

Controlled motion of polar vortex boundary in oxide heterostructures

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This manuscript has been previously reviewed at another journal. This document only contains information relating to versions considered at Nature Communications.

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This file contains all reviewer reports in order by version, followed by all author rebuttals in order by version.

Version 0:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

This manuscript addresses a key aspect of these dipolar topological objects forming and stable at room temperature, which have been investigated much in the last 10 years. Namely how these nanoobjects respond to electric field and if they can be moved in a controllable manner. This is indeed of high interest for applications and nontrivial task to demonstrate how it works.

I reviewed the paper when it was submitted before and now I have appreciated that the authors made great efforts to revise the paper and to address my comments and concerns.

Reading through the response to the reviewers, I got puzzled by the values of the lattice parameters used by the authors for PbTiO₃. I myself have been working on PbTiO₃ for a long time and the values I had in mind were about 3.90 Å and 4.12 Å for the tetragonal unit cell at RT. The same I found again in some literature, see the attached data supporting my assumption. Also XRD data that I have for my PbTiO₃ films support these values.

Could the authors justify why they consider rather 3.9574 Å for bulk PbTiO₃ and thus compressive strain when PbTiO₃ is grown on DyScO₃? I expected that PbTiO₃ is under tensile strain on DyScO₃(110): see also the following paper

<https://doi.org/10.1063/1.4823536>

This is a different situation and may affect the predictions of the phase-field simulations accordingly.

Another point is that the paper is still very loaded with information that does not strongly justify itself. I was critical in my past paper evaluation about the choice of samples, saying that a sample with mixed states of vortices and a₁/a₂ domains is not a good idea, as the focus is on control of vortex motion. Thus studying the trilayer sample would suffice for a very good and thorough study. The reader will then easier get the point and not become tired of the many data on different samples, etc.

The authors replied in their response that:

"The trilayer sample stabilizes a pure vortex phase, while the superlattice sample contains a mixed vortex and a₁/a₂ domain phase. Studying both architectures enables the identification of the various energy terms governing vortex motion and demonstrates that deterministic, reversible manipulation of vortices is achievable even in the presence of competing ferroelastic-ferroelectric domains.

This comparative approach establishes the universality and robustness of electrically driven vortex motion across different structural and energetic configurations."

I am in favor to "less is more", and for clear focus of the message, and I strongly suggest the authors to get their message across with summarizing the data and the conclusions resulting from the study of the trilayer sample only.

I think that upon thoughtful revision this manuscript can be improved and become suitable for Nature Communications.

Reviewer #2

(Remarks to the Author)

This work investigates the electric-field-driven controllable motion of polar vortices in PTO/STO oxide heterostructures. The authors utilize localized pulsed and trailing electric fields via a conductive AFM tip to claim the realization of reversible and long-distance movement of polar vortices. Combining piezoresponse force microscopy (PFM), in-situ scanning transmission electron microscopy (STEM) and phase-field simulations, the authors state that they have manipulated vortices over a distance up to ~3000 nm. Higher-order topological structures including vortex rings and vortex ring pairs are also observed, and the long-term stability of displaced vortex structures is verified.

However, there is a critical conceptual misunderstanding throughout the whole paper: the authors repeatedly equate vortex boundary displacement with the motion of polar vortices. The large displacement values reported in the text only describe boundary position changes, rather than the long-range motion of individual or assembled polar vortices. The core claim of this work — controlled motion of polar vortices — is not achieved as stated. This fundamental flaw invalidates the main conclusion of the manuscript.

Specifically, all PFM, STEM results and phase-field simulations only demonstrate the lateral shift of vortex domain boundaries between zig-zag vortex structures and transitioned near-flat vortex configurations, rather than the translational motion of individual or assembled polar vortex lattices as claimed in the title, abstract, main text and figures. The reported displacement values (e.g., ~1170–2000 nm, ~2240–3000 nm, ~3100 nm) exclusively refer to the position change of boundaries between adjacent vortex domains, instead of the long-range movement of complete polar vortex units. In fact, the applied electric field mainly induces vertical reconfiguration of vortices along the [001] direction as shown in Figure 2: external fields drive vortex cores to approach one another and transform the original zig-zag arrangement into a flat structure. In addition, the novelty of this work is insufficient. A number of prior publications have already reported field-induced motion and dynamics of polar vortices, such as *Science Advances*, 8 (1), eabj8030 and *Acta Materialia* 152 (2018) 155e161. The present study fails to deliver distinct new findings compared with these established works.

The authors repeatedly mention "topologically protected controlled motion and manipulation" in the abstract and main text, but they never clarify how the polar vortices gain topological protection during the movement. The dynamic behavior of ferroelectric polar vortices is fundamentally different from magnetic topological textures. Magnetic skyrmions and vortices are stabilized and topologically protected by intrinsic Dzyaloshinskii–Moriya (DM) interaction, while such protection mechanism is absent in the ferroelectric vortex system discussed here.

For the PTO/STO superlattice sample where vortices coexist with a1/a2 domains, the electric field application only triggers transition of vortex domains, instead of genuine vortex motion. The LPFM signals merely reflect the shift of transition boundaries, and cannot be used to quantify the translational moving distance of vortices.

In Figure 2i, the authors applied a 17 V bias using an in-situ tungsten tip on the trilayer sample with a size below 10 nm. According to previous literature, an electric field of such magnitude is strong enough to destroy polar vortex structures. It is confusing why only vortex reconfiguration rather than vortex annihilation was observed in this experiment. Furthermore, the authors do not specify the radius of the tungsten probe. Only initial and final states before and after bias application are presented; the full dynamic evolution process is missing, which makes the experimental evidence incomplete.

The authors propose that the coupling between net in-plane polarization and in-plane trailing electric field drives vortex motion. Nevertheless, the electric field applied in the in-situ STEM test (Figure 2i) is along the out-of-plane direction. To validate the proposed mechanism, dedicated in-situ experiments with pure in-plane electric fields are strongly required, which are not provided in this manuscript.

Moreover, this work suffers from problematic data presentation. Readers will struggle to comprehend the five intricate figures composed of numerous sub-panels, which cover two different sample systems simultaneously. The authors must completely revise their data layout to be more succinct and persuasive.

Version 1:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

The authors addressed in great detail my remarks. I think the paper is much easier to read and follow now.

The only concern I have is that now, as prompted by the other reviewer's comments, the value of the findings has decreased: the title (following the content) changed to "Controlled motion of polar vortex boundary in oxide heterostructures". That the boundaries of the vortices are the mobile objects (and not the vortices as individual objects can be manipulated in a controlled manner) does not appear to be a great finding to me, and of any serious technological impact, the described findings have mere academic relevance.

Nevertheless, it is a comprehensive study, with multiple experimental efforts carefully conducted, so the manuscript is worth publishing.

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Response letter for NCOMMS-26-040921-T

Reviewer #1

Remark: This manuscript addresses a key aspect of these dipolar topological objects forming and stable at room temperature, which have been investigated much in the last 10 years. Namely how these nanoobjects respond to electric field and if they can be moved in a controllable manner. This is indeed of high interest for applications and nontrivial task to demonstrate how it works. I reviewed the paper when it was submitted before and now I have appreciated that the authors made great efforts to revise the paper and to address my comments and concerns.

Response: We thank the reviewer for appreciating and highlighting the importance of our work. Also, we thank review for appreciating our efforts to improve the manuscript. Indeed his/her suggestion were very useful to improve the clarity and quality of the manuscript. Following we have provided the point-by-point answers to reviewer's comments.

Comment 1: Reading through the response to the reviewers, I got puzzled by the values of the lattice parmeters used by the authors for PbTiO₃. I myself have been working on PbTiO₃ for a long time and the values I had in minde were about 3.90 Angstrom and 4.12 Angstrom for the tetragonal unit cell at RT. The same I found again in some literature, see the attached data supporting my assumption. Also XRD data that I have for my PbTiO₃ films support these values. Could the authors justify why they consider rather 3.9574 Å for bulk PbTiO₃ and thus compressive strain when PbTiO₃ is grown on DyScO₃? I expected that PbTiO₃ is under tensile strain on DyScO₃(110): see also the following paper

<https://doi.org/10.1063/1.4823536>

This is a different situation and may affect the predictions of the phase-field simulations accordingly.

Response: We sincerely apologize for the confusion caused by referencing the value of 3.9574 Å in our previous response. This value does not represent the relaxed room-temperature tetragonal lattice parameter. Rather, in our phase-field simulations, the reference state for defining mechanical equilibrium and thermodynamic energies is based on the pseudocubic lattice parameters. Following the classic thermodynamic theory of PbTiO₃ formulated by Haun M.J. *et al. J. Appl. Phys.* **62**, 3331–3338 (1987), the reference pseudocubic lattice constant for PbTiO₃ is taken as ~3.9547–3.9574 Å.

This methodology for setting the reference lattice constants (e.g., ~3.9574 Å for PbTiO₃ and 3.905 Å for SrTiO₃) to compute misfit strain is the standard theoretical framework for simulating polar topologies. It was fundamentally established in the seminal Nature paper by Yadav A.K. *et al. Nature* **530**, 198–201 (2016), which pioneered the observation and phase-field modeling of polar vortices in oxide superlattices, and it continues to be employed in recent topological studies by Zheng, S. *et al. Nano Lett.* **25**, 4, 1397–1403 (2025).

In our simulations, we adopted this exact foundational framework. By utilizing the reference pseudocubic lattice parameter of 3.9574 Å for PbTiO₃ against the anisotropic DyScO₃ substrate (3.952 Å and 3.947 Å), the phase-field model inherently and accurately captures the stabilization of the vortex topology in PTO/STO heterostructure.

Comment 2: Another point is that the paper is still very loaded with information that does not strongly justify itself. I was critical in my past paper evaluation about the choice of samples, saying that a sample with mixed states of vortices and a_1/a_2 domains is not a good idea, as the focus is on control of vortex motion. Thus studying the trilayer sample would suffice for a very good and thorough study. The reader will then easier get the point and not become tired of the many data on different samples, etc. The authors replied in their response that:

“The trilayer sample stabilizes a pure vortex phase, while the superlattice sample contains a mixed vortex and a_1/a_2 domain phase. Studying both architectures enables the identification of the various energy terms governing vortex motion and demonstrates that deterministic, reversible manipulation of vortices is achievable even in the presence of competing ferroelastic-ferroelectric domains. This comparative approach establishes the universality and robustness of electrically driven vortex motion across different structural and energetic configurations.”

Response: We thank the reviewer for this constructive and helpful suggestion. We understand the reviewer’s concern that including samples with mixed vortex and a_1/a_2 ferroelastic domain states may obscure the central message of the manuscript, which is the deterministic control of vortex motion. After carefully reconsidering the structure and focus of the paper, we agree that the trilayer sample, which stabilizes a well-defined vortex phase, provides a cleaner and more direct platform to demonstrate electrically controlled vortex displacement.

Comment: I am in favor to "less is more", and for clear focus of the message, and I strongly suggest the authors to get their message across with summarizing the data and the conclusions resulting from the study of the trilayer sample only.

Response: In the revised manuscript, we have removed the data related to the superlattice sample that exhibited mixed vortex and a_1/a_2 domain configurations. The revised manuscript now focuses exclusively on the trilayer architecture, where the vortex phase is clearly established and its motion can be analyzed without the additional complexity introduced by competing ferroelastic-ferroelectric domains. We believe this revision significantly improves the clarity, coherence, and impact of the study.

