

Electromechanical properties of the 180° domain wall in PbTiO₃

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We analyze the electromechanical response of the 180° ferroelectric domain wall in tetragonal PbTiO₃ by combining first-principles calculations with a Landau-Ginzburg-Devonshire (LGD) description. Using regular multidomain structures with varying domain-wall density, we extract polarization profiles and lattice distortions and map them onto the continuum model to determine conventional (homogeneous) and gradient (inhomogeneous) electrostriction. Conventional electrostriction yields only a small negative length change of the sample, whereas gradient electrostriction—arising from the coupling between strain and polarization gradients—produces a positive contribution nearly an order of magnitude larger and localized at the wall core. Our results demonstrate that gradient electrostriction dominates the electromechanical response of 180° walls in PbTiO₃, supporting its inclusion in LGD models that stabilize Bloch-type domain wall structures.

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I. INTRODUCTION

The tensorial properties of domain walls (DWs) in ferroic materials have attracted growing interest owing to experimental and computational advances that enable the fabrication and characterization of submicron and nanoscale structures. A broad spectrum of theoretical approaches is available to describe DWs [1], including first-principles calculations [2], machine-learned force fields [3], phase-field modeling [4], and phenomenological Landau-Ginzburg-Devonshire (LGD) theory [5]. These methods are closely connected to symmetry-based analyses using layer groups, which provide a rigorous framework for classifying DW structures and their associated tensor properties [6–10].

Local polarization confined to DWs has been theoretically predicted in several perovskite systems [11,12]. Such DW-localized order can arise from couplings including flexoelectricity [13], rotapolar interactions [8,14], and biquadratic couplings between primary and secondary order parameters [15]. These mechanisms highlight the intricate interplay between structural distortions, polarization gradients, and strain at ferroic boundaries.

In PbTiO₃ (PTO), the possible existence and structure of a polar 180° DW have been extensively discussed, but the microscopic nature of the wall remains under debate. Several *ab initio* studies have reported a stable Bloch-like configuration with switchable local polarization and reduced macroscopic

symmetry compared to the nonpolar Ising-type wall [2,16–19]. This suggests that the transformation between Ising and Bloch DWs should be describable within the LGD framework. However, within conventional LGD models that include only standard homogeneous electrostriction, the Bloch configuration was found to be unstable [17,19,20]. To resolve this discrepancy, the LGD free energy was extended to incorporate a coupling between strain and polarization gradients of the form $e(\partial P/\partial x)^2$, referred to as gradient [21] or inhomogeneous electrostriction [20]. While the usual electrostriction term dominates inside the domains, the gradient coupling becomes significant within the DW and enables stabilization of the Bloch-like configuration in PTO [20].

In this work, we employ *ab initio* calculations to analyze the electromechanical properties of the 180° DW in PbTiO₃ and to quantify the role of inhomogeneous electrostriction. We construct a periodic equidistant array of 180° DWs, extract layer-resolved polarization profiles and DW-induced lattice distortions, and map the results onto a continuum LGD description of electrostriction. This allows us to (i) estimate the effective tensor combination describing gradient electrostriction, (ii) separate the homogeneous and inhomogeneous electrostriction contributions to the DW-induced strain, and (iii) demonstrate that gradient electrostriction provides the dominant contribution to the longitudinal elongation localized at the DW core.

II. ARRAY OF 180° DOMAIN WALLS

PbTiO₃ undergoes a uniaxial ferroelectric phase transition from the cubic to the tetragonal structure without multiplication of the unit cell. The symmetry reduction from *Pm3m* to *P4mm* yields six tetragonal domain states (DSs): $1_1 \equiv (-P_s, 0, 0)$, $2_1 \equiv (0, -P_s, 0)$, $3_1 \equiv (0, 0, -P_s)$, and their counterparts $1_2, 2_2, 3_2$ with opposite polarization.

