

Strain-induced emergent polar skyrmion and topological
structures: Multiphysics phase-field simulations

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Contents

Chapter 1	Introduction.....	1
1.1	Ferroelectric materials and polarization orders	1
1.2	Overview of this thesis	6
	References.....	8
Chapter 2	Emergence of higher-order two-dimensional topological polar structures via singular stress field	13
2.1	Introduction	13
2.2	Simulation method	14
2.2.1	Phase-field modeling.....	14
2.2.2	Simulation model	17
2.2.3	Simulation procedure	18
2.3	Results and discussion.....	19
2.3.1	Singular stress field-induced topological polar nano-region.....	19
2.3.2	Electromechanical formation mechanism of topological polar structures	22
2.3.3	Mechanical control of topological polar structures.....	25
2.4	Conclusion.....	27
	References.....	28
Chapter 3	Emergence of isolated polar skyrmion and quasi two-dimensional topological states by nanoindentation.....	31
3.1	Introduction	31
3.2	Simulation method	32
3.2.1	Phase-field modeling.....	32
3.2.2	Simulation model and procedure.....	34
3.3	Results and discussion.....	37
3.3.1	Mechanical writing of polar skyrmion	37
3.3.2	Mechanism of polarization switching	40
3.3.3	Electric erasing of polar skyrmion	43
3.3.4	Mechanical and electrical control of polar topological structures	44
3.3.5	Indentation into in-plane 90-degree polarization domain	45
3.4	Conclusion.....	48

References.....	49
Chapter 4 Emergence of collective polar skyrmion lattice and related topological phases via strain engineering	55
4.1 Introduction	55
4.2 Simulation method	56
4.2.1 Phase-field modeling.....	56
4.2.2 Simulation model and procedure.....	60
4.3 Results and discussion.....	61
4.4 Conclusion.....	68
References.....	69
Chapter 5 Dislocation structures in emergent polar skyrmion lattices	73
5.1 Introduction	73
5.2 Method	75
5.2.1 Phase-field modeling.....	75
5.2.2 Simulation model and procedure.....	77
5.3 Results and Discussion.....	79
5.3.1 Quasiparticle-specific core structure of skyrmion dislocations	79
5.3.2 Inherent strain field and breakdown of Volterra's elasticity theory	82
5.3.3 Quasiparticle-specific lattice mechanics in polar skyrmion lattices	84
5.4 Conclusion.....	88
References.....	89
Chapter 6 Grain boundary structures in emergent polar skyrmion lattices.....	95
6.1 Introduction	95
6.2 Simulation method	96
6.2.1 Phase-field modeling.....	97
6.2.2 Simulation model and procedure.....	99
6.3 Results and discussion.....	101
6.3.1 Skyrmion Grain Boundary Structure under High Electric Field (12 MV/cm)..	101
6.3.2 Skyrmion grain boundary structure under medium electric field (9 MV/cm) ..	103
6.3.3 Skyrmion grain boundary structure under low electric field (6 MV/cm)	103
6.3.4 Temperature dependence of skyrmion grain boundary structure	104
6.3.5 Quasiparticle-based relaxation mechanism in skyrmion grain boundaries	105
6.3.6 Structures and features of zigzag grain boundaries.....	108

6.3.7	Energetic and structural characteristics of skyrmion grain boundaries.....	109
6.4	Conclusion.....	111
	References.....	112
Chapter 7	Conclusion	117
7.1	Summary of thesis.....	117
7.2	Prospects.....	118
	List of publications and conferences	121
	Acknowledgement	127

Chapter 1 Introduction

1.1 Ferroelectric materials and polarization orders

Ferroelectric perovskite-oxide materials, such as PbTiO_3 , BaTiO_3 , and $\text{Pb}(\text{Zr,Ti})\text{O}_3$, possess spontaneous electric polarization arising from their asymmetric crystal structures, which can be reversed by an applied electric field, a property known as ferroelectricity. They also have the ability to interconvert electrical and mechanical energies mutually, generating electricity from external forces and deforming in response to an applied electric field. Owing to these intrinsic functionalities, they have been widely utilized in various applications, including non-volatile memories, sensors, actuators, and other electromechanical devices [1–6]. Moreover, they are expected to play essential roles in next-generation nanoscale electromechanical devices, energy harvesting from environmental sources such as mechanical vibrations, and advanced neuromorphic computing components that may serve as alternatives to conventional semiconductors [7–11]. Therefore, enhancing and developing the functionalities of ferroelectrics are essential for technological innovation.

The origin of functionalities of ferroelectrics lies in the microscopic collective ordering of

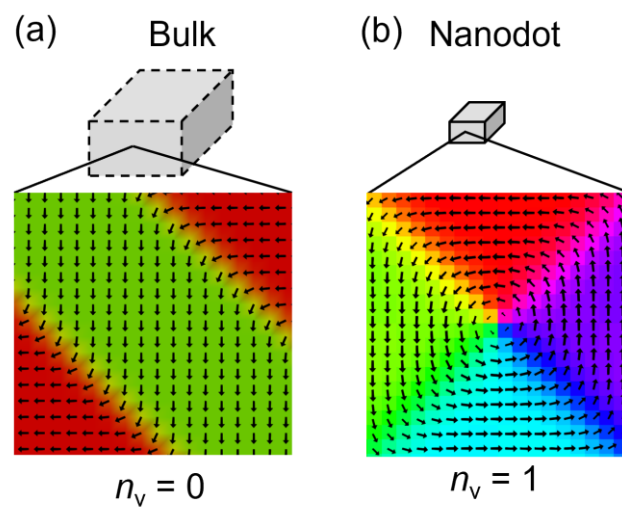


Figure 1-1 Schematic of polarization orders. (a) 90-degree domains in bulk ferroelectrics. (b) Vortex in nanoscale ferroelectrics.

spontaneous polarizations. Understanding and controlling this polarization order are therefore crucial for improving material performance and realizing advanced applications. In conventional bulk ferroelectrics, a linear 180-degree or 90-degree domain structure (Figure 1-1(a)) typically forms to optimize both elastic and electrical energies. These macroscopic domain structures enable macroscopic (net) polarization switching by an applied electric field while maintaining a remanent polarization after its removal, which is called ferroelectricity. In recent years, due to advances in material fabrication techniques and the demand for device miniaturization, nanoscale ferroelectrics have been developed. Generally, nanoscale materials exhibit physical properties different from those of bulk materials due to size effects, where the proportion of the surface area becomes significantly larger. In nanoscale ferroelectrics such as nanodots and ultrathin films, pronounced surface effects tend to suppress polarization components perpendicular to the surface due to a depolarizing field arising from surface polarization charge. As a result of reconfiguration of the polarization structure to reduce the electrostatic energy due to the depolarizing field, the nanoscale ferroelectric system adopts to emerge a nanoscale vortex polarization order [12] (Figure 1-1(b)). In such a polar vortex, the toroidal moments, defined by the vortex geometry, can be switched by a circulating current, giving rise to an emergent functionality termed ferrotoroidicity [12–14]. This phenomenon demonstrates that novel polarization orders can provide novel functionalities. Therefore, the exploration and creation of such unconventional polarization orders, as well as the elucidation of associated electromechanical functions, are of scientific and engineering significance [15–17].

Such microscopic polarization orders can be classified by the concept of topology [18–20]. Topology provides a mathematical framework for characterizing whether two vector fields can be transformed into one another continuously without essential features. In other words, configurations that can be continuously deformed into one another are regarded as topologically equivalent. Quantities that remain invariant under such continuous transforms are called topological charges, which are used to classify topological orders. In the case of two-dimensional vector fields, a representative topological charge is the winding number, defined as [21]

$$n_v = \frac{1}{2\pi} \oint_C \nabla \theta dr, \quad (1-1)$$

where θ is the counterclockwise rotation angle of vectors, C is an integral path around the interest area. For instance, the 90-degree domain structure corresponds to $n_v = 0$, whereas the vortex structure corresponds to $n_v = 1$. Nanoscale vector field configurations with inherent

topological characteristics have widely attracted considerable attention in the field of condensed matter physics and are called topological structures or topological orders. In particular, topological magnetic spin orders in ferromagnets have been extensively studied, with typical examples including skyrmions [22–24], merons [25,26], and hedgehogs [27,28] (Figure 1-2). These complex spin structures possess intrinsic physical properties owing to their geometry and have attracted considerable interest in their potential applications. Among them, skyrmions, classified as one of quasi-two-dimensional topological orders in which three-dimensional vectors are continuously distributed in a two-dimensional space (plane), have been particularly well studied. A topological charge classifying quasi-two-dimensional topological orders is the skyrmion number, defined as [20]

$$n_{\text{sk}} = \frac{1}{4\pi} \iint \mathbf{n} \cdot \left(\frac{\partial \mathbf{n}}{\partial x} \times \frac{\partial \mathbf{n}}{\partial y} \right) \text{d}r^2, \quad (1-2)$$

where $\mathbf{n}$ is a normalized vector. Owing to their unique features, such as nanoscale size, robustness against external perturbations due to topological protection, and emergent electromechanical field, magnetic skyrmions have been proposed for diverse device applications, including magnetic memories [29], racetrack memories [30], logic gates [31], and synaptic devices [32]. Furthermore, one of the most intriguing aspects of skyrmions is their particle-like nature and collective behaviors. Individual skyrmions behave as discrete quasiparticles that can be driven by external fields such as electric current and magnetic fields, maintaining their individuality even during deformation, and can be created or annihilated under strong external field excitations [33–35]. In addition, skyrmions tend to self-organize into periodic arrays, forming skyrmion lattices. These skyrmion lattices host lattice defects such as dislocations [36,37] and grain boundaries [38,39], and exhibit defect-mediated lattice dynamics

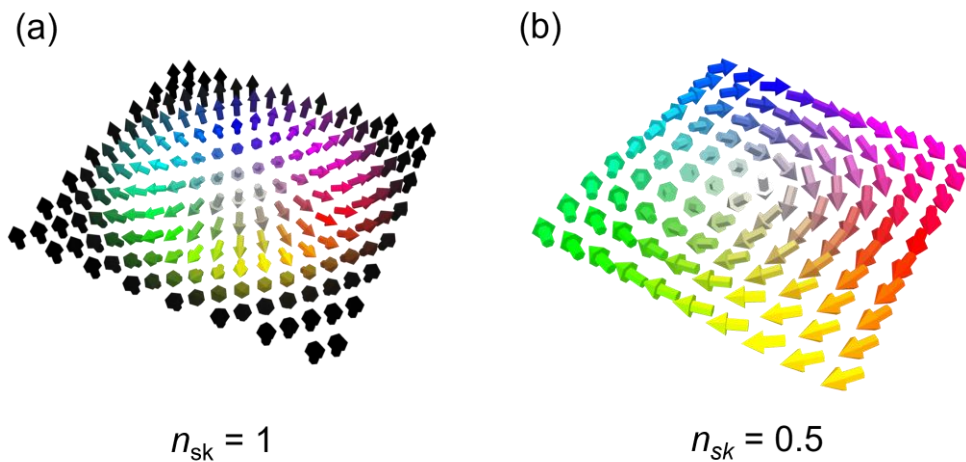


Figure 1-2 The schematic of (a) skyrmion and (b) meron.

analogous to those observed in atomic crystals [40,41]. Such emergent behaviors demonstrate that assemblies of skyrmions constitute a new form of material, a lattice of topological quasiparticles in vector fields, fundamentally distinct from conventional spin textures.

By analogy with such topological magnetic orders, ferroelectric polarization orders are also expected to host various topological structures with unique physical functionalities. Recent studies have theoretically proposed and experimentally demonstrated the formation of topological polar structures, including super helices [42], polar merons [43], and polar skyrmions [44] by exploiting complex structures and boundary conditions of nanoscale ferroelectrics such as ultrathin films and nanoscale superlattice systems. Although direct observations remain limited, several intrinsic physical properties absent in their magnetic counterparts have been reported. In particular, polar skyrmions exhibit local negative permittivity [45,46], leading to an enhanced dielectric response of the material. Other intriguing behaviors, such as teleportation [47] and high optical responses [48,49], have also been reported. Furthermore, polar skyrmions can exist in ferroelectrics even below the critical size at which spontaneous polarization usually vanishes, with characteristic diameters of only a few nanometers [50,51], significantly smaller than those of magnetic skyrmions. Such an extremely small dimension of polar skyrmions enables ultra-high density information storage and readout, making the polar skyrmions highly promising for next-generation ferroelectric devices that surpass the performance of conventional ferroelectrics.

Despite possessing such intriguing potential, research on topological polar structures still lags behind that on magnetic counterparts. This scarcity primarily arises from the absence of driving forces for their spontaneous formation. In ferromagnets, the major driving force for the emergence of topological structures is the Dzyaloshinskii-Moriya interaction (DMI) [52,53], which induces chiral spins twisting. Although DMI-like interactions have been theoretically predicted in ferroelectrics, their magnitudes are insufficient to drive polarization twisting [54]. Indeed, intricate factors such as electrical boundary conditions, surface effects, and strain conditions. Consequently, the few observed topological polar structures are mostly determined by extrinsic factors such as sample geometry or boundary conditions, making their types and locations essentially uncontrollable. Given this background, although topological polar structures hold great potential, the central challenge remains how to create and control them. While magnetic topologies have already been established as controllable quasiparticles with rich device concepts, realizing the approach to generation and manipulation of polar topological structures, particularly polar skyrmions, would open an entirely new route toward next-

generation nanodevice using electromechanical responses specific to ferroelectric polarizations. Moreover, achieving polar skyrmions in a manner that allows direct comparison with magnetic skyrmions holds great scientific implications for elucidating the contrast between magnetism and electricity.

To establish a novel strategy to achieve the emergence and control polar topological structures including the skyrmions, here, I focus on unique electromechanical coupling effects in ferroelectrics. Ferroelectrics exhibit the electrical responses to mechanical strain originating from the strong connection between microscopic polarization due to asymmetric crystal structure and mechanical deformations. This phenomenon, known as the piezoelectric effect, describes a linear increase or decrease in polarization with applied strain. In addition, recent research reported the influence of the flexoelectric effect, particularly in nanoscale ferroelectrics. The flexoelectric effect refers to the inducing polarization by strain gradient [55]. Although its influence is negligible in bulk ferroelectrics, it becomes significant and even dominant in the nanoscale counterpart, where extremely large strain gradients may arise due to size effect. Based on these electromechanical effects, previous studies reported mechanical creation and writing of polarization domains in ferroelectrics [56]. Therefore, strain and/or strain gradient fields can be a promising driving force to induce polarization and even design the polarization distribution in ferroelectrics. Moreover, by combining strain fields and structural factors, it is expected that complex topological polar structures can be arbitrarily created and controlled [57]. Nevertheless, the fundamental theoretical understanding of how mechanical fields can generate and control such topological polar structures and what nature they have remains largely unexplored.

Building on this concept, exploring the strain field-induced topological polarization requires a theoretical framework that can simultaneously describe polarization, strain, electric field, and interactions between them. For this purpose, the multiphysics phase-field method [58,59] provides a powerful approach. In the phase-field simulation, the polarization vector $\mathbf{P} = (P_1, P_2, P_3)$ is treated as an order parameter, allowing various electromechanical phenomena in ferroelectrics to be explicitly incorporated. The effects of strain and electric field can be included as coupling energy terms. Using the phase-field approach, stabilization and dynamics of conventional domain structures have been successfully investigated [60]. Furthermore, specific boundary conditions such as surface effects can be modeled as additional interfacial energy terms [43,61]. This framework thus enables the elucidation of underlying mechanisms of mechanically induced polarization structures and thus provides a predictive platform for designing emergent topological structures.

1.2 Overview of this thesis

This thesis aims both to propose a novel strategy for the mechanical creation and control of emergent topological polar structures by establishing an understanding of the interaction between strain fields and polar topological orders in nanoscale ferroelectric systems using multiphysics phase-field simulations and to elucidate the fundamental structural properties of topological polar structures. Although conventional ferroelectrics have been utilized widely, they are primarily governed by linear domain switching, which limits the tunability and application range. The exotic topological polar structures overcoming these limitations can be realized by strain field engineering. Through electromechanical coupling effects, various topological polar structures are successfully induced by strain fields, especially reflecting the geometrical characteristics of the strain field. This approach extends the possibilities for polarization structures beyond conventional continuous domains toward discrete quasiparticles, such as polar skyrmions. Furthermore, intrinsic collective phenomena and emergent properties arising from the aggregation of quasiparticle polar skyrmions are revealed from the perspective of lattice mechanics. The thesis is organized as follows.

Chapter 2 investigates the creation and control of two-dimensional higher-order topological polar structures using singular stress fields at the tip of a crack under mixed-mode loading. This validates strain-field engineering as an effective method for creating and controlling topological polar structures through electromechanical coupling.

Chapter 3 explores the possibility of creation and manipulation of emergent quasi two-dimensional topological polar structures, such as polar skyrmions and merons, induced by nanoindentation on ferroelectric thin films. I show that the combination of strain field and structural factors compensates for the absence of driving forces for spontaneous quasiparticle-like structure formations and extends the range of accessible topological polar structures from the conventional domains to novel quasiparticle-like structures.

Chapter 4 examines strain-induced topological transitions from a trivial stripe phase to skyrmion bubble and skyrmion lattice phases in a ferroelectric superlattice system. I demonstrate that strain fields are also effective for controlling the collective formation of emergent topological polar quasiparticles, providing a new platform of polar skyrmion lattices.

Chapter 5 focuses on one of the representative lattice defects, a dislocation, in polar skyrmion lattices, and investigates their structural characteristics and impacts on the lattice properties based on Volterra's elasticity theory. I reveal the emergence of quasiparticle-specific lattice

mechanics arising from skyrmion deformations. These results demonstrate novel phenomena inherent to collective discrete quasiparticles, which are completely different from atomic crystals and conventional continuous domain polar structures.

Chapter 6 investigates another important lattice defect, a grain boundary, in polar skyrmion lattices. I also reveal the quasiparticle-specific structures and stabilization mechanism due to skyrmion deformations, similarly observed in the dislocation in polar skyrmion lattices. The findings provide an insight into the relaxation processes of lattice mismatch and defect-mediated unique dynamics of polar skyrmion lattices.

Finally, Chapter 7 presents the conclusions and prospects of this thesis.

References

- [1] J. F. Scott, Applications of Modern Ferroelectrics, *Science* **315**, 954 (2007).
- [2] N. Li, K. Xie, and H. Huang, Ferroelectric Materials for High Energy Density Batteries: Progress and Outlook, *ACS Energy Lett.* **8**, 4357 (2023).
- [3] M. Yan et al., Ferroelectric Synaptic Transistor Network for Associative Memory, *Advanced Electronic Materials* **7**, 2001276 (2021).
- [4] L. Liu et al., Control the Neural Stem Cell Fate with Biohybrid Piezoelectrical Magnetite Micromotors, *Nano Lett.* **21**, 3518 (2021).
- [5] K. Wang, V. Bheemarasetty, J. Duan, S. Zhou, and G. Xiao, Fundamental physics and applications of skyrmions: A review, *Journal of Magnetism and Magnetic Materials* **563**, 169905 (2022).
- [6] H. W. Park, J.-G. Lee, and C. S. Hwang, Review of ferroelectric field-effect transistors for three-dimensional storage applications, *Nano Select* **2**, 1187 (2021).
- [7] Z. Chen et al., All-ferroelectric implementation of reservoir computing, *Nat Commun* **14**, 3585 (2023).
- [8] X. Wei, N. Domingo, Y. Sun, N. Balke, R. E. Dunin-Borkowski, and J. Mayer, Progress on Emerging Ferroelectric Materials for Energy Harvesting, Storage and Conversion, *Advanced Energy Materials* **12**, 2201199 (2022).
- [9] Y. Zou, L. Bo, and Z. Li, Recent progress in human body energy harvesting for smart bioelectronic system, *Fundamental Research* **1**, 364 (2021).
- [10] K. Xu, L. Yang, J. Wang, and H. Huang, Design of high energy storage ferroelectric materials by phase-field simulations, *J. Mater. Inf.* **5**, (2025).
- [11] C. Liang et al., Ferroelectric Charged Domain-Wall Synapse for Neuromorphic Computing, (n.d.).
- [12] I. I. Naumov, L. Bellaiche, and H. Fu, Unusual phase transitions in ferroelectric nanodisks and nanorods, *Nature* **432**, 7018 (2004).
- [13] G. Pilania, S. P. Alpay, and R. Ramprasad, *Ab initio* study of ferroelectricity in BaTiO₃ nanowires, *Phys. Rev. B* **80**, 014113 (2009).
- [14] T. Shimada, X. Wang, S. Tomoda, P. Marton, C. Elsässer, and T. Kitamura, Coexistence of rectilinear and vortex polarizations at twist boundaries in ferroelectric PbTiO₃ from first principles, *Phys. Rev. B* **83**, 094121 (2011).

- [15] L. Van Lich, T. Shimada, S. Sepideh, J. Wang, and T. Kitamura, Multilevel hysteresis loop engineered with ferroelectric nano-metamaterials, *Acta Materialia* **125**, 202 (2017).
- [16] X.-K. Wei, K. Xu, K. Vaideeswaran, J. Mayer, and H. Huang, Observation of Atomic-Scale Polar Vortex–Antivortex Pairs in Antiferroelectric PbZrO_3 Thin Films, *Nano Lett.* [acs.nanolett.5c01506](https://doi.org/10.1021/acs.nanolett.5c01506) (2025).
- [17] Z. Jiang and J. Wang, Stability of chiral polarization vortex in strained ferroelectric superlattices, *Journal of Applied Physics* **131**, 164101 (2022).
- [18] A. F. Ramada, A topological theory of the electromagnetic field, 10 (n.d.).
- [19] J. V. Selinger, *Introduction to Topological Defects and Solitons: In Liquid Crystals, Magnets, and Related Materials*, Vol. 1032 (Springer Nature Switzerland, Cham, 2024).
- [20] N. Nagaosa and Y. Tokura, Topological properties and dynamics of magnetic skyrmions, *Nature Nanotech* **8**, 899 (2013).
- [21] N. D. Mermin, The topological theory of defects in ordered media, *Rev. Mod. Phys.* **51**, 591 (1979).
- [22] S. Mühlbauer, B. Binz, F. Jonietz, C. Pfleiderer, A. Rosch, A. Neubauer, R. Georgii, and P. Böni, Skyrmion Lattice in a Chiral Magnet, *Science* **323**, 915 (2009).
- [23] Y. Tokura and N. Kanazawa, Magnetic Skyrmion Materials, *Chem. Rev.* **121**, 2857 (2021).
- [24] B. Göbel, I. Mertig, and O. A. Tretiakov, Beyond skyrmions: Review and perspectives of alternative magnetic quasiparticles, *Physics Reports* **895**, 1 (2021).
- [25] H. Jani et al., Antiferromagnetic half-skyrmions and bimerons at room temperature, *Nature* **590**, 74 (2021).
- [26] X. Z. Yu, W. Koshibae, Y. Tokunaga, K. Shibata, Y. Taguchi, N. Nagaosa, and Y. Tokura, Transformation between meron and skyrmion topological spin textures in a chiral magnet, *Nature* **564**, 7734 (2018).
- [27] S. Watanabe, Magnetic dynamics of hedgehog in icosahedral quasicrystal, *Sci Rep* **12**, 15514 (2022).
- [28] Y. Fujishiro et al., Topological transitions among skyrmion- and hedgehog-lattice states in cubic chiral magnets, *Nat Commun* **10**, 1059 (2019).
- [29] N. Nagaosa and Y. Tokura, Topological properties and dynamics of magnetic skyrmions, *Nature Nanotech* **8**, 899 (2013).
- [30] A. Fert, V. Cros, and J. Sampaio, Skyrmions on the track, *Nature Nanotech* **8**, 152 (2013).
- [31] X. Zhang, M. Ezawa, and Y. Zhou, Magnetic skyrmion logic gates: conversion, duplication and merging of skyrmions, *Sci Rep* **5**, 9400 (2015).

- [32] Y. Huang, W. Kang, X. Zhang, Y. Zhou, and W. Zhao, Magnetic skyrmion-based synaptic devices, *Nanotechnology* **28**, 08LT02 (2017).
- [33] C. Reichhardt, C. J. O. Reichhardt, and M. V. Milošević, Statics and dynamics of skyrmions interacting with disorder and nanostructures, *Rev. Mod. Phys.* **94**, 035005 (2022).
- [34] Z. Chen, X. Zhang, Y. Zhou, and Q. Shao, Skyrmion Dynamics in the Presence of Deformation, *Phys. Rev. Applied* **17**, L011002 (2022).
- [35] J. Zang, M. Mostovoy, J. H. Han, and N. Nagaosa, Dynamics of Skyrmion Crystals in Metallic Thin Films, *Phys. Rev. Lett.* **107**, 136804 (2011).
- [36] R. Gruber, J. Rothörl, S. M. Fröhlich, M. A. Brems, M.-A. Syskaki, E. M. Jefremovas, S. Krishnia, A. Sudbø, P. Virnau, and M. Kläui, Imaging Topological Defect Dynamics Mediating 2D Skyrmion Lattice Melting, (n.d.).
- [37] P. Huang, T. Schönenberger, M. Cantoni, L. Heinen, A. Magrez, A. Rosch, F. Carbone, and H. M. Rønnow, Melting of a skyrmion lattice to a skyrmion liquid via a hexatic phase, *Nat. Nanotechnol.* **15**, 761 (2020).
- [38] T. Matsumoto, Y.-G. So, Y. Kohno, H. Sawada, Y. Ikuhara, and N. Shibata, Direct observation of $\Sigma 7$ domain boundary core structure in magnetic skyrmion lattice, *Sci. Adv.* **2**, e1501280 (2016).
- [39] T. Schönenberger, P. Huang, L. D. Brun, L. Guanhao, A. Magrez, F. Carbone, and H. M. Rønnow, Direct Visualisation of Skyrmion Lattice Defect Alignment at Grain Boundaries, *Nanoscale Res Lett* **17**, 20 (2022).
- [40] R. Brearton, L. A. Turnbull, J. A. T. Verezhak, G. Balakrishnan, P. D. Hatton, G. Van Der Laan, and T. Hesjedal, Deriving the skyrmion Hall angle from skyrmion lattice dynamics, *Nat Commun* **12**, 2723 (2021).
- [41] S. Pöllath et al., Dynamical Defects in Rotating Magnetic Skyrmion Lattices, *Phys. Rev. Lett.* **118**, 207205 (2017).
- [42] T. Shimada, L. V. Lich, K. Nagano, J.-S. Wang, J. Wang, and T. Kitamura, Polar Superhelices in Ferroelectric Chiral Nanosprings, *Sci Rep* **6**, 1 (2016).
- [43] Y. J. Wang et al., Polar meron lattice in strained oxide ferroelectrics, *Nat. Mater.* **19**, 881 (2020).
- [44] S. Das et al., Observation of room-temperature polar skyrmions, *Nature* **568**, 368 (2019).
- [45] C. Guo, S. Tang, D. Liang, J. Wang, and H. Huang, Continuously Tuning Negative Capacitance via Field-Driven Polar Skyrmions in Ferroelectric Trilayer Wrinkled Films, *ACS Nano* **acsnano.4c16350** (2025).

- [46] S. Das et al., Local negative permittivity and topological phase transition in polar skyrmions, *Nat. Mater.* **20**, 194 (2021).
- [47] S. Prokhorenko, Y. Nahas, V. Govinden, Q. Zhang, N. Valanoor, and L. Bellaiche, Motion and teleportation of polar bubbles in low-dimensional ferroelectrics, *Nat Commun* **15**, 412 (2024).
- [48] L. Gao, S. Prokhorenko, Y. Nahas, and L. Bellaiche, Dynamical Control of Topology in Polar Skyrmions via Twisted Light, *Phys. Rev. Lett.* **132**, 026902 (2024).
- [49] Q. Li and J. Sun, Twisted light with a designed polar topology, *Nat. Nanotechnol.* (2025).
- [50] F.-H. Gong, Y.-L. Tang, Y.-J. Wang, Y.-T. Chen, B. Wu, L.-X. Yang, Y.-L. Zhu, and X.-L. Ma, Absence of critical thickness for polar skyrmions with breaking the Kittel's law, *Nat Commun* **14**, 3376 (2023).
- [51] L. Han et al., High-density switchable skyrmion-like polar nanodomains integrated on silicon, *Nature* **603**, 7899 (2022).
- [52] I. Dzyaloshinsky, A thermodynamic theory of “weak” ferromagnetism of antiferromagnetics, *Journal of Physics and Chemistry of Solids* **4**, 241 (1958).
- [53] T. Moriya, Anisotropic Superexchange Interaction and Weak Ferromagnetism, *Phys. Rev.* **120**, 91 (1960).
- [54] Y. Nahas, S. Prokhorenko, L. Louis, Z. Gui, I. Kornev, and L. Bellaiche, Discovery of stable skyrmionic state in ferroelectric nanocomposites, *Nat Commun* **6**, 8542 (2015).
- [55] W. Zhou, P. Chen, and B. Chu, Flexoelectricity in ferroelectric materials, *IET Nanodielectrics* **2**, 83 (2019).
- [56] H. Lu, C.-W. Bark, D. Esque de los Ojos, J. Alcala, C. B. Eom, G. Catalan, and A. Gruverman, Mechanical Writing of Ferroelectric Polarization, *Science* **336**, 59 (2012).
- [57] T. Shimada, Y. Wang, T. Hamaguchi, K. Kasai, K. Masuda, L. Van Lich, T. Xu, J. Wang, and H. Hirakata, Emergence of non-trivial polar topologies hidden in singular stress field in SrTiO_3 : topological strain-field engineering, *J. Phys.: Condens. Matter* **33**, 505301 (2021).
- [58] L.-Q. Chen, Phase-Field Method of Phase Transitions/Domain Structures in Ferroelectric Thin Films: A Review, *J American Ceramic Society* **91**, 1835 (2008).
- [59] Y. L. Li, S. Y. Hu, Z. K. Liu, and L. Q. Chen, Phase-field model of domain structures in ferroelectric thin films, *Appl. Phys. Lett.* **78**, 3878 (2001).
- [60] A. K. Soh, Y. C. Song, and Y. Ni, Phase Field Simulations of Hysteresis and Butterfly Loops in Ferroelectrics Subjected to Electro-Mechanical Coupled Loading, *J American Ceramic Society* **89**, 652 (2006).

- [61] S. Yuan, Z. Chen, S. Prokhorenko, Y. Nahas, L. Bellaiche, C. Liu, B. Xu, and L. Chen, Hexagonal Close-packed Skyrmion Lattice in Ultrathin Ferroelectric PbTiO_3 Films, *Physical Review Letters* **130**, 226801 (2023).

Chapter 2 Emergence of higher-order two-dimensional topological polar structures via singular stress field

2.1 Introduction

Topological structures have great potential for applications in nanodevice due to their properties such as behaving as nanoscale quasi-particles, stability from topological protection, and unique interaction with external fields. In the realm of ferromagnetics, extensive researches have been conducted on topological spin structures, such as vortex [1,2], meron [3,4], and skyrmion [5–8] primarily driven by the Dzyaloshinskii-Moriya interaction (DMI) [9], in which spins twist each other. On the other hand, in ferroelectrics, relatively simple structures like vortex [10,11] have been commonly observed due to the absence of effects like DMI [12]. Even though, boundary conditions of nanomaterials such as surface effect and multi-physics properties of ferroelectrics facilitate the formation of various topological polar structures. Recent observations have highlighted the presence of topological polar structures, such as skyrmion [13,14] in $\text{PbTiO}_3/\text{SrTiO}_3$ superlattice, meron [15,16] in PbTiO_3 thin film and $\text{BiFeO}_3/\text{SrTiO}_3$ superlattice. However, the formation of most of these topological polar structures are formed dependent on the device shape, indicating that the arbitrary creation of topological structures in ferroelectric materials remains elusive. Thus, the development of methods to manipulate the formation and topological properties of polarization structures is imperative for engineering applications.

Here, I focus on the singular stress field near the crack tip and flexoelectric effect [17,18]. The singular stress field exhibits stress concentration around the crack tip, accompanied by a distribution of strain in the angular direction around the crack tip. Moreover, intense concentration generates strain gradients, leading to the dominance of the flexoelectric effect, resulting in potential for topological polar structure creation. Based on this idea, our research group has demonstrated that mode I load (in-plane tensile load) into a crack in SrTiO_3 can create a non-trivial two-dimensional topological polar structure [19]. Furthermore, I have established that the topological features hidden in singular strain gradient field are mirrored in the

polarization field. This result provided the concept of mechanical control of functional topological polar structures by strain field, named ‘topological strain-field engineering’. However, it is not sufficient to demonstrate that this approach is generally effective, and results in geometrically different and more complex mechanical fields are necessary.

In this chapter, I investigate the polarization distribution induced by the mode II singular stress field near the crack tip in SrTiO₃ to demonstrate the versatility of the approach in creating topological polarization structure using geometric mechanical fields via flexoelectric effect. Additionally, I demonstrate a mechanically induced topological transition of polar structure through mixed mode I and mode II loading. This result indicates the general effectiveness of ‘topological strain-field engineering’

2.2 Simulation method

2.2.1 Phase-field modeling

In phase-field simulation for ferroelectrics, taking the polarization $\mathbf{P} = (P_1, P_2, P_3)$ as the order parameters, the total free energy F can be expressed as

$$F = \int_V f dV = \int_V (f_{\text{Landau}} + f_{\text{grad}} + f_{\text{electo}} + f_{\text{elastic}} + f_{\text{ecoup}} + f_{\text{flexo}}) dV \quad (2.1)$$

where $f, f_{\text{Landau}}, f_{\text{grad}}, f_{\text{electo}}, f_{\text{elastic}}, f_{\text{ecoup}}, f_{\text{flexo}}$ are the total free energy density, the Landau energy density, the gradient energy density, electrostatic energy density, elastic energy density, electrostrictive energy density, and the flexoelectric energy density, respectively.

The Landau energy density, which describes the temperature-dependent ferroelectric phase transition based on Landau phenomenological theory [18], is expressed as

$$\begin{aligned} f_{\text{Landau}} &= \alpha_{ij} P_i P_j + \alpha_{ijkl} P_i P_j P_k P_l \\ &= \alpha_1 (P_1^2 + P_2^2 + P_3^2) + \alpha_{11} (P_1^4 + P_2^4 + P_3^4) + \alpha_{12} (P_1^2 P_2^2 + P_1^2 P_3^2 + P_2^2 P_3^2) \end{aligned} \quad (2.2)$$

where α_{ij} and α_{ijkl} are the Landau expansion coefficients and in particular α_{11} is a function of temperature T . The gradient energy density is related with formation of domains and domain walls, and is expressed as

$$\begin{aligned} f_{\text{grad}} &= \frac{1}{2} G_{ijkl} P_{i,j} P_{k,l} \\ &= \frac{1}{2} G_{11} (P_{1,1}^2 + P_{2,2}^2 + P_{3,3}^2) + G_{12} (P_{1,1} P_{2,2} + P_{2,2} P_{3,3} + P_{3,3} P_{1,1}) \\ &\quad + \frac{1}{2} G_{44} [(P_{1,2} + P_{2,1})^2 + (P_{2,3} + P_{3,2})^2 + (P_{1,3} + P_{3,1})^2] \end{aligned} \quad (2.3)$$

where G_{ijkl} is the gradient energy coefficient in Voigt notation and $P_{i,j} = \frac{\partial P_i}{\partial x_j}$ is polarization gradient. The electrostatic energy density is expressed as

$$\begin{aligned} f_{\text{electo}} &= -\frac{1}{2}\kappa_0 E_i E_i - E_i P_i \\ &= -\frac{1}{2}\kappa_0 (E_1^2 + E_2^2 + E_3^2) - E_1 P_1 - E_2 P_2 - E_3 P_3 \end{aligned} \quad (2.4)$$

where κ_0 is the relative permittivity and E_i is electric field. The elastic energy density is expressed as

$$\begin{aligned} f_{\text{elastic}} &= \frac{1}{2} c_{ijkl} \varepsilon_{ij} \varepsilon_{kl} \\ &= \frac{1}{2} c_{11} (\varepsilon_{11}^2 + \varepsilon_{22}^2 + \varepsilon_{33}^2) + c_{12} (\varepsilon_{11} \varepsilon_{22} + \varepsilon_{22} \varepsilon_{33} + \varepsilon_{33} \varepsilon_{11}) \\ &\quad + 2c_{44} (\varepsilon_{12}^2 + \varepsilon_{23}^2 + \varepsilon_{13}^2) \end{aligned} \quad (2.5)$$

where c_{ijkl} is the elastic constant and ε_{ij} is elastic strain. The electrostrictive energy density, which describes the electrostrictive effect [20], is expressed as

$$\begin{aligned} f_{\text{ecoup}} &= -q_{ijkl} \varepsilon_{ij} P_k P_l \\ &= -(q_{11} \varepsilon_{11} + q_{12} \varepsilon_{22} + q_{12} \varepsilon_{33}) P_1^2 - (q_{12} \varepsilon_{11} + q_{11} \varepsilon_{22} + q_{12} \varepsilon_{33}) P_2^2 \\ &\quad - (q_{12} \varepsilon_{11} + q_{12} \varepsilon_{22} + q_{11} \varepsilon_{33}) P_3^2 - 2q_{44} (\varepsilon_{12} P_1 P_2 + \varepsilon_{23} P_2 P_3 + \varepsilon_{13} P_1 P_3) \end{aligned} \quad (2.6)$$

where q_{ijkl} is the electrostrictive coefficient. The flexoelectric energy density describes the flexoelectric effect [20,21] which includes strain induced by polarization gradient and polarization induced by strain gradient, and is expressed as

$$\begin{aligned} f_{\text{flexo}} &= \frac{1}{2} f_{ijkl} (\varepsilon_{ij} P_{k,l} - P_k \varepsilon_{ij,l}) \\ &= \frac{1}{2} \left\{ \begin{aligned} &f_{11} (P_{1,1} \varepsilon_{11} + P_{2,2} \varepsilon_{22} + P_{3,3} \varepsilon_{33}) \\ &+ f_{12} (P_{2,2} \varepsilon_{11} + P_{3,3} \varepsilon_{11} + P_{1,1} \varepsilon_{22} + P_{3,3} \varepsilon_{22} + P_{1,1} \varepsilon_{33} + P_{2,2} \varepsilon_{33}) \\ &+ f_{44} [\varepsilon_{23} (P_{2,3} + P_{3,2}) + \varepsilon_{12} (P_{1,3} + P_{3,1}) + \varepsilon_{12} (P_{1,2} + P_{2,1})] \end{aligned} \right\} \\ &\quad - \frac{1}{2} \left\{ \begin{aligned} &f_{11} (\varepsilon_{11,1} P_1 + \varepsilon_{22,2} P_2 + \varepsilon_{33,3} P_3) \\ &+ f_{12} [P_1 (\varepsilon_{22,1} + \varepsilon_{33,1}) + P_2 (\varepsilon_{11,2} + \varepsilon_{33,2}) + P_3 (\varepsilon_{11,3} + \varepsilon_{22,3})] \\ &+ f_{44} [P_1 (\varepsilon_{12,2} + \varepsilon_{13,3}) + P_2 (\varepsilon_{12,1} + \varepsilon_{23,3}) + P_3 (\varepsilon_{13,1} + \varepsilon_{23,2})] \end{aligned} \right\} \end{aligned} \quad (2.7)$$

where $P_{i,j}$, $\varepsilon_{ij,k}$, and f_{ijkl} are polarization gradient, strain gradient, and the flexoelectric coupling coefficient. All material parameters [22–24] are shown in Table 2-1.

The temporal and spatial evolution of polarization distribution is calculated by the time-dependent Ginzburg-Landau (TDGL) equation written as

$$\frac{\partial P_i(x, t)}{\partial t} = -L \frac{\delta F}{\delta P_i} \quad (2.8)$$

where t represents time. L is the kinetic coefficient. The right side of equation (2.8) means the thermodynamic driving force for polarization evolution. The stress mechanical equilibrium equation written as

$$\sigma_{ij,j} = \left(\frac{\partial f}{\partial \varepsilon_{ij}} \right)_{,j} = 0 \quad (2.9)$$

and Maxwell-Gauss equation written as

$$D_{i,j} = \left(\frac{\partial f}{\partial E_i} \right)_{,j} = 0 \quad (2.10)$$

must be satisfied simultaneously.

To solve these governing equations with finite element method, using virtual work principle, they are combined into the weak form written as

$$\begin{aligned} & \int_V \left(\frac{\partial P_i}{\partial t} \delta P_i + L \frac{\partial F}{\partial P_i} \delta P_i + L \frac{\partial F}{\partial P_{i,j}} \delta P_{i,j} + \frac{\partial F}{\partial E_i} \delta E_i + \frac{\partial F}{\partial \varepsilon_{ij}} \delta \varepsilon_{ij} \right) dV \\ &= \int_S \left(\frac{\partial F}{\partial P_{i,j}} n_j \delta P_i - \omega \delta \varphi + t_{ij} \delta u_i \right) dS \end{aligned} \quad (2.11)$$

where n_j , ω , and t_{ij} are the normal vector of surfaces, surface charge, and the surface traction, respectively.

Table 2-1 Material constants for SrTiO₃ [22–24].

Landau coefficient			
α_1 [C ⁻² ·m ² ·N]	$4.05 \times 10^7 [\coth(54/T) - \coth(54/30)]$		
α_{11} [C ⁻⁴ ·m ⁶ ·N]	1.7×10^9		
α_{12} [C ⁻⁴ ·m ⁶ ·N]	3.926×10^9		
Gradient constants		Electrostrictive coefficient	
g_{11} [C ⁻² ·m ⁴]	1.0×10^{-11}	q_{11} [m·F ⁻¹]	-1.09×10^9
g_{12} [C ⁻² ·m ⁴]	0.5×10^{-11}	q_{12} [m·F ⁻¹]	12.46×10^9
g_{44} [C ⁻² ·m ⁴]	1.0×10^{-11}	q_{44} [m·F ⁻¹]	2.42×10^9
Elastic constants		Flexoelectric coupling coefficient	
c_{11} [m ⁻² ·N]	3.36×10^{11}	f_{11} [V]	-3.21
c_{12} [m ⁻² ·N]	1.07×10^{11}	f_{12} [V]	1.47
c_{44} [m ⁻² ·N]	1.27×10^{11}	f_{44} [V]	1.07
Relative dielectric constant			
ϵ_b	43		

2.2.2 Simulation model

Figure 2-1 (a) shows the schematic of the simulation model for phase-field simulation. The x_1 , x_2 , and x_3 axes correspond to the [100], [010], and [001] directions of the SrTiO₃ crystallographic axes, respectively. Three-dimensional periodic boundary conditions are applied for polarization, electric potential, and displacement. A crack with a length of $2a = 60$ nm is introduced in the center of the model. The size of model is set to $200 \text{ nm} \times 150 \text{ nm} \times 1 \text{ nm}$, which is large enough to avoid interference between singular stress fields under the periodic boundary condition. To eliminate factors other than the singular stress field that affect the polarization distribution such as depolarization field on the crack surface, the crack is treated as filled with permeable fluid, with the elastic constant set to 0 and relative permittivity to 10^6 inside the crack [25].

