

Supplementary Information

Controlled motion of vortex boundary in oxide heterostructures

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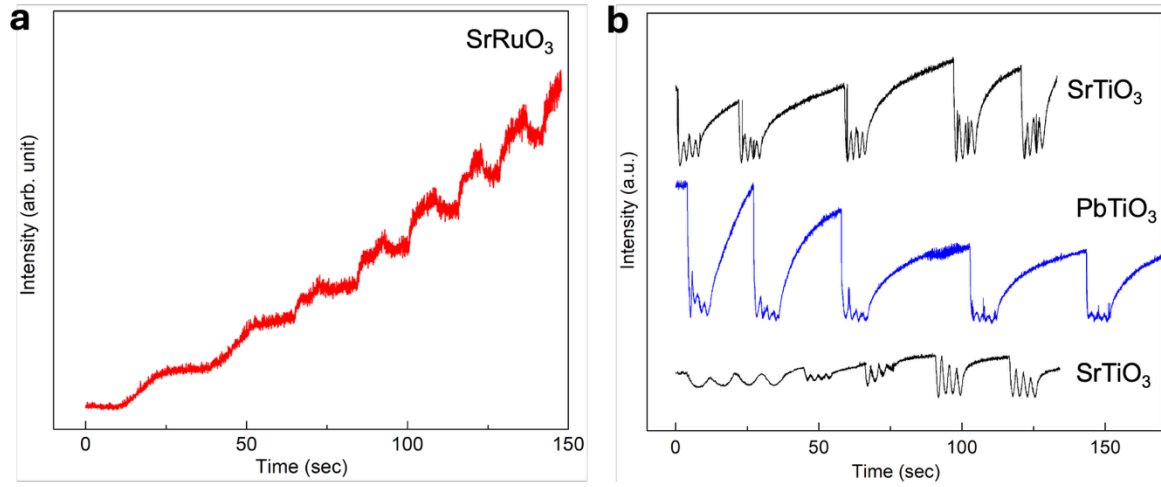
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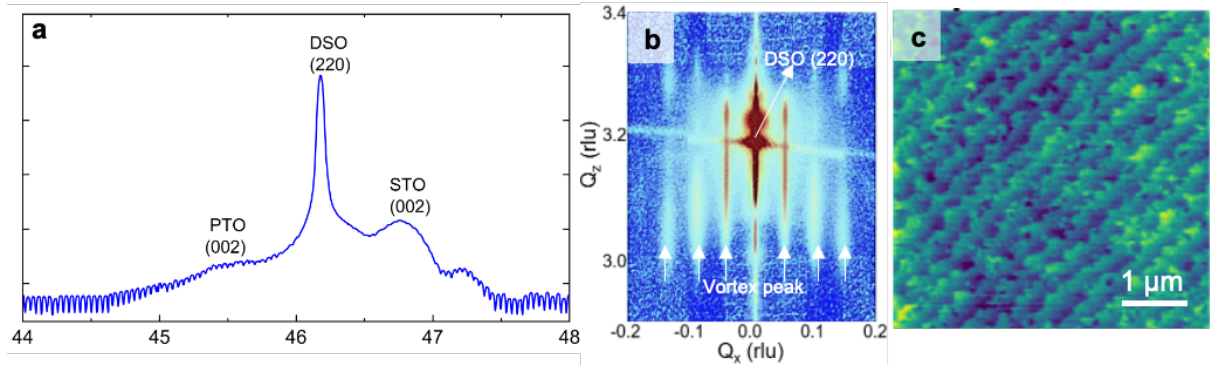
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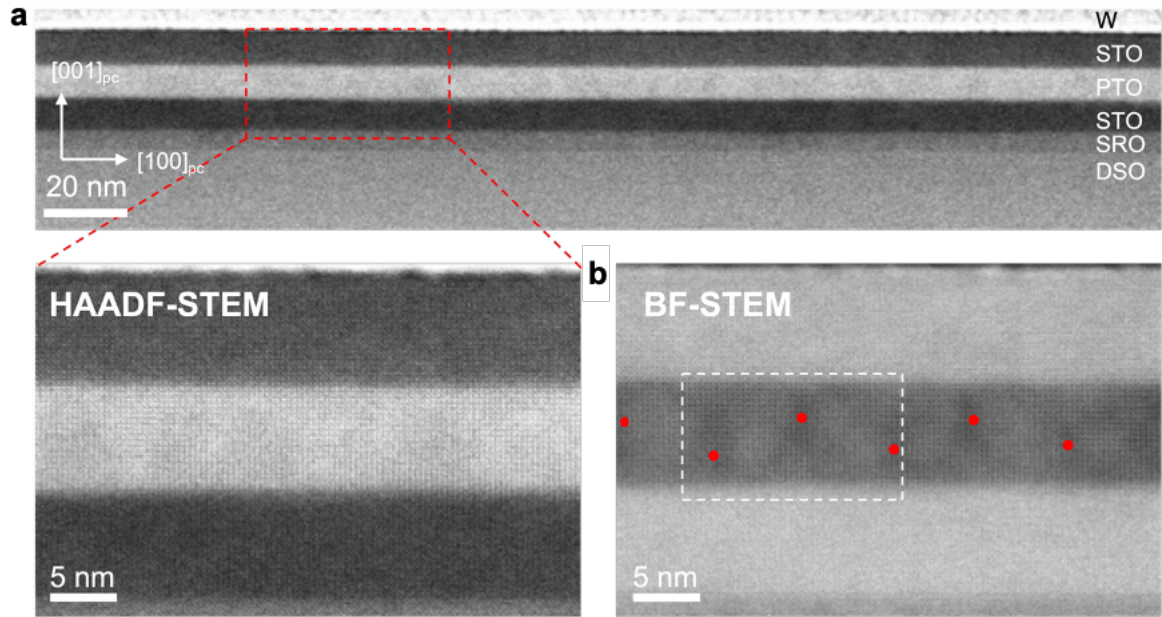
Supplementary Figures