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We focus on the 180° DW ($3_1 | \mathbf{n}, \mathbf{p} | 3_2$) separating the DSs $3_1 \equiv (0, 0, -P_s)$ and $3_2 \equiv (0, 0, P_s)$, with wall normal $\mathbf{n} = [1, 0, 0]$. The microscopic position $\mathbf{p}$ within the unit cell [8,9] is energetically favored at the Pb site, at least at low temperature. In the LGD description such microscopic detail is not resolved, and the layer-group symmetry of the DW twin ($3_1 | \mathbf{n} | 3_2$) is the four-element group

$$T_{12} = \mathbf{T}\{1, m_y, 2_y, \bar{1}\}, \quad (1)$$

where $\mathbf{T}$ denotes translations parallel to the DW plane [9,20]. This symmetry enforces an antisymmetric Néel component, $P_1(x) = -P_1(-x)$, which may remain nonzero throughout the entire temperature range below T_c . In contrast, the Bloch component is forbidden by symmetry, since the operation m_y yields $P_2(x) = -P_2(x) = 0$. A nonzero Bloch component can therefore arise only after a symmetry-lowering transition to

$$T'_{12} = \mathbf{T}\{1, 2_y\}, \quad (2)$$

which allows a symmetric and switchable component $P_2(x) = P_2(-x) \neq 0$.

Two distinct polarization structures may thus occur in the wall: (i) an Ising-like (Ising-Néel) profile ($P_1(x), 0, P_3(x)$) with $P_2 = 0$, and (ii) a Bloch-like profile ($P_1(x), P_2(x), P_3(x)$) with a nonzero, symmetric, and switchable Bloch component $P_2(x)$. The Néel component $P_1(x)$, although always present, is antisymmetric, nonswitchable, and typically very small due to internal electric fields, and may often be neglected.

The typical profiles are well represented by hyperbolic functions ($P_1 \approx 0$):

$$\begin{aligned} P_3(x) &= P_s \tanh(kx), \\ P_2(x) &= P_B \cosh^{-1}(kx), \end{aligned} \quad (3)$$

where P_s is the spontaneous polarization, P_B is the amplitude of the Bloch component, $k = 2/\xi$, and ξ is the DW thickness. The values of P_s , P_B , and k are determined from *ab initio* calculations.

Because the VASP package (see Appendix) enforces periodic boundary conditions, the system must be represented by a periodic model. We therefore employ a supercell containing two symmetry-related, energetically equivalent DW variants. The supercell is periodically repeated along the x axis, producing an infinite, equidistant array of domain walls.

The supercell comprises n unit cells arranged as $n \times 1 \times 1$, corresponding to a periodicity of n unit cells along the x axis and a single unit cell along the y and z axes. The separation between adjacent DWs is thus $n/2$ unit cells.

In real-space units, the tetragonal homogeneous (DW-free) reference structure has lattice dimensions $L \times a_t \times c_t$, where $L = na_t$. Introducing DWs modifies the supercell dimensions to $(L + \Delta L) \times b \times c$, with $b = a_t + \Delta b$ and $c = c_t + \Delta c$. The overall geometric distortion induced by the DWs can therefore be characterized by the volume change $\Delta L \times \Delta b \times \Delta c$.

To conveniently parametrize different supercell sizes and DW spacings, we define the dimensionless DW density

$$\rho = \frac{2}{n},$$

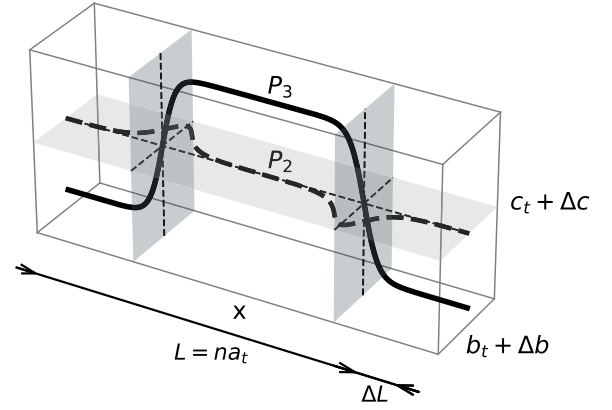


FIG. 1. Schematic illustration of the periodic supercell containing two 180° domain walls in PbTiO_3 . The supercell is repeated along the x axis, forming an equidistant array of DWs separated by $n/2$ unit cells.

which gives the number of DWs per unit cell. The change of supercell dimensions generally depends on ρ . In the dilute limit $n \rightarrow \infty$ (i.e., $\rho \rightarrow 0$), the periodic DW interaction vanishes, corresponding to two infinitely separated and independent DWs in an effectively infinite supercell. In this regime, only the longitudinal lattice parameter is perturbed, and the dimension modification reduces to $\Delta L \times 0 \times 0$. Equation (3) then provides a good approximation to the DW profile.