Figure 2-1 (b) shows the element division for performing finite element phase-field analysis on the model. The region measuring $8 \text{ nm} \times 7 \text{ nm}$ near the crack tip, where significant strain gradient and polarization gradient are expected, undergoes subdivision into fine meshes of 0.1 nm , and the other regions are subdivided into meshes ranging from 0.1 nm to 4 nm

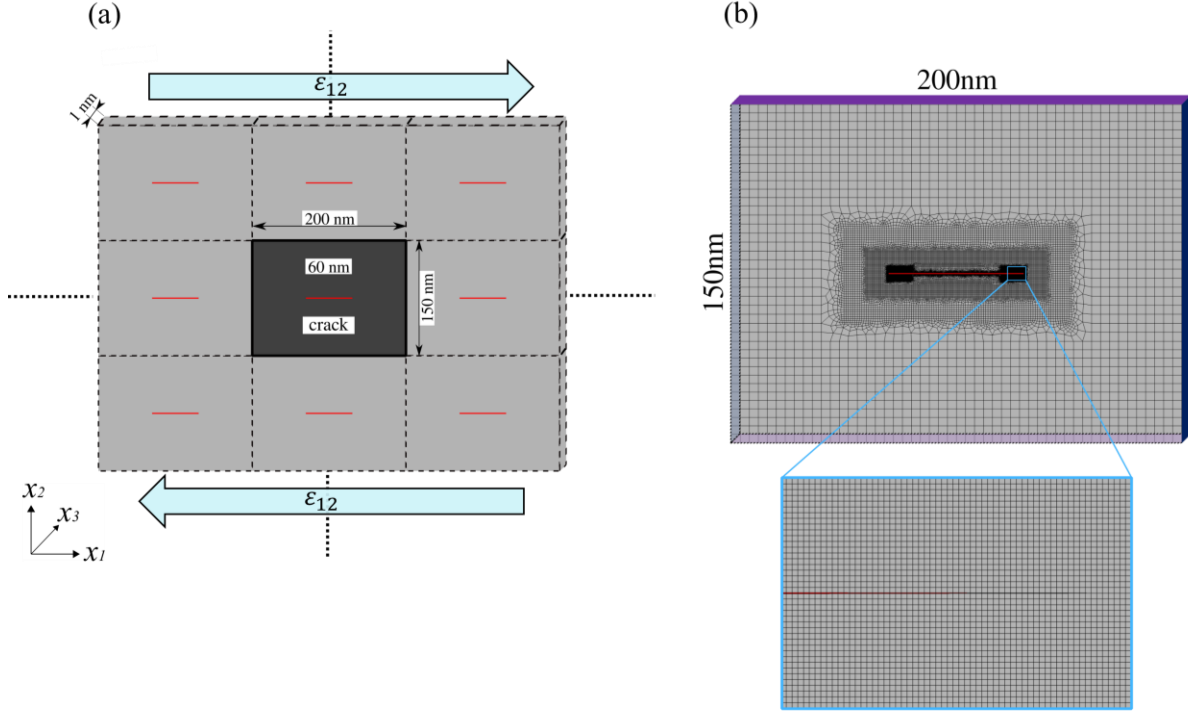


Figure 2-1 (a) Schematic illustration of a crack in SrTiO₃ with three-dimensional periodic boundary conditions. (b) Mesh divisions of the simulation model. The mesh size around the crack is 0.1 nm .

2.2.3 Simulation procedure

First, the thermodynamic equilibrium state with no applied load is calculated. Random polarization P_1 and P_2 with a small magnitude of $-1.0 \sim 1.0 \text{ C} \cdot \text{m}^{-2}$ is given as an initial state. The governing equations are solved using the finite element method with a time step of $\Delta t = 0.01 \text{ ns}$. Iterations of phase-field method continue until the change in total free energy density per unit time step time is less than $1 \text{ J} \cdot \text{m}^{-3}$ to achieve thermodynamic equilibrium state.

Next, a small mode II load [in-plane shear stress; see Figure 1(a)] is applied to the model and the thermodynamic equilibrium state under mode II load is obtained using the same iterative procedure. Repeating these procedures of applying an incremental mode II load and calculating the thermodynamics equilibrium state, a quasi-static mode II loading simulation progresses until

the mode II load reaches $K_{II} = 0.6 \text{ MPa}\sqrt{\text{m}}$, a value below the fracture toughness of SrTiO_3 [26]. These simulations are conducted at room temperature (298K), using the general-purpose finite element method software COMSOL (ver. 5. 4).

2.3 Results and discussion

2.3.1 Singular stress field-induced topological polar nano-region

Polarization distribution of SrTiO_3 with an edge crack is investigated using phase-field simulation. In order to extract only the influence of the singular stress fields, the crack is assumed to be filled with permeable fluid avoiding effects of depolarization field of crack surface. Figure 2-2 shows the polarization distribution in the vicinity of the crack tip under mode II loading. Gray areas in the figure indicate the paraelectric phase (i.e., $\mathbf{P} = 0$), while colored areas indicate the ferroelectric phase. Contours in the figure indicate the polarization magnitude. Before loading ($K_{II} = 0 \text{ MPa}\sqrt{\text{m}}$)[Figure 2-2(a)], there is no polarization, and the system remains in the paraelectric phase. On the other hand, when mode II loaded and the stress intensity factor reaches $K_{II} = 0.2 \text{ MPa}\sqrt{\text{m}}$, spontaneous polarization appears, resulting in a paraelectric-to-ferroelectric transition near the crack tip. The emerging polar region is

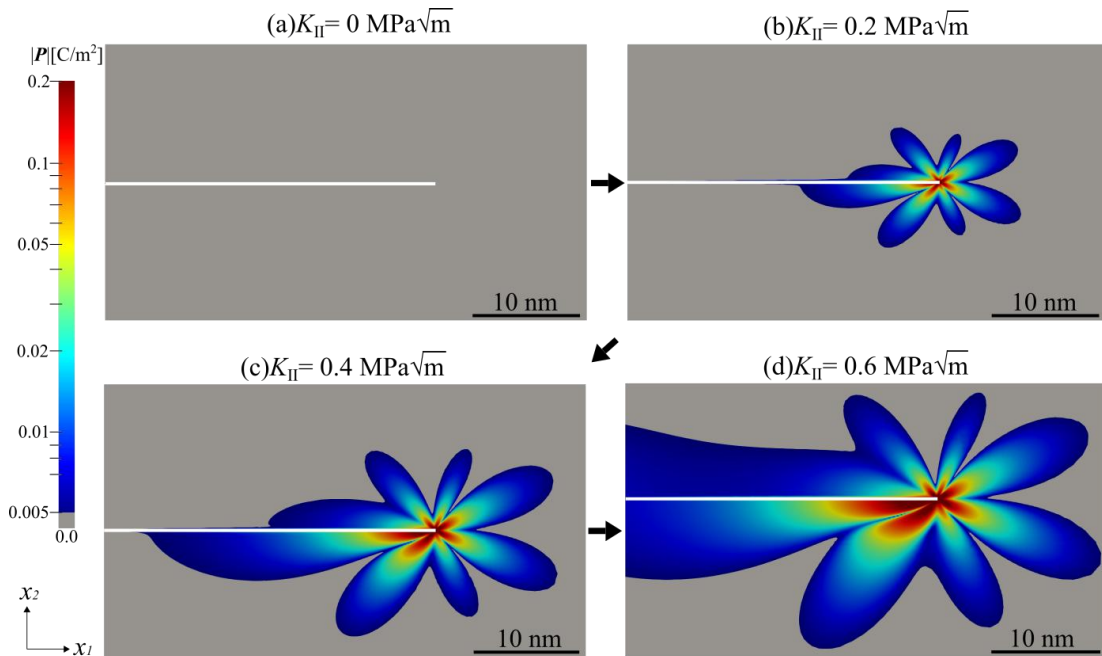


Figure 2-2 Polarization distribution near the crack tip in SrTiO_3 subjected to mode II load. The contour color indicates the polarization magnitude. The white line represents the crack.

approximately 5 nm in size [Figure 2-2(b)]. Within this polar nano-region, the magnitude of polarization is higher near the crack tip and diminishes with distance, akin to a singular stress field concentrated around the crack tip, suggesting its induction by the singular stress field. Moreover, the shape of the polar nano-region consists of eight small elliptical regions arranged in a radially around the crack tip. With increasing load to $K_{II} = 0.6 \text{ MPa}\sqrt{\text{m}}$, the polar nano-region expands [Figure 2-2(c)(d)], and the polarization magnitude inside the region also increases, indicating its significant dependence on the load level (stress intensity factor). This load value is below the fracture toughness of SrTiO_3 [26], and crack propagation would not occur. Surrounded by a paraelectric area without polarization, the polar nano-region can be regarded as an isolated nano ferroelectric material embedded within the paraelectric material. Notably, this polar nano-region does not exist in unloaded state but emerges near the crack tip under stress concentration by external force, suggesting its induction by the singular stress field. Therefore, these results indicate that the singular stress field induced by mode II load can mechanically create ferroelectric polar region in paraelectric SrTiO_3 .

Figure 2-3(a) shows the polarization directions in the polar nano-region at $K_{II} = 0.2 \text{ MPa}\sqrt{\text{m}}$. The gray area denotes absence of polarization, and the color scale indicates the in-plane angle of the polarization from the x_1 axis. Eight polar domains are present, with white arrows indicating

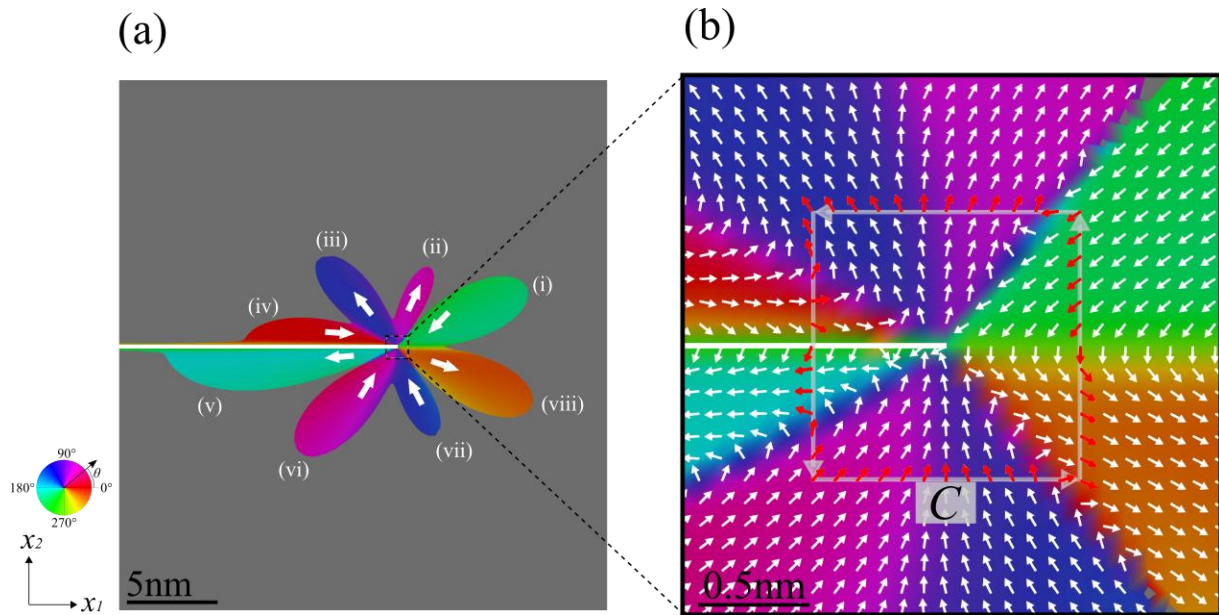


Figure 2-3 Polarization direction distribution of polar nano-region formed near the crack tip at $K_{II} = 0.2 \text{ MPa}\sqrt{\text{m}}$. (a) overview and (b) enlarged view. The white lines represent the crack. The contour color indicates the direction of the polarization. I set the integration path C as illustrated to calculate the winding number.

the polarization direction in each domain. For instance, the polarization direction of the domains (i) and (ii) are 220 and 80 degrees, and at the boundary of these domains, polarizations rotate 140 degrees. As magnified in Figure 2-3(b), near the crack tip, adjacent domains (i) and (ii) are connected, forming a domain wall through continuous polarization transition. Similar continuous angular changes in polarization direction occur at the boundaries between other domains, ranging from 70 to 180 degrees, exceeding 90-degree domain walls observed in typical ferroelectrics [27]. All domains and domain walls converge at the crack tip, where the polarization become discontinuous, resulting in a singularity. Given the presence of singularities in typical two-dimensional topological structures such as vortices [28], this polar nano-region may constitute a two-dimensional topological structure.

In order to investigate the topological property of the polar nano-region induced by mode II load, I evaluate the winding number n_v defined as [29]

$$n_v = \frac{1}{2\pi} \oint_C \nabla \theta dr, \quad (2.12)$$

where θ is the counterclockwise angle of polarization from x_1 axis, and C is integral path. For the polarization distribution at $K_{II} = 0.2 \text{ MPa}\sqrt{\text{m}}$, the integral path C around the crack tip is delineated [gray line in Figure 2-3(b)], yielding a calculated winding number of $n_v = -2$. This differs from the single-domain polarization ($n_v = 0$) observed in macroscale ferroelectrics and the vortex ($n_v = 1$) in nanoscale ferroelectrics [10], indicating a unique non-trivial topological feature in the polar nano-region. Therefore, this result indicates that the mode II loading induces a novel two-dimensional topological structure with $n_v = -2$ via the singular stress field formed near the crack tip in SrTiO_3 .

Figure 2-4 shows the polarization direction in the polar nano-regions at $K_{II} = 0.4$, $0.6 \text{ MPa}\sqrt{\text{m}}$. When the mode II load increases to $K_{II} = 0.4 \text{ MPa}\sqrt{\text{m}}$ [Figure 2-4(a)], each ferroelectric domain expands, accompanied by a corresponding expansion of the domain walls. Notably, a polar vortex with a topological number of $n_v = 1$ forms in the area indicated by the gray box while the non-trivial $n_v = -2$ polar structure persists at the crack tip. An additional polar vortex emerges as the mode II load increases to $0.6 \text{ MPa}\sqrt{\text{m}}$ [Figure 2-4(b)]. These results indicate that higher external loads induce more topological structures. According to the topology theory, a conservation law holds for the winding number, ensuring that the overall winding number of an order containing multiple orders equals to the sum of the winding numbers of the individual orders. The relationship between the total winding numbers and external loads is depicted in Figure 2-5. As the mode II load increases from 0 to 0.2, 0.4, and $0.6 \text{ MPa}\sqrt{\text{m}}$, the

total winding number discontinuously switches from 0, -2 , -1 , and 0. This result implies mechanical tuning of the topological features of two-dimensional polar structure.

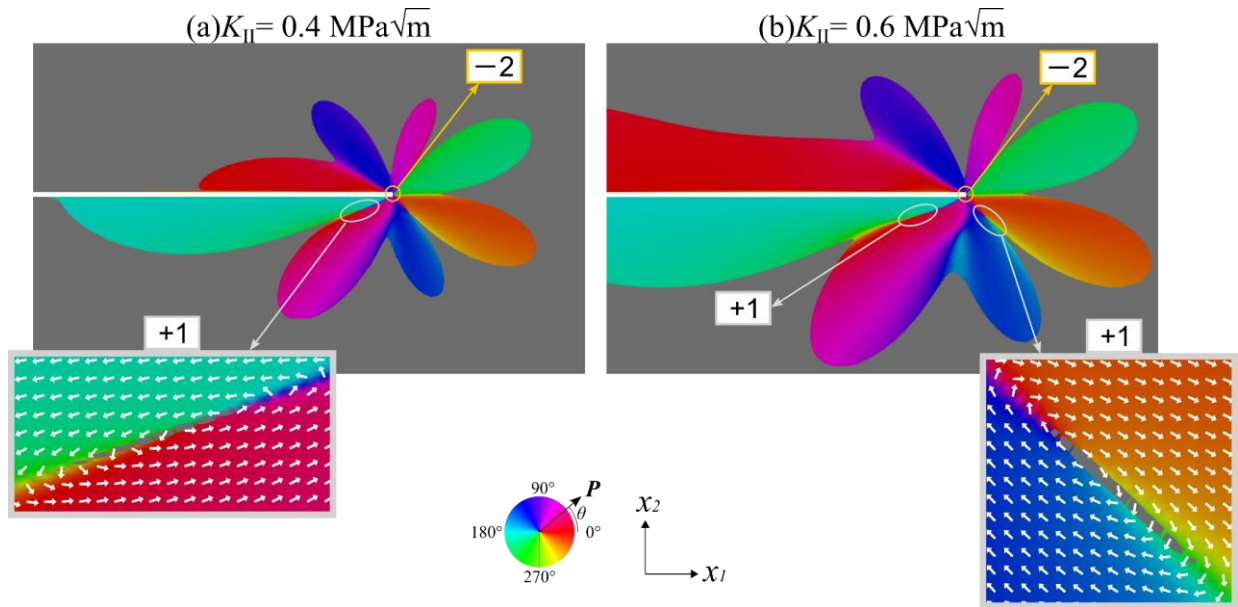


Figure 2-4 Polarization direction distribution of polar nano-region near the crack tip at $K_{II} = 0.4$ and $0.6 \text{ MPa}\sqrt{\text{m}}$. The white line represents the crack. The contour color indicates the angle between the polarization vector and the x direction.

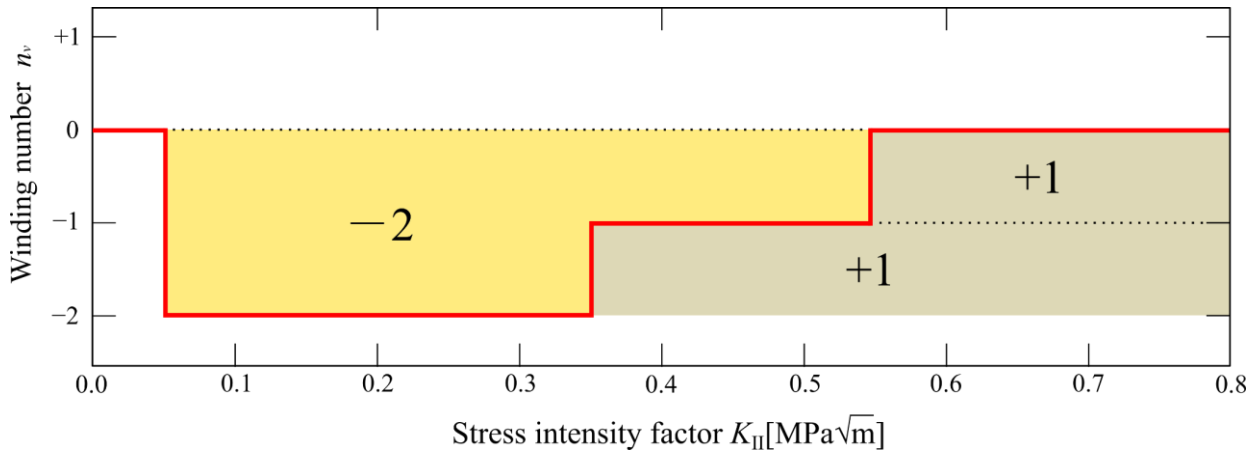


Figure 2-5 The total winding number of polar nano-region in increasing external loads.

2.3.2 Electromechanical formation mechanism of topological polar structures

To comprehend the mechanism behind the formation of non-trivial topological polar structures induced by mode II load, I focus on two mechanical-electrical coupling effects: the

electrostrictive effect and the flexoelectric effect. To separate the contribution of these effects, two models are prepared: model E considers only electrostrictive effect, and model F considers only flexoelectric effect. Figure 2-7 shows the polarization direction distribution in model E and model F. At $K_{II} = 0.2 \text{ MPa}\sqrt{\text{m}}$, as shown in Figure 2-7(a-1), eight elliptical domains appear in the model F. The shape of this polarization area and polarization directions are closely corresponding to the polar nano-region emerged in the model that includes both effects. Additionally, the winding number of this polarization area is calculated as $n_v = -2$. In contrast, in model E, almost no polarization is observed. Therefore, the $n_v = -2$ structure at crack tip is mainly attributed to the flexoelectric effect. At $K_{II} = 0.4$ and $0.6 \text{ MPa}\sqrt{\text{m}}$, while in model F eight small domains expand and $n_v = -2$ structure remains, in model E, polarization area appears near the crack tip. This polarization area is consistent with the vortices formed with increasing load shown in Figure 2-4. These results indicate that the flexoelectric effect shapes the topological structure of $n_v = -2$, and both effects contribute to the vortex formation.

Then, how flexoelectric effects drive two-dimensional topological structure is described in detail. The polarization induced by flexoelectric effect is expressed as $P_i = \mu_{klij} \partial \varepsilon_{kl} / \partial x_j$ and its components can be written as

$$P_1 = \mu_{11} \frac{\partial \varepsilon_{11}}{\partial x_1} + \mu_{12} \frac{\partial \varepsilon_{22}}{\partial x_1} + \mu_{44} \frac{\partial \varepsilon_{12}}{\partial x_2}, \quad (2.13)$$

$$P_2 = \mu_{11} \frac{\partial \varepsilon_{22}}{\partial x_2} + \mu_{12} \frac{\partial \varepsilon_{11}}{\partial x_2} + \mu_{44} \frac{\partial \varepsilon_{12}}{\partial x_1}, \quad (2.14)$$

where μ_{11} , μ_{12} , and μ_{44} are flexoelectric coefficients [24]. Defining the strain gradient vectors $\mathbf{S}_1 = \left(\frac{\partial \varepsilon_{11}}{\partial x_1}, \frac{\partial \varepsilon_{22}}{\partial x_2} \right)$, $\mathbf{S}_2 = \left(\frac{\partial \varepsilon_{22}}{\partial x_1}, \frac{\partial \varepsilon_{11}}{\partial x_2} \right)$, and $\mathbf{S}_3 = \left(\frac{\partial \varepsilon_{12}}{\partial x_2}, \frac{\partial \varepsilon_{12}}{\partial x_1} \right)$, Eqs. (2) and (3) can be written as

$$\mathbf{P} = \mu_{11} \mathbf{S}_1 + \mu_{12} \mathbf{S}_2 + \mu_{44} \mathbf{S}_3 \quad (2.15)$$

where $\mathbf{P} = (P_1, P_2)$ is the flexoelectric polarization vector induced by strain gradient. Figure 2-6 shows the distribution of polarization $\mathbf{P}$ and three strain gradient vectors $\mathbf{S}_1$, $\mathbf{S}_2$, and $\mathbf{S}_3$ in the nano-region at $K_{II} = 0.2 \text{ MPa}\sqrt{\text{m}}$. For instance, in domain (i), the directions of $\mathbf{S}_1$, $\mathbf{S}_2$, and $\mathbf{S}_3$ are approximately 220 degrees, aligning with the polarization direction. In domain (ii), which has 50 degrees polarization, $\mathbf{S}_2$ is same direction as polarization. Similarly, for domains (iii) ~ (viii), at least one of the strain gradient vectors align with the polarization vectors. This alignment indicates that the strain gradient from the mode II singular stress field induces a topological polar structure near the crack tip via the flexoelectric effect. In addition, each strain gradient vector undergoes significant transition in angle around the crack tip. In other words, the topological features hidden in the strain gradient field near the crack tip are reflected in the

polarization field. Similar discussions can apply to the case of mode I load [19], indicating that manipulating the topological feature of singular stress fields holds promise for creating higher-order and non-trivial topological polar structures.

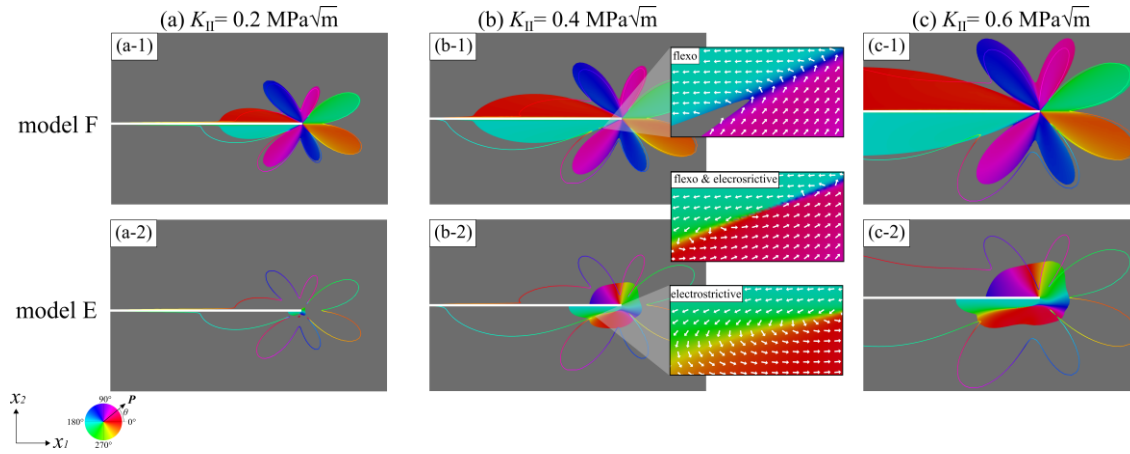


Figure 2-7 Polarization direction of polar nano-region induced by mode II load of (1) model F (only flexoelectric effect) (2) model E (only electrostrictive effect). As a comparison, the colored lines represent the outline of nano polar region with both flexoelectric and electrostrictive effect.

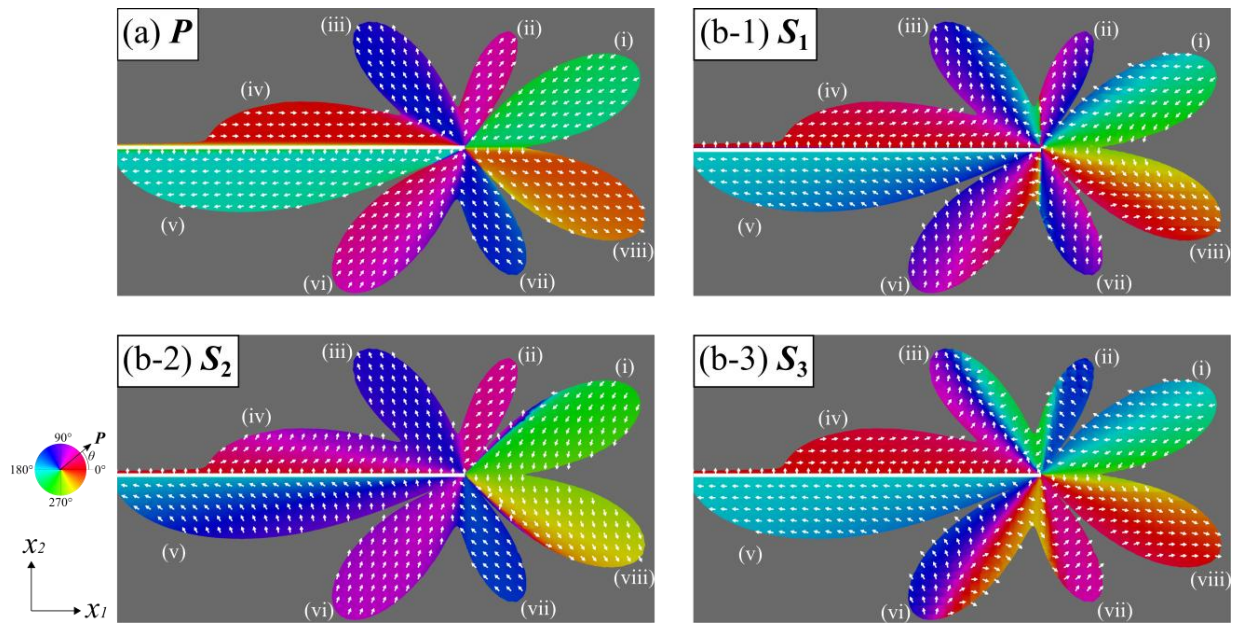


Figure 2-6 (a) Polarization direction and (b) strain gradient vector distribution of polar nano-region induced by mode II load. The white line represents the crack. The contour color indicates the angle between each vector and the x_1 axis. Note that considering the negative flexoelectric coefficient μ_{11} , S_1 is shown reversed to make clear the contribution to the polarization.

2.3.3 Mechanical control of topological polar structures

To demonstrate a more diverse control of the topological properties of the polar structures induced by geometric mechanical fields, I simulate the polarization distribution formed by mixed mode load of mode I and mode II. As an example, cracks in various directions have been observed experimentally in SrTiO₃ thin films [30,31], suggesting that mixed-mode loading is possible by epitaxial strain and other factors. The simulation model is same as mode II case, and only direction of load is changed. Generally, the magnitude of the mixed mode load is expressed by $K = \sqrt{K_I^2 + K_{II}^2}$ using the stress intensity factors K_I and K_{II} for each single mode load. By using K and the mode ratio $\alpha = K_{II}/K_I$, mixed mode load conditions are determined. In this chapter, K is fixed at 0.2 and only the mode ratio α is changed from 0 to ∞ . Similar to mode I and mode II loadings, the mixed mode loading can induce the polar nano-region consisting of multiple domains near the crack tip as shown in Figure 2-8(b), while the number of domains is larger than either, 10. Moreover, the winding number represented in Eqs. (1) is calculated as $n_v = -4$. This value not only differs from typical polar structure such as linear polarization or vortex, but also different from polarization distribution induced by mode I and mode II load. Therefore, this simulation demonstrates the mechanically creation of novel two-dimensional topological polar structure with high winding number by mixed mode load into a nano crack in SrTiO₃.

To compare the polarization distributions across various mode ratios, Figure 2-8(a) and (c) shows the polarization direction distribution in the vicinity of the crack tip under mode I load [19] (mode ratio $\alpha = 0$), and mode II load ($\alpha = \infty$) at $K = 0.2 \text{ MPa}\sqrt{\text{m}}$, respectively. Among the ten domains in mixed mode, (i) and (ii), marked in the figures, appear in the same positions and polarization direction as those in mode I. Similarly, (iii) correspond to that in mode II, while the other domains are observed across all mode ratios. In other words, the polarization distribution induced by mixed mode load encompasses all the domains present in both mode I and mode II load. This reflects the geometric properties of the mechanical field by mixed mode load which is superposition of mode I and mode II load. These polarization distributions have different winding numbers, $n_v = -3$ for mode I, -4 for mixed mode, and -2 for mode II, due to the difference in domain structures.

Figure 2-9 shows the polarization domain configuration surrounding the crack tip at various mode ratios $\alpha = 0, 1, 2, \infty$ alongside the winding number n_v . As the mode ratio changes from mode I ($\alpha = 0$) to mode II ($\alpha = \infty$), the winding number transitions as $n_v = -3, -4, -3, -2$.

Thus, the topological attributes of polarization distribution depend on mode ratios. In Figure 2-9, dashed lines indicate polarization domains arranged radially around the crack tip, with intervening domain walls. Some domains indicated by dashed black lines are maintained across all mode ratios. These domains and the domain walls between them do not affect the winding number because the transition in polarization direction around the crack tip remains consistent for each mode ratio. On the other hand, domains (1), (2), and (3), indicated by dashed white lines, only appear at specific mode ratios, leading to changes in the polarization direction around the crack tip. Consequently, alternations in the polarization distribution around the crack tip due to the appearance or disappearance of these domains results in the changes of winding number.

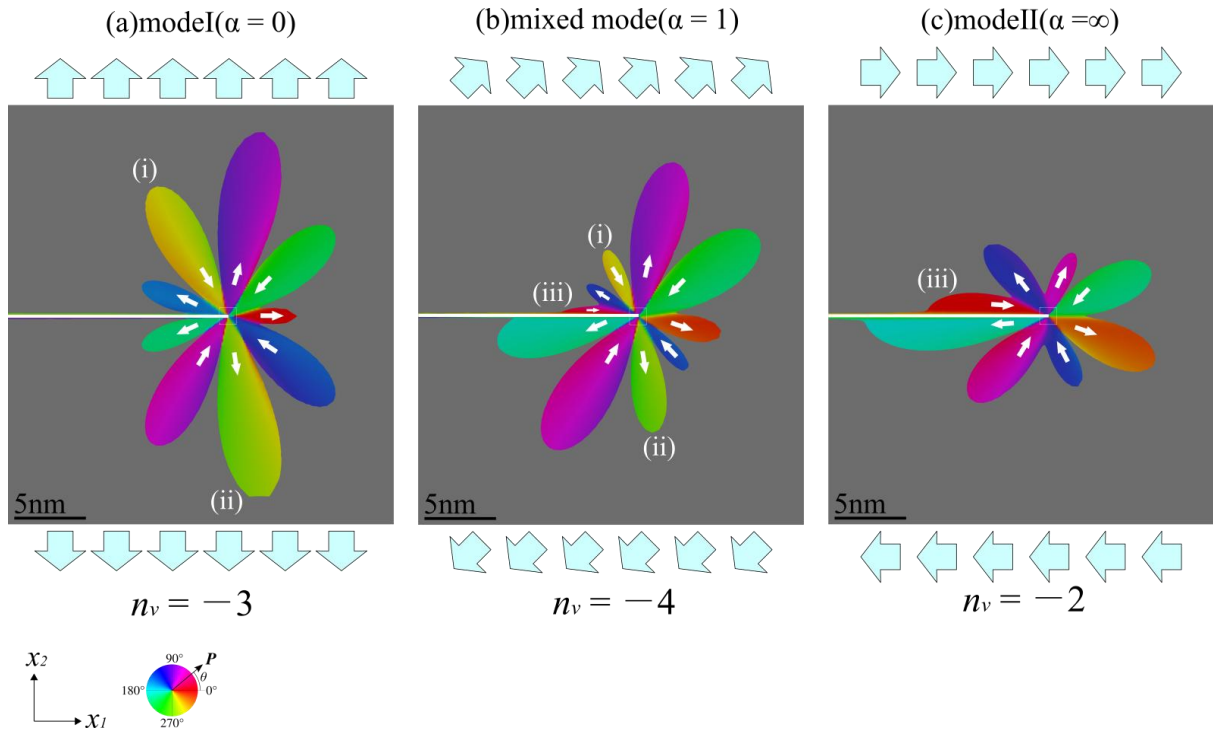


Figure 2-8 Polarization direction distribution of polar nano-region near the crack tip under (a) mode I [17] (b) mixed mode (c) mode II load at $K = 0.2 \text{ MPa}\sqrt{\text{m}}$. The contour color indicates the direction of polarization, and the white lines represent the cracks.

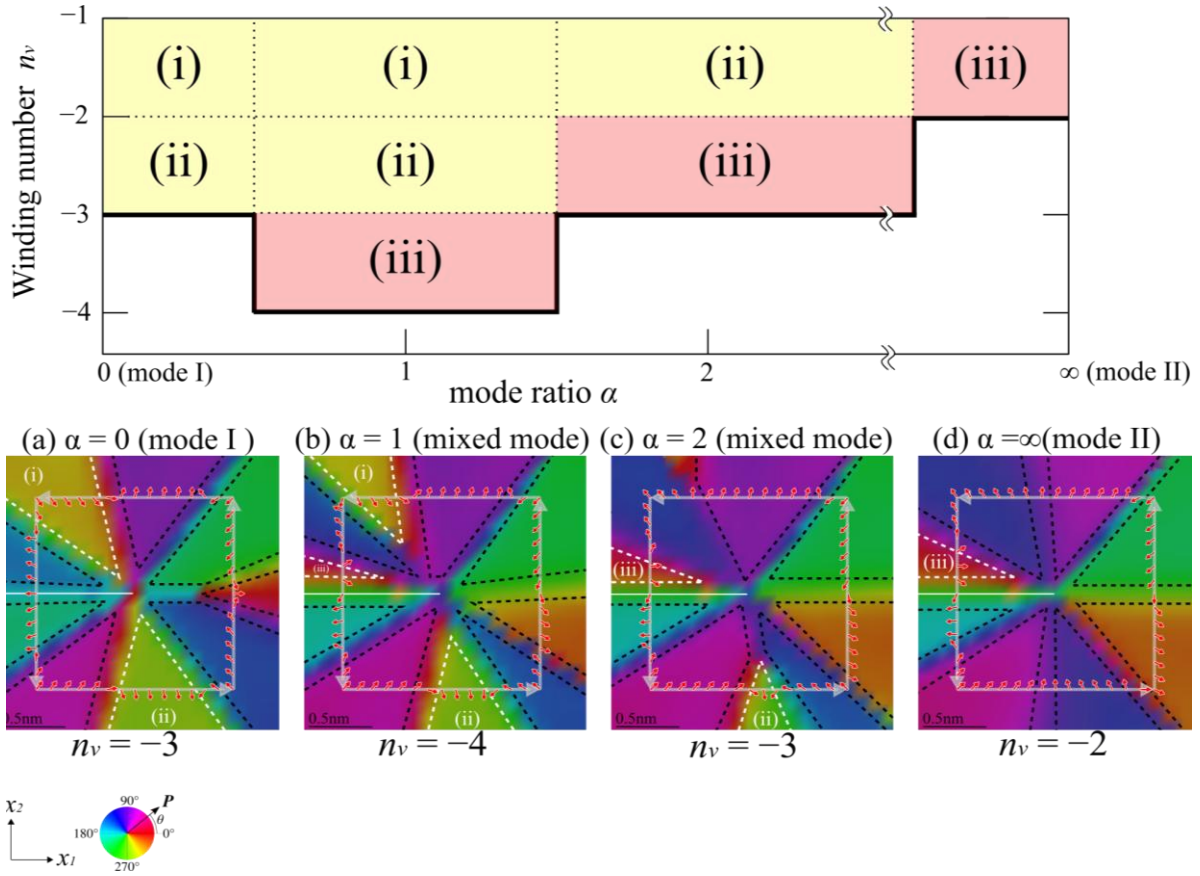


Figure 2-9 Transition of winding number of polarization distributions near the crack tip in different mode ratio.

2.4 Conclusion

In summary, by using phase-field simulations, I demonstrated the mechanical creation and control of two-dimensional topological polar structures by mode II and mixed mode load into nano crack in SrTiO₃. The strain fields of various magnitudes and distributions induce a variety of topological polar structures, mirroring the topological features of mechanical strain and strain gradient fields. Since mixed mode loading with geometrically complex mechanical fields produce the various topological polar structures including one with high topological number ($n_v = -4$), superposition of additional strain fields may produce much higher topological number structures. Furthermore, strain and strain gradient fields from any sources can also induce polar structure reflecting the topological features of them. The results provide a path to design and control functional topological polar structures in ferroelectrics through ‘topological strain field engineering’.

References

- [1] T. Nagai, H. Yamada, M. Konoto, T. Arima, M. Kawasaki, K. Kimoto, Y. Matsui, and Y. Tokura, Direct observation of the spin structures of vortex domain walls in ferromagnetic nanowires, *Phys. Rev. B* **78**, 180414 (2008).
- [2] J. Wang and J. Zhang, A real-space phase field model for the domain evolution of ferromagnetic materials, *International Journal of Solids and Structures* **50**, 3597 (2013).
- [3] X. Z. Yu, W. Koshibae, Y. Tokunaga, K. Shibata, Y. Taguchi, N. Nagaosa, and Y. Tokura, Transformation between meron and skyrmion topological spin textures in a chiral magnet, *Nature* **564**, 7734 (2018).
- [4] H. Jani et al., Antiferromagnetic half-skyrmions and bimerons at room temperature, *Nature* **590**, 74 (2021).
- [5] S. Mühlbauer, B. Binz, F. Jonietz, C. Pfleiderer, A. Rosch, A. Neubauer, R. Georgii, and P. Böni, Skyrmion Lattice in a Chiral Magnet, *Science* **323**, 915 (2009).
- [6] X. Z. Yu, Y. Onose, N. Kanazawa, J. H. Park, J. H. Han, Y. Matsui, N. Nagaosa, and Y. Tokura, Real-space observation of a two-dimensional skyrmion crystal, *Nature* **465**, 901 (2010).
- [7] A. Fert, V. Cros, and J. Sampaio, Skyrmions on the track, *Nature Nanotech* **8**, 152 (2013).
- [8] A. Neubauer, C. Pfleiderer, B. Binz, A. Rosch, R. Ritz, P. G. Niklowitz, and P. Böni, Topological Hall Effect in the A Phase of MnSi, *Phys. Rev. Lett.* **102**, 186602 (2009).
- [9] C. Ederer and N. A. Spaldin, Weak ferromagnetism and magnetoelectric coupling in bismuth ferrite, *Phys. Rev. B* **71**, 060401 (2005).
- [10] B. J. Rodriguez, X. S. Gao, L. F. Liu, W. Lee, I. I. Naumov, A. M. Bratkovsky, D. Hesse, and M. Alexe, Vortex Polarization States in Nanoscale Ferroelectric Arrays, *Nano Lett.* **9**, 1127 (2009).
- [11] Z. Jiang and J. Wang, Stability of chiral polarization vortex in strained ferroelectric superlattices, *Journal of Applied Physics* **131**, 164101 (2022).
- [12] Y. Nahas, S. Prokhorenko, L. Louis, Z. Gui, I. Kornev, and L. Bellaiche, Discovery of stable skyrmionic state in ferroelectric nanocomposites, *Nat Commun* **6**, 8542 (2015).
- [13] S. Das et al., Observation of room-temperature polar skyrmions, *Nature* **568**, 368 (2019).
- [14] Y.-T. Shao et al., Emergent chirality in a polar meron to skyrmion phase transition., *Nat Commun* **14**, 1355 (2023).

- [15] Y. J. Wang et al., Polar meron lattice in strained oxide ferroelectrics, *Nat. Mater.* **19**, 881 (2020).
- [16] V. Govinden et al., Ferroelectric solitons crafted in epitaxial bismuth ferrite superlattices, *Nat Commun* **14**, 4178 (2023).
- [17] W. Ma, A study of flexoelectric coupling associated internal electric field and stress in thin film ferroelectrics, *Physica Status Solidi (b)* **245**, 761 (2008).
- [18] E. A. Eliseev, A. N. Morozovska, M. D. Glinchuk, and R. Blinc, Spontaneous flexoelectric/flexomagnetic effect in nanoferroics, *Phys. Rev. B* **79**, 165433 (2009).
- [19] T. Shimada, Y. Wang, T. Hamaguchi, K. Kasai, K. Masuda, L. Van Lich, T. Xu, J. Wang, and H. Hirakata, Emergence of non-trivial polar topologies hidden in singular stress field in SrTiO_3 : topological strain-field engineering, *J. Phys.: Condens. Matter* **33**, 505301 (2021).
- [20] P. V. Yudin and A. K. Tagantsev, Fundamentals of flexoelectricity in solids, *Nanotechnology* **24**, 432001 (2013).
- [21] W. Ma, A study of flexoelectric coupling associated internal electric field and stress in thin film ferroelectrics, *Phys. Stat. Sol. (b)* **245**, 761 (2008).
- [22] J. H. Haeni et al., Room-temperature ferroelectricity in strained SrTiO_3 , **430**, 4 (2004).
- [23] G. Sheng, Y. L. Li, J. X. Zhang, S. Choudhury, Q. X. Jia, V. Gopalan, D. G. Schlom, Z. K. Liu, and L. Q. Chen, A modified Landau–Devonshire thermodynamic potential for strontium titanate, *Appl. Phys. Lett.* **96**, 232902 (2010).
- [24] P. Zubko, G. Catalan, A. Buckley, P. R. L. Welche, and J. F. Scott, Strain-Gradient-Induced Polarization in SrTiO_3 Single Crystals, *Phys. Rev. Lett.* **99**, 167601 (2007).
- [25] J. Wang and M. Kamlah, Effect of electrical boundary conditions on the polarization distribution around a crack embedded in a ferroelectric single domain, *Engineering Fracture Mechanics* **77**, 3658 (2010).
- [26] B.-K. Park, J.-W. Lee, S.-B. Lee, T.-H. Lim, S.-J. Park, R.-H. Song, W. B. Im, and D.-R. Shin, La-doped SrTiO_3 interconnect materials for anode-supported flat-tubular solid oxide fuel cells, *International Journal of Hydrogen Energy* **37**, 4319 (2012).
- [27] J. Wang, S.-Q. Shi, L.-Q. Chen, Y. Li, and T.-Y. Zhang, Phase-field simulations of ferroelectric/ferroelastic polarization switching, *Acta Materialia* **52**, 749 (2004).
- [28] J. C. Y. Teo and T. L. Hughes, Topological Defects in Symmetry-Protected Topological Phases, *Annu. Rev. Condens. Matter Phys.* **8**, 211 (2017).
- [29] N. D. Mermin, The topological theory of defects in ordered media, *Rev. Mod. Phys.* **51**, 591 (1979).

- [30] T. Ostapchuk et al., Origin of soft-mode stiffening and reduced dielectric response in SrTiO_3 thin films, *Phys. Rev. B* **66**, 235406 (2002).
- [31] E. Hollmann, J. Schubert, R. Kutzner, and R. Wördenweber, Stress generated modifications of epitaxial ferroelectric SrTiO_3 films on sapphire, *Journal of Applied Physics* **105**, 114104 (2009).

Chapter 3 Emergence of isolated polar skyrmion and quasi two-dimensional topological states by nanoindentation

3.1 Introduction

In chapter 2, mechanical creation and control of two-dimensional topological polar structure is demonstrated. This chapter aims to apply the proposed method into three-dimensional systems and realize mechanically induced quasi-two-dimensional topological structures.

