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Mild and wild ferroelectrics and their potential role in neuromorphic computation

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ABSTRACT

In this Perspective, two interrelated new developments are discussed. The first relates to a much better understanding of the actual movement of domain walls during switching. Ferroelectric and ferroelastic domain movements proceed via the combination of jerky and smooth displacements of domain walls. A careful separation of these two mechanisms into "wild" and "mild" is crucial for the understanding of avalanches in ferroelectrics. Avalanche switching involves jerky domain wall movements and leads to singularities in the switching current. During avalanches, domain walls enhance and localize atomic transport and generate magnetism emerging from mobile kinks in the walls. The second development is based on the transport of dopants inside domain walls during nano-fabrication of devices. Progressing domain walls in electric fields can then—mainly in the case of wild wall movements—connect defect "reservoirs" similar to synapses connecting neurons in the brain. The walls take the role of synapses, and the defect clusters take that of neurons. The combination of fast moving domain walls and chemical transport inside the walls constitutes, therefore, ingredients for memristive device elements in neuromorphic computers. This application is predicted to play a major future role in ferroelectricity.

I. INTRODUCTION

Ferroelectricity is defined by the switching of polarization and, thus, by the movement of ferroelectric domain boundaries. The equivalent definition holds for ferroelastic materials where the switching occurs between different strain states (Salje, 2012). Many ferroic materials, such as archetypal $BaTiO_3$, are both ferroelastic and ferroelectric with respect to the switching of 90° boundaries, while they are almost exclusively ferroelectric for 180° boundaries. The switching of 90° boundaries (Ishibashi and Salje, 2002) often requires more strain energy than electrical energy so that the evaluation of the energy transfer $\int PdE$ over the full hysteresis loop tells us more about the inherent strain interactions than about the electric interactions. Moreover, weak ferroelectricity occurs in specific ferroelastic patterns, while the bulk material is purely ferroelastic. Ferroelectricity and piezoelectricity stem in these cases entirely from rearrangements of polar ferroelastic twin walls (Lu *et al.*, 2020a; 2019a; 2019b; 2019c) and are unrelated to the crystal structure of the bulk. Wall polarity in non-polar materials was predicted already by Goncalves-Ferreira *et al.* (2008) and Salje (2010) and first direct observed by Van Aert *et al.* (2012). It is also now established that the vast majority of those ferroelastic domain walls that have been tested contain polarity (switchable or not) (e.g., Hlinka *et al.*, 2016) depending on the symmetry of the bulk and the domain walls (Salje *et al.*, 2016a; Schiaffino and Stengel, 2017). Ferroelectric switching is not only relevant in the case of binary switching. A ferroelectric thin film deposited with some strain mismatch on substrates develops complex domain patterns (Tagantsev *et al.*, 2002; Pertsev *et al.*, 1998) including a multitude of intermediate (meta-)stable configurations, which allow for non-binary ferroelectric switching. In adaptive domain structures, these multi-state logical applications are at the heart of the design of domain walls with three states in most single crystals and more states in realistic domain configurations close to the morphotropic phase boundary (Viehland and Salje, 2014). Multibit hysteresis in such configurations was described in detail by Baudry *et al.* (2017).

This result demonstrates that multistate switching is possible in twinned ferroelectric materials when deposited as thin films. Multistate effects are based on the complex energy landscape of the ferroelectric device. This complex behavior does not, however, explain the possible dynamics of the switching where possible pathways may not be realized simply because they are dynamically blocked by high activation energies or other reasons. Domain wall dynamics is now considered in its most basic approach.

II. MILD AND WILD HYSTERESIS

Several new developments have invigorated the investigation of domain switching in ferroelectrics and ferroelastics. I wish to elucidate here some further developments that are likely to have a major effect on the evolution of neuromorphic (brain-like) computation. In order to do so, let me first elucidate some necessary related developments, which are crucial for the use of domain wall movements for such device applications. They all relate to the movements of the domain walls under fields and the way they can connect chemical dopants between reservoirs (Lee and Salje, 2005; Lee *et al.*, 2005).

