

Dislocation-induced modulation of polarization switching in bulk ferroelectric perovskites

Sepideh Kavousi,¹ Hamed Nobarani²,^{1,*} and Mohsen Asle Zaeem^{1,*}

¹Department of Materials Science and Engineering, *The University of Tennessee*, Knoxville, Tennessee 37916, USA

²Department of Mechanical Engineering, *Colorado School of Mines*, Golden, Colorado 80401, USA



(Received 3 July 2025; revised 28 October 2025; accepted 4 February 2026; published 24 February 2026)

Dislocations in perovskites are believed to disrupt their ferroelectric properties. In this study, a novel molecular dynamics approach utilizing a modified embedded-atom method with charge equilibration interatomic potential is employed to provide qualitative new insights into how dislocations in the bulk can modulate the ferroelectric behavior of perovskites. Results show that stress gradients induced by various dislocation configurations/densities influence the polarization distribution pattern and the domain switching process. This modulation affects the shape of the hysteresis loop. The dynamics of the intrinsic charge-lattice coupling mechanism reveal that changes in polarization patterns are more pronounced in regions under larger compressive or tensile stress. Specific stress thresholds are identified that mark significant changes in polarization behavior under different loading conditions, offering a significant step towards understanding how the heterogeneous stress/strain state of a material influences its ferroelectric properties. Additionally, we qualitatively uncovered the effect of dislocation density on the energy characteristics, offering material design guidelines for specific electronic applications. Lower dislocation densities widen the hysteresis response and enhance the polarization properties, desirable for random-access memory applications, that require long-term data preservation. In contrast, higher dislocation densities result in narrower hysteresis loops and sharper switching responses, optimizing the conditions for high-speed computing and power-sensitive logic devices.

DOI: [10.1103/7m6v-pcrq](https://doi.org/10.1103/7m6v-pcrq)

I. INTRODUCTION

Under an external electric field, the spatial distribution of bound charges in ferroelectric materials shifts, a phenomenon known as polarization. These materials retain their polarization even after the electric field is removed and need a specific threshold of energy to change their polarization magnitude and direction. Ferroelectric materials are characterized by a polarization hysteresis loop that defines critical properties like the saturated field (E_s), the remanent polarization (P_r), and the coercive field (E_c). Due to their polarization switching ability, ferroelectrics are used in a broad spectrum of nanoelectronics applications [1–3], such as nonvolatile memory devices [4,5] and logic devices [6,7].

The desired shape of the polarization-electric field hysteresis loop, along with specific ferroelectric characteristics, varies based on the target application. A rectangular-shaped hysteresis loop is often considered ideal for memory devices. A clear distinction between the polarized and depolarized stable states with a well-defined electric field required for initiating the switching allows for precise control over the switching process. In contrast, the optimum hysteresis shape

for computational logic devices depends on the specific requirements of the application, including factors such as speed, energy efficiency, scalability, and reliability. A narrower hysteresis loop with smaller P_r and E_c likely enables faster switching but can compromise reliability and retention due to the challenges in maintaining stable logic states. On the contrary, a wider hysteresis indicates a higher coercive field, which enhances the stability of the logic states but causes polarization reversal to occur later in the electric loading cycle, effectively slowing the overall switching response. Ultimately, achieving a hysteresis loop with reliable and repeatable performance depends on the specific trade-offs and often involves careful engineering of the material, structure, and operating conditions of the device.

Previous studies highlighted the profound effect of mechanical strain on the polarization characteristics of ferroelectric materials [8–12]. The strain can arise from either the mismatch in properties between the substrate and the perovskite film (such as lattice parameter or thermal expansion mismatch) or from growth conditions (substrate's temperature, growth flux, and growth chamber conditions [11,13]). The in-plane compression strain typically widens the hysteresis loop, improves the polarization stability, and enhances spontaneous polarization [8–10]. Some studies observed an increase in both the remanent polarization [10,12] and the coercive field [10] due to the external compressive strain field. Conversely, the tensile strain makes the hysteresis loops narrower until they finally degenerate into lines, potentially compromising device functionality. Thus, the strain engineering of ferroelectric materials is

*Contact author: mzaeem@utk.edu

introduced as a tool to control and alter their ferroelectric properties [11].

When the perovskite film is under a stress field and its thickness exceeds a critical value, dislocations start forming to relax the structure toward a zero-strain state. Studies on misfit edge dislocations propose a strong coupling between the dislocation strain field and the polarization, which leads to high local polarization gradients [14–20]. Depending on the dislocation core location and its Burgers vector, the strain field around dislocations is generally known to locally alter the film's ferroelectric response, reducing its spontaneous polarization up to 48% [17,19] and degrading its properties based on the overall hysteresis behavior of the system [8,21,22]. The primary reason is that dislocations may pin the domain walls and inhibit their motion under the electric field. Moreover, the regions near the dislocation core do not contribute to the polarization response. The strong depolarizing fields existing around dislocation cores ultimately suppress ferroelectricity [22,23]. However, depending on the dislocation type, they can also widen the hysteresis loop and increase E_c and P_r , respectively [24].

The dislocation defect near the ferroelectric-substrate interface, caused by lattice mismatch, can be an edge dislocation or a dislocation pair [23]. One critical point overlooked in previous studies is that they mainly focus on dislocations occurring at the interface between the substrate and film due to property mismatches. For instance, the results suggest that the degradation of long-range polarization due to the presence of dislocations can be explained by focusing on the material of choice and the substrate. Chu *et al.* [8] experimentally investigated the growth of $\text{Pb}(\text{Zr}_{0.52}\text{Ti}_{0.48})\text{O}_3$ (PZT) on cubic Nb-doped $\text{SrTiO}_3(001)$ substrates, and their quantitative measurements of the strain field showed that a tensile-strained region extends predominantly into the PZT. Thus, the impact of the tensile stress field in degenerating the hysteresis loop into lines was predictable. Similar conclusions can be made from the computational phase-field simulations in the literature [21,22]. For instance, the phase-field simulations performed by Alpay *et al.* [21] predicted the existence of strong depolarizing fields that suppress the polarization around dislocations. The lattice mismatch between the film (PbTiO_3) and the substrate (LaAlO_3) presents interface dislocations with an extended tensile field inside the film. Thus, suppressed polarization was expected, and this observation did not specifically depend on the polarization gradients.

In an unconventional approach, some investigations propose the potential of imprinting dislocations in bulk ferroelectric materials to increase polarization switching response. Wu *et al.* [25] performed phase-field simulations to show the critical role of domain configuration and dislocation density on the polarization switching behavior. Their results showed that there exists a critical dislocation density below which the polarization hysteresis loop of the material gets narrower as we increase the dislocation density. However, further increasing the dislocation density above the threshold makes the polarization hysteresis loop wider [25]. Furthermore, these studies mostly focus on the interactive effect of dislocations in bulk ferroelectric material with the domain walls and their pinning effect on the domain wall motion [26–29].

Several strategies can be applied to enhance dislocation density in perovskite oxides, such as interface engineering [30,31], modifying manufacturing conditions [32], doping [33], or postprocessing techniques such as annealing [34]. We believe that understanding the localized interactions between the stress-polarization pair, considering their magnitude and gradient, is essential to unlocking the potential effect of dislocation configuration on improving ferroelectric performance for different applications. In this work, we study the effect of the strain fields around the dislocation cores on the intrinsic charge-lattice and strain-polarization coupling mechanisms. Most studies in the literature have focused on investigating the isolated role of the dislocations and how they alter the overall polarization and hysteresis loop characteristics of perovskites. We propose to utilize the molecular dynamics (MD) approach, a computational tool with profound capabilities for studying nanoscale mechanisms and predicting the ferroelectric properties and behaviors of different material systems under electrical stimuli. MD simulations enable *in situ* investigation of the dynamic and coupled response of the localized stress field and polarization under electric loading [35].

This paper aims to study the impact of edge dislocations on the polarization behavior and hysteresis loop of BiFeO_3 . Our atomistic simulations of the ferroelectric domain evolution under cyclic electrical loading provide nanoscale insights into domain switching, enhancing our understanding of the relationship between the domain configurations and ferroelectric hysteresis characteristics. The insights gained from this work can guide the design of ferroelectric nanostructures with tailored dislocation configurations for improved switching reliability and reduced energy loss.

II. METHODS AND SIMULATION DETAILS

We utilize MD simulations in this study within the framework of Large-scale Atomic/Molecular Massively Parallel Simulator developed by Sandia National Laboratories [36]. In MD simulations, the accuracy of the interatomic potential, which is used to describe atomic interactions, dictates the fidelity of the results [37–40]. Most studies in literature use the core-shell potential to study the polarization response of ferroelectric materials [41–47]. This model is a type of ion-pair potential [48] which decomposes all atoms into a core with a static charge located at a fixed atomic position and a shell with variable charge and position [49]. Core-shell models are highly effective for purely ionic materials because they explicitly capture electronic polarization through core-shell displacement. However, because core-shell potentials rely on fixed charges, they are less suitable for systems such as BiFeO_3 [50] that exhibit mixed ionic-covalent bonding, charge transfer, and bond directionality [51]. Its reliability is supported by successful applications to a wide range of material systems with diverse bonding characteristics, including ionic (Al–O [52], Ti–O [53]), covalent (Si–O [53]), and mixed ionic-covalent systems (Li–Co–O [54], Li–Mn–O [55], Li–Ni–O [56], Li–Mn–Co–Ni–O [57], and Hf–Ti–O [58]).

The modified embedded atom method (MEAM) potential models the electronic structure and interatomic forces based on the electron density of atoms and their neighbors [59], considering both pairwise and embedded energy terms

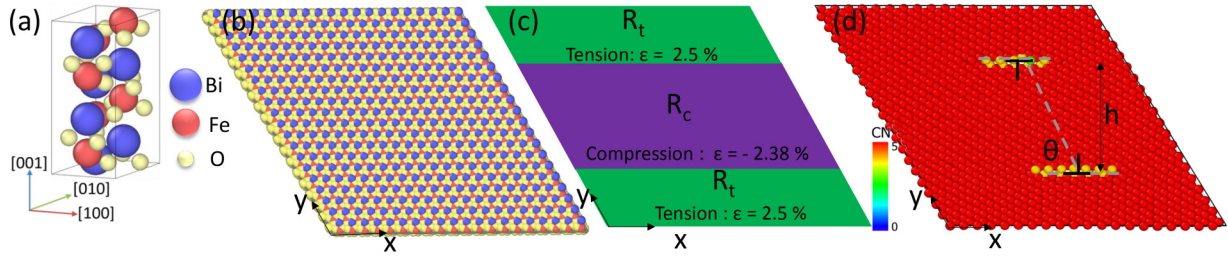


FIG. 1. (a) The crystal structure of BiFeO_3 . The coloring is based on the atomic type; blue, red, and yellow represent Bi, Fe, and O atoms, respectively. (b) Snapshot of defect-free simulation system. (c) Schematic representation of strain field applied to the simulation system to create a system with one pair of dislocations (dislocation density of $\rho_l = 0.017 \text{ nm}^{-2}$; DD1 system). (d) Graphical representation of the equilibrated MD simulation system with one pair of dislocations, where the coloring is based on the coordination number (CN).