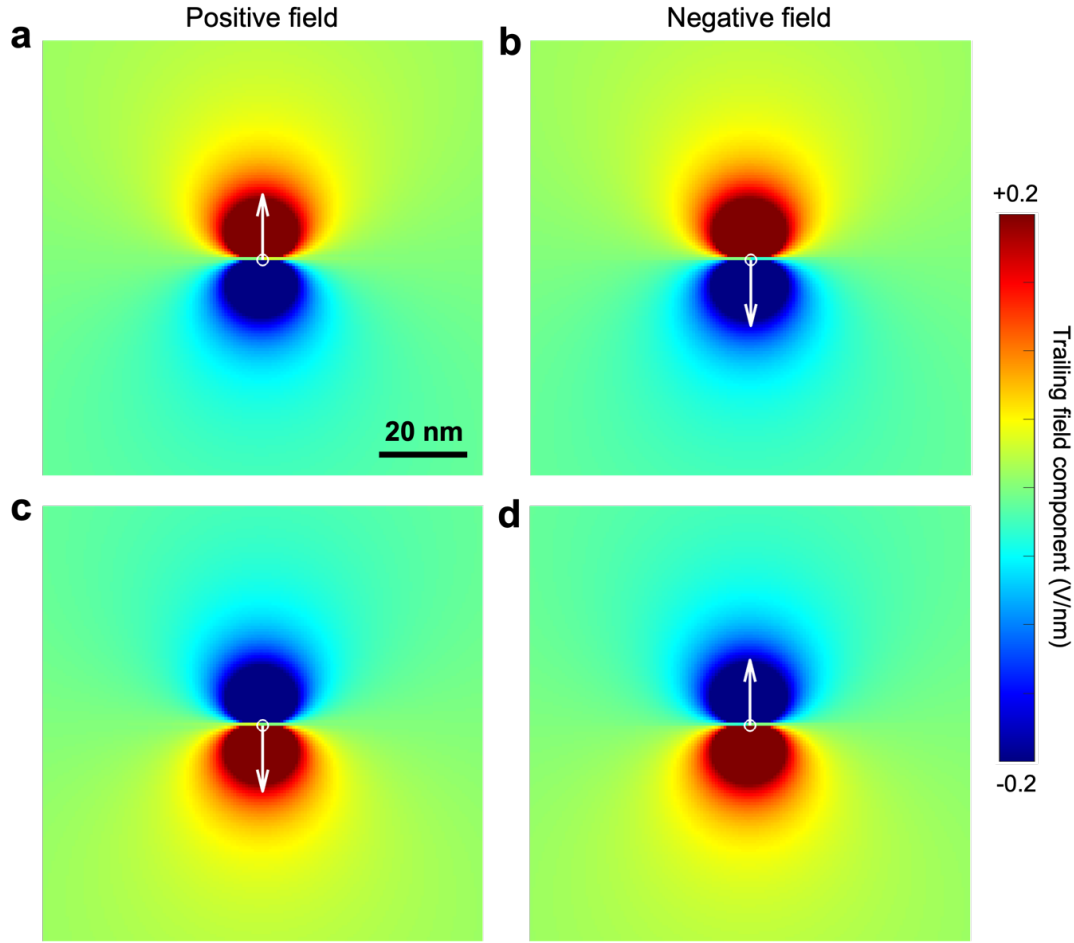
Supplementary Fig. 1| RHEED oscillations for the trilayer sample on $\text{DyScO}_3 (001)_{\text{pc}}$ substrates. **a, RHEED oscillations during the growth of bottom SRO layer in the trilayer. **b**, RHEED oscillation during the growth of STO, PTO and STO in the STO/PTO/STO trilayer samples. After the growth of every four-unit cell laser was pause for 5 second allows atoms to relax.**



