Supporting Information:

Misfit Relaxation Mechanisms and Domain Ordering in Anisotropically Strained Manganite Thin Films

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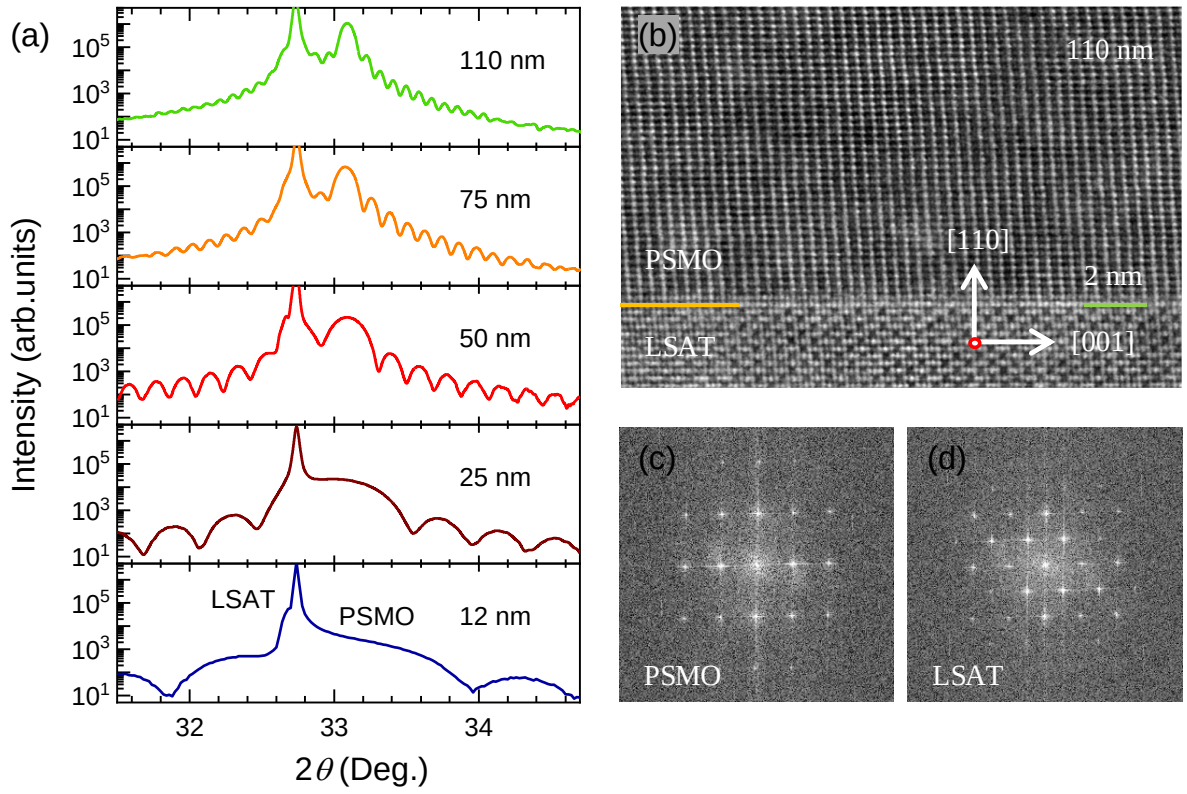

Figure S1 (a) XRD 2θ-ω linear scans measured from PSMO/LSAT(110) films with various thicknesses. (b) HAADF-STEM image viewed along the [1-10] direction of 110 nm PSMO/LSAT(110) film. (c,d) The fast Fourier transformation of PSMO and LSAT in (b), respectively.