III. ELECTROMECHANICAL PROPERTIES

In order to describe change of the size of the supercell in Fig. 1 caused by the DWs, we use the continuum constitutive relation between strain and polarization:

$$e_{ij} = f_{ijkl} \frac{\partial P_k}{\partial x_l} + Q_{ijkl} P_k P_l + R_{ijklmn} \frac{\partial P_k}{\partial x_m} \frac{\partial P_l}{\partial x_n} + S_{ijkl} \sigma_{kl}. \quad (4)$$

The piezoelectricity is zero in the cubic system. Using 2-suffix notation, the flexoelectric term with three independent components f_{11}, f_{12}, f_{44} plays a minor role due to depolarizing field. The second term is the ordinary electrostriction with three independent components, Q_{11}, Q_{12}, Q_{44} . The third one is the quadratic term representing “gradient (inhomogeneous) electrostriction” under discussion. The tensor R has 15 independent components, but considering the DW along the x axis there are only seven components contributing and R_{ijkl11} with symmetric (i, j) and (k, l) pairs can be expressed in the 2-suffix notation:

$$R_{ab1} = \begin{pmatrix} R_{111} & R_{121} & R_{121} & 0 & 0 & 0 \\ R_{211} & R_{221} & R_{231} & 0 & 0 & 0 \\ R_{211} & R_{231} & R_{221} & 0 & 0 & 0 \\ 0 & 0 & 0 & R_{441} & 0 & 0 \\ 0 & 0 & 0 & 0 & R_{551} & 0 \\ 0 & 0 & 0 & 0 & 0 & R_{551} \end{pmatrix}, \quad (5)$$

where a refers to the strain component, b to the quadratic (bilinear) coupling in P , and the last index “1” to the bilinear derivative along x .

For a cubic crystal with a one-dimensional polarization profile varying only along $\mathbf{n} = (1, 0, 0)$, so that all gradients reduce to ∂_x , the symmetry-allowed strain components are

$$e_1 = f_{11}\partial_x P_1 + P_1^2 Q_{11} + (P_2^2 + P_3^2)Q_{12} + R_{111}(\partial_x P_1)^2 + R_{121}[(\partial_x P_2)^2 + (\partial_x P_3)^2] + \sigma_1 S_{11} + (\sigma_2 + \sigma_3)S_{12}, \quad (6a)$$

$$e_2 = f_{12}\partial_x P_1 + P_2^2 Q_{11} + (P_1^2 + P_3^2)Q_{12} + R_{221}(\partial_x P_2)^2 + R_{211}(\partial_x P_1)^2 + R_{231}(\partial_x P_3)^2 + \sigma_2 S_{11} + (\sigma_1 + \sigma_3)S_{12}, \quad (6b)$$

$$e_3 = f_{12}\partial_x P_1 + P_3^2 Q_{11} + (P_1^2 + P_2^2)Q_{12} + R_{221}(\partial_x P_3)^2 + R_{211}(\partial_x P_1)^2 + R_{231}(\partial_x P_2)^2 + \sigma_3 S_{11} + (\sigma_1 + \sigma_2)S_{12}, \quad (6c)$$

$$e_4 = Q_{44}P_2 P_3 + R_{441}(\partial_x P_2)(\partial_x P_3) + \sigma_4 S_{44}, \quad (6d)$$

$$e_5 = f_{44}\partial_x P_3 + Q_{44}P_1 P_3 + R_{551}(\partial_x P_1)(\partial_x P_3) + \sigma_5 S_{44}, \quad (6e)$$

$$e_6 = f_{44}\partial_x P_2 + Q_{44}P_1 P_2 + R_{551}(\partial_x P_1)(\partial_x P_2) + \sigma_6 S_{44}. \quad (6f)$$

For a single 180° domain wall ($3_1|\mathbf{n}|3_2$) with normal $\mathbf{n} = (1, 0, 0)$, mechanical equilibrium requires $\sigma_1 = \sigma_5 = \sigma_6 = 0$.