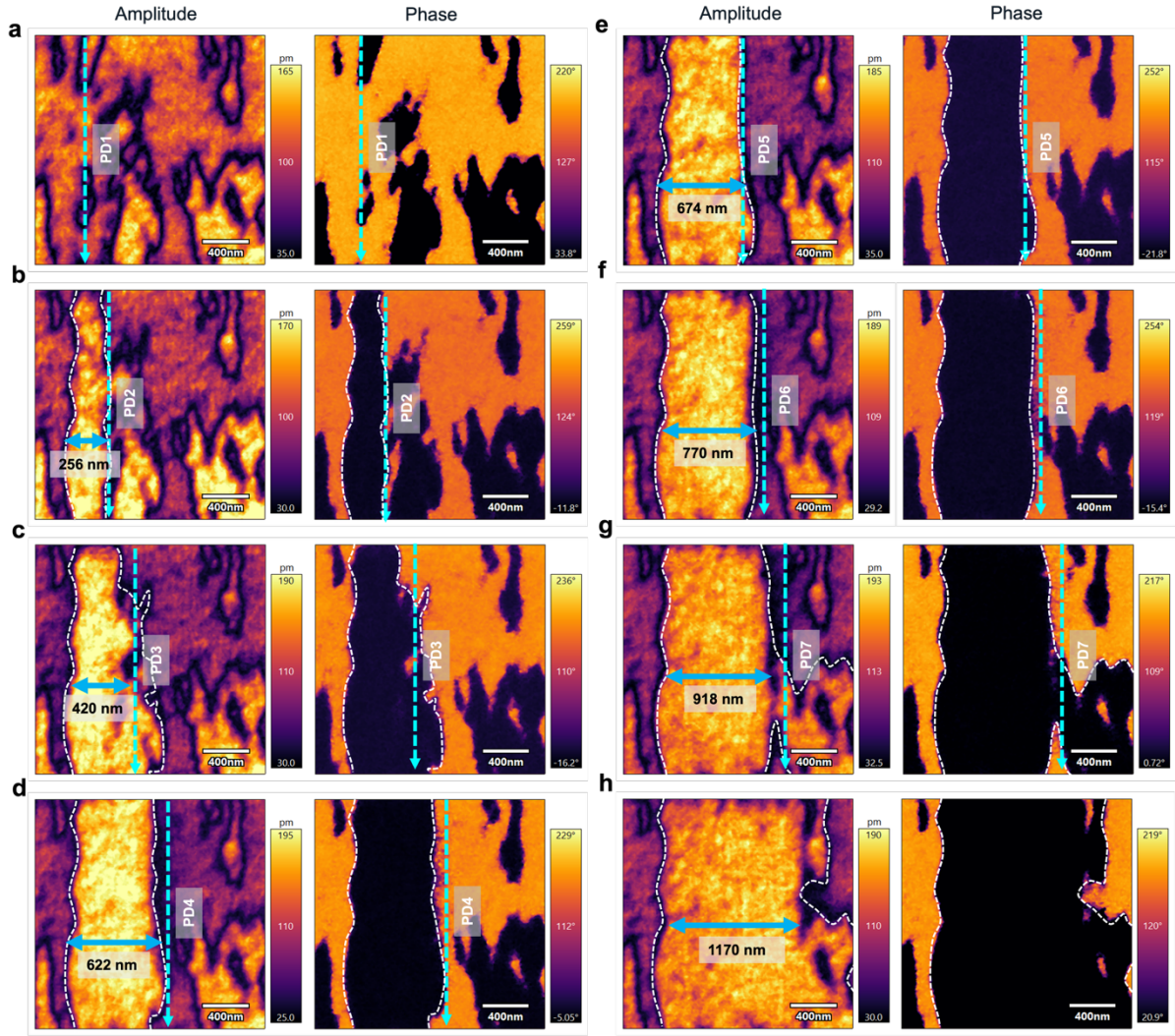
Supplementary Fig. 2| X-ray diffraction based structural characterization for trilayer. a, High-resolution θ - 2θ symmetric scan of a 20 uc STO/20 uc PTO/20 uc STO trilayer. **b,** Reciprocal space mapping for the trilayer around the $(002)_{\text{pc}}$ -reflection peak reveals the presence of pure vortex phase (satellite peaks along the in-plane directions) in the trilayer having the periodicity ~ 14 nm. **c,** Topography of the trilayer structure measured with atomic force microscopy (AFM). The presence of step heights confirm layer-by-layer growth and smooth surface.



