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# Interlocking Antiphase Boundary with 180° Domain Wall in PbTiO<sub>3</sub> – Antiphase Ferroelectric Boundary

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The ferroelectric domain wall, serving as the boundary between separate data carriers based on domains, has attracted widespread interest due to its distinctive physical properties. Although the domain walls in ferroelectric materials are narrower than those in magnetic materials due to their higher lattice anisotropy, they still account for a considerable proportion in ultrathin films, reducing storage efficiency to some extent. Here, ultrathin antiphase ferroelectric boundaries (APFBs) are presented and validated their feasibility as ferroelectric domain walls. The naturally formed APFB shows a sharp and straight morphology, with the characteristic of interlocking between the antiphase boundary (APB) and conventional 180° domain wall. The calculations from the density functional theory demonstrate that the APFBs undergo a significant but localized change in electronic structure. They largely retain the characteristics that are consistent with those of conventional domain walls, such as enhanced conductivity, irregular oxygen vacancy trapping energy, and vacancy-tunable physical properties. Finally, as techniques for precisely controlling the nucleation of APB developing, configurations with out-of-plane APFBs used as dividers may provide a promising strategy for miniaturizing ferroelectric devices.

# 1. Introduction

Ferroelectric materials<sup>[1,2]</sup> are characterized by spontaneous polarizations that can be reversed by an external electric field. By

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utilizing different polarization states, it is possible to achieve bistate or even multistate<sup>[3–5]</sup> storage. In general, the size of domains in ferroelectric films adheres to Kittel's law,<sup>[6,7]</sup> which delineates an empirical relationship indicating that the size of ferroelectric domains decreases with the square root of film thickness decreasing, which makes it possible to achieve ultra-high density storage on a nanometer scale in ferroelectric materials.

The regions separating domains with different polarization orientations are referred to as ferroelectric domain walls. Compared with domains, domain walls possess lower dimensionality and localized symmetry breaking, which exhibits unconventional physical properties<sup>[8–11]</sup> that are different from those displayed by the domain regions, and thus attract significant attention. Despite that domain walls in ferroelectric materials exhibit smaller characteristic widths<sup>[12]</sup> than in magnetic materials due to higher lattice anisotropy, the

widths of domain walls are still in a range of 1–10 nm. In ultrathin films, domain walls can occupy an astonishingly high proportion,<sup>[6]</sup> even up to 50%.<sup>[13]</sup> Moreover, conventional domain walls are susceptible to thermal disturbances, which may lead to storage failure.<sup>[14]</sup> Therefore, the exploration of novel ferroelectric domain walls with narrow characteristic widths and strong resistance to thermal disturbances is critical for improving storage efficiency, and thus providing more design options for ferroelectric materials.

The unique function of defects<sup>[15]</sup> in ferroelectric materials provides infinite possibilities for nanoscale applications, such as polarization retention enhancement,<sup>[16]</sup> ferroelectricity enhancement,<sup>[17]</sup> and domain structure regulation.<sup>[18]</sup> As a common planar defect in perovskite oxides, the main characteristic of antiphase boundaries (APBs) observed under electron microscopy is the displacement of the half-unit cell relative to adjacent regions. However, due to the non-equilibrium characteristics, the location and morphology of APBs are unpredictable,<sup>[19,20]</sup> making it difficult to explain the projection of APBs along the electron beam direction.<sup>[21]</sup> This unpredictability also limits the exploration of the mechanisms and potential applications of APBs.

In this work, we validate the potential of antiphase ferroelectric boundary (APFB) to serve as ferroelectric domain walls.





**Figure 1.** a) A low magnification diffraction contrast dark filed TEM image using (001) g-vector. b) A high magnification HAADF-STEM image with an APB inside the green rectangular box. Atomically resolved EDS mapping around the APB core corresponding to Figure S3b (Supporting Information) for c) Pb, d) Ti, e) O, and d) Pb+Ti+O, respectively. g) Relative EDS intensity of Pb and Ti elements around the APB core. h) EDS intensity of O element around the APB core.

