

Domains and domain walls in ferroic materials

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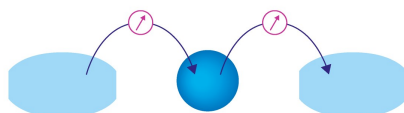
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I. INTRODUCTION

Ferroic materials exhibit long-range order with respect to their elastic, electric, or magnetic properties, giving rise to fascinating physics and functional properties that are used in, e.g., state-of-the-art sensor technology, energy harvesting, and medical diagnosis. Despite more than 100 years of research on ferroics, this class of materials remains an exciting playground for both fundamental and applied research studies, and it is safe to predict that this trend will continue. Recent examples for the special behaviors of ferroic materials that have inspired the community are the discovery of topologically protected electric¹ and magnetic^{2,3} skyrmions, negative capacitance,^{4,5} and resistive switching.^{6,7} Progress in the field of ferroics is propelled by the remarkable evolution that has taken place in both experimental investigations and theory, making it possible to explore the physical properties of ferroic materials with unprecedented completeness down to the length scale of individual atoms. In addition, improved synthesis methods and *in situ* characterization tools applied during growth allow for stabilizing novel exotic phases and artificial heterostructures to engineer ferroic properties.^{8,9} These capabilities have opened the door for new science and conceptually different technologies, promoting innovative fields such as low-energy spintronics¹⁰ and multi-level data storage for neuromorphic computing and next-generation nanotechnology.

Many of the current challenges, however, still fall into the basic research sector, and it is clear that we have only scratched the tip of the iceberg regarding the rich emergent nanoscale phenomena in ferroics. This is the motivation for the Special Topic on Domains and Domain Walls in Ferroic Materials. The functional response of any ferroic material inevitably relates to the formation and/or manipulation of domains and domain walls. Thus, their control via the crystallographic structure, size effects, strain, and

electrostatic conditions is one of the key aspects within the field. In this Guest Editorial, we will provide a short introduction to the basic concepts and the fundamental questions that drive the modern research on ferroic domains and domain walls. In addition, a short overview of different topics covered by the articles in the Special Topic will be provided.

II. BACKGROUND

Ferroics are defined by their elastic, electric, or magnetic order, which spontaneously arise across a non-disruptive phase transition.¹¹ In ferroelectrics, for example, electric dipoles align giving rise to spontaneous polarization, whereas collinear arrangements of spins lead to macroscopic magnetization in ferromagnets. Depending on the microscopic interactions that drive the order, however, much more complex structures can arise. Going beyond the primary types of ferroic order—namely, ferromagnetism, ferroelectricity, ferroelasticity, and ferrotoroidicity¹²—a plethora of anti-ferroic phases exists. In fact, the majority of magnetically ordered systems are antiferromagnetic. Another interesting class of ferroic materials are multiferroics.¹³ In multiferroics, at least two types of ferroic order are present in the same phase, which can lead to unusual correlation phenomena, including pronounced magnetoelectric and magnetocapacitance effects.¹⁴ Independent of the nature of the long-range order, the order parameter(s) that describe the ferroic phase can point in at least two symmetrically equivalent directions between which it can be switched by the application of a conjugate field. When cooling into the ordered state, the symmetry equivalent directions have the same energy so that two or more order parameter orientations will occur. Regions with the same orientation of the order parameter are called *domains*, and the interfaces that separate them are the *domain walls*.^{15,16}

Originally, ferroics-based device concepts mainly utilized the magnetic domains. Here, the basic idea is to use ferroic domains with opposite order parameters to store information, representing information bits “1” and “0.” This approach enabled magnetic recording with magnetic wires and tapes and, later, in magnetic hard disk drives.¹⁷ Eventually, this concept developed as the foundation for ferroelectric random-access memory and field-effect transistors,^{18,19} where the polarization of ferroelectrics is utilized for data storage. Promising new candidate materials that have the potential to revolutionize these technologies are two-dimensional (2D) electric²⁰ and magnetic²¹ ferroics, offering novel opportunities for domain control and the design of power-efficient electronics devices.