[60]. MEAM interatomic potentials provide an effective and accurate prediction of a wide range of properties, including mechanical, thermal, and electrical properties [37,61–63]. In this work, we utilize our in-house interatomic potential, which is a second-nearest-neighbor modified embedded atom method potential with charge equilibrium (2NN-MEAM+QEq) [64]. Integrating MEAM with the charge equilibration (QEq) method [65] enables dynamic charge transfer by minimizing the system's electrostatic energy with respect to atomic partial charges within a local interaction cutoff [53,66]. This coupling extends MEAM's applicability to systems with significant charge redistribution, providing broader chemical transferability. Unlike the fixed-charge core-shell model, MEAM+QEq can self-consistently describe covalent, ionic, and metallic bonding on an equal footing [67]. Therefore, the 2NN-MEAM+QEq potential provides a more accurate description of the interactions between bismuth, iron, and oxygen atoms. Also, it predicts the structures of different phases, elastic properties, phase transformations, defect formation energies, and charge equilibration, making it highly desirable for investigating the nanoscale polarization-switching mechanisms and the mechanical behavior of BiFeO_3 under different environmental loading conditions [52–54].

The rhombohedral structure of BiFeO_3 consists of two pseudocubic perovskite blocks connected along the diagonal in the $\langle 111 \rangle$ direction. The unit cell of the BiFeO_3 rhombohedral crystal structure, shown in Fig. 1(a), has a space group of R_3c with hexagonal lattice parameters of $a = 5.58 \text{ \AA}$ and $c = 13.90 \text{ \AA}$ [68]. Bi, Fe, and O atoms are shown in blue, red, and yellow, respectively. The initial polarization in the material is obtained by the phase transition from a high-temperature paraelectric state to a ferroelectric polar state at the Curie temperature [69]. The initial polarization patterns are created by displacing the iron atoms in the z direction. The total displacement is less than 1 angstrom to keep the stability of the structure. However, the displacement should be large enough to avoid the iron atom returning to its reference/unpolarized structure. During relaxation, all the cations and anions move to reach their new minimized positions in the polarized structure. The equilibrated locations of dislocations are demonstrated by h (the vertical distance between the dislocation lines) and θ (the angle between the line connecting the dislocation lines and the x direction) in Fig. 1(c).

The spontaneous polarization of a ferroelectric material is given by the difference between the polarization of the

ferroelectric state and a reference paraelectric state. A small atomic displacement in a ferroelectric material linearly changes the dipole moment by altering the atomic charge distribution. Atomic charges can be discussed from either a static or dynamic point of view [65]. In the concept of static charge, the delocalized electron density is replaced with localized point charges associated with each atom. On the other hand, the Born effective charge is a dynamic concept that describes how the polarization responds to atomic displacement. If the effective charges are roughly constant, then the polarization of each unit cell can be calculated by Eq. (1) [70]:

$$p(t) = \frac{1}{V_u} \left[\frac{1}{8} Z_{\text{Bi}}^* \times \sum_{i=1}^8 r_{\text{Bi},i}(t) + Z_{\text{Fe}}^* \times r_{\text{Fe}}(t) + \frac{1}{2} Z_{\text{O}}^* \times \sum_{i=1}^6 r_{\text{O},i}(t) \right], \quad (1)$$

where V_u is the volume of a unit cell. The displacement of Bi, Fe, and O atoms with respect to the reference paraelectric structure is represented by r_{Bi} , r_{Fe} , and r_{O} , respectively. Z_{Bi}^* , Z_{Fe}^* , and Z_{O}^* are the Born effective charges. While MEAM+QEq potentials use charge transfer during MD simulations to identify the atomic trajectories, we use the constant Born effective charges [71], as 4.38, 3.61, and -2.66 for Bi, Fe, and O, respectively, to reduce the noise. The Python scripting embedded in OVITO [72] is used to calculate the relative polarization value for each primitive cell [72]. First, the 14 nearest neighbors of each iron atom are determined. We need to ensure that these nearest neighbors consist of six O atoms and eight Bi atoms and then calculate the relative atomic displacement with respect to the unpolarized reference structure. Finally, using Eq. (1), we quantitatively calculate the magnitude of the polarization vector.

Edge dislocations are frequently found in BiFeO_3 and are expected to alter the polarization due to the presence of anisotropic strain fields [17]. The edge dislocations in BiFeO_3 , which have the Burgers vector of $\mathbf{b} = [100]$ and dislocation line of $b\mathbf{f}\zeta = [001]$, appear mostly in pairs [73]. So the simulation system has $[100]$, $[010]$, and $[001]$ orientations in the x , y , and z directions, respectively. In this study, the simulation box remains periodic in all three directions. Thus, we introduce a lattice mismatch between two crystals to form the dislocation. For this, we first construct the bottom of the simulation system by creating a region sized $20 \times 5 \times 3$ unit cells [as shown in Fig. 1(c)] and elongate it by half a unit cell

along the x direction (strain = 2.5%); this region is referred to as the R_t region. We then construct the R_c region (under compression loading) in the simulation system with dimensions of $21 \times 10 \times 3$ unit cells and compress it by half a unit cell along the x direction (corresponding to a strain of -2.38%). Afterwards, we stack R_t - R_c - R_t regions along the y direction. This structure contains two crystals with a mismatch along the x direction and does not yet contain a dislocation. By equilibrating this structure, one pair of dislocations with the dislocation density of $\rho_l = 0.017 \text{ nm}^{-2}$ (DD1 system) will form, as shown in Fig. 1(d). Coordination number (CN) embedded in OVITO [72] was used to characterize local atomic coordination and detect dislocation cores by distinguishing atoms with locally distorted coordination from those in the perfect lattice. The radial distribution functions for Bi-Bi pairs have two peaks located at 3.98 Å and 5.7 Å, which denote the approximate sites of the first and second nearest neighbors. We set the cutoff radius in the coordination analysis to be 4.8 Å to identify the dislocations in the structure. Throughout the paper, we either conduct discussions on the entire simulation cell or perform separate analyses on regions under compression (R_c) or tension (R_t).

Similarly, to create the DD2 system, we followed the same procedure but constructed a multilayer configuration consisting of two $21 \times 6 \times 3$ unit-cell regions compressed by half a unit cell (denoted as SYS1), one $21 \times 6 \times 3$ unit-cell region elongated by half a unit cell (SYS2), and two $20 \times 3 \times 3$ unit-cell regions elongated by half a unit cell (SYS3). These were stacked in the sequence SYS3-SYS1-SYS2-SYS1-SYS3 along the y direction, producing two pairs of dislocations with a total dislocation density of $\rho_l = 0.034 \text{ nm}^{-2}$. Using this method, the dislocation structures are uniformly and periodically distributed throughout the simulation cell.

For all simulation setups, the initial polarization is applied by displacing the iron atoms by -1 Å in the z direction. Then the system is equilibrated in two steps. First, we fix the positions of the iron atoms and equilibrate the system under the isothermal-isobaric ensemble (NPT) at room temperature for 1 ps. For setting the temperature in the simulation box, the Nose-Hoover thermostat [74] was used with a temperature damping parameter of 0.01 ps. Then we remove the constraint on iron atoms and let the system equilibrate under the NPT for an additional 10 ps. In order to investigate how the ferroelectric material responds to an applied electric field and calculate the polarization-electric field hysteresis loop, we applied electric fields ranging from -0.1 V/Å to 0.1 V/Å with an increment of 0.02 V/Å along the z direction. The duration time of each increment is 4 ps. Due to the inherent time and length scales in MD simulations, observing meaningful system responses within practical simulation durations requires higher external forces or fields to accelerate the dynamics. Furthermore, extensive sampling in MD timescales is required to observe many rare events (e.g., phase transformations). Applying stronger stimuli increases the event frequency, improving statistical significance within the limited number of MD runs. Using the same interatomic potential, Nobarani and Asle Zaeem [35] showed that within the time limit of electric loading for a couple of ps, the switching was not observed using smaller electric field values. In this study, an electric field larger than the experimental range is applied over a

shorter period to obtain a distinct response within a reasonable computational cost. We have ensured that the system was equilibrated at the end of 4 ps (Fig. S3 in the Supplemental Material [75]) and remained stable under various loading conditions (Fig. S4 in the Supplemental Material [75]). As a result, the MD-calculated coercive electric field is much larger than the experimental observations [35,76–78].

MD simulation has numerous advantages in studying nanoscale mechanisms and behaviors, but it has time and length scale limitations and a relatively high computational cost. For example, the magnitude of the electric field applied in MD simulations [35,79] needs to be higher than that typically employed in experiments [76–78,80–83]. The rate of the applied electric field in MD simulations is also relatively high. In this study, we performed each loading increment followed by a 4-ps equilibration period. The evolution of total energy over time during each increment, shown in Fig. S3 in the Supplemental Material [75], confirms that 4 ps is sufficient for the system to reach equilibrium after each loading. Also, experimental studies typically report dislocation densities on the order of 10^{13} – 10^{14} m^{-2} [84,85], while our simulations have higher densities. Although it is usual for MD simulations to employ higher dislocation densities, the dislocation pairs tend to accumulate in specific regions, particularly near substrate-perovskite interfaces [73], rather than being uniformly distributed, creating localized regions of high dislocation density and strong strain gradients. Such localized clusters of dislocations can lead to significant variations in the material's polarization behavior. In this study, we focus on understanding how these localized high dislocation regions affect polarization. The DD1 and DD2 configurations in this study serve as simplified analogs of these experimentally observed defect clusters, enabling quantitative analysis of how dislocation-induced strain fields affect ferroelectric switching behavior. A promising future research direction in this area is the integration of atomistic insights into microscale phase-field modeling [86,87] to accurately capture the ferroelectric response, including the effects of dislocation structures, interface phenomena, and defect-driven polarization variations.

III. RESULTS AND DISCUSSIONS

We will first assess the effect of system size on the polarization distribution and hysteresis loop within the MD simulation framework. The electric loading is applied along the z direction, and the evolution of the polarization along this axis is investigated. We have calculated the dynamic response of polarization to cyclic electric loading and have observed that both the size and shape of the hysteresis loop converge as the system dimensions along the x , y , and z axes (length, width, and thickness) increase. Figure 2(a) shows the hysteresis loop of four different system sizes. For the system size of $10 \times 10 \times 3$ unit cells, the hysteresis loop is obviously narrower than in the other cases, with almost zero remanent polarization and coercive field values. However, the hysteresis loop parameters converged beyond the system size of $20 \times 20 \times 3$ unit cells, which was selected as the baseline for all simulations.