With this change, the manuscript now presents a more streamlined and focused demonstration of controlled vortex motion. The revised figure set has also been reorganized accordingly, with the new figures in the revised manuscript as follows:

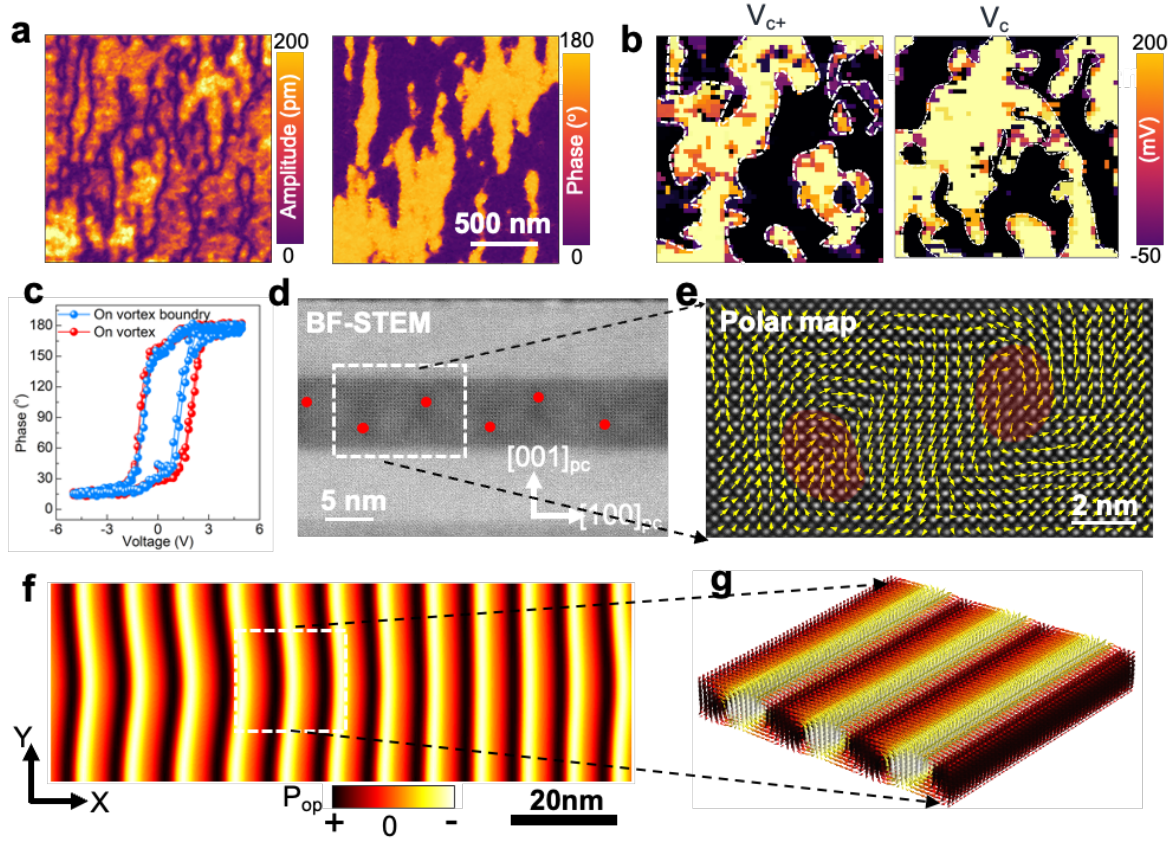


Figure 1| Structure and stabilization of polar vortices in STO/PTO/STO trilayer grown on DyScO₃ (001)_{pc} substrates. **a**, LPMF scan for the trilayer with amplitude (left) and phase (right). The dark contrast in the amplitude represents the boundary of the vortex. **b**, SS-PFM mapping for trilayer reveals spatial distribution of switching behaviour for vortex. The positive nucleation bias (V_{c+} , left panel) showing domains with different polarity having different contrast (marked by white dashed line) and the average coercive bias (V_c , right panel) of same region shows vortex boundary having significantly lower coercive field. **c**, The hysteresis loops extracted from the vortex and near vortex boundary for trilayer illustrating vortex boundary having lower coercive bias compared to the vortex demonstrating vortex boundary motion can be controlled using lower electric field. **d**, Cross-sectional bright field (BF) STEM, and **e**, high-resolution polarization mapping images of white mark region for the trilayer samples show CW and CCW continuous polarization rotation within the PTO layers corresponding to polar structures. Yellow arrows indicate the local polar directions, while red dots denote the vortex cores. **f-g**, The phase field simulated images for the trilayer. The zoomed image shown in the right side is the 3D view of the vortices.

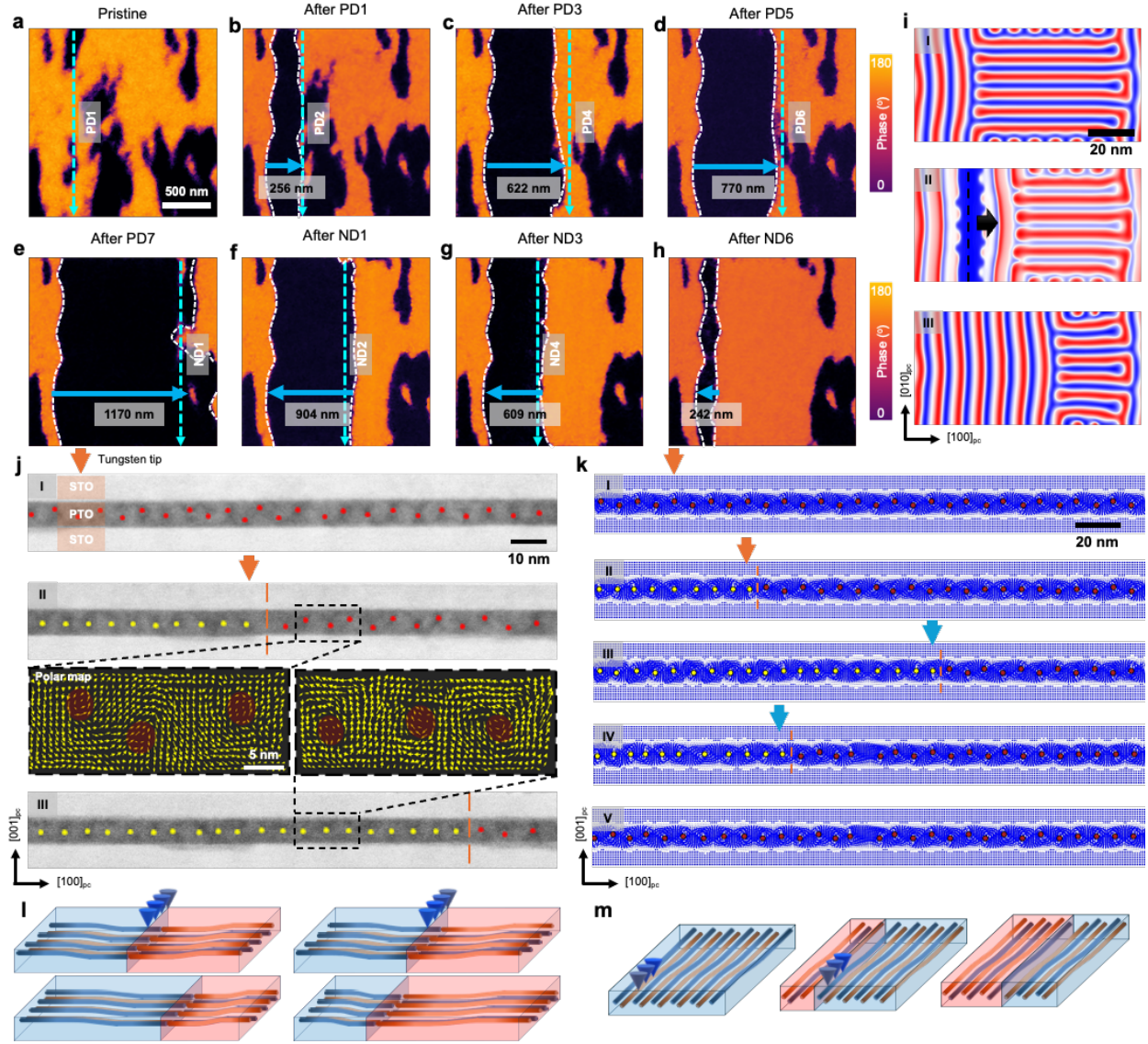


Figure 2| Deterministic motion of the vortex boundary in the STO/ PTO/ STO trilayer. **a**, Pristine LPM images show well-defined vortex arrays. **b–e**, Successive application of positive trailing fields (PD1–PD7; +5 V) moves the vortex boundary rightward, reaching a total displacement of ~1170 nm. **f–h**, Applying negative trailing fields (ND1–ND6; -5 V) reverses the motion, producing a cumulative leftward movement. **i**, Plane view phase-field simulations illustrating the motion of polar vortex and vortex boundary with application of electric field (I–III). **j**, Cross-sectional *in-situ* STEM confirms the vortex motion (I–III) in the trilayer. Initially, the vortex cores are arranged in an up-down-up configuration (I, red dots). Application of a +17 V electric field via a tungsten tip (denoted by orange arrow) aligns the vortex cores into a near-flat configuration (II, yellow dots), forming a boundary between the up-down arrangement and the near-flat configuration (orange dashed line). A subsequent +17 V application moved the boundary further rightward (III), consistent with the PFM results. Field-induced switching reverses the zigzag vortex arrangement while preserving topological state as shown by polarization mapping before and after application of electric field (black dashed region). This demonstrates the topologically protected motion of polar vortices during field-induced motion. **k**, Cross-sectional

phase-field simulations illustrating the initial polar vortex configuration (I). The orange arrow marks the initial position of the simulated electrode where a positive electric field is applied (I). Upon applying the electric field, the polar vortex shifts to the right and the near-flat region expands (II–III), consistent with in situ STEM observations. To reversibly move the polar vortex, a negative electric field is applied (III–IV; blue arrow). Sequential phase-field simulations further illustrate the reversible motion of the polar vortex (IV–V). **l**, CW and CCW polar vortices are marked in blue and red, respectively. Their zigzag arrangement generates regions with opposite net in-plane polarization. When the AFM probe moves downward under a positive bias, vortices in the blue region expand, shifting the vortex boundary to the right (left panel in **l**). Reversing the bias reverses the favored vortex expansion, causing vortices in the red region to expand and shifting the boundary to the left (right panel in **l**). **m**, Motion of the vortex boundary when the electric field is applied perpendicular to the vortex tubes, leading to the rearrangement of the polar-vortex tubes.

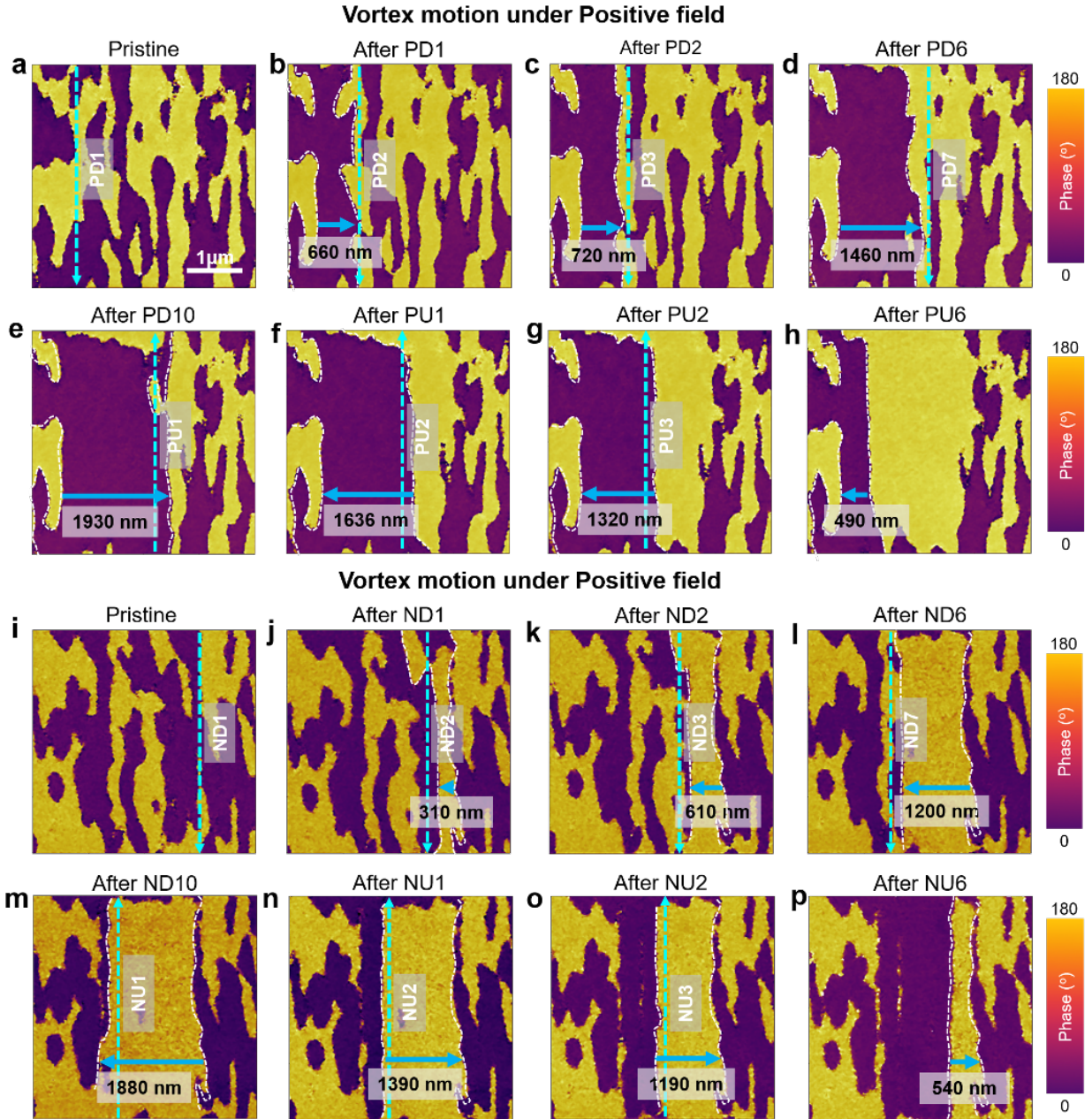


Figure 3| Directional control of the vortex boundary motion in the trilayer. **a**, Pristine vortex state. The cyan arrow indicates the path along which the AFM probe is moved (PD: tip motion up to down with positive polarity of bias). **b**, After the removal of PD1, the PFM scans reveal a distinct rightward shift in the vortex boundary, marked by the blue arrow. **c-e**, Vortex boundary 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 boundary can be moved reversibly i.e. leftward shift. **i**, 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). **j**, After the removal of ND1, the PFM scans reveal a distinct leftward shift in the vortex boundary, marked by the blue arrow. **k-m**, Shows the vortex boundary movement after ND2-ND10 with total movement

of ~ 1880 nm. **n-p**, 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.