The strains e_2 , e_3 , and e_4 retain their bulk values, and we neglect P_1 and the shear strains e_5 , e_6 , consistent with the DFT results presented in the next section. Eliminating σ_2 and σ_3 in favor of e_2 and e_3 , Eq. (6) reduces to

$$e_1 = (P_2^2 + P_3^2)Q_{12} + \frac{S_{12}}{S_{11} + S_{12}}[e_2 + e_3 - (P_2^2 + P_3^2)(Q_{11} + Q_{12})] + [(\partial_x P_2)^2 + (\partial_x P_3)^2]\left(R_{121} - \frac{S_{12}}{S_{11} + S_{12}}(R_{221} + R_{231})\right), \quad (7)$$

$$e_2 = e_{2,S} = P_S^2 Q_{12}, \quad e_3 = e_{3,S} = P_S^2 Q_{11}, \quad e_4 = e_5 = e_6 = 0. \quad (8)$$

Only the longitudinal strain $e_1(x)$ varies across the wall. In the bulk domain $e_{1,S} = P_S^2 Q_{12}$, and the wall-induced strain deviation $\Delta e_1 \equiv e_1 - e_{1,S}$ separates as

$$\begin{aligned} \Delta e_1 &= \Delta e_{1,eh} + \Delta e_{1,en} \\ &= [(P_{2,S}^2 - P_2^2) + (P_{3,S}^2 - P_3^2)]I_Q - [(\partial_x P_2)^2 + (\partial_x P_3)^2]I_R, \end{aligned} \quad (9)$$

where the spontaneous values are $P_{2,S} = 0$, $P_{3,S} = P_S$, and

$$I_Q = \frac{S_{12}}{S_{11} + S_{12}}(Q_{22} + Q_{23}) - Q_{12}, \quad (10)$$

$$I_R = \frac{S_{12}}{S_{11} + S_{12}}(R_{221} + R_{231}) - R_{121}, \quad (11)$$

with $Q_{22} = Q_{11}$ and $Q_{23} = Q_{12}$. Note that these equations share a similar form with respect to the two indices. The corresponding change in sample length is

$$\Delta \mathcal{L} = \int_{-\infty}^{+\infty} \Delta e_1(x) dx = \Delta \mathcal{L}_{eh} + \Delta \mathcal{L}_{en}, \quad (12)$$

where $\Delta \mathcal{L}_{eh}$, containing the Q tensor, represents the contribution of homogeneous electrostriction, while $\Delta \mathcal{L}_{en}$, containing the R tensor, represents the contribution of inhomogeneous electrostriction. For a supercell containing two well-separated walls described by Eq. (3), the total length change is

$$\begin{aligned} \Delta L &= \Delta \mathcal{L}_{eh} + \Delta \mathcal{L}_{en} = 2\Delta \mathcal{L} \\ &= \frac{4}{k}(P_S^2 - P_B^2)I_Q - \frac{8k}{3}\left(P_S^2 + \frac{P_B^2}{2}\right)I_R, \end{aligned} \quad (13)$$

where $k = 2/\xi$. Equation (13) applies in the long-supercell limit, where the two walls do not interact ($\rho \approx 0$) and the transverse lattice parameters also remain unchanged, $\Delta b = \Delta c = 0$ (see Fig. 1). The conventional electrostriction contribution (associated with I_Q) scales with the DW thickness ξ , whereas the gradient contribution (associated with I_R) scales as ξ^{-1} , highlighting the dominant role of gradient electrostriction in thin DWs.

IV. AB INITIO MODEL

Equation (13) expresses the domain-wall-induced length change ΔL in terms of the spontaneous polarization P_S , the Bloch amplitude P_B , the inverse wall thickness k , and the effective combinations I_Q and I_R of electrostrictive and gradient-electrostrictive coefficients. The purpose of this section is to show how these quantities are obtained from first-principles calculations for PbTiO_3 and how they connect to the continuum description of Sec. III (electromechanical properties); technical details of the electronic-structure calculations are given in the Appendix.