Topological structures are local configurations with topological features in condensed matters. Magnetic skyrmions [1,2] are one of the typical topological structures in ferromagnetics. They were first considered theoretically [1], and later observed experimentally [2]. They exhibit quasiparticle behavior such as stability due to topological protection, Hall effect [3], and current-driven motion with ultralow current density [4]. Because of these properties, magnetic skyrmions are expected to be applied to racetrack memory [5] and logical gates [6]. Also, in the realm of ferroelectrics, topological structures offer considerable potential for nanotechnology devices, such as dielectric energy storage [7].

The presence of intrinsic non-collinear interaction, i.e., Dzyaloshinskii–Moriya interaction (DMI), in certain magnets [8,9] facilitates the emergence of intricate topological structures like magnetic merons [10,11] and skyrmions [2,12]. Conversely, the polarization arrangements in ferroelectrics exhibit relatively monotonous (such as polar vortex [13–15]). This may be due to the absence of DMI-like effects in ferroelectrics, however, recent theoretical studies predict the existence of non-collinear interaction analogous to DMI [16,17], and the emergence of various nontrivial polar topologies are expected. In literature, with the help of sophisticated boundary conditions and specimen structures, several polar topological structures, such as chiral domain wall [18], chiral vortex [19], polar bubbles [20], superhelices [21], merons [22,23], and skyrmions [24,25] are created in ferroelectrics. Especially, due to the rich electro-elastic coupling and complex strain distribution from the interfaces, the multilayer PbTiO_3 (PTO) and

SrTiO₃ (STO) heterostructures have become a popular playground for polar topologies. Tang et al [26] observed the flux-closure polar structures in the PTO/STO superlattices at first. Then, Yadav et al. [27] observed electrical controlled vortex-antivortex, and Shafer et al. [28] demonstrated their chirality. Das et al. [29] found that the large strain can even create the room-temperature polar skyrmions in the PTO/STO superlattices. These findings stimulate the continuing quest for polar topologies. However, previous polar chiral vortex or skyrmions are strongly dependent on the specimen structure or strict boundary conditions [14] and hard to be controlled, which leads to critical limitations in application. Therefore, creating and controlling polar topologies in a relatively simple system such as ferroelectric thin film is highly desired for their further application.

Here, mechanical loading can be an effective tool for creating polar topologies. Recent experimental work by Lu et. al. [30] showed the switching of the local polarizations by a mechanical indentation via the flexoelectric effect in a ferroelectric thin film, and they find the switched area is stable as a domain pattern. Meanwhile, Shimada et al [31] demonstrated the flexoelectric effect can provide the topological drive for the mechanical design of polar topologies in theory as “topological strain-field engineering”. In addition, Morozovska et. al. [32] pointed out the similarities between the flexoelectric effect and the DMI, and control of chiral polarization by flexoelectric effect has been studied [33]. These studies suggest the potential of flexoelectric effect as an extrinsic DMI-like inducer of polar topologies. Accordingly, this chapter aims to introduce the flexoelectric effect as a non-collinear interaction and demonstrate mechanical designing and controlling room-temperature polar topologies like skyrmion in the simple ferroelectric thin film.

3.2 Simulation method

3.2.1 Phase-field modeling

The total free energy of a ferroelectric system is $F = \int f dV$, where the f is the total free energy density and can be expressed as

$$f = f_{\text{Landau}} + f_{\text{grad}} + f_{\text{electo}} + f_{\text{elastic}} + f_{\text{ecoup}} + f_{\text{flexo}}. \quad (3.1)$$

The Landau energy density [34], which describes the temperature-dependent ferroelectric phase transition, can be written as

$$\begin{aligned}
f_{\text{Landau}} = & \alpha_{ij}P_iP_j + \alpha_{ijkl}P_iP_jP_kP_l + \alpha_{ijklmn}P_iP_jP_kP_lP_mP_n = \alpha_1(P_1^2 + \\
& P_2^2 + P_3^2) + \alpha_{11}(P_1^4 + P_2^4 + P_3^4) + \alpha_{12}(P_1^2P_2^2 + P_1^2P_3^2 + P_2^2P_3^2) + \\
& \alpha_{111}(P_1^6 + P_2^6 + P_3^6) + \alpha_{112}(P_1^4(P_2^2 + P_3^2) + P_2^4(P_1^2 + P_3^2) + P_3^4(P_1^2 + \\
& P_2^2)) + \alpha_{123}(P_1^2P_2^2P_3^2),
\end{aligned} \tag{3.2}$$

where α_{ij} , α_{ijkl} , and α_{ijklmn} are the Landau expansion coefficients. The gradient energy density, which conducts the formation of the polarization domains, can be written as

$$\begin{aligned}
f_{\text{grad}} = & \frac{1}{2}G_{ijkl}P_{i,j}P_{k,l} = \frac{1}{2}G_{11}(P_{1,1}^2 + P_{2,2}^2 + P_{3,3}^2) + G_{12}(P_{1,1}P_{2,2} + \\
& P_{2,2}P_{3,3} + P_{3,3}P_{1,1}) + \frac{1}{2}G_{44}[(P_{1,2} + P_{2,1})^2 + (P_{2,3} + P_{3,2})^2 + (P_{1,3} + \\
& P_{3,1})^2],
\end{aligned} \tag{3.3}$$

where G_{ijkl} is the gradient energy coefficient. The electrostatic energy density can be written as

$$\begin{aligned}
f_{\text{electo}} = & -\frac{1}{2}\kappa_0 E_i E_i - E_i P_i = -\frac{1}{2}\kappa_0(E_1^2 + E_2^2 + E_3^2) - E_1 P_1 - E_2 P_2 - \\
& E_3 P_3,
\end{aligned} \tag{3.4}$$

where κ_0 is the relative permittivity. The elastic energy density can be written as

$$\begin{aligned}
f_{\text{elastic}} = & \frac{1}{2}c_{ijkl}\varepsilon_{ij}\varepsilon_{kl} = \frac{1}{2}c_{11}(\varepsilon_{11}^2 + \varepsilon_{22}^2 + \varepsilon_{33}^2) + c_{12}(\varepsilon_{11}\varepsilon_{22} + \varepsilon_{22}\varepsilon_{33} + \\
& \varepsilon_{33}\varepsilon_{11}) + 2c_{44}(\varepsilon_{12}^2 + \varepsilon_{23}^2 + \varepsilon_{13}^2),
\end{aligned} \tag{3.5}$$

where c_{ijkl} is the elastic constant. The electrostrictive energy density, which describes the electrostrictive effect [35], can be written as

$$\begin{aligned}
f_{\text{ecoup}} = & -q_{ijkl}\varepsilon_{ij}P_kP_l = -(q_{11}\varepsilon_{11} + q_{12}\varepsilon_{22} + q_{12}\varepsilon_{33})P_1^2 - (q_{12}\varepsilon_{11} + \\
& q_{11}\varepsilon_{22} + q_{12}\varepsilon_{33})P_2^2 - (q_{12}\varepsilon_{11} + q_{12}\varepsilon_{22} + q_{11}\varepsilon_{33})P_3^2 - 2q_{44}(\varepsilon_{12}P_1P_2 + \\
& \varepsilon_{23}P_2P_3 + \varepsilon_{13}P_1P_3),
\end{aligned} \tag{3.6}$$

where q_{ijkl} is the electrostrictive coefficient. Eventually, the flexoelectric energy density, which describes the flexoelectric effect [35–37] by the Lifshitz invariant [38], can be written as

$$\begin{aligned}
f_{\text{flexo}} = & \frac{1}{2}f_{ijkl}(\varepsilon_{ij}P_{k,l} - P_k\varepsilon_{ij,l}) = \frac{1}{2}\{f_{11}(P_{1,1}\varepsilon_{11} + P_{2,2}\varepsilon_{22} + P_{3,3}\varepsilon_{33}) + \\
& f_{12}(P_{2,2}\varepsilon_{11} + P_{3,3}\varepsilon_{11} + P_{1,1}\varepsilon_{22} + P_{3,3}\varepsilon_{22} + P_{1,1}\varepsilon_{33} + P_{2,2}\varepsilon_{33}) + \\
& f_{44}[\varepsilon_{23}(P_{2,3} + P_{3,2}) + \varepsilon_{12}(P_{1,3} + P_{3,1}) + \varepsilon_{12}(P_{1,2} + P_{2,1})]\} - \\
& \frac{1}{2}\{f_{11}(\varepsilon_{11,1}P_1 + \varepsilon_{22,2}P_2 + \varepsilon_{33,3}P_3) + f_{12}[P_1(\varepsilon_{22,1} + \varepsilon_{33,1}) + P_2(\varepsilon_{11,2} + \\
& \varepsilon_{33,2}) + P_3(\varepsilon_{11,3} + \varepsilon_{22,3})] + f_{44}[P_1(\varepsilon_{12,2} + \varepsilon_{13,3}) + P_2(\varepsilon_{12,1} + \varepsilon_{23,3}) + \\
& P_3(\varepsilon_{13,1} + \varepsilon_{23,2})]\},
\end{aligned} \tag{3.7}$$

where f_{ijkl} is the flexoelectric coupling coefficient. All of material parameters used in this chapter are shown in Table 3-1 and Table 3-2. By following the previous study [39], the

paraelectric SrTiO₃ is treated as a simple elastic body, and the polarization is assumed to be always zero.

Table 3-1 The material parameters of PbTiO₃ [35,40–43].

Landau expansion coefficients			
$\alpha_1 [C^{-2} \cdot m^2 \cdot N]$	$3.8 \times (T - 753) \times 10^5$	$\alpha_{111} [C^{-6} \cdot m^{10} \cdot N]$	2.6×10^8
$\alpha_{11} [C^{-4} \cdot m^6 \cdot N]$	-7.3×10^7	$\alpha_{112} [C^{-6} \cdot m^{10} \cdot N]$	6.1×10^8
$\alpha_{12} [C^{-4} \cdot m^6 \cdot N]$	7.5×10^8	$\alpha_{123} [C^{-6} \cdot m^{10} \cdot N]$	-3.7×10^9
Elastic constants		Electrostrictive coefficients	
$c_{11} [m^{-2} \cdot N]$	1.75×10^{11}	$q_{11} [C^{-2} \cdot m^2 \cdot N]$	1.1446×10^{10}
$c_{12} [m^{-2} \cdot N]$	7.94×10^{10}	$q_{12} [C^{-2} \cdot m^2 \cdot N]$	4.522×10^8
$c_{44} [m^{-2} \cdot N]$	1.11×10^{11}	$q_{44} [C^{-2} \cdot m^2 \cdot N]$	7.548×10^9
Flexoelectric coupling coefficients		Gradient energy coefficients	
$f_{11} [V]$	0.6	$G_{11} [C^{-2} \cdot m^4 \cdot N]$	1.04×10^{-10}
$f_{12} [V]$	5	$G_{12} [C^{-2} \cdot m^4 \cdot N]$	-1.04×10^{-10}
$f_{44} [V]$	2	$G_{44} [C^{-2} \cdot m^4 \cdot N]$	1.04×10^{-10}

T is temperature (K).

Table 3-2 The material parameters of SrTiO₃ [31].

Elastic constants	
$c_{11} [m^{-2} \cdot N]$	3.36×10^{11}
$c_{12} [m^{-2} \cdot N]$	1.07×10^{11}
$c_{44} [m^{-2} \cdot N]$	1.27×10^{11}

3.2.2 Simulation model and procedure

Figure 3-1 shows the schematic of the simulation model in this chapter. The thickness of PTO thin film is set to 5 nm [30]. To consider a substrate with a constrained bottom surface, the displacement of the bottom surface of the STO substrate in the x_3 -direction is constrained. The thickness of the STO substrate is set to 50 nm as a sufficiently thick substrate to ensure that the restraint of the bottom surface of the substrate does not affect the mechanical state in the PTO

thin film during indentation loading. The length and the width of the PTO thin film and STO substrate are set to 80 nm, which is large enough compared with the size of the loading area. Additionally, the PTO thin film is subjected to short-circuit electrical boundary conditions, with the electrostatic potentials at the top surface and interface are set to 0 V [22]. This follows the situation of actual experimental conditions, where the compensation of surface polarization charge by impurities adsorbed on the thin film surface results in the absence of depolarization field inside the thin film. When the PTO thin film is epitaxially grown on the STO substrate, an in-plane misfit strain occurs due to the difference in lattice constants between the two materials. This misfit strain is accounted for by applying an in-plane biaxial strain of -1.6% [44] to the entire model. This value corresponds to an experimentally measured strain of 0.1% [44].

Figure 3-2(a) shows the mesh divisions of the simulation model for the finite element calculation in this chapter. For the x_1 - x_2 plane, the circular indentation area and its surroundings are divided by 1 nm, while the other areas are divided by up to 4 nm square. For the x_3 direction, the PTO thin film is divided by 0.5 nm and STO substrate is divided by 4 nm. Note that in the preliminary calculations using 2 nm square mesh for the entire of thin film, the mechanical writing process is reproduced, indicating the independence of the results from mesh division. For the calculations of moving indenter, the simulation model is extended by up to 40 nm in the x_1 and x_2 directions, and divided by 2 nm squares in the in-plane direction.

The method of applying the in-plane biaxial strain with the periodical boundary conditions is shown in Figure 3-2(b). As shown in Figure 1b, the solid lines represent the real boundaries of the specimen, and dashed lines represent the virtual boundaries. The position vectors of the left-

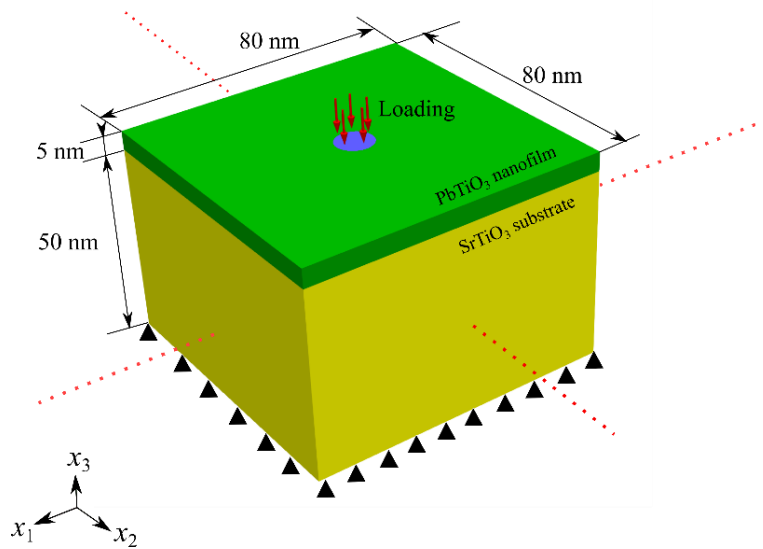


Figure 3-1 Schematic of simulation model of PbTiO_3 nanofilm grown on SrTiO_3 substrate. The blue circle represents the loading area.

bottom point of the specimen ($\vec{u}^0$) and the corresponding point ($\vec{u}^1$) on the virtual boundary are marked by red circles in Figure 3-2(b). And the vector connecting these two points ($\Delta\xi$) is used for setting the displacement in the following equation

$$S_{ij} = \frac{u_i^1 - u_i^0}{\Delta\xi_j}. \quad (3.8)$$

In this way, a displacement criterion is established. Based on this criterion, the positions at the nodes on imaginary boundaries are determined, and a uniform displacement is applied to each node. Since the displacement criterion is based on a set of nodes on the initial boundary and the imaginary boundary, there is a possibility that the displacement criterion may be discontinuous due to the interaction with order parameters (like polarizations). Therefore, the averaged displacement criterion is calculated by calculating the displacement criterion across the boundary nodes as

$$\overline{S}_{ij} = \frac{\overline{u_i^1} - \overline{u_i^0}}{\Delta\xi_j}. \quad (3.9)$$

This enables the reproduction of the loading conditions of the macroscopic specimen with periodic boundary conditions. Note that a simple elastic analysis was performed in a previous study [30]. In this chapter, I consider the time evolution and thermodynamical equilibrium state of polarization distribution by explicitly taking into account the complicated interactions among the polarization, strain (gradient), and electric field.

For an application of local electric field is based on the previous experimental study [30], in which by placing interfacial electrode between the film and interfaces shown in Figure 3-2(c). The simulation of electric field application in this chapter mimics and reproduces such a

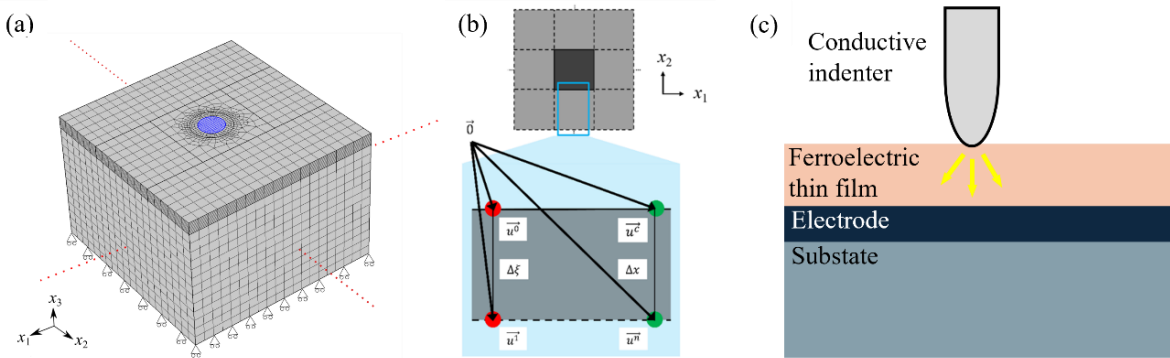


Figure 3-2 (a) The mesh divisions of the simulation model. The blue circle area represents the indentation area. (b) The real boundary and the virtual boundary with the continuous periodic boundary condition. (c) Schematic of experimentally manufactured ferroelectric thin film in previous study[30]. Interfacial electrode is placed between the film and substrate.

practical (experimental) situation, in which on the top of the film, only the contact area with the indenter tip is set as specific value of electric potential, while other area is set as same as bottom interface.

The general-purpose finite element method software COMSOL (ver. 5.4) is used for calculations. To correctly include the flexoelectric effect, which is a function of strain gradient, I used second-order element to represent the strain gradient.

3.3 Results and discussion

3.3.1 Mechanical writing of polar skyrmion

As shown in Figure 3-3(a), at room-temperature (298 K), the initial polarization distribution in PTO thin film is a single domain paralleled to the x_3 -direction ($P_3 = 0.76 \text{ C/m}^2$), which is contributed by the misfit strain-induced out-of-plane easy axis and the lack of depolarization field. Then, local mechanical loads assuming the use of indenter with 10 nm diameter is applied to the thin film up to a maximum of 5 GPa (equivalent to the value in previous experimental study [45]). When a 5 GPa stress load is applied to the center of the thin film, polarizations at the loading area are switched from up to down like Figure 3-3(b). After removing the indenter like Figure 3-3(c), the switched area further forms a stable downward-polarized cylinder domain

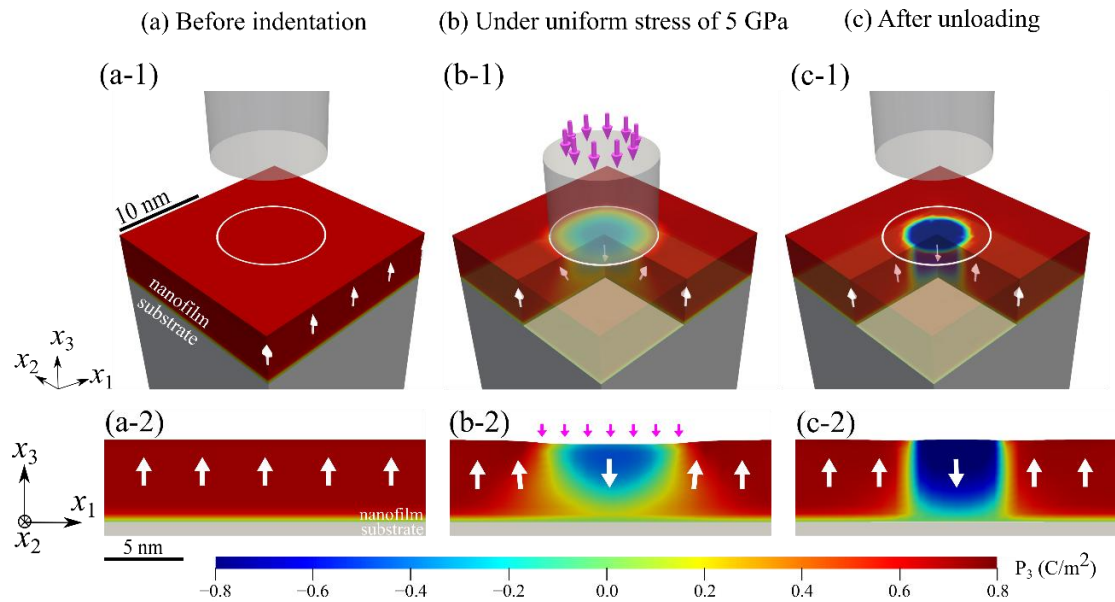


Figure 3-3 Polarization distribution of PbTiO_3 nano-thin film (a) before indentation, (b) under indentation loading, and (c) after unloading. The contour color indicates the value of P_3 , and white arrows indicate the direction of polarization. The white circle and the pink arrows represent the indentation area.

with a diameter of around 6 nm. This process of the mechanical switching of polarization is consistent with the previous experimental observation [45].

The detailed polarization structure of this mechanically switched domain is shown in Figure 3-4. The color of the arrows denotes the in-plane angle θ between polarization and x_1 -axis, and the darkness of the arrows indicates the out-of-plane component of polarization. Therefore, the luminous polarizations in the center are downward ($-x_3$), the peripheral dark polarizations are upward ($+x_3$), and the domain wall between them shows a vortex configuration in the sections parallel to the x_3 -axis. Figure 3-4(b)~(d) present the polarization distributions in cross-sections at $x_3 = 0.5, 2.5$, and 5.0 nm respectively, clearly showing the Neel-type skyrmion configuration at the top, middle, and bottom surfaces of the mechanical switched domain. Such Neel-type configurations have been observed in magnetic skyrmions in lacunar spinel [46]. To further verify the skyrmion structure and its topological feature, the skyrmion number $n_{sk} = 1/4\pi \int \mathbf{P} \cdot (\partial \mathbf{P} / \partial x \times \partial \mathbf{P} / \partial y) dx dy$ (where $\mathbf{P}$ is the unit vector of the polarization) of top, middle, and bottom surfaces are calculated to be -1 (which is the value of skyrmion structure). It is worth mentioning that while previous polar skyrmion observed in PTO/STO superlattice is a hybrid one, with Neel skyrmion on the top and bottom surfaces and Bloch skyrmion at the middle surface, the polar skyrmion in the simple PTO thin film solely comprises Neel skyrmion. In ferromagnetics, there are two competitive energies of spin rotation, the DMI energy and exchange energy, and the competition of them stabilizes the Bloch-type skyrmion with chiral spin rotation [47]. On the other hand, in ferroelectrics, only the gradient energy, which conflicts with chiral rotation, contributes to the spatial change of polarization, and this situation makes only Néel type skyrmions stable, as obtained in the present simulation [48–50]. Note that, the polar skyrmion observed in PTO/STO superlattice [24] is a hybrid type with Néel skyrmion the top and bottom surfaces and Bloch skyrmion at the middle surface. The differences of complex boundary condition between the PTO/STO superlattice and our thin film model may lead the different types of polar skyrmion. Additionally, the distributions of Pontryagin density, which is the integrand in equation of skyrmion number, are shown in Figure 3-4(b-2)~(d-2). The Pontryagin density increases in the regions of significant polarization transition, forming ring-like distributions at $x_3 = 2.5$ and 5.0 nm, and circular distribution at $x_3 = 0.5$ nm. As the comparison, the magnetic skyrmion observed in conventional magnetic materials have uniform distribution⁴¹ of Pontryagin density in the skyrmion because they are formed by the intrinsic DMI acting homogeneously in the material. On the other hand, polar skyrmion consists of nearly uniform domains (with the positive and negative polarizations at the inside and outside of

skyrmion) and continuous polarization rotations among these domains. Only this transition region has “skyrmionic” change in polarization direction, resulting in an inhomogeneous distribution of Pontryagin charge density.

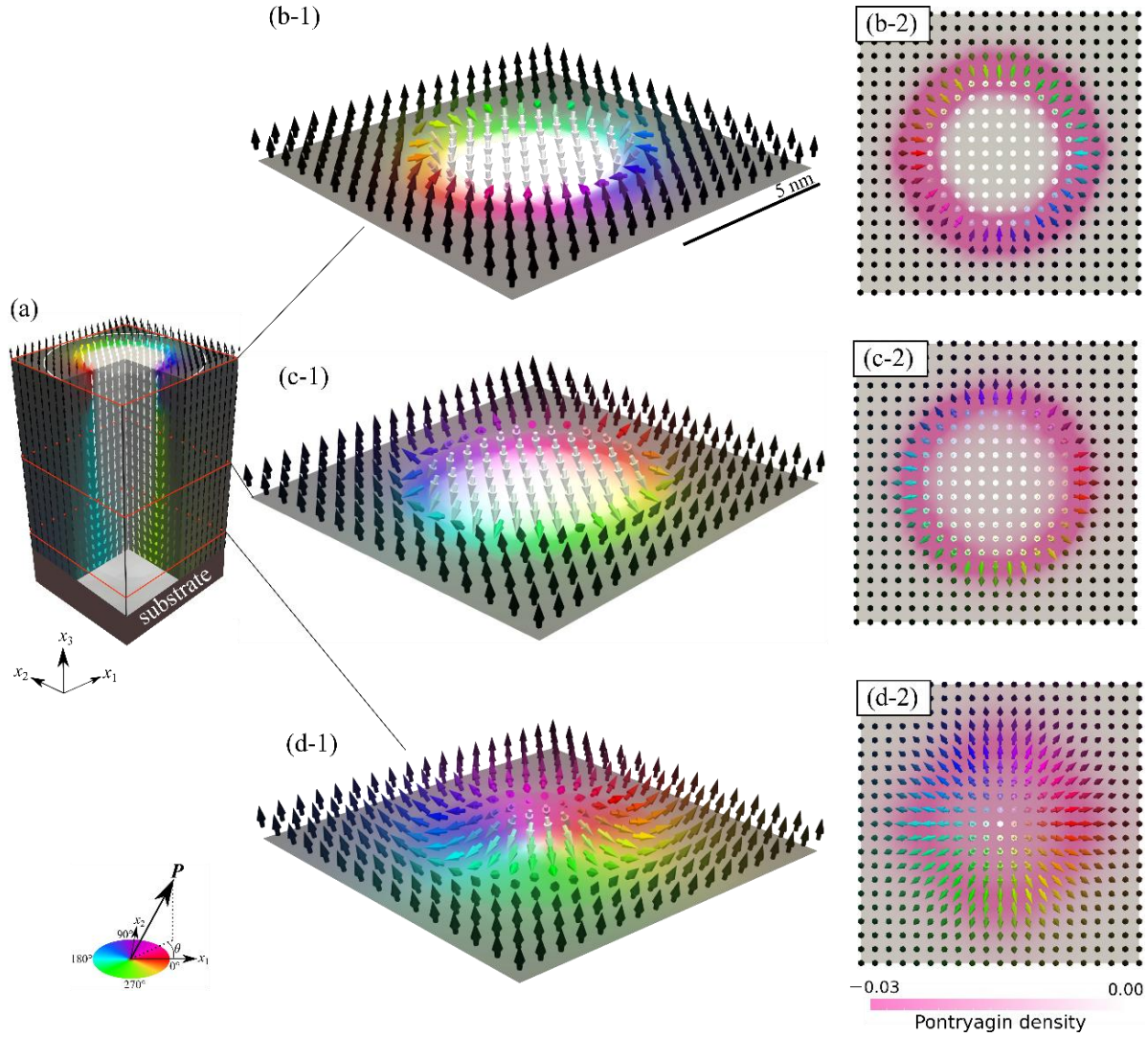


Figure 3-4 Polarization distribution (a) under the indentation area, and its cross section in x_1 - x_2 plane at (b) $x_3 = 5.0$ nm, (c) $x_3 = 2.5$ nm, (d) $x_3 = 0.5$ nm. The color of arrows indicates the in-plane angle θ between direction of polarization and x_1 direction, and the darkness indicates out-of-plane components of polarization. Since polarization is quite small at $x_3 = 0$ nm, while the polarization magnitude is normalized in figure, $x_3 = 0.5$ nm is used as representative of the (approximate) bottom of the polar skyrmion bubble. At any cross section, polarization constitutes a skyrmion structure.

3.3.2 Mechanism of polarization switching

To investigate the formation mechanism of mechanically written skyrmion, the time evolutions of the polarization switching process in the x_1 - x_3 section are shown in Figure 3-5. The loading process is quasi-static due to the nanosecond order of polarization switching. In simulations, I applied stress load to the thin film time by time with the increment of 1 GPa until 5 GPa in the end. In Figure 3-5(a), a homogeneous stress load is gradually applied to the initial single domain. The polarizations have obviously switched at the stress load reaches a critical value of 3 GPa, and maintain the similar polarization configuration until the maximum of 5 GPa. The detailed polarization switching process under critical load of 3 GPa is shown in Figure 3-5(b). At first, the external press load along with the in-plane compressive misfit strain induces a triaxial compressive area beneath the external load, decreasing the polarization magnitude due to the electrostrictive effect. Since the polarization magnitude is almost zero, this change deviates from the usual continuous deformation and can be a topological phase transition.

Subsequently, the boundary of this area exhibits in-plane polarization, and the polarization inside the area initiates switching from upward to downward by some “driving force”. In the end, the area beneath the external load finished the switching and the bottom area near the

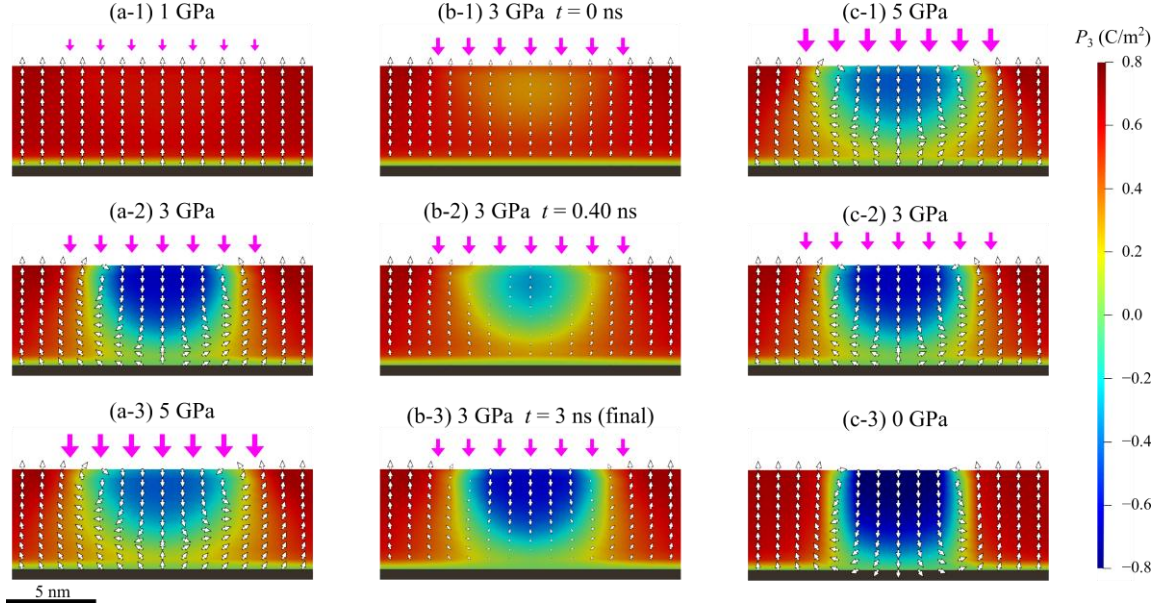


Figure 3-5 The distribution of polarization around the indentation area during the (a) loading process, (b) 3 GPa loading, and (c) unloading process. The contour color indicates the value of P_3 . The pink arrows indicate the indentation area, and the white arrows indicates the direction of polarization. In (a) and (c) white arrows are normalized, and in (b) the size of white arrows represents the magnitude of polarization.

interface of PTO/STO retains upward polarization, indicating that such a “driving force” near the interface area is insufficient. The unloading process, shown in Figure 3-5(c), involves gradual removal of the external stress load, the compressive strain is decreased and the polarization magnitude is increased. As the “driving force” disappears gradually, the head-to-head polarization configuration near the interface becomes unstable, prompting the upward polarization near the interface to switch to the downward and the domain wall forms the vortex configuration to reach the energy stable state. In the end, after the total removal of the indenter, the switching area becomes a polar skyrmion structure as shown in Figure 3-3. Note that all mechanical deformation is assumed to be within the range of elastic deformation and there is no permanent stress after unloading. In addition, since the indentation depth is small, surface effects such as nanohardness [51] are not taken into account.

The stability of polar skyrmion was already discussed in previous studies [24,52]; there is an energy barrier between the topologically nontrivial uniform state and skyrmion state makes the stability of skyrmion, which is called topological protection. Therefore, in this switching process, decreasing of polarization magnitude by electrostrictive effect and driving force from flexoelectric effect break the topological protection, resulting formation of a polar skyrmion. I just mentioned that a “driving force” helps the polarizations switch to a complex way more than just the conventional electrostrictive effect-induced magnitude decrease. What is the true color of this driving force? A mechanical load has two general couplings with polarizations. One is the electrostrictive effect, which provides strain-induced polarization, and the other is the flexoelectric effect, which generates strain gradient-induced polarizations. The magnitude decrease of the polarizations beneath the loading area is contributed by the electrostrictive effect, since the triaxial compressive strain suppresses polarizations here. Meanwhile, the complex polarization switching under the mechanical load should be contributed by the flexoelectric effect, since Lu et al [30] demonstrated that the nano-indenter on the nano-thin film can generate a large local strain gradient field near the loading point. Although the detailed mechanism of the flexoelectric effect in nanoscale ferroelectrics is still not fully understood [53], the strain gradient-induced flexoelectric polarization can be expressed as [54]

$$P_i^{flexo} = \mu_{klij} \frac{\partial \varepsilon_{kl}}{\partial x_j}, \quad (3.10)$$

where μ_{klij} is the flexoelectric coefficient, and $\partial \varepsilon_{kl} / \partial x_j$ is the strain gradient. The in-plane P_1^{flexo} and out-of-plane components P_3^{flexo} of Equation (3.10) are

$$P_1^{flexo} = \mu_{11} \frac{\partial \varepsilon_{11}}{\partial x_1} + \mu_{12} \left(\frac{\partial \varepsilon_{22}}{\partial x_1} + \frac{\partial \varepsilon_{33}}{\partial x_1} \right) + \mu_{44} \left(\frac{\partial \varepsilon_{12}}{\partial x_2} + \frac{\partial \varepsilon_{13}}{\partial x_3} \right), \quad (3.11.1)$$

$$P_3^{flexo} = \mu_{11} \frac{\partial \varepsilon_{33}}{\partial x_3} + \mu_{12} \left(\frac{\partial \varepsilon_{11}}{\partial x_3} + \frac{\partial \varepsilon_{22}}{\partial x_3} \right) + \mu_{44} \left(\frac{\partial \varepsilon_{31}}{\partial x_1} + \frac{\partial \varepsilon_{32}}{\partial x_2} \right). \quad (3.11.2)$$

Figure 3-6 shows the distribution of each term on the right side of Equation (3.11) in the specimen when the external load is 3 GPa (corresponding to Figure 3-5(b)). Firstly, all contributions of P_1^{flexo} are shown in Figure 3-6(a). Under the local stress load, strain ε_{33} has a great difference in x_1 -direction at the boundary of the loading area, which generates the large strain gradient $\partial \varepsilon_{33} / \partial x_1$ at the edge of the loading area. Therefore, the according $\mu_{12} (\partial \varepsilon_{33} / \partial x_1)$ term induced an in-plane flexoelectric polarization P_1 at the boundary of the loading area (shown by white arrows). This result is consistent with the emergence of in-plane polarization at the boundary of the loading area in Figure 3-5(b), which confirms the flexoelectric effect is indeed the “driving force” mentioned in the previous part for the complex

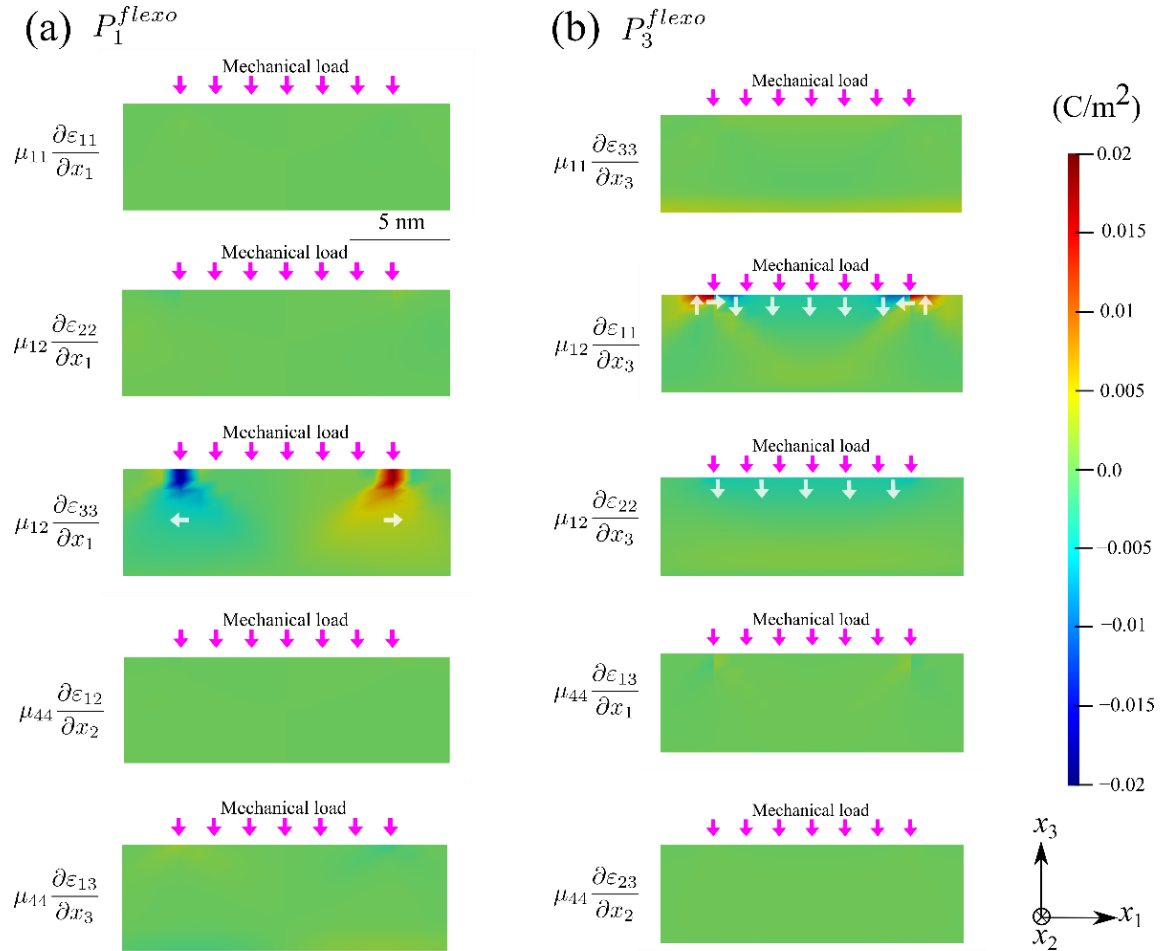


Figure 3-6 The distribution of the components of strain gradient-induced flexoelectric polarization under the indentation area with 3 GPa stress (a) P_1^{flexo} , (b) P_3^{flexo} . The contour color indicates the values of polarization components. The pink arrows indicate the indentation area.

polarization switching. Similarly, flexoelectric contributions of out-of-plane polarization P_3^{flexo} are shown in Figure 3-6(b). The area beneath the external load has an obvious strain gradient $\partial\epsilon_{11}/\partial x_3$ and $\partial\epsilon_{22}/\partial x_3$, which provides the driving force for the downward polarization $-P_3$ by the flexoelectric term of $\mu_{12}(\partial\epsilon_{11}/\partial x_3 + \partial\epsilon_{22}/\partial x_3)$ of Equation (3.11.2). It is consistent with the upward to downward polarization switching process in Figure 3-5(b). This driving force (shown in the blue area in Figure 3-6(b)) is large and the polarization magnitude is small near the loading surface, therefore the polarization here easily switched to downward but the polarization at the bottom area (near the PTO/STO interface) is still upward as initial. Further, the term $\mu_{12}(\partial\epsilon_{11}/\partial x_3 + \partial\epsilon_{22}/\partial x_3)$ even provide a driving force for the upward out-of-plane polarization $+P_3$ near the interface and the outside of the loading area (red area in Figure 3-6(b)), therefore the upward polarization and the head-to-head polarization configuration near the interface in Figure 3-5(b) are contributed by this driving force. In summary, the flexoelectric effect contributes to the complex polarization switching process under an external mechanical load, providing a non-collinear interaction within the simple ferroelectric system, which acts as a driving force for the emergence of polar skyrmion. In addition, the free energy calculation indicates that flexoelectric driving force alone cannot exceed the energy barrier for polarization reversal. However, under triaxial compression, it can exceed the barrier and polarization switching occurs. Therefore, for polarization switching, both reduction of energy barrier by compressive strain and flexoelectric driving field are necessary.

3.3.3 Electric erasing of polar skyrmion

As shown in Figure 3-7, a local electric field is applied to the thin film model containing a polar skyrmion by contacting the conductive tip to the same area where the indentation load was applied. The local electric field is reproduced by setting the electric potential at the interface to 0 V and at the contact area to -3 V (below the magnitude in the previous experimental study [45]). Assuming that thin film is under short-circuit conditions, the electric potential at the film surface excluding the contact area remains at 0 V. Under an electric field, polarization under the contact area reorients to $+x_3$ direction shown in Figure 3-7(b). Subsequent removal of the conductive tip results in a complete restoration of the polarization distribution to its pre-indentation state, as shown in Figure 3-7(c). In this process, the skyrmion number n_{sk} under the tip changes from -1 to 0 for any x_1 - x_2 section. Thus, this result suggests that a mechanically written polar skyrmion can be electrically erased.

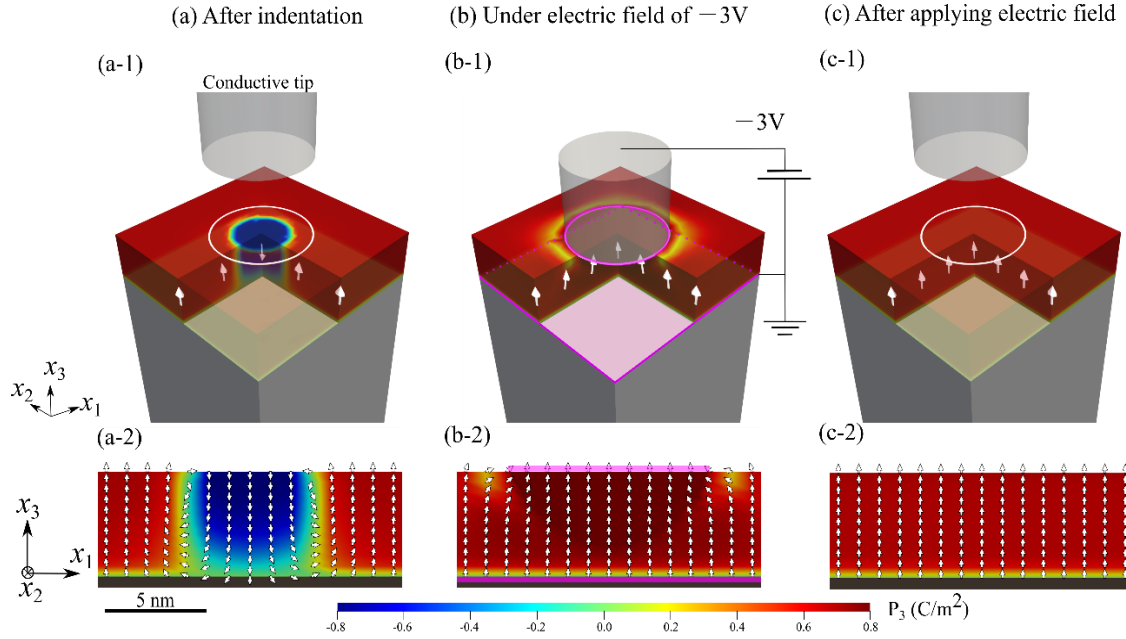


Figure 3-7 Polarization distribution of PbTiO_3 nano-thin film (a) after indentation, (b) under an electric field, (c) after applying electric field.