Edge dislocations are frequently found in BiFeO₃ and are expected to alter the polarization due to the presence of

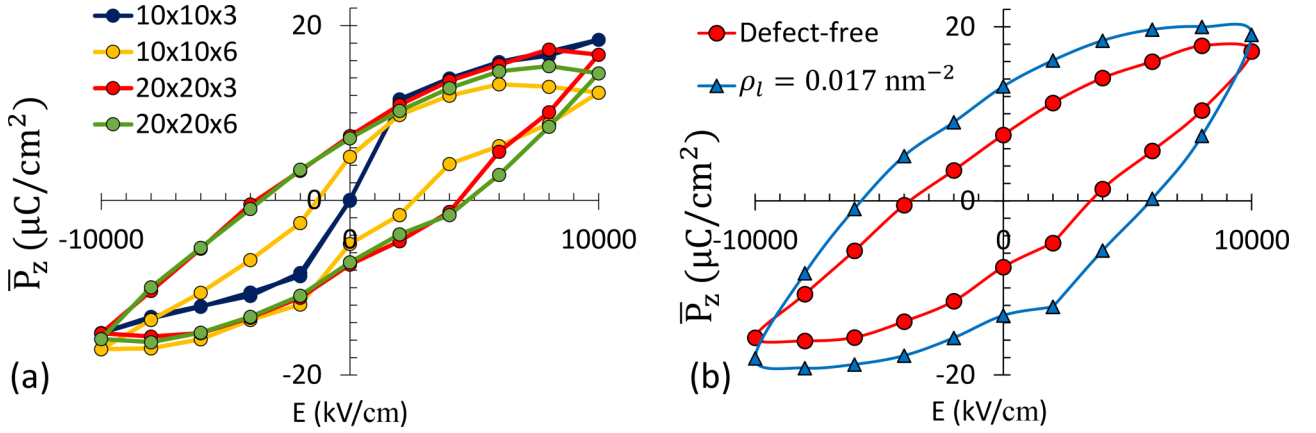


FIG. 2. (a) Graphical representation of the size convergence for the hysteresis loop for a defect-free system, based on the size of the simulation system (number of unit cells), and (b) the hysteresis loop for a defect-free simulation system compared with that for a system with a dislocation density of $\rho_l = 0.017 \text{ nm}^{-2}$.

heterogeneous strain fields [17]. Simulations with a defect-free bulk BiFeO_3 supercell allow the separation of the intrinsic response from those due to defects. Figure 2(b) shows the effect of dislocations (DD1 system) on the polarization-electric field hysteresis loop. The hysteresis loop for polarization across the z direction, P_z , is plotted under an incrementally applied electric field along the thickness direction of the simulation system. Both the defect-free and DD1 cases result in the expected shape of spontaneous polarization-electric field switching. It can be seen from Fig. 2(b) that the positive and negative remanent polarizations and the coercive fields for the hysteresis loops of both structural configurations are almost symmetrical. Compared to the perfect structure, the DD1 system shows a broader hysteresis loop, indicating a higher coercive field, remanent polarization, saturation polarization, and possibly higher energy loss. The strong dependence of spontaneous polarization on existing dislocation defects can be explained by strong lattice-charge coupling. Any modification in the atomic configuration alters the distribution of charges and the resultant polarization [Eq. (1)]. Zhuo *et al.* [27] studied the hysteresis loops of a perfect structure and one containing dislocations in BaTiO_3 , suggesting that defects reduce spontaneous polarization while enhancing the coercive field. However, there are two points that should be emphasized before drawing any conclusions from their study. Zhuo *et al.* [27] mostly focused on the interactions of dislocations and domain walls, which is different from the case study in this paper. Furthermore, they did not investigate how the dislocation density impacts the results.

To uncover the role of dislocations in altering hysteresis behavior, we take a deeper look at the polarization distribution of the DD1 system. As shown in Figs. 3(a) and 3(b), regions experiencing overall tensile or compressive stresses are identified. The extra half-plane of atoms inserted into the crystal structure causes atoms in the lattice surrounding the dislocation core in the central part of the simulation system [indicated as “compression” in Fig. 3(a)] to be under a compressive field. Similarly, the atoms in the lattice near the top and bottom regions are subjected to tensile stress. The stress field around the dislocations evolves with electrical loading rather than remaining constant, indicating a dynamic coupling between

local stress and polarization responses. Previous research also has indicated that a piezoelectric material under compressive stress displays enhanced hysteresis characteristics, including P_r , E_c , and saturation polarization [88]. Figure 3(c) compares the overall polarization of DD1 with that of the compressive and tensile regions. We find that the hysteresis in areas under tension is narrower than the overall hysteresis of the system, with comparably smaller P_r , E_c , and saturation polarization. Moving from the tensile to the compressive region in Fig. 3(c) widens the hysteresis loop with increased P_r , E_c , and saturation polarization. The results for the in-plane tensile stress align well with the experimental observations of ferroelectric properties of lead zirconate titanate (PZT) [89,90] under in-plane tensile loading perpendicular to the electric field direction.

To examine whether polarization remains uniform and homogeneous in the presence of dislocations, we analyzed the evolution of average polarization and stress ($\bar{P}_z$ and $\bar{S}_{xx}$) along the in-plane direction (x axis). The system was divided into small bins [gray dashed parallelogram in Fig. 3(a)] to evaluate local variations under four loading conditions (A–D) in both tensile and compressive regions, denoted by “t” and “c” subscripts, respectively. For each bin, the polarization was averaged along the z direction, consistent with the direction of applied electric field; the simulation cell was intentionally kept thin (three unit cells) along z to minimize statistical noise in local polarization values while preserving physical fidelity. In general, the dislocation pair significantly modifies both the magnitude and spatial distribution of polarization, leading to a pronounced inhomogeneity between tensile and compressive regions. The evolution of $\bar{P}_z$ and $\bar{S}_{xx}$ in the tension is shown in Figs. 3(d1)–3(e1). A slight increase in $\bar{S}_{xx}$ is observed as the loading progresses from D_t to A_t/C_t , and to B_t . Changes in $\bar{P}_z$ are more pronounced, particularly near the bins highlighted by the green dashed region, where B_t shows an upward trend and D_t a downward one. These opposite trends emerge once the local $\bar{S}_{xx}$ exceeds a threshold of about 1.9 GPa, indicating the onset of stress-induced divergence in polarization response. In the compression region Figs. 3(d2)–3(e2), a similar threshold effect is found when $\bar{S}_{xx}$ drops below -1.0 GPa. Bins enclosed by the purple dashed area exhibit the largest $\bar{P}_z$

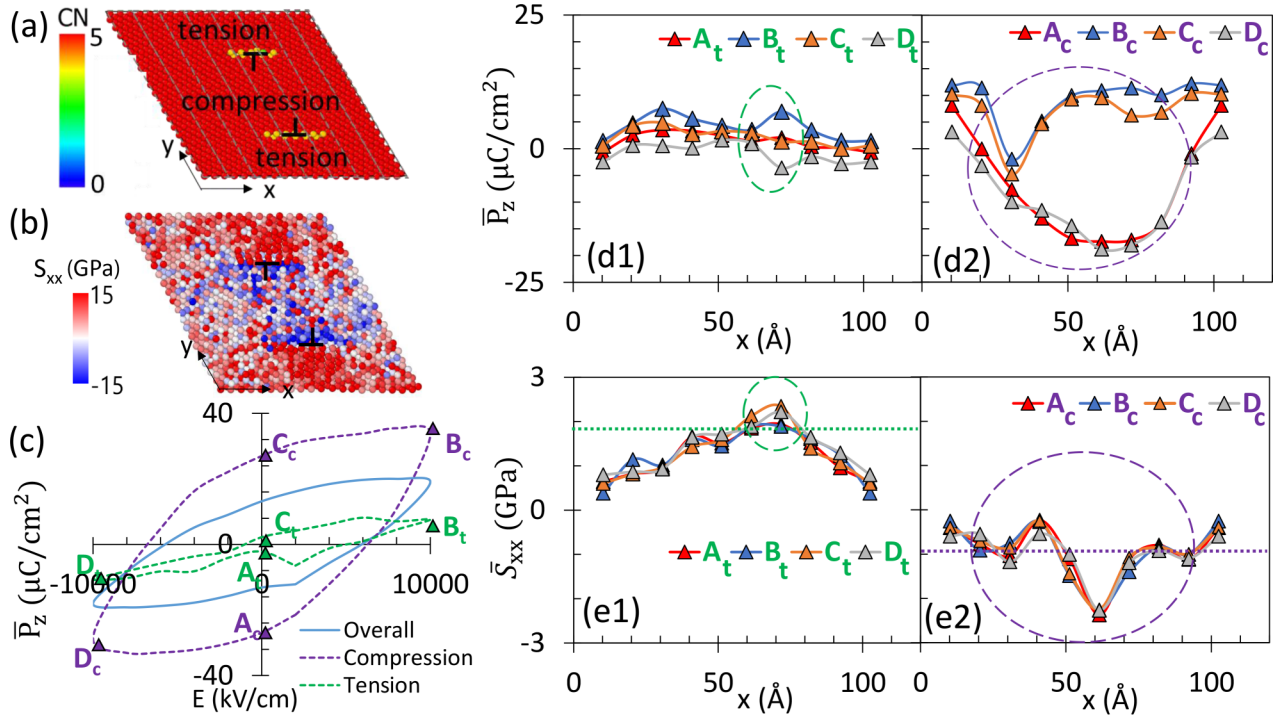


FIG. 3. [(a) and (b)] Graphical representation of the system with a dislocation density of 0.017 nm^{-2} (DD1 system). The coloring is based on CN (a) and stress S_{xx} (b). The gray dotted parallelograms are schematic representations of bins used in this study. (c) The overall hysteresis loop compared with the one for the regions under compression and tension stress for the DD1 system [shown in (a) and (b)]. The evolution of $\bar{P}_z$ along the x direction for four points on the hysteresis loop for (d1) the region under tension (A_t, B_t, C_t, D_t) and (d2) the region under compression (A_c, B_c, C_c, D_c). Average polarization is calculated by averaging P_z in the $y-z$ plane. The evolution of $\bar{S}_{xx}$ along the x direction for four points on the hysteresis loop for (e1) the region under tension (A_t, B_t, C_t, D_t) and (e2) the region under compression (A_c, B_c, C_c, D_c). Average stress is calculated by averaging S_{xx} in the $y-z$ plane.

variations and respond most strongly to electrical loading. Overall, these thresholds mark the transition from uniform to heterogeneous switching behavior driven by dislocation-induced stress gradients. Although not directly comparable to experimental values, they provide a quantitative measure for evaluating local stress-polarization coupling.

