Quantitative study of the pinning effect of the edge dislocation on domain wall motion in Barium Titanate thin films

Shuai Wang[a] and Li-Hua Shao *[a]

[a]National Key Laboratory of Strength and Structural Integrity, Institute of Solid Mechanics, School of Aeronautic Science and Engineering, Beihang University, Beijing 100191, China

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Abstract

Dislocation is a very important one-dimensional defect in ferroelectrics. This work introduces an easy and flexible model of implementing the edge dislocation by introducing eigenstrain at the interface, and it could be easily extended to incorporate the surface stress to refine the analysis of ferroelectric thin films. The influence of dislocations on the ferroelectric domain wall motion and hysteresis loop including the remanent polarization and coercive field using phase-field simulations is analyzed. The pinning effect of the dislocation on the domain wall motion is discussed and whether the domain wall is pined is the competition between the external loading and the magnitude of the burgers vector of the dislocation. This work could contribute to the understanding of the pining effect of the dislocation and provide guidance for the fabrication of ferroelectric thin films.

Ferroelectrics are essential components in a wide spectrum of applications due to their unique electro-mechanical coupling properties. [1] The nonlinear properties of the electromechancial coupling effect of ferroelectrics originate from the spontaneous polarization. The spontaneous polarization can be switched by external field or mechanical loading via the nucleation and growth of a more energetically favored domains through a highly inhomogeneous process, whereby local variations in free energy from defects dominate the switching kinetics. [2] The ferroelectric domain wall represents the transition region of the polarization between neighbouring domains and the domain wall motion is responsible for the non-linear dielectric, piezoelectric and elastic properties of ferroelectrics. Defects and material inhomogeneities such as oxygen vacancies, point defects and dislocations play a crucial role when explaining the abnormal phenomenons during the domain wall motion such as pining effect at the micro scale. The pining effect of the domain wall could be found in either bicrystal grain boundary [3] polycrystalline gain boundaries [4] or at the defect regions in thin films.

*Corresponding author:shaolihua@buaa.edu.cn

For instance, Dragan *et al.* provide experimental evidence that the domain-wall movement in $BiFeO_3$ is strongly inhibited by charged defects and the domain-wall mobility can be considerably increased by preventing the defects from migrating into their stable configuration. [5]

Compared with their bulk counterparts, ferroelectric thin films are essential components in applications at micro/nano scale, such as microsystems [6], memory devices [7], and high frequency electrical components[8]. The divergence between the properties of bulk ferroelectrics and thin films comes from the surface effect, large depolarization field, film/substrate interface, epitaxial stress and so on. Dislocations are one-dimensional defects that commonly exist in polycrystallines and thin films. [9] The dislocation densities in bulk ferroelectric single crystals [10] and ceramics mainly affect the mechanical properties and hysteresis through the plastic deformation. While for the epitaxial films and heterostructures, the dislocations are almost unavoidable and has a direct influence on the domain topology. The dislocations in thin films has been extensively studied. [11] The strain due to the misfit strain between the thin film layer and a substrate may be accommodated by the misfit dislocations at the interface. [12] The role of the misfit dislocation is to release the mechanical energy caused by the misfit strain. The existence of the dislocations make the ferroelectric thin films excellent candidates for innovative device concepts, ranging from dislocation-based memory devices to light emission diodes [13].

The role of the dislocations on the domain wall motion and macroscopic properties in ferroelectric thin films has been extensively studied in the past decades. Dai *et al.* [14] first point out the that the dislocations act as pinning sites for the domain boundaries. Antonios and Landis [15] use a phase-field method to systematically investigate the influence of the edge dislocation on the domain configuration. In their model, the dislocation regions are represented by the mechanical/electric boundary conditions. In a recent series of works by Zhou et al. [16, 17, 18] regard dislocations as order parameter to study the interaction between dislocation and domains. Cheng et al. combines STEM and 2-dimensional phase-field simulation and find dislocation pairs are more favorable for the retention of a domains by playing a pinning role. [19] Jiang et al. observes the pining effect of the dislocation on the domain wall in BTO thin films. [20]