Ferroic domain walls began to move into focus with the advent of racetrack-type memory technology. In the original concept, information is still stored in ferromagnetic domains;²² however, it is the domain walls and their specific interactions with spin currents that enable the motion of bits along the track, which distinguished this technology from previous approaches. Today, it is established that ferroic domain walls represent outstanding functional nano-entities with unique physical properties, which are exploited in the rapidly expanding field of domain-wall nanoelectronics.^{16,23} Due to their low local symmetry and the distinct structural, electric, or magnetic environment, ferroic domain walls can behave substantially differently than the surrounding domains. For example, ferromagnetic domain walls can form in otherwise anti-ferromagnetic systems,^{24,25} and insulating materials can develop highly conducting domain walls (Fig. 1).^{26–28} These discoveries are driving the research activities on ferroics toward the local scale and facilitate conceptually new nanotechnology concepts;^{16,23,29} ferroic domain walls are either leveraged as active elements within devices^{30–32} or the domain walls themselves become the device, emulating the behavior of electronic components at the atomic scale.^{33,34} In a more recent development, this trend has been expanded toward other types of nanoscale objects in ferroics, exploring local functional properties that arise due to specific types of point defects and topologically non-trivial structures, such as disclinations, dislocations, vortices, and skyrmions,^{1,23,35–40} offering completely new and exciting perspectives for both fundamental and applied research on ferroic materials.

III. OVERVIEW OF TOPICS

A. Ferroic domains

Ferroic domains are of direct interest as small-sized units for processing or storing information as briefly discussed in Sec. II. Technological opportunities are, however, much wider and domains offer multiple avenues for the development of tunable devices. Thus, understanding their formation and local responses to external stimuli is of essential interest for the field. Kondovych *et al.* study the domain formation of ferroelectric domains under local electric fields in thin films with in-plane polarization.⁴¹ Govinden *et al.* investigate the impact of depolarization fields on ferroelectric domains,⁴² while strain⁴³ and thickness⁴⁴ effects are discussed in the works by Ichinose *et al.* and Feng *et al.*, respectively. Utilizing the impact of varying structural boundary conditions on ferroelectric domains, Geng *et al.* control polar nanodomains in BiFeO₃

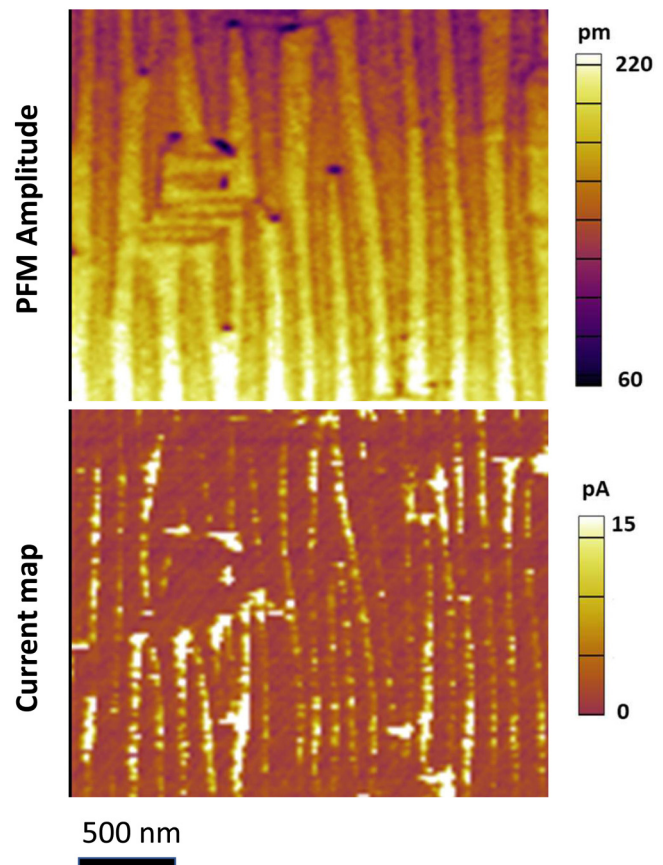


FIG. 1. Piezoresponse force microscopy (PFM) amplitude (top) and conductive atomic force microscopy (cAFM) current map (bottom) of an epitaxial BiFeO₃ thin film deposited on the La_{0.67}Sr_{0.33}MnO₃-buffered SrTiO₃ (001) substrate, showing conductive domain walls. Image courtesy of Q. Zhang and D. Sando, University of New South Wales (UNSW).