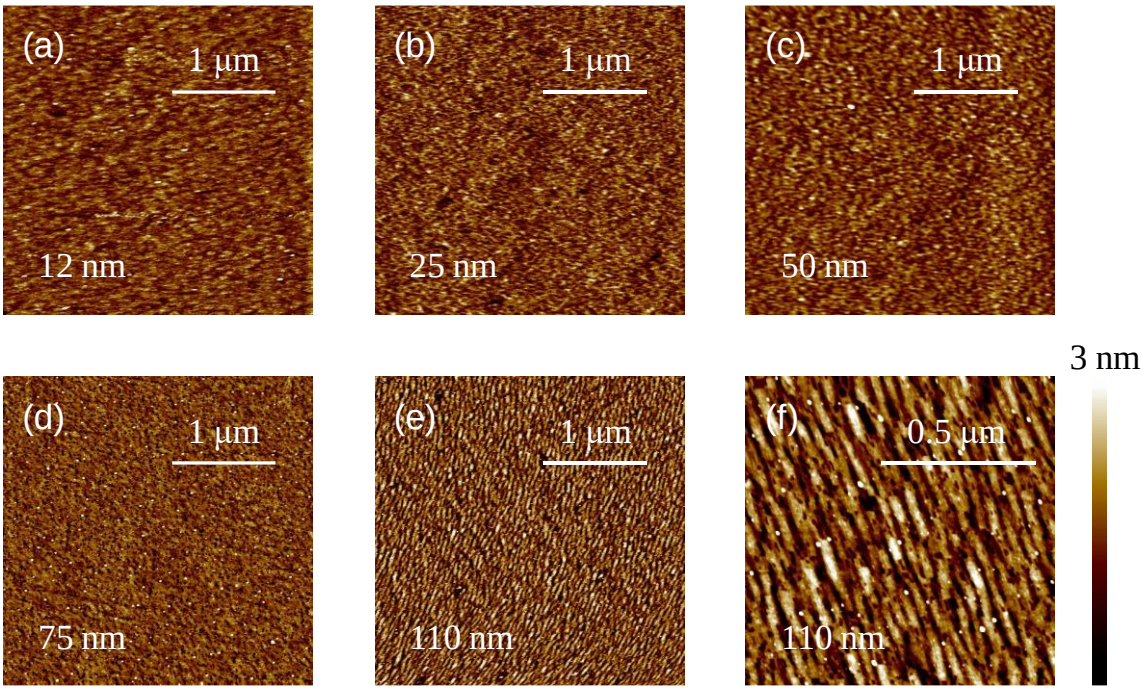

Figure S2 The surface morphologies measured from (a) 12 nm, (b) 25 nm, (c) 50 nm, (d) 75 nm, and (e,f) 110 nm PSMO/LSAT(110) films. The size of (a-e) [(f)] is 3 ×3 (1 ×1) μm^2.

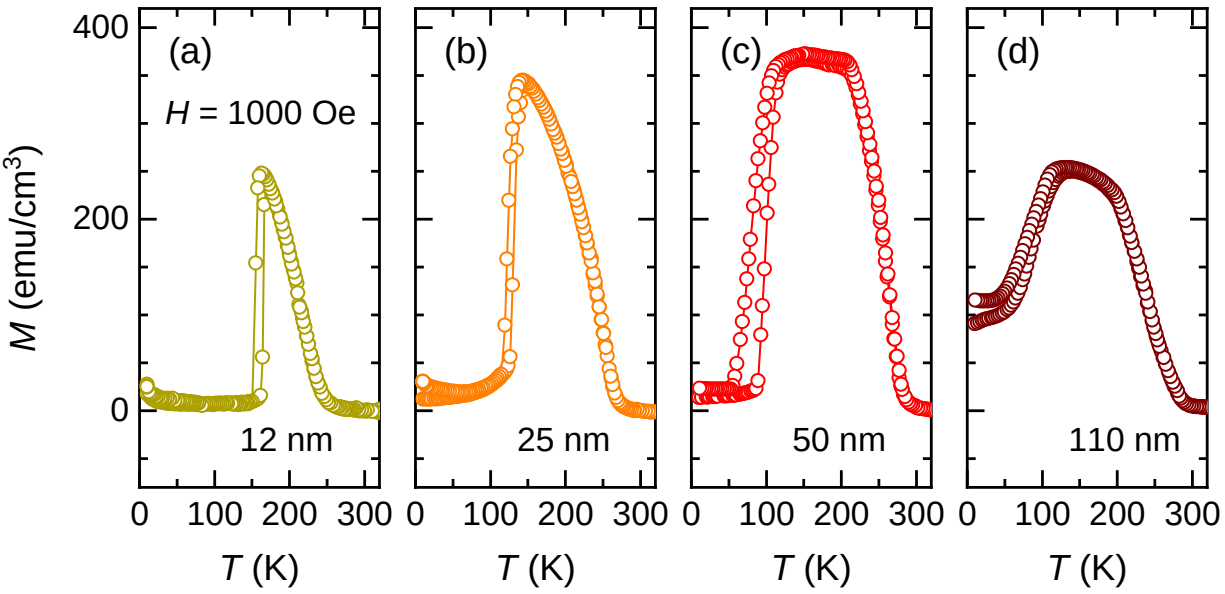

Figure S3 *M*-*T* curves measured from the (a) 12 nm, (b) 25 nm, (c) 50 nm, and (d) 110 nm PSMO/LSAT(110) films at 1000 Oe.