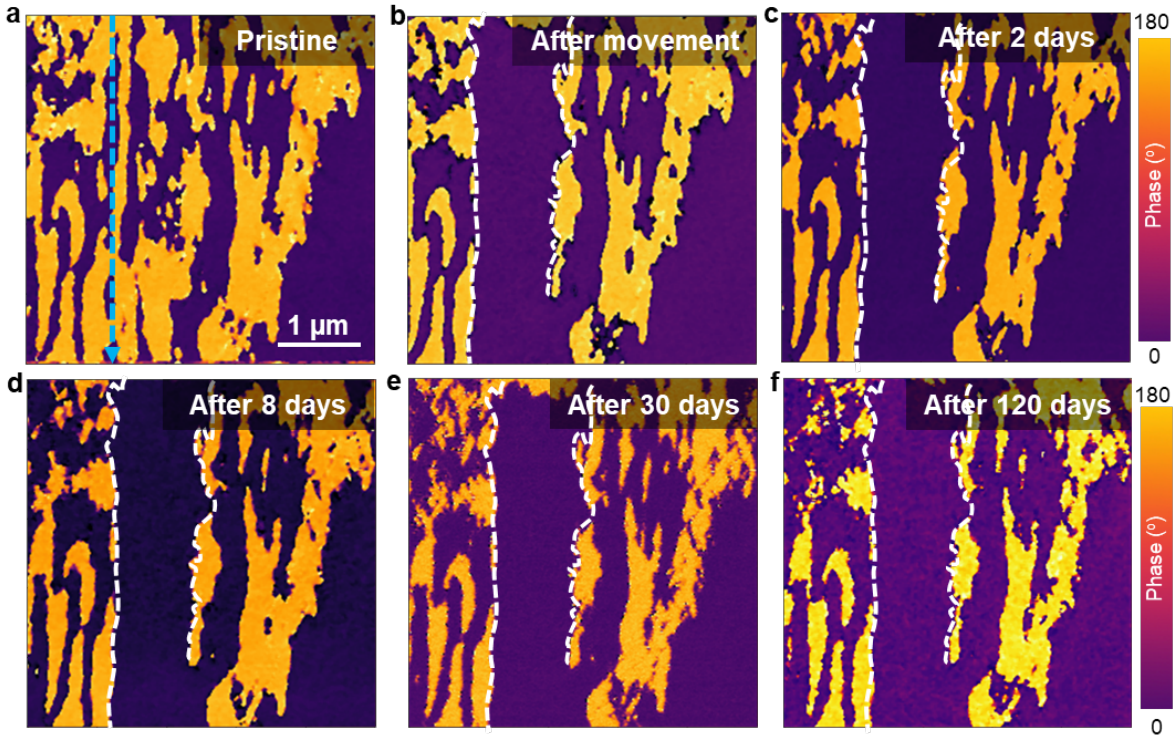


Figure 4| Stability of vortex boundary motion. **a**, Pristine LPFM images for the PTO/STO/PTO trilayer confirm presence of well-defined vortices. Following the application of trailing field from up to down, as described in earlier experiments, the vortex boundary shifted from left to right. Subsequent PFM scans conducted at various intervals reveal the long-term stability of the moved vortex boundary: **c**, after 2 days; **d**, after 8 days; **e**, after 30 days; and **f**, after 120 days. Each scan demonstrates the sustained robustness of the moved vortex boundary over time.

We again thank the reviewer for this valuable suggestion. We fully agree that a more focused presentation will strengthen the manuscript and allow the central message to emerge more clearly. In the revised version, we have therefore followed the reviewer's recommendation and restricted the discussion and data presentation to the trilayer sample only.

This revision removes the additional complexity associated with mixed vortex and a_1/a_2 domain states in the superlattice samples, and instead provides a concise and coherent demonstration of electrically controlled vortex motion in the trilayer system.

Final comment: I think that upon thoughtful revision this manuscript can be improved and become suitable for Nature Communications.

Response: We thank reviewer for this final remark. We believe with all these changes the referee will be satisfied with the revised manuscript and recommend for the publication in Nature communications.

Reviewer #2

Remark: This work investigates the electric-field-driven controllable motion of polar vortices in PTO/STO oxide heterostructures. The authors utilize localized pulsed and trailing electric fields via a conductive AFM tip to claim the realization of reversible and long-distance movement of polar vortices. Combining piezoresponse force microscopy (PFM), in-situ scanning transmission electron microscopy (STEM) and phase-field simulations, the authors state that they have manipulated vortices over a distance up to ~3000 nm. Higher-order topological structures including vortex rings and vortex ring pairs are also observed, and the long-term stability of displaced vortex structures is verified.

Response: We thank reviewer for going through the manuscript and providing his/her insightful remark. We believe that with the incorporation of the suggestions provided by the reviewer, our manuscript further strengthen. Here we have provided point-by-point answers to the concerns raised by the reviewer.

Comment 1: However, there is a critical conceptual misunderstanding throughout the whole paper: the authors repeatedly equate vortex boundary displacement with the motion of polar vortices. The large displacement values reported in the text only describe boundary position changes, rather than the long-range motion of individual or assembled polar vortices. The core claim of this work — controlled motion of polar vortices — is not achieved as stated. This fundamental flaw invalidates the main conclusion of the manuscript.

Response: We thank the reviewer for this critical comment and appreciate the concern raised. We agree that the PFM images primarily show the motion of domain-like boundaries under the applied electric field, rather than direct imaging of individual polar vortices.

We have revised the manuscript to clarify this point. The contrast observed by PFM corresponds to mesoscopic polar-vortex domains, not individual vortex tubes. These mesoscopic domains arise from collective arrangements of polar vortices, where the vortex sequence changes from counterclockwise-up/clockwise-down to clockwise-up/counterclockwise-down, resulting in a change of the net in-plane polarization, as established by Behera *et al.* (*Science Advances* **8**, eabj8030, 2022).

To make this distinction clear, we have added a summary figure, Fig. R2.1. Plane-view STEM images in Fig. R2.1a resolve individual polar-vortex tubes and the boundaries separating different vortex assemblies. In contrast, the corresponding PFM images show larger mesoscopic domains, which represent the collective polar-vortex arrangements rather than the individual vortex tubes. Therefore, throughout the revised manuscript, we now describe the experimentally observed effect as **polar-vortex-boundary motion**.

Under an applied electric field from the moving AFM probe, the polar-vortex boundary is displaced. This boundary displacement reflects the collective expansion and contraction of neighbouring vortex assemblies on either side of the boundary. Thus, while PFM directly tracks vortex-boundary motion, the underlying physical picture involves redistribution of the adjacent polar-vortex tubes. We have clarified this relationship in Fig. R2.1c,d using schematic illustrations.

As additional support, we show that the boundary couples to the trailing electric field of the moving AFM probe. Depending on the voltage polarity and scan direction, one vortex assembly expands while the neighbouring assembly contracts, resulting in a collective displacement of the polar-vortex boundary. This interpretation is consistent with the in-situ cross-sectional-view STEM observations, phase-field simulations and finite-element analysis of the trailing electric field shown in Fig. R2.2 and the revised Fig. 2 of the main manuscript.

We acknowledge that the original manuscript did not clearly distinguish between direct PFM observation and microscopic interpretation. We have therefore revised the relevant text, figures and schematics to state explicitly that PFM measures polar-vortex-boundary motion, while the associated vortex-tube expansion/contraction provides the microscopic mechanism for this boundary displacement.

We have added below text in the revised main text (page 4) to make this clear, read as,

“We observe that the polar-vortex cores in $\text{PbTiO}_3/\text{SrTiO}_3$ heterostructures adopt a zigzag, rather than collinear, arrangement, giving rise to a finite net in-plane polarization. By deterministically controlling the motion of a biased AFM probe, either through the probe polarity or scan direction, we modulate the in-plane trailing electric field that couples to this net polarization and drives polar-vortex-boundary motion. These results establish a reproducible route for electrically controlled displacement of polar vortex boundaries and reveal the role of local electrostatic energy landscapes in governing vortex dynamics. This deterministic and reversible control of polar vortex-boundary motion provides a foundation for reconfigurable oxide nanoelectronics based on topological polarization textures.”

We have added below text in the revised main text (page 6) to make this clear, read as,

“Reversing the vortex boundary motion (right to left) by applying the opposite polarity of the trailing field or reversing the probe motion direction (Fig. 2-3, Supplementary Fig. 5-8).”

“Phase-field simulations were further performed in the plane-view geometry, which show that the polar vortices rearrange along with boundary motion after the application of a voltage pulse (Fig. 2i, I–III).”

We have incorporated the following text in the revised main text (page 7-8), read as,

“The experimental results are further summarized using schematic (Fig. 2l-m). First schematics illustrate (Fig. 2l) polar-vortex reconfiguration when the electric field is applied along the vortex tubes, resulting in motion of the boundaries through vortex expansion. The second schematic (Fig. 2m) shows the corresponding response when the field is applied perpendicular to the vortex tubes, producing clear motion of the vortex boundaries.”

