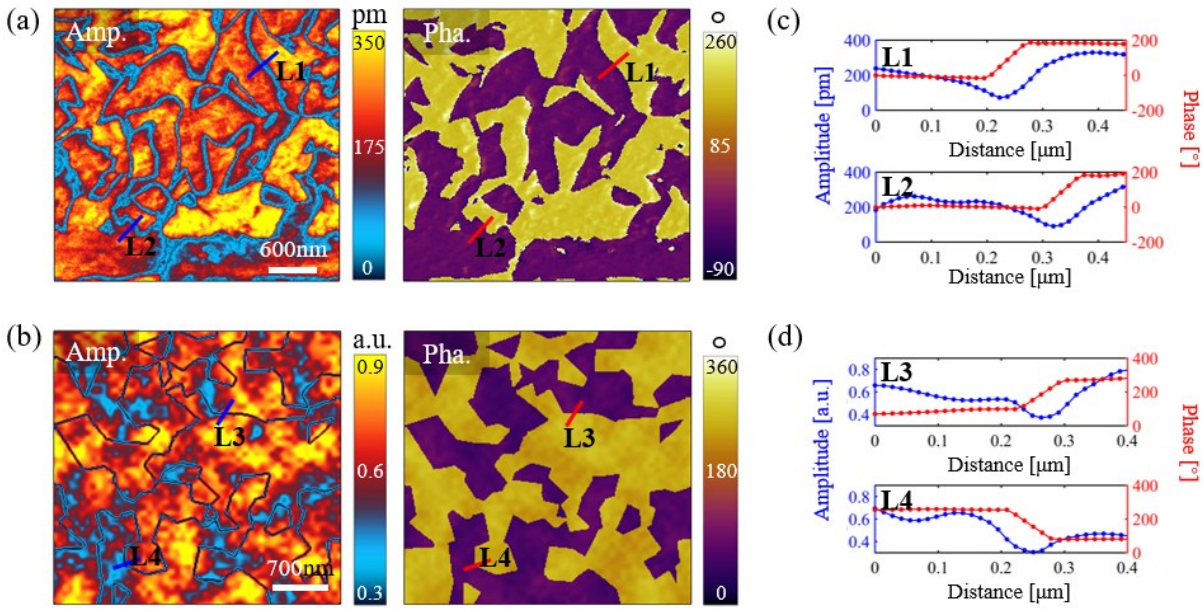

Fig. S2 (a) Real PFM maps of a PMN-PT sample; (b) simulated PFM maps of a FE sample. Corresponding amplitude and phase scans of: (c) lines L1 and L2, (d) lines L3 and L4.