The first breakthrough was the quantitative observation of the dynamics of domain switching. After first observations of jerk-like switching, similar to Barkhausen noise (Shur *et al.*, 2002) and the measurement of jerky propagation of needle domains under stress in ferroelastics (Harrison and Salje, 2010) and the investigation of acoustic noise in martensites (Salje *et al.*, 2011), a full set of dynamical switching parameters in $BaTiO_3$ and some other ferroelectric materials were measured (Salje *et al.*, 2019; Xu *et al.*, 2020). In most studies, the switching currents constituted a first indicator for avalanche switching dynamics (Tan *et al.*, 2019; Casals *et al.*, 2020). The time resolution of more detailed investigations was massively improved by using acoustic emission (AE) techniques to measure the switching energy, amplitude, time sequence, aftershock probability, and correlations (Salje *et al.*, 2019). The results of these experiments show that switching proceeds by avalanches of correlated domain wall movements where a full set of avalanche parameters could be determined (Salje and Dahmen, 2014). In addition, much milder, smoother domain propagation (Zhang *et al.*, 2020) coexists. This schism is captured by the notion that we observe "wild" and "mild" processes where wild means that spiky energy emission, the so-called "jerks," dominates the domain wall movements (Yang *et al.*, 2020; Weiss *et al.*, 2015). These movements constitute avalanches in the description of the work of Salje and Dahmen (2014). Coexistence of mild and wild movements is well known for restructuring processes in many materials under external forcing, such as ice (Weiss, 2019) and martensites (Chen *et al.*, 2019) and in crack propagation (Bonamy *et al.*, 2008; Laurson *et al.*, 2010). Mild processes are much more difficult to observe than spiky jerks (Casals *et al.*, 2019) where the optical observation of domain wall movements proved particularly useful (Casals *et al.*, 2020). Mild movements produce very little strain although they are potentially visible in AE at a very low noise level.

In summary, avalanche switching generates jerks in the depolarization current, the acoustic emission, and other macroscopic parameters, which are manifest by spikes in almost all response functions. In contrast to these wild events, mild events do not generate significant spikes and are often seen as background noise. Nevertheless, they also relate to domain wall movements, albeit these movements are smoother but still change the fractal dimension of the domain pattern (Catalan *et al.*, 2008).

III. DOMAIN WALLS CARRY CHEMICAL CURRENTS

The second development relates to the current in domain walls and associated chemical changes. Ever since the discovery of superconductivity in domain walls (Aird and Salje, 1998) and subsequent studies of highly conducting walls (Seidel *et al.*, 2010), the concept of domain wall electronics was developed rapidly and was reviewed by Catalan *et al.* (2012) and Evans *et al.* (2020).

An important step forward was the idea that domain wall transport includes chemical changes during electronic conduction. This impacts on the origin of memristor properties of ferroelastic domain walls (Bibes *et al.*, 2008; Garcia *et al.*, 2009). In fact, networks of ferroelectric domains have the same properties as arrays of memristors (Chanthbouala *et al.*, 2012). One typical nanostructure is a needle domain approaching a perpendicular wall, leading to the formation of a junction between domain walls and tweed microstructures (Hayward and Salje, 1998; Salje and Parlinski, 1991; and Salje *et al.*, 2016b). If the walls are superconducting, the connection between the two walls' orientations (Fig. 1) constitutes a Josephson junction. An additional effect is that the current provokes chemical changes in the walls or in the needle domain itself. Such modifications may lead to a percolation criticality when the needle touches an orthogonal wall, an interface, or the surface of the sample (Novak *et al.*, 2002).

A typical example is the case of WO_3 where oxygen depletion promotes superconductivity. Even at the room temperature, domain boundaries are highly conducting, while the bulk is a wide gap semiconductor (Kim *et al.*, 2010), as shown in Figs. 2 and 3. In a different approach, Aird and Salje (2000) systematically doped twinned WO_3 crystals with Na. The enrichment proceeds via the diffusion from a Na-gas phase surrounding the sample and is easily detected optically and via chemical microprobe analysis (see Fig. 4). The dark colored domain walls in Fig. 4 are doped with Na, and the doping level is