multilayers,⁴⁵ Chen *et al.* manipulate ferroelectric domains in patterned arrays of Pb(Zr_{0.7}Ti_{0.3})O₃ (PZT) nanoislands,⁴⁶ and Picht *et al.* apply grain size effects in PZT ceramics.⁴⁷ Domain engineering in LiNbO₃ by electron beam irradiation is discussed by Kokhanchik and co-workers,⁴⁸ and the relation between the temperature and the electric-field response of ferroelectric domains in LiNbO₃ is studied by Liu *et al.*⁴⁹ Monodomain states in ferroelectric heterostructures⁵⁰ and new opportunities for controlling the ferroelectric order via superlattice structures⁵¹ are addressed in the work by Mahjoub *et al.* and Strkalj *et al.*, respectively. Going beyond local aspects, domain engineering is well-established and widely applied to control the response of ferroic materials at the macro-scale. To better understand the relation between structure, mesoscopic-scale domains, and macroscopic properties, Zhao *et al.* study the domain structure under electric fields in (1-x)Bi_{0.5}Na_{0.5}TiO₃-(x)BaTiO₃ ceramics⁵² and Nishiyama *et al.* measure the large-signal piezoelectric properties of (Li,Na,K)NbO₃-based ceramics under combined electrical and mechanical loadings.⁵³

The collection of research articles is extended toward magnetism by Tianen and Jin, who revisit the cellular magnetic domains in Fe–Ga alloys,⁵⁴ and antiferromagnetic disks are analyzed by Silva *et al.* with a focus on magnetic skyrmions and their dynamics.⁵⁵ Cross coupling phenomena between electric fields and magnetic domains are tackled by Antipin *et al.* by studying the electric-field-induced formation of magnetic domains in iron garnet thin films.⁵⁶ Ghidini and co-workers transfer voltage-driven strain to polycrystalline Ni films using a single-crystal PMN–PT substrate, demonstrating a new potential pathway toward low-power magnetoelectric memory technology.⁵⁷ The coexistence of multiple (anti-)ferroic order parameters and related magnetic domains in hexaferrites are investigated by Ueda *et al.*, shedding new light on magnetoelectric coupling in multiferroics.⁵⁸ Using pulsed laser deposition, Liu *et al.* synthesize multiferroic Bi₂FeCrO₆ thin films and study the ferroelectric domain structures and their response to electrical fields.⁵⁹

In addition to the fundamental physical properties of ferroic domains, their characterization by different microscopy tools is covered in the Special Topic. A comprehensive tutorial about piezoresponse force microscopy (PFM) imaging and local hysteresis loop measurement is given by Hong.⁶⁰ Hiranaga *et al.* propose scanning nonlinear dielectric microscopy for visualizing ferroelectric switching,⁶¹ and Solís Canto and co-workers explore the impact of the cantilever spring constant and frequency in PFM-based domain studies using BiFeO₃ as an instructive model system.⁶² Furthermore, the perspective of Park *et al.* takes an application-oriented view of devices with a focus on fluoride-structured ferroelectrics, discussing the state of the art of the field, open challenges, and future opportunities.⁶³