In the work by Kröner [21], the elastic field with defects in solids is mathematically described, which led to solving the stress field by taking the dislocation induced strain as eigenstrain. In this letter, we perform a quantitative analysis of the pinning effect of the edge dislocation on domain wall motion in ferroelectric thin films. Different from the work of Landis or Zhou, the dislocation in thin film is represented by the misfit eigenstrain at the film-substrate interface. The influence of the dislocation on the hysteresis loops and the domain wall motion is quantitatively analyzed. Finally the criteria of the domain wall-dislocation pinning is given to give a guidance for the ferroelectric thin film fabrication and dislocation-engineered domain tuning.

To model the influence effect of the dislocation on the ferroelectric thin film, three assumptions are proposed:
(1) Only the edge dislocation occurs in the ferroelectric thin film;
(2) The position of the dislocation do not change during the domain evolution;
(3) The influence of the dislocation on the polarization mainly originates from the stress field of the dislocation.

We assume a right-handed Cartesian coordinate system centered at the core of a single, straight dislocation. The dislocation is characterized by the dislocation line vector ξ (with unit length) and the Burgers vector $\boldsymbol{b}$ (with length b). For simplicity, we consider in the current paper only edge dislocations with a line vector oriented in z-direction, $\xi = [0, 0, 1]$, and a Burgers vector pointing into the x-direction, $\boldsymbol{b} = [0, 0, b]$. The dislocation eigenstrain $\varepsilon_{ij}^{\text{dis}}$ is prescribed as one row finite element mesh on the slip plane in the x-direction. The normal component of $\varepsilon_{ij}^{\text{dis}}$ is zero and the shear component of $\varepsilon_{ij}^{\text{dis}} = \frac{b}{l_0}$, where l_0 is the width of the mesh in the y-direction (see Fig. 1(a)). Here we use a uniform size of a four node linear Lagrange element throughout the paper.

Figure 1 shows the contour plots of the stress components σ_{xx}, σ_{yy} and σ_{xy} around the single edge dislocation in the unloaded body. The analytical solutions are taken from the literature [22]. It is seen that the dislocation model proposed here produces stresses that coincide very well with the analytical solution. The normal stress components coincide equally well. The dislocation core is the singularity of the stress field. Differences between modeling results and analytical solution only occur near the dislocation core.

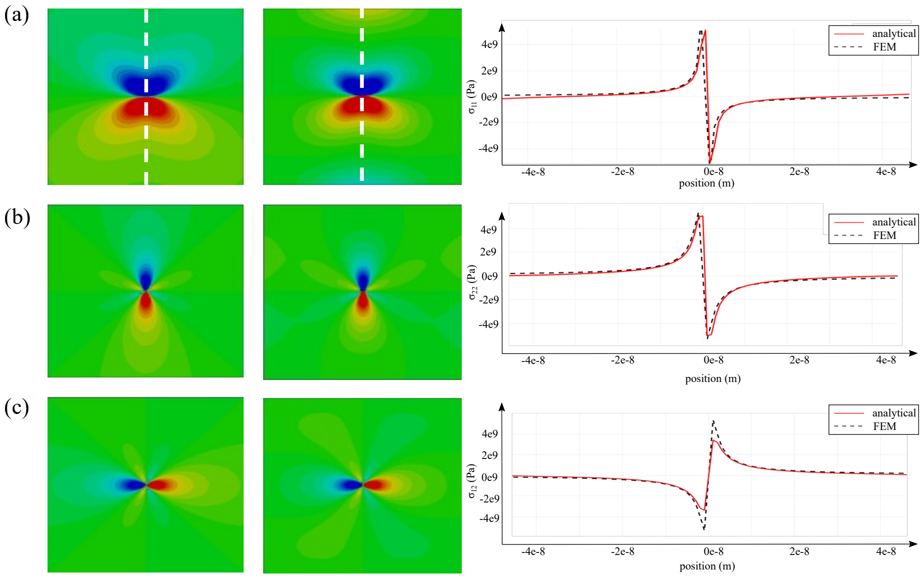

Figure 1: Contour plots of the stress components around a single edge dislocation in an unloaded body: Comparison between analytic solutions: (a) σ_{xx} (b) σ_{yy} (c) σ_{xy}.