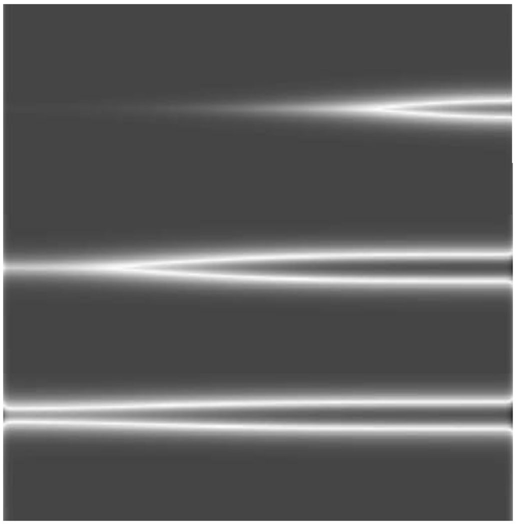

FIG. 1. Attraction of a needle domain to a surface or an interface. A conductivity spike occurs when the connection is made between the walls and the interface if atoms are transported inside the domain walls (computation after the work of Novak *et al.*, 2002).

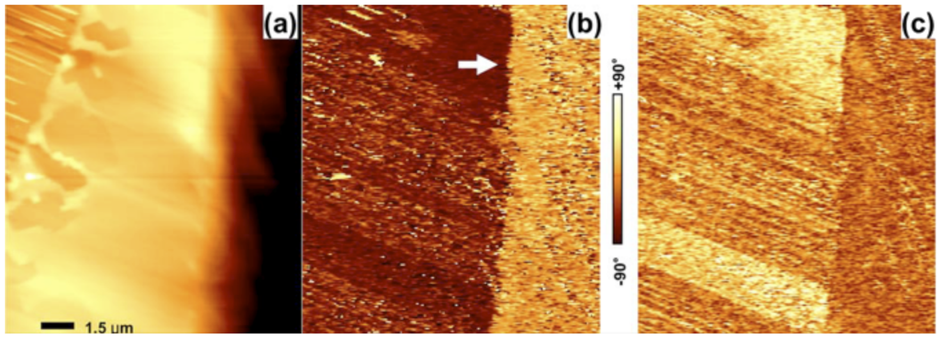

FIG. 2. PFM images of WO3 at room temperature: (a) Topography, (b) VPFM phase, and (c) VPFM amplitude of a reduced tungsten oxide crystal. The twin boundary is clearly visible by the phase change in the VPFM images. No significant change in the in-plane component at the twin boundary occurs in LPFM images. The white arrow in panel (b) represents the position of the twin boundaries. After Kim *et al.* (2010).

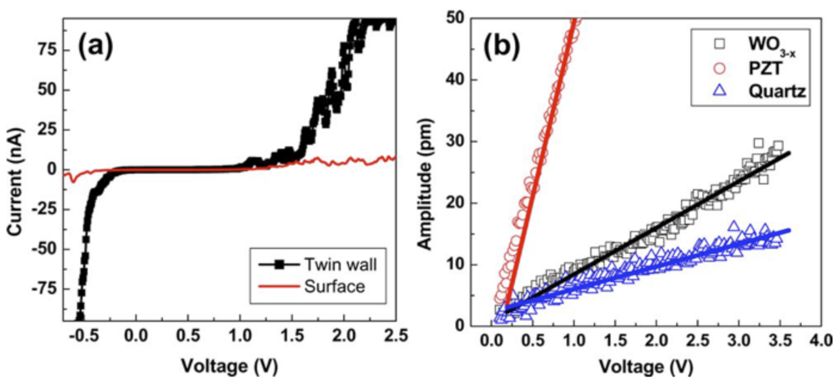

FIG. 3. I–V characteristics acquired on the twin wall (a) and on the free surface of a freshly reduced WO_{3-x} crystal. The diode effect is very large with critical voltages near −0.1 V and +1.7 V. Memristive behavior is superimposed on a piezoelectric response. (b) The piezoresponse amplitude is a function of the ac voltage applied to the conductive probe of a PZT epitaxial thin film, an x-cut quartz, and the reduced surface layer of a WO3−x single crystal. After Kim *et al.* (2010).

maximum 12% (Na/W ratio). Both oxygen and Na are mobile above the room temperature so that WO_3 domain boundaries can be utilized to inject or retract defects into un-doped domain boundaries and thereby dramatically modulate the conductivity of an array of domain boundaries. It may be mentioned that other ions such as Li and K equally generate superconductivity.