First, the bulk elastic compliances S_{11} and S_{12} (listed as S_{11} and S_{12} in Table I) and the electrostriction coefficients Q_{11} and Q_{12} were determined for cubic PbTiO_3 from homogeneous strain states. The spontaneous polarization P_S was obtained from the fully relaxed tetragonal ferroelectric phase. These bulk calculations provide the input for the homogeneous electrostriction contribution to Eq. (13) and allow us to evaluate I_Q via Eq. (10). The resulting values of S_{ij} , Q_{ij} , I_Q , and P_S are summarized in Table I.

As reference structures, we consider homogeneous tetragonal supercells of $n \times 1 \times 1$ unit cells, with $n = 8, 10, \dots, 22$. The relaxed cell parameters are $L \times a_t \times c_t$, where $L = na_t$, $a_t = b_t = 3.88 \text{ \AA}$, and $c_t = 4.18 \text{ \AA}$.

To access the domain-wall contribution, we then considered the periodic array of 180° DWs introduced in Sec. II (array of 180° domain walls). Each supercell of size $n \times 1 \times 1$ contains two symmetry-related DWs separated by $n/2$ unit cells along the x axis, corresponding to a wall density $\rho = 2/n$. For each n , a full structural relaxation under zero external stress was performed, yielding the relaxed ρ -dependent lattice parameters of the supercell $L + \Delta L(\rho) \times a_t + \Delta b(\rho) \times c_t + \Delta c(\rho)$ and the microscopic ionic displacements. Comparing it with the homogeneous tetragonal reference supercell containing the same number of unit cells, the supercell size change $\Delta L(\rho)$, $\Delta b(\rho)$, and $\Delta c(\rho)$ was calculated; see Figs. 1 and 4.

TABLE I. Material constants (independent of the DWs) and the characteristics of the Ising and Bloch domain walls. Most quantities were obtained from *ab initio* calculations, while ΔL_{eh} , ΔL_{en} , and I_R were derived from the continuum model, Eq. (13).

Component	Value	Unit
Material constants		
S_{11}	3.998×10^{-3}	GPa^{-1}
S_{12}	-1.128×10^{-3}	GPa^{-1}
S_{44}	9.845×10^{-3}	GPa^{-1}
Q_{12}	-1.857×10^{-2}	$\text{m}^4 \text{C}^{-2}$
Q_{11}	7.900×10^{-2}	$\text{m}^4 \text{C}^{-2}$
I_Q	-5.181×10^{-3}	$\text{m}^4 \text{C}^{-2}$
I_R	-6.019×10^{-21}	$\text{m}^6 \text{C}^{-2}$
P_S	0.86	C m^{-2}
Ising DW		
ΔL	3.9×10^{-11}	m (0.39 Å)
ΔL_{eh}	-0.42×10^{-11}	m (-0.042 Å)
ΔL_{en}	4.3×10^{-11}	m (0.43 Å)
k	3.64×10^9	m^{-1} (0.364 Å $^{-1}$)
Bloch DW		
ΔL	4.2×10^{-11}	m (0.42 Å)
ΔL_{eh}	-0.38×10^{-11}	m (-0.038 Å)
ΔL_{en}	4.6×10^{-11}	m (0.46 Å)
k	3.66×10^9	m^{-1} (0.366 Å $^{-1}$)
P_B	0.26	C m^{-2}

From the relaxed structures we extracted the layer-resolved polarization components $P_1(x)$, $P_2(x)$, and $P_3(x)$ across the supercell. An example of the relaxed Bloch profile for $n = 20$ is shown in Fig. 2; the Ising profile is similar, with $P_2 = 0$; compare also Refs. [20,22]. The points represent the positions of alternating Pb and Ti planes, the DW centers are at the Pb planes corresponding to the lowest energy. These profiles provide, for each wall density ρ , the maximum polarization $P_{3,\max}(\rho)$ in the domain, the amplitude $P_{2,\max}(\rho)$

