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ABSTRACT

Grain boundaries (GBs) are one of the main factors influencing the polar domain evolution of polycrystalline ferroelectrics. However, domain switching from GBs to grains remains an unsolved aspect. Previous microscopic GB assumptions hinder such theoretical investigations, assuming that the structure and properties of GB are independent of the misorientation of adjacent grains. This work investigates the competition between the energy densities and domain-switching pathways based on the formation mechanism of the GB model. It is found that the domain-switching pathways in polycrystalline ferroelectrics follow three rules: (1) domain switching occurs near low-energy-density GBs; (2) the development of domain-switching pathway originates near the low-energy-density GBs. This pathway ultimately influences the overall domain-switching process, which follows the energy minimization principle; and (3) the domain-switching trend expands to both sides of the pathways after complete formation. The domain evolution rules for polycrystalline ferroelectric materials proposed in this work are conducive to improving the performance of ferroelectric ceramics via GB engineering.

Ferroelectric ceramics are widely used in microelectronic devices due to their excellent electromechanical coupling properties.[1] The electromechanical coupling behavior of ferroelectric ceramics is related to the evolution of polarization domains. For most polycrystalline ferroelectric ceramics, the mechanism of how the domain switches from the grain boundaries (GBs) to inside the grain body remains an area to be investigated. The domain configuration in polycrystalline ferroelectrics can be regulated by an applied electric field or mechanical stress.[2–4] Domain evolution is related to complex energy-density competitive processes,[5–7] causing a macroscopic nonlinear response. There is a consensus that the origin of domain evolution occurs near the GBs.[8] The lack of understanding of the mechanism for domain switching from the GBs to inside the grain body limits the applications of grain boundary engineering in regulating the domain structure and electromechanical properties.

The evolution of polar domains has been investigated in several microscopic theoretical studies. Hwang *et al.* summarized the energy-based domain switching criterion through the Preisach hysteresis model and demonstrated that changes in electric and elastic energy density are the main factors inducing domain switching.[9] Huber *et al.* developed a constitutive model, simulated the domain switching process, and estimated the macroscopic response of tetragonal ferroelectric crystals under different loading conditions.[10] Su and Landis developed a two-level micromechanical model coupling the nonlinear response effect.[11] However, the above-mentioned theoretical models focused on the effect of grains on domain switching, which is insufficient when the influence of GBs becomes more significant. For instance, it is important to analyze the role of GBs in the energy storage and electrical breakdown performances of capacitor materials.[12,13]

Hence, an appropriate description of GBs is crucial. According to thermodynamic principles, a phase-field method was introduced to investigate the domain evolution processes. In accordance with the mathematical boundary assumption of GB, Choudhury *et al.* investigated the process of domain evolution and found that it strongly correlates with the orientation of the grains in polycrystalline ferroelectric materials.[14,15] However, the atomic-scale structure of GBs is complex, with random distributions of oxygen vacancies, as well as dislocations and migration of defects,[16] which reduces the lattice symmetry and ferroelectricity of GBs. Therefore, the "dead layer" hypothesis has been proposed to describe GBs as a dielectric material with the spontaneous polarization of zero. Based on this hypothesis, it has been reported that domain switching occurs in regions near large-angle GBs.[17] Further discussions about these assumptions and the structures of GB are included in the supplementary material. Su *et al.* assumed the GBs to be homogeneous and weakly ferroelectric when studying their rate-dependent domain switching.[18] This assumption has some advantages, as it depicts a continuum of domain polarization inside the GBs. Nevertheless, the aforementioned results were obtained under the postulation that the structures of the GBs are homogeneous and independent of the grains, which is not consistent with the experimental findings.[19]

The structure of the GBs plays an important role in domain evolution.[20,21] An appropriate assumption of GBs that should be taken into account is the influence of the misorientation between adjacent grains on the structure of GBs, as this has a significant impact on their mechanical and electrical properties. The establishment of mathematical requirements for domain continuity over GBs[22] and the corresponding experimental observations[23] necessitate the study of domain evolution in polycrystalline ferroelectrics from a more comprehensive perspective.