Phase-field models have been used extensively to describe various aspects of the behaviors of the ferroelectric thin films. [23] The framework of the ferroelectric model in this letter is based on the previous work of Xu *et al.* [24] and Wang *et al.* [25]. The paraelectric-to-ferroelectric phase transition occurs in a ferroelectric material when its temperature is lower than its Curie point. The spontaneous polarization $\boldsymbol{P}$ is adopted as the order parameter. In phase field simulations, the time-dependent Ginzburg–Landau equation is used to describe the polarization evolution and thus the domain configuration change,

$$\frac{\partial P_i(\boldsymbol{x},t)}{\partial t} = -M\frac{\delta \mathcal{H}}{\delta P_i(\boldsymbol{x},t)} \tag{1}$$

where M governs the mobility of the polarization vector, $\mathcal{H}$ is the electric enthalpy of the system. In the model, the electrical enthalpy is constituted with four parts, i.e.

$$\mathcal{H} = \mathcal{H}^{ela} + \mathcal{H}^{ele} + \mathcal{H}^{coup} + \mathcal{H}^{bulk} + \mathcal{H}^{grad} \tag{2}$$

in which $\mathcal{H}^{ela}$, $\mathcal{H}^{ele}$, $\mathcal{H}^{coup}$, $\mathcal{H}^{bulk}$ and $\mathcal{H}^{grad}$ represent elastic energy density, electrical energy density, mechanical-electric coupling energy density, Landau free energy density and gradient energy, respectively. These energy densities are given in the following form,

$$\begin{cases} \mathcal{H}^{ela} &= \frac{1}{2}c_{ijkl}\varepsilon^e_{ij}\varepsilon^e_{kl} \\ \mathcal{H}^{ele} &= -\frac{1}{2}k_{ij}E_iE_j - P_iE_i \\ \mathcal{H}^{coup} &= -b_{ijk}\varepsilon_{ij}E_k \\ \mathcal{H}^{bulk} &= \beta_1(G,\lambda)(a_{ij}P_iP_j + a_{ijkl}P_iP_jP_kP_l) + a_{ijklmn}P_iP_jP_kP_lP_mP_n \\ \mathcal{H}^{grad} &= \beta_2(G,\lambda)(P_{i,j}P_{k,l}) \end{cases} \tag{3}$$

where $\varepsilon^e_{ij} = (\varepsilon_{ij} - \varepsilon^0_{ij}(\boldsymbol{P}) - \varepsilon^{\mathrm{dis}}_{ij})$ is the elastic contribution to the strain. ε^i_{ij} is the total strain and $\varepsilon^0_{ij}(\boldsymbol{P})$ is the remnant strain induced by the remnant polarization, β_1 and β_2 are the coefficients related to the domain wall energy G and the domain wall thickness λ[26]. Here $\varepsilon^0_{ij}(\boldsymbol{P})$ are calculated following the work by Huo and Jiang [27].

$$\varepsilon^0_{ij}(\boldsymbol{P}) = \frac{3}{2}\varepsilon_{sat}\frac{\sqrt{P_iP_i}}{P_{sat}}(n_in_j - \frac{1}{3}\delta_{i,j}) \tag{4}$$

where n_i is the unit vector of P, ε_{sat} is the maximum remnant strain and P_{sat} is the maximum remnant polarization. Strain tensor c_{ijkl} and permittivity tensor k_{ij} are the same as macroscopic ones. For piezoelectric tensor b_{ijk}, we used the representation

$$\begin{aligned} b_{ijk}(\boldsymbol{P}) =& \frac{\sqrt{P_iP_i}}{P_{sat}}\{d_{33}n_in_jn_k + d_{31}(\delta_{ij} - n_in_j)n_k \\ &+ \frac{1}{2}d_{15}\left[(\delta_{ki} - n_kn_i)n_j + (\delta_{kj} - n_kn_j)n_i)\right)]\} \end{aligned} \tag{5}$$

adopted by Kamlah[28].