As per reviewer suggestion and make it clear, now we have changed the manuscript title in the revised manuscript, read as, “**Controlled motion of polar vortex boundary in oxide heterostructures**”

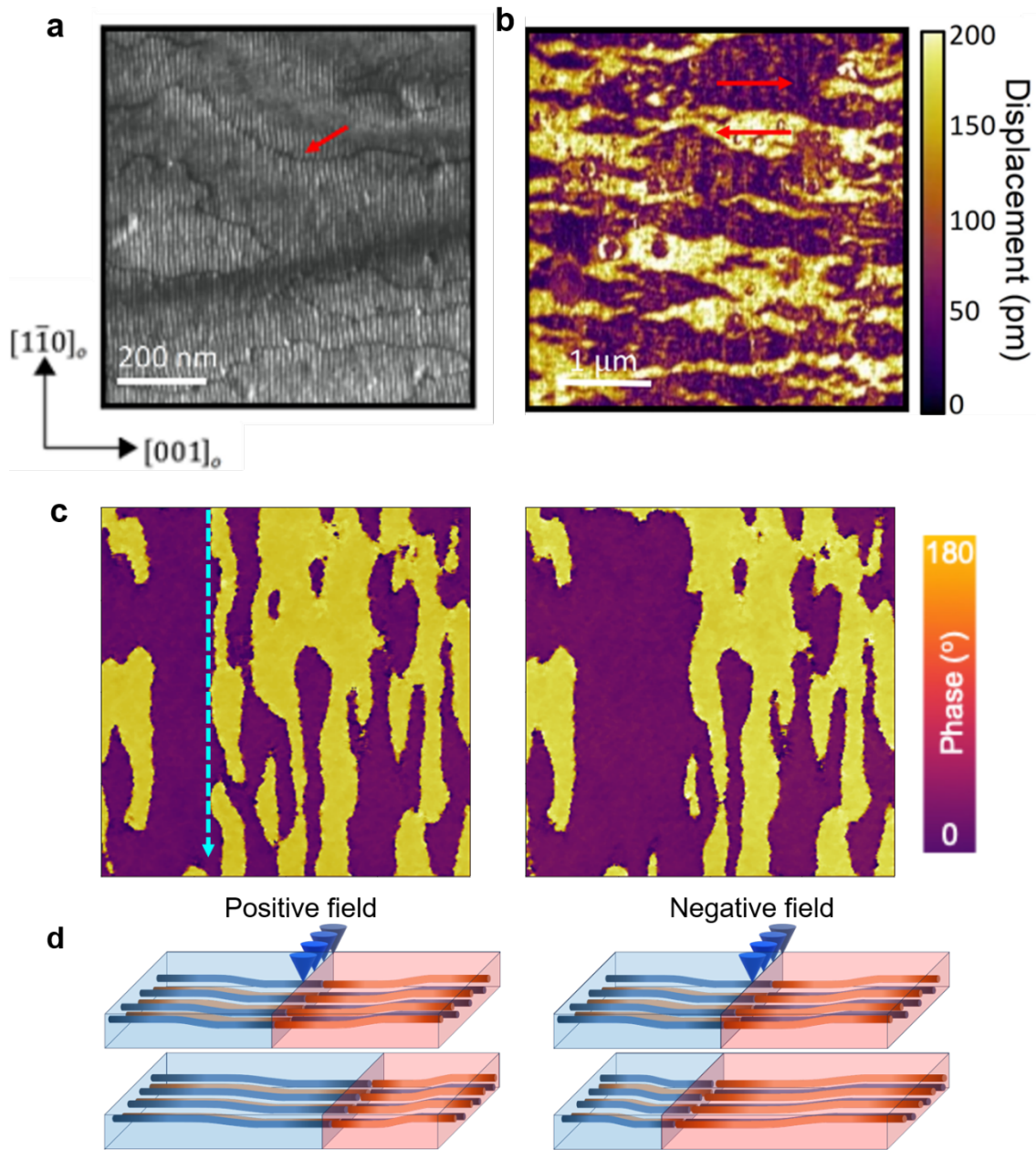


Fig.R2.1| PFM contrast in polar vortex in PTO-STO heterostructure **a**, Plane view STEM images showing the polar vortex boundary (marked by red arrow) across which the zizzag arrangement of polar vortex leading to net in-plane polarization from Behra *et al.* **b-c**, PFM images showing the contrast from the vortex domains from Behra *et al.* (b) and our study for (c) and showing how with electric field the polar vortex boundary can be moved. **d**, schematic representation the underlying physics of polar vortex-boundary motion.

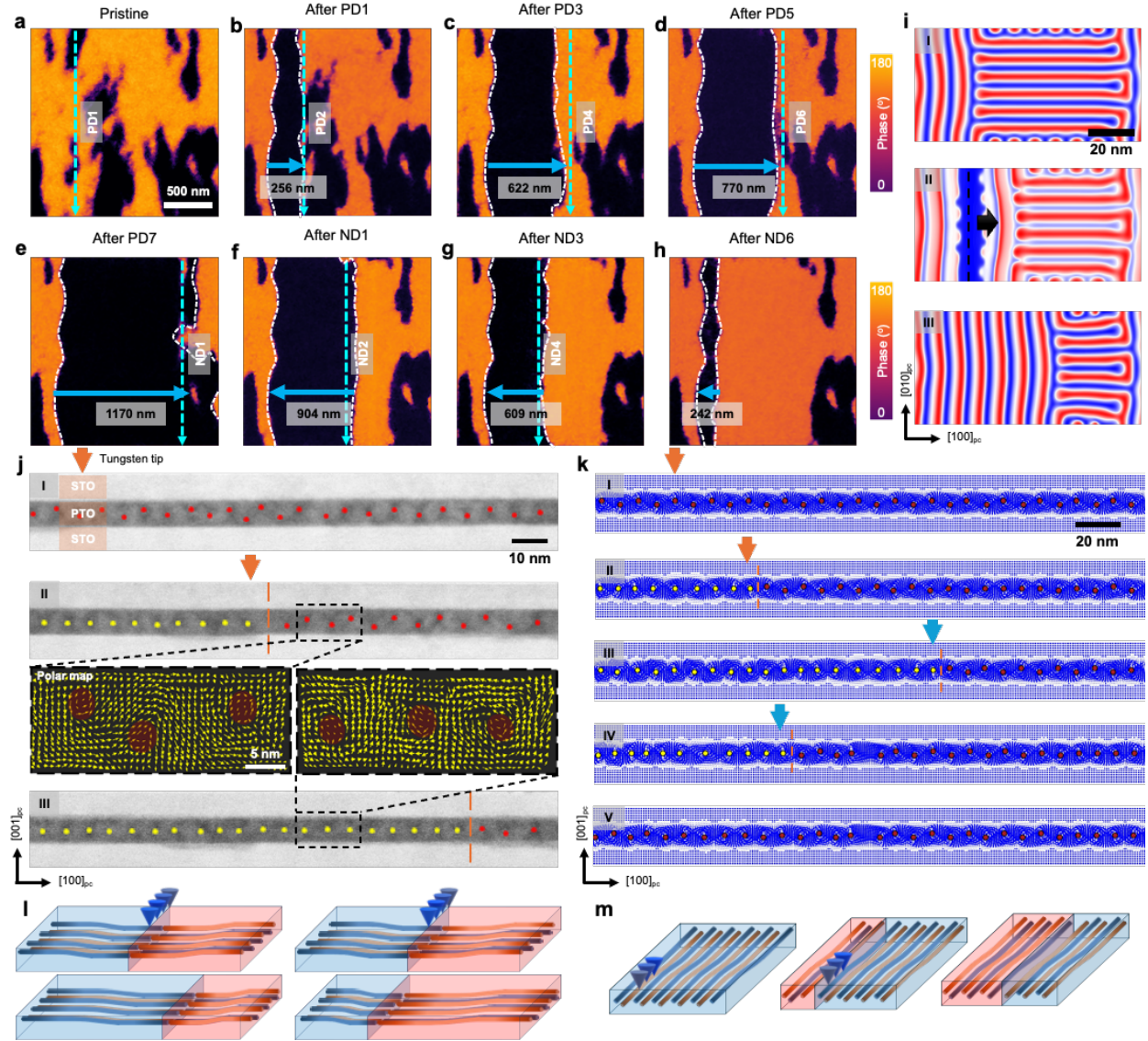


Fig.R2.2| Deterministic motion of the vortex boundary in the STO/ PTO/ STO trilayer. **a**, Pristine LPM images show well-defined vortex arrays. **b–e**, Successive application of positive trailing fields (PD1–PD7; +5 V) moves the vortex boundary rightward, reaching a total displacement of ~1170 nm. **f–h**, Applying negative trailing fields (ND1–ND6; -5 V) reverses the motion, producing a cumulative leftward movement. **i**, Plane view phase-field simulations illustrating the motion of polar vortex and vortex boundary with application of electric field (I-III). **j**, Cross-sectional *in-situ* STEM confirms the vortex motion (I-III) in the trilayer. Initially, the vortex cores are arranged in an up-down-up configuration (I, red dots). Application of a +17 V electric field via a tungsten tip (denoted by orange arrow) aligns the vortex cores into a near-flat configuration (II, yellow dots), forming a boundary between the up-down arrangement and the near-flat configuration (orange dashed line). A subsequent +17 V application moved the boundary further rightward (III), consistent with the PFM results. Field-induced switching reverses the zigzag vortex arrangement while preserving topological state as shown by polarization mapping before and after application of electric field (black dashed region). This demonstrates the topologically protected motion of polar vortices during field-induced motion. **k**, Cross-sectional

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Comment 2: Specifically, all PFM, STEM results and phase-field simulations only demonstrate the lateral shift of vortex domain boundaries between zig-zag vortex structures and transitioned near-flat vortex configurations, rather than the translational motion of individual or assembled polar vortex lattices as claimed in the title, abstract, main text and figures. The reported displacement values (e.g., ~1170–2000 nm, ~2240–3000 nm, ~3100 nm) exclusively refer to the position change of boundaries between adjacent vortex domains, instead of the long-range movement of complete polar vortex units. In fact, the applied electric field mainly induces vertical reconfiguration of vortices along the [001] direction as shown in Figure 2: external fields drive vortex cores to approach one another and transform the original zig-zag arrangement into a flat structure.

Response: We thank the reviewer for this critical and insightful comment. We fully appreciate the seriousness of the concern and agree that the original manuscript contained a significant conceptual ambiguity regarding the relationship between vortex-boundary displacement and the motion of the polar vortices themselves.

In the original version, the large displacement values reported in the text were obtained from the position changes of the boundaries observed in PFM. As the reviewer correctly notes, the TEM/STEM data primarily showed a transformation from a zig-zag to a near-flat vortex configuration, which can appear as a simple rearrangement rather than long-range translational motion of the vortices. We acknowledge that this discrepancy, together with the language used in the original manuscript, created the impression that boundary motion was being equated with vortex motion without sufficient justification.

To resolve this issue rigorously we have performed additional phase-field simulations and added a clarifying schematic (now included in Fig. R2.2 and the revised main-text as figure 2). These new results highlight a crucial experimental distinction:

- When the electric field is applied **perpendicular** the vortex tubes (the geometry relevant to the original TEM/STEM experiments), the dominant response is longitudinal reconfiguration of the vortex cores, with limited lateral translation.
- In contrast, when the electric field is applied parallel to the vortex tubes, corresponding to the PFM geometry, the response involves lateral displacement of the polar-vortex ensemble. In this case, vortices on one side of the boundary elongate while those on the opposite side contract, producing a collective displacement of the vortex boundary. To avoid any possible

ambiguity, and in line with the reviewer's suggestion, we now describe the experimentally observed process as polar-vortex-boundary motion.

- We have also performed additional phase-field simulations to support the interpretation of vortex and vortex-boundary motion. These results have been incorporated into the revised main text as Fig. 2. In addition, we have added a schematic to Fig. 2 to clarify how different electric-field configurations lead to different vortex responses and boundary-displacement pathways.

In response to the reviewer's comment, we have revised the manuscript title and the relevant text on pages 4 and 8 to improve the precision of the language. We are grateful to the reviewer for highlighting this important point. The additional simulations and schematic now allow us to clarify the geometry-dependent nature of the vortex response and vortex-boundary motion more accurately.

Comment 3: In addition, the novelty of this work is insufficient. A number of prior publications have already reported field-induced motion and dynamics of polar vortices, such as *Science Advances*, 8 (1), eabj8030 and *Acta Materialia* 152 (2018) 155e161. The present study fails to deliver distinct new findings compared with these established works.

Response: We thank reviewer for raising his/her concern about novelty, and providing previous relevant work. Here we would like to bring this in reviewer's attention that the provided references are some how related to the author of this work as well, therefore we are well aware about both of these works with many of our current co-authors are part of these work, which makes it easier to explain how the works provided by reviewer's are different from the current work and how current work advances the presented work.

1. We would like to highlight the novelty of the present work relative to the important early study by Hong et al., *Acta Materialia* 152, 155–161 (2018), titled “Blowing polar skyrmion bubbles in oxide superlattices”. That elegant work, in which one of the co-authors of the current manuscript also participated, primarily focused on the electric-field-driven manipulation and transformation of polar vortices into polar skyrmions. However, it did not focused on the demonstration of vortex/vortex boundary motion.

To the best of our knowledge, the present study provides the first experimental demonstration of controlled motion of polar vortex boundray driven by a local electric field. This advance clearly distinguishes our work from previous reports and underscores its novelty.

2. The work by Behera et al. (*Science Advances* 8, eabj8030 (2022); <https://doi.org/10.1126/sciadv.abj8030>), titled “Electric field control of chirality,” primarily focuses on the electric-field control of material chirality. This is achieved through a change in the zigzag arrangement of polar vortices, which produces a net change in chirality (handedness). By applying an in-plane electric field, the authors successfully switch the chirality of the polar vortices. However, that study does not address the motion of polar vortices/boundaries, nor does it present any direct or indirect evidence of such motion.