To explore how changing the dislocation density impacts the shape and characteristic properties of the hysteresis loop, we conducted a similar simulation and analysis for a system with a higher dislocation density (DD2). As shown in Fig. 4, among the three systems examined, DD1 exhibits the widest hysteresis loop. Generally, compared to regions under tension, P_r values for regions under compression exhibit a higher sensitivity to changes in dislocation density. Increasing the dislocation density from 0 (defect-free) to 0.017 nm^{-2} enhances both the overall P_r and E_c . However, a further increase in dislocation density to 0.034 nm^{-2} yields a more complex response. The narrowest response among all the systems is observed in the system with DD2. While P_r drastically drops with an increase in dislocation density, the difference in E_c between DD1 and DD2 is minimal. A comparison of P_r and E_c in regions under compression and tension reveals that at a lower dislocation density (0.017 nm^{-2}), there is a large gap between the hysteresis response and characteristic properties of these regions. Specifically, moving from the tensile to the compression regions, P_r increases from 2.2 to $26.97 \mu\text{C}/\text{cm}^2$ and E_c also rises from 5100 to $6300 \text{ kV}/\text{cm}$. A similar trend

is observed for the system with a larger dislocation density of 0.034 nm^{-2} , although the changes in P_r are less pronounced, while the variations in E_c are more noticeable. Particularly, moving from the tensile to the compression regions in the DD2 system, P_r increases from 0.74 to $7.04 \mu\text{C}/\text{cm}^2$ and E_c also increases from 3100 to $7200 \text{ kV}/\text{cm}$.

When comparing polarization in a defect-free system with DD1 and DD2 cases, we observed that strain gradients near dislocations significantly impact the polarization response. As described in the methodology section, the strains in both tension and compression regions in DD1 and DD2 are approximately $\pm 2.4\%$, but the key difference lies in the strain gradient along the y axis. Experimental results for BiFeO_3 at 80 K [91] and room temperature [31] demonstrated that the in-plane compressive strain linearly increases the remanent polarization while tensile strain decreases it. Our results for the DD1 case align with these findings, with regions under compression and tension in DD1 showing substantial changes in P_r , reaching 26.97 and $2.2 \mu\text{C}/\text{cm}^2$, respectively, compared to the defect-free case of $7.6 \mu\text{C}/\text{cm}^2$. However, the dislocation configuration and stress distribution around the dislocation core complicate the interpretation, as nearby dislocations generate stress fields that lead to nonlinear variations in the hysteresis loop. Contrary to expectations, the trends in P_r between tension and compression regions in DD2 do not fully align with experimental observations, suggesting that both plane strain and strain gradients, particularly near dislocation

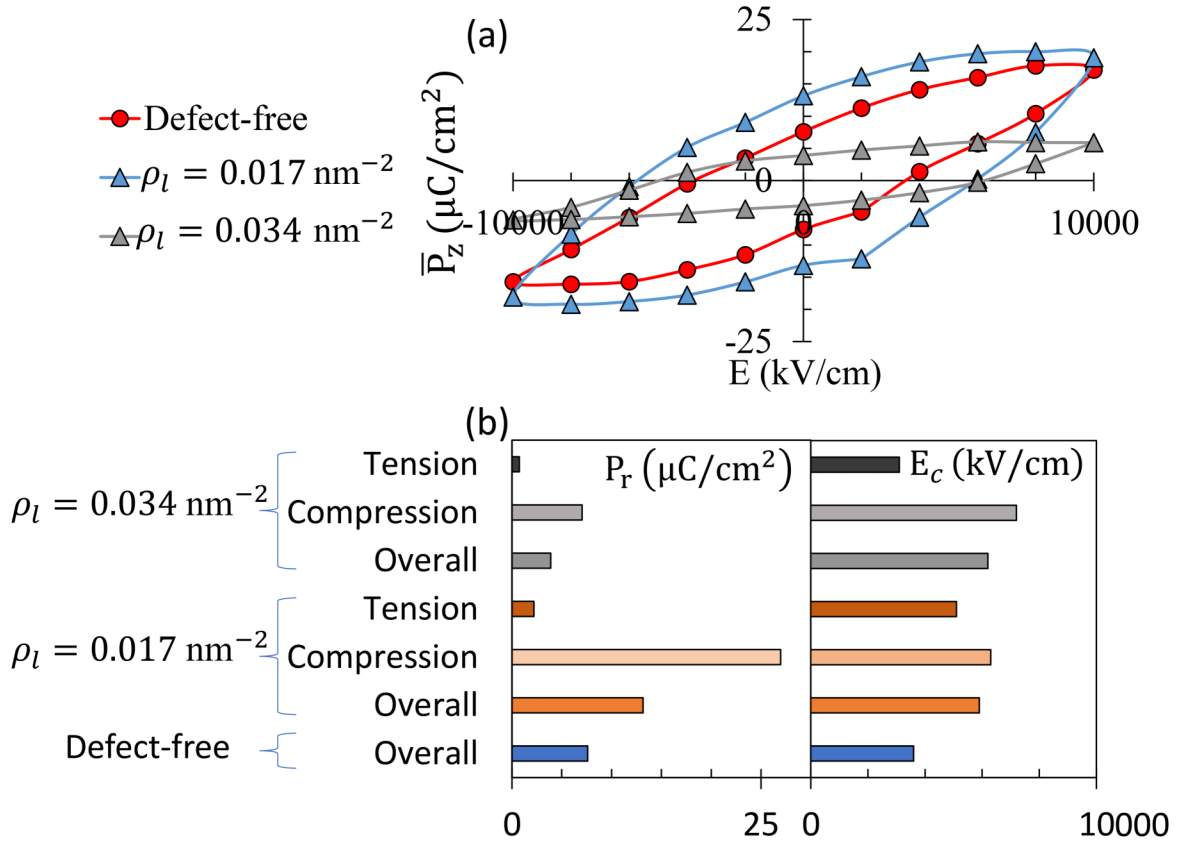


FIG. 4. (a) The hysteresis loop for a defect-free simulation system compared with that for systems with dislocation densities of $\rho_l = 0.017 \text{ nm}^{-2}$ and 0.034 nm^{-2} . (b) The remanent polarization P_r ($\mu\text{C}/\text{cm}^2$) and coercive electric field E_c (kV/cm) for a defect-free system compared with the corresponding values for systems with dislocation densities of $\rho_l = 0.017 \text{ nm}^{-2}$ and 0.034 nm^{-2} . P_r and E_c are calculated separately based on the overall hysteresis response and for the regions under compression and tension stress.

cores, play crucial roles in polarization behavior. Therefore, both factors must be considered for a comprehensive understanding of polarization in these systems.

Making meaningful comparisons between these results and experiments is difficult because the hysteresis loop depends on various factors such as nano/microstructure, maximum applied electric field, simulation system size, and so on [82]. However, the experimental results for P_r cover a wide range of 5–95 $\mu\text{C}/\text{cm}^2$ for maximum electric fields ranging between 75 and 3000 kV/cm [76–78,80–82,92]. It is challenging to draw direct quantitative comparisons with experimental data from the literature, as the shape and magnitude of the hysteresis loop depend strongly on factors such as the film's nano/microstructure, maximum applied electric field, thickness, and measurement conditions. Moreover, MD simulations typically require the application of much larger electric fields to achieve polarization responses comparable to experimental observations, due to the limited timescales accessible in atomistic simulations. Nevertheless, we summarized representative experimental findings in Table S1 of the Supplemental Material [75], and our calculated remnant polarization values remain in reasonable agreement with those reported experimentally. First-principles density functional calculations have shown that the existing distinct switching paths between two states of structures may cause polarization results to spread over a wide range [93,94].

A higher dislocation density increases the interactions between the dislocations and ferroelectric domains, with two mechanisms contributing to the increase in remanent and maximum polarization. First, the strain field associated with a dislocation modifies the lattice structure around the dislocation core and induces local polarization. This modification creates favorable regions for polarization alignment, contributing to a higher remanent polarization. Second, the atomic distortion near the dislocation core leads to localized electric fields. These fields enhance polarization switching behavior, thereby increasing remanent polarization and maximum polarization. Both mechanisms are magnified as the dislocation density increases, resulting in stronger polarization responses. However, when increasing the dislocation density beyond a critical point, it can degrade the polarization behavior. We have identified three mechanisms responsible for this:

(i) At high dislocation densities, the strain fields from adjacent dislocations begin to overlap. This overlapping can distort the local lattice structure and reduce the ability of the material to maintain stable polarization. Thus, the aligned polarization may be disrupted, leading to a decrease in P_r .

(ii) At high dislocation densities, the strain fields become more nonuniform, leading to regions of tensile and compressive strain that may cancel each other out, thereby decreasing the overall net polarization.

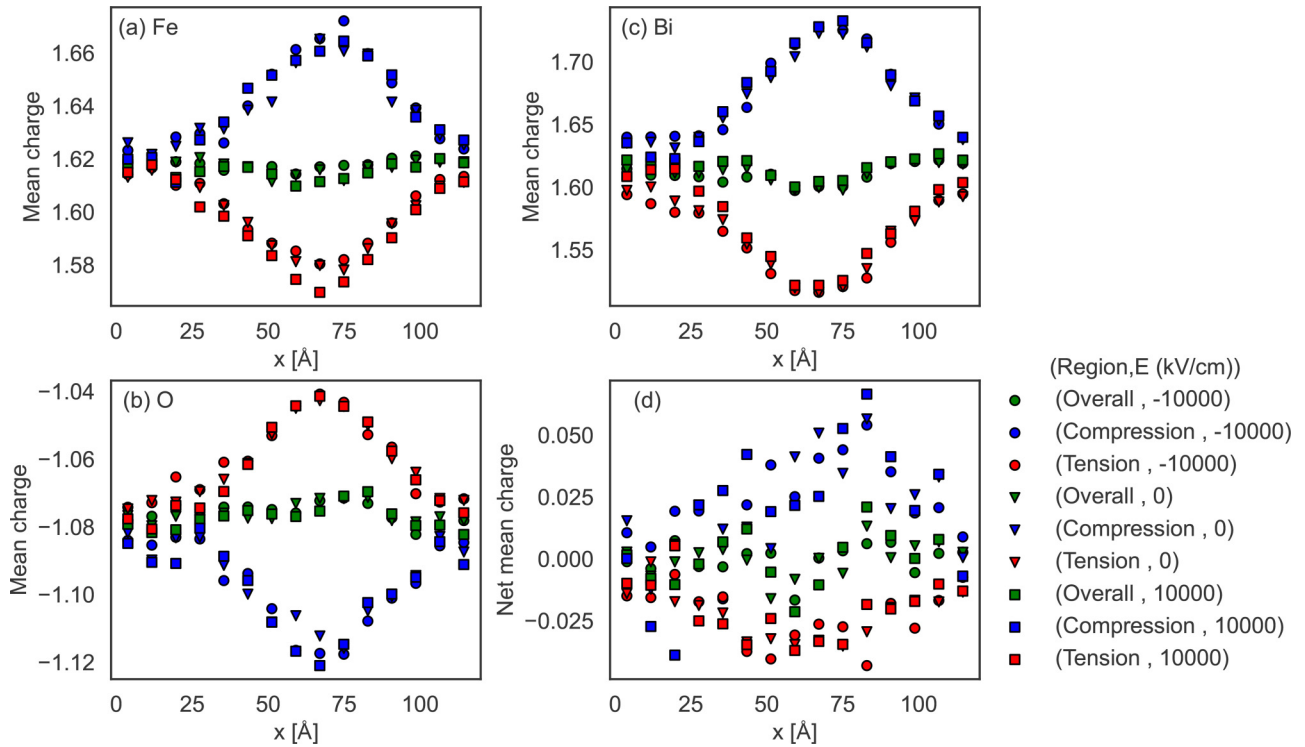


FIG. 5. The evolution of average charge along the x direction for (a) Fe, (b) O, (c) Bi, and (d) net charge. The charges are calculated separately for the overall simulation system and for regions under tension and compression. The evolution of charge calculations is also calculated for three different loadings of $E = -10000$, 0 , and 10000 kV/cm. The average charge is calculated by averaging the charge for different bins along the $y-z$ plane.