Here we use the parameters for barium titanate for the simulation. [29] In the Figure 2 shows the domain configuration of a BTO thin film with different edge dislocation at temperature ranges from 300 K 500 K to 800 K. The change-free boundary and stress-free condition is set for all boundaries and the initial polarization distribution is set random. Figure 2(a) shows that the final configuration of the domain is a vortex. As the temperature rises, the magnitude of the polarization decreases and the energy barrier is lowered, which indicates that the domain configuration is very sensitive to the external stimuli at elevated temperature. If the edge dislocation is introduced to the system (see Fig. 2(b) and Fig. 2(c)), the vortex is twisted near the core and the dislocation with burger's vector pointed to the right pushes the polarization to the vortex center, while the dislocation in with the burger's vector in the opposite direction attracts the polarization near the dislocation core. As the temperature rises, the vortex become less distinct.

Figure 3 shows influences of the dislocation with different direction on the

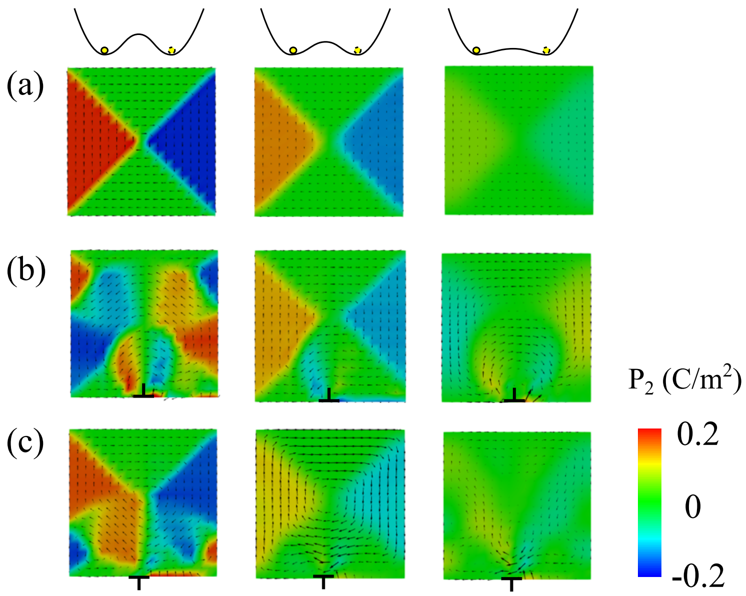

Figure 2: Influence of the edge dislocation on the domain vortex at different temperature. (a)without the edge dislocation. (b) with the edge dislocation at the bottom boundary of the thin film. (Burgers vector pointed to the right) (c) with the edge dislocation at the bottom boundary of the thin film. (Burgers vector pointed to the left) from left to right: 300 K, 500 K, 800 K.

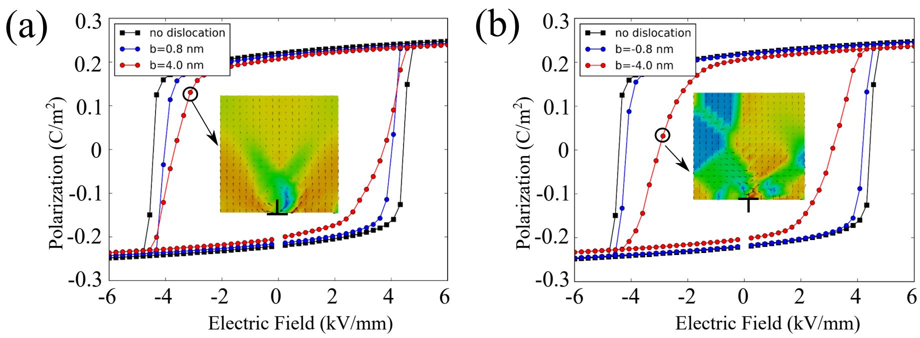

Figure 3: Influence of edge dislocation on the polarization hysteresis loop of the BTO thin film. (a) Burgers vector pointed to the right. (b) Burgers vector pointed to the left