3. In addition to the paper cited by the reviewer, P. Behera et al. (*Adv. Mater.* 2025, 37, 2410149) also reported in-situ electric-field-dependent second-harmonic generation (SHG) measurements performed with an in-plane electric field applied through interdigitated electrodes (IDEs) on polar vortex. We have included these results as Fig. R2.3. The data clearly show that the application of an in-plane electric field does not drive domain-wall motion. Instead, the domain-wall positions remain fixed while the chirality is switched, leaving the overall domain configuration unchanged at both $+150 \text{ kV cm}^{-1}$ and -150 kV cm^{-1} .

These observations underscore that achieving motion of polar vortices/boundaries is a non-trivial task that cannot be realized simply with conventional in-plane electrode geometries. More creative approaches are required to enable non-volatile motion of polar vortices and their associated boundaries.

In the present manuscript we demonstrate precisely this capability: the application of a local electric field (via a scanning AFM probe) drives the motion of vortex boundaries. Moreover, we achieve unipolar electric-field control of this motion. Unipolar control of domain walls is already non-trivial even for conventional ferroelectric domains and, to the best of our knowledge, has been reported in only one prior study on a van der Waals material (Jiang, X. *et al. Nat Commun* **16**, 7607 (2025)). That work, while unique, relies on ion migration, which alters the material topography. In contrast, our polar-vortex system realizes unipolar control simply by reversing the AFM probe scan direction, at comparatively low fields and in a non-destructive manner.

With all these we believe that this manuscript possesses sufficient novelty and fits to the audience of Nature Communication family.

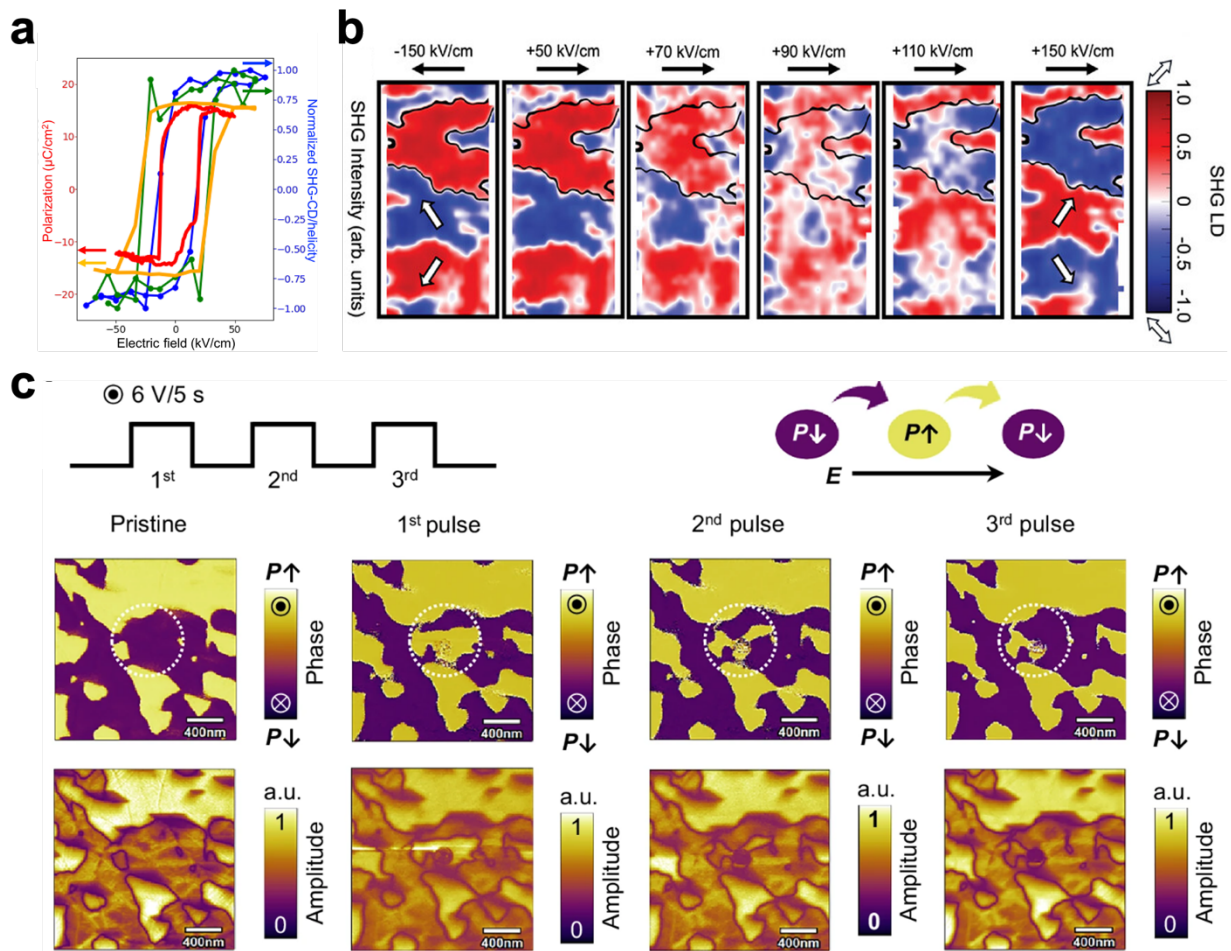


Fig. R2.3 | In-plane electric-field control of chirality and unipolar electric-field control of polarization. **a-b**, Probing the in-plane electric-field-induced change in chirality using second-harmonic generation (SHG). The data show that while an applied electric field can tune the chirality, the domain walls remain pinned. **c**, Unipolar control of ferroelectric domains, where subsequent electric-field pulses of the same polarity can switch the polarization into both up and down configurations.

Comment 5: The authors repeatedly mention "topologically protected controlled motion and manipulation" in the abstract and main text, but they never clarify how the polar vortices gain topological protection during the movement. The dynamic behavior of ferroelectric polar vortices is fundamentally different from magnetic topological textures. Magnetic skyrmions and vortices are stabilized and topologically protected by intrinsic Dzyaloshinskii–Moriya (DM) interaction, while such protection mechanism is absent in the ferroelectric vortex system discussed here.

Response: We thank the reviewer for raising this important point on the stability and topological protection of polar vortices during motion. The key requirement for potential device applications, such as racetrack memory, is that the topological state remains robust under external perturbations.

In magnetic systems (some cases), skyrmions and vortices can be annihilated or lose their protection when driven by fields or currents while many works have shown motion of magnetic skyrmions and vortex with their topological charge intact. In our case, we do not observe annihilation or collapse of the polar vortex structure. Instead, the vortices remain topologically stable while being displaced, demonstrating robust motion without loss of their characteristic topology.

Our results therefore show that polar vortices can be displaced over micrometer distances under applied electric fields while maintaining their topology, highlighting their robustness compared to other topological quasiparticles. When the AFM probe applies a trailing electric field, the vortices couple to this field and undergo the displacement (Fig. R2.2 above). The protection of the topological state is confirmed by both cross-sectional STEM and phase-field simulations (Fig. R2.2 above, Fig.2 main revised manuscript). During motion induced by the electric field, the vortex state remains intact, with no conversion into other competing ferroelectric phases, consistent with simulations demonstrating that the topological state remains constant after displacement. Furthermore, cross-sectional polarization mapping, combined with phase-field simulations, directly reveals that the vortex arrangement after motion is preserved without any loss of topological character. This robustness of the vortex topology under external driving fields is a key requirement for the realization of topological device concepts such as racetrack memory.

Additionally, we would like to clarify that our phase-field simulations explicitly include the Dzyaloshinskii–Moriya–like (DM-like) interaction, and its effect is clearly reflected in the gradient energy evolution under bias. This mechanism indeed extends beyond a purely conventional phenomenological picture by incorporating the microscopic chiral stabilization within the overall energy competition framework.

In our simulations, the DM-like interaction is implemented as an additional effective field term in the time-dependent Ginzburg–Landau equations, following Zhao *et al.* (*Nat. Mater.* 20, 341–345, 2021). The antisymmetric gradient coupling

$$f_{\text{DMI},i} = -\alpha D_N \frac{\partial P_z}{\partial x_i} + (1 - \alpha) D_B (\nabla \times \mathbf{P})_i$$

is computed at every time step and directly contributes to the polarization dynamics. Although the DMI energy is not output as a separate term, its influence manifests in the gradient energy evolution, which records how the system’s spatial polarization texture responds to the electric field.

The evolution of the gradient energy (Fig. R2.5) demonstrates the dynamic competition between the DM-like interaction and the external bias. Initially, when the field is applied to a pure zigzag vortex array, the gradient energy drops sharply, indicating that the electric field temporarily suppresses the chiral rotation favored by the DMI term—flattening the polarization modulation and lowering the local gradient penalty. Upon removal of the field (around step 5000), the system spontaneously reorganizes into a mixed 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. This behavior repeats in subsequent bias cycles, with each field-on/off sequence producing a reproducible energy dip followed by a recovery.

This non-monotonic evolution provides direct evidence of the field–DMI interplay, the external electric field momentarily suppresses the DMI-induced chirality (leading to energy drop), while the intrinsic DMI interaction subsequently drives the system toward a new metastable chiral configuration after the field is withdrawn (energy recovery). The hysteretic pattern of the gradient energy confirms that the system retains a memory of its previous chiral state, consistent with the reversible yet history-dependent vortex displacement observed experimentally.

Therefore, the observed vortex motion in our simulations still originates from the competition among electrostatic, elastic, Landau, and gradient (including DMI) energies, but with the DMI-like interaction explicitly incorporated into the gradient-energy term. This inclusion ensures that the interplay between chiral stabilization and electric-field–driven polarization reorientation is fully captured within our phase-field framework.

The corresponding gradient-energy evolution curve is added to the revised manuscript to explicitly demonstrate how the DMI-like interaction is already included in our model and naturally affects the vortex reconfiguration under applied fields (Supplementary Fig. 9).

About the topological protection, a subsequent application voltage, moved the boundary rightward, consistent with the PFM, STEM and Phase field results. Field-induced switching reverses the zigzag vortex arrangement while preserving topological state as shown by polarization mapping before and after application of electric field (black dashed region in Fig.2j main manuscript). This demonstrates the topologically protected motion of polar vortices during field-induced motion.

On page 8-9, in the main manuscript we have incorporated the text, reads as, “The energetic aspects of vortex motion are illustrated in Supplementary Fig. 9. The total energy profile exhibits a pronounced peak during the boundary displacement, corresponding to the transient state in which the vortex configuration is distorted under the applied field. Upon subsequent relaxation, the energy decreases toward a local minimum as the vortices stabilize at a new position. This behavior indicates that the injected electric energy enables the system to overcome the energetic barrier associated with vortex-boundary motion between two nearly degenerate configurations.

Further insight is provided by the evolution of the gradient energy, which directly reflects changes in the vortex polarization texture. During field application, the gradient energy (including Dzyaloshinskii–Moriya (DMI)) decreases as the vortex rotation is partially suppressed and the vortex cores become flattened. After removal of the field, the gradient energy rises sharply as the vortices reconstruct and the system traverses the switching pathway before relaxing into a mixed configuration (Supplementary Fig. 9b). Notably, a maximum in the energy is observed when the electric field polarity switches, which can be attributed to the transient antiparallel alignment between the local polarization and the applied field, resulting in an energy penalty prior to stabilization (Supplementary Fig. 9a–c). Together, these results establish a clear energetic pathway underlying the reversible motion of vortex boundaries driven by trailing fields.”

In page 18-19, in the revised main manuscript Method section read as “**Phase field Simulation.**

The spontaneous polarization vector ($\vec{P}$, $i = 1, 3$) $\vec{P} = (P_1, P_2, P_3)$ was chosen as the order parameter. The polarization dynamics were determined by resolving the time-dependent Ginzburg-Landau equation^{27,28}:

$$\frac{d\vec{P}}{dt} = -L \frac{\delta F(\vec{P})}{\delta \vec{P}}$$

Where $\vec{P}$ is the spontaneous polarization vector, and t is the evolution time step. The kinetic coefficient L is related to the domain wall mobility. F is the free energy functional of the system, consisting of four contributions (the Landau, mechanical, electric, and gradient energy densities), integrated over the entire volume (V) of the film:

$$F(\vec{P}) = \int (f_{elas} + f_{elec} + f_{grad} + f_{land}) dV$$

Details of the free energies and the simulation parameters can be found in previous literature.⁶⁻⁹

The gradient energy term includes an additional Dzyaloshinskii–Moriya–like (DM-like) interaction, implemented as an antisymmetric coupling between polarization components following Zhao *et al.*²⁹:

$$f_{DMI,i} = -\alpha D_N \frac{\partial P_z}{\partial x_i} + (1 - \alpha) D_B (\nabla \times \mathbf{P})_i.$$

This term is computed at every time step and directly contributes to the polarization dynamics, capturing the intrinsic chiral stabilization mechanism observed experimentally. In this study, the coefficients were set to $D_N = 0.1$ and $D_B = 0.3$, which provide a quantitatively consistent description of the experimentally observed vortex chirality and stability.