Supplementary Fig. 3| STEM images of the 20 uc STO/20 uc PTO/20 uc STO on DyScO₃ (001)_{pc} substrate. a, Cross-sectional STEM image of the STO/PTO/STO trilayer structure, zoomed part of the image is shown in the bottom panel, indicating the clear interface. **b,** The BF-STEM image of the trilayer, the contrast in the PTO layer is relevant to polar structures, which can be used to identify the vortices.

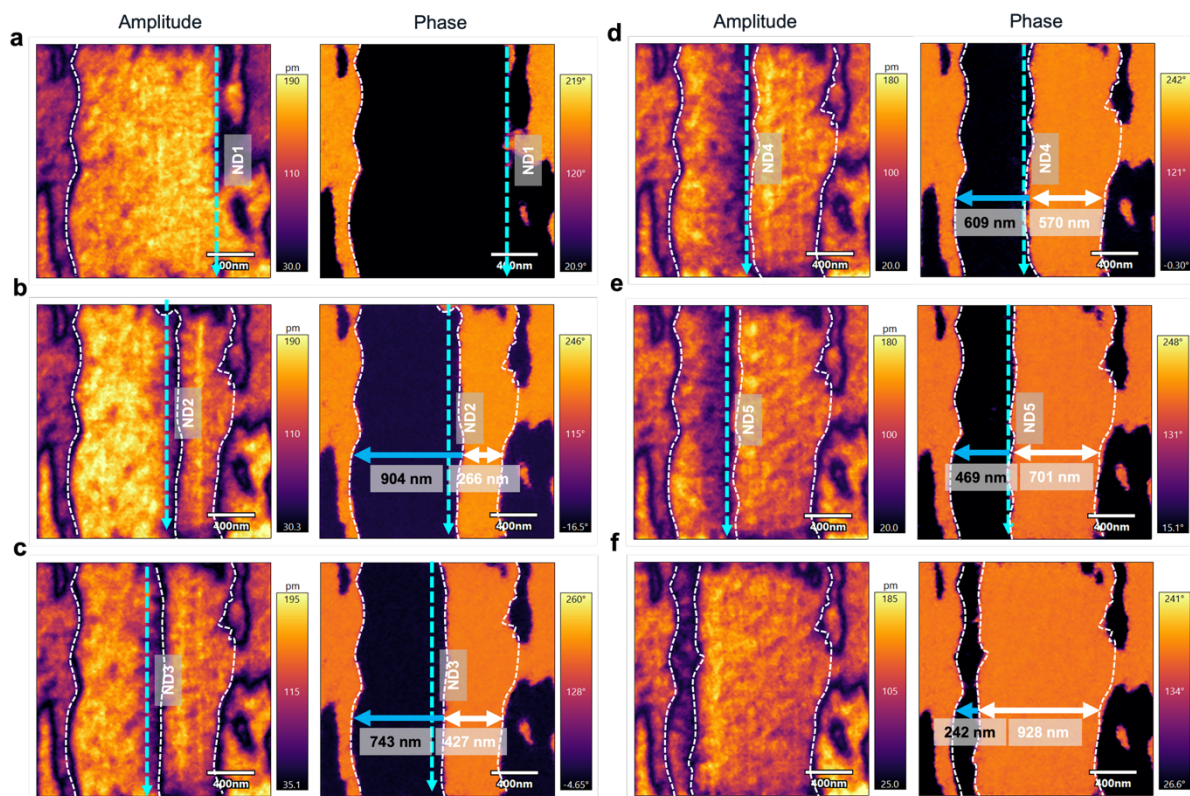


Supplementary Fig. 4| Finite element modelling (FEM) of trailing electric field distributions generated by a moving AFM probe. a-b, Calculated trailing field profiles as an AFM probe moves vertically (a: up, b: down) with applied electric bias of +5 V (a) or -5 V (b), respectively. The field distribution couples to the vortex in-plane polarization and can drive the motion of polar vortices. Notably, the trailing field for 5 V upward motion (a) mirrors that of -5 V downward motion. **c-d,** Trailing field distributions for reversed probe scanning directions +5 V bias with downward motion (c), -5 V bias with upward motion (d). These panels highlight that changing either the probe motion direction or the voltage polarity results in a distinct change in the trailing field distribution. This FEM demonstrates that the trailing field can be manipulated by tuning the probe direction or voltage polarity, consistent with our bias and probe motion dependent experimental observations.