(iii) Finally, excessive dislocations lead to an overaccumulation of space charge near dislocation cores. This can cause screening of the local electric fields that would otherwise enhance domain stability. When the local electric field is screened or neutralized, the material becomes less responsive to the external electric field, reducing both P_r and E_c .

Further detailed investigations are needed to elucidate how these changes in the hysteresis response influence the material's energetic characteristics.

To better understand the dynamic response of polarization, we explore how the relative positioning of atoms and charges evolves, focusing on the overall simulation system as well as regions subjected to either tension or compression under various loading scenarios. Figures 5(a)–5(c) presents the evolution of the average charge for Fe, Bi, and O species along the x direction. This average charge is determined by taking the mean dynamic charge of atoms across 15 bins on the $y-z$ plane. First, we assess whether loading conditions ($E = -10000$, 0 , and 10000 kV/cm) affect the dynamic behavior of the average charge along the x direction. From Figs. 5(a)–5(c), it is evident that while the evolution of average charge in the overall system is distinct from that in regions under tension and compression, the loading condition does not significantly impact the dynamic response of the average charge. Therefore, our subsequent analysis will disregard the effect of loading conditions.

The average charge of Fe, Bi, and O in the overall simulation system changes slightly along the x direction, and the loading condition does not have a significant impact on it. The net mean charge in each bin, as presented in Fig. 5(d), is

calculated by adding the mean charge of all atoms (Bi, Fe, and O) in each bin's tension and compression regions. The graph corresponding to the overall evolution of net charge along the x direction typically hovers around zero. However, the mean charge responses in regions under tension and compression exhibit distinct trends. In compression regions, there is a notable increase in the net charge values for Fe and Bi, peaking near the bins that contain the dislocation cores and declining as we move farther away from the dislocation core. This suggests that a high compressive stress field, existing near the dislocation core in Figs. 3(d) and 3(e) and diminishing away from the core, is the governing factor for observing this trend. For O atoms, the mean charge evolution appears inverted, showing a dip near the dislocation core. However, it is important to note that O atoms carry a negative charge, implying that the compressive stress field actually increases the absolute value of the mean charge. However, the extent of the change in mean charges for Fe, Bi, and O varies, leading to a positive final net charge near the dislocation core in the compression region. We also calculated the $x-y$ charge distribution averaged along the z direction. The 2D graphical representation of the CN and charge distribution for O, Fe, and Bi atoms across the $x-y$ plane is shown in Fig. S5 in the Supplemental Material [75], with corresponding data provided in Table S2 of the Supplemental Material [75]. Although our approach models dislocations without directly modifying the atomic structure, the atomic rearrangement during equilibration suggests that the dislocation cores carry a net negative charge.

Conversely, in regions under tension, the mean charge dynamics for Fe and Bi atoms have a dip near the dislocation

core. However, a peak is observed for O atoms with a negative charge, indicating a decrease in the absolute mean charge near the dislocation core. However, the extent of the change of mean charges of Fe, Bi, and O varies, leading to a negative final net charge near the dislocation core. When comparing the evolution of net mean charge, it is evident that the extent of net charge in the compression region is greater than that in the regions under tension.

Before speculating on how the dynamic response of the mean charge alters the polarization response, we first assess whether the loading conditions impact the relative positioning of atoms. For this, we use the radial distribution function, $g(r)$, which measures the probability of finding a particle at a distance (r) from the reference atoms. The findings, presented in Figs. S1(a)–S1(c) in the Supplemental Material [75], show that the interparticle distance between pairs of iron (Fe-Fe), oxygen (O-O), and bismuth (Bi-Bi) atoms remains relatively consistent across the entire simulation system as well as regions under tension and compression.

Essentially, the dynamic shifts in net mean charge, when considered alongside the consistent interatomic distances as shown by $g(r)$, suggest that the polarization changes induced by electrical loading are a direct response to the defects. This relationship highlights the complex interaction between mechanical stress and electronic structure in materials, showing how defects can drastically influence material properties by altering atomic arrangements and redistributing charge.

The surface integral of the polarization-electric field hysteresis loop, represented by Fig. 6(a), can be used to estimate various energy characteristics for ferroelectric materials, known as energy loss (U_l), energy storage (U), and energy storage efficiency (η), given by

$$\eta = \frac{U}{U + U_l}. \quad (2)$$

Figures 6(b)–6(d) panels represent the comparison of U_l , U , and η across different regions (overall, compression, and tension regions, respectively) of simulation systems with defects versus a defect-free system. Figure 6(b) shows that the presence of defects generally has a negative impact on energy storage efficiency. In comparison to the perfect structure with $\eta = 0.27$, DD1 and DD2 have efficiencies of 0.11 and 0.12, respectively. In the DD1 case, U reduces from 34.4 to 18.4 J/cm³, and U_l increases from 91.0 to 146.2, demonstrating a significant detrimental effect on all the energy storage characteristics at a dislocation density of 0.017 nm⁻². The trend observed for DD2 shows a very slight increase in efficiency. As the dislocation density increases from 0.017 nm⁻² to 0.034 nm⁻², the stored energy further decreases to 6.0 J/cm³. The slight increase in efficiency is due to the reduction in energy loss U_l to 44.0 J/cm³, which is even lower than that of the defect-free system.

Despite a drop in efficiency from 0.27 for a defect-free system to 0.11 for the DD1 system, further analysis of different regions of DD1 [shown in Fig. 6(c)] reveals interesting trends in η , U , and U_l . The regions under compression, which exhibit a wider hysteresis response compared to the overall and tensile regions, have a larger U (26.4 J/cm³) and U_l (270.1 J/cm³). However, the resulting efficiency ($\eta = 0.09$) is significantly lower than that of the defect-free system

($\eta = 0.27$) and the overall efficiency of the DD1 system ($\eta = 0.11$). Meanwhile, the region under tension has a much better efficiency (0.19) than the overall region and the region under compression. The improved efficiency, despite the drop in stored energy, is observed because the region under tension has a dramatically smaller U_l (42.3 J/cm³), even lower than that of the defect-free system.

The detailed analysis of the DD2 system reveals an even more interesting insight into the role of defect density in the energy storage characteristics. While the defect-free system displays higher efficiency than the DD2 case, the region under tension for DD2 exhibits an even greater efficiency, achieving $\eta = 0.32$. This counterintuitive trend happens despite the drop in energy stored in all regions of the DD2 system (overall $U = 6.0$ J/cm³, tension $U = 7.0$ J/cm³, compression $U = 5.3$ J/cm³) in comparison to the defect-free system that has $U = 34.4$ J/cm³. The enhanced efficiency in the tensile region of the DD2 system can be attributed to its relatively low energy loss ($U_l = 14.7$ J/cm³), which is significantly smaller than that in the defect-free system ($U_l = 91.0$ J/cm³). This smaller energy loss is the key factor driving the improved efficiency in this region.

In order to link the energy storage characteristics to the material's hysteresis response, Fig. 6(e) represents a scatter plot depicting the evolution of hysteresis characteristics P_r versus E_c across all regions (overall, tension, and compression) in the investigated structures (defect-free, DD1, and DD2). The marker sizes in these plots represent the values of U_l (e1), U (e2), and η (e3). In general, the narrower the hysteresis response is, the smaller P_r and E_c tend to be. Details from Fig. 6(e1) suggest that there is almost a direct correlation between P_r and/or E_c and energy loss. Either larger P_r or E_c leads to a higher U_l . This explains the large energy loss observed for regions under compression in comparison to the overall system. Similarly, the small energy loss in tensile regions can be attributed to their smaller hysteresis characteristics. Figure 6(e2) shows the direct impact of P_r on U , and E_c , with an inverse correlation with U . Despite its larger P_r , the high E_c value for the overall DD1 system results in a lower stored energy compared to the overall hysteresis of the defect-free structure. A similar observation applies to the results for the system with DD2. Although there is a significant drop in P_r for the overall DD2 case, the increase in E_c counteracts the potential benefits of a higher P_r value on energy storage. This suggests that the relationship between various hysteresis characteristics and energy storage properties is complex, necessitating precise calculations for definitive conclusions. Figure 6(e3) summarizes the correlation between P_r , E_c , and η . Our dataset indicates that decreases in P_r and/or E_c (narrower hysteresis) positively impact the efficiency of the designed structure. This clarifies why regions under tension exhibit higher efficiency compared to the overall region or regions under compression. We also observed that the DD2 system displays a slightly better efficiency compared to the DD1 case, highlighting distinct and complex differences in how dislocation density impacts ferroelectric properties.