Periodic boundary conditions are applied along the in-plane directions, while a superposition method is used for the out-of-plane (z) direction. In this method, the total electrostatic potential is expressed as the superposition of a bulk solution (obtained via 3D Fourier transforms for inhomogeneous polarization) and an analytical plate solution that satisfies the electrostatic boundary conditions along z . The plate solution is derived using 2D Fourier transforms in the x - y plane combined with exponential functions in z , ensuring an accurate and stable treatment of the non-periodic out-of-plane boundaries.

The SrRuO₃ (SRO) bottom electrode is implicitly modeled via a fixed potential boundary condition at the bottom interface, representing a metallic contact. The epitaxial strain from the DyScO₃ (DSO) substrate is introduced through mechanical boundary conditions that fix the in-plane lattice constants while allowing out-of-plane relaxation. In the simulations, the DSO substrate is assigned lattice constants of 3.952 Å (x -direction) and 3.947 Å (y -direction), while the bulk lattice parameters of STO and PTO are 3.905 Å and 3.9574 Å, respectively. This setup produces an effective misfit strain equivalent to the experimental epitaxial environment and introduces a small in-plane anisotropy that helps stabilize the experimentally observed zigzag vortex arrangement.

The simulation setup is modeled using a three-dimensional grid, where each point represents a unit cell. Periodic boundary conditions are applied in the in-plane directions, while a superposition method is used for the out-of-plane direction. For the trilayer sample, the structure consists of 20 unit cells (uc) of SrTiO₃ (STO), 20 uc of PbTiO₃ (PTO), and another 20 uc of STO, all grown on a DyScO₃ (DSO) substrate. The simulation grid is $400 \times 200 \times 120$ $800 \times 200 \times 150$, which includes 30 grid layers for the substrate, 60 layers for the trilayer film, and the remaining layers for air. The grid spacing is set to 0.4 nm. An iterative perturbation method is used to consider the elastic

anisotropy for the PTO and STO layers.³⁰ The normalized time step is set as 0.01 0.02 in this study.”

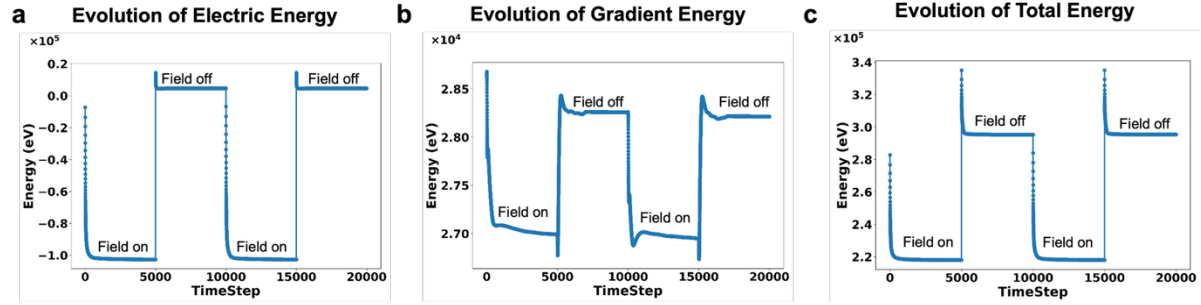


Fig. R2.5| Evolution of electric, gradient and total energies corresponding to the vortex motion. **a**, Time-dependent electric energy during two bias cycles (5 V tip field). **b-c**, Corresponding gradient and total-energy evolution showing field-on stabilization and the post-bias barrier-crossing 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.

Comment 6: For the PTO/STO superlattice sample where vortices coexist with a1/a2 domains, the electric field application only triggers transition of vortex domains, instead of genuine vortex motion. The LPFM signals merely reflect the shift of transition boundaries, and cannot be used to quantify the translational moving distance of vortices.

Response: We thank the reviewer for this insightful comment and fully appreciate the concern raised regarding the distinction between genuine vortex motion and the shift of transition boundaries in the superlattice samples.

Following careful consideration, and consistent with a similar suggestion from Reviewer 1, we have removed the data from the PTO/STO superlattice samples (where vortices coexist with a1/a2

domains). The presence of mixed phases significantly complicates the interpretation of the observed dynamics. To provide a clearer and more definitive demonstration, we now focus exclusively on the trilayer sample, which exhibits a pure vortex phase. This revision results in a much cleaner presentation of the key findings.

We believe these focused revisions, together with the additional simulations and schematics, deliver a clearer message while fully addressing the reviewer's concern. All changes are highlighted in the revised manuscript.

Comment 7: In Figure 2i, the authors applied a 17 V bias using an in-situ tungsten tip on the trilayer sample with a size below 10 nm. According to previous literature, an electric field of such magnitude is strong enough to destroy polar vortex structures. It is confusing why only vortex reconfiguration rather than vortex annihilation was observed in this experiment. Furthermore, the authors do not specify the radius of the tungsten probe. Only initial and final states before and after bias application are presented; the full dynamic evolution process is missing, which makes the experimental evidence incomplete.

Response: We thank the reviewer for this important and insightful remark.

We clarify that the technical details of the tungsten probe used for the in-situ biasing experiments have been provided in the Methods section. The probe has a tip radius below 50 nm, with a typical apex radius of approximately 10 nm. To improve clarity, we have now explicitly reiterated this information in the revised manuscript.

We agree that a nominal bias of 17 V appears relatively high and could, in principle, be expected to destabilize or annihilate polar vortex structures. However, two experimental factors substantially reduce the effective electric field experienced by the ferroelectric layer:

1. The measurements are performed under a floating-ground geometry. Consequently, the applied voltage is not fully dropped across the ferroelectric trilayer in the vertical direction. Instead, the voltage is distributed across the tungsten probe, the sample, and the surrounding electrical circuit. The effective field acting on the polar vortices is therefore considerably lower than that estimated from the nominal applied bias.
2. The sample environments of the two techniques differ markedly. PFM experiments were conducted under ambient conditions, where residual moisture and adsorbates reduce the dielectric gap between the probe and the sample surface, enabling switching at lower voltages (~ 5 V). In contrast, in-situ STEM measurements were carried out under ultrahigh vacuum. As demonstrated by Eom et al. (*Advanced Functional Materials* **35**, 2406944 (2025)), the dielectric environment surrounding the tip strongly influences the effective bias required for polarization switching. Their comparative experiments under distilled water, silicone oil, and ambient air (Fig. R2.6) show that lower-permittivity or vacuum-like conditions increase the effective tunnelling barrier across the dielectric gap and therefore require higher applied voltages. In our UHV STEM geometry the absence of surface adsorbates similarly increases the effective dielectric gap, necessitating a higher nominal voltage (~ 17 V) to achieve an equivalent local field.

Taken together, these factors explain why the nominal 17 V bias induces vortex boundary motion rather than annihilation, and why the apparent voltage difference between PFM and STEM arises naturally from environmental effects rather than from any intrinsic difference in the switching mechanism. Both measurements probe the same underlying ferroelectric response under appropriately scaled local electric fields.

Regarding the dynamic evolution, continuous atomic-resolution imaging during voltage application is extremely challenging in in-situ STEM because of acquisition-speed limitations, specimen drift, and electron-dose constraints. The images shown in Fig. 2i in main revised manuscript therefore represent the equilibrium configurations before and after bias application. The observed structural transformation is, however, fully reproducible across multiple independent experiments.

To avoid any further confusion we have clarified these points in the revised manuscript (pages 7–8). The relevant text now reads:

We have already clarified this difference in the voltage in the manuscript in page 7, which read as,

“The difference in electric-field magnitude observed in PFM and STEM studies arises primarily from differences in the sample environment. Previous work shows that changes in the local environment can modify the effective dielectric gap around the sample, which in turn alters how the applied voltage is distributed.^{26, 27} As the dielectric spacing changes, the same external bias can generate substantially different local electric fields.”

In the revised manuscript, the text below in the Method section clarifies the procedure and tip radius and reads as follows, “***In-situ* biasing TEM.** The samples for *in-situ* experiments were prepared by focused ion beam (FIB) milling. Thin slices were fabricated with the widest faces perpendicular to the [010]_{pc} direction (cross-sectional samples) and the [001]_{pc} direction (plan-view samples).

In-situ TEM experiments were conducted using a PicoFemto electrical holder and control system (Zeptools Co., Ltd). The SrRuO₃ (SRO) layer served as the bottom electrode, while a movable tungsten probe acted as the counter top electrode. The tungsten tip was fabricated via electrochemical etching, with an apex radius typically below 50 nm (and in some cases ~10 nm). The probe movement was controlled by a piezoelectric actuator with a positioning precision within 10 nm.

An electric field was applied between the sharp conductive tungsten tip (of apex 10-50 nm prepared using electrochemical etching) and the SRO conducting layer, which was connected to the holder ground to establish a stable reference potential. The input voltage was supplied through an external Source Measure Unit (SMU). During the entire *in-situ* experiment, the incident electron beam was blocked whenever imaging was not required to minimize possible electron-beam irradiation effects.”

[Figure Redacted]

Fig. R2.6| a–c, Comparative PFM amplitude, phase, and piezoresponse measurements performed in distilled water, silicone oil, and ambient air. Experiments used platinum- and iridium-coated silicon probes pre-dried at 170 °C for 30 min. The results show a clear variation in coercive voltage with changing sample environment, attributed to differences in the effective dielectric gap between the probe and sample surface. Adapted from Eom *et al.* (*Advanced Functional Materials* 35.11 (2025): 2406944)

Comment 8: The authors propose that the coupling between net in-plane polarization and in-plane trailing electric field drives vortex motion. Nevertheless, the electric field applied in the in-situ STEM test (Figure 2i) is along the out-of-plane direction. To validate the proposed mechanism, dedicated in-situ experiments with pure in-plane electric fields are strongly required, which are not provided in this manuscript.

Response: We thank the reviewer for this valuable suggestion and for highlighting the importance of the driving-field geometry.

We agree that a dedicated in-situ STEM experiment with a purely in-plane electric field would provide an additional platform to probe the proposed mechanism. However, we respectfully note that such an experiment is not strictly required to support the conclusions of the present work.

Our proposed mechanism relies on the coupling between the net in-plane polarization of the polar-vortex domains and the *in-plane electric-field component* generated by the local probe. Importantly, because of the small size of the tungsten tip used in the STEM experiments (apex

radius ≈ 10 nm), the applied out-of-plane bias generates a highly nonuniform electric field in the vicinity of the tip. This nonuniformity naturally produces a significant net in-plane field component that can couple to the net in-plane polarization of the polar vortices, thereby driving the observed boundary motion and collective vortex displacement. Our finite-element modeling of the probe-induced field distribution (see revised Fig. R2.7 and supplementary Fig. 4) explicitly demonstrates the presence of this in-plane component under the experimental tip geometry.

In contrast, pure in-plane electric-field experiments do not produce vortex/vortex boundary motion. Behera et al. (*Science Advances* 8, eabj8030 (2022), *Adv. Mater.*, 37, 2410149 (2025)) applied a uniform in-plane electric field using interdigitated electrodes and successfully switched the chirality (handedness) of the polar vortices through changes in their zigzag arrangement. However, they did not observe vortex motion or domain-wall displacement, consistent with in-situ SHG data (Fig. R2.8), which show that uniform in-plane fields primarily switch chirality while leaving domain boundaries fixed. This further supports that the *local, inhomogeneous field* generated by a sharp scanning probe (with both out-of-plane and in-plane components arising from field nonuniformity) is essential for driving the vortex motion reported here.