For AFM/PFM studies, it is important that the symmetry of the surface layer given by Kim *et al.* (2010) was found to be higher than that of the core. The orthogonal orientation of the twin boundaries indicates orthorhombic or tetragonal symmetry. A direct experimental evidence for the identity of the surface layer stems from the observation of a strong local piezoelectric activity measured by PFM, as shown in Fig. 2. The triclinic phase of the core does not have any transverse or vertical piezoelectric activity; thus, no piezoelectric effect is to be expected. However, a strong piezoresponse was clearly observed in the vertical out-of-plane PFM mode. The piezoelectric activity in the VPFM images shown in Figs. 2(a) and 2(c) is due to a genuine surface effect originating from the reduced surface layer rather than from the crystal bulk. All but two phases in WO_3 are centrosymmetric, and the only piezoelectric phases are the monoclinic ε phase (with weak piezoelectricity) and the tetragonal phase $P42_1m$ with very strong piezoelectricity (Hamdi *et al.*, 2016). The chemical composition of the tetragonal phase was reported to be around $WO_{2.95}$ depending on the reducing conditions during the sample preparation. When this surface is contacted with another sample, the chemical potential gradient will induce ionic transport, which can be modulated by electric fields. This biased transport is the key ingredient for memristive devices.

It is then left to the ingenuity of the engineers to design the appropriate chemical changes in such a way that the percolation induces a strong increase in the filamentary conductivity between walls. When the wall retracts, the circuit is broken so that an asymmetric memory device can be constructed. This fundamental idea was already proposed in 2012 by Chanthbouala *et al.* and has been worked upon ever since. The key ingredient is now that chemical modifications can, in principle, be constrained to the domain walls where the chemical mobility is high, while the mobility remains low inside the bulk. Erasable conductive domain walls in insulating ferroelectric thin films can hence be used for the non-destructive electrical readout of the polarization states in ferroelectric memories. However, domain-wall currents based on these devices seem not to have reached the intensity and stability required to drive readout circuits operating at high speeds. First results using specific domain-wall configurations in epitaxial $BiFeO_3$ thin films show great promise (Jiang *et al.*, 2018).

IV. NEUROMORPHIC COMPUTATION

Combining the two developments, namely, the recognition of wild and mild mobilities of the domains and the targeted

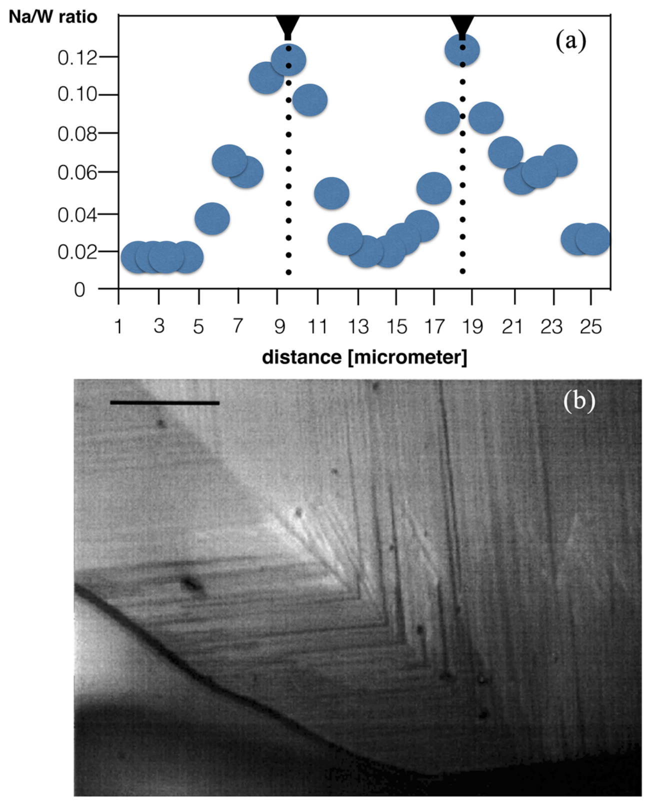

FIG. 4. (a) Chemical composition and (b) optical image of reacted monoclinic–orthorhombic boundaries in WO_3. The scale bar is 50 μm. The black lines indicate the Na charged regions. Note the intersection between the horizontal and vertical twin boundaries in a diagonal boundary where Na transport occurred between the two twin systems. The graph (a) shows the enrichment by Na over W in two adjacent domain walls indicated by the dotted lines. After Aird and Salje (2000).