Using scanning transmission electron microscopy (STEM) and density functional theory (DFT) calculations, we demonstrate the interlocking feature of the conventional 180° domain wall and the antiphase boundary in APFB, thereby explaining the feature of straight and sharp morphology. Due to the high lattice anisotropy, the APFBs exhibit a wall thickness that is significantly smaller than that of conventional domain walls. The mutual evolution relationship between APFB and the conventional 180° domain wall is demonstrated by STEM, and the polarization switching process of the domains separated by APFB is verified by DFT calculations. A phenomenological explanation is built for the formation mechanism of the APFB and the polarization rotation near the APFB is also elucidated based on the flexoelectric effect. It is found that AFPB retains a high degree of similarity to conventional domain walls, such as the enhanced conductivity, the irregular vacancy trapping energy, and the vacancy-tunable electrical transport properties and magnetic properties.<sup>[22]</sup> These results of the APFB provide a possibility for novel design of ferroelectric materials.

#### 2. Results

#### 2.1. Morphological Characterization of APFB

We grew PbTiO<sub>3</sub> films with a thickness of 25 nm on the pseudocubic (001)<sub>PC</sub> LaAlO<sub>3</sub> substrate, with a mismatch of  $\approx$ 3% (Table S1, Supporting Information). The selected area electron diffraction (SAED) and X-ray diffraction (XRD) pattern (Figures S1a and

S2, Supporting Information) shows the high crystalline quality of the film. The dark-field image in **Figure 1**a shows the typical a/cdomain configuration, which is also verified in the SAED pattern shown in Figure S1(b,c) (Supporting Information). In addition, we can also observe some localized dark stripes perpendicular to the interface, which do not satisfy the diffraction conditions in (001) (Figure 1a) nor (010) (Figure S3a, Supporting Information). To further investigate the origin of these dark stripes, we performed STEM high-angle annular dark-field (HAADF) imaging in the region marked by the white dashed rectangle in Figure 1a. As shown in Figure 1b, the dark stripes originate from an outof-plane APB configuration, characterized by a sharp interface and a half-unit cell shift relative to adjacent regions. After multiple observations, it is found that the configuration of APBs is not unique, with most of them being Type-I and Type-II APBs (Figure S3c,d, Supporting Information), which is similar to the findings in in-plane charged APBs.<sup>[23]</sup> The characteristic of Type-I APBs is a relative displacement of c/2 between separated domains, with the core consisting of a Pb-O bilayer, whereas the core of Type-II APBs consists of a Pb-O tetralayer. (Please note that Type-II APBs may also arise from Type-I APBs propagating along [100], but with a bend occurring in the [010] direction (Figure S4, Supporting Information)). We performed energy dispersive X-ray spectroscopy (EDS) mapping near the core of the Type-I APB as shown in Figure S3b (Supporting Information), and the results are exhibited in Figure 1c-f. The characteristic half-unit-cell shifts on both sides of the APB are clearly visible. In addition, as shown in Figure 1g,h, compared with adjacent



**Figure 2.** a) A high magnification HAADF-STEM image with an overlay of the polar vectors, while a Type-II APFB is indicated by a blue arrow. In order to show the comparison between APFB and conventional domain walls, the region of the coexistence of both domain walls is chosen, while a region with pure APFB domain wall is displayed in Figure S12d (Supporting Information). b) The spatial distribution of the polar displacement along the *c* direction near the Type-II APFB. c) A high magnification ABF-STEM image of a Type-I APFB. d) The relative energy of APFB in different step lengths with the configurations of up-down, up-up, head-tail, and head-head, respectively (These calculations were also performed on SrTiO<sub>3</sub>). The sketches of various configurations are inserted in panel (e). e) NEB energy path diagram for the state switches from up-up to up-down and head-tail to head-head.

domains, Pb enrichment, and Ti deficiency occur around the APB core, accompanied by oxygen vacancy enrichment with a characteristic length of  $\approx$ 2 nm.

In perovskite materials, APB is a common type of planar defect, they can be induced by various mechanisms, including non-stoichiometric growth,<sup>[24,25]</sup> stress,<sup>[26,27]</sup> and specific interface design.<sup>[28]</sup> Despite the differences in formation mechanisms, the nucleation sites and morphology<sup>[20,23,28,29]</sup> of APBs are largely random due to their non-equilibrium features, leading to the coexistence of a wide variety of irregular and multidirectional APBs. In the LaAlO<sub>3</sub>/PbTiO<sub>3</sub> system, the out-of-plane APBs indeed exhibit positional uncertainty. However, what is unusual is their highly uniform morphology, characterized by sharp and straight interface.