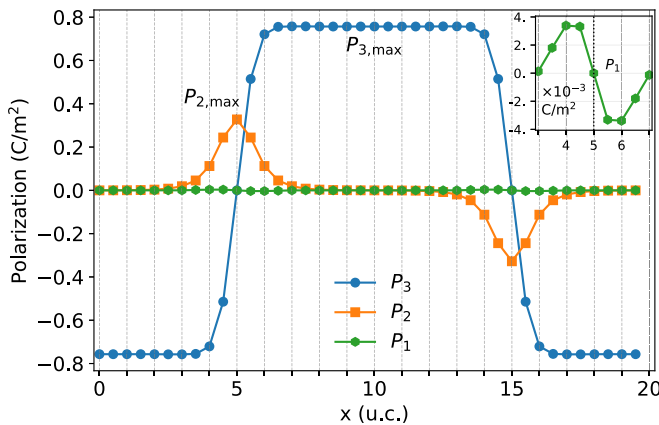


FIG. 2. Layer-resolved polarization components across the Bloch-type 180° DW for $n = 20$. Integer indices correspond to Pb planes, while half-integer indices (e.g., 0.5, 1.5, ...) correspond to Ti planes.

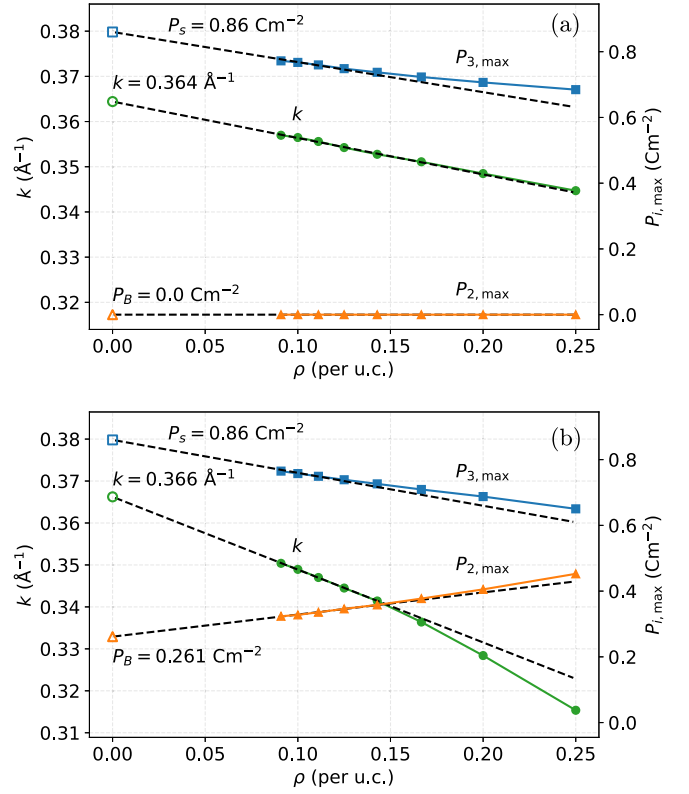


FIG. 3. Dependence of the domain polarization $P_{3,\max}$, the Bloch component $P_{2,\max}$, and the inverse DW thickness k on the DW density ρ . (a) Ising case; (b) Bloch case. The values for independent DWs occur at $\rho = 0$.

of the Bloch component, and the inverse wall thickness $k(\rho)$, defined as $k(\rho) = P_{3,\max}^{-1} \partial_x P_3(x)|_{x=0}$. So we have six quantities characterizing DWs array: the profile parameters $P_{3,\max}(\rho)$, $P_{2,\max}(\rho)$, and $k(\rho)$ shown in Fig. 3 and the supercell size changes $\Delta L(\rho)$, $\Delta b(\rho)$, and $\Delta c(\rho)$ plotted in Fig. 4. To get a description of isolated DWs we need to extrapolate the curves to $\rho = 0$, yielding parameters in Eq. (13): $P_{3,\max}(\rho) \rightarrow P_S$,

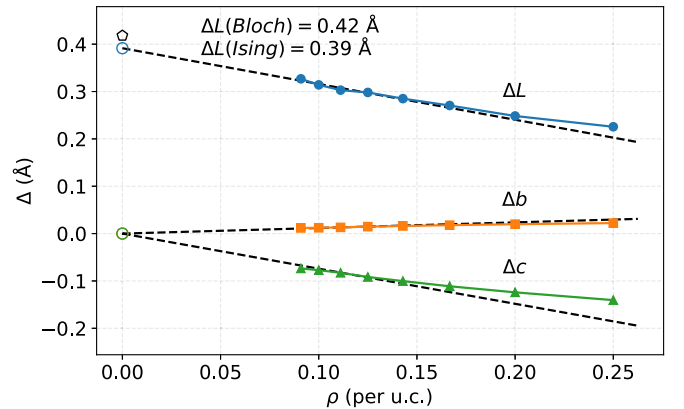


FIG. 4. The longitudinal elongation ΔL , as well as the transverse expansion Δb and contraction Δc , are shown as functions of ρ for the Ising domain wall (DW). The corresponding Bloch DW curves are very similar; only ΔL for the Bloch case at $\rho = 0$ is shown. The values for independent DWs occur at $\rho = 0$.