Generally, the DD1 case with a dislocation density of $\rho_l = 0.017$ nm⁻² exhibits lower efficiency and energy storage, along with a higher energy loss value. However, their influence on the hysteresis characteristics and the

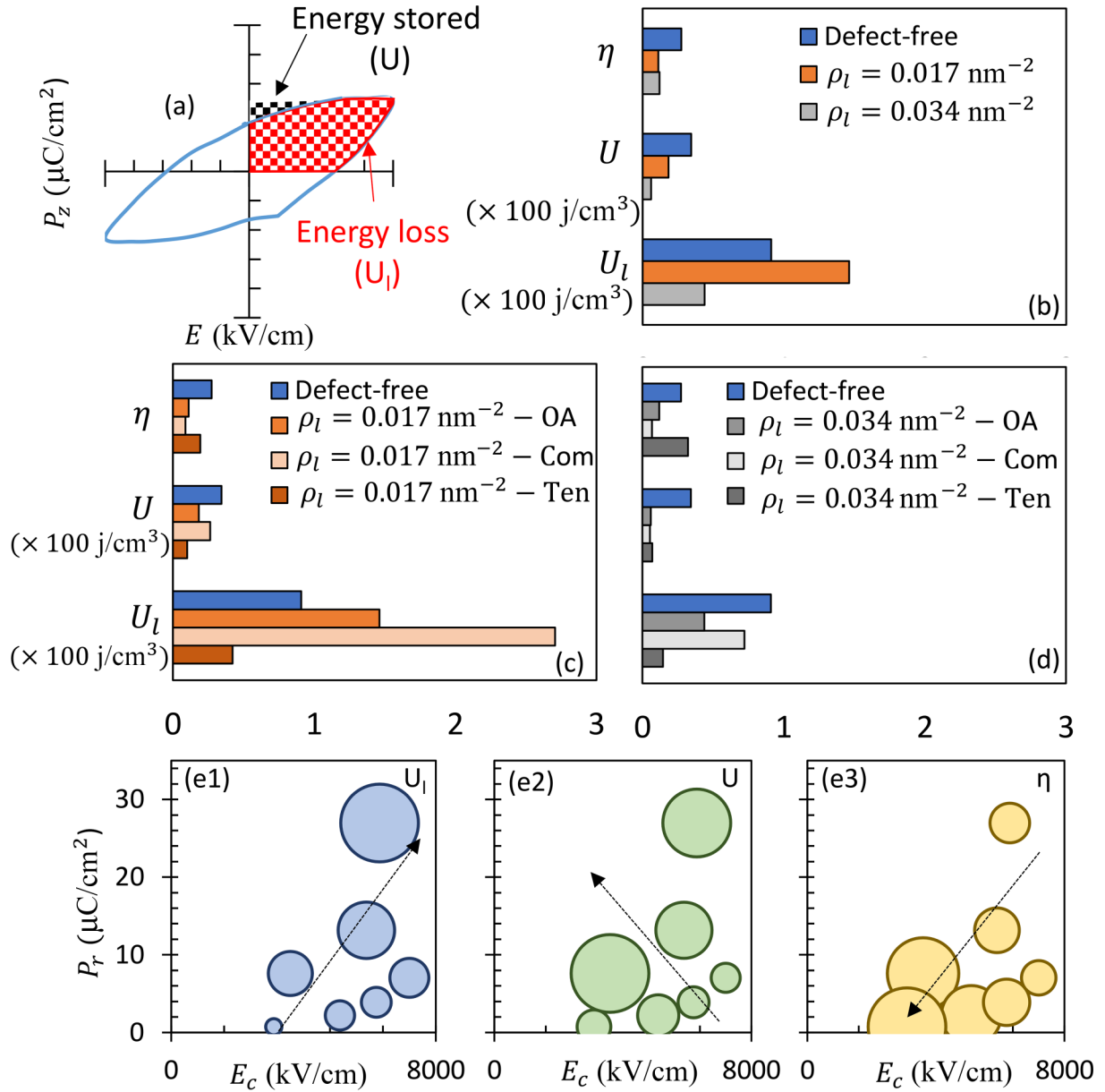


FIG. 6. (a) Schematic description of the energy storage characteristics for ferroelectric materials and (b) the role of dislocation density on the energy storage characteristics of the material, represented by energy loss U_l , energy stored U , and efficiency η . The comparison of U_l , U , and η for a defect-free simulation system with the overall, compression, and tension regions in simulation systems with two dislocation densities: (c) $\rho_l = 0.017 \text{ nm}^{-2}$ and (d) $\rho_l = 0.034 \text{ nm}^{-2}$. And (e) the variation of P_r versus E_c with the marker size representing (e1) U_l , (e2) U , and (e3) η . The arrows represent the approximate trend lines.

resulting impact on the energy properties of ferroelectric materials, as depicted in Fig. 6(e), suggests that dislocations negatively affect overall U_l , U , and η . A narrower hysteresis loop with a sharper switching response (i.e., complete switching at a lower electric field) can improve energy efficiency in nanoelectronic devices such as ferroelectric field-effect transistors [6,7]. Conversely, wider loops with greater remanent polarization are advantages for long-term data retention in ferroelectric random-access memory devices [4,5].

This study suggests that defects (such as dislocation pairs) can be strategically advantageous when the target is to reduce the energy costs associated with the switching process, even

if this does not necessarily lead to improvements in efficiency. For applications such as memory devices, the decrease in energy loss and a reasonably acceptable efficiency achieved by introducing a higher dislocation density (as in the DD2 system) may compensate for the reduced energy storage. This approach aligns with the objective of minimizing energy costs in data storage applications. Our investigation on the polarization retention (given in Fig. S2 in the Supplemental Material [75]) suggests that dislocations assist the ferroelectric material in retaining its polarized state for extended periods. Further detailed investigations into polarization retention, aging, and fatigue during operational cycles are planned for future research.

IV. CONCLUSIONS

We investigated the atomistic-scale polarization switching in BiFeO₃ under an external electric field to obtain the polarization-electric field hysteresis loop. We studied the complex interplay between dislocation density, mechanical stress, and ferroelectric properties through detailed MD simulations. The results supported the existence of a correlation among the dislocation configuration, dislocation-induced inhomogeneous stress, and the polarization characteristics of ferroelectric material. In particular, the following conclusions can be drawn:

(i) *Dislocation-induced stress and local polarization.* Dislocations introduce compressive and tensile stresses in different regions of the material, significantly impacting polarization behavior. Our investigations reveal that the mechanical stress induced by dislocations, whether compressive or tensile, leads to notable localized changes in polarization dynamics under different electrical loading conditions. The inhomogeneous stress fields, which depend on the dislocation configurations and are dynamically linked to the electrical loading, alter the polarization distribution across the material. However, there is a variance in the hysteresis response of regions under compression and tension. Specifically, regions under compression exhibit wider hysteresis loops with increased remanent polarization, coercive field, and saturation polarization, compared to narrower loops in the tensile regions.

(ii) *Stress thresholds for response contrast.* This study established specific stress thresholds that correlate with notable changes in polarization behavior, providing a quantitative framework to predict how dislocation-induced stresses affect ferroelectric properties. This finding is pivotal as it quantifies the mechanical-electrical interaction within ferroelectric materials for the first time. Although these thresholds do not directly correspond to experimental values, they offer a significant step towards understanding the mechanical influences on ferroelectric properties.

(iii) *Energy characteristics and dislocation density.* Dislocations in ferroelectric materials generally reduce energy storage efficiency and increase energy loss. But the key findings in this work show that as the dislocation density increases, its detrimental effect on efficiency fades. Different regions (under tension or compression) within the system, each with different dislocation densities, exhibit different

responses. Regions under compression, despite having higher energy storage and loss, show a much lower efficiency than regions under tension, which have a lower energy loss. The tension regions within the system with a higher dislocation density show a higher efficiency comparable with the defect-free system, which is attributed to its significantly lower energy loss.

(iv) *Linking hysteresis shape to energy metrics.* This study correlated the hysteresis loop shape and characteristics with the energy characteristics. Narrower loops tend to have smaller P_r and E_c , which positively impacts energy efficiency. However, there is a complex relationship between hysteresis characteristics and energy storage, where higher P_r and E_c values can diminish the benefits for energy storage. But it can be advantageous for high-speed computing and energy-efficient switching in devices like ferroelectric field-effect transistors. Larger P_r and/or E_c values generally result in higher energy loss, especially in compression regions. Such a hysteresis response can be beneficial where the energy loss is compensated for by the importance of long-term data preservation in memory devices.

The new insights provided by this work not only deepen the understanding of dislocation effects on the hysteresis behaviors of ferroelectric materials but also enable the tailoring of material properties for specific applications. This is achieved by controlling the density and contribution of dislocations to manipulate the polarization-electric field hysteresis response.

ACKNOWLEDGMENTS

This work was supported by the U.S. Department of Energy, Office of Science, Basic Energy Sciences, under Award No. DE-SC0019279. The authors are grateful for the supercomputing time allocation provided by the National Science Foundation's ACCESS (Advanced Cyberinfrastructure Coordination Ecosystem: Services & Support), Award No. TG-DMR140008.

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.

-
- [1] J. Scott, Applications of modern ferroelectrics, *Science* **315**, 954 (2007).
 - [2] J. Hoffman, X. Pan, J. W. Reiner, F. J. Walker, J. Han, C. H. Ahn, and T. Ma, Ferroelectric field effect transistors for memory applications, *Adv. Mater.* **22**, 2957 (2010).
 - [3] L. W. Martin and A. M. Rappe, Thin-film ferroelectric materials and their applications, *Nat. Rev. Mater.* **2**, 16087 (2016).
 - [4] D. Bondurant, Ferroelectric ram memory family for critical data storage, *Ferroelectrics* **112**, 273 (1990).
 - [5] Y. Arimoto and H. Ishiwara, Current status of ferroelectric random-access memory, *MRS Bull.* **29**, 823 (2004).
 - [6] S. H. Kuk, S. Han, D. H. Lee, B. H. Kim, J. Shim, M. H. Park, J. H. Han, and S. H. Kim, Logic and memory ferroelectric field-effect-transistor using reversible and irreversible domain wall polarization, *IEEE Electron Device Lett.* **44**, 36 (2023).
 - [7] Y. Xunzhao, A. Aziz, J. Nahas, S. Datta, S. Gupta, M. Niemier, and H. Xiaobo Sharon, Exploiting ferroelectric FETs for low-power non-volatile logic-in-memory circuits, in *IEEE/ACM International Conference on Computer-Aided Design (ICCAD)* (IEEE, Los Alamitos, CA, 2016), pp. 1–8.
 - [8] M.-W. Chu, I. Szafraniak, R. Scholz, C. Harnagea, D. Hesse, M. Alexe, and U. Gösele, Impact of misfit dislocations on the