the hysteresis loop. The hysteresis becomes slimmer as the magnitude of the burgers vector increases regardless the direction of the burgers vector. The domain configuration at the same electric field (3 KV/mm) is plotted as the inset figures. Compared with Fig. 3(a), the domain configuration in Fig. 3(b) is more randomized due to the attraction effect from the dislocation core, which makes the overall polarization smaller.

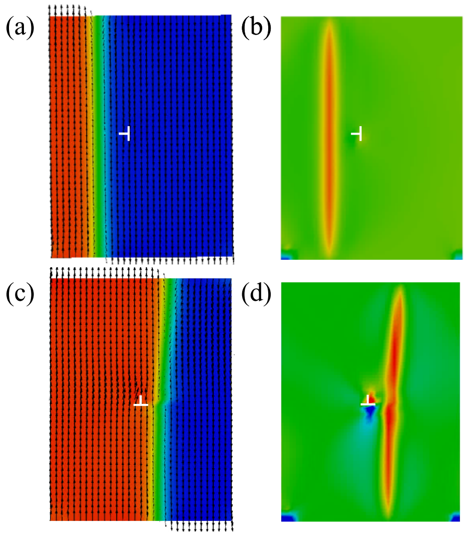

Figure 4: Domain wall motion at the edge dislocation with direction. (a)Burgers vector parallel to the domain wall, legend: P_y (b) Burgers vector perpendicular to the domain wall, legend: σ_{yy}

The last simulation shows the domain wall motion at the edge dislocation. At the initial state, a 180°domain wall is assumed in the y-direction which separates the left domain (upright) from the right domain (downward). An electric potential is given at the top surface to give the driving force for the domain wall motion. Here we use the coercive field E_c as the reference field value and one unit cell length ($b_0 = a = 0.4$ nm) as the reference burgers vector's magnitude.

We first examine the effect of the edge dislocation with burgers vector pointed to the positive y-direction on the domain wall motion. For the applied electric field $E = 3E_c$ and $b = b_0$, the domain wall motion is hindered at the dislocation core. While for the case where the burgers vector pointed to the positive x-direction, the domain wall will pass through the dislocation core. This proves that not only the magnitude but also the direction of the edge dislocation determines the difficulty of domain switching. For the first case shown in Fig.4(a), if one increase the external field to $E = 6E_c$. The domain wall can pass through the edge dislocation call. More simulation cases can be found in the Supplementary Material. To further investigate the antagonism between the strength of the dislocation and the external field strength, a series of simulation is carried out and whether the domain wall successfully pass through the dislocation core is illustrated in Fig. 5. The burgers vector and the external fields are all normalized by b_0 and E_c ($b^\star = b/b_0$ and $E^\star = E/E_c$). For the case without external field, the domain wall does not move. At the coercive field ($E^\star = 1$), the domain wall can only move without edge dislocation. For the same burgers vector, there is a threshold field where the domain wall can pass the dislocation core. For the same external field stimuli, the domain wall can only pass the dislocation core at the lower burgers vector. We use the nonlinear regression algorithm fit the watershed as a dash line in Fig. 5. The exponential fitting of the watershed is $b^\star = -0.088 + (0.024E^\star)^{1.12}$. The slope of the curve is much smaller when $E^\star < 3$.

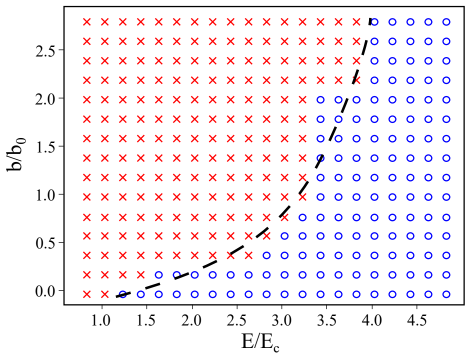

Figure 5: Threshold of Burgers vector strength for pinning effect of the dislocation on the domain wall motion. The red cross mark represent the simulation case that the domain wall does not pass through the edge dislocation and the hollow circle indicates that the domain wall pass through the edge dislocation.