$P_{2,\max}(\rho) \rightarrow P_B$ and $k(\rho) \rightarrow k$, $\Delta L(\rho) \rightarrow \Delta L$, $\Delta b(\rho) \rightarrow 0$ and $\Delta c(\rho) \rightarrow 0$, Table I. The extrapolations were done by linear fits of the lowest four points. The only unknown quantity, the gradient electrostriction coefficient I_R , is calculated from Eq. (13).

The dimensions of the simulation box and their variation (see Fig. 1) can be used to express the macroscopic (average) strain $\overline{\Delta e_1}$ of a sample with a finite DW density:

$$\overline{\Delta e_1} = \frac{\Delta L(\rho)}{L} = \frac{\Delta L(\rho)}{2a_t} \rho = \frac{\Delta L(\rho)}{2} \hat{\rho}, \quad (14)$$

where $\hat{\rho} = \rho a_t^{-1}$ is the DW number density (per unit length). As shown in Fig. 4, ΔL decreases slightly with density; however, in the dilute regime, $\overline{\Delta e_1} \propto \hat{\rho}$ and the macroscopic strain increases with the DW density, as expected. To quantitatively estimate the macroscopic strain, we consider, as an example, $\rho = 0.1$ in Fig. 4. This yields $\Delta L = 0.31 \text{ \AA}$ and $\overline{\Delta e_1} = 0.4\%$. The separation between adjacent DWs is ten unit cells, i.e., $10a_t$.

V. DISCUSSION

The dependencies of various quantities on the DW density in the multidomain sample, together with their fits, allow us to extract very small yet reliable values, such as a longitudinal elongation on the order of a fraction of an ångström. At finite DW density ρ , the walls exert forces (averaged over the supercell) on the bulk lattice, expanding it along the b axis and compressing it along the polar c axis. This reduces the tetragonality and leads to a decrease in the domain polarization, ($P_{3,\max} < P_S$). As $\rho \rightarrow 0$, the transverse dimensions remain unchanged ($\Delta b = \Delta c = 0$), and the longitudinal elongation ΔL , originating from two independent DWs, remains only a fraction of an ångström.

The continuum electromechanical equation, Eq. (4), enables us to separate the contributions of conventional and gradient electrostriction to ΔL , as expressed in Eq. (13). Electrostriction in the standard LGD framework is governed solely by the tensor Q , and in our case its contribution ΔL_{eh} is very small and negative; thus, it cannot account for the observed positive elongation ($\Delta L > 0$). The remaining, dominant contribution arises from gradient electrostriction, ΔL_{en} , which is positive and an order of magnitude larger than the small negative ΔL_{eh} (see Table I). This underscores the crucial role of gradient electrostriction at the DW center. Consistently, the electrostriction parameter I_R extracted from Eq. (13) is identical for both Ising and Bloch DWs.

The difference in elongation between Ising and Bloch DWs is very small, $\approx 0.03 \text{ \AA}$, indicating a weak dependence on the precise DW profile. Accordingly, the extremely small Néel component $P_1 \approx 10^{-3} \text{ C/m}^2$ has negligible influence (see inset in Fig. 2). The longitudinal elongation ΔL is highly localized: about 97% of the effect originates within two unit cells at the DW center.

VI. SUMMARY

We have investigated the electromechanical properties of the 180° ferroelectric domain wall in PbTiO_3 by combining first-principles calculations with a continuum

Landau-Ginzburg-Devonshire description that includes inhomogeneous electrostriction. Symmetry analysis using layer groups shows that, in the high-symmetry DW state, only an antisymmetric Néel component of the polarization is allowed, while the symmetric and switchable Bloch component requires a symmetry-lowering transition within the DW.