Based on the microscopic formation mechanism of the GB model, which was proposed in the previous work,[24] this work investigates the competitive relationship of energy densities and analyzes the mechanism of domain switching from GB to the grain body during the domain evolution process. The aim is to provide a perspective for regulating domain evolution with rational grain distribution. The details of the new GB model used in this work are presented in the supplementary material. Domain evolution in polycrystalline ferroelectrics has received increasing attention, and understanding the internal mechanisms of domain evolution will aid in designing ferroelectric ceramic devices.

A phase-field method reliant on the time-dependent Ginzburg–Landau (TDGL) equation is adopted to investigate the domain evolution.[25] The free-energy density of polycrystalline ferroelectrics can be expressed as a function of local polarization (P_i^L), polarization gradient ($P_{i,j}^L$), strain (ε_{ij}^L), and electric field (E_i^L), where $i, j = 1, 2, 3$, and the superscript L denotes the local coordinates of the individual grain and GBs.[26] The free-energy density can be expressed as the sum of the bulk free energy density (f_{bulk}), the elastic energy density (f_{elas}), the coupling energy density (f_{coup}), the gradient energy density (f_{grad}), and the electric energy density (f_{elec}) as[27]

$$f(P_i^L, P_{i,j}^L, \varepsilon_{ij}^L, E_i^L) = f_{bulk}(P_i^L) + f_{elas}(\varepsilon_{ij}^L) + f_{coup}(P_i^L, \varepsilon_{ij}^L) + f_{grad}(P_{i,j}^L) + f_{elec}(P_i^L, E_i^L). \quad (1)$$

The specific expression of Eq. (1) follows the previous literature,[28] which has been added to the supplementary material.

To investigate the impact of GBs on the electromechanical coupling properties of polycrystalline ferroelectrics, it is essential to consider the domain distribution within the GB. In this work, the symmetric tilt [001] GB is regarded as weakly ferroelectric, and its structure is related to the orientation of adjacent grains.[29] Hence, the ferroelectric, mechanical, and electrical properties [e.g., the Curie–Weiss temperature (T_{gb}), elastic coefficients (c_{gb}), and permittivity (κ_{gb})] of the GBs are affected by the grain orientation. The specific expression of those properties follows the formation mechanism of the GB model, which is represented in the supplementary material.[24] The accumulated space-charge density induced by defect of the GB is described as

$$Q_{gb(m-n)} = Q \ \sin(2\Delta\theta_{(m-n)}), \quad (2)$$

where Q is the maximum space-charge density in the GBs, and $\Delta\theta_{(m-n)}$ denotes the misorientation of adjacent grains $\boldsymbol{m}$ and $\boldsymbol{n}$.

The orientation of the grains and GBs are represented by a set of Euler angles, and the order parameters are used in a conversion formula to convert local coordinates into global coordinates.[30]

The total free energy of a polycrystalline ferroelectric (F) can be expressed by integrating f over its whole volume V. The evolution of P can be described as[31]

$$\frac{\partial P_i(\mathbf{r}, t)}{\partial t} = -L\frac{\delta F}{\delta P_i(r, t)}, \quad (3)$$

where L is the kinetic coefficient of the domain switching. The mechanical equilibrium equation $\partial(\partial f/\partial\varepsilon_{ij})/\partial x_i = 0$ and Maxwell's equation $\partial(-\partial f/\partial E_i)/\partial x_i = 0$ must also be satisfied.

In this work, a GB formation mechanism model using high symmetry tetragonal $BaTiO_3$ with strong continuity is constructed to investigate the effects of GBs on domain evolution. The discussion about the continuity of the GBs is represented in the supplementary material. The periodic polycrystalline ferroelectric model is shown in Fig. 1(a), and the orientations of the grains are set to 0°, 30°, 45°, and 15°. The GB structure is influenced by adjacent grains, and differences in the GB structure leads to changes in the domain distribution between grains.