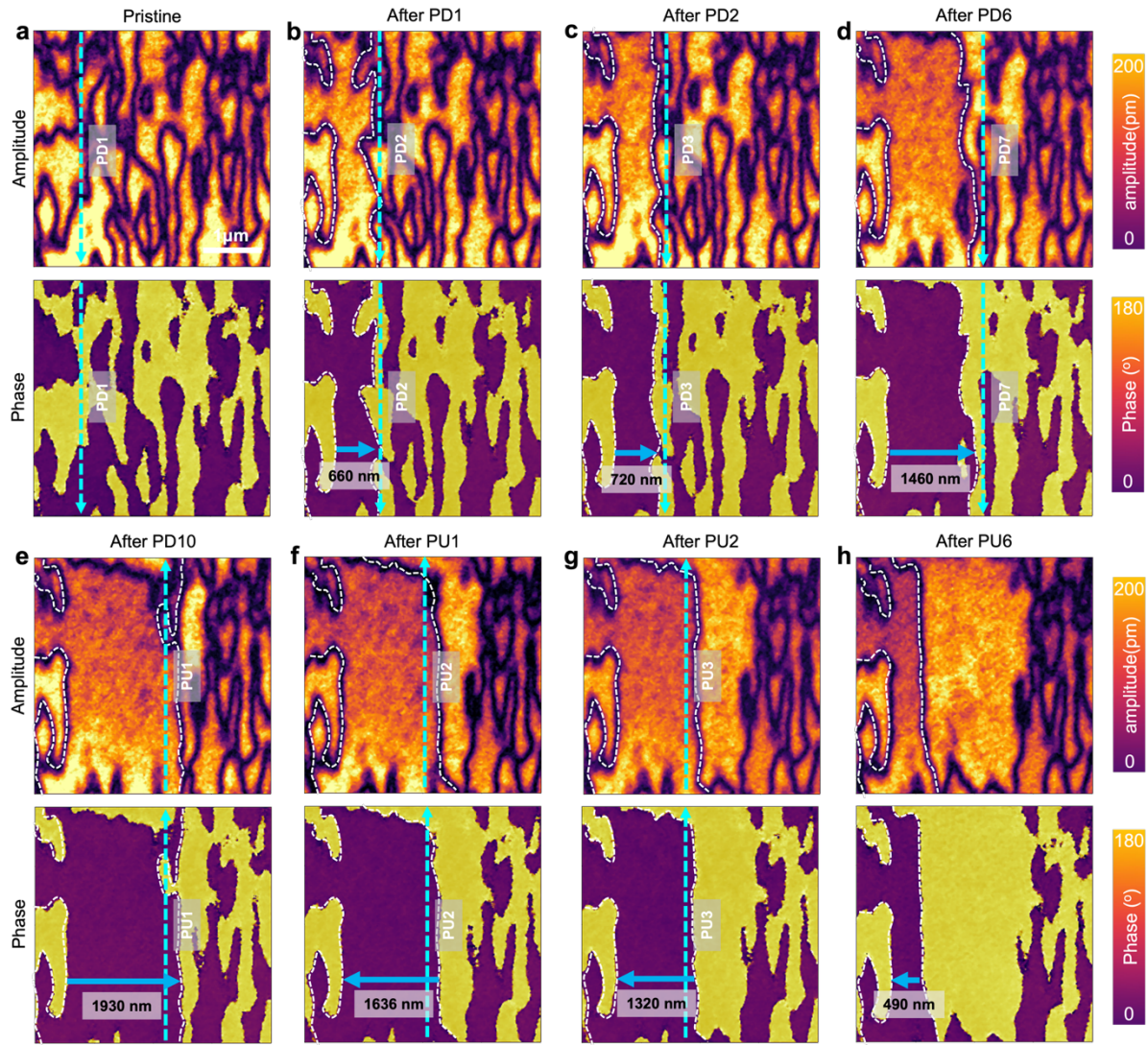


Supplementary Fig. 5| Deterministic motion of the vortex boundary in the trilayer under positive trailing field. **a**, Pristine LPFM scans (amplitude, left; phase, right) reveal the presence of well-defined pure vortex structures in the trilayer. The cyan arrow denotes the path along which the first positive trailing field (PD1) was applied for one minute. The PFM scans were acquired after the removal of the field. **b**, Post-field removal PFM imaging following the first trailing field (PD1) indicates the initial rightward displacement of the vortex boundary (marked by the blue arrow). A second positive trailing field (+5 V) was then applied for one minute along a subsequent path (PD2). **c–h**, Sequential PFM scans illustrate the progressive rightward movement of the vortex boundary, observed after each trailing field application. Following the removal of PD3 (c), a further shift to the right is evident. This pattern persists with the application of a third trailing field (+5 V) along path PD3 (d), a fourth field (+5 V) along PD4 (e), and continues through subsequent trailing field applications (PD5–PD7). This cumulative effect results in a pronounced vortex boundary shift of ~1170 nm after the seventh