3.3.4 Mechanical and electrical control of polar topological structures

Since the mechanical load can trigger polarization by switching by the flexoelectric effect and leave a stable topological structure (like polar skyrmions) in the thin film, I anticipate that the arbitrary patterns or more complex polar topologies can be mechanically written by the indenter. In Figure 3-8(a), I let the indenter press the thin film and then move it along the x_2 -direction under a constant 5 GPa stress in the simulation model. As the indenter traversed, polarizations continuously switched, forming a stable pattern of trajectory, upon its removal. The width of this stripe domain is 6 ~ 7 nm, which consistent with the value of previous study [55]. Furthermore, when the indenter traced a circular path on the thin film, a donut-like pattern is created as shown in Figure 3-8(b). By calculating its skyrmion number, I identified this pattern as a polar “skyrmionium”. Skyrmionium is also called “ 2π -skyrmions” [56], in which a small skyrmion ($n_{sk} = +1$) inside a bigger opposite skyrmion ($n_{sk} = -1$). Our simulation demonstrated writing several polar topologies such as skyrmion point, polar helical line, and skyrmionium pattern on room-temperature PTO thin film. Then, by using the same indenter, I conducted the electrical “erase” of the topological pattern. In Figure 3-8(c), by contacting the conducted indenter at the right part of the polar skyrmionium and applying a voltage of -3 V between the indenter and substrate, the local polarization is reoriented to $+x_3$ -direction. Consequently, after removing the conducted indenter, the polar skyrmionium exhibits a gap, while the rest of the

pattern remains intact. These polar topologies are thermodynamically stable and maintain their shapes.

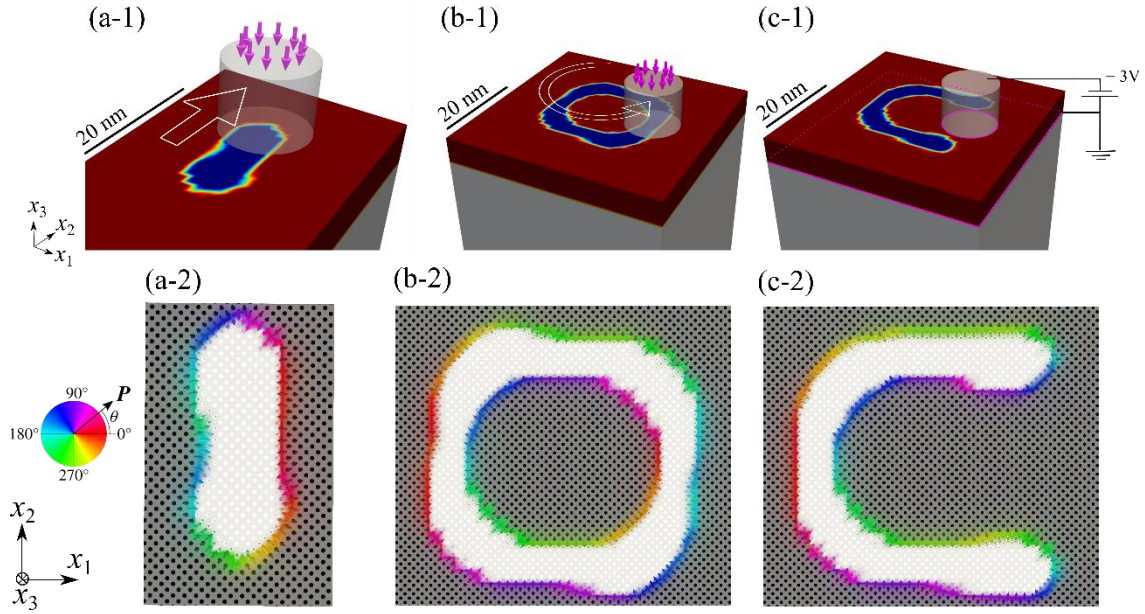


Figure 3-8 Polarization distribution when indenter is moved (a) linearly in x_2 direction, (b) circularly, and (c) when part of the circle is erased by electric field. Skyrmion number of these structures are calculated as (a) $n_{sk} = -1$, (b) $n_{sk} = -1 + 1$ (skyrmionium), and (c) $n_{sk} = -1$.

3.3.5 Indentation into in-plane 90-degree polarization domain

In previous sections, skyrmion is formed by indentation into the thin film with uniform upward polarization. Using different background polarization, other polar topologies such as meron are likely created. In addition to skyrmion ($n_{sk} = \pm 1$), meron ($n_{sk} = \pm 1/2$) is another typical quasi-two-dimensional topological structure. In contrast to skyrmion reversing vectors at the periphery and inside, meron has in-plane vector at the periphery and the out-of-plane components rotate 90 degrees from the center. Considering this, stable in-plane domains are necessary for the creation of polar melons. In previous study [22], Wang et.al showed that in PTO thin film epitaxially grown on SmScO₃(SSO) substrate, in-plane and out-of-plane polarization domains coexist due to appropriate misfit strain. Furthermore, where polarization directions of adjacent in-plane domains are opposite to each other, polarization indicates out-of-plane direction at junction, forming quasi-two-dimensional topological structures such as merons and antimerons. Therefore, generating adjacent domains in which in-plane polarization faces each other may create polar meron or antimeron. In the previous sections, I revealed that

the mechanically switched region in PTO nano thin film has continuous transition of polarization between outside and inside of indentation area via flexoelectric effect. In other words, Indentation load can induce in-plane domain in PTO thin film. Based on these, I calculate polarization distribution during indentation process on PTO thin film with in-plane 90-degree polarization domain. Figure 3-9 shows the polarization distribution of surface of PTO

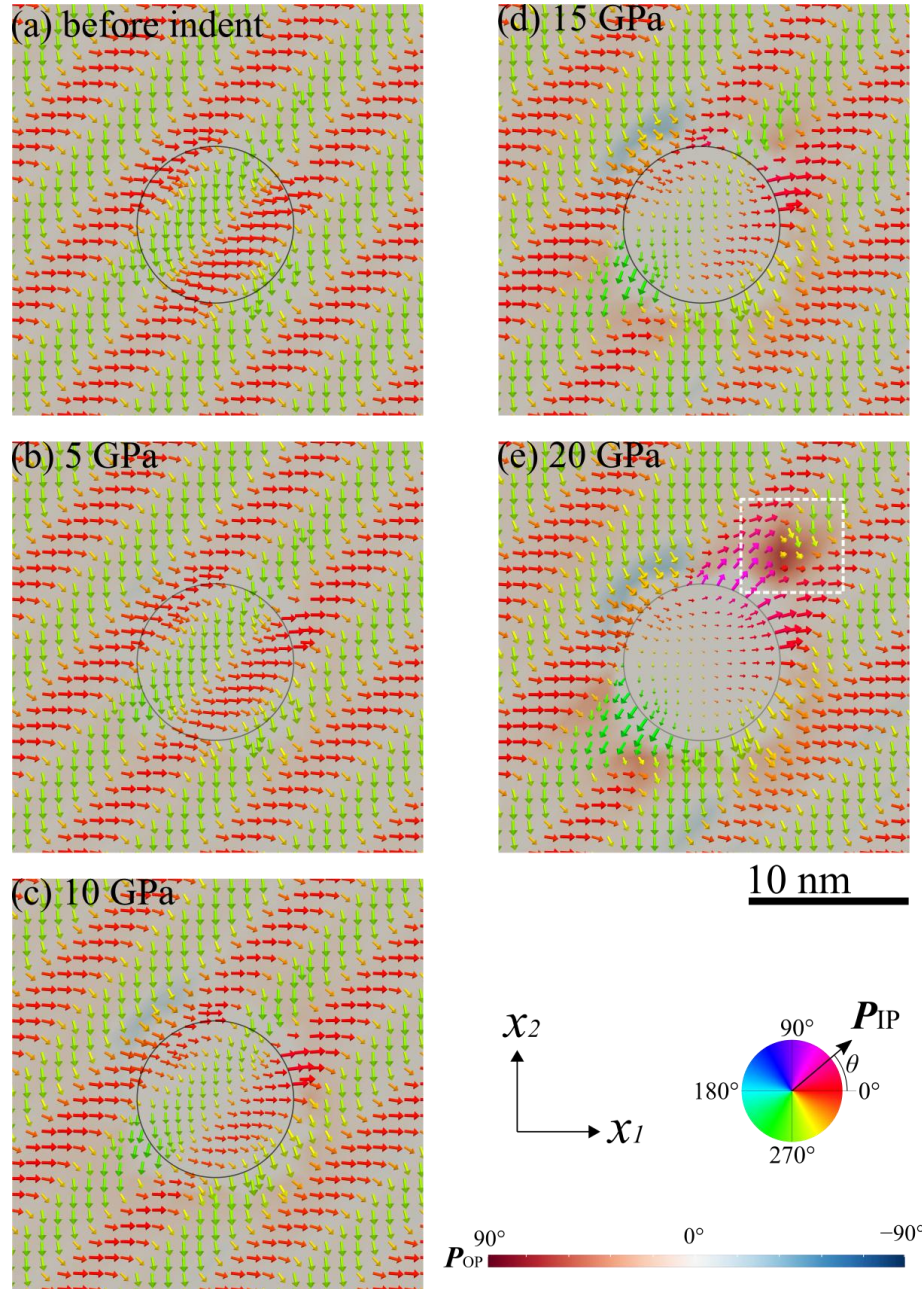


Figure 3-9 The polarization distribution during the indentation into PbTiO_3 thin film with in-plane polarization. The arrow indicates the in-plane direction and magnitude of polarization. The contour color indicates the out-of-plane direction of polarization.

nano film near the indentation area. The arrows indicate the direction and magnitude of polarization. As shown in Figure 3-9, when no load is applied, only in-plane polarization exists with two types of domains of positive x_1 or negative x_2 direction. Between these domains, polarization direction changes continuously, forming 90-degree domain walls. When indentation load is applied to this model and reached 20 GPa, polarization distribution changed as shown in Figure 3-9 (e). Compared to the that before indentation, the magnitude of polarization just below the load region is decreased. More importantly, new radial direction polarization appears at the outer periphery of the indentation region. This is considered to be induced by the strain gradient and flexoelectric effect. Furthermore, in the area indicated in white dash line, new in-plane polarization direction and existing domain face each other, and polarization of positive x_3 direction appears. Therefore, new in-plane and out-of-plane polarization can be created by indentation load into a PTO nano thin film.

Figure 3-10(a) shows the polarization distribution and domain configuration at 20 GPa in the dotted box in Figure 3-9(e). The out-of-plane domain in the center is surrounded by in-plane directional domains. The out-of-plane directional domain in the center is surrounded by four in-plane directional domains. Two of the four in-plane domains are oriented in same direction, and the other two are oriented in opposite directions. This feature is consistent with the structure of MAC (meron-antimeron-combination), one of the two-dimensional topological orders [22]. Actually, this domain structure is consistent with that shown in o Extended Data Fig. 5(e) in the previous study [22]. Furthermore, the polarization distribution has continuous transitions and different out-of-plane components in the central and peripheral regions, suggesting the formation of a quasi-two-dimensional polar topology. To investigate the topological properties

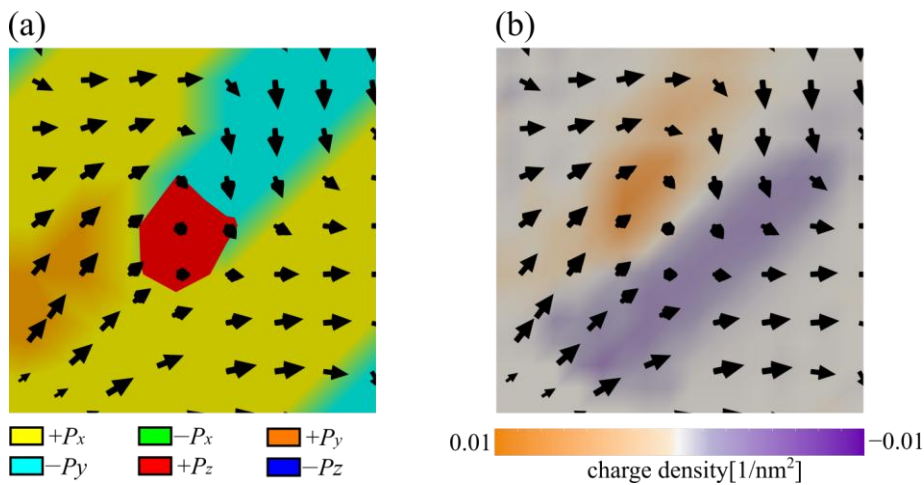


Figure 3-10 (a)The polarization domain structure of MAC.

(b)Topological charge density distribution of MAC.

of this polarization distribution in detail, I calculate skyrmion number and find it to be 0. Since a MAC is a quasi-two-dimensional topological order consisting of overlapping meron ($n_{sk} = 1/2$) and antimeron ($n_{sk} = -1/2$) the topological charge of MAC is the sum, $n_{sk} = 0$. Furthermore, evaluating pontyagin charge density, as shown in Figure 3-10(b), we can see that the polarization distribution is divided into two regions, one with positive value of charge density and the other with negative values, indicating that this polarization is MAC. This result demonstrates that indentation loading into PbTiO₃ nanofilms can create different types of quasi-two-dimensional topological polar structure, MAC.

3.4 Conclusion

In summary, our simulations demonstrate that polar topologies including skyrmion and skyrmionium can be manipulated using simple methods: mechanical writing and electrical erasing. Since PbTiO₃ is prototype of ferroelectric perovskite oxides, our approach of writing and erasing polar topologies can be effective for other ferroelectrics in general. While several studies reported the stabilization of polar topologies by the intrinsic ferroelectric DMI [16,17] and DMI-like flexoelectric effect [32,33], my simulations demonstrated writing of polar skyrmions in arbitral positions by using flexoelectric effect as extrinsic driving force. In addition, a previous study³⁹ has read nanoscale skyrmions using PFM, suggesting that the various topological orders shown in this chapter are equally readable. The inherent advantages of these polar topologies over magnetic counterparts are smaller scale, field independence, and existence at room-temperature, positioning them as promising candidates for ultra-high density memory devices. Furthermore, the major advantage of this method is that it allows for local manipulation of individual topologies. Combined with previous work [57] that shows the manipulation of global topological pattern, we can manipulate polar topologies over wide scale range. Therefore, our findings of the ability to easily write and erase various polar topologies will drive their potential for applications. Furthermore, our simulations not only demonstrate the efficacy of the strain (gradient) field driving for the local polarization switching via the flexoelectric effect as a non-collinear interaction, but also suggest the feasibility of the mechanical design of functional topologies in simple ferroelectric systems. I envisage that these findings will inform future research focused on the manipulation of polar topologies by mechanical fields or other physics fields with topological nature.

References

- [1] A. Bogdanov and A. Hubert, Thermodynamically stable magnetic vortex states in magnetic crystals, *Journal of Magnetism and Magnetic Materials* **138**, 255 (1994).
- [2] S. Mühlbauer, B. Binz, F. Jonietz, C. Pfleiderer, A. Rosch, A. Neubauer, R. Georgii, and P. Böni, Skyrmion Lattice in a Chiral Magnet, *Science* **323**, 915 (2009).
- [3] J. Zang, M. Mostovoy, J. H. Han, and N. Nagaosa, Dynamics of Skyrmion Crystals in Metallic Thin Films, *Phys. Rev. Lett.* **107**, 136804 (2011).
- [4] N. Nagaosa and Y. Tokura, Topological properties and dynamics of magnetic skyrmions, *Nature Nanotech* **8**, 899 (2013).
- [5] J. Seidel, R. K. Vasudevan, and N. Valanoor, Topological Structures in Multiferroics – Domain Walls, Skyrmions and Vortices, *Advanced Electronic Materials* **2**, 1500292 (2016).
- [6] S. S. P. Parkin, M. Hayashi, and L. Thomas, Magnetic Domain-Wall Racetrack Memory, *Science* **320**, 190 (2008).
- [7] Y. Liu, J. Liu, H. Pan, X. Cheng, Z. Hong, B. Xu, L.-Q. Chen, C.-W. Nan, and Y.-H. Lin, Phase-Field Simulations of Tunable Polar Topologies in Lead-Free Ferroelectric/Paraelectric Multilayers with Ultrahigh Energy-Storage Performance, *Advanced Materials* **34**, 2108772 (2022).
- [8] I. Dzyaloshinsky, A thermodynamic theory of “weak” ferromagnetism of antiferromagnetics, *Journal of Physics and Chemistry of Solids* **4**, 241 (1958).
- [9] T. Moriya, Anisotropic Superexchange Interaction and Weak Ferromagnetism, *Phys. Rev.* **120**, 91 (1960).
- [10] X. Z. Yu, W. Koshibae, Y. Tokunaga, K. Shibata, Y. Taguchi, N. Nagaosa, and Y. Tokura, Transformation between meron and skyrmion topological spin textures in a chiral magnet, *Nature* **564**, 95 (2018).
- [11] A. Petković and M. V. Milovanović, Fractionalization into Merons in Quantum Dots, *Phys. Rev. Lett.* **98**, 066808 (2007).
- [12] X. Z. Yu, Y. Onose, N. Kanazawa, J. H. Park, J. H. Han, Y. Matsui, N. Nagaosa, and Y. Tokura, Real-space observation of a two-dimensional skyrmion crystal, *Nature* **465**, 901 (2010).
- [13] L. Van Lich, T. Shimada, K. Nagano, Y. Hongjun, J. Wang, K. Huang, and T. Kitamura, Anomalous toughening in nanoscale ferroelectrics with polarization vortices, *Acta Materialia* **88**, 147 (2015).

- [14] Z. Hong et al., Stability of Polar Vortex Lattice in Ferroelectric Superlattices, *Nano Lett.* **17**, 2246 (2017).
- [15] S.-L. Hsu, M. R. McCarter, C. Dai, Z. Hong, L.-Q. Chen, C. T. Nelson, L. W. Martin, and R. Ramesh, Emergence of the Vortex State in Confined Ferroelectric Heterostructures, *Advanced Materials* **31**, 1901014 (2019).
- [16] H. J. Zhao, P. Chen, S. Prosandeev, S. Artyukhin, and L. Bellaiche, Dzyaloshinskii–Moriya-like interaction in ferroelectrics and antiferroelectrics, *Nat. Mater.* **20**, 341 (2021).
- [17] P. Chen, Microscopic origin of the electric Dzyaloshinskii-Moriya interaction, *Physical Review B* **106**, 224101 (2022).
- [18] X. Wang, F.-T. Huang, J. Yang, Y. S. Oh, and S.-W. Cheong, Interlocked chiral/polar domain walls and large optical rotation in Ni_3TeO_6 , *APL Materials* **3**, 076105 (2015).
- [19] Z. Jiang and J. Wang, Stability of chiral polarization vortex in strained ferroelectric superlattices, *Journal of Applied Physics* **131**, 164101 (2022).
- [20] M. A. Pereira Gonçalves, C. Escorihuela-Sayalero, P. Garca-Fernández, J. Junquera, and J. Íñiguez, Theoretical guidelines to create and tune electric skyrmion bubbles, *Sci. Adv.* **5**, eaau7023 (2019).
- [21] T. Shimada, L. V. Lich, K. Nagano, J.-S. Wang, J. Wang, and T. Kitamura, Polar Superhelices in Ferroelectric Chiral Nanosprings, *Sci Rep* **6**, 1 (2016).
- [22] Y. J. Wang et al., Polar meron lattice in strained oxide ferroelectrics, *Nat. Mater.* **19**, 881 (2020).
- [23] Y.-T. Shao et al., Emergent chirality in a polar meron to skyrmion phase transition., *Nat Commun* **14**, 1355 (2023).
- [24] S. Das et al., Observation of room-temperature polar skyrmions, *Nature* **568**, 368 (2019).
- [25] Z. Wang and L.-Q. Chen, Reversible Phase Transition between Vortex Lattice and Hexagonal Polar Skyrmion Crystals, *Nano Lett.* **23**, 9907 (2023).
- [26] Y. L. Tang et al., Observation of a periodic array of flux-closure quadrants in strained ferroelectric PbTiO_3 films, *Science* **348**, 547 (2015).
- [27] A. K. Yadav et al., Observation of polar vortices in oxide superlattices, *Nature* **530**, 7589 (2016).
- [28] P. Shafer et al., Emergent chirality in the electric polarization texture of titanate superlattices, *Proceedings of the National Academy of Sciences* **115**, 915 (2018).
- [29] S. Das et al., Observation of room-temperature polar skyrmions, *Nature* **568**, 368 (2019).

- [30] H. Lu, C.-W. Bark, D. Esque de los Ojos, J. Alcala, C. B. Eom, G. Catalan, and A. Gruverman, Mechanical Writing of Ferroelectric Polarization, *Science* **336**, 59 (2012).
- [31] T. Shimada, Y. Wang, T. Hamaguchi, K. Kasai, K. Masuda, L. Van Lich, T. Xu, J. Wang, and H. Hirakata, Emergence of non-trivial polar topologies hidden in singular stress field in SrTiO_3 : topological strain-field engineering, *J. Phys.: Condens. Matter* **33**, 505301 (2021).
- [32] A. N. Morozovska, R. Hertel, S. Cherifi-Hertel, V. Yu. Reshetnyak, E. A. Eliseev, and D. R. Evans, Chiral polarization textures induced by the flexoelectric effect in ferroelectric nanocylinders, *Phys. Rev. B* **104**, 054118 (2021).
- [33] A. N. Morozovska, E. A. Eliseev, O. A. Kovalenko, and D. R. Evans, Influence of chemical strains on the electrocaloric response, polarization morphology, tetragonality, and negative-capacitance effect of ferroelectric core-shell nanorods and nanowires, *Phys. Rev. Applied* **21**, 054035 (2024).
- [34] G. Sheng, Y. L. Li, J. X. Zhang, S. Choudhury, Q. X. Jia, V. Gopalan, D. G. Schlom, Z. K. Liu, and L. Q. Chen, A modified Landau-Devonshire thermodynamic potential for strontium titanate, *Applied Physics Letters* **96**, 108 (2010).
- [35] C. Liu, J. Wang, G. Xu, M. Kamlah, and T. Y. Zhang, An isogeometric approach to flexoelectric effect in ferroelectric materials, *International Journal of Solids and Structures* **162**, 198 (2019).
- [36] W. Ma, A study of flexoelectric coupling associated internal electric field and stress in thin film ferroelectrics, *Physica Status Solidi (B) Basic Research* **245**, 761 (2008).
- [37] A. N. Morozovska, E. A. Eliseev, S. L. Bravina, A. Y. Borisevich, and S. V. Kalinin, Roto-flexoelectric coupling impact on the phase diagrams and pyroelectricity of thin SrTiO_3 films, *Journal of Applied Physics* **112**, (2012).
- [38] E. A. Eliseev, A. N. Morozovska, M. D. Glinchuk, and R. Blinc, Spontaneous flexoelectric/flexomagnetic effect in nanoferroics, *Physical Review B - Condensed Matter and Materials Physics* **79**, 1 (2009).
- [39] Y. Gu, Z. Hong, J. Britson, and L.-Q. Chen, Nanoscale mechanical switching of ferroelectric polarization via flexoelectricity, *Appl. Phys. Lett.* **106**, 022904 (2015).
- [40] Y. J. Wang et al., Polar meron lattice in strained oxide ferroelectrics, *Nature Materials* **19**, 881 (2020).
- [41] Y. L. Li, S. Y. Hu, Z. K. Liu, and L. Q. Chen, Effect of substrate constraint on the stability and evolution of ferroelectric domain structures in thin films, *Acta Materialia* **50**, 395 (2002).
- [42] S. Das et al., Observation of room-temperature polar skyrmions, *Nature* **568**, 368 (2019).

- [43] L. Jiang, J. Tang, Y. Zhou, Q. Yang, Y. Zhang, L. Guo, and X. Zhong, Simulations of local-mechanical-stress-induced ferroelectric polarization switching by a multi-field coupling model of flexoelectric effect, *Computational Materials Science* **108**, 309 (2015).
- [44] S. Li, Y. J. Wang, Y. L. Zhu, Y. L. Tang, Y. Liu, J. Y. Ma, M. J. Han, B. Wu, and X. L. Ma, Evolution of flux-closure domain arrays in oxide multilayers with misfit strain, *Acta Materialia* **171**, 176 (2019).
- [45] H. Lu, C.-W. Bark, D. Esque de los Ojos, J. Alcala, C. B. Eom, G. Catalan, and A. Gruverman, Mechanical Writing of Ferroelectric Polarization, *Science* **336**, 59 (2012).
- [46] I. Kézsmárki et al., Néel-type skyrmion lattice with confined orientation in the polar magnetic semiconductor GaV_4S_8 , *Nature Mater* **14**, 1116 (2015).
- [47] B. Göbel, I. Mertig, and O. A. Tretiakov, Beyond skyrmions: Review and perspectives of alternative magnetic quasiparticles, *Physics Reports* **895**, 1 (2021).
- [48] L. Han et al., High-density switchable skyrmion-like polar nanodomains integrated on silicon, *Nature* **603**, 7899 (2022).
- [49] S. Yuan, Z. Chen, S. Prokhorenko, Y. Nahas, L. Bellaiche, C. Liu, B. Xu, and L. Chen, Hexagonal Close-packed Skyrmion Lattice in Ultrathin Ferroelectric PbTiO_3 Films, *Physical Review Letters* **130**, 226801 (2023).
- [50] L. Desplat, J.-V. Kim, and R. L. Stamps, Paths to annihilation of first- and second-order (anti)skyrmions via (anti)meron nucleation on the frustrated square lattice, *Phys. Rev. B* **99**, 174409 (2019).
- [51] T.-Y. Zhang and W.-H. Xu, Surface Effects on Nanoindentation, *J. Mater. Res.* **17**, 1715 (2002).
- [52] Y. Zhang, Q. Li, H. Huang, J. Hong, and X. Wang, Strain manipulation of ferroelectric skyrmion bubbles in a freestanding PbTiO_3 film: A phase field simulation, *Phys. Rev. B* **105**, 224101 (2022).
- [53] W. Zhou, P. Chen, and B. Chu, Flexoelectricity in ferroelectric materials, *IET Nanodielectrics* **2**, 83 (2019).
- [54] P. V. Yudin and A. K. Tagantsev, Fundamentals of flexoelectricity in solids, *Nanotechnology* **24**, 432001 (2013).
- [55] R. Takahashi, Ø. Dahl, E. Eberg, J. K. Grepstad, and T. Tybell, Ferroelectric stripe domains in PbTiO_3 thin films: Depolarization field and domain randomness, *Journal of Applied Physics* **104**, 064109 (2008).

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- [56] S. Zhang, F. Kronast, G. Van Der Laan, and T. Hesjedal, Real-Space Observation of Skyrmionium in a Ferromagnet-Magnetic Topological Insulator Heterostructure, *Nano Lett.* **18**, 1057 (2018).
- [57] T. Xu, C. Wu, S. Zheng, Y. Wang, J. Wang, H. Hirakata, T. Kitamura, and T. Shimada, Mechanical Rippling for Diverse Ferroelectric Topologies in Otherwise Nonferroelectric SrTiO₃ Nanofilms, *Phys. Rev. Lett.* **132**, 086801 (2024).

Chapter 4 Emergence of collective polar skyrmion lattice and related topological phases via strain engineering

4.1 Introduction

In Chapter 3, the mechanical generation of isolated quasi-two-dimensional topological structures including skyrmion were achieved. This chapter focuses on methods for forming condensed skyrmions and skyrmion lattices.

Ferroelectrics have been applied to non-volatile memory, sensors, actuators, and other devices [1–5] due to their mechanical and electrical response properties, which arise from internal nanoscale polarization orders. These orders are characterized by topology, meaning that orders with different topologies have different functions [6–8]. For this reason, substantial research efforts have been directed toward exploring, creating, and controlling topological polarization orders. Among these, polar skyrmions [9,10], recently discovered as a novel topological polarization order, have drawn considerable attention due to their distinctive properties and potential applications [11–14]. Possessing both deformability and stability due to topological protection, skyrmions behave like single particles, even being an order of polarization vectors. This quasiparticle-like feature distinguishes them from conventional polarization orders. Previously, skyrmions have been studied as a magnetic moment order in the field of magnetic materials [15–18]. The unique properties of magnetic skyrmions, such as emergent electromagnetic fields and topological Hall effects have been clarified, and their potential applications have been proposed for racetrack memory [19], artificial intelligence devices [20,21], and others [22]. Current research on polar skyrmions has revealed their unique physical properties and potential applications, including local negative permittivity in response to an electric field [11], writability by twisted light [12], and ultrahigh-density arrangement for non-volatile memory [13]. Furthermore, polar skyrmions exist below the critical size of polarization in typical ferroelectrics [14], indicating their potential application for nanodevices.

Nevertheless, creating polar skyrmions remains challenging. Their stabilization relies on intricate factors, including electrical boundary conditions, in-plane strain conditions, and

polarization gradient energy effects [10,23], whereas magnetic skyrmions are stabilized by Dzyaloshinskii–Moriya interaction (DMI) [24,25] and have been observed in many cases. For this reason, there are still very few observations of polar skyrmions, and most of them are limited to ferroelectric superlattice systems [14,26,27], where polar skyrmions were first discovered [10]. Therefore, establishing reliable stabilization conditions and underlying the stabilization mechanism is essential for advancing polar skyrmion research and applications.

Previous studies investigating stabilizing conditions for polar skyrmions have highlighted the importance of in-plane strain. Cheng et al. [28] investigated the stable state depending on the in-plane strain condition in PbTiO₃/SrTiO₃(PTO/STO) superlattices and demonstrated that skyrmions are stable under uniaxial compressive strain of 1%. Zhang et al. [29] predicted that a transition from the skyrmion phase to the stripe phase is possible by continuous uniaxial tensile strain loading in free-standing PbTiO₃ thin films. In addition, Shao et al. [27] experimentally observed the transition between the skyrmion phase and the non-skyrmion phase in PTO/STO superlattices by varying the temperature conditions, suggesting that in-plane strain conditions depending on temperature are significant for the formations of skyrmions. These findings suggest that in-plane strain is the key to understanding the formation mechanism of polar skyrmions. However, the formation process of polar skyrmions, despite its importance for elucidating the fundamental mechanism, remains elusive.

In this chapter, I demonstrate the formation process of polar skyrmions and the topological transition in SrTiO₃/PbTiO₃ superlattices under different in-plane strain conditions using phase-field simulations. Furthermore, I clarify that in-plane strain is a critical factor governing the formation process of topological polarization orders.

4.2 Simulation method

4.2.1 Phase-field modeling

The phase-field simulations in this chapter consist of three governing equations: time-dependent Ginzburg-Landau (TDGL), mechanical equilibrium, and Maxwell's equations, i.e.,

$$\frac{\partial P_i}{\partial t} = -L \frac{\delta F}{\delta P_i}, \quad (4.1)$$

$$\sigma_{ij,j} = \left(\frac{\partial f}{\partial \varepsilon_{ij}} \right)_{,j} = 0, \quad (4.2)$$

$$D_{i,i} = \left(-\frac{\partial f}{\partial E_i} \right)_{,i} = 0, \quad (4.3)$$

where P_i is polarizations, t is time, L is the kinetic coefficient, $F = \int f dV$ is total free energy, in which f is total free energy density, $\sigma_{ij,j}$ is the stress gradient, ε_{ij} is strain, $D_{i,i}$ is the electric displacement gradient, and E_i is the electric field in the material. The total free energy of a ferroelectric system consists of Landau energy, gradient energy, electrostatic energy, and elastic energy. The total free energy density f is expressed as

$$f = f_{\text{Landau}} + f_{\text{grad}} + f_{\text{electo}} + f_{\text{elast}}. \quad (4.4)$$

The Landau energy density, which describes the temperature-dependent ferroelectric phase transition, is written as [30,31]

$$\begin{aligned} f_{\text{Landau}} &= \alpha_{ij}P_iP_j + \alpha_{ijkl}P_iP_jP_kP_l + \alpha_{ijklmn}P_iP_jP_kP_lP_mP_n \\ &= \alpha_1(P_1^2 + P_2^2 + P_3^2) + \alpha_{11}(P_1^4 + P_2^4 + P_3^4) + \alpha_{12}(P_1^2P_2^2 + \\ &\quad P_1^2P_3^2 + P_2^2P_3^2) + \alpha_{111}(P_1^6 + P_2^6 + P_3^6) + \alpha_{112}(P_1^4(P_2^2 + P_3^2) + \\ &\quad P_2^4(P_1^2 + P_3^2) + P_3^4(P_1^2 + P_2^2)) + \alpha_{123}(P_1^2P_2^2P_3^2), \end{aligned} \quad (4.5)$$

where α_{ij} , α_{ijkl} , and α_{ijklmn} are the Landau expansion coefficients. The gradient energy density, which conducts the formation of the polarization domains, is written as [32]

$$\begin{aligned} f_{\text{grad}} &= \frac{1}{2}G_{ijkl}P_{i,j}P_{k,l} \\ &= \frac{1}{2}G_{11}(P_{1,1}^2 + P_{2,2}^2 + P_{3,3}^2) + G_{12}(P_{1,1}P_{2,2} + P_{2,2}P_{3,3} + P_{3,3}P_{1,1}) + \\ &\quad \frac{1}{2}G_{44}[(P_{1,2} + P_{2,1})^2 + (P_{2,3} + P_{3,2})^2 + (P_{1,3} + P_{3,1})^2], \end{aligned} \quad (4.6)$$

where G_{ijkl} is the gradient energy coefficient. The electrostatic energy density, which describes the effects of electric fields on polarization, is written as

$$\begin{aligned} f_{\text{electo}} &= -\frac{1}{2}\kappa_0 E_i E_i - E_i P_i \\ &= -\frac{1}{2}\kappa_0(E_1^2 + E_2^2 + E_3^2) - E_1 P_1 - E_2 P_2 - E_3 P_3, \end{aligned} \quad (4.7)$$

where κ_0 is the relative permittivity of materials. The elastic strain energy density is written as

$$f_{\text{elast}} = \frac{1}{2}c_{ijkl}\varepsilon_{ij}^{\text{elast}}\varepsilon_{kl}^{\text{elast}}, \quad (4.8)$$

where c_{ijkl} is the elastic constant and $\varepsilon_{ij}^{\text{elast}}$ is elastic strain. In addition to elastic strain, ferroelectric materials exhibit eigen strain caused by spontaneous polarization. The eigen strain ε_{ij}^0 can be written as [31]

$$\varepsilon_{ij}^0 = Q_{ijkl}P_kP_l. \quad (4.9)$$

where Q_{ijkl} is electrostrictive coefficients. The total strain ε_{ij} , defined as the displacement gradient in the material, is the sum of elastic strain and eigen strain, which can be written as

$$\varepsilon_{ij} = \varepsilon_{ij}^{\text{elast}} + \varepsilon_{ij}^0. \quad (4.10)$$

Substituting Eq. (4.10), f_{elast} is rewritten as

$$\begin{aligned} f_{\text{elast}} &= \frac{1}{2} c_{ijkl} \varepsilon_{ij}^{\text{elast}} \varepsilon_{kl}^{\text{elast}} \\ &= \frac{1}{2} c_{ijkl} (\varepsilon_{ij} - \varepsilon_{ij}^0) (\varepsilon_{kl} - \varepsilon_{kl}^0) \\ &= \frac{1}{2} c_{ijkl} \varepsilon_{ij} \varepsilon_{kl} - \frac{1}{2} c_{ijkl} (\varepsilon_{ij} \varepsilon_{kl}^0 + \varepsilon_{ij}^0 \varepsilon_{kl}) + \frac{1}{2} c_{ijkl} \varepsilon_{ij}^0 \varepsilon_{kl}^0. \end{aligned} \quad (4.11)$$

Substituting Eq. (4.9) into the second term using $c_{ijkl} = c_{klij}$ in for isotropic materials, the term is rewritten as

$$\begin{aligned} -\frac{1}{2} c_{ijkl} (\varepsilon_{ij} \varepsilon_{kl}^0 + \varepsilon_{ij}^0 \varepsilon_{kl}) &= -\frac{1}{2} c_{ijkl} (\varepsilon_{ij} Q_{klmn} P_m P_n + Q_{ijmn} P_m P_n \varepsilon_{kl}) \\ &= -\frac{1}{2} (c_{ijmn} + c_{mni j}) Q_{mnkl} \varepsilon_{ij} P_k P_l \\ &= -c_{ijmn} Q_{mnkl} \varepsilon_{ij} P_k P_l \\ &= -q_{ijkl} \varepsilon_{ij} P_k P_l, \end{aligned} \quad (4.12)$$

where $q_{ijkl} (= c_{ijmn} Q_{mnkl})$ is piezoelectric coupling coefficients. The third term of Eq. (4.11) is

$$\begin{aligned} \frac{1}{2} c_{ijkl} \varepsilon_{ij}^0 \varepsilon_{kl}^0 &= \frac{1}{2} c_{ijkl} Q_{ijmn} P_m P_n Q_{klmn} P_m P_n \\ &= \frac{1}{2} c_{mnop} Q_{mni j} Q_{opkl} P_i P_j P_k P_l. \end{aligned} \quad (4.13)$$

The right side of this equation is the fourth-order term of P_i , which can be incorporated into Landau energy density. Therefore, f_{elast} is expressed by

$$f_{\text{elast}} = \frac{1}{2} c_{ijkl} \varepsilon_{ij} \varepsilon_{kl} - q_{ijkl} \varepsilon_{ij} P_k P_l. \quad (4.14)$$

The second term of Eq. (4.14) represents the energy when strain and polarization are presented simultaneously, describing the piezoelectric effect in ferroelectric materials. Table 4-1 shows all of the material parameters used in this chapter.

Table 4-1 Material parameters of SrTiO₃ and PbTiO₃ [33–35]. T [K] is temperature.

Parameters (SI units)		SrTiO ₃	PbTiO ₃
Landau coefficients	α_1 ($10^5 \text{C}^{-2} \text{m}^2 \text{N}$)	$7.06(T - 35.5)$	$3.8(T - 479)$
	α_{11} ($10^8 \text{C}^{-4} \text{m}^6 \text{N}$)	17.0	−0.7252
	α_{12} ($10^8 \text{C}^{-4} \text{m}^6 \text{N}$)	44.52	7.5
	α_{111} ($10^8 \text{C}^{-6} \text{m}^{10} \text{N}$)	0.0	2.606
	α_{112} ($10^8 \text{C}^{-6} \text{m}^{10} \text{N}$)	0.0	6.1
	α_{123} ($10^8 \text{C}^{-6} \text{m}^{10} \text{N}$)	0.0	−37
Gradient energy coefficients	G_{11} ($10^{-10} \text{C}^{-2} \text{m}^4 \text{N}$)	0.734	1.7252
	G_{12} ($10^{-10} \text{C}^{-2} \text{m}^4 \text{N}$)	0.0	0.0
	G_{44} ($10^{-10} \text{C}^{-2} \text{m}^4 \text{N}$)	0.367	0.8626
Elastic constants	c_{11} ($10^{11} \text{m}^{-2} \text{N}$)	3.36	1.746
	c_{12} ($10^{11} \text{m}^{-2} \text{N}$)	1.07	0.7937
	c_{44} ($10^{11} \text{m}^{-2} \text{N}$)	1.27	1.111
Electrostrictive coefficients	Q_{11} ($10^{-2} \text{C}^{-2} \text{m}^4$)	4.57	8.9
	Q_{12} ($10^{-2} \text{C}^{-2} \text{m}^4$)	−1.35	−2.6
	Q_{44} ($10^{-2} \text{C}^{-2} \text{m}^4$)	0.957	6.75
Background dielectric constants	$\kappa_{11}, \kappa_{22}, \kappa_{33}$ (Dimensionless)	40.0	40.0
	$\kappa_{12}, \kappa_{23}, \kappa_{31}$ (Dimensionless)	0.0	0.0
Lattice constants	a_1 (10^{-10}m)	3.905 ($T = 300 \text{ K}$)	3.957 ($T = 300 \text{ K}$)
		3.909 ($T = 373 \text{ K}$)	3.961 ($T = 373 \text{ K}$)
	a_2 (10^{-10}m)	3.905 ($T = 300 \text{ K}$)	3.957 ($T = 300 \text{ K}$)
		3.909 ($T = 373 \text{ K}$)	3.961 ($T = 373 \text{ K}$)

4.2.2 Simulation model and procedure

Figure 4-1 shows the simulation model mimicking the experimentally fabricated $\text{SrTiO}_3/\text{PbTiO}_3$ superlattice on the SrTiO_3 substrate. The number of layers is 9 for each layer, following a previous study [10,27] in which $[(\text{SrTiO}_3)_{16}/(\text{PbTiO}_3)_{16}]_9$ superlattice model was used to simulate polar skyrmion, consistent with experimental observation. The in-plane size is $80 \text{ nm} \times 80 \text{ nm}$, the thickness of each layer is 6.4 nm , and the substrate thickness is 12 nm . A three-dimensional periodical boundary condition is applied, and a 12 nm thickness vacuum layer is added to the top of the model to avoid interactions with adjacent superlattices in image cells. The entire model is divided by a uniform 0.4 nm mesh. To simulate the closed-circuit conditions of experimental specimens [10,27], the electric potentials at the bottom and top of the superlattice are set to be uniform. A misfit strain due to lattice mismatch between materials is

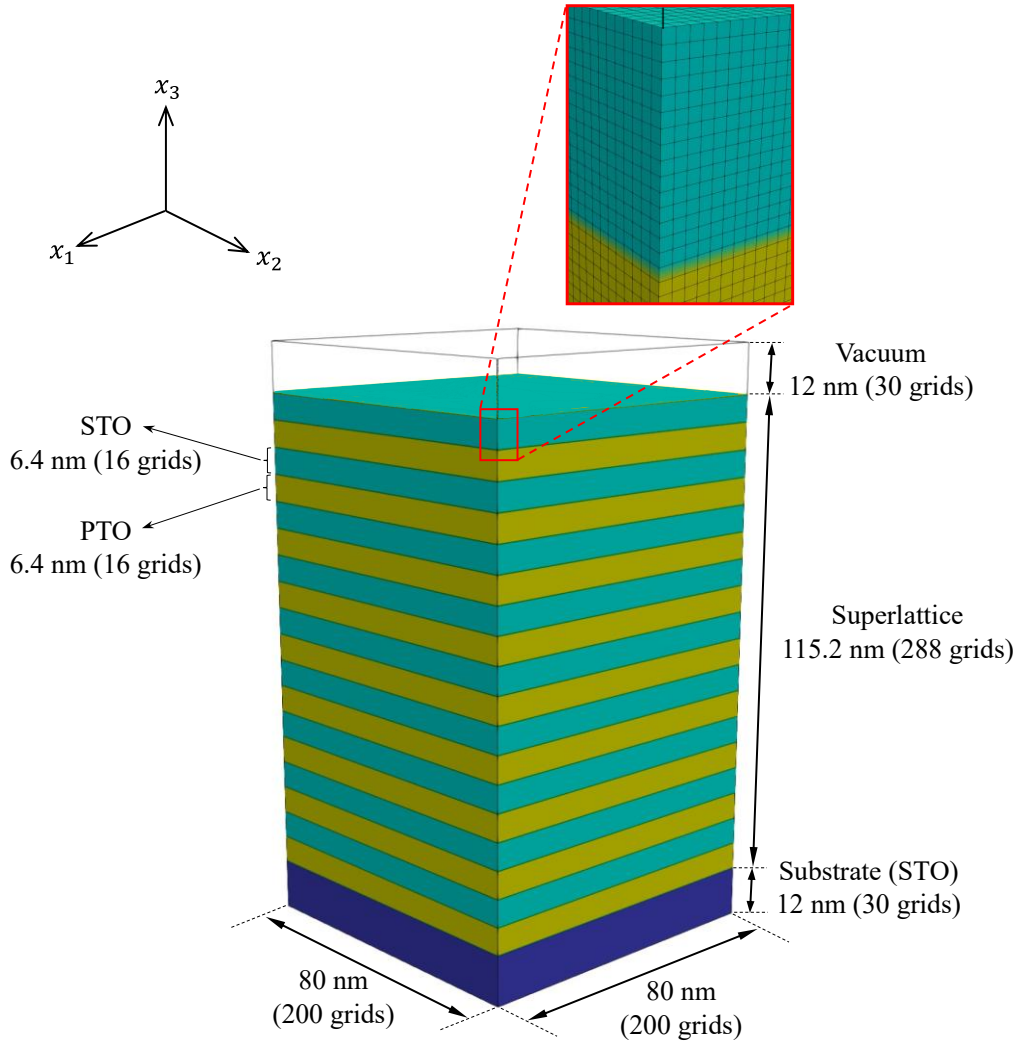


Figure 4-1 Schematic of the simulation model and mesh divisions of $[(\text{SrTiO}_3)_{16}/(\text{PbTiO}_3)_{16}]_9$ superlattice on SrTiO_3 substrate.

considered by applying uniform in-plane eigen strain [36]. Within the superlattice, the PbTiO_3 and SrTiO_3 films induce in-plane tensile and compressive strain on each other due to differences in their lattice constants. This is a temperature-dependent constant calculated by lattice constants given in Table 4-1. Additionally, the entire superlattice, which has an average lattice constant of PbTiO_3 and SrTiO_3 , is subjected to misfit strain due to the mismatch with the substrate. While both strains are considered in calculations, only the latter is written in the manuscript as an in-plane misfit strain.