- polarization instability of epitaxial nanostructured ferroelectric perovskites, *Nat. Mater.* **3**, 87 (2004).
- [9] A. G. Zembilgotov, N. A. Pertsev, H. Kohlstedt, and R. Waser, Ultrathin epitaxial ferroelectric films grown on compressive substrates: Competition between the surface and strain effects, *J. Appl. Phys.* **91**, 2247 (2002).
- [10] Y. Sang, B. Liu, and D. Fang, The size and strain effects on the electric-field-induced domain evolution and hysteresis loop in ferroelectric BaTiO₃ nanofilms, *Comput. Mater. Sci.* **44**, 404 (2008).
- [11] D. G. Schlom, L.-Q. Chen, C.-B. Eom, K. M. Rabe, S. K. Streiffer, and J.-M. Triscone, Strain tuning of ferroelectric thin films*, *Annu. Rev. Mater. Res.* **37**, 589 (2007).
- [12] K.-J. Choi, M. Bieganski, Y. L. Li, A. Sharan, J. Schubert, R. Uecker, P. Reiche, Y. B. Chen, X. Pan, V. Gopalan, L. Q. Chen, D. Schlom, and C. Eom, Enhancement of ferroelectricity in strained BaTiO₃ thin films, *Science* **306**, 1005 (2004).
- [13] L. B. Freund and S. Suresh, *Thin Film Materials: Stress, Defect Formation and Surface Evolution* (Cambridge University Press, Cambridge, UK, 2004).
- [14] A. Y. Emelyanov and N. A. Pertsev, Abrupt changes and hysteretic behavior of 90° domains in epitaxial ferroelectric thin films with misfit dislocations, *Phys. Rev. B* **68**, 214103 (2003).
- [15] M.-W. Chu, I. Szafraniak, D. Hesse, M. Alexe, and U. Gösele, Elastic coupling between 90 twin walls and interfacial dislocations in epitaxial ferroelectric perovskites: A quantitative high-resolution transmission electron microscopy study, *Phys. Rev. B* **72**, 174112 (2005).
- [16] D. Su, Q. Meng, C. Vaz, M.-G. Han, Y. Segal, F. J. Walker, M. Sawicki, C. Broadbridge, and C. H. Ahn, Origin of 90° domain wall pinning in Pb(Zr_{0.2}Ti_{0.8})O₃ heteroepitaxial thin films, *Appl. Phys. Lett.* **99**, 102902 (2011).
- [17] A. Lubk, M. Rossell, J. Seidel, Y. Chu, R. Ramesh, M. Hÿtch, and E. Snoeck, Electromechanical coupling among edge dislocations, domain walls, and nanodomains in BiFeO₃ revealed by unit-cell-wise strain and polarization maps, *Nano Lett.* **13**, 1410 (2013).
- [18] Y. Zheng, B. Wang, and C. Woo, Effects of interface dislocations on properties of ferroelectric thin films, *J. Mech. Phys. Solids* **55**, 1661 (2007).
- [19] C. L. Jia, S. B. Mi, K. Urban, I. Vrejoiu, M. Alexe, and D. Hesse, Effect of a single dislocation in a heterostructure layer on the local polarization of a ferroelectric layer, *Phys. Rev. Lett.* **102**, 117601 (2009).
- [20] N. Li, R. Zhu, X. Cheng, H.-J. Liu, Z. Zhang, Y.-L. Huang, Y.-H. Chu, L.-Q. Chen, Y. Ikuhara, and P. Gao, Dislocation-induced large local polarization inhomogeneity of ferroelectric materials, *Scr. Mater.* **194**, 113624 (2021).
- [21] S. P. Alpay, I. B. Misirlioglu, V. Nagarajan, and R. Ramesh, Can interface dislocations degrade ferroelectric properties? *Appl. Phys. Lett.* **85**, 2044 (2004).
- [22] V. Nagarajan, C. Jia, H. Kohlstedt, R. Waser, I. Misirlioglu, S. Alpay, and R. Ramesh, Misfit dislocations in nanoscale ferroelectric heterostructures, *Appl. Phys. Lett.* **86**, 192910 (2005).
- [23] I. B. Misirlioglu, A. L. Vasiliev, S. P. Alpay, M. Aindow, and R. Ramesh, Defect microstructures in epitaxial PbZr_{0.2}Ti_{0.8}O₃ films grown on (001) SrTiO₃ by pulsed laser deposition, *J. Mater. Sci.* **41**, 697 (2006).
- [24] Y. L. Li, S. Y. Hu, S. Choudhury, M. I. Baskes, A. Saxena, T. Lookman, Q. X. Jia, D. G. Schlom, and L. Q. Chen, Influence of interfacial dislocations on hysteresis loops of ferroelectric films, *J. Appl. Phys.* **104**, 104110 (2008).
- [25] H. H. Wu, J. Wang, S. G. Cao, and T. Y. Zhang, Effect of dislocation walls on the polarization switching of a ferroelectric single crystal, *Appl. Phys. Lett.* **102**, 232904 (2013).
- [26] A. Kontsos and C. M. Landis, Computational modeling of domain wall interactions with dislocations in ferroelectric crystals, *Int. J. Solids Struct.* **46**, 1491 (2009).
- [27] F. Zhuo, X. Zhou, S. Gao, M. Höfling, F. Dietrich, P. B. Groszewicz, L. Fulanović, P. Breckner, A. Wohninsland, B.-X. Xu, H.-J. Kleebe, X. Tan, J. Koruza, D. Damjanovic, and J. Rödel, Anisotropic dislocation-domain wall interactions in ferroelectrics, *Nat. Commun.* **13**, 6676 (2022).
- [28] F. Zhuo, X. Zhou, S. Gao, F. Dietrich, P. B. Groszewicz, L. Fulanović, P. Breckner, B.-X. Xu, H.-J. Kleebe, D. Damjanovic, and J. Rödel, Intrinsic-strain engineering by dislocation imprint in bulk ferroelectrics, *Phys. Rev. Lett.* **131**, 016801 (2023).
- [29] M. Höfling, X. Zhou, L. M. Riemer, E. Bruder, B. Liu, L. Zhou, P. B. Groszewicz, F. Zhuo, B.-X. Xu, K. Durst, X. Tan, D. Damjanovic, J. Koruza, and J. Rödel, Control of polarization in bulk ferroelectrics by mechanical dislocation imprint, *Science* **372**, 961 (2021).
- [30] H. U. Habermeier, Substrate surface engineering for functional ceramic thin film growth, *J. Electroceram.* **13**, 23 (2004).
- [31] H. W. Jang, S. H. Baek, D. Ortiz, C. M. Folkman, R. R. Das, Y. H. Chu, P. Shafer, J. X. Zhang, S. Choudhury, V. Vaithyanathan, *et al.*, Strain-induced polarization rotation in epitaxial (001) BiFeO₃ thin films, *Phys. Rev. Lett.* **101**, 107602 (2008).
- [32] Q. Lin, R. Ding, Q. Li, Y. Y. Tay, D. Wang, Y. Liu, Y. Huang, and S. Li, Large piezoelectricity and ferroelectricity in Mn-Doped (Bi_{0.5}Na_{0.5})TiO₃ – BaTiO₃ thin film prepared by pulsed laser deposition, *J. Am. Ceram. Soc.* **99**, 2347 (2016).
- [33] L. Porz, 60 years of dislocations in ceramics: A conceptual framework for dislocation mechanics in ceramics, *Int. J. Ceram. Eng. Sci.* **4**, 214 (2022).
- [34] L. V. Maneeshya, V. S. Anitha, S. S. Lekshmy, I. John Berlin, P. B. Nair, G. P. Daniel, P. V. Thomas, and K. Joy, Influence of annealing temperature and oxygen atmosphere on the optical and photoluminescence properties of BaTiO₃ amorphous thin films prepared by sol–gel method, *J. Mater. Sci. Mater. Electron.* **24**, 848 (2013).
- [35] H. Nobarani and M. Asle Zaeem, A molecular dynamics study of domain switching in BiFeO₃ nanofilm under DC electric field, *Comput. Mater. Sci.* **199**, 110718 (2021).
- [36] S. Plimpton, Fast parallel algorithms for short-range molecular dynamics, *J. Comput. Phys.* **117**, 1 (1995).
- [37] S. Kavousi, B. R. Novak, M. I. Baskes, M. Asle Zaeem, and D. Moldovan, Modified embedded-atom method potential for high-temperature crystal-melt properties of Ti–Ni alloys and its application to phase field simulation of solidification, *Modell. Simul. Mater. Sci. Eng.* **28**, 015006 (2020).
- [38] E. Asadi, M. Asle Zaeem, S. Nouranian, and M. I. Baskes, Two-phase solid–liquid coexistence of Ni, Cu, and Al by molecular dynamics simulations using the modified embedded-atom method, *Acta Mater.* **86**, 169 (2015).
- [39] I. Aslam, M. I. Baskes, D. E. Dickel, S. Adibi, B. Li, H. Rhee, M. Asle Zaeem, and M. F. Horstemeyer, Thermodynamic and kinetic behavior of low-alloy steels: An atomic level study using an Fe–Mn–Si–C modified embedded atom method (MEAM) potential, *Materialia* **8**, 100473 (2019).