The GB is regarded as weakly ferroelectric, and the saturation polarization (P_s) and the coercive electric field (E_c) are smaller than those of the grains.[16] The switching criteria of the domain structure are represented as[9]

$$\sigma_{ij}\Delta\varepsilon_{ij} + E_i\Delta P_i \geq 2P_sE_c, \quad (4)$$

where σ_{ij} and ε_{ij} represent the stress and strain tensors, and E_i and P_i denote the electric field and polarization intensity. The first term on the left side of Eq. (4) represents the change in the elastic energy density, and the second term is the change in the electrostatic energy density. Equation (4) indicates that the elastic and electrostatic energy densities induce domain switching. These two energy densities exhibit a competing relationship during the switching process. The evolution of the energy densities and polarization with the varying electric field strength are shown in Figs. 1(b) and 1(c).

With a decrease in electric field strength, the electrostatic energy density decreases significantly. In contrast, the elastic energy density increases slowly, and the change in both curves is approximately linear (period I). During this period, almost all domains point in the direction of the electric field, and the linear attenuation of the electrostatic

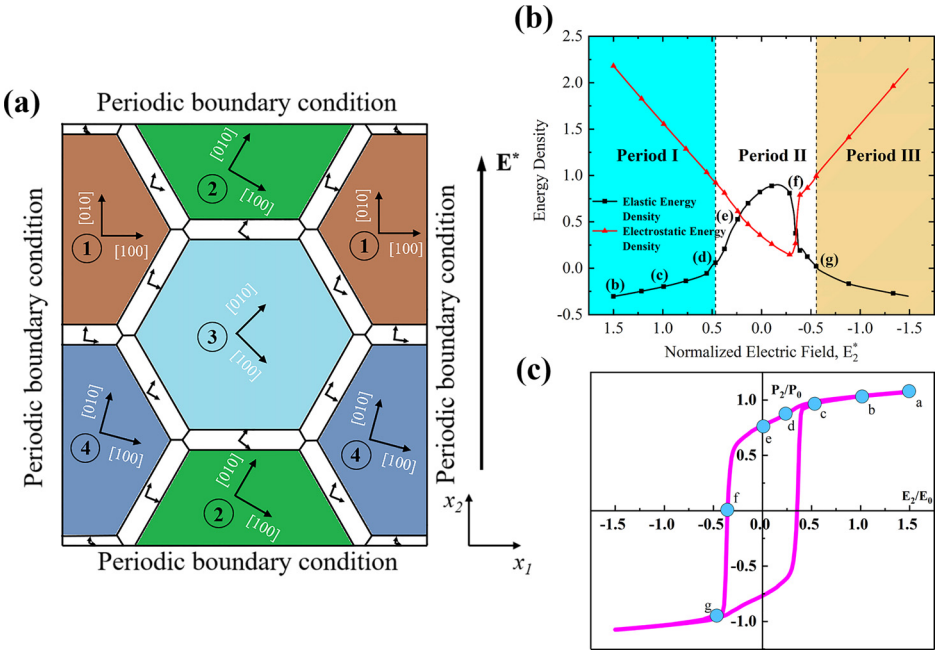

FIG. 1. The periodic polycrystalline ferroelectric model and the evolution of energy densities and polarization with varying electric field strength. (a) An x_2-direction electric field is applied, and the size of the simulation model is 265 × 306 nm. The proportion of the GB is expanded to 18.6% to capture the phenomenon in the GB. (b) The evolution of elastic energy density (the bold black line) and the electrostatic energy density (the bold red line) with the electric field. (c) The P–E hysteresis loop curve ($E_0 = 96.5$ kV/cm and $P_0 = 0.26$ C/m^2).

energy density leads to a decay of the polarization intensity along the GBs. The large energy density difference between the GBs and grains hinders domain switching from the GBs to the grains, as shown in Figs. 2(a)–2(c). The nonlinear trend of the elastic energy density curve increases as the electric field decreases to $E/E_0 = 0.495$ (period II). During this period, the increasing elastic energy density of the grains decreases the difference in energy density between the grains and GBs, which promotes the spreading of domain switching into the grains. The elastic energy density is an important factor that complicates the domain structure. The energy density curves reach a maximum rate of