Eqs. (4.1)~(4.3) are numerically solved by the fast-Fourie transform method [37,38] to obtain time evolution and equilibrium state of polarization. A dimensionless time step is $\Delta t^* = 0.05$. Simulations are performed under temperatures of $T = 373$ K, at which the skyrmion lattice phase was observed experimentally [27]. Assuming the poling process, the initial polarization distribution is set to a uniform out-of-plane direction ($P_3 = 0.10$ C/m²) plus a small amount of 3-dimensional random noise with a magnitude of 0.010 C/m². To investigate the effect of in-plane strain, calculations are performed for isotropic in-plane strains from 0.0% to -0.8% in every 0.1% and for anisotropic in-plane strains of $(\varepsilon_{11}, \varepsilon_{22}) = (-1.0\%, -0.5\%), (-1.5\%, 0.0\%)$.

4.3 Results and discussion

Figure 4-2 presents the polarization distributions at the thermodynamic equilibrium state in top PTO layers under various in-plane strain conditions. Overall, downward polarization domains exist within upward polarization regions. Under low compressive isotropic in-plane strain below -0.2% shown in Figure 4-2 (a-1), elongated and intricate domains of about 10 nm width emerge. As shown in Figure 4-2 (b-1), the polarization vectors at the outer edges of these domains continuously transition between upward and downward through in-plane direction. Other domains exhibit similar characteristics, indicating that these domains are elongated skyrmions, intermingled in the entire PTO film. This feature corresponds to the labyrinth phase [28]. As the strain increases, although several small domains appear, most are still elongated domains. On the other hand, under the high compressive isotropic strain above -0.3% shown in Figure 4-2(a-2), many small circular domains of less than 10 nm in diameter are formed. As shown in Figure 4-2(b-2), each circular domain is skyrmion with continuous polarization rotation similar to that of the small strain conditions. Therefore, there are numerous circular skyrmions in the entire film, indicating a formation of a skyrmion bubble phase. Furthermore, at -0.8% , some skyrmions arrange into a square lattice. The aligned skyrmions

have almost circular shapes, with a radius is about 10 nm and center-to-center distance of about 15 nm. These features are consistent with the previously observed skyrmion lattice [27], confirming the signifiy the formation of a skyrmion lattice phase. Therefore, increasing isotropic compressive strain causes the topological phase transition from labyrinth phases to skyrmion bubble and skyrmion lattice phases. These results are consistent with those of a previous study [28], which reported an in-plane strain-dependent topological transition from the labyrinth

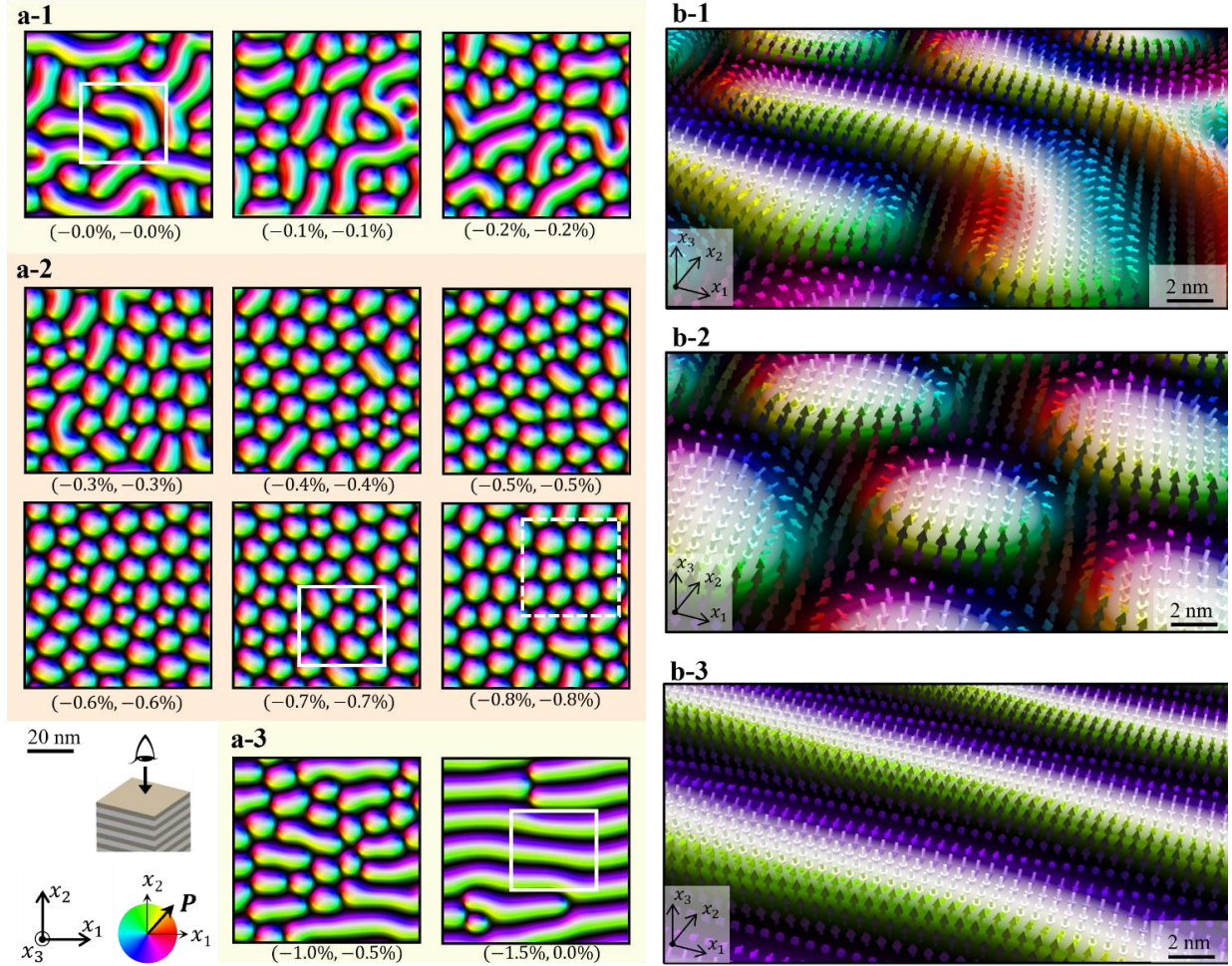


Figure 4-2 Polarization distribution of the top PbTiO_3 layers in $\text{PbTiO}_3/\text{SrTiO}_3$ superlattices. (a) Distributions in the x_1 - x_2 plane under different in-plane strain conditions: (a-1) small isotropic strain, (a-2) large isotropic strain, and (a-3) anisotropic strain. The values of in-plane strain $(\epsilon_{11}, \epsilon_{22})$ are indicated below each figure. (b) Detailed polarization distributions within the regions highlighted by white squares in (a). The color map represents the in-plane polarization direction, while the black and white regions correspond to upward and downward out-of-plane polarization.

phase to the skyrmion phase. Note that a vortex phase, commonly observed in STO/PTO superlattices [28,39], does not appear in our case, because the misfit strain substantially applied to the PTO films is below the range for its formation. In addition, polarization distribution also depends on strain direction under anisotropic strain, as shown in Figure 4-2(a-3). At $(\varepsilon_{11}, \varepsilon_{22}) = (-1.0\%, -0.5\%)$, stripe domains aligned with the x_1 axis coexist with skyrmions. The higher strain anisotropy as $(\varepsilon_{11}, \varepsilon_{22}) = (-1.5\%, 0.0\%)$ tends to eliminate circular skyrmions, resulting in dominant stripe domains. Therefore, increasing the in-plane strain anisotropy promotes stripe phase formation.

These results confirm that in-plane strain significantly influences skyrmion and phase formation. In the experimental observations in the previous study [27], substrate-superlattice lattice mismatches cause strain variations, producing skyrmion bubble phases at $(\varepsilon_{11}, \varepsilon_{22}) = (-0.7\%, -0.7\%)$, skyrmion lattice phases at $(-0.9\%, -0.9\%)$, and stripe phases at $(-1.3\%, -1.1\%)$. Furthermore, the width of each stripe domain was approximately 10 nm, and the diameters of the skyrmions varied from approximately 5 to 10 nm. Our results are quantitatively consistent with this experimental observation and demonstrate the validity of the simulation approach to the skyrmion formation process. In the following, the polarization evolution process under each strain condition is described in detail.

At first, as a representative example of the formation process of labyrinth phase, the temporal evolution of polarization under the strain-free condition is shown in Figure 4-3. As shown in Figure 4-3(a), the polarization at the initial stage ($t = 5$ ns) is nearly homogeneous without any domains. By $t = 10$ ns, downward polarization domains emerge across all PTO layers. As time progresses further, domains grow and connect with other domains, eventually forming the labyrinth phase. To examine the domain formation process in detail, the early stages of polarization evolution in the top PTO layer are shown in Figure 4-3(b). At the initial stage ($t = 5$ ns), P_3 has small positive values overall, among which some local regions show relatively small values. These regions act as nuclei for polarization inversion, with P_3 transitioning to negative values at $t = 8$ ns. By $t = 10$ ns, the magnitude of P_3 increases, and the polarization inverted domains expand. At this stage, adjacent domains begin to connect with each other. By $t = 15$ ns, the domains are fully connected, resulting in the formation of elongated domains. Thus, under the strain-free condition, the polarization inversion domains grow from relatively small value regions and connect with adjacent domains, eventually forming the labyrinth phase consisting of elongated and intricate domains. This process is similar in other PTO layers, where the labyrinth phase develops simultaneously across all layers.

On the other hand, skyrmion formation under isotropic in-plane strain conditions occurs via a different process. Figure 4-4(a) shows the polarization evolution at $(\varepsilon_{11}, \varepsilon_{22}) = (-0.7\%, -0.7\%)$, where the formation of well-defined skyrmion bubble phase is observed. At the initial stage ($t = 10$ ns), polarization inverted domains appear only in the lowest layer. Toward $t = 20$ ns, domain formation extends to the upper layers, and by $t = 25$ ns, domains are present throughout all layers. Subsequently, each domain expands slightly but remains isolated, rarely connecting with others. This gradual domain formation, progressing from the lower to upper layers, contrasts with the simultaneous domain formation observed under the strain-free

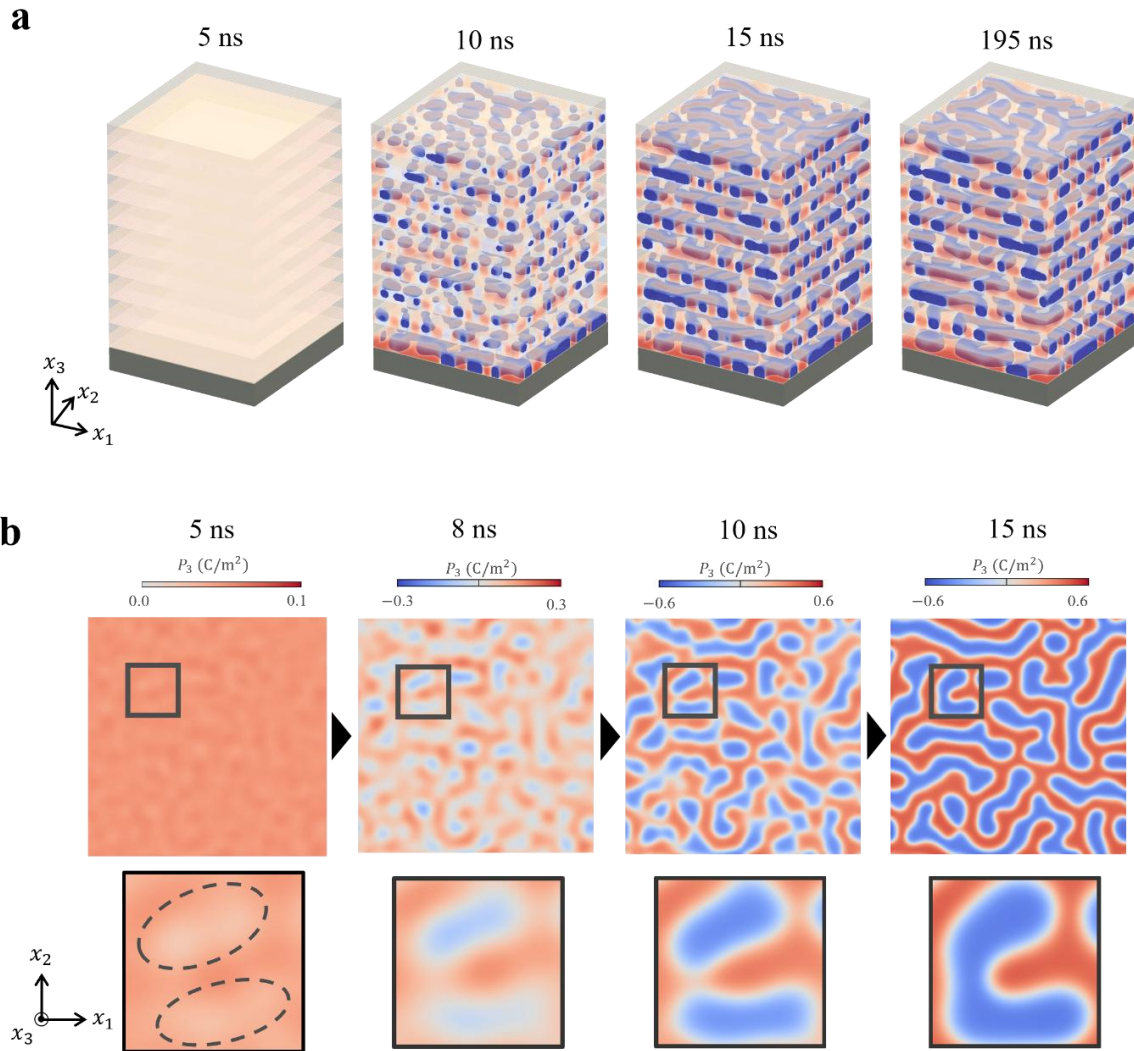


Figure 4-3 Time evolution of out-of-plane polarization during formation process of labyrinth phase at $(\varepsilon_{11}, \varepsilon_{22}) = (0.0\%, 0.0\%)$. (a) Evolution across the entire superlattice, with blue areas indicating downward polarization domains. (b) Evolution in the top PbTiO_3 layer. The color map represents the out-of-plane polarization P_3

condition. To investigate the process in more detail, Figure 4-4(b) focuses on the top PTO layer. Similar to the strain-free condition, polarization inversions begin in local regions with small values. However, the inversion timing varies across domains, avoiding their connection during growth. Consequently, numerous isolated domains become circular skyrmions, resulting in the formation of the skyrmion bubble phase. These features are more obvious at $(\varepsilon_{11}, \varepsilon_{22}) = (-0.8\%, -0.8\%)$ as shown in Figure 4-5, emphasizing that magnitude of in-plane strain affects the domain growth velocities.

In these processes, the polarization inversion appears to be suppressed. In other words, some

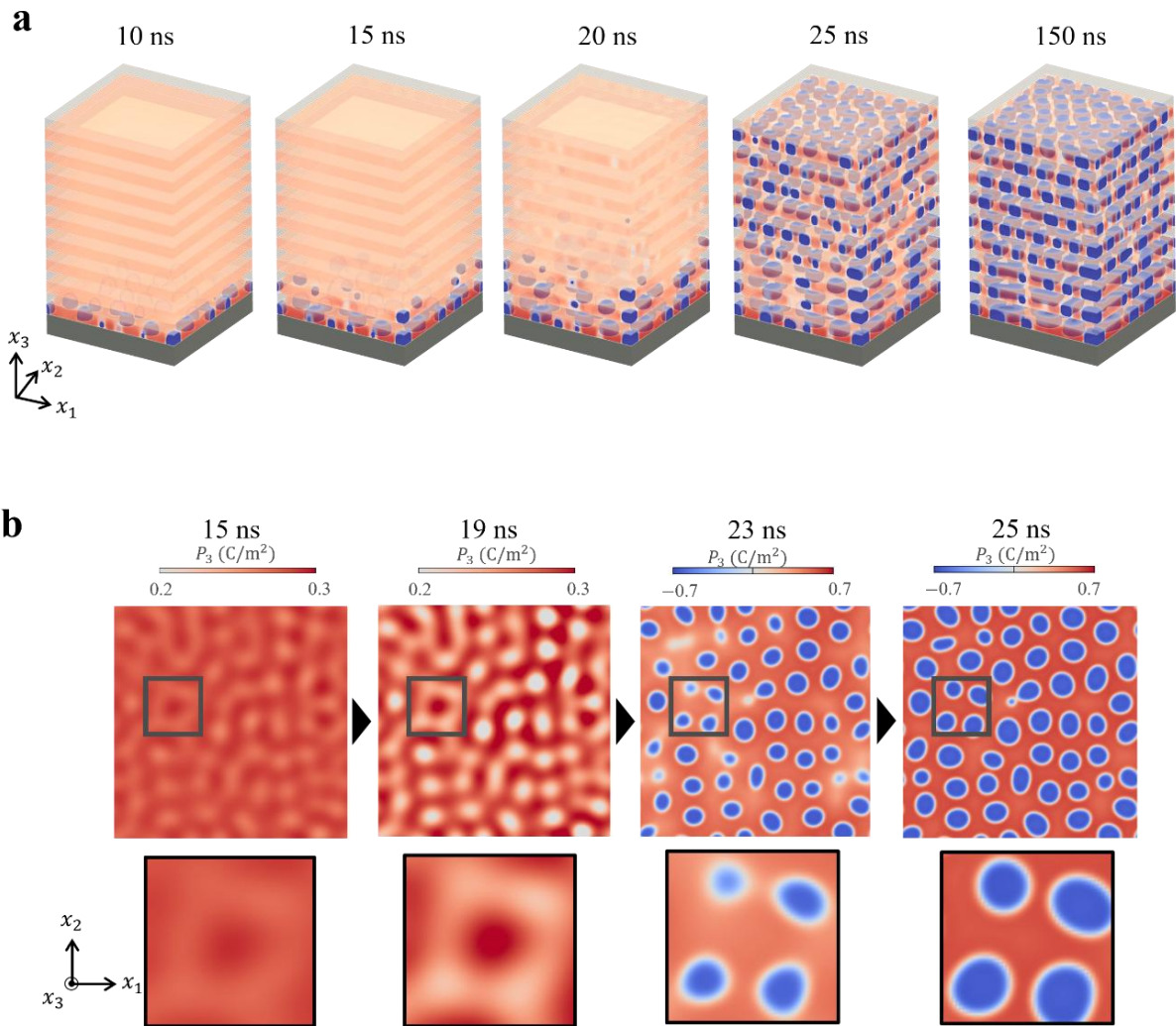


Figure 4-4 Time evolution of out-of-plane polarization during formation process of skyrmion bubble phase at $(\varepsilon_{11}, \varepsilon_{22}) = (-0.7\%, -0.7\%)$. (a) Evolution across the entire superlattice, with blue areas indicating downward polarization domains. (b) Evolution in the top PbTiO_3 layer. The color map represents the out-of-plane polarization P_3 .

trigger is likely required to initiate the inversion. Comparing Figure 4-4(b) and Figure 4-3(b), at the initial stage, P_3 values are higher under compressive strain, indicating the stabilization of out-of-plane polarization by in-plane compressive strain, which inhibits polarization inversion. In this time, polarization inversion is possible only in the bottom PTO layer due to the mechanical constraint of substrate. Furthermore, inverted domains tend to form directly above those domains in the lower layers and gradually propagate upward, suggesting that polarization inversion in the lower layer drives subsequent inversions in the upper layer. Therefore, under in-plane strain, stabilization of out-of-plane polarization inhibits inversion, resulting in the

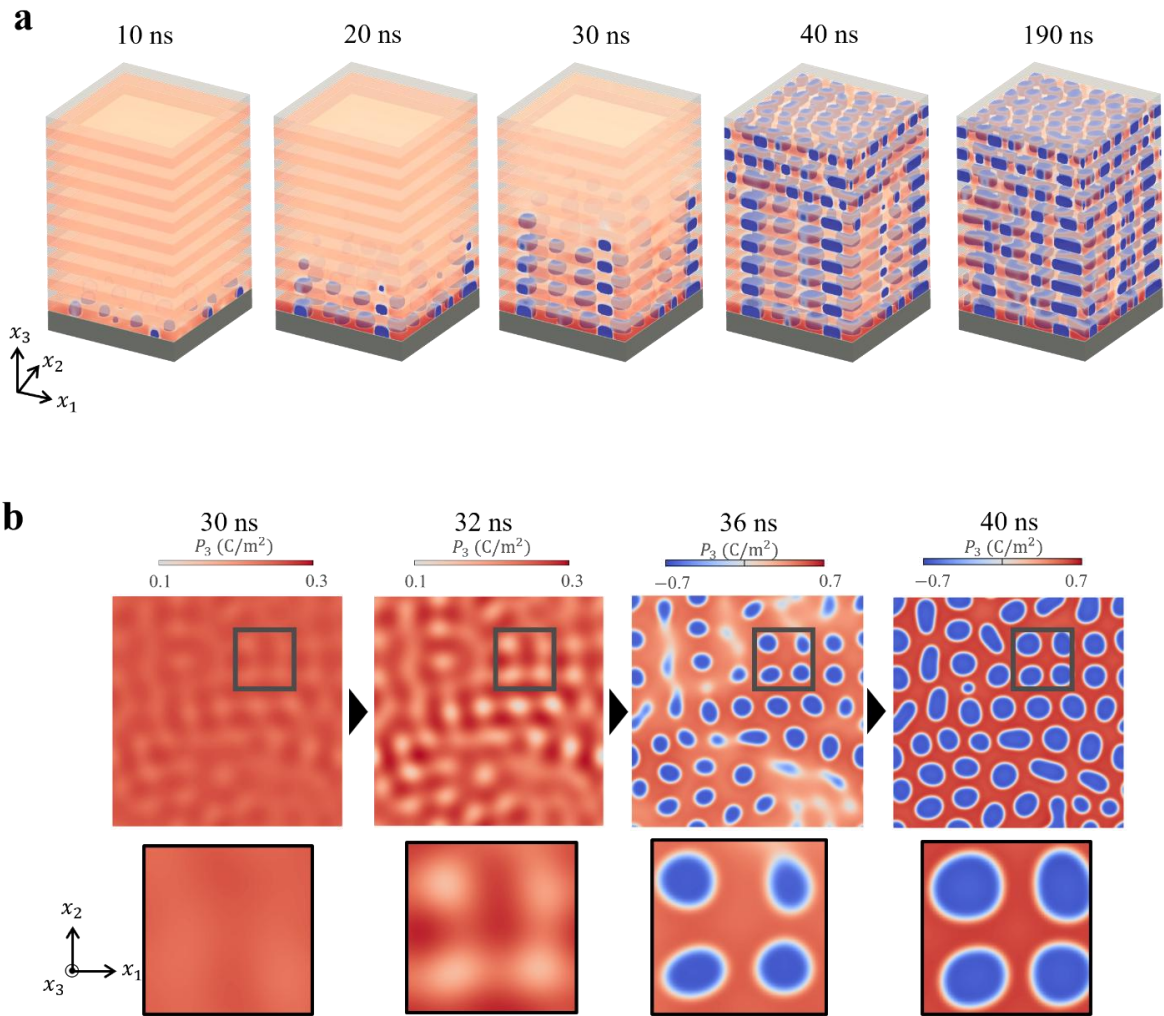


Figure 4-5 Time evolution of out-of-plane polarization during formation process of skyrmion bubble phase at $(\varepsilon_{11}, \varepsilon_{22}) = (-0.8\%, -0.8\%)$. (a) Evolution across the entire superlattice, with blue areas indicating downward polarization domains. (b) Evolution in the top PbTiO₃ layer. The color map represents the out-of-plane polarization P_3 .

formation of skyrmions from individually grown domains.

The formation process of the stripe phase under anisotropic in-plane strain condition shown in Figure 4-6 also indicates the influence of in-plane strain on the polarization evolution. As shown in Figure 4-6(a), the first polarization inversion occurs in the lowest layer at $t = 10$ ns, and by $t = 25$ ns, inverted domains have developed across all layers. These domains connect exclusively in the x_1 direction, producing narrow, elongated domains that form the stripe phase. The detailed views shown in Figure 4-6(b) indicate that inverted domains emerge from the nuclei of small P_3 regions, consistent with results under other conditions. However, at $t = 15$ ns,

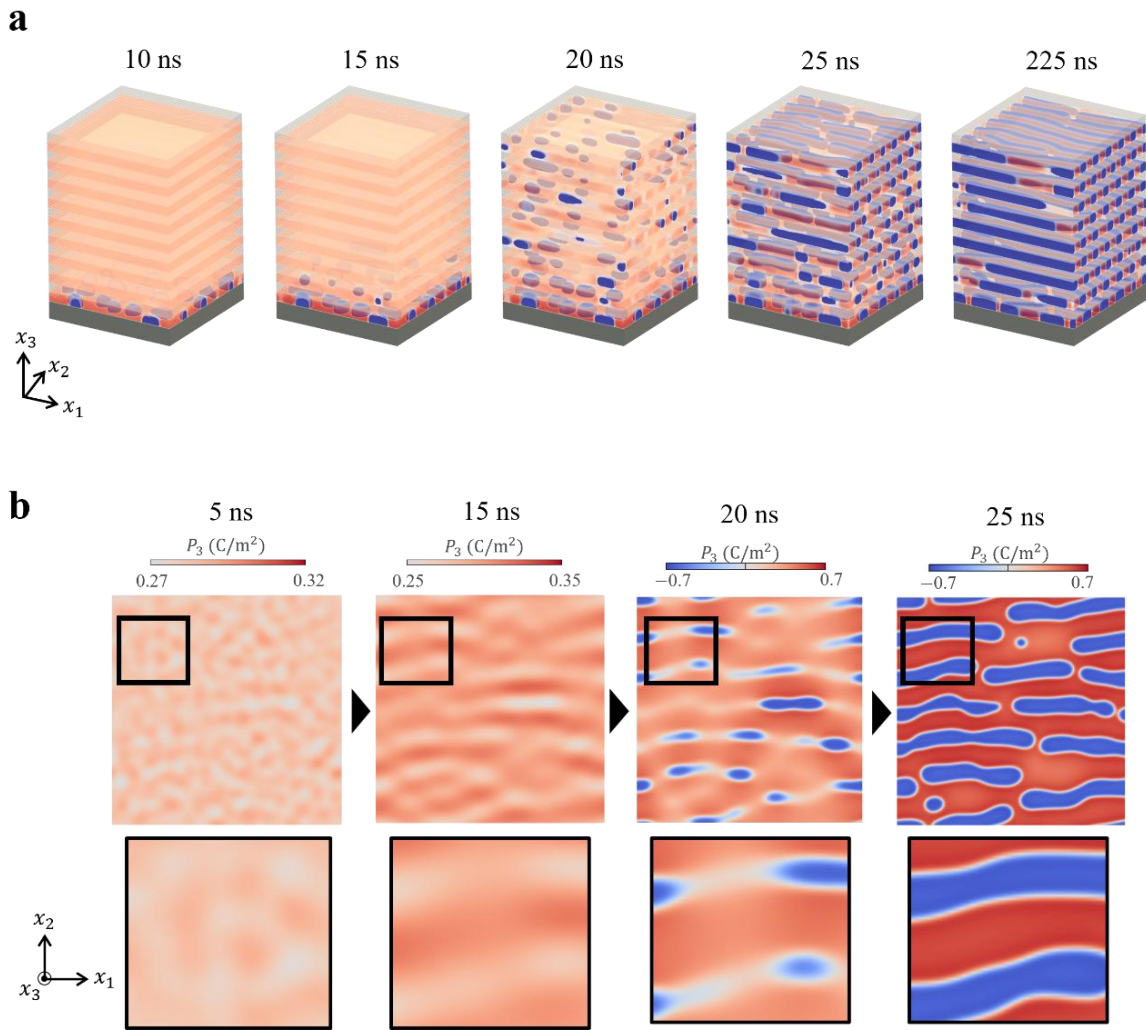


Figure 4-6 Time evolution of out-of-plane polarization during formation process of stripe phase at $(\varepsilon_{11}, \varepsilon_{22}) = (-1.5\%, 0.0\%)$. (a) Evolution across the entire superlattice, with blue areas indicating downward polarization domains. (b) Evolution in the top PbTiO_3 layer. The color map represents the out-of-plane polarization P_3 .

these nuclei are already elongated along the x_1 direction. This behavior is attributed to the application of substantial compressive strain in the x_1 direction, which suppresses P_1 . These results indicate that in-plane strain strongly affects the polarization inversion even in the early stages. Consequently, the anisotropy of in-plane strain determines the direction of stripe domains, suggesting the potential for mechanical and selective control of domain formation.

4.4 Conclusion

In summary, in-plane strain plays a pivotal role in the skyrmion formation process and the topological transition in PTO/STO superlattices. Under low in-plane strain conditions, polarization inversion occurs readily, leading to the simultaneous growth of numerous domains. During growth, domains connect to each other, resulting in the formation of the labyrinth phase. On the other hand, higher in-plane strains suppress polarization inversion, forcing independent domain growth that culminates in skyrmion formation. Furthermore, the anisotropy of in-plane strain affects the direction of domain growth, and strong anisotropy promotes the formation of the stripe domain. Although strain-induced topological polarization orders in PTO/STO have been studied extensively, our results reveal their formation processes for the first time. These findings advance the understanding of the formation mechanisms of polar skyrmions and offer valuable guidance for the development of devices based on topological polarization orders.

References

- [1] J. F. Scott, Applications of Modern Ferroelectrics, *Science* **315**, 954 (2007).
- [2] N. Li, K. Xie, and H. Huang, Ferroelectric Materials for High Energy Density Batteries: Progress and Outlook, *ACS Energy Lett.* **8**, 4357 (2023).
- [3] Z. Chen et al., All-ferroelectric implementation of reservoir computing, *Nat Commun* **14**, 3585 (2023).
- [4] M. Yan et al., Ferroelectric Synaptic Transistor Network for Associative Memory, *Advanced Electronic Materials* **7**, 2001276 (2021).
- [5] L. Liu et al., Control the Neural Stem Cell Fate with Biohybrid Piezoelectrical Magnetite Micromotors, *Nano Lett.* **21**, 3518 (2021).
- [6] I. I. Naumov, L. Bellaiche, and H. Fu, Unusual phase transitions in ferroelectric nanodisks and nanorods, *Nature* **432**, 7018 (2004).
- [7] B. J. Rodriguez, X. S. Gao, L. F. Liu, W. Lee, I. I. Naumov, A. M. Bratkovsky, D. Hesse, and M. Alexe, Vortex Polarization States in Nanoscale Ferroelectric Arrays, *Nano Lett.* **9**, 1127 (2009).
- [8] J. Junquera et al., Topological phases in polar oxide nanostructures, *Rev. Mod. Phys.* **95**, 025001 (2023).
- [9] Y. Nahas, S. Prokhorenko, L. Louis, Z. Gui, I. Kornev, and L. Bellaiche, Discovery of stable skyrmionic state in ferroelectric nanocomposites, *Nat Commun* **6**, 8542 (2015).
- [10] S. Das et al., Observation of room-temperature polar skyrmions, *Nature* **568**, 368 (2019).
- [11] S. Das et al., Local negative permittivity and topological phase transition in polar skyrmions, *Nat. Mater.* **20**, 194 (2021).
- [12] L. Gao, S. Prokhorenko, Y. Nahas, and L. Bellaiche, Dynamical Control of Topology in Polar Skyrmions via Twisted Light, *Phys. Rev. Lett.* **132**, 026902 (2024).
- [13] L. Han et al., High-density switchable skyrmion-like polar nanodomains integrated on silicon, *Nature* **603**, 7899 (2022).
- [14] F.-H. Gong, Y.-L. Tang, Y.-J. Wang, Y.-T. Chen, B. Wu, L.-X. Yang, Y.-L. Zhu, and X.-L. Ma, Absence of critical thickness for polar skyrmions with breaking the Kittel's law, *Nat Commun* **14**, 3376 (2023).
- [15] S. Mühlbauer, B. Binz, F. Jonietz, C. Pfleiderer, A. Rosch, A. Neubauer, R. Georgii, and P. Böni, Skyrmion Lattice in a Chiral Magnet, *Science* **323**, 915 (2009).

- [16] X. Z. Yu, Y. Onose, N. Kanazawa, J. H. Park, J. H. Han, Y. Matsui, N. Nagaosa, and Y. Tokura, Real-space observation of a two-dimensional skyrmion crystal, *Nature* **465**, 901 (2010).
- [17] B. Göbel, I. Mertig, and O. A. Tretiakov, Beyond skyrmions: Review and perspectives of alternative magnetic quasiparticles, *Physics Reports* **895**, 1 (2021).
- [18] K. Wang, V. Bheemarasetty, J. Duan, S. Zhou, and G. Xiao, Fundamental physics and applications of skyrmions: A review, *Journal of Magnetism and Magnetic Materials* **563**, 169905 (2022).
- [19] A. Fert, V. Cros, and J. Sampaio, Skyrmions on the track, *Nature Nanotech* **8**, 152 (2013).
- [20] C. H. Marrows, J. Barker, T. A. Moore, and T. Moorsom, Neuromorphic computing with spintronics, *Npj Spintronics* **2**, 12 (2024).
- [21] M.-K. Lee and M. Mochizuki, Handwritten digit recognition by spin waves in a Skyrmion reservoir, *Sci Rep* **13**, 19423 (2023).
- [22] S. Li, X. Wang, and T. Rasing, Magnetic skyrmions: Basic properties and potential applications, *Interdisciplinary Materials* **2**, 260 (2023).
- [23] S. Sen and S. S. Mandal, *Theory of Polar Skyrmions in Layered Structure of Ferroelectric Perovskites*, arXiv:2409.16100.
- [24] I. Dzyaloshinsky, A thermodynamic theory of “weak” ferromagnetism of antiferromagnetics, *Journal of Physics and Chemistry of Solids* **4**, 241 (1958).
- [25] T. Moriya, Anisotropic Superexchange Interaction and Weak Ferromagnetism, *Phys. Rev.* **120**, 91 (1960).
- [26] R. Zhu et al., Dynamics of Polar Skyrmion Bubbles under Electric Fields, *Phys. Rev. Lett.* **129**, 107601 (2022).
- [27] Y.-T. Shao et al., Emergent chirality in a polar meron to skyrmion phase transition., *Nat Commun* **14**, 1355 (2023).
- [28] C. Dai, Z. Hong, S. Das, Y.-L. Tang, L. W. Martin, R. Ramesh, and L.-Q. Chen, Strain effects on stability of topological ferroelectric polar configurations in $(\text{PbTiO}_3)_n / (\text{SrTiO}_3)_n$ superlattices, *Applied Physics Letters* **123**, 052903 (2023).
- [29] Y. Zhang, Q. Li, H. Huang, J. Hong, and X. Wang, Strain manipulation of ferroelectric skyrmion bubbles in a freestanding PbTiO_3 film: A phase field simulation, *Phys. Rev. B* **105**, 224101 (2022).

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- [30] G. Sheng, Y. L. Li, J. X. Zhang, S. Choudhury, Q. X. Jia, V. Gopalan, D. G. Schlom, Z. K. Liu, and L. Q. Chen, A modified Landau–Devonshire thermodynamic potential for strontium titanate, *Appl. Phys. Lett.* **96**, 232902 (2010).
- [31] Y. L. Li, S. Y. Hu, Z. K. Liu, and L. Q. Chen, Effect of substrate constraint on the stability and evolution of ferroelectric domain structures in thin films, *Acta Materialia* **50**, 395 (2002).
- [32] C. H. Woo and Y. Zheng, Depolarization in modeling nano-scale ferroelectrics using the Landau free energy functional, *Appl. Phys. A* **91**, 59 (2008).
- [33] G. Sheng, Y. L. Li, J. X. Zhang, S. Choudhury, Q. X. Jia, V. Gopalan, D. G. Schlom, Z. K. Liu, and L. Q. Chen, A modified Landau–Devonshire thermodynamic potential for strontium titanate, *Appl. Phys. Lett.* **96**, 232902 (2010).
- [34] Y. L. Li, S. Y. Hu, Z. K. Liu, and L. Q. Chen, Effect of substrate constraint on the stability and evolution of ferroelectric domain structures in thin films, *Acta Materialia* **50**, 395 (2002).
- [35] M. J. Haun, E. Furman, S. J. Jang, H. A. McKinstry, and L. E. Cross, Thermodynamic theory of PbTiO_3 , *Journal of Applied Physics* **62**, 3331 (1987).
- [36] L.-Q. Chen, Phase-Field Method of Phase Transitions/Domain Structures in Ferroelectric Thin Films: A Review, *J American Ceramic Society* **91**, 1835 (2008).
- [37] H.-L. Hu and L.-Q. Chen, Three-Dimensional Computer Simulation of Ferroelectric Domain Formation, *Journal of the American Ceramic Society* **81**, 492 (2005).
- [38] L. Q. Chen and J. Shen, Applications of semi-implicit Fourier-spectral method to phase field equations, *Computer Physics Communications* **108**, 147 (1998).
- [39] C. Tan et al., Engineering polar vortex from topologically trivial domain architecture, *Nat Commun* **12**, 4620 (2021).

Chapter 5 Dislocation structures in emergent polar skyrmion lattices

5.1 Introduction

In Chapter 4, I investigated the collective generations of polar skyrmions and successfully demonstrated the emergence of a lattice phase. In this chapter, I focus on this skyrmion lattice and attempt to elucidate its properties from the perspective of lattice mechanics. I will target dislocations, which are typical lattice defects, and clarify their structural characteristics and effects on skyrmion lattice.

Skyrmions are an emerging nanoscale topological configuration of an electric dipole, magnetic moment, or other physical vectors observed in various physical systems [1–4], and they are also treated as particles, called quasi-particles. Extensive researches [1,5–10] on magnetic skyrmions in ferromagnetics have unveiled their intriguing features such as stability due to the topological protection, ability to deform, annihilate, generate, and be driven by external fields [8,11–15], and exploited potential applications including racetrack memory, reservoir computing devices, and logic gates [7,16–18]. Following the great success of magnetic skyrmion, a polar skyrmion [2,19–29], which was first discovered in $\text{PbTiO}_3/\text{SrTiO}_3$ superlattices very recently [2], is also anticipated to exhibit valuable properties and applications. In ferroelectrics, the dynamics of microscopic polarization domain patterns determine the macroscopic electrical and mechanical responses, providing potential for various applications such as programmable devices [30], energy storage [31], and neuromorphic computing devices [32]. However, the limitation on the specimen's thickness for ferroelectricity restricts the enhancement of domain density and device properties. The polar skyrmion is reported to overcome this limitation, existing in PbTiO_3 film with a thickness of only two unit cells while retaining ferroelectricity [22]. Additionally, several studies reported that the polar skyrmion exhibits unique electrical responses such as negative permittivity [20], readability with high-density [21], and high optical response [33], sparking growing interest from both scientific and engineering fields. In addition, the skyrmion is treated as one of topological solitons [34]. The

individual and collective response of solitonic particles are one of important researches in condensed matter physics [35,36].

Skyrmions are arranged in triangles or squares, to form skyrmion lattices (also called skyrmion crystals (SkX)) [24,37], indicating that it is essential to understand the fundamental properties and electric or mechanical response of a polar skyrmion lattice, rather than individual one. It has been reported that uniaxial tensile strain deforms magnetic skyrmion lattices with large anisotropic deformation of individual skyrmions [38], indicating the unique behavior of skyrmion lattices distinct from conventional lattices such as atomic crystal lattices. Moreover, in many experimental observations and theoretical simulations, magnetic skyrmion lattices contain lattice defects such as vacancies [39], dislocations [40,41], and grain boundaries [42,43] similar to those found in atomic systems. Analogous to atomic crystal lattices, these lattice defects dominate the dynamic behavior of skyrmion lattices [40,44]. In particular, SkX dislocations are intensively studied among the defects because their dynamics contribute to a plethora of unique behaviors of skyrmion lattices such as rotations and deformations [40], melting [44], and stabilization [41]. In particular, the melting of the skyrmion lattice is discussed in terms of a two-dimensional phase transition [45,46]. Based on KTHNY (Kosterlitz, Thouless, Halperin, Nelson, and Young) theory, dislocations contribute significantly to melting of lattice [44,47]. Dislocations in polar skyrmion lattices, therefore, are expected to play an important role in unique lattice dynamics arising from the quasi-particle nature of skyrmion. A dislocation-like lattice mismatch has also been experimentally observed [24] and the emergence of dislocation has been theoretically predicted [48] in polar skyrmion lattices, while the detail and fundamental properties of the dislocation remain unrevealed. In general, dislocations exhibit an inherent elastic strain field that follows Volterra's law [49]. This elastic field can be described in a form proportional to $1/r$ (r is distance from a dislocation core), and this means that dislocations affect far from dislocation core each other (i.e., long-range interactions). Based on the Volterra's law, dislocations have unique interaction and dynamics, which lead important mechanical properties and responses such as yielding, hardening, and plastic deformation [50]. Therefore, understanding the fundamental properties of dislocation structures in polar skyrmion lattices, particularly whether the Volterra's law holds or not, is the key to investigating both static and dynamic properties of polar skyrmion lattices, including their novel responses to external fields. Although dislocations in skyrmion lattice have been observed experimentally, to the best of the author's knowledge, there is still no study focused on such elastic fields.

Here, I demonstrate that there exist the unique SkX dislocation structure and its inherent singular strain field in skyrmion lattice, which critically differ from those of conventional dislocation in atomic crystals and even fall outside of Volterra's elasticity theory of dislocation. Our phase-field simulation reveals that skyrmions around the dislocation core deform largely by more than 50% depending on external electric fields and temperature. Furthermore, a quasiparticle-strain analysis based on the continuum mechanics theory provides the detailed deformation properties of skyrmions and their lattices, which discovers novel mechanics hidden in skyrmion lattices.