- [40] A. Mahata and M. Asle Zaeem, Size effect in molecular dynamics simulation of nucleation process during solidification of pure metals: Investigating modified embedded atom method interatomic potentials, *Modell. Simul. Mater. Sci. Eng.* **27**, 085015 (2019).
- [41] Y. Chen, B. Liu, Y. Ma, and Y. Zhou, Modification of a shell model for the study of the radiation effects in BaTiO₃, *Nucl. Instrum. Methods B* **267**, 3090 (2009).
- [42] J. Wang, Y. Shen, F. Song, F. Ke, Y. Bai, and C. Lu, Effects of oxygen vacancies on polarization stability of barium titanate, *Sci. China: Phys. Mech. Astron.* **59**, 634602 (2016).
- [43] V. Boddu, F. Endres, and P. Steinmann, Molecular dynamics study of ferroelectric domain nucleation and domain switching dynamics, *Sci. Rep.* **7**, 806 (2017).
- [44] A. J. Klomp, R. Khachatryan, T. Wallis, K. Albe, and A. Grünebohm, Thermal stability of nanoscale ferroelectric domains by molecular dynamics modeling, *Phys. Rev. Mater.* **6**, 104411 (2022).
- [45] Y. Zhang, J. Hong, B. Liu, and D. Fang, Molecular dynamics investigations on the size-dependent ferroelectric behavior of BaTiO₃ nanowires, *Nanotechnology* **20**, 405703 (2009).
- [46] T. Tsuzuki, S. Ogata, R. Kobayashi, M. Uranagase, S. Shimoi, D. Durdiev, and F. Wendler, Vacancy-assisted ferroelectric domain growth in BaTiO₃ under an applied electric field: A molecular dynamics study, *J. Appl. Phys.* **131**, 194101 (2022).
- [47] H. Azuma, S. Ogata, R. Kobayashi, M. Uranagase, T. Tsuzuki, D. Durdiev, and F. Wendler, Microscopic structure and migration of 90° ferroelectric domain wall in BaTiO₃ determined via molecular dynamics simulations, *J. Appl. Phys.* **133**, 104101 (2023).
- [48] K.-P. Schröder and J. Sauer, Potential functions for silica and zeolite catalysts based on ab initio calculations. 3. A shell model ion pair potential for silica and aluminosilicates, *J. Phys. Chem.* **100**, 11043 (1996).
- [49] J. S. Tan, S. X. Boerrigter, R. P. Scaringe, and K. R. Morris, Core-shell potential-derived point charges, *J. Comput. Chem.* **33**, 950 (2012).
- [50] F. Ahmad, I. Naz, J. K. Jang, and J. Rhee, Stability of the crystal structure of α -BiFeO₃, *J. Kor. Phys. Soc.* **70**, 394 (2017).
- [51] J. Guo, *Computational Study of Infrared Spectra of Silica Polymorphs Via Classical Mechanics* (University of Missouri, Columbia, 2017).
- [52] J.-S. Lee, J. Ji, S.-H. Oh, and B.-J. Lee, A second nearest-neighbor modified embedded-atom method combined with a charge equilibration interatomic potential for the Al-O binary system, *Comput. Mater. Sci.* **230**, 112505 (2023).
- [53] E. Lee, K.-R. Lee, M. I. Baskes, and B.-J. Lee, A modified embedded-atom method interatomic potential for ionic systems: 2NNMEAM+Qeq, *Phys. Rev. B* **93**, 144110 (2016).
- [54] E. Lee, K.-R. Lee, and B.-J. Lee, An interatomic potential for the Li-Co-O ternary system, *Comput. Mater. Sci.* **142**, 47 (2018).
- [55] E. Lee, K.-R. Lee, and B.-J. Lee, Interatomic potential of Li-Mn-O and molecular dynamics simulations on Li diffusion in spinel Li_{1-x}Mn₂O₄, *J. Phys. Chem. C* **121**, 13008 (2017).
- [56] J. Ji and B.-J. Lee, A second nearest-neighbor modified embedded-atom method combined with a charge equilibration interatomic potential for the Li-Ni-O ternary system and Li diffusion in lithium-ion battery cathode structure, *J. Power Sources* **542**, 231790 (2022).
- [57] J. Ji and B.-J. Lee, Analyzing the effect of Li/Ni intermixing on Ni-rich layered cathode structures using atomistic simulation of the Li-Ni-Mn-Co-O quinary system, *J. Power Sources* **556**, 232535 (2023).
- [58] S. Lahkar, V. Bragaglia, B. Bagheri, D. Falcone, M. Galetta, M. Sousa, and A. Todri-Sanial, Decoupling local electrostatic potential and temperature-driven atomistic forming mechanisms in TaO_x/HfO₂-based ReRAMs using reactive molecular dynamics simulations, *ACS Appl. Mater.* **18**, 8369 (2026).
- [59] B.-J. Lee and M. I. Baskes, Second nearest-neighbor modified embedded-atom-method potential, *Phys. Rev. B* **62**, 8564 (2000).
- [60] M. I. Baskes, Modified embedded-atom potentials for cubic materials and impurities, *Phys. Rev. B* **46**, 2727 (1992).
- [61] S.-G. Kim, M. Horstemeyer, M. Baskes, M. Rais-Rohani, S. Kim, B. Jelinek, J. Houze, A. Moitra, and L. Liyanage, Semi-empirical potential methods for atomistic simulations of metals and their construction procedures, *J. Eng. Mater. Technol.* **131**, 041210 (2009).
- [62] B. Jelinek, S. Groh, M. F. Horstemeyer, J. Houze, S.-G. Kim, G. J. Wagner, A. Moitra, and M. I. Baskes, Modified embedded atom method potential for Al, Si, Mg, Cu, and Fe alloys, *Phys. Rev. B* **85**, 245102 (2012).
- [63] J. A. Harrison, J. D. Schall, S. Maskey, P. T. Mikulski, M. T. Knippenberg, and B. H. Morrow, Review of force fields and intermolecular potentials used in atomistic computational materials research, *Appl. Phys. Rev.* **5**, 031104 (2018).
- [64] H. Zhou, Interatomic potential of multiphase materials using modified embedded atom method (MEAM) and artificial neural network (RANN), Theses and Dissertations 6727, Mississippi State University, 2025, <https://scholarsjunction.msstate.edu/td/6727>.
- [65] A. K. Rappe and W. A. Goddard, III, Charge equilibration for molecular dynamics simulations, *J. Phys. Chem.* **95**, 3358 (1991).
- [66] P. Ghosez, First-principles study of the dielectric and dynamical properties of barium titanate, Ph.D. thesis, Université Catholique de Louvain, 1997.
- [67] M. H. Müser, S. V. Sukhomlinov, and L. Pastewka, Interatomic potentials: Achievements and challenges, *Adv. Phys. X* **8**, 2093129 (2023).
- [68] D. C. Arnold, Composition-driven structural phase transitions in rare-earth-doped BiFeO₃ ceramics: A review, *IEEE Trans. Ultrason. Ferroelectr. Freq. Control* **62**, 62 (2015).
- [69] J.-J. Wang, B. Wang, and L.-Q. Chen, Understanding, predicting, and designing ferroelectric domain structures and switching guided by the phase-field method, *Annu. Rev. Mater. Res.* **49**, 127 (2019).
- [70] P. Ghosez, J. P. Michenaud, and X. Gonze, Dynamical atomic charges: The case of ABO₃ compounds, *Phys. Rev. B* **58**, 6224 (1998).
- [71] Y. Okuno and Y. Sakashita, Born effective charges and piezoelectric coefficients of BiXO₃, *Jpn. J. Appl. Phys.* **48**, 09KF04 (2009).
- [72] A. Stukowski, Visualization and analysis of atomistic simulation data with OVITO—the open visualization tool, *Modell. Simul. Mater. Sci. Eng.* **18**, 015012 (2010).
- [73] P. Agrawal, M. Campanini, A. Rappe, S. Liu, V. Grillo, C. Hébert, R. Erni, D. Passerone, and M. D. Rossell, Structure and

- properties of edge dislocations in BiFeO₃, *Phys. Rev. Mater.* **3**, 034410 (2019).
- [74] D. J. Evans and B. L. Holian, The nose–hoover thermostat, *J. Chem. Phys.* **83**, 4069 (1985).
- [75] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/7m6v-pcrq> for the radial distribution function between Fe-Fe, Bi-Bi, and O-O pairs (Fig. S1); comparison of the time evolution of polarization change for a defect free system and overall DD1 system (Fig. S2); time evolution of total energy during an incremental increase of electric loading (Fig. S3); the radial distribution function for the MD simulation system before/after application of electric field (Fig. S4); the evolution of average CN and charge across the $x-y$ plane (Fig. S5); summary of the hysteresis characteristics from literature (Table S1); the evolution of average charge in various regions for DD1 system (Table S2).
- [76] X. Qi, W.-C. Chang, J.-C. Kuo, I.-G. Chen, Y.-C. Chen, C.-H. Ko, and J.-C.-A. Huang, Growth and characterisation of multiferroic BiFeO₃ films with fully saturated ferroelectric hysteresis loops and large remanent polarisations, *J. Eur. Ceram. Soc.* **30**, 283 (2010).
- [77] T. Rojac, M. Kosec, B. Budic, N. Setter, and D. Damjanovic, Strong ferroelectric domain-wall pinning in BiFeO₃ ceramics, *J. Appl. Phys.* **108**, 074107 (2010).
- [78] R. R. Das, D. M. Kim, S. H. Baek, C. B. Eom, F. Zavaliche, S. Y. Yang, R. Ramesh, Y. B. Chen, X. Q. Pan, X. Ke, M. S. Rzchowski, and S. K. Streiffer, Synthesis and ferroelectric properties of epitaxial BiFeO₃ thin films grown by sputtering, *Appl. Phys. Lett.* **88**, 242904 (2006).
- [79] M. L. Jablonski, S. Liu, C. R. Winkler, A. R. Damodaran, I. Grinberg, L. W. Martin, A. M. Rappe, and M. L. Taheri, Asymmetric response of ferroelastic domain-wall motion under applied bias, *ACS Appl. Mater. Interfaces* **8**, 2935 (2016).
- [80] G. L. Yuan, Y. Yang, and S. W. Or, Aging-induced double ferroelectric hysteresis loops in BiFeO₃ multiferroic ceramic, *Appl. Phys. Lett.* **91**, 122907 (2007).
- [81] J. Wu, J. Wang, D. Xiao, and J. Zhu, Resistive hysteresis in BiFeO₃ thin films, *Mater. Res. Bull.* **46**, 2183 (2011).
- [82] H. Liu and X. Wang, Rectangular and saturated hysteresis loops of BiFeO₃ film on LaNiO₃ bottom electrode, *J. Alloys Compd.* **485**, 769 (2009).
- [83] A. Anwar, M. A. Basith, and S. Choudhury, From bulk to nano: A comparative investigation of structural, ferroelectric and magnetic properties of Sm and Ti co-doped BiFeO₃ multiferroics, *Mater. Res. Bull.* **111**, 93 (2019).
- [84] F. Zhuo, X. Zhou, F. Dietrich, M. Soleimany, P. Breckner, P. B. Groszewicz, B.-X. Xu, G. Buntkowsky, and J. Rödel, Dislocation density-mediated functionality in single-crystal BaTiO₃, *Adv. Sci.* **11**, 2403550 (2024).
- [85] D. Wang, P. Wang, S. Mondal, Y. Xiao, M. Hu, and Z. Mi, Impact of dislocation density on the ferroelectric properties of ScAlN grown by molecular beam epitaxy, *Appl. Phys. Lett.* **121**, 042108 (2022).
- [86] G. Cao, H. Huang, and X. Ma, Thickness dependence of switching behavior in ferroelectric BiFeO₃ thin films: A phase-field simulation, *Appl. Sci.* **7**, 1162 (2017).
- [87] R.-C. Peng, X. Cheng, B. Peng, Z. Zhou, L.-Q. Chen, and M. Liu, Domain patterns and super-elasticity of freestanding BiFeO₃ membranes via phase-field simulations, *Acta Mater.* **208**, 116689 (2021).
- [88] N. Aruchamy, T. Schenk, V. Kovacova, S. Glinsek, E. Defay, and T. Granzow, Influence of tensile vs. compressive stress on fatigue of lead zirconate titanate thin films, *J. Eur. Ceram. Soc.* **41**, 6991 (2021).
- [89] H. Zhu, D. P. Chu, N. A. Fleck, I. Pane, J. E. Huber, and E. Natori, Polarization change of pzt ferroelectric thin films under uniform in-plane tensile stress, *Integr. Ferroelectr.* **95**, 117 (2007).
- [90] T. Kumazawa, Y. Kumagai, H. Miura, M. Kitano, and K. Kushida, Effect of external stress on polarization in ferroelectric thin films, *Appl. Phys. Lett.* **72**, 608 (1998).
- [91] M. D. Biegalski, D. H. Kim, S. Choudhury, L. Q. Chen, H. M. Christen, and K. Dörr, Strong strain dependence of ferroelectric coercivity in a BiFeO₃ film, *Appl. Phys. Lett.* **98**, 142902 (2011).
- [92] Y. Nakamura, S. Nakashima, and M. Okuyama, BiFeO₃ thin films prepared by chemical solution deposition with approaches for improvement of ferroelectricity, in *Ferroelectrics—Material Aspects*, edited by M. Lallart (IntechOpen, London, 2011).
- [93] P. Ravindran, R. Vidya, A. Kjekshus, H. Fjellvåg, and O. Eriksson, Theoretical investigation of magnetoelectric behavior in BiFeO₃, *Phys. Rev. B* **74**, 224412 (2006).
- [94] J. B. Neaton, C. Ederer, U. V. Waghmare, N. A. Spaldin, and K. M. Rabe, First-principles study of spontaneous polarization in multiferroic BiFeO₃, *Phys. Rev. B* **71**, 014113 (2005).