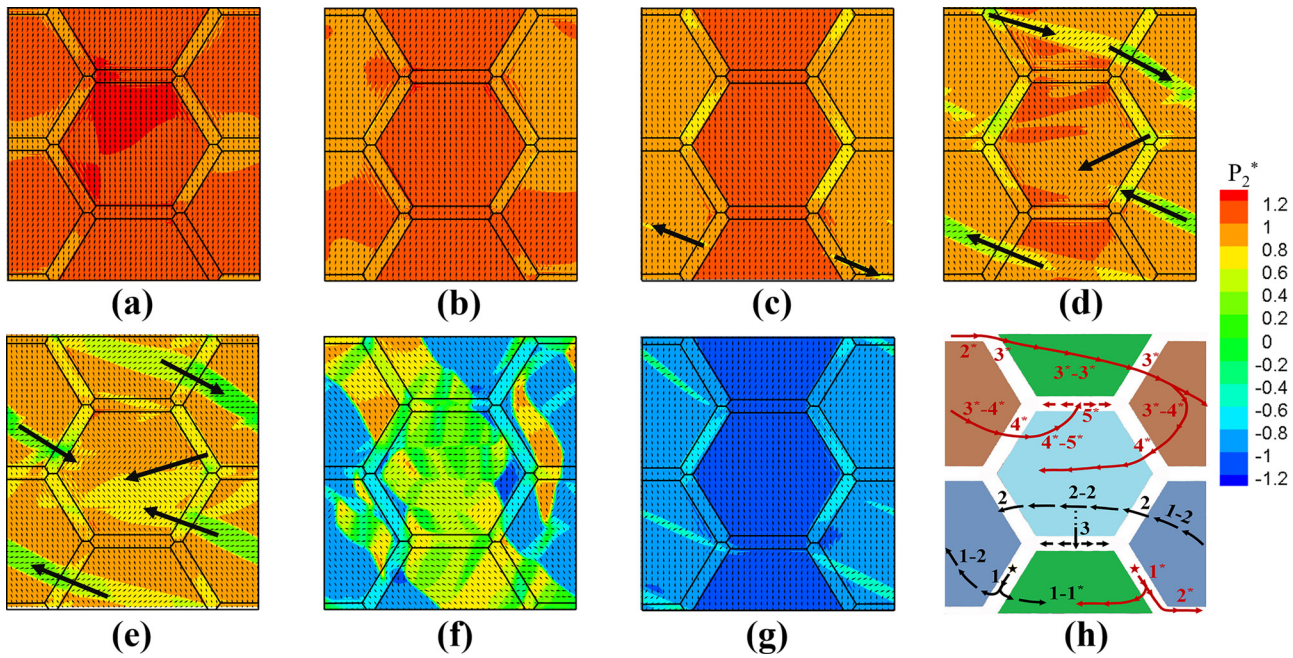

FIG. 2. The domain switching process with an applied electric field. The domain structure pattern with (a) $E_2/E_0 = 1.5$, (b) $E_2/E_0 = 1.065$, (c) $E_2/E_0 = 0.525$, (d) $E_2/E_0 = 0.24$, (e) $E_2/E_0 = 0$, (f) $E_2/E_0 = -0.345$, and (g) $E_2/E_0 = -0.525$ (h) Two domain switching pathways (the bold black line and bold red line), where the number indicates the domain switching order at the GBs.