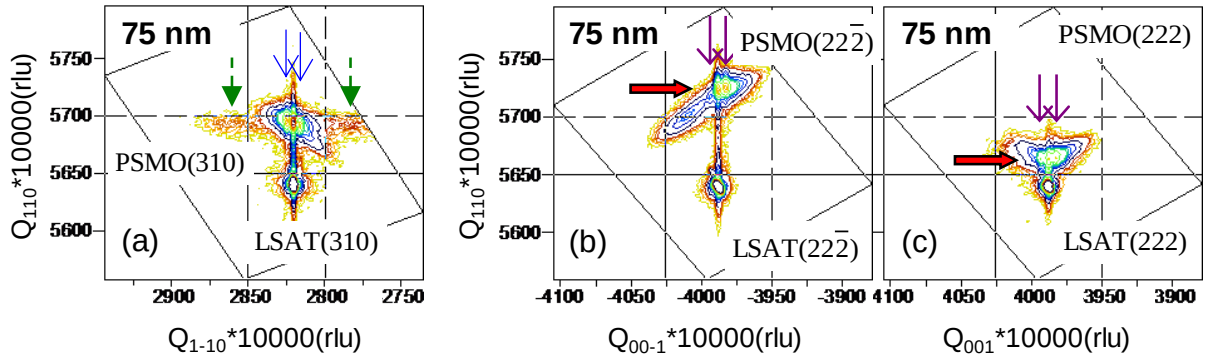

Figure S4 RSMs on the (a) (310), (b) (22-2), and (c) (222) reflections in the high-resolution scattering zone of the 75 nm PSMO/LSAT(110) film. The horizontal arrows indicate the positions of the main Bragg (22-2) or (222) reflection of PSMO films. The satellites are denoted by the downward arrows. The grid in each panel is just a guide to the eye.

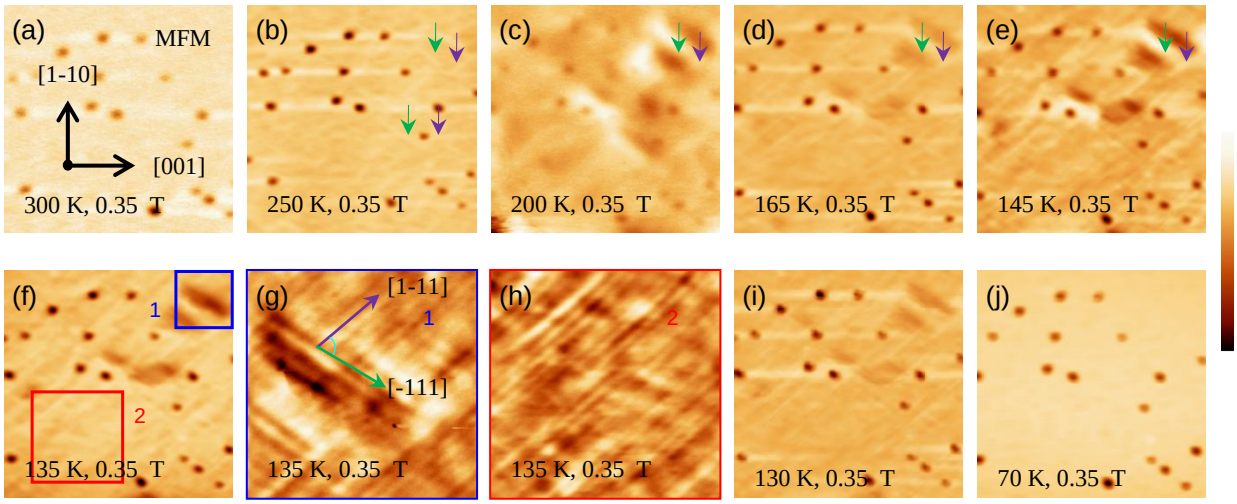

Figure S5 (a-j) MFM images taken at 300, 250, 200, 165, 145, 135, 135, 135, 130, and 70 K, respectively. A small field of 0.35 T is applied perpendicular to the film plane to enhance the magnetic signal. Images were taken at the same location. (g,h) Enlarged views of the square regions in (f), showing the orientation-ordered domain patterns. The sizes of (a-f,i,j), (g), and (h) images are 5×5, 1.25×1.25, and 2×2 μm^2, respectively. The color scales are 0.22, 1.49, 362, 2.62, 1.11, 1.78, 0.84, 0.62, 4.0, and 2.43 mHz for (a-j), respectively.