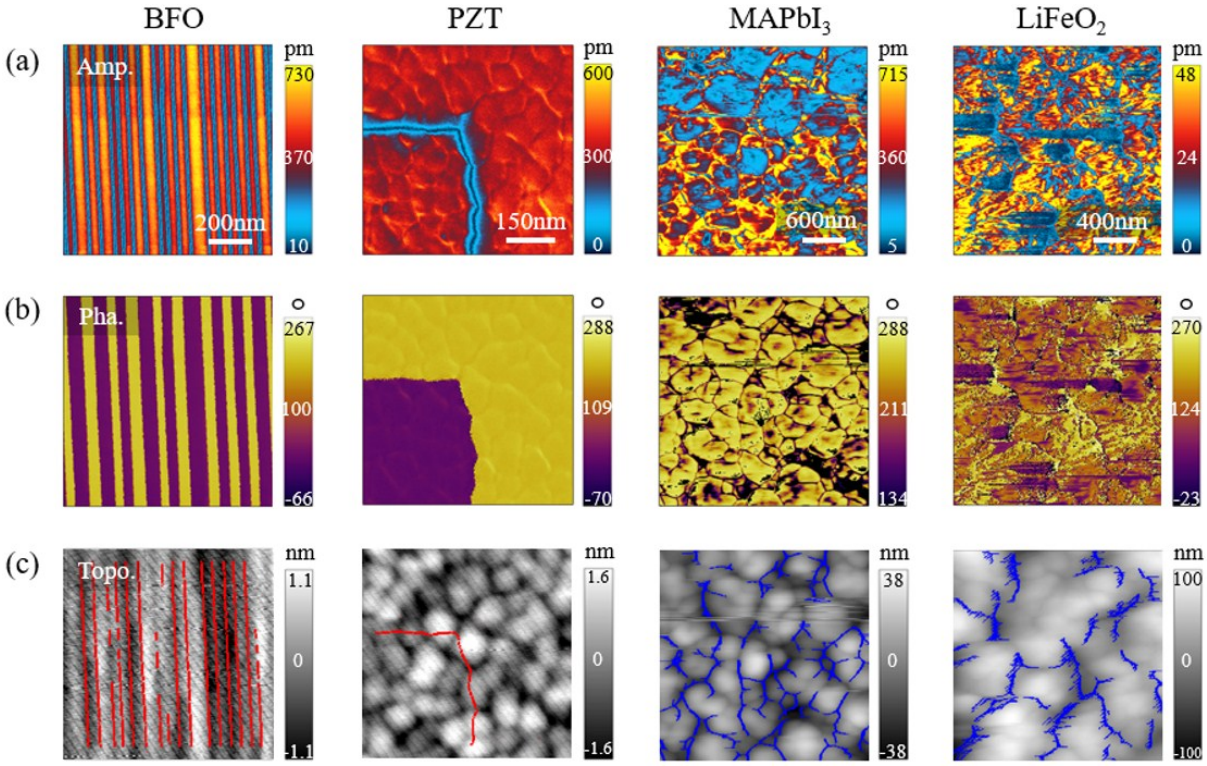

Fig. S3 Other typical ferroelectric and electrochemical mappings determined by the AI.

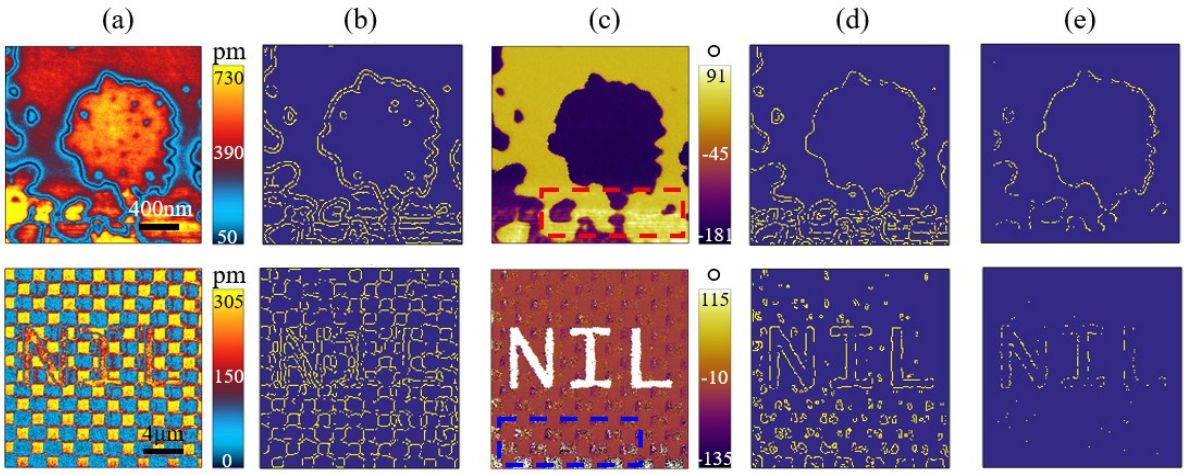

Fig. S4 Failure of edge-detector algorithm. Comparison between the DWs identified by Canny edge detector and AI algorithms. Real PFM maps are columns (a) Amplitude and (c) Phase, the corresponding edges detected by Canny are shown in columns (b) and (d), separately; (e) DWs identified by AI algorithms.

Even though the detected DWs sometimes could overlap the boundaries of phase maps as in **Fig. 3(b)** and first two columns of **Fig. S3**, it is not always the case. We compare the output of the AI model with a popular Canny edge detector in **Fig. S4**, where columns (b, d) are edges identified by the Canny algorithm from columns (a) Amplitude and (b) Phase maps, separately. Since edge detector are highly sensitive to the gradient of a map, slight phase distortion due to scanning disturbance (marked by red dash rectangular), impurities or artificial pattern (marked by blue dash rectangular) may cause redundant markers, which is not applicable for extracting DWs. In this regard, the DWs highlighted by the AI model will not suffer from such problems and outperform the edge detector, as shown in **Fig. S4(e)**.

Mov. S1 Movie of AI-AFM testing on a ferroelectric PMN-Pt sample unknown to AI-AFM in advance.

Mov. S2 Movie of AI-AFM testing on a non-ferroelectric Ceria sample unknown to AI-AFM in advance.