To understand the unique APB morphology in the LaAlO<sub>3</sub>/PbTiO<sub>3</sub> system, we first examined the polar behaviors on both sides of the out-of-plane APB. The polar structure is determined by analyzing the localized noncentrosymmetric of the atomic columns between the Pb and Ti sublattices in the HAADF image, as is often done in the literature.<sup>[30,31]</sup> **Figure 2a**,b show the spatial distribution of the polar phase and amplitude for the Type-II domain wall, respectively. Sur-

prisingly, it is found that the polar amplitudes on both sides of the APB are similar, but the phases are opposite. This characteristic is also applicable to the Type-I APB, as demonstrated by the annular bright-field (ABF) image in Figure 2c, where the Ti-O octahedra on either side of the APB can be clearly seen to move in opposite directions. In other words, the crystallographic APB simultaneously acts as a conventional 180° domain wall, which can be referred to as the APFB. The interlocking feature of APB and ferroelectric domain wall is a fundamental characteristic in improper hexagonal manganite and ferrite ferroelectric material.[32,33] According to this interlocking feature, we constructed APFB configurations with varying numbers of steps as shown in Figure 2d. It can be seen that the formation energy of the stepped configurations with opposite polarizations across the APFB is significantly higher than that of other configurations. This is attributed to the formation of the charged wall at the step, which is generally considered to have a high energy cost. Therefore, the interlocking between APB and ferroelectric domain wall is the key reason for the sharp and straight morphology. (The in-plane propagation of APFB might exhibit distinct characteristics compared with out-of-plane counterparts. These patterns are partially

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discussed in Supplementary Information (Figure S5, Supporting Information).

Due to the higher lattice anisotropy of ferroelectric domain walls than that of magnetic domain walls, the characteristic lengths of ferroelectric domain walls, which are believed to be in a range of 1–10 nm,<sup>[6,34]</sup> are approximately one-tenth smaller than those of the magnet. In the LaAlO<sub>3</sub>/PbTiO<sub>3</sub> system with a conventional 180° domain wall, the width is  $\approx$ 3 nm (Figure S6, Supporting Information). The width and density of conventional domain walls are constrained by Kittel's law. As a rearranged term of Kittle's law,<sup>[6,7]</sup>  $\delta/w = \sqrt{\delta/Gd}$  can be regarded as the proportion of the domain wall in a film, where  $\delta$ , w, G, and d denote the domain wall width, domain width, adimensional parameter, and film thickness, respectively. It is foreseeable that as the thickness of the film decreases, the proportion of the domain wall in the film increases. In films with a thickness of 100 nm, this proportion can reach 6–20%,<sup>[6]</sup> and even 50% in ultrathin films.<sup>[13]</sup> However, it seems that APFB can break through the limitation of Kittel's law, allowing for a narrower wall width. The width of the Type-II domain wall is 6.1 Å, while the width of the Type-I domain wall is as narrow as 2.3 Å (Figure S3e, Supporting Information), exhibiting a sub-unit cell thickness, which is only one-tenth of the width of a conventional 180° domain wall. Moreover, due to the higher lattice anisotropy of APFB, it exhibits a significant pinning characteristic, specifically, the property of resistance to thermal disturbance. This implies that the APFB holds the potential for achieving higher domain utilization efficiency and, thus storage efficiency, without compromising domain wall density, thereby enabling the miniaturization of ferroelectric devices.

# 2.2. Ferroelectric Switching Process of Domain Separated by APFB

To verify whether polarization switching can occur in the domain regions separated by APFBs, we performed box-in-box pattern writing in PFM with reversed DC bias. Although distinguishing APFBs from conventional out-of-phase 180° domain walls is challenging in PFM phase image, it can be observed that the polarization in the domains they separate has both undergone switching (Figures S7 and S8, Supporting Information). To verify the feasibility of the rewritten mechanism separated by APFB at the atomic scale, we constructed four possible APFB models based on the lattice constraints imposed by the APB. According to the polar directions on either side of the APFB, we defined four configurations: up-up (down-down), up-down (down-up), headhead (tail-tail), and head-tail (tail-head). The schematic diagrams of these four configurations are shown in the insets of Figure 2e. Among them, the up-up and up-down configurations are referred to as c-APFB where the polar vector is perpendicular to the interface, while the head-head and head-tail configurations are referred to as *a*-APFB where the polar vector is parallel to the interface. We performed nudged elastic band (NEB) calculations for polar switching in *c*-APFB and *a*-APFB. As for *c*-APFB, it is found that the up-down configuration is more energetically favorable, as it could reduce the depolarization energy to some extent. This explains the observed interlocking feature between the ferroelectric domain wall and APB. The configuration of up-up is a metastable state, with its energy being 8 meV atom<sup>-1</sup> higher than that of the up-down state. The switching barrier between the up-up and up-down states is  $\approx 90$  meV atom<sup>-1</sup>. (Only half of the supercell participated in the polarization switching). This means that the data storage can be achieved by switching between different domains separated by APFB. Compared with the conventional domain wall in the bulk<sup>[35]</sup> (41 meV atom<sup>-1</sup>), the switching barrier of the domain based on APFB increases, indicating that APFB imposes a pinning effect on polarization switching. For the *a*-APFB, their energies are generally higher than that of the c-APFB, and the charged head-head configuration is unstable. In our experiments, we also rarely observed the natural formation of pure *a*-APFB. However, interestingly, when a domain crosses an APFB, it forms a localized a-APFB. In this configuration, to prevent the formation of 90° charged domain walls, the tail-tail charged a-APFB with the energetically less favorable tends to form (Figure S9, Supporting Information), causing the APFB to simultaneously act as both a tail-tail charged a-APFB and an up-down neutral *c*-APFB. Moreover, the APFB is annihilated into a conventional 180° domain after interacting with a defect, which deserves to be further investigated.

