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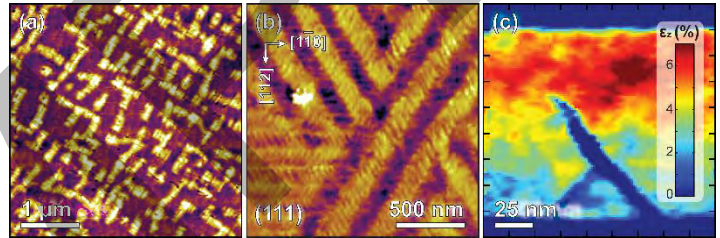
# Multidimensional Probes of Complex Functional Ferroelectric Domain Structures

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Advances in the control of interface chemistry and *in situ* monitoring/control of the growth processes have provided researchers the tools necessary to design complex ferroelectric domain structures. Leveraging these approaches, researchers have produced a wealth of polar structures including: ordered ferroelastic and mixed-phase domain structures, topological defects, and others. Understanding the structure and response of these complex domain structures requires the ability to perturb and observe their responses under relevant length- and time-scales. In turn, researchers are forced to conduct multidimensional experiments which in general are difficult to analyze “by hand.” Here, we discuss our recent progress using “big data” analytics to interpret multidimensional scanning transmission electron microscopy and scanning probe microscopy experiments to understand the structure and response ferroelectric materials. Particularly we focus our attention on three interesting candidate structures: a mixed-phase  $\text{PbTiO}_3$  structure, collective behavior in (111)-oriented  $\text{PbZrTiO}_3$ , and the shape, structure, and response of “springy” ferroelastic domains in compositionally-graded  $\text{PbZr}_{1-x}\text{Ti}_x\text{O}_3$ .

First, we explore mixed-phase in-plane oriented  $a_1/a_2/a_1/a_2$  and dense  $c/a/c/a$  domain structures in highly tensile strained  $\text{PbTiO}_3$  (Fig. 1a). Using band-excitation switching spectroscopy (BE-SS) we observe that it is possible to write out-of-plane oriented  $c$  structures using a locally bias; and explore the changes in the local switching that occur during these ferroelastic transitions. We demonstrate the local interconvertibility of in-plane and out-of-plane oriented nanoscale structures. The ability to controllably write and erase out-of-plane polarized domains in an in-plane polarized matrix creates local structures with largely varying properties (*i.e.* piezoelectric, dielectric, elastic modulus, etc.) which are readily adaptable for memory devices.



**Figure 1.** (a) Topography showing mixed-phase  $\text{PbTiO}_3$  on  $\text{SmScO}_3$  (110). (b) Highly ordered nanotwinned domain structure in (111)-oriented  $\text{PbZr}_{0.2}\text{Ti}_{0.8}\text{O}_3$ . (c) Out-of-plane strain map of compositionally-graded  $\text{PbZr}_{1-x}\text{Ti}_x\text{O}_3$ .

Second, using BE-SS we explore collective switching behavior in (111)-oriented  $\text{PbZr}_{0.2}\text{Ti}_{0.8}\text{O}_3$  consisting of highly ordered nanotwinned domain structure (Fig. 1b). We observe a homogeneous nucleation bias inside the twin band across a number of nanodomains, different from the inhomogeneous nucleation typically observed in conventional ferroelectrics. This phenomenon suggests the presence of possible strong domain-domain coupling between nano-domains resulting in collective switching. This work provides new insights into the fundamental understanding of ferroelastic switching in ferroelectrics.

Finally, we utilize detailed nanobeam diffraction strain mapping (ND-SM) and BE-SS to explore the fine structure and response of ferroelastic domains in compositionally-graded  $\text{PbZr}_{1-x}\text{Ti}_x\text{O}_3$ . ND-SM reveals that the compositional and strain gradients modify the morphology of the ferroelastic domains making them appear more needle-like rather than parallelepiped-like (Fig. 1c). Subsequent studies of electric-field induced switching show that these needle-like domains are highly mobile; expanding (or contracting) from the free surface depending on the direction of the applied bias.<sup>1</sup> This unusual field-dependent response renders these domains highly mobile in the out-of-plane direction yet spatially localized in the plane; prompting them to be called “springy” in nature.<sup>2</sup> Such work highlights the ability to utilize strain gradients to finely control the structure, response, and properties of ferroelastic domains.

1. J. C. Agar, *et al.*, Highly-mobile ferroelastic domain walls in compositionally-graded ferroelectric thin films. *Nature Mater.* **15**, 549-556 (2016).
2. K. Dorr, Ferroelastic domains: springy expansion. *Nature Mater.* **15**, 497-498 (2016).