In summary, this study provides a simple while effective way to implement the edge dislocation to the phase field ferroelectric thin film model. The competitive of the external electric field loading and edge dislocation burgers vector is quantitative studied and the criteria for the domain wall pinning is given. Normally for the epitaxial growth ferroelectric thin film, the burgers vector is one unit cell and the distance between two dislocation cores are far enough that one can ignore the interaction between two different dislocations. The proposed criteria may guide the domain wall tuning for the thin films where the dislocations is introduced. The above exponential relation between $E^\star$ and $b^\star$ is only valid for barium titanate. For other material systems, the parameters should be tuned since the energy barrier high of the polarization switching and the piezoelectric coefficients are different. Moreover, since the stress/strain field near the dislocation core changes dramatically and the strain-gradient effect and flexoelectric effect should be considered for a more accurate analysis.

Conflict of Interest

The authors have no conflicts to disclose.

Data availibility

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

Acknowledgments

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References

[1] JF Scott. Applications of modern ferroelectrics. *science*, 315(5814):954–959, 2007.

[2] Peng Gao, Christopher T Nelson, Jacob R Jokisaari, Seung-Hyub Baek, Chung Wung Bark, Yi Zhang, Enge Wang, Darrell G Schlom, Chang-Beom Eom, and Xiaoqing Pan. Revealing the role of defects in ferroelectric switching with atomic resolution. *Nature communications*, 2(1):591, 2011.

[3] Brian J Rodriguez, YH Chu, R Ramesh, and Sergei V Kalinin. Ferroelectric domain wall pinning at a bicrystal grain boundary in bismuth ferrite. *Applied Physics Letters*, 93(14), 2008.

[4] J Schultheiß, S Checchia, H Uršič, T Frömling, JE Daniels, B Malič, T Rojac, and J Koruza. Domain wall-grain boundary interactions in polycrystalline pb (zr0. 7ti0. 3) o3 piezoceramics. *Journal of the European Ceramic Society*, 40(12):3965–3973, 2020.

[5] Tadej Rojac, Marija Kosec, Bojan Budic, Nava Setter, and Dragan Damjanovic. Strong ferroelectric domain-wall pinning in bifeo3 ceramics. *Journal of Applied Physics*, 108(7), 2010.

[6] Emmanuel Defaÿ. Integration of ferroelectric and piezoelectric thin films: concepts and applications for microsystems. 2013.

[7] T Mikolajick, U Schroeder, and S Slesazeck. The past, the present, and the future of ferroelectric memories. *IEEE Transactions on Electron Devices*, 67(4):1434–1443, 2020.

[8] Yang Zou, Chao Gao, Jie Zhou, Yan Liu, Qinwen Xu, Yuanhang Qu, Wen-Juan Liu, Jeffrey Bo Woon Soon, Yao Cai, and Chengliang Sun. Aluminum scandium nitride thin-film bulk acoustic resonators for 5g wideband applications. *Microsystems & Nanoengineering*, 8(1):124, 2022.

[9] Wei Cai and William D Nix. *Imperfections in Crystalline Solids*. Springer, 2018.

[10] Marion Höfling, Maximilian Trapp, Lukas Porz, Hana Uršič, Enrico Bruder, Hans-Joachim Kleebe, Jürgen Rödel, and Jurij Koruza. Large plastic deformability of bulk ferroelectric knbo3 single crystals. *Journal of the European Ceramic Society*, 41(7):4098–4107, 2021.

[11] William D Nix. Mechanical properties of thin films. *Metallurgical transactions A*, 20:2217–2245, 1989.

[12] JW Matthews and AE Blakeslee. Defects in epitaxial multilayers: I. misfit dislocations. *Journal of Crystal growth*, 27:118–125, 1974.