5.2 Method

5.2.1 Phase-field modeling

The total free energy of a ferroelectric system is $F = \int f dV$, where the f is the total free energy density and written as

$$f = f_{\text{Landau}} + f_{\text{grad}} + f_{\text{electo}} + f_{\text{elast}}. \quad (5.1)$$

The first term is Landau energy density, describing the ferroelectric phase transition depending on temperature, is expressed by [51]

$$f_{\text{Landau}} = \alpha_{ij}P_iP_j + \alpha_{ijkl}P_iP_jP_kP_l + \alpha_{ijklmn}P_iP_jP_kP_lP_mP_n, \quad (5.2)$$

where α_{ij} , α_{ijkl} , and α_{ijklmn} are the Landau coefficients. The second term is gradient energy density, describing the formation of polarization domain walls, is expressed by [52]

$$f_{\text{grad}} = \frac{1}{2}G_{ijkl}P_{i,j}P_{k,l}, \quad (5.3)$$

where G_{ijkl} is the gradient energy coefficient. The third term is electrostatic energy density is expressed by [53]

$$f_{\text{electo}} = -\frac{1}{2}\varepsilon_b E_i E_i - E_i P_i, \quad (5.4)$$

where ε_b is the background permittivity. In this study, to reproduce the electrical boundary conditions of thin film surface where polar skyrmion appears in the previous study [54], screening factor θ [55], which represents the amount of shielding of the surface charge, is introduced, and electric field is expressed by

$$E_i = E_i^{\text{ext}} + (1 - \theta)E_i^{\text{dep}}, \quad (5.5)$$

where E_i^{ext} is external electric field and E_i^{dep} is depolarization field. The fourth term is elastic energy density is expressed by

$$f_{\text{elast}} = \frac{1}{2} c_{ijkl} \varepsilon_{ij}^{\text{elast}} \varepsilon_{kl}^{\text{elast}}, \quad (5.6)$$

where c_{ijkl} is the elastic constant and $\varepsilon_{ij}^{\text{elast}}$ is elastic strain. In addition to elastic strain, ferroelectric materials exhibit eigen strain (intrinsic strain) caused by spontaneous polarization. The eigen strain ε_{ij}^0 can be written as [52]

$$\varepsilon_{ij}^0 = Q_{ijkl} P_k P_l, \quad (5.7)$$

where Q_{ijkl} is electrostrictive coefficients. The total strain ε_{ij} , defined as the displacement gradient in the material, is the sum of elastic strain and eigen strain, can be written as

$$\varepsilon_{ij}^{\text{elast}} = \varepsilon_{ij} - \varepsilon_{ij}^0. \quad (5.8)$$

Substituting Eq. (5.8), f_{elast} is

$$\begin{aligned} f_{\text{elast}} &= \frac{1}{2} c_{ijkl} \varepsilon_{ij}^{\text{elast}} \varepsilon_{kl}^{\text{elast}} \\ &= \frac{1}{2} c_{ijkl} (\varepsilon_{ij} - \varepsilon_{ij}^0) (\varepsilon_{kl} - \varepsilon_{kl}^0) \\ &= \frac{1}{2} c_{ijkl} \varepsilon_{ij} \varepsilon_{kl} - \frac{1}{2} c_{ijkl} (\varepsilon_{ij} \varepsilon_{kl}^0 + \varepsilon_{ij}^0 \varepsilon_{kl}) + \frac{1}{2} c_{ijkl} \varepsilon_{ij}^0 \varepsilon_{kl}^0. \end{aligned} \quad (5.9)$$

Substituting Eq. (5.7) into the second term, using $c_{ijkl} = c_{klij}$ in for isotropic materials,

$$\begin{aligned} -\frac{1}{2} c_{ijkl} (\varepsilon_{ij} \varepsilon_{kl}^0 + \varepsilon_{ij}^0 \varepsilon_{kl}) &= -\frac{1}{2} c_{ijkl} (\varepsilon_{ij} Q_{klmn} P_m P_n + Q_{ijmn} P_m P_n \varepsilon_{kl}) \\ &= -\frac{1}{2} (c_{ijmn} + c_{mni j}) Q_{mnkl} \varepsilon_{ij} P_k P_l \\ &= -c_{ijmn} Q_{mnkl} \varepsilon_{ij} P_k P_l \\ &= -q_{ijkl} \varepsilon_{ij} P_k P_l, \end{aligned} \quad (5.10)$$

where $q_{ijkl} (= c_{ijmn} Q_{mnkl})$ is piezoelectric coupling coefficients. And the third term is

$$\begin{aligned} \frac{1}{2} c_{ijkl} \varepsilon_{ij}^0 \varepsilon_{kl}^0 &= \frac{1}{2} c_{ijkl} Q_{ijmn} P_m P_n Q_{klmn} P_m P_n \\ &= \frac{1}{2} c_{mnop} Q_{mni j} Q_{opkl} P_i P_j P_k P_l. \end{aligned} \quad (5.11)$$

The right side of this equation is the fourth-order term of P_i , which can be incorporated into Landau energy density. Therefore, f_{elast} is expressed by

$$f_{\text{elast}} = \frac{1}{2} c_{ijkl} \varepsilon_{ij} \varepsilon_{kl} - q_{ijkl} \varepsilon_{ij} P_k P_l. \quad (5.12)$$

The second term of Eq. (5.12) represents the energy when strain and polarization are present simultaneously, describing the piezoelectric effect in ferroelectric materials. All of material parameters used in this study are shown in Table 5-1.

Time-dependent Ginzburg-Landau (TDGL) equation is

$$\partial P_i / \partial t = -L(\delta F / \delta P_i), \quad (5.13)$$

where t is time, L is the kinetic coefficient and $F = \int f dV$ is total free energy. Mechanical equilibrium equation is

$$\sigma_{ij,j} = (\partial f / \partial \varepsilon_{ij})_{,j} = 0, \quad (5.14)$$

where $\sigma_{ij,j}$ is the stress gradient. Maxwell's equations is

$$D_{i,i} = (-\partial f / \partial E_i)_{,i} = 0, \quad (5.15)$$

where $D_{i,i}$ is the electric displacement gradient in the material. Eqs. (5.13), (5.14), and (5.15) are the governing equations of the phase-field simulation.

Table 5-1 Material parameters for PbTiO₃ [54,56]. T is temperature.

Landau coefficients			
α_1 [C ⁻² m ² N]	$3.8(T - 752) \times 10^5$	α_{111} [C ⁻⁶ m ¹⁰ N]	2.606×10^8
α_{11} [C ⁻⁴ m ⁶ N]	-0.7252×10^8	α_{112} [C ⁻⁶ m ¹⁰ N]	6.1×10^8
α_{12} [C ⁻⁴ m ⁶ N]	7.5×10^8	α_{123} [C ⁻⁶ m ¹⁰ N]	-3.7×10^9
Gradient energy coefficients		Elastic constants	
G_{11} [C ⁻² m ⁴ N]	3.4504×10^{-10}	c_{11} [m ⁻² N]	1.746×10^{11}
G_{12} [C ⁻² m ⁴ N]	0.0	c_{12} [m ⁻² N]	0.7937×10^{11}
G_{44} [C ⁻² m ⁴ N]	1.7252×10^{-10}	c_{44} [m ⁻² N]	1.111×10^{11}
Electrostrictive coefficients		Background permittivity	
Q_{11} [C ⁻² m ⁴]	8.9×10^{-2}	ε_b	66
Q_{12} [C ⁻² m ⁴]	-2.6×10^{-2}	Screening factor	
Q_{44} [C ⁻² m ⁴]	3.375×10^{-2}	θ	0.2

5.2.2 Simulation model and procedure

Polar skyrmions have been directly observed in PbTiO₃/SrTiO₃ superlattices [2,24], and bubble-like domains with similar shapes have been observed on PbTiO₃ nanofilms [25]. It has also been predicted analytically that a skyrmion perfect lattice can be realized on PbTiO₃

nanofilms by adjusting the electrical boundary conditions of the surface [37]. Based on these previous studies, I focus on the skyrmion lattice and skyrmion dislocations on PbTiO_3 nanofilms.

Figure 5-1(a) shows the unit lattice of a skyrmion triangular lattice. The unit lattice has length a in the x_1 direction and length $\sqrt{3}a$ in the x_2 direction. Since the lattice constant (distance between skyrmions) depends on the electric field and temperature, $a(T, E)$ is obtained by preliminary simulations of a unit lattice in the perfect triangular lattice as shown in Table 5-2 and Table 5-3. The thin film model shown in Figure 5-1(b) consists of 28 polar skyrmion lattices in the x_1 direction and 17 in the x_2 direction. The size of the model is $28a$ [nm] $\times$ $17\sqrt{3}a$ [nm] $\times$ 20 [nm], and periodic boundary conditions are applied in the x_1 , x_2 , x_3 directions. The thickness of the thin film is 8 nm, and vacuum layer is placed in the x_3 direction to prevent interplay with adjacent image cells. For numerical calculations, the entire model is divided equally in the x_1 , x_2 , x_3 directions with grid length of approximately 1 nm. To reproduce the misfit strain caused by difference of lattice parameter between film and substrate, an in-plane compressive strain of -1% is applied in the x_1 and x_2 directions[37].

As an initial state, a skyrmion lattice including for dislocation cores symmetrically is placed referencing quadrupole model used in atomic systems [57–59]. This arrangement holds geometrical consistency of the model under periodical boundary conditions and cancels the effect of the strain field by adjacent dislocations in the vicinity of each dislocation core, allowing the evaluation of isolated dislocation core. The coordinates of skyrmions in quadrupole configuration are obtained referencing molecular static (MS) simulations [60]. The TDGL equation is numerically solved using Semi-Implicit Fourier-Spectral method [61,62] with

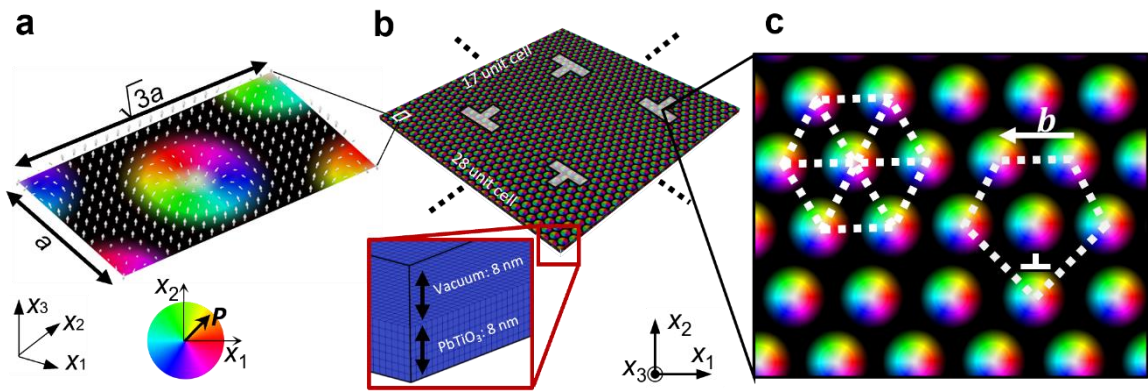


Figure 5-1 (a) Unit triangular lattice of skyrmion lattice. (b) Simulation model of quadrupole configuration under periodic boundary conditions and mesh divisions for calculation. (c) Skyrmion configuration near the dislocation core in the initial state. The white arrow represents a burgers vector of the dislocation.

normalized timestep $\Delta t = 0.04$, and mechanical equilibrium equation and Maxwell's equations are solved simultaneously by FSIPM (Fourier spectral iterative perturbation method) [63,64] to calculate the time evolution of the polarization. The out-of-plane electric field is applied in the 3-12 MV/cm, which is the range of skyrmion phase maintained. Note that this range is very sensitive to material parameters and conditions. The effect of the electric field change is expected to remain qualitatively constant.

Table 5-2 Lattice constants of polar SkX at $T = 300$ K.

E [MV/cm]	3	6	9	12	15
a [nm]	15.8	15.4	15.0	14.6	14.2

Table 5-3 Lattice constants of polar SkX at $E = 6$ MV/cm.

T [K]	100	200	300	400	500
a [nm]	16.6	16.1	15.4	14.7	13.8

5.3 Results and Discussion

5.3.1 Quasiparticle-specific core structure of skyrmion dislocations

Figure 5-2(a)(b) show the detailed polarization configuration and skyrmion structure near the dislocation core at different out-of-plane external electric field $E = 12, 9$, and 6 MV/cm and constant temperature $T=300$ K. As shown in the three-dimensional(3D) view of Figure 5-2(b), these skyrmions are the Néel type and they are stacked in the film thickness direction forming skyrmion bubbles, which is in good agreement with the previous studies [25,37]. As shown in Figure 5-2(a-1), at $E = 12$ MV/cm, circular skyrmions are regularly arranged to form a triangular lattice, except in the center of the figure where 5-fold and 7-fold skyrmions are present with lattice mismatch. This is a typical edge dislocation structure in a conventional two-dimensional triangular lattice, called a 5-7 pair [65]. When the electric field is decreased to $E = 9$ MV/cm, as shown in Figure 5-2(a-2), each skyrmion slightly deforms. Particularly, the 7-fold skyrmion is elongated along the x_1 -direction, resulting in an elliptical shape. At an even lower electric field of $E = 6$ MV/cm [Figure 5-2(a-3)], each skyrmion deforms even larger, and the 7-fold skyrmion is elongated significantly by 56% in the x_1 -direction and compressed by 18% in the x_2 -direction. On the other hand, the 5-fold skyrmion is compressed by 10% in both the x_1

and x_2 directions. Other skyrmions around the dislocation core also deform into elliptical shapes with various directions, while skyrmions far from the dislocation core deform slightly.

To discuss the detailed geometrical features of these enlarged skyrmions, I calculate a Pontryagin charge density $\mathbf{P}(\partial\mathbf{P}/\partial x_1 \times \partial\mathbf{P}/\partial x_2)$, where $\mathbf{P}$ is the normalized polarization vector. The summation of Pontryagin charge density is an invariant of the topological texture (i.e., skyrmion number), and it is used to identify the topological characteristics of domain

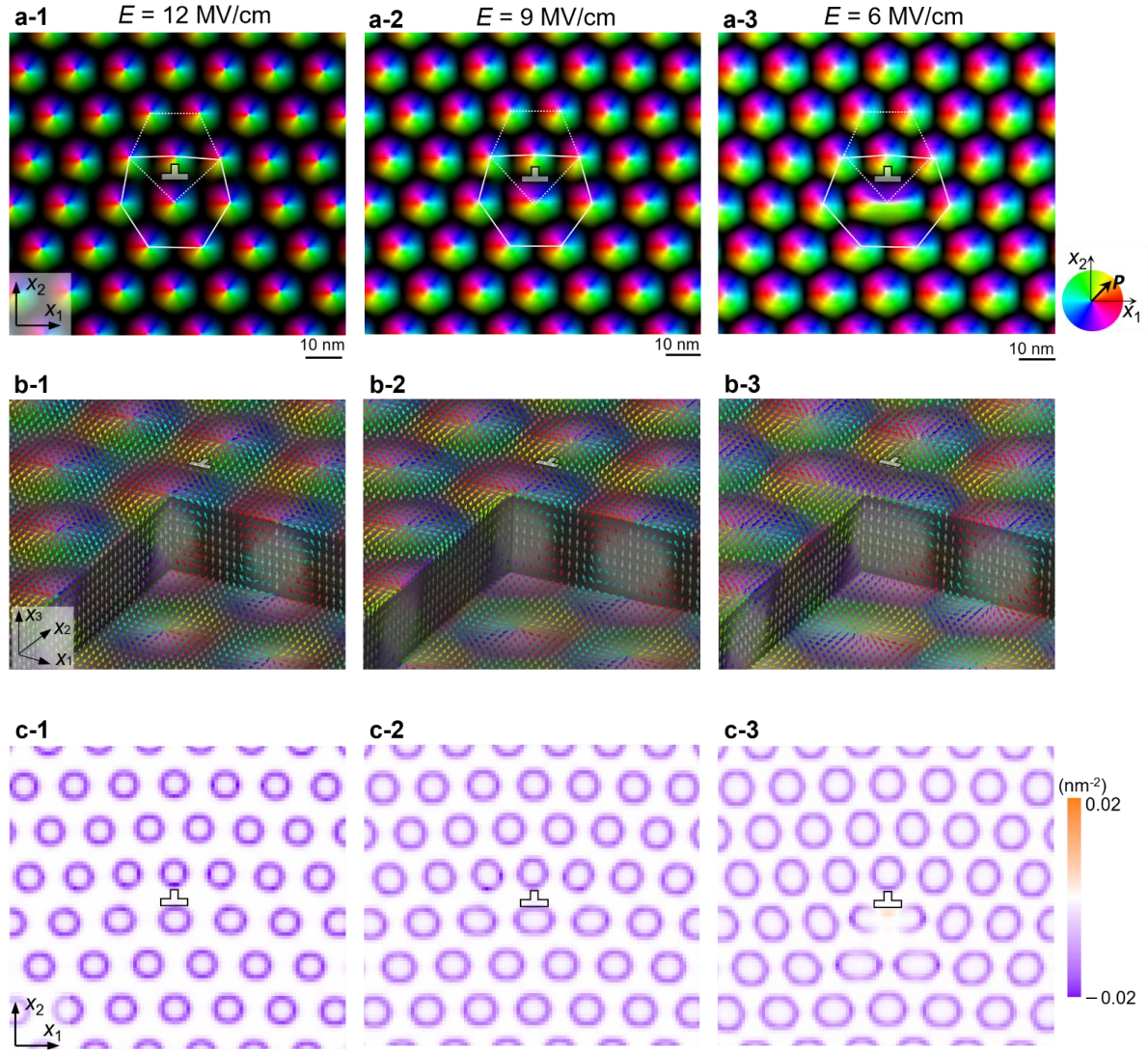


Figure 5-2 Polarization distribution in (a) planer view (b) 3D view, distribution near the dislocation core under different electric fields $E = 12, 9$, and 6 MV/cm . The black area represents out-of-plane polarization and colored circular regions represent in-plane polarization forming skyrmions. The white dashed lines indicate 5-fold and 7-fold skyrmion lattices that comprise the dislocation. (c) Pontryagin charge density distribution in the same area as (a).

structures [66]. As shown in Figure 5-2(c), the Pontryagin charge density is concentrated at the edge of skyrmions, even in enlarged skyrmions. However, the most enlarged (core-site) skyrmion at $E = 6$ MV/cm [Figure 5-2(c-3)], has a different distribution from others. The Pontryagin charge density is concentrated at the right and left sides of the skyrmion, with slightly positive values in the middle, while the overall amount of topological charge remains unchanged. In other words, this skyrmion is splitting into two skyrmions, while it still remains a single skyrmion. Considering that the polar skyrmions in PbTiO_3 thin film become circular shapes forming a perfect triangular lattice [37], the dislocation leads to substantial deformations of the skyrmions, and the deformation increases with a lower electric field. Furthermore, the skyrmions change not only their shapes but also their positions depending on the electric field. At $E = 12$ MV/cm, the positions of skyrmions do not change much from the initial positions of conventional dislocation, while at $E = 6$ MV/cm the skyrmions around the dislocation core move largely. Particularly, at $E = 6$ MV/cm, the skyrmions on the left and right sides of the 7-fold skyrmion spread out significantly with the large deformation in the x_1 -direction of the center 7-fold skyrmion. In other words, under high electric field conditions, the skyrmions slightly deform and the dislocation keeps a conventional 5-7 pair structure, while under lower electric field conditions, the skyrmions around the dislocation core, especially the 7-fold skyrmion, significantly deform and change their positions, resulting in unique dislocation structure.

In addition to the effect of electric fields, temperature also invokes changes in the dislocation structure with the deformation of skyrmions. The polarization distribution at $E = 6$ MV/cm, $T = 500, 300$, and 100 K near the dislocation core are shown in Figure 5-3(a). At high temperature of $T = 500$ K shown in Figure 5-3(a-1), the size of skyrmions is small and no deformations without the vicinity of dislocation core. This is similar to that of high electric field condition, shown in Figure 5-2 (a-1). On the other hand, at low temperature as shown in Figure 5-3(a-3), skyrmion are large and deformed, particularly in the vicinity of dislocation core. Figure 5-3(b) shows the polarization magnitude distribution, indicating that the polarization tends to be larger in low temperature and smaller in high temperature, reflecting the effect of Landau energy describing the temperature-dependent ferroelectric-paraelectric phase transition. This result suggests that at high temperature polarization magnitude is relatively small and effect of electric field is large, resulting the similar dislocation structure as that under high electric field. At low temperature, the effect of electric field is considered to be low due to large polarization magnitude, however, the amount of deformation is as same as in $T = 300$ K. Therefore, the

effects of electric field and temperature on skyrmions are not monotonic, suggesting the existence of intricate mechanisms.

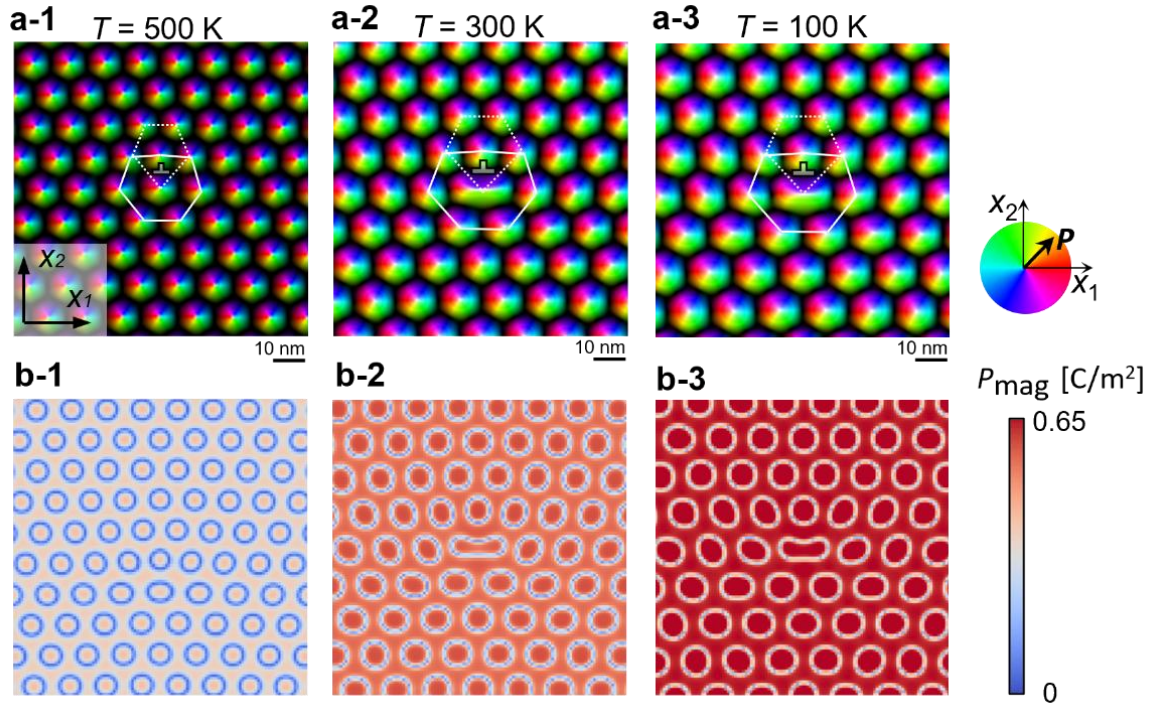


Figure 5-3 (a) Distribution of polarization direction near the dislocation core at $E = 6$ MV/cm, $T = 500$, 300, and 100 K. The black area represents out-of-plane polarization and colored circular regions represent in-plane polarization forming skyrmions. The white dashed lines indicate 5-fold and 7-fold skyrmion lattices that comprise the dislocation. (b) Distribution of polarization magnitude in the same area as (a).

5.3.2 Inherent strain field and breakdown of Volterra's elasticity theory

To deeply understand the unique feature of the dislocation structure in skyrmion lattices, here I focus on an inherent elastic field of dislocation. According to Volterra's elasticity theory of dislocations [49], the elastic field of a dislocation, which is introduced the inherent lattice mismatch of dislocation, is given by $1/r$ -law (r is the distance from a dislocation core). This elastic field of dislocation is of central importance because it determines the mechanics, dynamics, and interaction of dislocations, which lead to plastic deformation of a material. To precisely investigate the elastic features of SkX dislocations and study whether Volterra's law is still applicable to the SkX dislocation, the lattice strain as the indicator of lattice deformations,

referencing a perfect hexagonal skyrmion lattice, $\epsilon_{ij}^{\text{lattice}}$ ($i, j = 1, 2$) [67], is evaluated and shown in Figure 5-4. For comparison, Figure 5-4(a-1) also shows the strain field of conventional

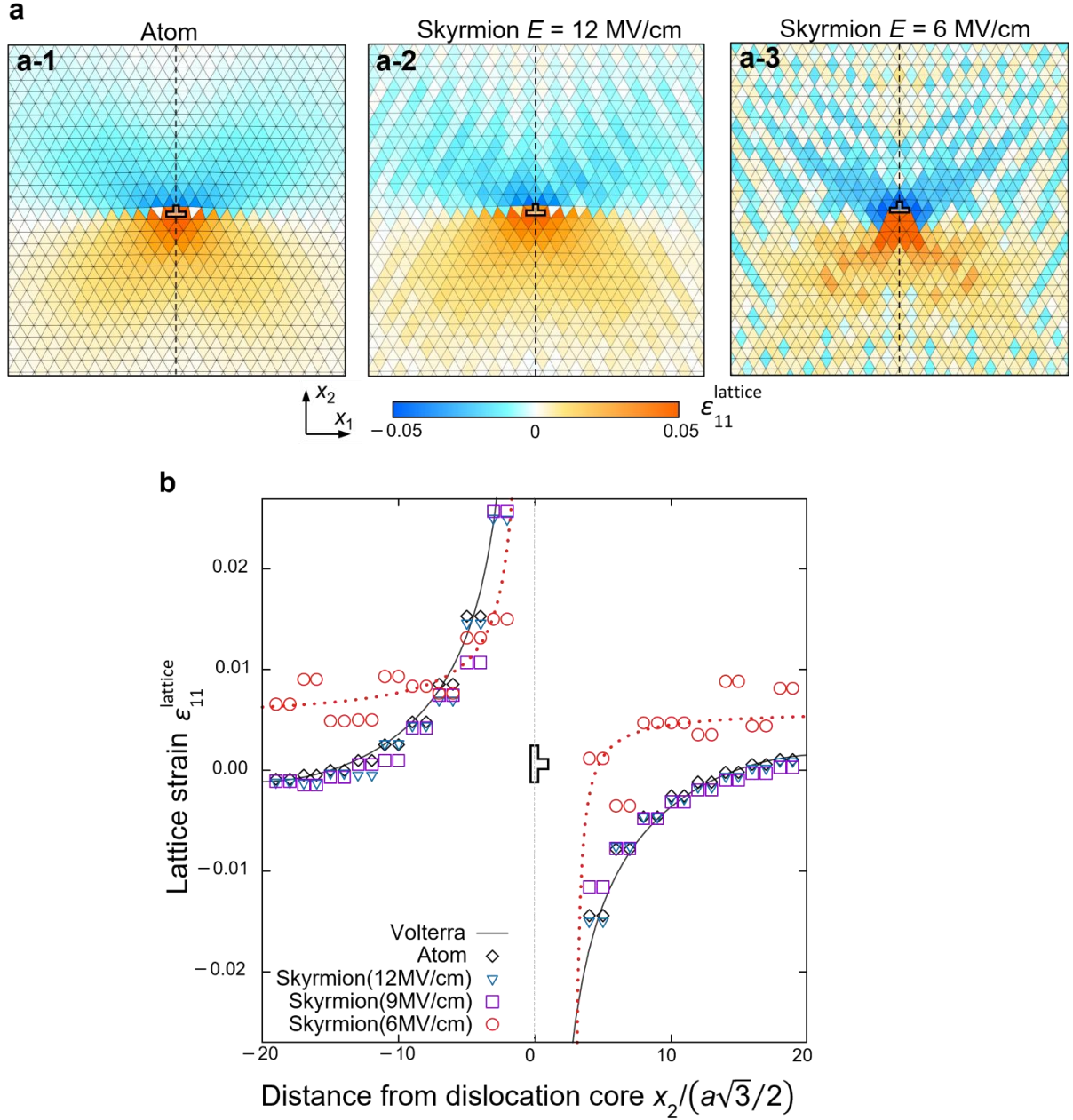


Figure 5-4 (a) Lattice strain distribution around the dislocation of (a-1) atom, (a-2) skyrmion at $E = 12 \text{ MV/cm}$, and (a-3) skyrmion at $E = 6 \text{ MV/cm}$. (b) Lattice strain as a function of the distance from the dislocation core on the centerline across the dislocation core [dashed lines in (a)]. For comparison, the red dotted line shows the function of Volterra's law with fitted to the value of $E = 6 \text{ MV/cm}$. To compare an atomic and skyrmion dislocation, the distance is normalized by the lattice constant a of each system.

dislocation in an atomic crystal, which is obtained from molecular static (MS) calculations. The conventional dislocation in the atomic crystal displays a strain distribution consistent with the $1/r$ -equation in Volterra's elastic theory: concentration at the dislocation core, monotonic and continuous decay with distance from the dislocation core, positive and negative value above and below the dislocation core. In the less deformed SkX dislocation at $E = 12$ MV/cm [Figure 5-4(a-2)], while the detail change of strains is slightly different, the overall strain distribution is almost same as that of atomic crystals and Volterra's law. On the other hand, at $E = 6$ MV/cm [Figure 5-4(a-3)], where skyrmions are much more deformable, local strain concentrations appear in some lattices. Notably, some regions with positive and negative strain values are intermingled, which clearly differs from the strain distribution in the atomic crystals and the SkX at $E = 12$ MV/cm. To compare these strain fields in detail, Figure 5-4 (b) shows the strain distribution along the center line across the dislocation core alongside the theoretical solution of Volterra's law for comparison. The strain distribution of SkX dislocation at $E = 12$ MV/cm agrees well with both that of atomic dislocation and Volterra's law ($1/r$ law), except for the vicinity of the dislocation core where the continuum solution becomes invalid due to the discrete structure of the core. At $E = 9$ MV/cm, the strain field still generally accords with those of atomic dislocations and a theoretical solution, while slight deviations appear. On the other hand, at $E = 6$ MV/cm, the strain rapidly decays with distance from the dislocation core and fluctuates between positive and negative values. As a result, the strain field of SkX dislocation at $E = 6$ MV/cm obviously deviates from that of the atomic crystal and Volterra's law. In other words, the well-known Volterra's law describing conventional dislocations within the framework of elasticity theory is no longer applicable to the polar SkX. Volterra's law is evidence that one dislocation strain field acts on the entire material, indicating the importance of dislocations in the mechanical behavior such as yielding at an extremely low value than ideal strength or hardening due to pinning of dislocations [68]. Therefore, this result suggests not only the existence of the inherent behavior of dislocation but also unique dynamic responses in the SkX. Moreover, since Volterra's law is based on the elastic theory, novel lattice mechanics in the skyrmion lattice may exist, in place of the traditional elastic mechanics.

5.3.3 Quasiparticle-specific lattice mechanics in polar skyrmion lattices

As shown above, such an unusual elastic strain field of SkX dislocation is pronounced under a low electric condition, where skyrmions exhibit a rather soft quasiparticle behavior with large deformation, suggesting that the deformation of the skyrmion is a key to understanding the

lattice strain fields. To discuss this point in detail, here I treat each skyrmion as a particle object and quantify the large deformation of a skyrmion as a “particle strain” based on the finite strain in the continuum mechanics theory [69] as follows (see also Figure 5-5). First, the normalized polarization vectors [positioning at $\mathbf{x}$ in Figure 5-5(a)] in a deformed skyrmion are mapped to the polarization vectors [positioning at $\mathbf{X}$ in Figure 5-5(a)] in a reference (undeformed) skyrmion. The displacement $\mathbf{u}$ of each corresponding polarization can be calculated by $\mathbf{u}(\mathbf{X}) = \mathbf{x} - \mathbf{X}$. Note that, since a polar skyrmion is a topologically characterized continuous polar texture, any normalized polarization vector in a deformed skyrmion is always present in the reference skyrmion. From the displacement fields of each vector, a deformation gradient tensor can be defined as $\mathbf{F}(\mathbf{X}) = \partial \mathbf{x} / \partial \mathbf{X}$ in the skyrmion. Since the deformation of skyrmion can be

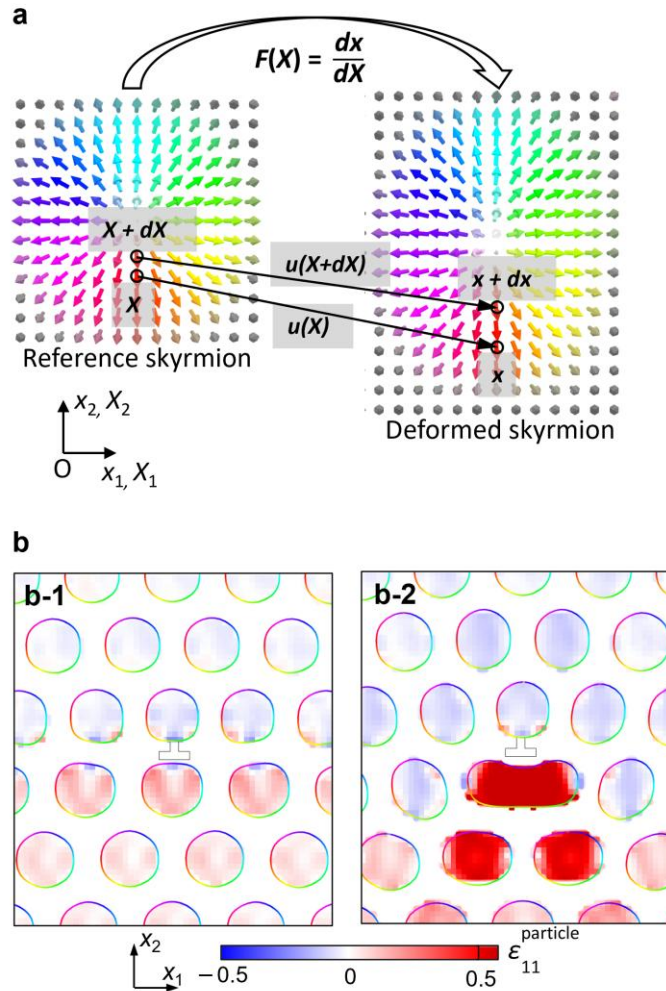


Figure 5-5 (a) Schematic illustration of deformation of skyrmion and definitions of displacements. (b) The distribution of particle strain in the skyrmions near the dislocation core at (b-1) $E = 12$ MV/cm and (b-2) $E = 6$ MV/cm.

large that the infinitesimal deformation assumption does not hold, here I use the Green-Lagrange strain tensor, which is the common finite strain defined as $\mathbf{E} = \frac{1}{2}(\mathbf{F}^T \mathbf{F} - \mathbf{I})$, to evaluate the finite large deformation of skyrmions. Figure 5-5(b) shows the distribution of particle Green-Lagrange strain $\varepsilon_{ij}^{\text{particle}}$ ($i, j = 1, 2$) in the skyrmions near the dislocation core at $E = 12$ and 6 MV/cm. At $E = 12$ MV/cm [Figure 5-5(b-1)], it can be seen that in the vicinity of the dislocation core, a positive and negative particle strain exists below and above the dislocation core, respectively, which is similar to the lattice strain distribution. On the other hand, at $E = 6$ MV/cm [Figure 5-5(b-2)], the skyrmions exhibit large particle strains, particularly the 7-fold skyrmion located below the dislocation core. Strain distribution is not uniform in the highly deformed skyrmion, extremely large value in the middle area, and a small value in the right and left edges. This distribution indicates that only the middle area of the skyrmion is elongated, consistent with the topological feature as shown in Figure 5-2(c-3).

To visualize the distribution of particle strain in the overall skyrmion lattices, the distribution of averaged particle strain within each skyrmion is illustrated in Figure 5-6. At $E = 12$ MV/cm [Figure 5-6(a)], the particle strain is generally small, except in the immediate vicinity of the dislocation core. On the other hand, at $E = 6$ MV/cm [Figure 5-6(b)], skyrmions with large positive and negative particle strains coexist in the entire lattice. Even the particle strain of skyrmions near the dislocation core is the largest, other skyrmions also have large strain. To discuss the relationship between the particle strain distribution and the elastic lattice strain field, Figure 5-6(c)(d) show the particle strain and lattice strain simultaneously. Focus on the region shortly above the dislocation core [Figure 5-6(c-2)(d-2)]. At $E = 12$ MV/cm [Figure 5-6(c-2)], the lattice strains are all negative, consistent with the Volterra's law, while the particle strains are quite small. This suggests that, when the skyrmion behaves like a rigid (undeformable) particle, the lattice follows the elastic law. On the other hand, at $E = 6$ MV/cm [Figure 5-6(d-2)], where the skyrmion is quite deformable and shows a non-trivial particle strain with positive and negative signs, the lattice strains also exhibit both positive and negative values, which are obviously out of the Volterra's law. Furthermore, by comparing the particle strain with the lattice strain at $E = 6$ MV/cm, I find that a lattice with a positive (negative) lattice strain is accompanied with skyrmions with positive (negative) particle strains. This coincidence indicates that a local (positive or negative) lattice strain, which is away from Volterra's law, arises from a local large deformation of skyrmions characteristically observed at the lower electric field condition. The same situation can also be seen at the lower area of the dislocation [Figure 5-6(c-3)(d-3)]; when the particle strain is large, the lattice strain is also large and its sign

coincides. In other words, the lattice strain field of dislocation (especially in the vicinity of the dislocation core) deforms the skyrmion, and each skyrmion deformation changes its local-site lattice strain, resulting in a unique strain distribution that deviates from conventional Volterra's

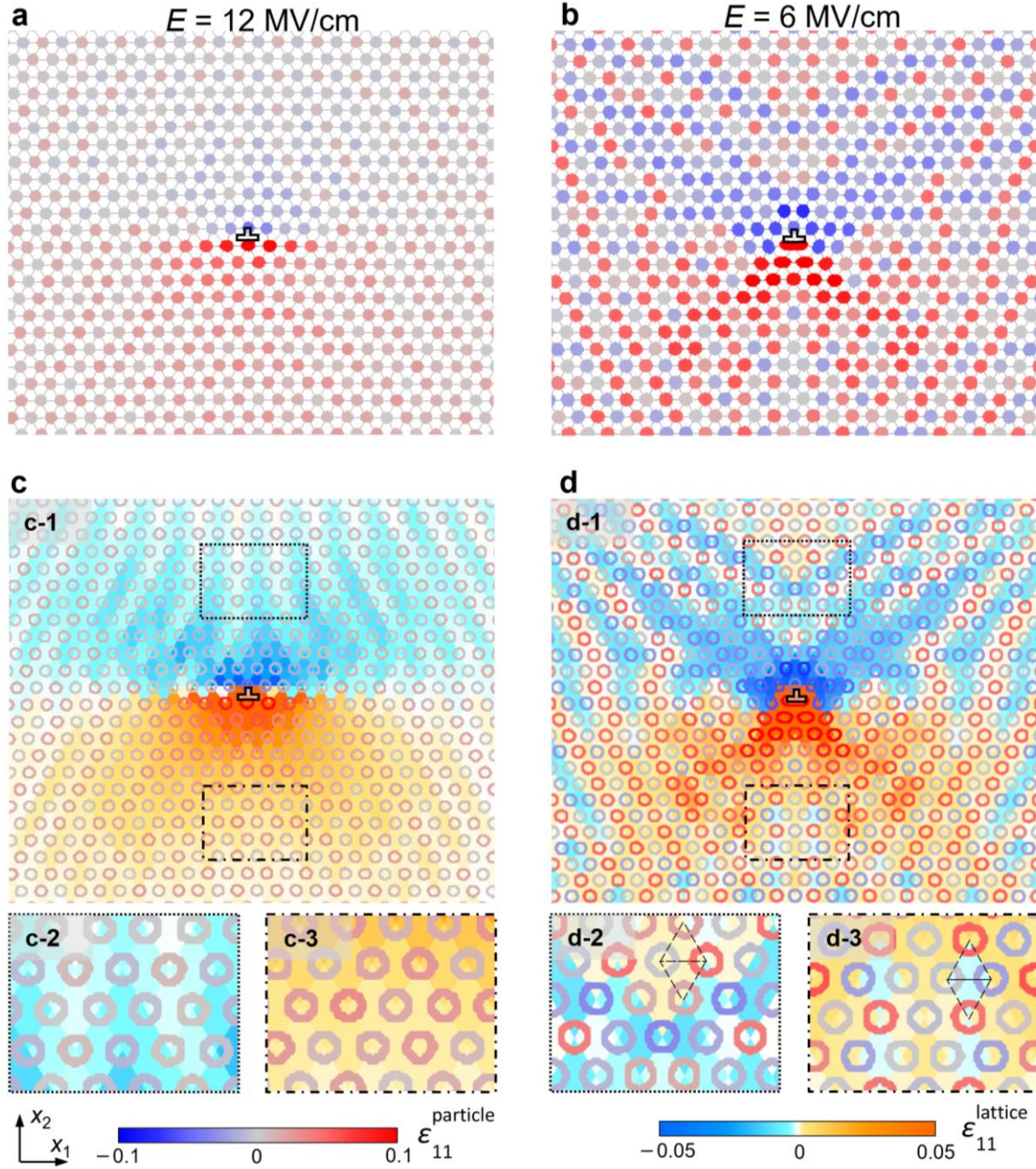


Figure 5-6 Particle strain distribution around the dislocation core at (a) $E = 12$ MV/cm and (b) $E = 6$ MV/cm. Distributions of both particle and lattice strains around the dislocation core at (c) $E = 12$ MV/cm and (d) $E = 6$ MV/cm. The color of circles and rings indicates the particle strain and the color of the fields represents the lattice strain. The dashed triangles in (d-2)(d-3) are example of lattices with different signs of strain from surroundings.

law and the elastic theory.

These results suggest that the deformability of skyrmions breaks the conventional elasticity law in skyrmion lattices and provides novel lattice mechanics involving both deformation and movement of skyrmions. This is in contrast to the conventional mechanics in atomic crystals with only the movement of atoms, which is consistent with the continuous elasticity law. In addition, the deformation of skyrmions and the strain field of dislocation are varied by external fields such as electric field and temperature, indicating the possibility of controlling these unique mechanics. Furthermore, the dislocation structures are significantly different from conventional ones, suggesting that the dynamic behaviors of SkX dislocations and entire skyrmion lattices are different from conventional lattices. Based on these dynamics, the collective quasiparticle behavior of skyrmions may lead to novel electromechanical responses of SkX, which are not present in conventional continuum polarization domains, opening up a new class of lattice mechanics hidden in skyrmion lattices.

5.4 Conclusion

In summary, I have investigated the dislocation structure in the polar skyrmion lattice distinct using phase-field simulations. I found that decreasing the external electric field or temperature causes a transition of skyrmion behavior from rigid particle-like to soft quasiparticle, forming distinct dislocation core structure with 50% significant skyrmion deformation. Furthermore, the strain field of soft skyrmion dislocation deviates from the elastic theory, resulting in the breakdown of conventional Volterra's law of dislocation. A quantitative evaluation of skyrmion deformation based on the continuum mechanics theory visualized the relationships between lattice strain and skyrmion deformation, suggesting that these unique features of skyrmion dislocation arise from inherent mechanics that mitigate the mismatch by both migration and deformation of skyrmion. Our findings not only indicate that skyrmion lattice may possess unique lattice properties, dynamics, and resulting in diverse electrical-mechanical responses, but also present the novel mechanics from a quasiparticle-like nature of skyrmions. Furthermore, from the perspective of the response behavior of skyrmions as topological solitons, this research also contributes to the field of soft solitonic particles. Also, from the viewpoint of two-dimensional phase transitions, the large deformation of skyrmion at dislocation core and the change of "elasticity" of skyrmion lattice may have a new impact on conventional melting process. Therefore, this study opens a new class of lattice mechanics where the quasiparticle properties of skyrmion lattices provide novel responses and functions.