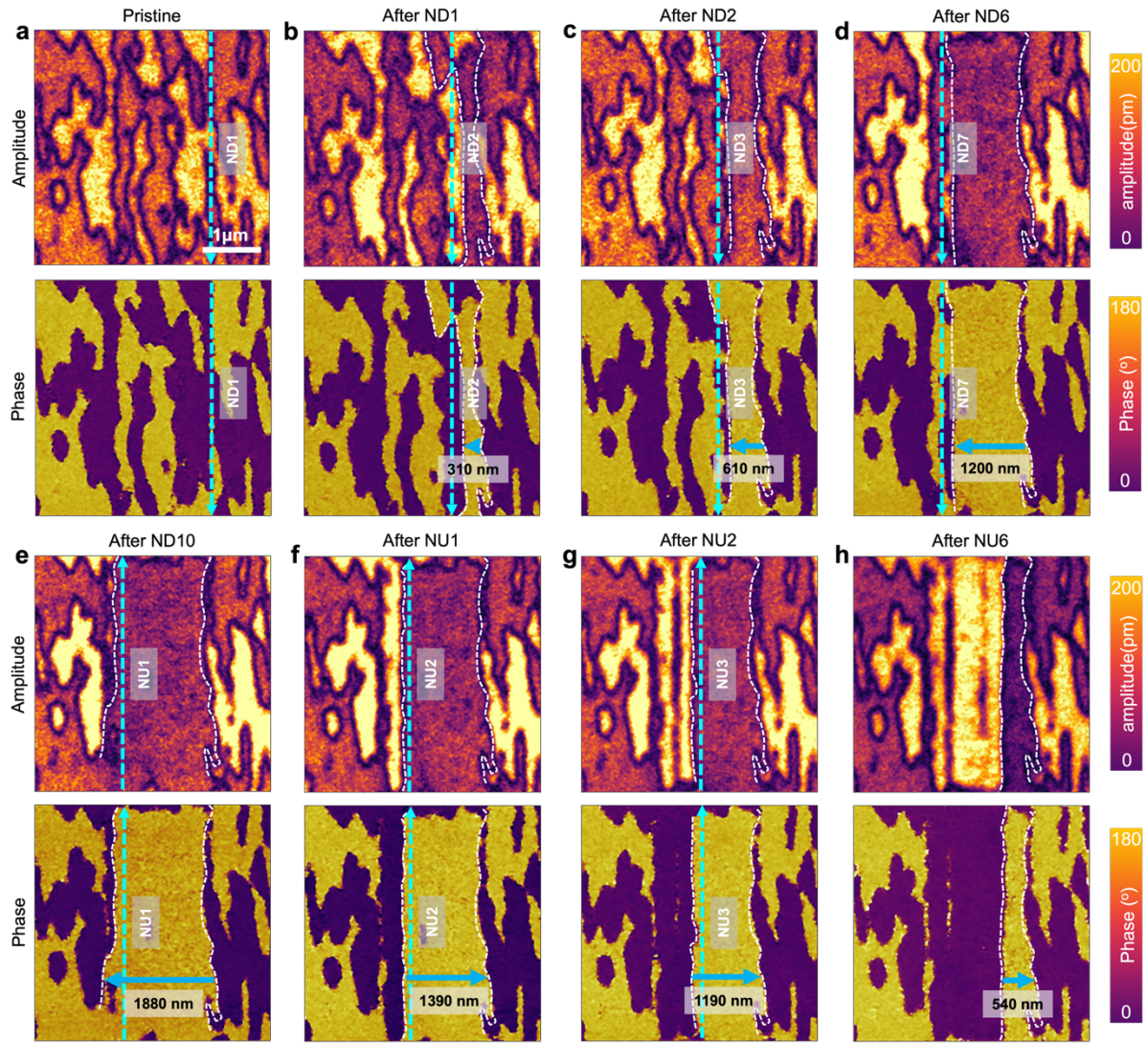
trailing field application from left to right side, culminating in a significant realignment of the vortex structure towards the right side (h).