From a practical standpoint, implementing a purely in-plane electric field in cross-sectional in-situ STEM is technically demanding. The electrode spacing in such geometries is typically several micrometers, requiring voltages of several hundred volts to reach the MV cm^{-1} fields used in our experiments. Such high voltages exceed the safe operating limits of standard in-situ biasing holders and can compromise sample stability and imaging quality. In contrast, the out-of-plane geometry with a sharp tip enables the required local fields (including the critical in-plane component arising from field nonuniformity) with only a few volts, owing to the thin (60 nm) ferroelectric layer. For these reasons, we employed the out-of-plane configuration in STEM, which allows atomic-scale visualization of the vortex evolution under conditions that naturally include the relevant in-plane field component.

We hope these additions fully address the reviewer's concern and convey the underlying physical mechanism more clearly.

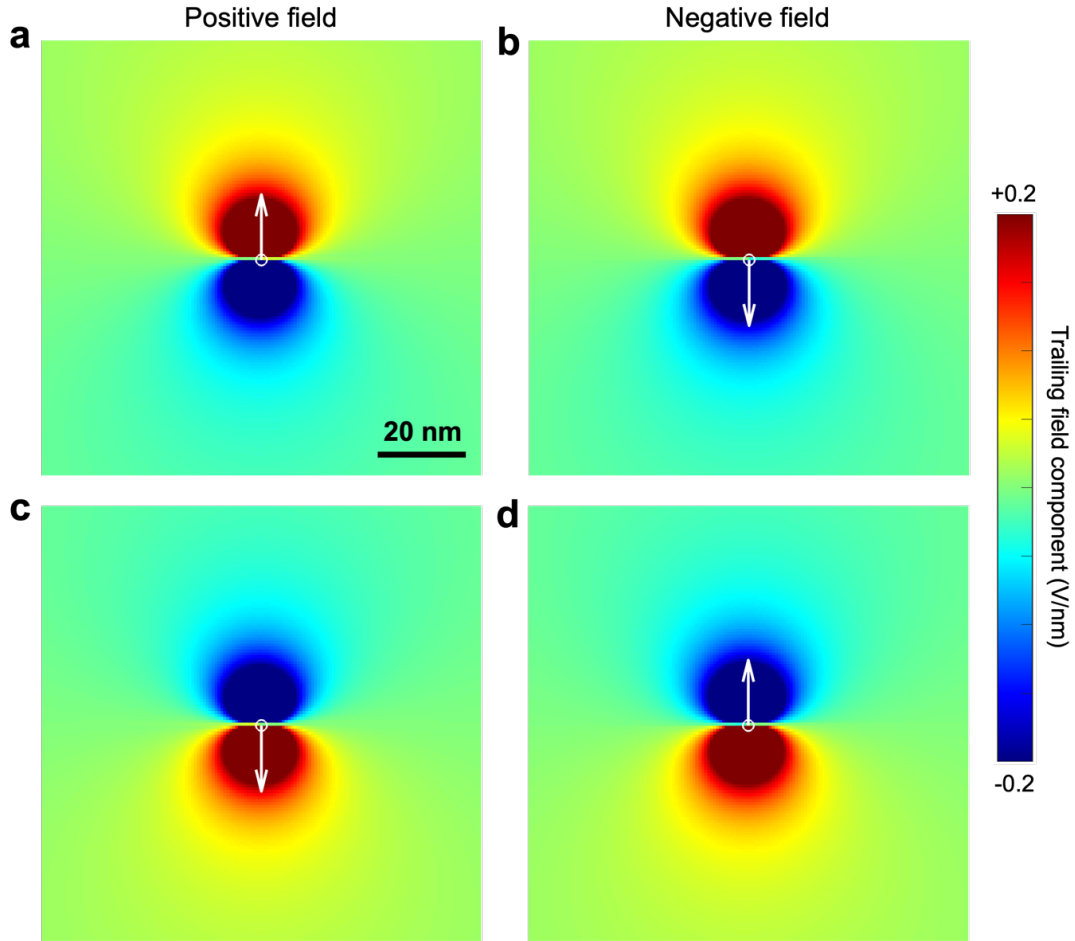


Fig. R2.7| Finite element modelling (FEM) of trailing electric field distributions generated by a moving AFM probe. **a-b,** Calculated trailing field profiles as an AFM probe moves vertically (a: up, b: down) with applied electric bias of +5 V (a) or -5 V (b), respectively. The field distribution couples 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.

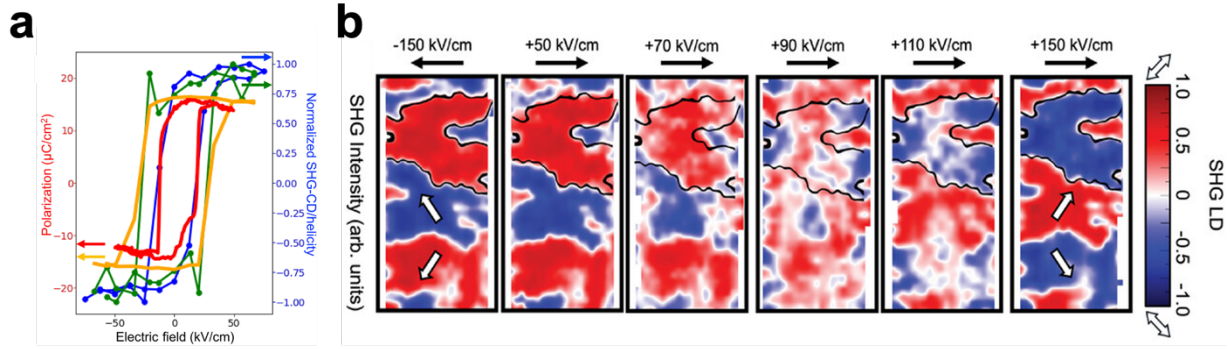


Fig. R2.8 | Electric-field control of chirality of polar vortex and pinned domain wall. a-b, Probing the electric-field-induced change in chirality using second-harmonic generation (SHG). The data show that while an applied electric field can tune the chirality, the domain walls remain pinned (b).

Comment 9: Moreover, this work suffers from problematic data presentation. Readers will struggle to comprehend the five intricate figures composed of numerous sub-panels, which cover two different sample systems simultaneously. The authors must completely revise their data layout to be more succinct and persuasive.

Response: We completely agree with the referee suggestion. We also believe that with the trilayer sample where pure vortex phase has been formed will be good enough to demonstrate the control vortex motion. We have revised the manuscript where only trilayer sample data have been presented, which looks much clean and clear. We believe with all these changes manuscript deliver clear message. The new figures are as following:

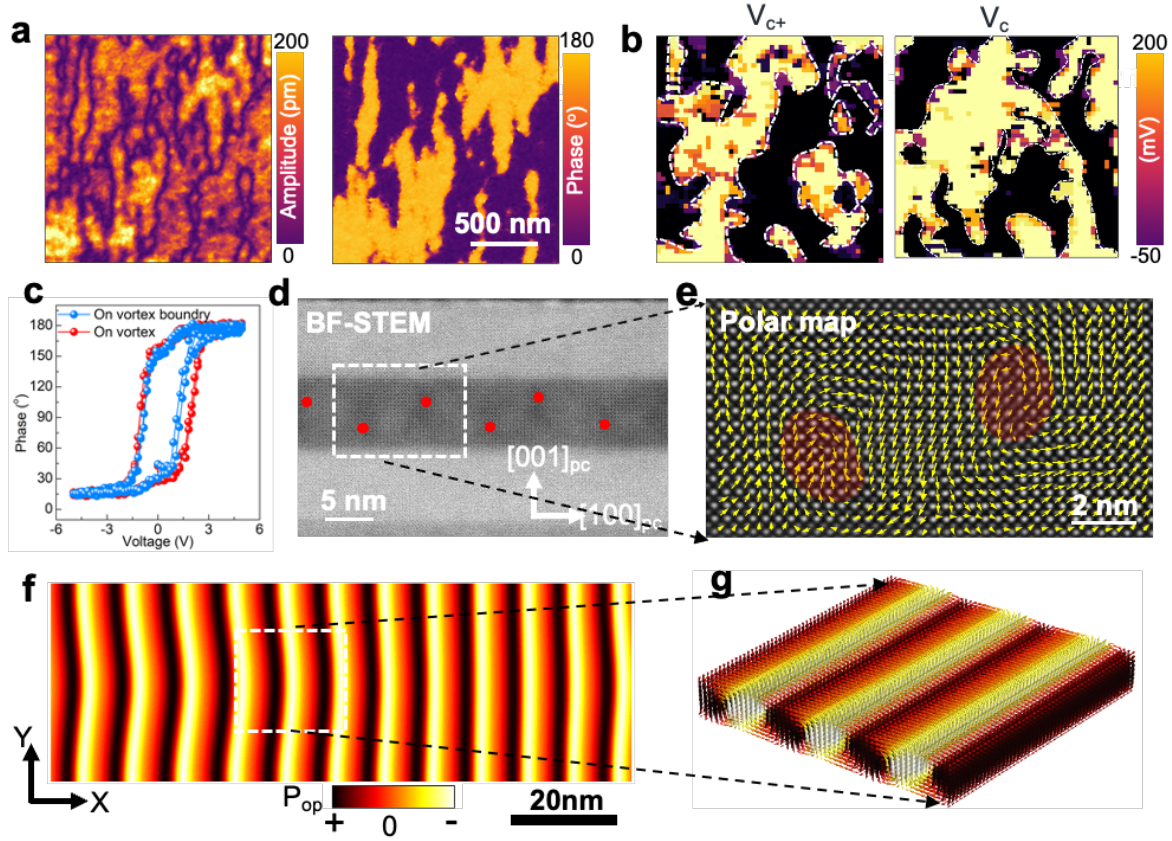


Figure 1| Structure and stabilization of polar vortices in STO/PTO/STO trilayer grown on DyScO_3 (001)_{pc} substrates. **a**, LPMF scan for the trilayer with amplitude (left) and phase (right). The dark contrast in the amplitude represents the boundary of the vortex. **b**, SS-PFM mapping for trilayer reveals spatial distribution of switching behaviour for vortex. The positive nucleation bias (V_{c+} , left panel) showing domains with different polarity having different contrast (marked by white dashed line) and the average coercive bias (V_c , right panel) of same region shows vortex boundary having significantly lower coercive field. **c**, The hysteresis loops extracted from the vortex and near vortex boundary for trilayer illustrating vortex boundary having lower coercive bias compared to the vortex demonstrating vortex boundary motion can be controlled using lower electric field. **d**, Cross-sectional bright field (BF) STEM, and **e**, high-resolution polarization mapping images of white mark region for the trilayer samples show CW and CCW continuous polarization rotation within the PTO layers corresponding to polar structures. Yellow arrows indicate the local polar directions, while red dots denote the vortex cores. **f-g**, The phase field simulated images for the trilayer. The zoomed image shown in the right side is the 3D view of the vortices.