References

- [1] S. Mühlbauer, B. Binz, F. Jonietz, C. Pfleiderer, A. Rosch, A. Neubauer, R. Georgii, and P. Böni, Skyrmion Lattice in a Chiral Magnet, *Science* **323**, 915 (2009).
- [2] S. Das et al., Observation of room-temperature polar skyrmions, *Nature* **568**, 368 (2019).
- [3] Y. Shen, Q. Zhang, P. Shi, L. Du, X. Yuan, and A. V. Zayats, Optical skyrmions and other topological quasiparticles of light, *Nat. Photon.* (2023).
- [4] H. Ge, X.-Y. Xu, L. Liu, R. Xu, Z.-K. Lin, S.-Y. Yu, M. Bao, J.-H. Jiang, M.-H. Lu, and Y.-F. Chen, Observation of Acoustic Skyrmions, *Phys. Rev. Lett.* **127**, 144502 (2021).
- [5] X. Z. Yu, Y. Onose, N. Kanazawa, J. H. Park, J. H. Han, Y. Matsui, N. Nagaosa, and Y. Tokura, Real-space observation of a two-dimensional skyrmion crystal, *Nature* **465**, 901 (2010).
- [6] B. Göbel, I. Mertig, and O. A. Tretiakov, Beyond skyrmions: Review and perspectives of alternative magnetic quasiparticles, *Physics Reports* **895**, 1 (2021).
- [7] A. Fert, V. Cros, and J. Sampaio, Skyrmions on the track, *Nature Nanotech* **8**, 152 (2013).
- [8] J. Sampaio, V. Cros, S. Rohart, A. Thiaville, and A. Fert, Nucleation, stability and current-induced motion of isolated magnetic skyrmions in nanostructures, *Nature Nanotech* **8**, 839 (2013).
- [9] C. H. Marrows, J. Barker, T. A. Moore, and T. Moorsom, Neuromorphic computing with spintronics, *Npj Spintronics* **2**, 12 (2024).
- [10] J. Kalin, S. Sievers, H. W. Schumacher, R. Abram, H. Fuser, M. Bieler, D. Kalin, A. Bauer, and C. Pfleiderer, Optical creation and annihilation of skyrmion patches in a chiral magnet, *Phys. Rev. Applied* **21**, 034065 (2024).
- [11] P. F. Bessarab, D. Yudin, D. R. Gulevich, P. Wadley, M. Titov, and O. A. Tretiakov, Stability and lifetime of antiferromagnetic skyrmions, *Phys. Rev. B* **99**, 140411 (2019).
- [12] Y. Liu, G. Yin, J. Zang, J. Shi, and R. K. Lake, Skyrmion creation and annihilation by spin waves, *Applied Physics Letters* **107**, 152411 (2015).
- [13] Y. Tchoe and J. H. Han, Skyrmion generation by current, *Phys. Rev. B* **85**, 174416 (2012).
- [14] A. Derras-Chouk, E. M. Chudnovsky, and D. A. Garanin, Thermal collapse of a skyrmion, *Journal of Applied Physics* **126**, 083901 (2019).
- [15] Z. Chen, X. Zhang, Y. Zhou, and Q. Shao, Skyrmion Dynamics in the Presence of Deformation, *Phys. Rev. Applied* **17**, L011002 (2022).

- [16] K. Everschor-Sitte, A. Majumdar, K. Wolk, and D. Meier, *Novel Implementations for Reservoir Computing -- from Spin to Charge*, arXiv:2311.11929.
- [17] T. Yokouchi, S. Sugimoto, B. Rana, S. Seki, N. Ogawa, Y. Shiomi, S. Kasai, and Y. Otani, Pattern recognition with neuromorphic computing using magnetic field-induced dynamics of skyrmions, *Sci. Adv.* **8**, eabq5652 (2022).
- [18] X. Zhang, M. Ezawa, and Y. Zhou, Magnetic skyrmion logic gates: conversion, duplication and merging of skyrmions, *Sci Rep* **5**, 9400 (2015).
- [19] Y. Nahas, S. Prokhorenko, L. Louis, Z. Gui, I. Kornev, and L. Bellaiche, Discovery of stable skyrmionic state in ferroelectric nanocomposites, *Nat Commun* **6**, 8542 (2015).
- [20] S. Das et al., Local negative permittivity and topological phase transition in polar skyrmions, *Nat. Mater.* **20**, 194 (2021).
- [21] L. Han et al., High-density switchable skyrmion-like polar nanodomains integrated on silicon, *Nature* **603**, 7899 (2022).
- [22] F.-H. Gong, Y.-L. Tang, Y.-J. Wang, Y.-T. Chen, B. Wu, L.-X. Yang, Y.-L. Zhu, and X.-L. Ma, Absence of critical thickness for polar skyrmions with breaking the Kittel's law, *Nat Commun* **14**, 3376 (2023).
- [23] S. Prokhorenko, Y. Nahas, Q. Zhang, V. Govinden, N. Valanoor, and L. Bellaiche, *Motion and Teleportation of Polar Bubbles in Ultra-Thin Ferroelectrics*, arXiv:2303.13883.
- [24] Y.-T. Shao et al., Emergent chirality in a polar meron to skyrmion phase transition., *Nat Commun* **14**, 1355 (2023).
- [25] X. Zhang et al., Creation and erasure of polar bubble domains in PbTiO_3 films by mechanical stress and light illuminations, *Journal of Materiomics* **9**, 626 (2023).
- [26] T. Xu, Y. Ichiki, K. Masuda, Y. Wang, H. Hirakata, and T. Shimada, Ultrasmall Polar Skyrmions and Merons in SrTiO_3 Heterostructures by Polaron Engineering, *ACS Nano* **17**, 10836 (2023).
- [27] L. Gao, S. Prokhorenko, Y. Nahas, and L. Bellaiche, Dynamical Control of Topology in Polar Skyrmions via Twisted Light, *Phys. Rev. Lett.* **132**, 026902 (2024).
- [28] C. Jeong et al., Revealing the three-dimensional arrangement of polar topology in nanoparticles, *Nat Commun* **15**, 3887 (2024).
- [29] S. Minami, Y. Ikeda, and T. Shimada, Spontaneous Atomic-Scale Polar Skyrmions and Merons on a SrTiO_3 (001) Surface: Defect Engineering for Emerging Topological Orders, *Nano Lett.* **24**, 3686 (2024).

-
- [30] A. Lipatov, T. Li, N. S. Vorobeva, A. Sinitskii, and A. Gruverman, Nanodomain Engineering for Programmable Ferroelectric Devices, *Nano Lett.* **19**, 3194 (2019).
- [31] Y. Ren, H. Cheng, J. Ouyang, O. Kurt, J. Wang, Q. Zhang, Y. Zhao, L. Gu, and L.-Q. Chen, Bimodal polymorphic nanodomains in ferroelectric films for giant energy storage, *Energy Storage Materials* **48**, 306 (2022).
- [32] Z. Chen et al., All-ferroelectric implementation of reservoir computing, *Nat Commun* **14**, 3585 (2023).
- [33] Q. Li et al., Giant Electric Field-Induced Second Harmonic Generation in Polar Skyrmions, preprint, In Review, 2023.
- [34] M. Nitta, Relations among topological solitons, *Phys. Rev. D* **105**, 105006 (2022).
- [35] Q. Zhang, P. J. Ackerman, Q. Liu, and I. I. Smalyukh, Ferromagnetic Switching of Knotted Vector Fields in Liquid Crystal Colloids, *Phys. Rev. Lett.* **115**, 097802 (2015).
- [36] J.-S. B. Tai and I. I. Smalyukh, Three-dimensional crystals of adaptive knots, *Science* **365**, 1449 (2019).
- [37] S. Yuan, Z. Chen, S. Prokhorenko, Y. Nahas, L. Bellaiche, C. Liu, B. Xu, and L. Chen, Hexagonal Close-packed Skyrmion Lattice in Ultrathin Ferroelectric PbTiO_3 Films, *Physical Review Letters* **130**, 226801 (2023).
- [38] K. Shibata et al., Large anisotropic deformation of skyrmions in strained crystal, *Nature Nanotech* **10**, 589 (2015).
- [39] X. Yu, D. Morikawa, T. Yokouchi, K. Shibata, N. Kanazawa, F. Kagawa, T. Arima, and Y. Tokura, Aggregation and collapse dynamics of skyrmions in a non-equilibrium state, *Nature Phys* **14**, 832 (2018).
- [40] S. Pöllath et al., Dynamical Defects in Rotating Magnetic Skyrmion Lattices, *Phys. Rev. Lett.* **118**, 207205 (2017).
- [41] R. Brearton, M. W. Olszewski, S. Zhang, M. R. Eskildsen, C. Reichhardt, C. J. O. Reichhardt, G. Van Der Laan, and T. Hesjedal, Skyrmions in anisotropic magnetic fields: strain and defect driven dynamics, *MRS Advances* **4**, 643 (2019).
- [42] T. Schönenberger, P. Huang, L. D. Brun, L. Guanhao, A. Magrez, F. Carbone, and H. M. Rønnow, Direct Visualisation of Skyrmion Lattice Defect Alignment at Grain Boundaries, *Nanoscale Res Lett* **17**, 20 (2022).
- [43] T. Matsumoto, Y.-G. So, Y. Kohno, H. Sawada, R. Ishikawa, Y. Ikuhara, and N. Shibata, Jointed magnetic skyrmion lattices at a small-angle grain boundary directly visualized by advanced electron microscopy, *Sci Rep* **6**, 1 (2016).

- [44] P. Huang, T. Schönenberger, M. Cantoni, L. Heinen, A. Magrez, A. Rosch, F. Carbone, and H. M. Rønnow, Melting of a skyrmion lattice to a skyrmion liquid via a hexatic phase, *Nat. Nanotechnol.* **15**, 761 (2020).
- [45] C. Timm, S. M. Girvin, and H. A. Fertig, Skyrmion lattice melting in the quantum Hall system, *Phys. Rev. B* **58**, 10634 (1998).
- [46] P. Baláž, M. Paściak, and J. Hlinka, Melting of Néel skyrmion lattice, *Phys. Rev. B* **103**, 174411 (2021).
- [47] D. R. Nelson and B. I. Halperin, Dislocation-mediated melting in two dimensions, *Phys. Rev. B* **19**, 2457 (1979).
- [48] Z. Wang and L.-Q. Chen, Reversible Phase Transition between Vortex Lattice and Hexagonal Polar Skyrmion Crystals, *Nano Lett.* **23**, 9907 (2023).
- [49] V. Volterra, Sur l'équilibre des corps élastiques multiplement connexes, *Ann. Sci. École Norm. Sup.* **24**, 401 (1907).
- [50] D. Kuhlmann-Wilsdorf, Theory of plastic deformation: - properties of low energy dislocation structures, *Materials Science and Engineering: A* **113**, 1 (1989).
- [51] G. Sheng, Y. L. Li, J. X. Zhang, S. Choudhury, Q. X. Jia, V. Gopalan, D. G. Schlom, Z. K. Liu, and L. Q. Chen, A modified Landau-Devonshire thermodynamic potential for strontium titanate, *Applied Physics Letters* **96**, 108 (2010).
- [52] Y. L. Li, S. Y. Hu, Z. K. Liu, and L. Q. Chen, Effect of substrate constraint on the stability and evolution of ferroelectric domain structures in thin films, *Acta Materialia* **50**, 395 (2002).
- [53] C. H. Woo and Y. Zheng, Depolarization in modeling nano-scale ferroelectrics using the Landau free energy functional, *Appl. Phys. A* **91**, 59 (2008).
- [54] S. Yuan, Z. Chen, S. Prokhorenko, Y. Nahas, L. Bellaiche, C. Liu, B. Xu, L. Chen, S. Das, and L. W. Martin, Hexagonal Close-Packed Polar-Skyrmion Lattice in Ultrathin Ferroelectric PbTiO₃ Films, *Phys. Rev. Lett.* **130**, 226801 (2023).
- [55] C. M. Wu, W. J. Chen, D. C. Ma, C. H. Woo, and Y. Zheng, Effects of the surface charge screening and temperature on the vortex domain patterns of ferroelectric nanodots, *Journal of Applied Physics* **112**, 104108 (2012).
- [56] M. J. Haun, E. Furman, S. J. Jang, H. A. McKinstry, and L. E. Cross, Thermodynamic theory of PbTiO₃, *Journal of Applied Physics* **62**, 3331 (1987).
- [57] J. R. K. Bigger, D. A. McInnes, A. P. Sutton, M. C. Payne, I. Stich, R. D. King-Smith, D. M. Bird, and L. J. Clarke, Atomic and electronic structures of the 90° partial dislocation in silicon, *Phys. Rev. Lett.* **69**, 2224 (1992).

-
- [58] H. S. Seung and D. R. Nelson, Defects in flexible membranes with crystalline order, *Phys. Rev. A* **38**, 1005 (1988).
- [59] S. K. Yadav, R. Ramprasad, A. Misra, and X.-Y. Liu, Core structure and Peierls stress of edge and screw dislocations in TiN: A density functional theory study, *Acta Materialia* **74**, 268 (2014).
- [60] M. Upmanyu, R. W. Smith, and D. J. Srolovitz, Atomistic Simulation of Curvature Driven Grain Boundary Migration, (1998).
- [61] L. Q. Chen and J. Shen, Applications of semi-implicit Fourier-spectral method to phase field equations, *Computer Physics Communications* **108**, 147 (1998).
- [62] H.-L. Hu and L.-Q. Chen, Three-Dimensional Computer Simulation of Ferroelectric Domain Formation, *Journal of the American Ceramic Society* **81**, 492 (2005).
- [63] J. J. Wang, S. Bhattacharyya, Q. Li, T. W. Heo, X. Q. Ma, and L.-Q. Chen, Elastic solutions with arbitrary elastic inhomogeneity and anisotropy, *Philosophical Magazine Letters* **1** (2012).
- [64] J. J. Wang, X. Q. Ma, Q. Li, J. Britson, and L.-Q. Chen, Phase transitions and domain structures of ferroelectric nanoparticles: Phase field model incorporating strong elastic and dielectric inhomogeneity, *Acta Materialia* **61**, 7591 (2013).
- [65] Y.-J. Lai and L. I, Packings and defects of strongly coupled two-dimensional Coulomb clusters: Numerical simulation, *Phys. Rev. E* **60**, 4743 (1999).
- [66] N. Nagaosa and Y. Tokura, Topological properties and dynamics of magnetic skyrmions, *Nature Nanotech* **8**, 899 (2013).
- [67] A. Stukowski, Computational Analysis Methods in Atomistic Modeling of Crystals, *JOM* **66**, 399 (2014).
- [68] P. M. Anderson, J. P. Hirth, and J. Lothe, *Theory of Dislocations*, 2017 edition (Cambridge University Press, Cambridge, 2017).
- [69] K. Bathe, E. Ramm, and E. L. Wilson, Finite element formulations for large deformation dynamic analysis, *Numerical Meth Engineering* **9**, 353 (1975).

Chapter 6 Grain boundary structures in emergent polar skyrmion lattices

6.1 Introduction

In Chapter 5, I revealed structural properties of dislocations in polar skyrmion lattice. Higher-order lattice defects are also expected to exhibit quasiparticle-specific characteristics. This chapter focuses on grain boundaries, which are complex defects composed of multiple aligned dislocations.

Skyrmions are novel topological orders arising from the arrangement of electric polarization in ferroelectrics [1,2], spin configurations in magnetic materials [3,4], or other physical vectors in various systems [5,6]. Due to the topological protection, skyrmions exist independently and stably [7]. Despite being organized as a continuous vector field, skyrmions behave as individual particles and are treated as quasiparticles. Since the discovery of magnetic skyrmions in 2009 [3], they have attracted significant attention due to their exotic physical properties, such as the topological Hall effect [8], and their potential applications as racetrack memory [9], logic devices [10,11], image recognition elements [12], and neuromorphic computing devices [13]. While extensive pioneering research has been conducted on magnetic skyrmions, an increasing number of studies have also focused on polar skyrmions [14–16]. Notably, polar skyrmions exhibit unique properties not observed in magnetic skyrmions, including high-density configurations for memory applications [17], local negative permittivity [18], and optical response properties [19]. These distinctive features of polar skyrmions make them promising candidates for electric devices and other advanced engineering applications.

Since skyrmions generally arrange in large numbers on a two-dimensional plane to form a lattice [4,20,21], not only on individual skyrmions but also on the collective properties of skyrmion lattices have been focused on. Similar to conventional atomic crystals, skyrmion lattices include lattice defects such as vacancies [22], dislocations [23,24], and grain boundaries [25]. Among them, grain boundaries are particularly important, as they influence the overall lattice structure and dynamics.

Grain boundaries are the typical planar lattice defects that form when two crystal grains with different orientations meet during crystal growth. In general, grain boundaries play a crucial role in plastic deformation by hindering or releasing dislocations and affecting their mobility [26,27]. Additionally, grain boundaries behave as source of other lattice defects [28,29]. As a result, the grain boundaries determines the overall mechanical behavior of polycrystalline materials [26,30]. Furthermore, grain boundaries exhibit lattice mismatches, leading to excess grain boundary energy compared to perfect crystals [31,32]. The local disorder of crystalline arrangement near grain boundaries can be diffusion pathways for vacancies and cause of segregation, making them potential initiation sites for material failure [33–36]. Given these characteristics, grain boundaries and their lattice mismatch significantly impact the overall mechanical properties of crystals, making them an important topic in materials engineering [37,38].

In skyrmion lattices, phenomena such as grain boundary-mediated rotation [23] have been reported, highlighting the importance of grain boundaries from a perspective of lattice mechanics. However, in polar skyrmion lattices, even the fundamental properties, such as structure and stabilization mechanisms of grain boundaries remains unknown. A previous study for dislocation [39] indicated that conventional elasticity theories cannot be applied to polar skyrmion lattices. Instead, a unique “quasiparticle lattice mechanics” emerges from the interaction between skyrmion deformation and lattice distortion. Given this, it is expected that grain boundaries in polar skyrmion lattices possess unique structures and stabilization mechanisms compared to those in atomic crystals.

In this chapter, I investigate the stable structure of $\Sigma 7$ grain boundaries in polar skyrmion lattices using phase-field simulations. Our results reveal that polar skyrmion grain boundaries undergo transitions into unique structures involving skyrmion deformation depending on electric fields and temperature conditions. Furthermore, through numerical evaluations of lattice and skyrmion deformation, I show a “quasiparticle-based” mismatch relaxation mechanism.

6.2 Simulation method

In general, grain boundary structures in two-dimensional crystals are classified based on the difference between two crystal orientations, often expressed in terms of Σ values, such as $\Sigma 7$ grain boundaries, etc. Although grain boundaries with various tilt angles have been reported [23,25,40], most of them are composed of 5-7 pair dislocations skyrmion lattices. In this chapter, I focus on the $\Sigma 7$ grain boundary structure as a representative configuration of 5-7

dislocation arrays and an intrinsically stable structure in polar skyrmion lattices. Polar skyrmions were first observed in a $\text{PbTiO}_3/\text{SrTiO}_3$ superlattice [2], and more recently, ferroelectric thin films have emerged as a promising platform exploring for polar skyrmions [17,41,42].

6.2.1 Phase-field modeling

To investigate the stable structures of polar skyrmion grain boundaries, I perform phase-field simulations of polarization distribution in PbTiO_3 thin film, using polarization $\mathbf{P} = (P_1, P_2, P_3)$ as an order parameter. In ferroelectric systems, the total free energy density F is written as

$$F = \int_V (f_{\text{Landau}} + f_{\text{grad}} + f_{\text{electo}} + f_{\text{elast}}) dV \quad (6.1)$$

where f_{Landau} , f_{grad} , f_{elec} , f_{elast} are the Landau energy density, the gradient energy density, the electrostatic energy density, and elastic energy density, respectively. The Landau energy density, describing the ferroelectric phase transition depending on temperature, is expressed by [43,44]

$$f_{\text{Landau}} = \alpha_{ij} P_i P_j + \alpha_{ijkl} P_i P_j P_k P_l + \alpha_{ijklmn} P_i P_j P_k P_l P_m P_n, \quad (6.2)$$

where α_{ij} , α_{ijkl} , and α_{ijklmn} are the Landau coefficients. The gradient energy density, describing the formation of polarization domain walls, is expressed by [43]

$$f_{\text{grad}} = \frac{1}{2} G_{ijkl} P_{i,j} P_{k,l}, \quad (6.3)$$

where G_{ijkl} is the gradient energy coefficient and $P_{i,j} = \frac{\partial P_i}{\partial x_j}$ is the spatial partial derivative of polarization. The electrostatic energy density is expressed by [45]

$$f_{\text{electo}} = -\frac{1}{2} \varepsilon_b E_i E_i - E_i P_i, \quad (6.4)$$

where ε_b is the background permittivity and E_i is electric field. In this chapter, screening factor β is introduced to reproduce the electrical boundary conditions of thin film surface where polar skyrmion appears in the previous study [41]. Thus, electric field is expressed by

$$E_i = E_i^{\text{ext}} + (1 - \beta) E_i^{\text{dep}}, \quad (6.5)$$

where E_i^{ext} is external electric field and E_i^{dep} is depolarization field. The elastic energy density is expressed by

$$f_{\text{elast}} = \frac{1}{2} c_{ijkl} \varepsilon_{ij}^{\text{elast}} \varepsilon_{kl}^{\text{elast}}, \quad (6.6)$$

where c_{ijkl} is the elastic constant and $\varepsilon_{ij}^{\text{elast}}$ is elastic strain. In addition to elastic strain, ferroelectric materials exhibit eigen strain (intrinsic strain) caused by spontaneous polarization. The eigen strain ε_{ij}^0 is expressed by [43]

$$\varepsilon_{ij}^0 = Q_{ijkl}P_kP_l, \quad (6.7)$$

where Q_{ijkl} is electrostrictive coefficients. The total strain ε_{ij} , defined as the displacement gradient in the material, is the sum of elastic strain and eigen strain and expressed by

$$\varepsilon_{ij}^{\text{elast}} = \varepsilon_{ij} - \varepsilon_{ij}^0. \quad (6.8)$$

Substituting Eq. (8), f_{elast} is

$$\begin{aligned} f_{\text{elast}} &= \frac{1}{2} c_{ijkl} \varepsilon_{ij}^{\text{elast}} \varepsilon_{kl}^{\text{elast}} \\ &= \frac{1}{2} c_{ijkl} (\varepsilon_{ij} - \varepsilon_{ij}^0) (\varepsilon_{kl} - \varepsilon_{kl}^0) \\ &= \frac{1}{2} c_{ijkl} \varepsilon_{ij} \varepsilon_{kl} - \frac{1}{2} c_{ijkl} (\varepsilon_{ij} \varepsilon_{kl}^0 + \varepsilon_{ij}^0 \varepsilon_{kl}) + \frac{1}{2} c_{ijkl} \varepsilon_{ij}^0 \varepsilon_{kl}^0. \end{aligned} \quad (6.9)$$

Substituting Eq. (7) into the second term, using $c_{ijkl} = c_{klij}$ in for isotropic materials,

$$\begin{aligned} -\frac{1}{2} c_{ijkl} (\varepsilon_{ij} \varepsilon_{kl}^0 + \varepsilon_{ij}^0 \varepsilon_{kl}) &= -\frac{1}{2} c_{ijkl} (\varepsilon_{ij} Q_{klmn} P_m P_n + Q_{ijmn} P_m P_n \varepsilon_{kl}) \\ &= -\frac{1}{2} (c_{ijmn} + c_{mni j}) Q_{mnkl} \varepsilon_{ij} P_k P_l \\ &= -c_{ijmn} Q_{mnkl} \varepsilon_{ij} P_k P_l \\ &= -q_{ijkl} \varepsilon_{ij} P_k P_l, \end{aligned} \quad (6.10)$$

where $q_{ijkl} (= c_{ijmn} Q_{mnkl})$ is piezoelectric coupling coefficients. And the third term is

$$\begin{aligned} \frac{1}{2} c_{ijkl} \varepsilon_{ij}^0 \varepsilon_{kl}^0 &= \frac{1}{2} c_{ijkl} Q_{ijmn} P_m P_n Q_{klmn} P_m P_n \\ &= \frac{1}{2} c_{mnop} Q_{mni j} Q_{opkl} P_i P_j P_k P_l. \end{aligned} \quad (6.11)$$

The right side of this equation is the fourth-order term of P_i , which can be incorporated into Landau energy density. Therefore, f_{elast} is expressed by

$$f_{\text{elast}} = \frac{1}{2} c_{ijkl} \varepsilon_{ij} \varepsilon_{kl} - q_{ijkl} \varepsilon_{ij} P_k P_l. \quad (6.12)$$

The second term of Eq. (9) describes the piezoelectric coupling effect in ferroelectric materials.

The governing equations of the phase-field simulation consist of the time-dependent Ginzburg-Landau (TDGL) equation, Mechanical equilibrium equation, and Maxwell's equation. TDGL equation is written as

$$\partial P_i / \partial t = -L(\delta F / \delta P_i), \quad (6.13)$$

where t is time, L is the kinetic coefficient and $F = \int f_{\text{total}} dV$ is total free energy of the system.

Mechanical equilibrium equation is

$$\sigma_{ij,j} = (\partial f / \partial \varepsilon_{ij})_{,j} = 0, \quad (6.14)$$

where $\sigma_{ij,j}$ is the stress gradient. Maxwell's equations is

$$D_{i,i} = (-\partial f / \partial E_i)_{,i} = 0, \quad (6.15)$$

where $D_{i,i}$ is the electric displacement gradient in the material. Eqs. (6.13), (6.14), and (6.15) are the governing equations of the phase-field simulation. All of the material parameters used in this chapter are listed in Table 6-1.

Table 6-1 Material parameters for PbTiO₃ [41,46]. T is temperature.

Landau coefficients			
α_1 [C ⁻² m ² N]	$3.8(T - 752) \times 10^5$	α_{111} [C ⁻⁶ m ¹⁰ N]	2.606×10^8
α_{11} [C ⁻⁴ m ⁶ N]	-0.7252×10^8	α_{112} [C ⁻⁶ m ¹⁰ N]	6.1×10^8
α_{12} [C ⁻⁴ m ⁶ N]	7.5×10^8	α_{123} [C ⁻⁶ m ¹⁰ N]	-3.7×10^9
Gradient energy coefficients		Elastic constants	
G_{11} [C ⁻² m ⁴ N]	3.4504×10^{-10}	c_{11} [m ⁻² N]	1.746×10^{11}
G_{12} [C ⁻² m ⁴ N]	0.0	c_{12} [m ⁻² N]	0.7937×10^{11}
G_{44} [C ⁻² m ⁴ N]	1.7252×10^{-10}	c_{44} [m ⁻² N]	1.111×10^{11}
Electrostrictive coefficients		Background permittivity	
Q_{11} [C ⁻² m ⁴]	8.9×10^{-2}	ε_b	66
Q_{12} [C ⁻² m ⁴]	-2.6×10^{-2}	Screening factor	
Q_{44} [C ⁻² m ⁴]	3.375×10^{-2}	β	0.2

6.2.2 Simulation model and procedure

As shown in Figure 6-1(a), the simulation model consists of an 8 nm thick PbTiO₃ film and a 12 nm vacuum layer in the x_3 direction. Assuming the presence of a substrate, a -1% misfit strain is applied to the PbTiO₃ film. The screening factor is set to $\beta = 0.2$. These external factors are selected to ensure that skyrmion phase remains stable over a wide range of temperatures and electric fields based on previous study⁴¹, enabling investigation of grain boundary structures depending on quasiparticle features of skyrmion. The initial polarization distribution in PbTiO₃

film forms two perfect hexagonal skyrmion lattices with different orientations, each having a lattice constant $a(E, T)$, as listed in Table 6-2 and Table 6-3. The lattice is tilted by θ ($= 10.9^\circ$) in the left half of the simulation model and $-\theta$ in the right half, creating a lattice mismatch corresponding to a $\Sigma 7$ grain boundary at the center and the edge of the model under 3D periodic boundary conditions (Figure 6-1(b)). The $\Sigma 7$ grain boundary structure, characterized by straightly aligned dislocations and commonly observed in magnetic skyrmion lattices, is adopted as the reference structure in this chapter. To minimize interactions between grain boundaries, I decided on the model size and initial polarization distribution so that 14 skyrmions aligned between the grain boundaries. To account for the free volume of grain boundaries, multiple simulations were performed by varying the model length in the x_1 direction, and the most stable grain boundary structure with the lowest total free energy was determined. The model length in the x_2 direction is set to match the repeating units under periodical boundary conditions. As a result, the actual model length in x_1 and x_2 directions are $(l_1, l_2) = (388.6 \text{ nm}, 38.5 \text{ nm})$, $(416.1 \text{ nm}, 39.7 \text{ nm})$, and $(436.3 \text{ nm}, 41.0 \text{ nm})$ for $E = 12, 9$, and 6 MV/cm , respectively. The entire simulation model is divided into grids of approximately 1 nm width. Eqs. (6.13)~(6.15) are numerically solved using Semi-Implicit Fourier-Spectral method [47,48] and Fourier spectral iterative perturbation method [49,50], under varied out-of-plane electric

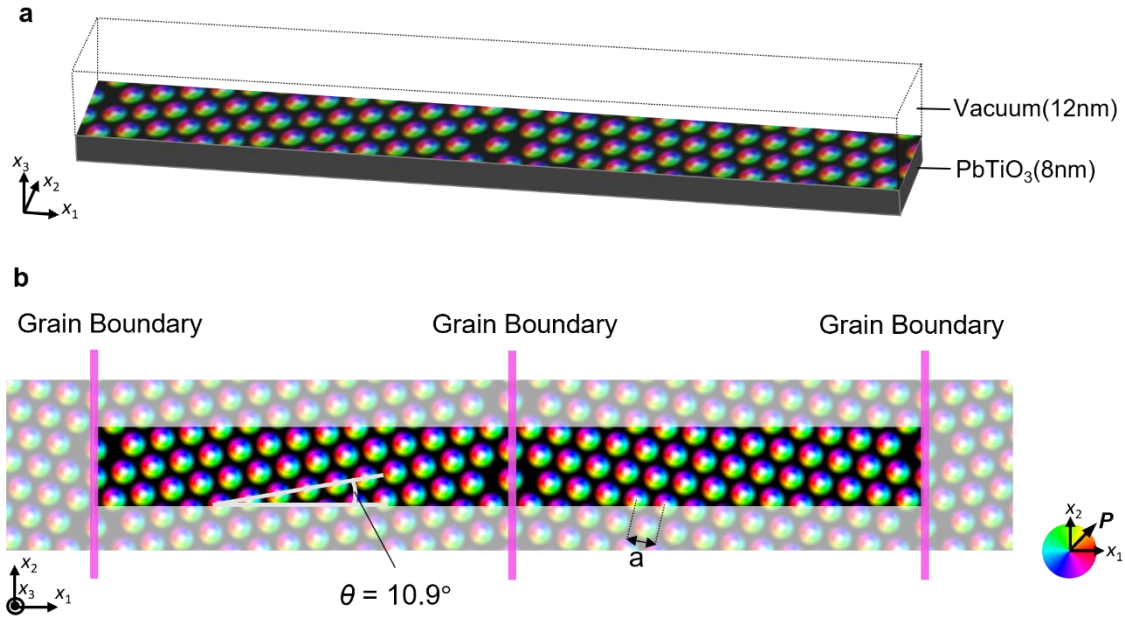


Figure 6-1 Schematic of simulation model in (a) 3D view and (b) 2D view. The pink lines represent the positions of the skyrmion grain boundaries. Light-colored areas represent the image cells under periodic boundary conditions.

field and temperature conditions. This method has been successfully used in previous studies to reproduce the emergence of polar skyrmions and skyrmion lattices [39,51].

Table 6-2 Lattice constants of polar SkX at $T = 300$ K.

E [MV/cm]	3	6	9	12
a [nm]	15.8	15.4	15.0	14.6

Table 6-3 Lattice constants of polar SkX at $E = 6$ MV/cm.

T [K]	100	300	500
a [nm]	16.6	15.4	13.8

6.3 Results and discussion

6.3.1 Skyrmion Grain Boundary Structure under High Electric Field (12 MV/cm)

Figure 6-2(a-1), (b-1), and (c-1) illustrate the three-dimensional polarization structures near the skyrmion grain boundary under electric fields of $E = 12$, 9, and 6 MV/cm, respectively. These figures show that regions with downward out-of-plane polarization exist within a surrounding upward out-of-plane region, with in-plane polarization at the intermediate transitions. Notably, near the upper surface, the polarization vectors continuously converge toward the skyrmion center, while near the lower surface, they diverge outward. This three-dimensional polar structure is characteristic of a skyrmion bubble, previously observed in $\text{PbTiO}_3/\text{SrTiO}_3$ superlattices [2]. In addition, the skyrmions have Néel type domain wall, consistent with other theoretical and experimental studies [17,41,52]. As shown in Figure 6-2(a-2), under a high electric field ($E = 12$ MV/cm), each skyrmion has circular shape without significant deformation, with an approximate diameter of 10 nm. At the grain boundary, 5-fold and 7-fold coordinated skyrmions are present and are arranged parallel to the grain boundary plane. This configuration is structurally analogous to a conventional $\Sigma 7$ grain boundary, where 5-7 pairs align sequentially. A notable aspect of this result is that the vector order polar skyrmions behave as discrete particles, forming a grain boundary structure similar to that observed in atomic crystals. To characterize the topological nature of the skyrmions, I calculated the Pontryagin charge density [53] q ,

$$q = \frac{1}{4\pi} \int_S \mathbf{P} \cdot \left(\frac{\partial \mathbf{P}}{\partial x} \times \frac{\partial \mathbf{P}}{\partial y} \right) d^2r \quad (6.16)$$

where $\mathbf{P}$ is the normalized polarization vector, and its distribution is shown in Figure 6-2(a-3). The negative q values form a ring-based pattern around the outer edges of each skyrmion, consistent with previous studies [2] on polar skyrmions. The same distribution is found for other skyrmions, indicating that they are topologically equivalent. Furthermore, to compare the lattice characteristics of this skyrmion grain boundary with those of atomic crystals, Figure 6-2(a-4) presents a comparison between individual skyrmion positions and atomic positions obtained from molecular statics (MS) simulations of an atomic grain boundary. With only minor

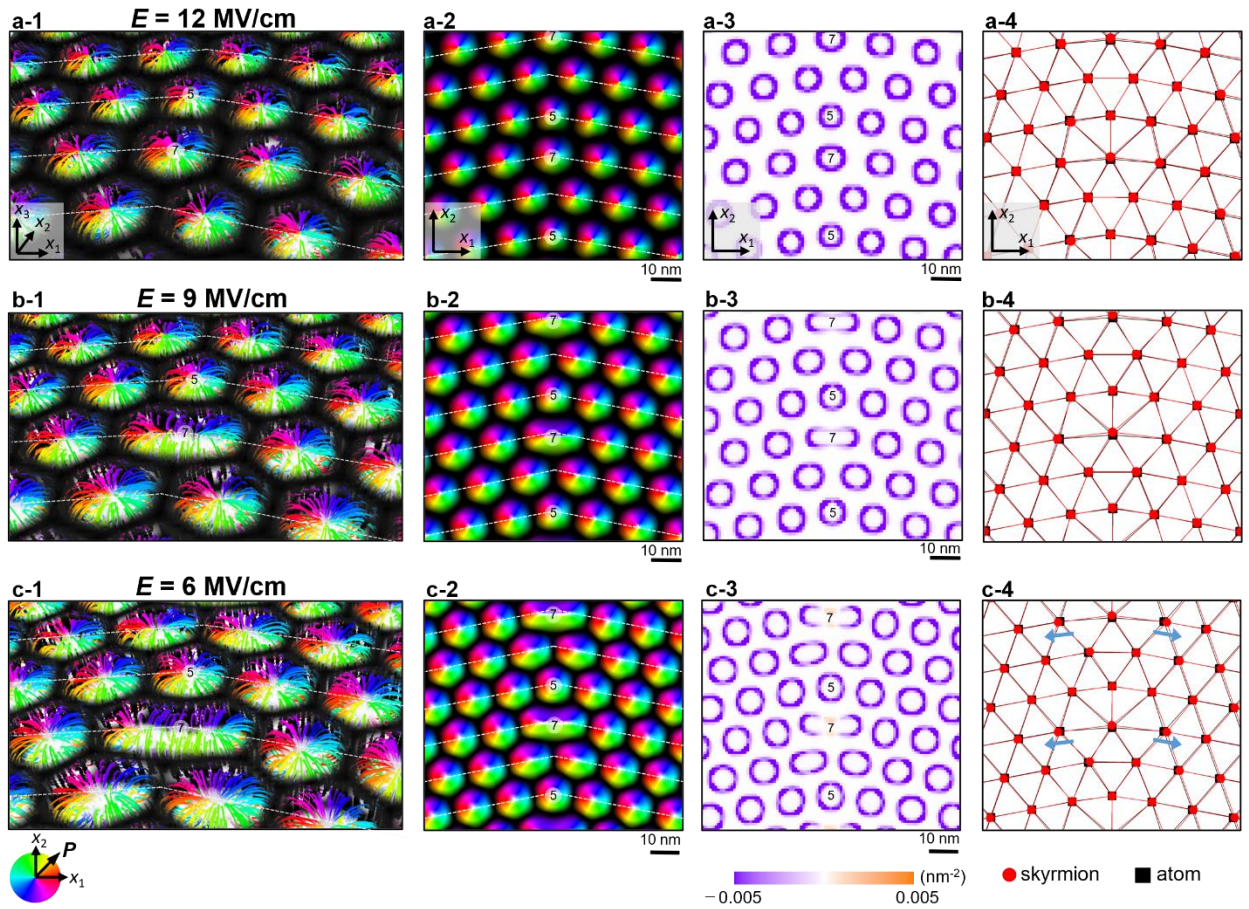


Figure 6-2 (a-1)-(c-1) Three-dimensional view and (a-2)-(c-2) two-dimensional view of polarization distribution at the skyrmion grain boundaries. (a-3)-(c-3) Pontryagin charge density near the skyrmion grain boundaries. (a-4)-(c-4) Differences in the positions of lattice points between the skyrmion grain boundaries and an atomic grain boundary. The length scales are normalized by lattice constants. In (a), (b), and (c), the external out-of-plane electric field is $E = 12, 9$, and 6 MV/cm, respectively.

exceptions, the lattice points in both structures align closely, confirming that the arrangement of skyrmions within the skyrmion grain boundary closely resembles that of atoms in an atomic crystal. Therefore, under high electric field conditions, circular skyrmions form a grain boundary structure analogous to that observed in atomic crystals.

6.3.2 Skyrmion grain boundary structure under medium electric field (9 MV/cm)

Under a medium electric field ($E = 9$ MV/cm), as shown in Figure 6-2(b-2), the 7-fold skyrmion elongates in the x_1 direction, while other skyrmions retain their circular shape. The major axis of the 7-fold skyrmion measures 18 nm, approximately two times that of other skyrmions. This corresponds to the fact that the 7-fold lattice points accommodate a larger area within the lattice mismatch region at the grain boundary. Similar elongation of 7-fold skyrmions has also been observed in magnetic skyrmions at grain boundaries [25]. From the topological charge distribution shown in Figure 6-2(b-3), a ring-based negative q distribution appears in correspondence with the elongation of the 7-fold skyrmion. This characteristic is similar to that of circular skyrmions, indicating that the topological properties remain equivalent. Furthermore, as shown in Figure 6-2(b-4), the skyrmion arrangement aligns closely with that of atomic crystals, similar to the high-field case, with no significant deviations in lattice point positioning at the grain boundary. Note that the grain boundary free volume change of skyrmion lattice is taken into account in the atomic arrangements. Therefore, under medium electric field conditions, even the 7-fold skyrmion undergoes elongation, the grain boundary structure remains analogous to that in atomic crystals.

6.3.3 Skyrmion grain boundary structure under low electric field (6 MV/cm)

On the other hand, as shown in Figure 6-2(c-2), under a low electric field ($E = 6$ MV/cm), skyrmion deformations at the grain boundary become significant. Notably, the 7-fold skyrmion is highly elongated in the x_1 direction, and the two skyrmions positioned below the 7-fold skyrmion are also stretched. In contrast, circular skyrmions are distributed in regions away from the grain boundary, suggesting that skyrmion deformation is induced by lattice mismatch at the

grain boundary. Even in these elongated skyrmions, as shown in Figure 6-2(c-3), the topological charge q remains distributed along their outer edges. However, unlike other skyrmions, the 7-fold skyrmion exhibits non-negative values in its intermediate section. This indicates that, the middle part of the 7-fold skyrmion formed by the elongation has a polarization structure similar to the stripe phase. In other words, a local topological transition occurs in highly elongated the 7-fold skyrmion. Furthermore, as shown in Figure 6-2(c-4), despite the large skyrmion deformations, the arrangement of skyrmions almost remains aligned with the atomic grain boundary. However, only the skyrmions beside the 7-fold skyrmion are slightly shifted in the x_1 direction, consistent with the deformation direction of the 7-fold skyrmion. These observations indicate that under low electric fields, although some skyrmion significantly deform with topological transitions, the fundamental grain boundary structure remains consistent with that of atoms, except in the vicinity of the most deformed skyrmion, the 7-fold skyrmion.

Based on these findings, in skyrmion GBs, although individual skyrmions exhibit electric field dependent deformation, the overall structural characteristics remain largely unchanged. Under high electric fields, skyrmion GB consist of rigid circular skyrmions, whereas the 7-fold skyrmion deform significantly as the electric field decreases. The arrangement of each skyrmion is similar to that of an atomic grain boundary, however, in the vicinity of the highly deformed 7-fold skyrmion, slight positional shifts of adjacent skyrmions occur.

6.3.4 Temperature dependence of skyrmion grain boundary structure

I also investigate the temperature dependence of polar skyrmion grain boundaries. Figure 6-3 presents the skyrmion grain boundaries at an electric field of $E = 6$ MV/cm and temperatures of $T = 100$ and 500 K. At $T = 100$ K, as shown in Figure 6-3(a-1)(a-2), the skyrmions are generally large and exhibit significant deformation near the grain boundary, similar to the distribution observed at $T = 300$ K. The topological charge density (Figure 6-3(a-3)) shows positive values at 7-fold skyrmion, indicating the topological phase transition. In contrast, at $T = 500$ K, as shown in Figure 6-3(b-1)(b-2), individual skyrmions are relatively smaller with less deformation.

The magnitude of polarization values $|P| = \sqrt{P_1^2 + P_2^2 + P_3^2}$ range from 0.145 to 0.448 C/m² at $T = 100$ K, 0.058 to 0.329 C/m² at $T = 300$ K, and 0.014 to 0.193 C/m² at $T = 500$ K. This indicates that polarization increases as temperature decreases, which can be attributed to the temperature dependence of polarization described by the Landau energy (Eq. (2)). Additionally, as shown in Figure 6-3(a-4)(b-4), the positions of each skyrmion consist with atomic grain

boundary in both temperature conditions. Overall, at lower temperatures, both the skyrmion size and deformation increase, whereas at higher temperatures, they decrease. Although this trend is less pronounced than the changes induced by variations in the electric field, it follows a similar pattern. This behavior can be explained by the fact that at lower temperatures, the absolute polarization is larger, making the relative influence of the electric field smaller, while at higher temperatures, the absolute polarization is smaller, making the relative influence of the electric field larger.

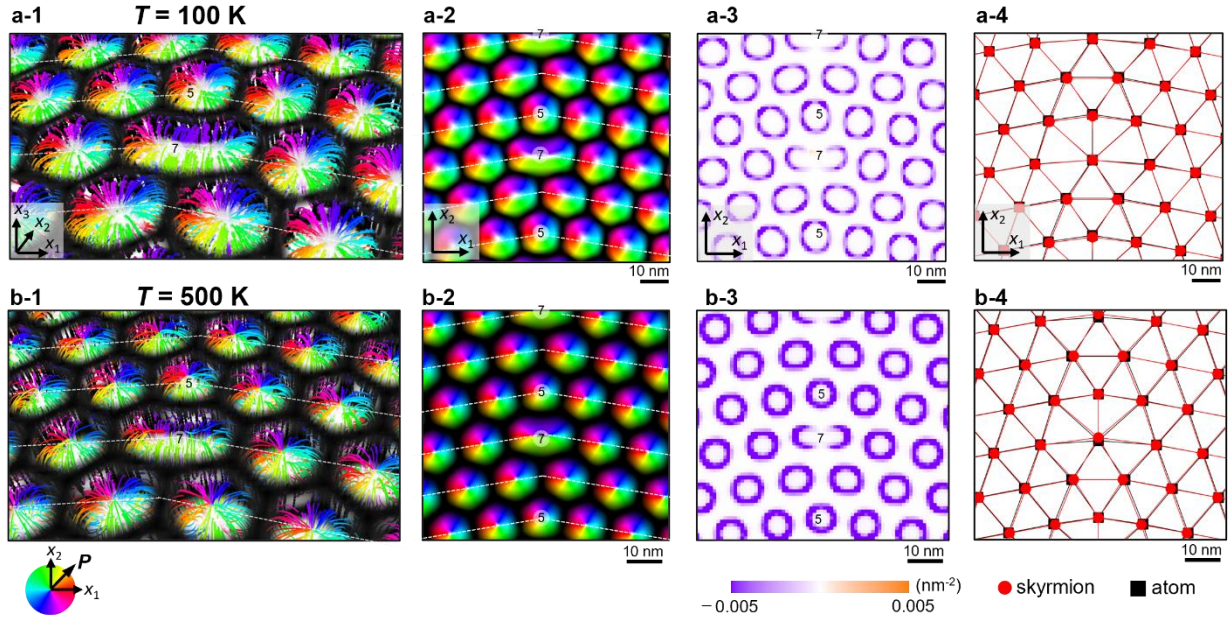


Figure 6-3 (a-1), (b-1) Three-dimensional view and (a-2), (b-2) two-dimensional view of polarization distribution at the skyrmion grain boundaries. (a-3), (b-3) Pontryagin charge density near the skyrmion grain boundaries. (a-4), (b-4) Differences in the positions of lattice points between the skyrmion grain boundaries and an atomic grain boundary. The length scales are normalized by lattice constants. In (a) and (b), the temperature is $T = 100$ and 500 K.