B. Ferroic domain walls

The emergence of superconductivity at ferroelastic domain walls,²⁶ enhanced electronic conductivity at ferroelectric domain walls,²⁷ and pronounced spin torque effects at ferromagnetic domain walls⁶⁴ are fascinating examples for the functional phenomena that arise at ferroic domain walls. For readers interested in a broader introduction to the fundamentals, the physical properties of ferroic domain walls and related device applications, we refer to recent review articles that address these particular aspects.^{16,23,29,31,32,65,66} In this Special Topic, a snapshot of current research directions is presented that reflects the broad activities, ranging from advanced characterization techniques and property analysis to novel application opportunities. In their tutorial, Cherifi-Hertel *et al.* discuss second harmonic generation (SHG) as a powerful tool for studying ferroelectric domain walls, which provides access to the three-dimensional domain wall structure and their local symmetry.⁶⁷ Yokota and Uesu apply SHG to ferroelastic walls, comparing the local nonlinear optical response at domain walls in CaTiO₃, LaAlO₃, Pb₃(PO₄)₂, and BiVO₄.⁶⁸ Novel insight into the contrast formation in SHG microscopy experiments at domain walls is presented by Sychala *et al.*⁶⁹ By combining PFM, three-dimensional x-ray diffraction, and phase-field simulations, Schmidbauer *et al.* study the 3D patterns of ferroelectric domain walls in K_{0.9}Na_{0.1}NbO₃.⁷⁰ A tutorial about scanning electron microscopy (SEM) for domain wall investigations and

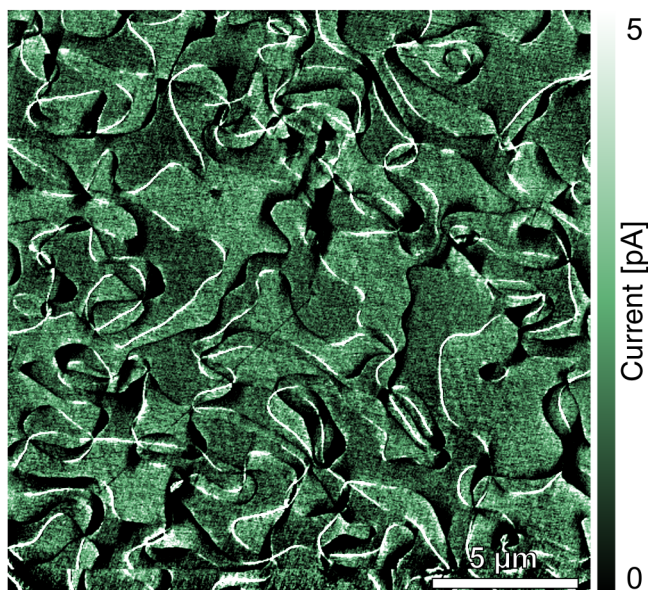


FIG. 2. Network of conducting (bright) and insulating (dark) ferroelectric domain walls in the hexagonal manganite ErMnO₃ mapped by conductive atomic force microscopy. Image courtesy of J. Schultheiß and D. Meier, Norwegian University of Science and Technology (NTNU).

opportunities that arise when correlating SEM with other techniques, such as focused ion beam (FIB) and atom probe tomography, is presented by Hunnstad *et al.*⁷¹ To resolve differences at the atomic scale between charged and uncharged domain walls in BiFeO₃ ceramics, Condurache *et al.* apply high-angle annular-dark-field scanning-transmission electron microscopy (HAADF-STEM).⁷²

Emergent physical phenomena at ferroic domain walls, the progress in measuring their properties, and the potential of ferroelastic domain walls for the development of new device concepts are discussed by Salje.⁷³ One interesting example is the polar properties that arise at anti-phase boundaries in SrTiO₃ and PbZrO₃, which are scrutinized by Schranz *et al.*⁷⁴ An innovative approach for stabilizing charged ferroelastic domain walls via self-assembled nano-islands in BiFeO₃ is presented by Chen *et al.*⁷⁵ Puntigam *et al.* demonstrate that insulating improper ferroelectric domain walls in hexagonal manganites (Fig. 2) give rise to an internal barrier layer capacitance, which may be used for engineering materials with colossal dielectric permittivities.⁷⁶ In Fe-doped BaTiO₃, Noguchi *et al.* report domain walls contributing to the overall photo-voltaic response⁷⁷ and Lundh *et al.* investigate local heating effects that arise due to electric-field driven domain wall movements in PZT thin films.⁷⁸ The possibility of pinning ferroelectric domain walls via defects in BiFeO₃ thin films to achieve improved polarization retention is explored by Zhang and co-workers.⁷⁹