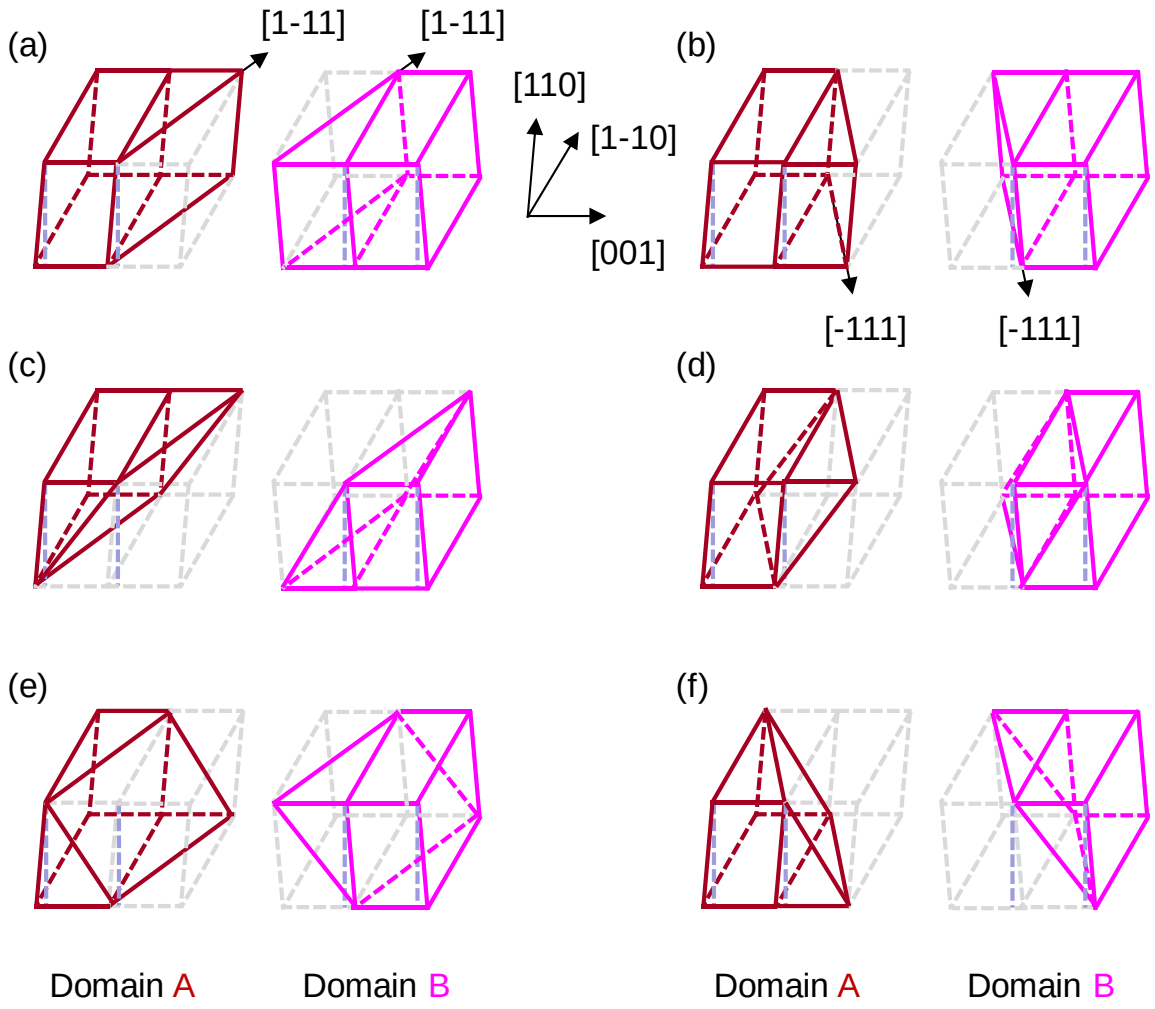

Figure S6 The schematic diagrams of compressive “head-to-head” domain walls formed between twin domains A and B. (a,c,e) The possible [1-11]-oriented domain walls. (b,d,f) The possible [-111]-oriented domain walls. The schematic diagrams of tensile “tail-to-tail” domain walls are similar to that of compressive “head-to-head” domain walls, and will not be shown.