Using density-functional theory, we constructed periodic arrays of 180° DWs in PbTiO_3 with varying wall density, relaxed the atomic and lattice degrees of freedom, and extracted layer-resolved polarization profiles and DW-induced lattice distortions. By analyzing the dependence on DW density in the dilute limit, we obtained the effective spontaneous polarization, Bloch amplitude, wall thickness, and longitudinal elongation associated with a single isolated DW, for both Ising-like and Bloch-like configurations.

Mapping these results onto the continuum constitutive relation (4) allowed us to separate the contributions of conventional (homogeneous) electrostriction and gradient electrostriction to the DW-induced length change. We find that the conventional electrostriction contribution is small and negative, whereas the gradient-electrostriction contribution is positive and nearly an order of magnitude larger, leading to a net elongation that is strongly localized within two unit cells around the DW core. The extracted gradient-electrostriction parameter, $I_R \approx 10^{-21} \text{ C}^{-2}\text{m}^6$, is nearly identical for Ising and Bloch DWs, and the DW-induced strain is only weakly sensitive to the precise profile shape or the tiny Néel component. Since I_R is a linear combination of the three R components, its value also provides a typical order of magnitude for these components, consistent with the value adopted in the phenomenological model of Ref. [20].

Furthermore, assuming a finite wall density, we expressed the macroscopic strain $\overline{\Delta e_1}$. In the dilute regime, this strain increases linearly with the DW density; for a representative DW separation of ten unit cells, we estimate a macroscopic strain of approximately 0.4%.

These results provide direct quantitative support for the central role of gradient (inhomogeneous) electrostriction in the electromechanical response [21] and structural stabilization of 180° domain walls in PbTiO_3 [20]. More broadly, they demonstrate how combining *ab initio* calculations with continuum modeling enables a systematic determination of gradient couplings in ferroic DWs.

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

APPENDIX: COMPUTATIONAL DETAILS

First-principles calculations were performed using Kohn-Sham density-functional theory as implemented in the VASP package [23–25]. The interaction between valence electrons and ionic cores was treated within the projector-augmented-wave (PAW) method, while exchange-correlation effects were described using the PBEsol generalized-gradient approximation. The PAW datasets treated 12 valence electrons for Ti ($3s^2 3p^6 3d^2 4s^2$), 14 for Pb ($5d^{10} 6s^2 6p^2$), and 6 for O ($2s^2 2p^4$). A plane-wave kinetic-energy cut-off of $\text{ENCUT} = 520$ eV and $\text{PREC} = \text{Accurate}$ were used throughout. Brillouin-zone integrations employed a $1 \times 6 \times 6$ Monkhorst-Pack mesh, chosen to reflect the elongated supercell geometry, with the long axis along x .

Structural relaxations were performed using the conjugate-gradient algorithm ($\text{IBRION} = 2$) for up to 300 ionic steps ($\text{NSW} = 300$). Both the atomic positions and the cell shape and volume were allowed to relax ($\text{ISIF} = 3$) under zero

external stress. The electronic self-consistency criterion was set to $\text{EDIFF} = 10^{-8}$ eV, and the ionic relaxations were converged when all Hellmann-Feynman forces were smaller than 0.001 eV $\text{\AA}^{-1}$ ($\text{EDIFFG} = -0.001$ eV $\text{\AA}^{-1}$). Gaussian smearing with $\text{ISMEAR} = 0$ and $\sigma = 0.02$ eV was employed, and real-space projectors were disabled ($\text{LREAL} = \text{.FALSE.}$) to ensure high accuracy.

The 180° domain walls were modeled using supercells containing n Pb, n Ti, and $3n$ O atoms, corresponding to $n \times 1 \times 1$ tetragonal units with n between 8 and 22, and lattice vectors of the form $L(= na_i) \times a_i \times c_i$ in the homogeneous reference structure. Periodic boundary conditions enforce an equidistant array of two symmetry-related walls along the x direction. For each n , the relaxed lattice parameters and ionic positions were used to determine the supercell length change $\Delta L(\rho)$ and the layer-resolved polarization components across the wall. Local polarization profiles were constructed from the relative cation-anion displacements with respect to the cubic structure with the help of Born effective charges.

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