#### 2.3. The Formation Mechanism of APFB

To reproduce APFBs in other materials, we need to clarify the formation mechanism of APFBs. We conduct HAADF observations of the APFB near the interface. As shown in **Figure 3**a,b, the two sides of the APFB correspond to two different epitaxial modes. On the left part of APFB, as shown in Figure 3a, which is in the conventional epitaxy mode, the LaO-terminated surface (Figure S10, Supporting Information) carries a positive charge, resulting in an energy preference for upward polarization in the epitaxial PbTiO<sub>3</sub>. On the right part, a magnified image is shown in Figure 3b, where the epitaxy is accompanied by a head-head in-plane charged APB (ICAPB). This is strikingly similar to the ICAPB observed in BiFeO<sub>3</sub>.<sup>[23]</sup> Although charged domain walls are generally considered to be unstable due to their high energy cost, the formation of ICAPB in PbTiO<sub>3</sub> seems reasonable. On the one hand, the LaO-terminated surface favors an upward polarization state. On the other hand, due to the energy preference for up-down configuration across the APFB, a downward polarization state on the right part is enforced. This creates frustration in the in-plane APB region, promoting the formation of ICAPB. And, the ICAPB in the right part results in the loss of an atomic row along the *c*-axis, providing crystallographic conditions conducive to the formation of the out-of-plane APFB.

Additionally, for the *c*-domain PbTiO<sub>3</sub> epitaxy on LaAlO<sub>3</sub>, the nominal compressive strain reaches  $s = \frac{a_{PTO} - a_{LAO}}{(a_{LAO} + a_{PTO})/2} \times 100\% = 3.4\%$ . In general, compressive strain is unfavorable for forming *a*-domains because it exacerbates lattice mismatch. However, previous experiments<sup>[36]</sup> confirmed that the dislocations or other defects can trigger the nucleation of *a*-domains. This phenomenon is also observed in LaAlO<sub>3</sub>/PbTiO<sub>3</sub> (Figure S11, Supporting Information). Therefore, the increase of elastic energy induced by intrinsic compressive strain and *a*-domains provides energetically favorable conditions for the formation of APFB.

Due to the presence of various boundary constraints, such as dislocations, APFBs, and ICAPBs, the strain near the APFB is





**Figure 3.** a) A high magnification HAADF-STEM image, where the blue arrow indicates a Type-II APFB and the white arrow indicates the polar direction. b) The corresponding magnified image of the white rectangular region in panel (a), overlayed with an atomic model schematic. Corresponding spatial distributions of lattice constants c) *a* and d) *c* of panel (a). Black stripes are missing values due to huge structure changes at the defect cores. e) The statistical results of lattice constants *a* and *c*, where the statistical directions are indicated in panels (c) and (d). f) The polar diagram of the phase and amplitude in PbTiO<sub>3</sub> near the APFB.

highly non-uniform as shown in Figure 3c,d. The different epitaxial modes on either side of the APFB create a strain gradient in the in-plane direction. Additionally, due to the joint effects of the interface, defects, and strain relaxation, a gradient also exists in the out-of-plane direction as shown in Figure 3e. Consequently, the in-plane strain gradient contributes a significant in-plane polarization component due to the flexoelectric effect. As a result, the statistical polar plot in Figure 3f shows that the polarizations on both sides of the APFB are not strictly antiparallel but present an angle of  $\approx 150^{\circ}$ .