chemical injection by domain walls (e.g., Lee and Salje, 2005), we can now discuss applications of ferroelectrics as proposed by Chaudhary *et al.* (2020), Chanthbouala *et al.* (2012), and McConville *et al.* (2020) in the field of neuromorphic computing. The memristor-type formation of filaments inducing conduction spikes near a percolation point (Dongale *et al.*, 2018) can, potentially, be replaced by injected chemical domain wall conductivity. This has several advantages. First, the chemical changes are strongly confined to interfaces (Nataf *et al.*, 2020; Salje, 2010) and can be identified by spectroscopy (Salje, 1992). Even though percolations will presumably generate Schottky barriers near junction areas, these barriers are limited to atomic size patches and will relax even at high operating frequencies. Moreover, ferroelectric walls and highly conducting walls (such as observed in WO_3) can help to overcome the bottleneck between memory and synaptic data transfer. Neuromorphic computers imitate the functionality of brain neurons and connecting synapses, where the synapses combine logical operations with memory effects. For this purpose, the dual functionality of the domain walls may be the key ingredient. Injected defects can stop transport currents in domain walls. Furthermore, synapses are dynamical elements, which is also the case for mobile, jerky domain walls.

The path of the electrical current can be switched at the percolation point. The percolation point is modified by shifting the position of the domain wall or of defects, which blocks the transport of atoms along the domain walls. An external signal by an electric field strain (e.g., via defect movements) can then greatly change the ionic current (Lu *et al.*, 2019a). The changes occur rapidly because the walls' movements in jerks are extremely fast (Zhang *et al.*, 2014). Equally, the connection and the blocking of atomic transport can occur over extremely short time intervals (Salje *et al.*, 2017; Sharma *et al.*, 2017; Jiang *et al.*, 2018; and Chai *et al.*, 2020). The work by Sharma *et al.* (2017) is particularly important and gives much hope for future developments. They demonstrated that by using nanofabricated electrodes and scanning probe techniques, a prototypic non-volatile ferroelectric memory element entirely based on domain boundaries can be constructed. The element was scalable to below 100 μm. The binary memory element is the conductivity which is present or absent in the boundary. They demonstrated that the device could be read non-destructively at less than 3 V with an on–off ratio of ~1000.

Much work has been directed into such use of ferroelectric domain boundary engineering in neuromorphic computation over the last decade, but several key issues still remain obscure. First, ferroelectric and ferroelastic domains and domain walls generate many complex domain patterns (Scott, 2020). Each domain boundary can carry specific chemical loading, which can be tailored to change the percolation point for the memristive carrier transport. This problem is well known, but no systematic assessment and catalogs of mobile defect species and their mobilities inside complex patterns of domain boundaries are yet available. Furthermore, ferroelectric and ferroelastic domain patterns are well described as post-mortem objects, while no robust concept exits, which allows us to understand the loading and unloading during pattern formation. It is also unknown how the loading of domain walls by mobile species (e.g., from the bulk) can be undertaken in a reliable way. Even additional magnetic interactions, generated by the polar and rough domain boundaries, have been postulated (Lu *et al.*, 2020b) but not confirmed experimentally.

In summary, research of classic ferroelectricity has come a long way to be understood theoretically. Furthermore, ferroelectric devices were designed, helped by simulations on various levels of sophistication, and commercialized. New challenges have also arisen, and new directions become clearer. Great progress is possible but is not certain, combining a more detailed knowledge of the behavior of ferroelectrics and ferroelastics and their domain boundary dynamics with the intricacies of neuromorphic computation. Novel tools exist in the field of ferroelectrics, and much progress has been made in understanding memristive switching. Some groundwork for computer applications has been laid, but the great breakthrough is still missing. It appears possible that synaptic ferroelectrics will be used in future neuromorphic computer designs, which greatly increases the applicability of ferroelectric materials. Applications in ferroelectricity are much more widespread than what could have been imagined 10 years ago. This includes the advent of ferroelectric Bloch-lines and highly structured ferroic surface layers (Salje and Carpenter, 2015; Salje and Scott, 2014). Research in ferroelectrics has greatly evolved, and novel questions have emerged. Research in ferroelectricity has branched out in many directions and will further expand vigorously.

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DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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