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Nanoscale studies of ferroelectric domain walls as pinned elastic interfaces

*Études nanoscopiques de parois de domaines ferroélectriques comme interfaces élastiques piégées*

Patrycja Paruch*, Jill Guyonnet

MaNEP-DPMC, Université de Genève, 24, quai Ernest-Ansermet, 1211 Geneva, Switzerland

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ABSTRACT

The competition between elasticity and pinning of an interface in a fluctuating potential energy landscape gives rise to characteristic self-affine roughening and a complex dynamic response to applied forces. This statistical physics approach provides a general framework in which the behaviour of systems as diverse as propagating fractures, wetting lines, burning fronts or surface growth can be described. Domain walls separating regions with different polarisation orientation in ferroelectric materials are another example of pinned elastic interfaces, and can serve as a particularly useful model system. Reciprocally, a better understanding of this fundamental physics allows key parameters controlling domain switching, growth, and stability to be determined, and used to improve the performance of ferroelectric materials in applications such as memories, sensors, and actuators. In this review, we focus on piezoresponse force microscopy measurements of individual ferroelectric domain walls, allowing their static configuration and dynamic response to be accessed with nanoscale resolution over multiple orders of length scale and velocity. Combined with precise control over the applied electric field, temperature, and strain, and the ability to influence the type and density of defects present in the sample, this experimental system has allowed not only a direct demonstration of creep motion and roughening, but provides an opportunity to test less-well-understood aspects of out-of-equilibrium behaviour, and the effects of greater complexity in the structure of both the interface and the disorder landscape pinning it.

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R É S U M É

La compétition entre l'élasticité d'une interface et le piégeage par un potentiel désordonné confère à cette dernière une configuration rugueuse auto-affine caractéristique ainsi qu'une réponse dynamique complexe aux forces externes. Cette approche de physique statistique fournit une description théorique générale du comportement de systèmes aussi divers que la propagation de fractures, les lignes de mouillage, les fronts de combustion et les phénomènes de croissance de surface. Dans les matériaux ferroélectriques, les parois de domaines, qui séparent les régions où la polarisation est orientée différemment, forment un autre exemple d'interfaces élastiques piégées, et constituent à ce titre un système modèle particulièrement utile. Réciproquement, une meilleure compréhension de ces propriétés physiques fondamentales permet de déterminer les paramètres-clés contrôlant

* Corresponding author.

E-mail address: Patrycja.Paruch@unige.ch (P. Paruch).

la nucléation, la croissance et la stabilité des domaines et, de ce fait, l'amélioration des performances des matériaux ferroélectriques pour des applications telles que mémoires, senseurs et actionneurs. Dans cette revue, nous nous focalisons sur des mesures de parois de domaines ferroélectriques individuelles par microscopie à force atomique en mode piézoréponse, qui permettent de déterminer leur configuration statique et leur réponse dynamique avec une résolution nanométrique sur plusieurs ordres de grandeur de longueur et de vitesse. Combiné au contrôle précis du champ électrique appliqué, de la température, de la contrainte, et de la nature et densité des défauts présents dans l'échantillon, ce système expérimental permet non seulement une démonstration directe des phénomènes de reptation et de rugosité, mais également d'appréhender certains aspects moins connus de phénomènes hors équilibre ainsi que les effets d'une structure plus complexe, tant au niveau de l'interface elle-même que du potentiel désordonné de piégeage.

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1. Introduction

Ferroelectric materials are characterised by symmetry-equivalent ground states with different orientation of the non-volatile macroscopic electric dipole moment, or polarisation, which may be switched by the application of an external electric field. Depending on the strain and electrostatic boundary conditions, as well as its switching history, a sample may present a uniformly polarised configuration, or coexisting regions with different polarisation orientation, known as domains, separated by domain walls [1–3]. As thin as 1–2 unit cells (<1 nm) [4,5], the domain walls, shown in Fig. 1, can be thought of as extended topological defects, with different electronic and structural symmetry from their parent materials as a result of the rotation or local absence of polarisation. The application of an external electric field along the polarisation axis asymmetrises the ferroelectric double-well potential, and promotes the growth of domains with the preferred orientation, thus driving domain wall motion.

Understanding the static and dynamic behaviour of domain walls, and specifically their interaction with commensurate (crystal lattice) or random (defects/disorder) pinning sites is key to predicting and controlling domain switching, growth and stability, all questions of high technological as well as fundamental interest. In particular, the switchable remanent polarisation, together with associated pyroelectric and piezoelectric properties, has led to the integration of ferroelectrics in devices ranging from memories [6,7] to micro- and nano-electromechanical sensors and actuators [8–10], and numerous electro-optic applications [11]. In addition, recent discoveries of unusual functional properties localised specifically at the domain walls [12–14] make them extremely promising as potential active device components in a future domain-wall-based nanoelectronics.

From a theoretical point of view, the effects of complex, heterogeneous disorder inherent to real samples at multiple orders of length scales are difficult to incorporate explicitly. Analytical solutions of Ginzburg–Landau mean-field-type calculations provide invaluable information about the thermodynamics and kinetics of defect-free domain switching [15–18], and models of domain walls [3] and their interactions with single defects [19], or with particularly regular, periodically ordered pinning potentials [20], but do not readily describe the inherent randomness. First-principles atomistic simulations, meanwhile, give extremely accurate estimates of domain wall energies and interactions with individual defects [4,21–23], but do not easily scale up beyond super cells of hundreds of atoms. Rather, to tackle the effects of random fluctuations at multiple length scales, a statistical description is necessary. Such a statistical, highly reductionist approach to domain walls within the general framework of disordered elastic systems has proven very powerful, allowing metastability and the glassy physics associated with an energy landscape characterised by multiple local minima to be addressed [24–26]. Conversely, as a result of the high resolution of scanned probe microscopy, combined with access to a broad range of control parameters such as the applied electric field, temperature, environmental (electrochemical) and strain boundary conditions, ferroelectric domain walls present a useful model system for testing theoretical predictions about pinned elastic interfaces.

Within this framework, the behaviour of such interfaces is governed by the competition between elasticity, which tends to flatten the interface, and pinning, due to fluctuations in the potential energy landscape, leading to roughening and a complex response to an applied force. The scaling of the roughness in the static equilibrium configuration, as well as of the quasi-static thermally activated dynamics for low driving forces, and of the transition to depinning at higher forces, are universal phenomena characterised by exponents whose values are linked to the dimensionality of the system and the universality class of the disorder. Although the equilibrium properties of such systems are relatively well established, much less is known about their out-of-equilibrium behaviour, including ageing and memory effects. In addition, the interaction of an interface with a more complex, highly heterogeneous disorder landscape, as well as the consequences of its potentially intricate internal structure remain poorly understood. Ferroelectric domain walls in epitaxial perovskite (multi)ferroic thin films, with potentially complex internal structure and coupling between different order parameters, and where a variety of defects can be introduced during or after growth can therefore allow these intriguing phenomena to be investigated, and form the subject of this review.

We begin with an overview of the physics of elastic interfaces in disordered media in Section 2, then discuss in Section 3 how this theoretical framework was specifically applied to ferroelectric domain walls, with initial indications of domain

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