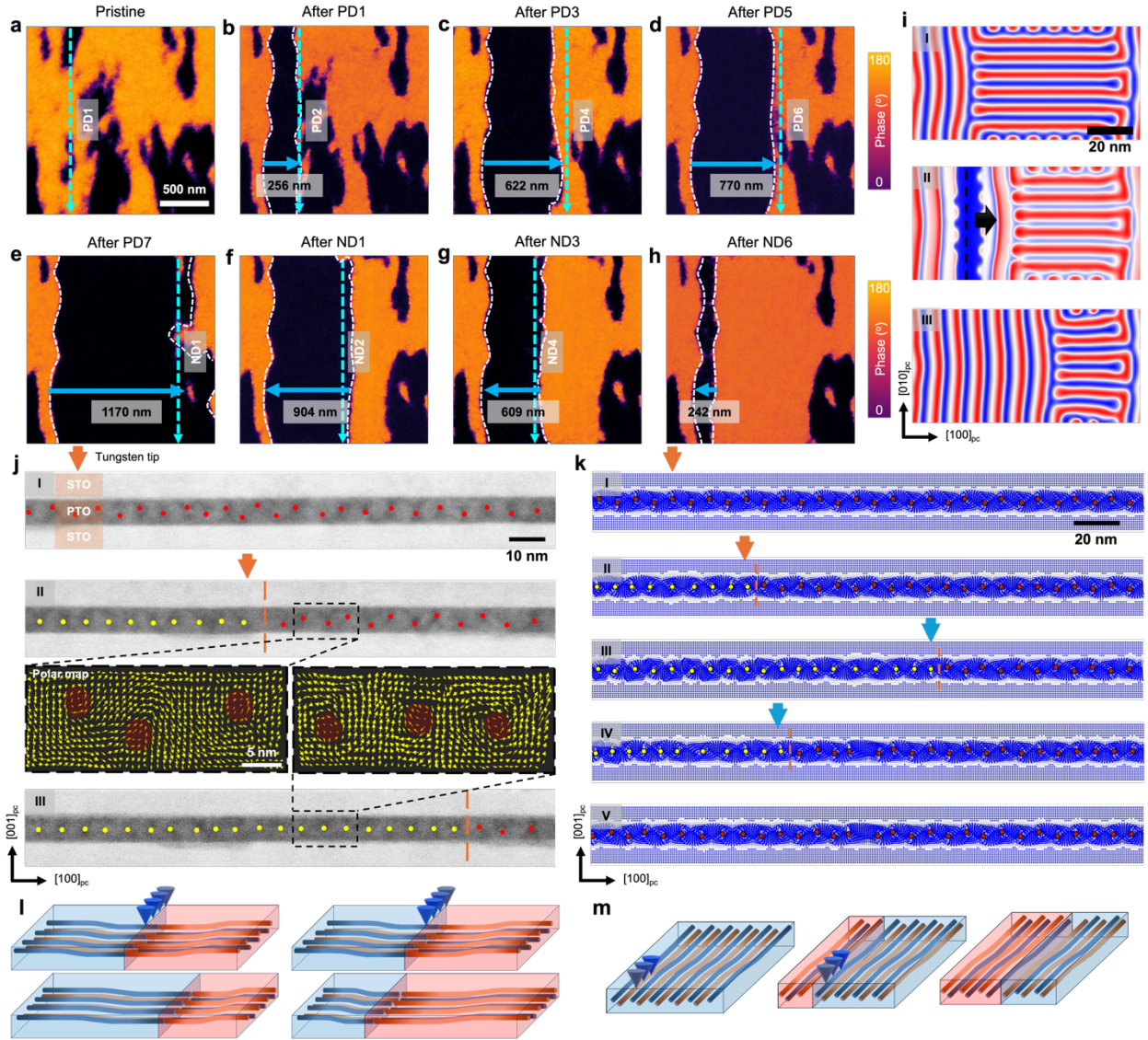


Figure 2|

Figure 2| Deterministic motion of the vortex boundary in the STO/ PTO/ STO trilayer. **a**, Pristine LFM images show well-defined vortex arrays. **b–e**, Successive application of positive trailing fields (PD1–PD7; +5 V) moves the vortex boundary rightward, reaching a total displacement of ~1170 nm. **f–h**, Applying negative trailing fields (ND1–ND6; -5 V) reverses the motion, producing a cumulative leftward movement. **i**, Plane view phase-field simulations illustrating the motion of polar vortex and vortex boundary with application of electric field (I–III). **j**, Cross-sectional *in-situ* STEM confirms the vortex motion (I–III) in the trilayer. Initially, the vortex cores are arranged in an up-down-up configuration (I, red dots). Application of a +17 V electric field via a tungsten tip (denoted by orange arrow) aligns the vortex cores into a near-flat configuration (II, yellow dots), forming a boundary between the up-down arrangement and the near-flat configuration (orange dashed line). A subsequent +17 V application moved the boundary further rightward (III), consistent with the PFM results. Field-induced switching reverses the zigzag vortex arrangement while preserving topological state as shown by polarization mapping

before and after application of electric field (black dashed region). This demonstrates the topologically protected motion of polar vortices during field-induced motion. **k**, Cross-sectional phase-field simulations illustrating the initial polar vortex configuration (I). The orange arrow marks the initial position of the simulated electrode where a positive electric field is applied (I). Upon applying the electric field, the polar vortex shifts to the right and the near-flat region expands (II–III), consistent with in situ STEM observations. To reversibly move the polar vortex, a negative electric field is applied (III–IV; blue arrow). Sequential phase-field simulations further illustrate the reversible motion of the polar vortex (IV–V). **l**, CW and CCW polar vortices are marked in blue and red, respectively. Their zigzag arrangement generates regions with opposite net in-plane polarization. When the AFM probe moves downward under a positive bias, vortices in the blue region expand, shifting the vortex boundary to the right (left panel in l). Reversing the bias reverses the favored vortex expansion, causing vortices in the red region to expand and shifting the boundary to the left (right panel in l). **m**, Motion of the vortex boundary when the electric field is applied perpendicular to the vortex tubes, leading to the rearrangement of the polar-vortex tubes.

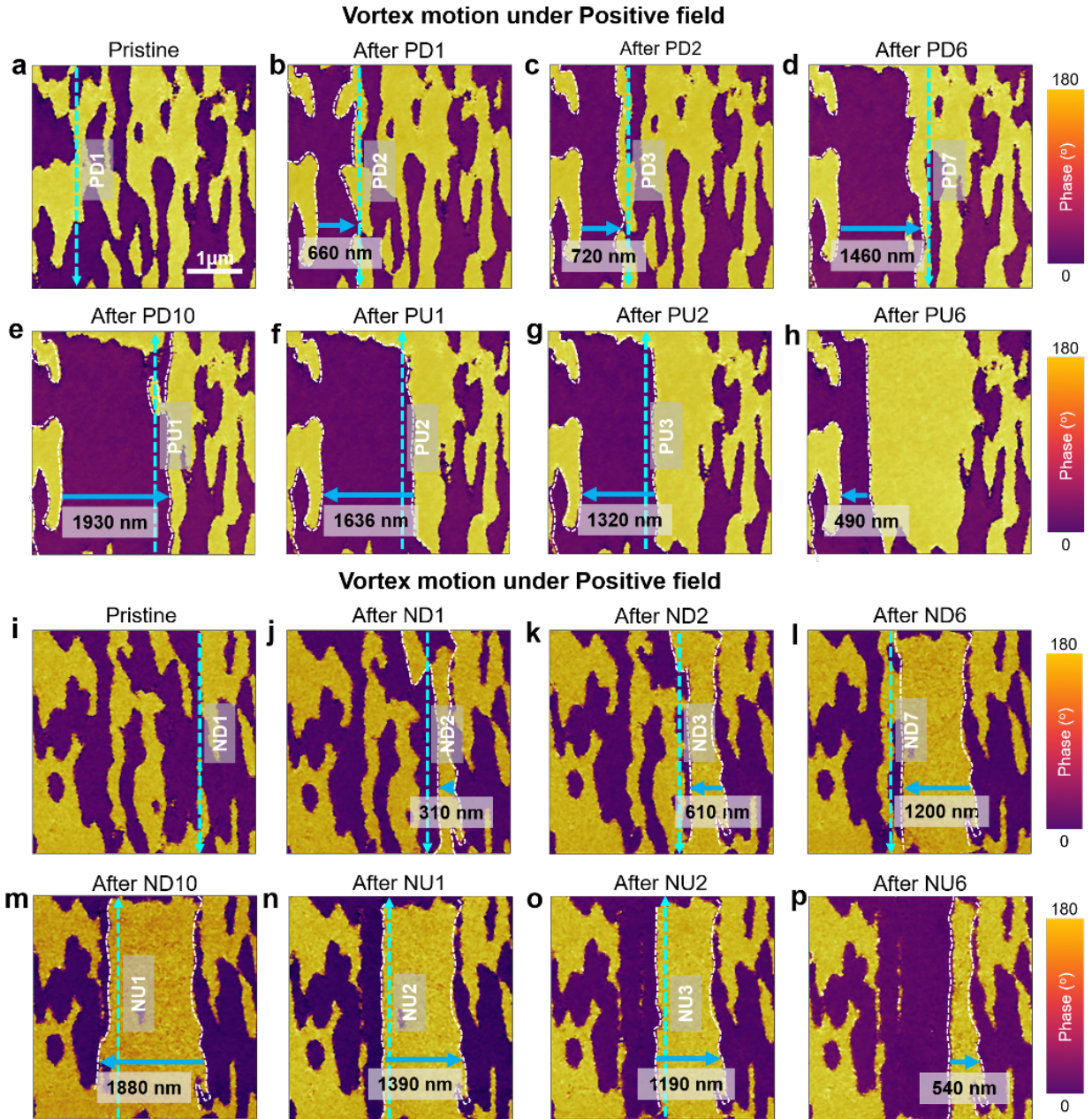


Figure 3| Directional control of the vortex boundary motion in the trilayer. **a**, Pristine vortex state. The cyan arrow indicates the path along which the AFM probe is moved (PD: tip motion up to down with positive polarity of bias). **b**, After the removal of PD1, the PFM scans reveal a distinct rightward shift in the vortex boundary, marked by the blue arrow. **c-e**, Vortex boundary 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 boundary can be moved reversibly i.e. leftward shift. **i**, 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). **j**, After the

removal of ND1, the PFM scans reveal a distinct leftward shift in the vortex boundary, marked by the blue arrow. **k-m**, Shows the vortex boundary movement after ND2-ND10 with total movement of ~1880 nm. **n-p**, 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.

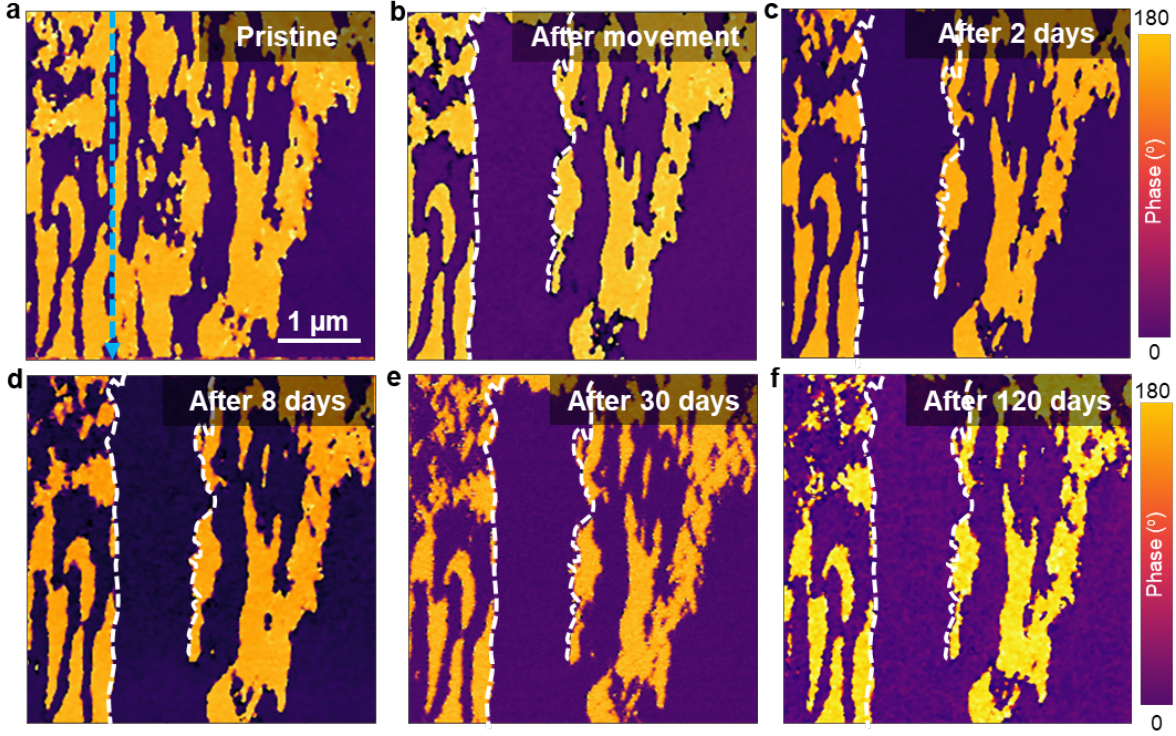


Figure 4| Stability of vortex boundary motion. **a**, Pristine LPFM images for the PTO/STO/PTO trilayer confirm presence of well-defined vortices. Following the application of trailing field from up to down, as described in earlier experiments, the vortex boundary shifted from left to right. Subsequent PFM scans conducted at various intervals reveal the long-term stability of the moved vortex boundary: **c**, after 2 days; **d**, after 8 days; **e**, after 30 days; and **f**, after 120 days. Each scan demonstrates the sustained robustness of the moved vortex boundary over time.

We believe with all these changes manuscript have strengthen and delivering clear message of vortex motion in oxide heterostructure. We hope reviewer will be convince with our efforts and recommend our manuscript for the publication in Nature Communication.