#### 2.4. The Change in the Electronic Structure of APFB

The change in crystal structure often drives the change in electronic structure, which is a key aspect of interface and surface engineering.<sup>[37–39]</sup> To explore the influence of APFBs on the electronic properties, we performed the electronic structure calculations of the APFB core marked in Figure 4a. The plots of the charge density difference map in Figure 4(b1-e1) evidently show significant charge redistributions near the cores of APFBs, specifically, Pb-O bonding across the APFBs. We integrate the charge transfer along the [100] direction and summarize it in Figure S12b (Supporting Information). It can be seen that the charge transfer is most pronounced in the *a*-APFB configuration. However, despite the intense charge redistribution, its spatial scale is extremely localized, occurring only within a 10 Å range around the core. Therefore, it is confirmed that APFBs show a significant but localized change in electronic structure. This also means that even if the spacing between two APFBs is close, there will be

no significant interaction which can lead to device failure. Currently, the smallest spacing between two naturally formed APFBs is  $\approx$ 15 nm as shown in Figure S12c (Supporting Information), and the APFBs maintain their morphological features.

Structural differences also often lead to shifts in the band gap, resulting in unconventional conductivity at domain walls. The unusual electrical transport properties of ferroelectric domain walls demonstrate the feasibility of developing domainwall-based electronic devices.<sup>[3,8,40]</sup> Therefore, we also calculated the density of states (DOS) at the cores of different APFB configurations (Figure 4(b2-e2)). The results show that the main contribution near the Fermi level comes from the hybridization of Pb 6s and O 2p orbitals (Figure S13, Supporting Information). The band gaps of the four APFB configurations shift due to electronic reconstruction at the domain walls. The band gaps for the up-down, up-up, and head-tail APFB configurations are 2.50, 2.47, and 1.61 eV, respectively, which are significantly lower than the 2.84 eV of bulk configuration (Figure S14, Supporting Information), even the head-head APFB configuration acts as a conductive domain wall. This further confirms the strong interactions at the cores of the domain walls. Unfortunately, despite the dramatic changes in the crystal structure, the Ti atoms essentially retain the original  $3d^0$  configuration. In the DOS, we did not observe any spin polarizations in the four types of APFBs. Thus, like conventional domain walls, the APFB is non-magnetic.

Oxygen vacancies are the most common and abundant defects in oxide materials, often inducing a variety of interesting phenomena. Conventional ferroelectric domain walls exhibit the ability to trap oxygen vacancies, thereby enabling the modulating of the physical properties<sup>[41–43]</sup> of the domain walls.

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**Figure 4.** a) A schematic diagram of the atomic model for the electronic structure calculations of APFB. b1-e1) Plots of charge density difference ( $\Delta \rho = \rho_{APFB} - \rho_{left} - \rho_{right}$ ) and b2-e2) the corresponding DOS of the four APFB configurations, where the calculation region is marked with black dashed rectangle in panel (a).

Therefore, we calculated the formation energy of oxygen vacancy near the APFB for the model shown in Figure S12a (Supporting Information). As illustrated in Figure 5a, the formation energy of oxygen vacancy at the core of APFBs shows a significant decrease compared with that in the domain region. The values of vacancy trapping energies  $E_{trap} (E_{trap} = E(V_O^{domain}) - E(V_O^{APFB}))$  for the up-down, up-up, head-tail, head-head and tail-tail APFB configurations at the APFB cores are 0.6, 0.7, 1.3, 1.4, and 3.5 eV, respectively, which are considerably higher than those of conventional 90° and 180° domain walls (on the order of 0.3 eV.<sup>[22,44,45]</sup>) This indicates that the APFBs have a higher capacity for trapping vacancies. The effect of APFBs on the formation energy of oxygen vacancy is localized to a spatial scale of  $\approx 10$  Å near the APFB core, consistent with the size of the vacancy region observed in EDS mapping (Figure 1h). The presence of oxygen vacancies leads the defect states to occur near the Fermi level, thereby enhancing its conductivity ((Figure 5b, and the enlarged plot is shown in Figure S15, Supporting Information). Moreover, for *a*-APFBs, the spin polarization results in a magnetic spin moment of  $\approx 0.27$ , 1.37 µB/APFB for the head-tail and head-head configurations, respectively. This suggests that there is the potential to exploit the selflocalization of oxygen vacancies at the APFB cores to modulate the electrical and magnetic properties.