Liu *et al.* expand the discussion toward magnetic domain walls and vortices by applying a six-state clock model, analogous to previous studies on ferroelectric domain walls and related topological

phenomena in hexagonal manganites.⁸⁰ A more general discussion about topological structures and related phenomena in ferroelectric oxides is given in the perspective by Ma and co-authors.⁸¹

C. Dynamical phenomena

The dynamic characteristics of ferroic domains and domain walls introduce the element of spatial mobility, allowing for real-time adjustment of their position, morphology, and density. This mobility in combination with the pronounced responses to external stimuli (e.g., strain and electric/magnetic fields) represents an important degree of flexibility that enables control of both the local and macroscopic properties of ferroic materials. Performing systematic studies on chemically doped BiFeO₃ ceramics, Makarovic *et al.* reveal how the density of conducting ferroelectric domain walls impacts the macroscopic nonlinear piezoelectric response.⁸² Liu *et al.* synthesize ferroelectric (K,Na)NbO₃ ceramics with different concentrations of point defects to control the domain wall motion and investigate the influence of the microstructure and electric properties,⁸³ while Adhikary and Ranjan demonstrate composition-driven changes in the electric field response of (x)Na_{0.5}Bi_{0.5}TiO₃-(1-x)K_{0.5}Bi_{0.5}TiO₃.⁸⁴ The growth dynamics of isolated ferroelectric domains with charged domain walls and the formation of self-assembled domain arrays in LiNbO₃ are presented by Shur *et al.*⁸⁵ By addressing the domain physics in relaxor ferroelectrics, Liu *et al.* explore the domain structures and electromechanical properties in the a.c. electric field-poled Pb(In_{0.5}Nb_{0.5})O₃-Pb(Mg_{0.33}Nb_{0.66})O₃-PbTiO₃.⁸⁶ In general, ferroic materials exhibit pronounced nonlinear responses, which Riemer and co-authors discuss for the case of different types of ferroelectric materials, including hard and soft systems, as well as relaxors; the physical origins of such nonlinearities, a mathematical formalism for their analysis, and experimental approaches are presented.⁸⁷ More fundamental theoretical investigations regarding the role of interface boundaries for the formation of two-phase states is presented by Levanyuk *et al.*⁸⁸ In addition, dynamical phenomena closely related to domain switching are investigated, such as charge injection and dynamics in PZT and the electroresistance effect in BaTiO₃, presented by Wang *et al.*⁸⁹ and Zhang *et al.*,⁹⁰ respectively.

Another important direction is the research on ferroic systems that do not belong to the perovskite family, searching for novel materials with superior functional properties. Here, hafnium oxide (HfO₂)-based compounds play an increasingly important role, promoting fast switching, high scalability, and reduced power consumption. A perspective discussing the switching behavior and the application potential of HfO₂-based ferroelectrics is given by Wang *et al.*⁹¹ Park and co-workers look into the specifics of defects in Si-doped HfO₂ thin films, concluding a substantial correlation between the switching dynamics and oxygen vacancies.⁹² Another promising class of materials in this context are ferroelectric wurtzite-type semiconductors, which exhibit a reversible ferroelectric polarization as Wolff *et al.* show at the unit cell length scale for Al_{0.75}Sc_{0.25}N thin films.⁹³

IV. CONCLUSIONS

In summary, the research on ferroic materials has substantially grown, but it is still far from being fully explored. Novel phases,

physical effects, and functional properties are continuously discovered, and the exploration of new systems often goes hand in hand with technological breakthroughs. Considering the trends in the field, the development of 2D ferroic materials sticks out as an appealing pathway toward a new generation of ferroic-based electronics as discussed in the perspective on 2D layered materials by Li *et al.*⁹⁴ In addition to improved electronic performance, sustainability aspects will play in the future a more and more important role for the research on ferroics and materials science in general. This new focus on sustainability will impose conceptually different challenges and requires new ways of thinking that inevitably will drive the field into unexplored directions; most importantly, it will give us the chance to revise our scientific strategies and establish novel functional ferroic materials for a more sustainable future.

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