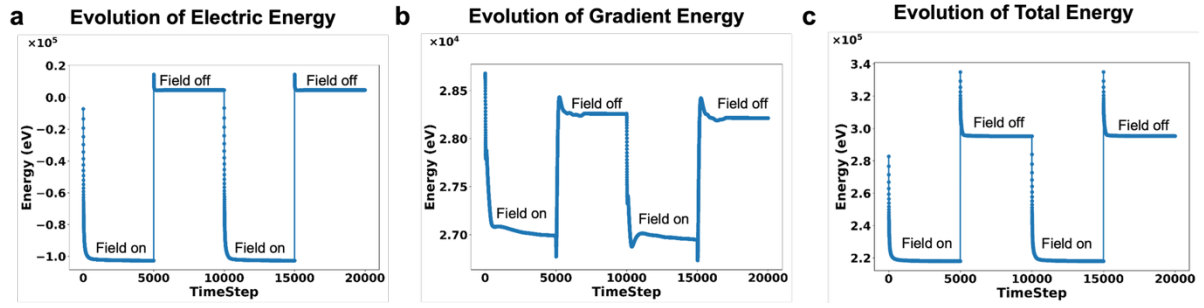
Supplementary Fig. 6 | Deterministic motion of the vortex boundary in the trilayer under negative trailing field. **a**, The image from Supplementary Fig. 4h serves as a reference point. The cyan arrow indicates the path along which the first negative trailing field (CHN1) (-5 V, 50 nm/s) was applied. PFM measurements were conducted immediately after the field was removed to capture the initial state of the vortex structure. **b**, After the removal of ND1, the PFM scans reveal a distinct leftward shift in the vortex boundary, marked by the blue arrow. This displacement originates from the right side, moving towards the left. A second negative trailing field (-5 V) was then applied along the same path (ND1) with 50 nm/s. **c–f**, This trend of leftward vortex boundary movement continues as further trailing fields are applied. The third trailing field (-5 V) along path ND3 in (c) results in an additional leftward shift, which is observed in panel (d). Similarly, the fourth trailing field (-5 V) along path ND4 in (d) leads to another leftward boundary shift (panel e). This pattern culminates with the fifth trailing field (-5 V) applied along path ND5 (e), driving the vortex boundary further to the left, as evidenced in panel (f). The magnitude of the boundary shift is noted in each respective panel. After six successive applications of negative trailing fields, the vortex boundary exhibits a significant leftward displacement of ~928 nm. This cumulative effect nearly returns the vortex boundary towards original position, indicating a consistent and measurable response to the applied negative fields.



Supplementary Fig. 7| Directional control of the vortex boundary motion in the trilayer with positive field. **a**, Shows the pristine vortex state. The cyan arrow indicates the path along which the AFM probe is moved (PD: tip motion up to down with positive polarity of bias). **b**, After the removal of PD1, the PFM scans reveal a distinct rightward shift in the vortex boundary, marked by the blue arrow. **c-e**, Shows the vortex movement after PD2-PD10 with total movement of ~ 1930 nm. **f-h**, The AFM probe motion direction was changed from down to up (PU) keeping polarity same (PU1-6). With changing the direction of probe motion keeping the field polarity same the vortex can be moved reversibly i.e. leftward shift.