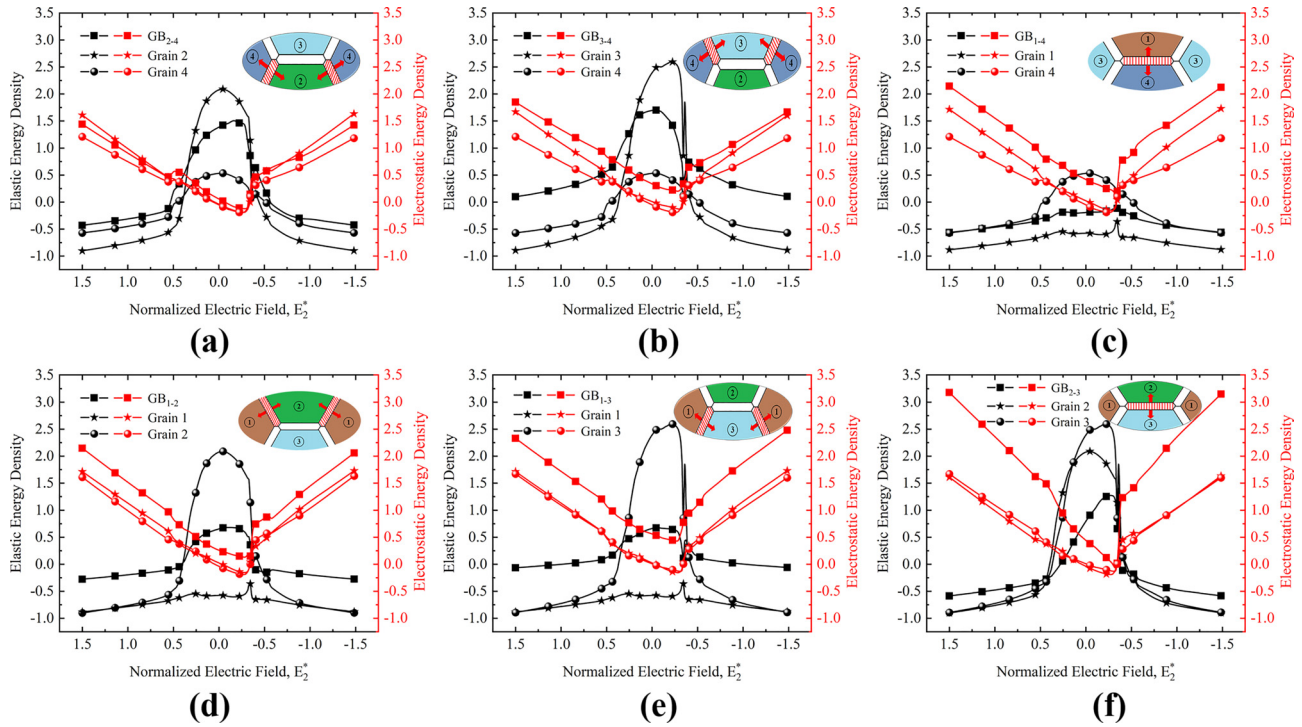

FIG. 3. The relationship between energy densities and external electric field of GBs and grains. (a) $GB_{2\text{-}4}$ and adjacent grains, (b) $GB_{3\text{-}4}$ and adjacent grains, (c) $GB_{1\text{-}4}$ and adjacent grains, (d) $GB_{1\text{-}2}$ and adjacent grains, (e) $GB_{1\text{-}3}$ and adjacent grains, and (f) $GB_{2\text{-}3}$ and adjacent grains.

change at $E/E_0 = -0.345$, which leads to the switching of the domain direction. In contrast, the total polarization intensity is approximately zero, and the domain structure presents the most complex state during the entire process, as shown in Figs. 2(d)–2(f). The effect of the elastic energy density gradually weakens as the electric field strength is further reduced to $E/E_0 = -0.525$ (period III). During this period, the electrostatic energy density becomes the main factor leading to the orientation of the domain structure to the electric field while also enhancing the polarization intensity, as shown in Fig. 2(g).

From the whole domain switching process, two switching pathways are formed as the electric field decreases [Fig. 2(h)], namely, $GB_{2-4} \rightarrow GB_{3-4}$ and $GB_{2-4} \rightarrow GB_{1-4} \rightarrow GB_{1-2} \rightarrow GB_{1-3} \rightarrow GB_{2-3}$. The energy evolution of the first domain-switching pathway, $GB_{2-4} \rightarrow GB_{3-4}$, is shown in Figs. 3(a) and 3(b). The electrostatic energy density of GB_{2-4} is much lower than that of the other GBs, indicating that domain switching is most likely to occur in this region. The switching trend spreads to the lower-energy-density grain, that is, grain 4, rather than through the triple-point GB. These mentioned observed results are consistent with recent experimental literature.[32] Two switching pathways are then formed because of the similar energy densities of GB_{3-4} and GB_{1-4}, as demonstrated by comparing Figs. 3(b) and 3(c). This phenomenon indicates that the domain-switching pathway spreads along the lower-energy direction.