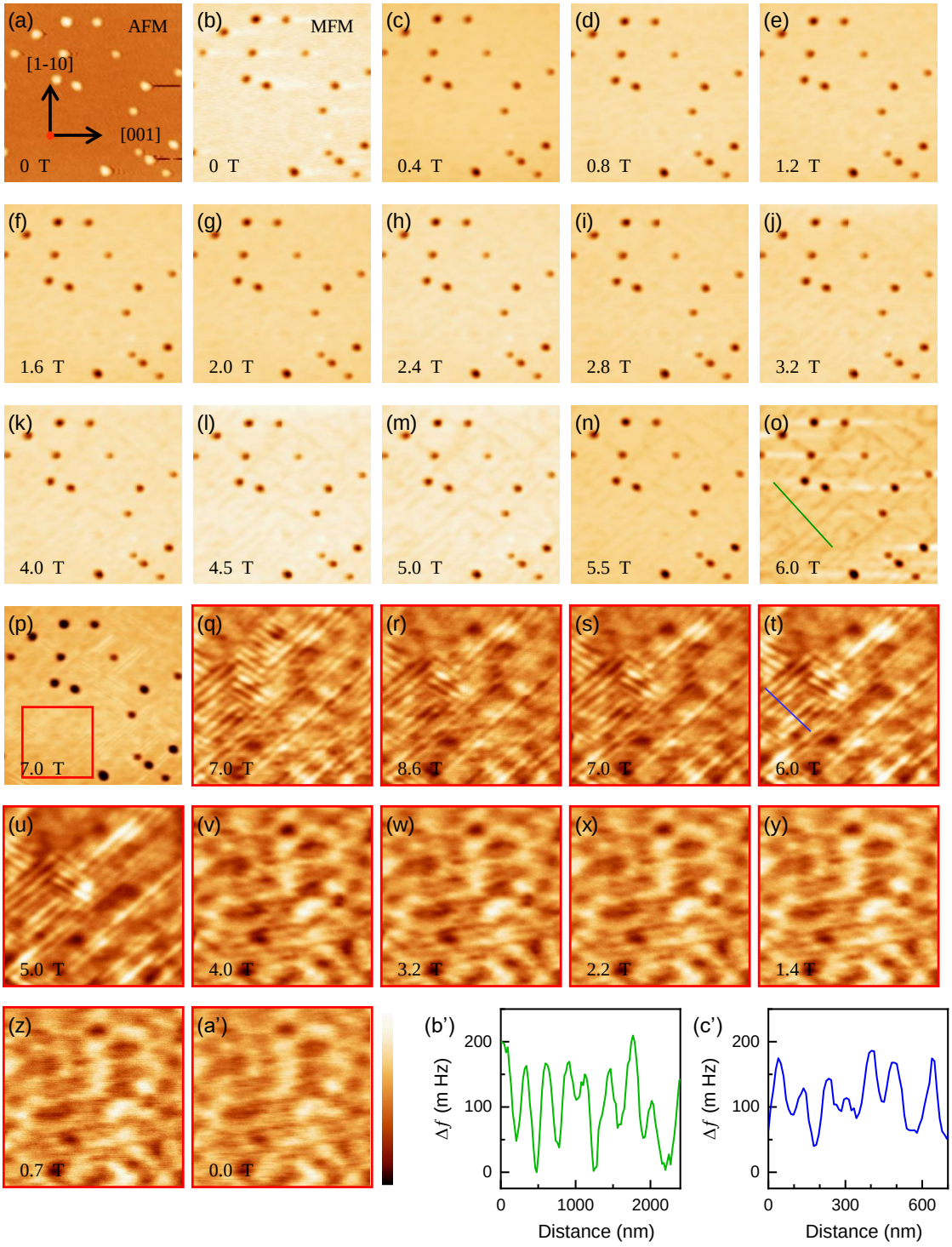

Figure S7 (a) The surface morphology of 50 nm PSMO film. (b-y) MFM images taken during field sweep at 50 K. Images of (a-p) were taken at the same location, and MFM images of (q-a') were taken at the same location marked as square red box in (p). The PSMO film is cooled from room temperature to 50K and then the magnetic field is applied perpendicular to the film plane. (b') [(c')] The profile taken along the green (blue) line in (o) [(t)].