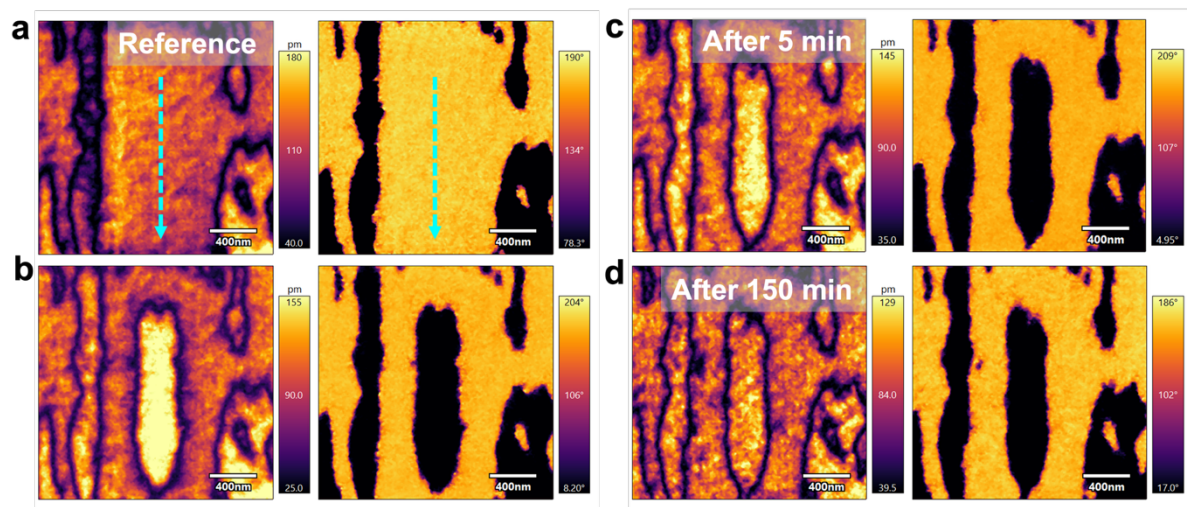
Supplementary Fig. 8| Directional control of the vortex boundary motion in the trilayer with negative field. **a**, Shows the pristine vortex state. The cyan arrow indicates the path along which the AFM probe is moved (ND: tip motion up to down with negative polarity of bias). **b**, After the removal of ND1, the PFM scans reveal a distinct leftward shift in the vortex boundary, marked by the blue arrow. **c-e**, Shows the vortex boundary movement after ND2-ND10 with total movement of ~ 1880 nm. **f-h**, The AFM probe motion direction was changed from down to up (NU) keeping polarity same (NU1-6). With changing the direction of probe motion keeping the field polarity same the vortex boundary can be moved reversibly i.e. rightward shift.



Supplementary Fig. 9| Evolution of electric, gradient and total energies corresponding to the vortex boundary motion. **a**, Time-dependent electric energy during two bias cycles (5 V tip field). **b-c**, Corresponding gradient and total-energy evolution showing field-on stabilization and the post-bias barrier-crossing relaxation process. In the phase-field simulations, the electric, gradient and total free energies were monitored over two cycles of applying and removing a 5 V tip bias. Upon field application (“Field on”), the system absorbs external electrical energy, with decrease in the gradient energy indicating the electric field suppresses the chiral rotation favoured by DMI term leading to flattening of the polarization modulation and lowering the gradient penalty. Upon removal of the field (around step 5000), the system spontaneously reorganizes into a mixed near flat + zigzag configuration, during which the gradient energy rises sharply as the vortices reconstruct under the residual DMI stabilization, then gradually relaxes to a slightly lower steady level. The total energy term decreases as the zigzag vortex configuration is transiently stabilized under the applied bias. When the field is removed (“Field off”), the total energy rises sharply, reaching a maximum of $\sim 3.37 \times 10^5$ eV—corresponding to the point at which the system traverses the switching barrier. The energy difference between the initial zigzag state (2.85×10^5 eV) and this transition peak (3.37×10^5 eV) defines an energy barrier of $\sim 5.2 \times 10^4$ eV. The system subsequently relaxes into a mixed near-flat + zigzag configuration at 2.95×10^5 eV, which is $\sim 1.0 \times 10^4$ eV higher in energy than the pristine zigzag state, consistent with their relative thermodynamic stability. These results demonstrate that the applied electric field reshapes the polarization energy landscape such that, after field removal, the residual electrostatic energy propels the system across the $\sim 5.2 \times 10^4$ eV barrier into the adjacent zigzag + near-flat configuration. The resulting energy-evolution trajectories thus offer a clear, quantitative picture of energy injection, barrier crossing, and subsequent relaxation, in agreement with the experimentally observed zigzag-to-flat transformation.

Temporal evolution of the vortex boundary in the trilayer.

The temporal evolution of a vortex boundary was examined in Fig. 4. Here, the AFM probe was moved with +5V bias along cyan arrow to create a new vortex boundary (Supplementary Fig. 10a,b). Subsequent scans conducted at 5 and 150 minutes post-formation (Supplementary Fig. 10c-d) confirmed the temporal stability of the newly formed vortex boundary, showing no significant variations over time. These findings demonstrate the stability of vortex boundaries under ambient conditions after external stimuli are applied, providing crucial insights into their robustness and reliability.



Supplementary Fig. 10| Temporal evolution of the vortex boundary in the trilayer. **a**, After harnessing the movement of the vortex boundary to the left, as shown in Fig. 4, a positive trailing field (+5V) was applied by moving the probe along the marked path (indicated in cyan). **b**, Following the removal of the field, a LPM scan revealed the formation of a new vortex boundary. **c-d**, The stability of this newly formed boundary was evaluated through successive PFM scans conducted 5 minutes (c) and 150 minutes (d) after its creation. The vortex boundary demonstrated exceptional stability over time, with no detectable changes observed post-formation.