The second domain switching pathway, $GB_{2-4} \rightarrow GB_{1-4} \rightarrow GB_{1-2} \rightarrow GB_{1-3} \rightarrow GB_{2-3}$, is shown in Figs. 3(c)–3(f). The similar energy density values of the grains cause the switching pathways in grains 1 and 2 to form approximately simultaneously, as shown in Figs. 3(d) and 2(d). Although the energy density of GB_{2-3} is significantly larger than that of GB_{1-3} [comparing Fig. 3(e) with Fig. 3(f)], the switching pathway in grain 1 is more likely to spread to GB_{1-3}, as shown in Fig. 2(e). As the electric field strength decreases further, the pathway spreads to GB_{2-3}, forming a complete switching pathway. Domain switching spreads along both sides of the pathway, as shown in Fig. 2(f). The formation of the second domain-switching pathway indicates that the energy minimization principle governs the domain evolution process of polycrystalline ferroelectrics.

Upon the foundation of the above-mentioned analysis, the domain evolution rules can be proposed as follows: (1) the nucleus of domain switching occurs near low-energy-density GBs; (2) the development of domain-switching pathway originates near the low-energy-density GBs. This pathway ultimately influences the overall domain-switching, which follows the energy minimization principle; and (3) domain switching expands to both sides of the pathway once complete pathways are formed.

These domain evolution rules help deepen our understanding of the mechanism of domain switching in polycrystalline ferroelectrics.

According to the above-mentioned domain evolution rules and the formation mechanism of the GB model, the switching of the domain from the GBs to the grain body results from a competition between elastic and electric energies. This work analyzed this competitive relationship and proposed three main rules for the formation of domain switching pathways, where two pathways for domain evolution from GBs into the grain body are formed. These results clarify the domain evolution of polycrystalline materials. It should be noted that

this study focuses on the domain switching of large-sized domains. However, as the grain size varies, domain variations become more complex which complicates the process of domain switching.[33] Thus, investigating the effect of domain size on the switching process would improve our understanding of domain switching. This work is conducive to improving the performance of ferroelectric ceramics through the rational distribution and design of grains and GBs.

See the supplementary material for energy equation of phase field method; dimensionless method in simulation and the material properties of $BaTiO_3$; the schematic diagram of mathematical interface and "dead layer" GB assumptions; the detail of formation mechanism GB model; the discussion about the complex structure of GBs; and the effect of GB's continuity on domain switching.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Xuhui Lou: Conceptualization (equal); Data curation (equal); Formal analysis (equal); Investigation (equal); Methodology (equal); Software (equal); Validation (equal); Visualization (equal); Writing – original draft (equal); Writing – review & editing (equal). **Xu Hou:** Data curation (equal); Formal analysis (equal); Methodology (equal); Software (equal); Validation (equal); Writing – review & editing (equal). **Yujun Chen:** Data curation (equal); Formal analysis (equal); Visualization (equal); Writing – review & editing (equal). **Shaohan Cui:** Data curation (equal); Validation (equal); Writing – review & editing (equal). **Jie Wang:** Data curation (equal); Formal analysis (equal); Methodology (supporting); Software (supporting); Writing – review & editing (equal). **Qingyuan Wang:** Resources (equal); Writing – review & editing (equal). **haidong fan:** Resources (equal); Writing – review & editing (equal). **Xiaobao Tian:** Conceptualization (equal); Data curation (equal); Formal analysis (equal); Funding acquisition (equal); Investigation (equal); Methodology (equal); Project administration (equal); Resources (equal); Supervision (equal); Validation (equal); Visualization (equal); Writing – review & editing (equal).

DATA AVAILABILITY

The data that support the findings of this study are available within the article and its supplementary material.

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