6.3.5 Quasiparticle-based relaxation mechanism in skyrmion grain boundaries

It is essential to discuss the characteristics of skyrmion grain boundaries from the perspective of lattice mechanics. In general, grain boundaries represent regions of lattice mismatch, and their relaxation plays a crucial role in determining the crystal structure, mechanical strength, and

other material properties. Therefore, understanding the mechanism by which skyrmion grain boundaries relax lattice mismatches is important. Here, I use the lattice strain [39,54] in skyrmion lattices as an indicator of lattice mismatch. To quantify this, I construct triangular lattice by treating the center points of each skyrmion as a lattice point and applying Delaunay triangulation. The local lattice strain is then calculated for each triangle by comparing it with an ideal equilateral triangle from a reference perfect lattice. Figure 6-4 shows the lattice strain distribution of skyrmion GBs at the initial state and the equilibrium states under electric fields of $E = 12$ and 6 MV/cm. At the initial configuration, as shown in Figure 6-4(a), skyrmions are regularly arranged, and locally high lattice strain appears only at the grain boundary region, while the rest of the lattice remains strain-free, similar to a perfect crystal. This indicates that the lattice mismatch is localized exclusively at the grain boundary region.

In contrast, at the equilibrium state under $E = 12$ MV/cm, as shown in Figure 6-4(b-1), strains are also distributed in the lattices near the grain boundary. While the lattice strain at the grain boundary itself has decreased compared to the initial state, previously unstrained surrounding lattices now exhibit positive strain values, with higher values observed closer to the boundary.

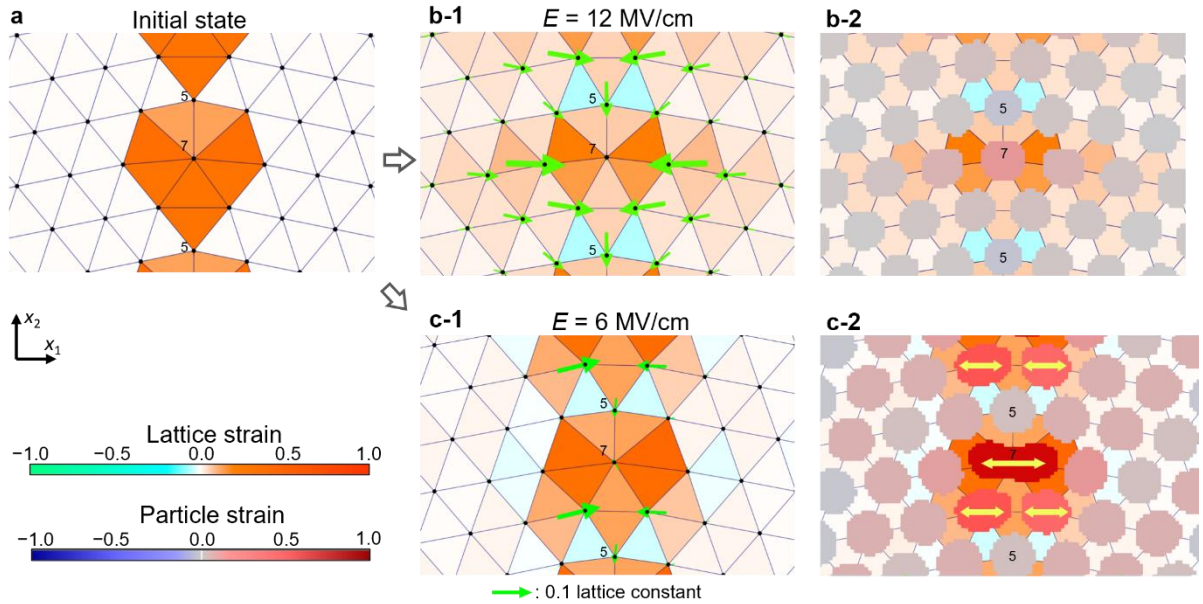


Figure 6-4 Distributions of lattice strain in the x_1 direction near the skyrmion grain boundaries at (a)initial configurations and (b-1)(c-1)equilibrium state of $E = 12$ and 6 MV/cm. The size and directions of the green arrows represent the displacements of each skyrmion from the initial configurations. (b-2)(c-2)The particle strains (rate of change in the long axis) are shown by the color of skyrmion particles. The yellow arrows indicate skyrmions with substantial deformation.

Additionally, as indicated by the green arrows in the figure, skyrmions near the grain boundary have moved significantly. These indicate that the position changes of skyrmions around the grain boundary disperses the local lattice strain from the boundary into the surrounding lattices, thereby relaxing the localized geometric mismatch of the grain boundary. Furthermore, as shown in Figure 6-4(b-2), individual skyrmions remain undeformed, with the most deformed 7-fold skyrmion exhibiting only 15 % deformation. This indicates that skyrmion deformation does not contribute to this relaxation process. Thus, under high electric field conditions, I confirm a lattice-stabilization mechanism similar to that in atomic crystals, where the geometric mismatch at the grain boundary is alleviated by the movement of nearby skyrmions.

On the other hand, under the lower electric field condition ($E = 6$ MV/cm), as shown in Figure 6-4(c-1), the high lattice strain at the grain boundary remains even in the equilibrium state. In particular, the strain in the lattice containing the 7-fold skyrmion remains almost unchanged from the initial state. Additionally, except for a few lattice points, the skyrmions have not moved significantly, indicating that the mismatch relaxation by skyrmion arrangement observed under higher electric field conditions does not occur under low electric field conditions. Furthermore, as shown in Figure 6-4(c-2), the deformation of skyrmions at the grain boundary is significantly large, with the most pronounced deformation occurring in the 7-fold skyrmion at 94 %, followed by the skyrmions below it at 24 %. Therefore, the surrounding skyrmions in regions of high lattice strain exhibit substantial deformation. In addition, as shown in Figure 6-5, the lattice strain in the x_2 direction is significantly small than that in x_1 direction,

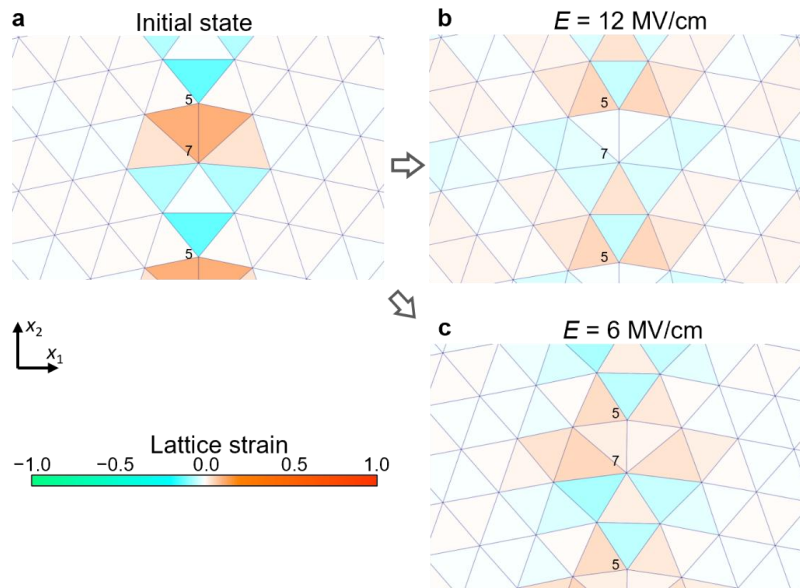


Figure 6-5 Distributions of lattice strain in the x_2 direction near the skyrmion grain boundaries at (a) initial configurations and (b)(c) equilibrium state of $E = 12$ and 6 MV/cm.

highlighting the strong correspondence between lattice strain and skyrmion deformation. These results indicate that under low electric field conditions, the maintenance of lattice mismatches by skyrmion deformations takes place instead of the relaxation of mismatches through skyrmion arrangement. This is a quasiparticle-based stabilization mechanism unique to skyrmions, which is fundamentally different from conventional lattice behavior.

6.3.6 Structures and features of zigzag grain boundaries

Experimentally observed atomic $\Sigma 7$ grain boundaries include zigzag grain boundaries formed by a series of dislocations oriented in different directions [55]. Similar $\Sigma 7$ grain boundaries have also been reported in magnetic skyrmion lattices [25]. Based on these observations, I also investigate zigzag polar skyrmion grain boundaries. Figure 6-6 shows the zigzag grain boundaries in polar skyrmion lattice at $E = 12, 9$, and 6 MV/cm. Similar to the straight grain boundary case, almost all skyrmions are circular at $E = 12$ MV/cm, while the skyrmion deformation near the grain boundary increases with decreasing electric field. The 7-fold skyrmion is the most deformed, supporting that the skyrmions covering a larger area are more deformed.

In addition, the lattice strain distribution differs with the electric field conditions. At $E = 12$ MV/cm, strain is distributed broadly around the grain boundary, while at $E = 6$ MV/cm, it becomes more concentrated at the grain boundary. These results indicate that the local mismatch maintained by quasiparticle mechanics discussed in section 6.3.5 also holds for zigzag grain boundaries. This suggests that such quasiparticle-based mechanics represent a general feature of polar skyrmion lattices.

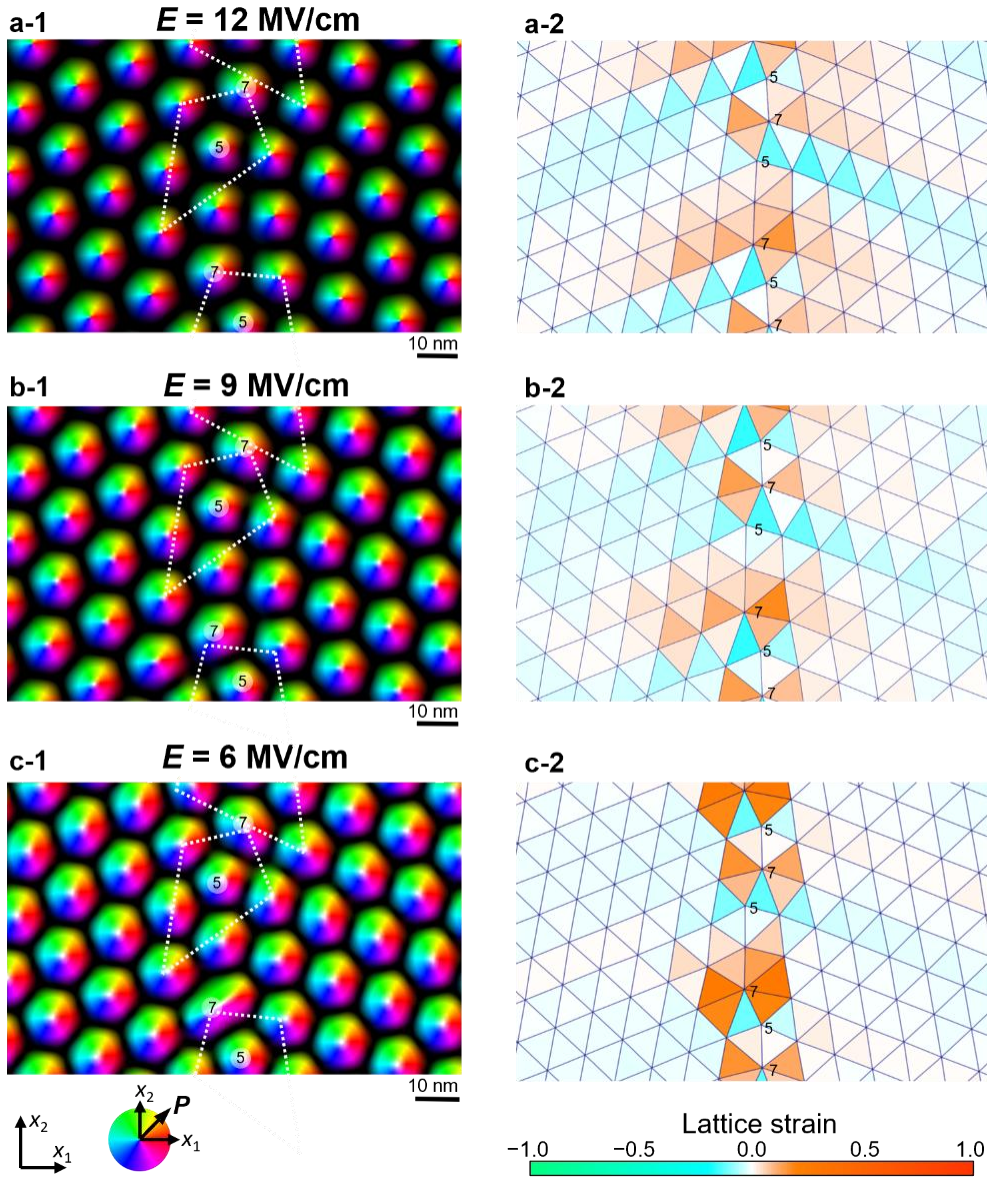


Figure 6-6 (a-1)-(c-1) Polarization distribution at the zigzag skyrmion grain boundaries. (a-2)-(c-2) Distributions of lattice strain near the skyrmion grain boundaries. In (a), (b), and (c), the external out-of-plane electric field is $E = 12, 9$, and 6 MV/cm, respectively.

6.3.7 Energetic and structural characteristics of skyrmion grain boundaries

To systematically characterize the skyrmion grain boundaries, I calculated two fundamental attribute of grain boundaries, grain boundary free volume (BFV) [56,57] and grain boundary energy (GBE) [31,58,59], under various conditions. BFV is the excess volume compared to the

perfect lattice due to the presence of grain boundaries and usually evaluated as an excess volume per unit grain boundary surface. GBE is excess free energy due to existence of grain boundaries, and also evaluated as values per grain boundary surface. To calculate these values, additional simulations of the perfect crystal model without grain boundaries were performed. Since skyrmion lattice is a two-dimensional crystal, both BFV and GBE here are calculated as values per grain boundary length. BFV δ is expressed as

$$\delta = \frac{l_x^{\text{GB}} - l_x^{\text{PL}}}{2l_y}, \quad (6.17)$$

where l_x^{GB} and l_x^{PL} are model length in the x_1 direction of grain boundary model and perfect lattice model, and l_y is the model length in the x_2 direction. GBE γ is expressed as

$$\gamma = \frac{F^{\text{GB}} - F^{\text{PL}}}{2l_y}, \quad (6.18)$$

where F^{GB} and F^{PL} are total free energy of grain boundary model and perfect lattice model. Table 6-4 presents the calculated values, indicating that as the electric field increases, the GBE increases while BFV decreases. A similar trend is observed with increasing temperature, shown in Table 6-5. In other words, under low electric field and low temperature conditions, where skyrmion deformation is more pronounced, BFV is larger, and GBE is lower. Notably, at $E = 6$ and 9 MV/cm, GBEs are negative. Although GBEs are generally positive in atomic crystals, previous studies have reported cases of negative defect formation energies under specific materials or external conditions [60,61], indicating that defect structures can form spontaneously. Therefore, the negative GBEs in skyrmion lattice suggest that decreasing electric field may lead to spontaneous grain boundary formation. In typical atomic crystals, GBE primarily arises from elastic energy due to atomic displacements. However, in skyrmion crystals, mismatch relaxation is not only governed by skyrmion movement but also by skyrmion deformation. This additional degree of freedom allows for a reduction in GBE, highlighting a distinct stabilization mechanism based on quasiparticle deformation, which is fundamentally different from classical grain boundary behavior.

Table 6-4 Grain boundary energy and grain boundary free volume of skyrmion grain boundaries under various electric field conditions with temperature $T = 300$ K.

E [MV/cm]	6	9	12
Grain boundary energy [10^{-11} J/m]	-1.31	-1.015	4.140
Grain boundary free volume	0.124	0.048	-0.149

Table 6-5 Grain boundary energy and grain boundary free volume of skyrmion grain boundaries with various temperature conditions under external electric field $E = 6 \text{ MV/cm}$.

$T \text{ [K]}$	100	300	500
Grain boundary energy [10^{-11} J/m]	−6.61	−1.31	−6.75
Grain boundary free volume	0.134	0.124	0.113

6.4 Conclusion

In summary, this chapter demonstrates that the grain boundaries of polar skyrmions undergo structural transitions from atomic-based configurations to unique skyrmion-specific structures depending on electric field and temperature, accompanied by changes in their stabilization mechanisms. Under high electric fields and high temperature conditions, skyrmions have circular shape and form grain boundary structures similar to those in atomic crystals. In this structure, the skyrmions near the grain boundaries adjust their positions to relax the lattice mismatches. In contrast, under low electric fields and low-temperature conditions, skyrmions at the grain boundary undergo significant deformation, maintaining large local lattice mismatches. This indicates a quasiparticle-based stabilization mechanism distinct from conventional crystals. Generally, in atomic crystal growth, atoms arrange themselves to minimize grain boundary mismatches, influencing the final crystal structure, mechanical strength, and other material properties [62]. The unique structures and stabilization mechanisms observed in polar skyrmion grain boundaries indicate the distinctive lattice dynamics of polar skyrmion lattices due to the grain boundary properties and suggest the potential emergence of novel electromechanical properties. Furthermore, the transition in stabilization mechanisms—from lattice relaxation via skyrmion arrangement to quasiparticle stabilization through skyrmion deformation—identified in these findings may also be relevant to the growth process of polar skyrmion lattices and their response to macroscopic external forces. Therefore, our findings provide fundamental insights into the general stabilization mechanisms of polar skyrmion lattices.

References

- [1] Y. Nahas, S. Prokhorenko, L. Louis, Z. Gui, I. Kornev, and L. Bellaiche, Discovery of stable skyrmionic state in ferroelectric nanocomposites, *Nat Commun* **6**, 8542 (2015).
- [2] S. Das et al., Observation of room-temperature polar skyrmions, *Nature* **568**, 368 (2019).
- [3] S. Mühlbauer, B. Binz, F. Jonietz, C. Pfleiderer, A. Rosch, A. Neubauer, R. Georgii, and P. Böni, Skyrmion Lattice in a Chiral Magnet, *Science* **323**, 915 (2009).
- [4] X. Z. Yu, Y. Onose, N. Kanazawa, J. H. Park, J. H. Han, Y. Matsui, N. Nagaosa, and Y. Tokura, Real-space observation of a two-dimensional skyrmion crystal, *Nature* **465**, 901 (2010).
- [5] H. Ge, X.-Y. Xu, L. Liu, R. Xu, Z.-K. Lin, S.-Y. Yu, M. Bao, J.-H. Jiang, M.-H. Lu, and Y.-F. Chen, Observation of Acoustic Skyrmions, *Phys. Rev. Lett.* **127**, 144502 (2021).
- [6] C. Liu, S. Zhang, S. A. Maier, and H. Ren, Disorder-Induced Topological State Transition in the Optical Skyrmion Family, *Phys. Rev. Lett.* **129**, 267401 (2022).
- [7] J. Sampaio, V. Cros, S. Rohart, A. Thiaville, and A. Fert, Nucleation, stability and current-induced motion of isolated magnetic skyrmions in nanostructures, *Nature Nanotech* **8**, 839 (2013).
- [8] A. Neubauer, C. Pfleiderer, B. Binz, A. Rosch, R. Ritz, P. G. Niklowitz, and P. Böni, Topological Hall Effect in the A Phase of MnSi, *Phys. Rev. Lett.* **102**, 186602 (2009).
- [9] A. Fert, V. Cros, and J. Sampaio, Skyrmions on the track, *Nature Nanotech* **8**, 152 (2013).
- [10] X. Zhang, M. Ezawa, and Y. Zhou, Magnetic skyrmion logic gates: conversion, duplication and merging of skyrmions, *Sci Rep* **5**, 9400 (2015).
- [11] H. Zhang, D. Zhu, W. Kang, Y. Zhang, and W. Zhao, Stochastic Computing Implemented by Skyrmionic Logic Devices, *Phys. Rev. Applied* **13**, 054049 (2020).
- [12] T. Yokouchi, S. Sugimoto, B. Rana, S. Seki, N. Ogawa, Y. Shiomi, S. Kasai, and Y. Otani, Pattern recognition with neuromorphic computing using magnetic field-induced dynamics of skyrmions, *Sci. Adv.* **8**, eabq5652 (2022).
- [13] M.-K. Lee and M. Mochizuki, Handwritten digit recognition by spin waves in a Skyrmion reservoir, *Sci Rep* **13**, 19423 (2023).
- [14] H. Aramberri and J. Íñiguez-González, Brownian Electric Bubble Quasiparticles, *Phys. Rev. Lett.* **132**, (2024).
- [15] M. A. P. Gonçalves, M. Paściak, and J. Hlinka, Antiskyrmions in Ferroelectric Barium Titanate, *Phys. Rev. Lett.* **133**, (2024).

- [16] M. A. Pereira Gonçalves, C. Escorihuela-Sayalero, P. Garca-Fernández, J. Junquera, and J. Íñiguez, Theoretical guidelines to create and tune electric skyrmion bubbles, *Sci. Adv.* **5**, (2019).
- [17] L. Han et al., High-density switchable skyrmion-like polar nanodomains integrated on silicon, *Nature* **603**, 7899 (2022).
- [18] S. Das et al., Local negative permittivity and topological phase transition in polar skyrmions, *Nat. Mater.* **20**, 194 (2021).
- [19] L. Gao, S. Prokhorenko, Y. Nahas, and L. Bellaiche, Dynamical Control of Topology in Polar Skyrmions via Twisted Light, *Phys. Rev. Lett.* **132**, 026902 (2024).
- [20] Y.-T. Shao et al., Emergent chirality in a polar meron to skyrmion phase transition., *Nat Commun* **14**, 1355 (2023).
- [21] W. Chen, J. Ren, L. Liu, F. Sun, Q. He, M. Wu, and Y. Zheng, *Emergence and Transformation of Polar Skyrmion Lattices via Flexoelectricity*.
- [22] X. Yu, D. Morikawa, T. Yokouchi, K. Shibata, N. Kanazawa, F. Kagawa, T. Arima, and Y. Tokura, Aggregation and collapse dynamics of skyrmions in a non-equilibrium state, *Nature Phys* **14**, 832 (2018).
- [23] S. Pöllath et al., Dynamical Defects in Rotating Magnetic Skyrmion Lattices, *Phys. Rev. Lett.* **118**, 207205 (2017).
- [24] R. Brearton, M. W. Olszewski, S. Zhang, M. R. Eskildsen, C. Reichhardt, C. J. O. Reichhardt, G. Van Der Laan, and T. Hesjedal, Skyrmions in anisotropic magnetic fields: strain and defect driven dynamics, *MRS Advances* **4**, 643 (2019).
- [25] T. Matsumoto, Y.-G. So, Y. Kohno, H. Sawada, Y. Ikuhara, and N. Shibata, Direct observation of $\Sigma 7$ domain boundary core structure in magnetic skyrmion lattice, *Sci. Adv.* **2**, e1501280 (2016).
- [26] J. P. Hirth, The influence of grain boundaries on mechanical properties, *Metall Trans* **3**, 3047 (1972).
- [27] J. Kacher, B. P. Eftink, B. Cui, and I. M. Robertson, Dislocation interactions with grain boundaries, *Current Opinion in Solid State and Materials Science* **18**, 227 (2014).
- [28] M. Upmanyu, D. J. Srolovitz, L. S. Shvindlerman, and G. Gottstein, Vacancy Generation During Grain Boundary Migration, (n.d.).
- [29] H. Gleiter, Grain boundaries as point defect sources or sinks—Diffusional creep, *Acta Metallurgica* **27**, 187 (1979).

- [30] Z. Gong, W. Zhao, K. Guan, P. Rao, Q. Zeng, J. Liu, and Z. Feng, Influence of grain boundary and grain size on the mechanical properties of polycrystalline ceramics: Grain-scale simulations, *J Am Ceram Soc* **103**, 5900 (2020).
- [31] M. E. Glicksman and R. A. Masumura, Grain boundary structure and energetics, *Metall Trans A* **8**, 1373 (1977).
- [32] T. Uesugi and K. Higashi, First-Principles Calculation of Grain Boundary Excess Volume and Free Volume in Nanocrystalline and Ultrafine-Grained Aluminum, *Mater. Trans.* **54**, 1597 (2013).
- [33] R. E. Hoffman and D. Turnbull, Lattice and Grain Boundary Self-Diffusion in Silver, *Journal of Applied Physics* **22**, 634 (1951).
- [34] J. C. Fisher, Calculation of Diffusion Penetration Curves for Surface and Grain Boundary Diffusion, *Journal of Applied Physics* **22**, 74 (1951).
- [35] M. P. Seah, Grain boundary segregation, (n.d.).
- [36] M. D. Sangid, H. J. Maier, and H. Schitoglu, The role of grain boundaries on fatigue crack initiation – An energy approach, *International Journal of Plasticity* **27**, 801 (2011).
- [37] T. Watanabe, The impact of grain boundary character distribution on fracture in polycrystals, *Materials Science and Engineering: A* **176**, 39 (1994).
- [38] A. Das, Grain boundary engineering: fatigue fracture, *Philosophical Magazine* **97**, 867 (2017).
- [39] K. Kasai, T. Xu, S. Minami, and T. Shimada, Breakdown of Volterra's Elasticity Theory of Dislocations in Polar Skyrmion Lattices, *Nano Lett.* **24**, 13247 (2024).
- [40] X. Fang, V. Viteri-Pflucker, A. H. King, J. Wang, J. Yan, L. Ke, and L. Zhou, Oscillating Grain Boundaries and Their Effects on Grain Growth: Observations in Skyrmion Bicrystals, *Acta Materialia* **290**, 120965 (2025).
- [41] S. Yuan, Z. Chen, S. Prokhorenko, Y. Nahas, L. Bellaiche, C. Liu, B. Xu, and L. Chen, Hexagonal Close-packed Skyrmion Lattice in Ultrathin Ferroelectric PbTiO₃ Films, *Physical Review Letters* **130**, 226801 (2023).
- [42] X. Zhang et al., Creation and erasure of polar bubble domains in PbTiO₃ films by mechanical stress and light illuminations, *Journal of Materiomics* **9**, 626 (2023).
- [43] Y. L. Li, S. Y. Hu, Z. K. Liu, and L. Q. Chen, Effect of substrate constraint on the stability and evolution of ferroelectric domain structures in thin films, *Acta Materialia* **50**, 395 (2002).

- [44] G. Sheng, Y. L. Li, J. X. Zhang, S. Choudhury, Q. X. Jia, V. Gopalan, D. G. Schlom, Z. K. Liu, and L. Q. Chen, A modified Landau–Devonshire thermodynamic potential for strontium titanate, *Appl. Phys. Lett.* **96**, 232902 (2010).
- [45] C. H. Woo and Y. Zheng, Depolarization in modeling nano-scale ferroelectrics using the Landau free energy functional, *Appl. Phys. A* **91**, 59 (2008).
- [46] M. J. Haun, E. Furman, S. J. Jang, H. A. McKinstry, and L. E. Cross, Thermodynamic theory of PbTiO_3 , *Journal of Applied Physics* **62**, 3331 (1987).
- [47] L. Q. Chen and J. Shen, Applications of semi-implicit Fourier-spectral method to phase field equations, *Computer Physics Communications* **108**, 147 (1998).
- [48] H.-L. Hu and L.-Q. Chen, Three-Dimensional Computer Simulation of Ferroelectric Domain Formation, *Journal of the American Ceramic Society* **81**, 492 (2005).
- [49] J. J. Wang, S. Bhattacharyya, Q. Li, T. W. Heo, X. Q. Ma, and L.-Q. Chen, Elastic solutions with arbitrary elastic inhomogeneity and anisotropy, *Philosophical Magazine Letters* **1** (2012).
- [50] J. J. Wang, X. Q. Ma, Q. Li, J. Britson, and L.-Q. Chen, Phase transitions and domain structures of ferroelectric nanoparticles: Phase field model incorporating strong elastic and dielectric inhomogeneity, *Acta Materialia* **61**, 7591 (2013).
- [51] K. Kasai, T. Itano, S. Minami, and T. Shimada, *Microscopic Formation Process of Polar Skyrmions and Strain-Dependent Topological Phase Transitions in $\text{PbTiO}_3/\text{SrTiO}_3$ Superlattices*.
- [52] Y. Zhang, Q. Li, H. Huang, J. Hong, and X. Wang, Strain manipulation of ferroelectric skyrmion bubbles in a freestanding PbTiO_3 film: A phase field simulation, *Phys. Rev. B* **105**, 224101 (2022).
- [53] N. Nagaosa and Y. Tokura, Topological properties and dynamics of magnetic skyrmions, *Nature Nanotech* **8**, 899 (2013).
- [54] A. Stukowski, Computational Analysis Methods in Atomistic Modeling of Crystals, *JOM* **66**, 399 (2014).
- [55] F. Oba, H. Ohta, Y. Sato, H. Hosono, T. Yamamoto, and Y. Ikumura, Atomic structure of [0001]-tilt grain boundaries in ZnO: A high-resolution TEM study of fiber-textured thin films, *PHYSICAL REVIEW B* (2004).
- [56] L. S. Shvindlerman and G. Gottstein, Unexplored topics and potentials of grain boundary engineering, *Scripta Materialia* **54**, 1041 (2006).

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- [57] S. Van Petegem, F. Dalla Torre, D. Segers, and H. Van Swygenhoven, Free volume in nanostructured Ni, *Scripta Materialia* **48**, 17 (2003).
- [58] W. Gui-Jin and V. Vitek, Relationships between grain boundary structure and energy, *Acta Metallurgica* **34**, 951 (1986).
- [59] G. S. Rohrer, The role of grain boundary energy in grain boundary complexion transitions, *Current Opinion in Solid State and Materials Science* **20**, 231 (2016).
- [60] T. Shimada, T. Ueda, J. Wang, and T. Kitamura, Hybrid Hartree-Fock density functional study of charged point defects in ferroelectric PbTiO₃, *Phys. Rev. B* **87**, (2013).
- [61] M. W. Cleveland and M. J. Demkowicz, Persistence of negative vacancy and self-interstitial formation energies in atomistic models of amorphous silicon, *Phys. Rev. Materials* **6**, (2022).
- [62] I. Adlakha, M. A. Tschopp, and K. N. Solanki, The role of grain boundary structure and crystal orientation on crack growth asymmetry in aluminum, *Materials Science and Engineering: A* **618**, 345 (2014).

Chapter 7 Conclusion

7.1 Summary of thesis

In this thesis, I have investigated emergent topological polar structures induced by mechanical fields using multiphysics phase-field simulations. This thesis establishes a novel strategy for the mechanical creation and control of emergent topological polar structures through electromechanical coupling effects, revealing how these effects cooperatively generate and stabilize exotic polarization configurations beyond conventional domains.

Chapter 2 demonstrates the mechanical creation and control of two-dimensional topological polar structures by singular stress fields around crack tips under mixed-mode loading. The topological characteristics of strain and strain gradient field are reflected in the resulting polarization distribution. Both piezoelectric and flexoelectric effects are shown to contribute to the formation and manipulation of topological polarization textures, which construct the basis of the proposed concept of “topological-strain field engineering”.

Chapter 3 extends the concept of strain-field engineering to higher-dimensional topological structures. The strain gradient fields induced by nanoindentation act as an effective driving force to generate rotational polarization, leading to the formation of quasi-two-dimensional polar structures, including polar skyrmions. By combining mechanical writing and electrical erasing under background polarization textures, various topological quasiparticles, such as merons and skyrmioniums, are demonstrated to be created in a controlled manner, confirming the universality of the proposed mechanical approach.

Chapter 4 investigates the collective formation of polar skyrmions, revealing strain-induced topological transitions from a trivial stripe phase to skyrmion bubble and skyrmion lattice phases. I successfully established a firm understanding of the formation process of these phases in ferroelectric heterostructures, providing theoretical support for their experimental realization. These results show that mechanical control is also effective for the collective organization of quasiparticle-like skyrmions, offering a realistic route toward the realization of emergent polar skyrmion lattices.

Chapter 5 focuses on the typical lattice defects, dislocations in polar skyrmion lattices. Dislocations are stabilized as a 5-7 pair configuration like those in atomic crystals, even though polar skyrmions are not real rigid particles of atoms but quasiparticles composed of polarization vector fields. The deformability of individual skyrmions significantly depends on external electric fields, leading to quasiparticle-specific core structures with markedly elongated skyrmions. Furthermore, these skyrmion deformations change the conventional elastic field intrinsic to the dislocation and cause the breakdown of Volterra's elasticity theory in polar skyrmion lattices. These findings suggest the emergence of a quasiparticle lattice mechanics, in which skyrmion deformation interacts with lattice strain.

Chapter 6 investigates another lattice defect, i.e., grain boundaries, in polar skyrmion lattices. I reveal a quasiparticle-specific structure and stabilization mechanism distinct from atomic crystals, providing novel insights into the collective behavior of topological polar skyrmions and skyrmion lattice mechanics.

7.2 Prospects

The fundamental understanding of topological polar structures and mechanical fields established in this thesis provides a universal framework for designing and controlling emergent topological polarization structures. Based on this foundation, further exploration of their novel response phenomena intrinsic to polar topological structures, such as optical activity, dielectric tunability, and electromechanical conversion, can be a promising research direction. Once these response properties are fully understood, it will open the possibility of realizing next-generation ferroelectric devices in which multiple functionalities associated with topological polar orders can be dynamically written and reconfigured. Such response properties of topological polar structures will also provide potential applications of multistate information storage, high electromechanical elements, and energy-efficient functional materials based on this work, for beyond the capabilities of conventional domain-based ferroelectrics.

Furthermore, the realization of polar topological structures in this thesis enables direct comparison with their magnetic counterparts, which have been extensively studied in ferromagnetics. A comparative investigation of polar and magnetic skyrmions will provide valuable insights into the fundamental physics behind ferroelectrics and ferromagnets. The contrast between the electromechanical interactions governing polar skyrmions and Dzyaloshinskii-Moriya interactions governing magnetic skyrmions will clarify how distinct

microscopic origins can lead to emergent phenomena across ferroic electric and magnetic systems.

Finally, this thesis opens a new research field of “quasiparticle lattice mechanics”. The breakdown of conventional elasticity in polar skyrmion lattices shown in this thesis also suggests the existence of unconventional mechanics in dynamic behaviors of skyrmion lattices. In conventional lattices such as atomic crystals, lattice defects and their dynamics, such as dislocation glides, play crucial roles in macroscopic mechanical properties, including plastic deformation. In contrast, polar skyrmion lattices are composed of discrete quasiparticles with internal degrees of freedom as intrinsic particle-deformability, suggesting that their lattice defect dynamics may obey entirely different principles. Elucidating how these quasiparticle-specific characteristics influence lattice mechanics will not only deepen our fundamental understanding of collective behavior in topological orders but also construct a way for utilizing these discrete features unlike conventional continuous domains. I have already started research in this direction and have preliminarily obtained distinctive lattice dynamics of skyrmion lattices. Further studies will advance the physics of collective quasiparticle systems.

List of publications and conferences

Publications

1. “Emergence of non-trivial polar topologies hidden in singular stress field in SrTiO₃: topological strain-field engineering”
Takahiro Shimada, Yu Wang, Takayuki Hamaguchi, Kohta Kasai, Kairi Masuda, Le Van Lich, Tao Xu, Jie Wang and Hiroyuki Hirakata
Journal of Physics: Condensed Matter 33 505301 (2021)
2. “Stability and deformation of a vacancy defect in skyrmion crystal under external magnetic and temperature fields”
Yu Wang, Ryosuke Manabe, Kohta Kasai, Tao Xu, Takahiro Shimada
Acta Materialia 281 120381 (2024)
3. “Singular stress field induced two-dimensional topological polar structures in SrTiO₃: Topological number manipulation via loading modes”
Kohta Kasai, Takayuki Hamaguchi, Yu Wang, Susumu Minami, Hiroyuki Hirakata, Takahiro Shimada
Computational Material Science 242 113070(2024)
4. “Mechanical Writing of Polar Skyrmionic Topological States via Extrinsic Dzyaloshinskii–Moriya-like Flexoelectricity in Ferroelectric Thin Films”
Kohta Kasai, Takashi Nojima, Yu Wang, Tao Xu, Hiroyuki Hirakata, Takahiro Shimada,
ACS Nano 18 47 32451-32457(2024)
5. “Breakdown of Volterra’s Elasticity Theory of Dislocations in Polar Skyrmion Lattices”
Kohta Kasai, Tao Xu, Susumu Minami, Takahiro Shimada
Nano Letters 24 13247-13254(2024)
6. “Microscopic formation process of polar skyrmions and strain-dependent topological phase transitions in PbTiO₃/SrTiO₃ superlattices”
Kohta Kasai, Toya Itano, Susumu Minami, Takahiro Shimada
Scripta Materialia 263 116680(2025)

7. “Grain boundaries stabilization of polar skyrmion lattice via quasiparticle-based mechanism”
Kohta Kasai, Shun Miyata, Susumu Minami, Takahiro Shimada
Physical Review B 112 125421(2025)
8. “Temperature and magnetic field dependent grain boundary structures in skyrmion lattice via quasiparticle-based mechanism”
Kohta Kasai, Chang Liu, Akihiro Uematsu, Tatsuki Kawakane, Yu Wang, Tao Xu, Susumu Minami, Takahiro Shimada
Physical Review Materials (accepted)
9. “Topological transition and emergent elasticity of dislocation in skyrmion lattice: Beyond Kittel's magnetic-polar analogy”
Kohta Kasai, Akihiro Uematsu, Yu Wang, Tao Xu, Chang Liu, Susumu Minami, Takahiro Shimada
Physical Review Letter (under review)
10. “Multiferroic Skyrmions in Composite Ferroic Heterostructures”
Tao Xu, Chensheng Wu, Yu Wang, Kohta Kasai, Jie Wang, Takahiro Shimada
Nano Letters (submitted)

International Conference

1. “Phase-field Simulation of Mechanically Induced Polar Skyrmions in PbTiO₃ Ultrathin Films”
Kohta Kasai, Takashi Nojima, Hiroyuki Hirakata, Takahiro Shimada
The 6th International Conference on Materials and Reliability, KDDI Ishin Hall, Yamaguchi, Japan, December 7-9, 2022 [Oral]
2. “Phase-field simulation of mechanical transition of polar skyrmion lattice”
Kohta Kasai, Toya Itano, Susumu Minami, Takahiro Shimada
34th IUPAP Conference on Computational Physics, Kobe International Conference Center, Kobe, Japan, August 4-8, 2023 [Poster]
3. “Phase-field simulation of polar skyrmion lattice and its defect in ferroelectric thin film”
Kohta Kasai, Yu Wang, Susumu Minami, Takahiro Shimada

- Joint-Symposium on Mechanics of Advanced Materials & Structures, Crowne Plaza
Chengdu West Hotel, Chengdu, China, December 1-4, 2023 [Oral]
4. “Quasiparticle-like structures and behaviors of magnetic skyrmion lattice defects from phase-field simulations”
Kohta Kasai, Yu Wang, Tatsuki Kawakane, Akihiro Uematsu, Ryosuke Manabe, Susumu Minami, and Takahiro Shimada
The 5th International Symposium on Phase-Field Modelling in Materials Science, Dragon Hotel, Hangzhou, China, May 19-23, 2024 [Poster]
 5. “Phase-field simulation of structure and motion of dislocation in polar skyrmion lattice in ferroelectric thin film”
Kohta Kasai, Susumu Minami, Takahiro Shimada
Symposium on Mechanics of Advanced Materials & Structures for Young Researchers 2024, Sago Royal Hotel, Hamamatsu, Japan, September 6-7, 2024 [Oral]
 6. “Phase-field simulation of dislocations in polar skyrmion lattice: Breakdown of Volterra’s elasticity theory”
Kohta Kasai, Susumu Minami, Takahiro Shimada
Asia-Pacific Conference on Fracture and Strength 2024, Kunibiki Messe, Shimane, Japan, November 25-29, 2024 [Oral]
 7. “Phase-field simulation of grain boundaries in polar skyrmion lattice”
Kohta Kasai, Shun Miyata, Susumu Minami, and Takahiro Shimada
The 10th Symposium for Multiscale Materials Mechanics: Mechanism and Modeling, Kyoto Research Park, Kyoto, Japan, June 7, 2025 [Poster]
 8. “Phase-field simulation of quasiparticle-based grain boundary structures in polar skyrmion lattice”
Kohta Kasai, Shun Miyata, Susumu Minami, Takahiro Shimada
The 8th Joint-Symposium on Mechanics of Advanced Materials & Structures, RIHGA Royal Hotel Kyoto, Kyoto, Japan, November 28-December 1, 2025 [Oral]

Domestic Conference

1. 「特異応力場による強誘電体内の高次トポロジカル秩序群の創出」
笠井恒汰, 濱口高征, 嶋田隆広, 平方寛之
第 6 回マルチスケール材料力学シンポジウム 山口大学 2021/5/24-28 [Online poster]
2. 「高次トポロジカルナノ強誘電体の創出：SrTiO₃ 内き裂への負荷に関する Phase-field 解析」
笠井恒汰, 濱口高征, 嶋田隆広, 平方寛之
第 7 回材料 WEEK 京都テルサ 2021/10/12-13 [Online oral]
3. 「強誘電スキルミオンのナノメカニカルライティング」
笠井恒汰, 野島崇志, 嶋田隆広
第 7 回マルチスケール材料力学シンポジウム 大阪科学技術センター
2022/5/29-31 [Online poster]
4. 「強誘電体ナノ薄膜への押し込みによる分極スキルミオン組織形成に関する Phase-field 解析」
笠井恒汰, 野島崇志, 平方 寛之, 嶋田隆広
第 35 回計算力学講演会 鹿児島大学 2022/11/16-18 [Online oral]
5. 「SrTiO₃/PbTiO₃ 超格子内の分極スキルミオン格子の力学誘起相転移に関する Phase-field 解析」
笠井恒汰, 板野統哉, 嶋田隆広
第 8 回マルチスケール材料力学シンポジウム つくば国際会議場 2023/5/28 [Poster]
6. 「強誘電ナノ薄膜における分極スキルミオン格子とその欠陥構造に関する Phase-field 解析」
笠井恒汰, 見波将, 嶋田隆広
M&M2023 材料力学カンファレンス 筑波大学 筑波キャンパス 第三エリア 3A 棟・3B 棟 2023/9/27-29 [Oral]

7. 「 PbTiO_3 ナノ薄膜における分極スキルミオン格子形成とそのトポロジカル欠陥構造に関する Phase-field 解析」
笠井恒汰, 見波将, 嶋田隆広
セラミックス協会 第 37 回秋季シンポジウム 名古屋大学 (東山キャンパス)
2024/09/10-12 [Oral]
8. 「強誘電ナノ薄膜における分極スキルミオン格子とその欠陥ダイナミクスに関する Phase-field 解析」
笠井恒汰, 見波将, 嶋田隆広
第 37 回計算力学講演会 トークネットホール仙台 2024/10/18-20 [Oral]
9. 「分極スキルミオン格子中の欠陥ダイナミクスに関する強誘電薄膜 phase-field 解析」
笠井恒汰, 佐竹隆成, 見波将, 嶋田隆広
日本金属学会 2025 年秋期(第 177 回)講演大会 北海道大学 2025/9/17-19 [Poster]
10. 「 MnSi 薄膜における磁性スキルミオン格子中の準粒子的欠陥運動に関する Phase-field 解析」
笠井恒汰, 川金立暉, 見波将, 嶋田隆広
第 38 回計算力学講演会 信州大学工学部 2025/9/24-26 [Oral]
11. 「強誘電ナノ薄膜中の分極スキルミオン格子における準粒子的すべり運動に関する Phase-field 解析」
笠井恒汰, 佐竹隆成, 見波将, 嶋田隆広
M&M2025 材料力学カンファレンス 熊本城ホール 2025/11/10-13 [Oral]

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