#### 3. Conclusion

We have identified an out-of-plane APFB in the LaAlO<sub>3</sub>/PbTiO<sub>3</sub> system, which is characterized by random nucleation sites but sharp and straight morphology, with interlocking APB and ferroelectric domain wall. The DFT calculations confirm the energy preference of *c*-APFB with the up-down configuration. It is proposed that different epitaxial modes and intrinsic high lattice mismatch lead to the formation of APFB, and due to the effects of

abundant general defects, the polar vectors near the APFB deviate from the antiparallel state because of the flexoelectric effect. Through DFT calculations, we predict the changes in bonding, band gap, magnetic property, and formation energy of oxygen vacancy at the APFB core, revealing a strong but localized influence. Furthermore, with the advancement of techniques for precisely controlling the nucleation sites of APBs, such as substrate with interface design,<sup>[28]</sup> substrate with miscut angle,<sup>[46]</sup> and ion implantation annealing,<sup>[23]</sup> the configurations with out-of-plane APFBs serving as dividers may provide a promising strategy for miniaturizing devices, due to their unique morphology, editability, and high storage efficiency.

#### 4. Experimental Section

Substrate Preparation and Thin-Film Growth: Pulsed laser deposition (PLD) was used for preparing PbTiO<sub>3</sub> films, with a KrF excimer laser ( $\lambda$  = 248 nm) with 1.5 J cm<sup>-2</sup> fluence and 2 Hz repetition rate to ablate a ceramic PbTiO<sub>3</sub> target. LaAlO<sub>3</sub> was adopted as the substrate for epitaxy of PbTiO<sub>3</sub>, with a lattice mismatch of about 3%. The setpoint temperature of the substrate holder was 700 °C. The oxygen partial pressure, p(O<sub>2</sub>), was 10 Pa. After growth, the films were cooled down to room temperature in the same p(O<sub>2</sub>) at a rate of 10 °C min<sup>-1</sup>.

STEM Observation and Analysis: The cross-sectional samples of the PbTiO<sub>3</sub> thin film were prepared by FEI Helios focused ion beam. Using special aberration-corrected JEOL ARM200F, we collected the HAADF images and EDS mappings of PbTiO<sub>3</sub> films along the [100] direction, under 200 kV accelerating voltage with  $\approx$ 25 mrad convergence semi-angle. The collection semi-angles for the ADF signal were set to 70–350 mrad. Peak pairs analysis was employed to analyze the positions of the atomic columns in the HAADF images.

DFT Calculations: VASP<sup>[47–49]</sup> was used to relax the crystal structure and obtain the electronic structure of APFBs in PbTiO<sub>3</sub>, and the PAW-PBE<sup>[50]</sup> method was used in the calculations. To ensure the rationality of the analysis, the cutoff energy of the plane wave was selected as 500 eV,





**Figure 5.** a) Position dependence of the relative formation energy of an oxygen vacancy around APFB, with a schematic diagram of two inequivalent atomic positions of O in the  $TiO_2$  plane. b) The corresponding DOS of the four APFB configurations.

and the energy convergence criterion was chosen to be  $10^{-6}$  eV. The K-space sampling point of  $0.04*2\pi/\forall$  was selected during the structural optimization and  $0.03*2\pi/\forall$  was adopted for calculating the DOS. Some results were visualized using the VASPKIT<sup>[51]</sup> software.

To model the AFPB and satisfy the periodic boundary condition, a  $1 \times 15 \times 1$  supercell containing 74 atoms was used, including two APFB cores. According to the periodic boundary condition, the APFB cores extend in the out-of-plane [001] direction and the in-plane [100] direction. Since the unit cells in the middle of two APFB cores have little distortion, we can thus infer that the interaction between the two APFB cores is negligible.

To evaluate the energy associated with forming stairs in the out-of-plane direction, a  $1 \times 14 \times 4$  supercell containing 276 atoms was used, including two APFB cores. The model was constructed based on TEM characterization reported in previous literature.<sup>[28,52]</sup> The APFB cores also extend in the out-of-plane [001] direction and the in-plane [100] direction, with the slip length of a single step being the lattice parameter *a* along the [010] direction.

# **Supporting Information**

Supporting Information is available from the Wiley Online Library or from the author.

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# **Conflict of Interest**

The authors declare no conflict of interest.

# **Data Availability Statement**

The data that support the findings of this study are available from the corresponding author upon reasonable request.

### **Keywords**

antiphase boundary, domain wall, electronic structure, ferroelectric material

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