[13] SD Lester, FA Ponce, MG Craford, and DA Steigerwald. High dislocation densities in high efficiency gan-based light-emitting diodes. *Applied Physics Letters*, 66(10):1249–1251, 1995.

[14] ZR Dai, ZL Wang, XF Duan, and Jiming Zhang. Link-up of 90° domain boundaries with interface dislocations in batio3/laalo3. *Applied physics letters*, 68(22):3093–3095, 1996.

[15] Antonios Kontsos and Chad M Landis. Computational modeling of domain wall interactions with dislocations in ferroelectric crystals. *International journal of solids and structures*, 46(6):1491–1498, 2009.

[16] Xiandong Zhou, Zhen Liu, and Bai-Xiang Xu. Influence of dislocations on domain walls in perovskite ferroelectrics: phase-field simulation and driving force calculation. *International Journal of Solids and Structures*, 238:111391, 2022.

[17] Xiandong Zhou, Christoph Reimuth, and Bai-Xiang Xu. Phase-field simulation of misfit dislocations in two-phase electrode particles: Driving force calculation and stability analysis. *International Journal of Solids and Structures*, 249:111688, 2022.

[18] Fangping Zhuo, Xiandong Zhou, Shuang Gao, Felix Dietrich, Pedro B Groszewicz, Lovro Fulanović, Patrick Breckner, Bai-Xiang Xu, Hans-Joachim Kleebe, Dragan Damjanovic, et al. Intrinsic-strain engineering by dislocation imprint in bulk ferroelectrics. *Physical review letters*, 131(1):016801, 2023.

[19] Long Cheng, Heng Zhang, Ran Xu, Kevin Co, Nicolas Guiblin, Mojca Otoničar, Charles Paillard, Yujia Wang, and Brahim Dkhil. Dislocations and a domains coupling in $pbtio_3$ thin films. *Applied Physics Letters*, 123(20), 2023.

[20] Tianshu Jiang, Fan Ni, Oscar Recalde-Benitez, Patrick Breckner, Leopoldo Molina-Luna, Fangping Zhuo, and Jürgen Rödel. Observation of dislocation-controlled domain nucleation and domain-wall pinning in single-crystal batio3. *Applied Physics Letters*, 123(20), 2023.

[21] Ekkehart Kröner et al. Continuum theory of defects. *Physics of defects*, 35:217–315, 1981.

[22] VA Lubarda and X Markenscoff. Configurational force on a lattice dislocation and the peierls stress. *Archive of Applied Mechanics*, 77:147–154, 2007.

[23] Long-Qing Chen. Phase-field method of phase transitions/domain structures in ferroelectric thin films: a review. *Journal of the American Ceramic Society*, 91(6):1835–1844, 2008.

[24] Bai-Xiang Xu, David Schrade, Ralf Müller, Dietmar Gross, Torsten Granzow, and Jürgen Rödel. Phase field simulation and experimental investigation of the electro-mechanical behavior of ferroelectrics. *Journal of Applied Mathematics and Mechanics/Zeitschrift für Angewandte Mathematik und Mechanik (ZAMM)*, 90(7):623, 2010.

[25] Shuai Wang, Min Yi, and Bai-Xiang Xu. A phase-field model of relaxor ferroelectrics based on random field theory. *International Journal of Solids and Structures*, 83:142–153, 2016.

[26] D Schrade, BX Xu, R Mueller, and D Gross. On phase field simulations of ferroelectrics: Parameter identification and verification. In *Proceedings of the ASME*, pages 301–308, 2008.

[27] Yongzhong Huo and Qing Jiang. Modeling of domain switching in polycrystalline ferroelectric ceramics. *Smart materials and structures*, 6(4):441, 1997.

[28] M Kamlah. Ferroelectric and ferroelastic piezoceramics–modeling of electromechanical hysteresis phenomena. *Continuum Mechanics and Thermodynamics*, 13(4):219–268, 2001.

[29] Long-Qing Chen. Appendix a–landau free-energy coefficients.