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Domain Structure and Properties in Inhomogeneously-Strained Ferroelectric Thin Films

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Epitaxial thin-film growth and the ability to deterministically apply lattice mismatch strain has enabled dramatic control over the structure and properties of a range of ferroelectric materials. Modern ferroelectric films, including bilayer and superlattice heterostructures, have also provided access to exotic structures and properties not available in the bulk. In this work, we focus on recent advances in our understanding of how strain can be manipulated and controlled to elicit new types of responses and new understandings about response in ferroelectric materials. In particular, we will explore new modalities of strain control of ferroelectric materials that go beyond traditional lattice mismatch effects and how this can be used to enhance performance, independently tune susceptibilities, and provide new insights into the nature of these complex materials. In particular, we will focus on the deterministic production of large strain gradients (on the order of $>10^{-5}$ m^{-1}) via purposeful compositional gradients. We will highlight work on compositionally-graded versions of $PbZr_{1-x}Ti_xO_3$ and $Ba_{1-x}Sr_xTiO_3$ where careful control of lattice mismatch and chemistry combine to produce large strain gradients, exotic properties, and new approaches to independently control traditionally coupled properties. As part of this discussion, we will explore the evolution of the crystal and domain structure as a function of the end-members of the compositional gradient, thickness of the film, and substrate. Advanced band-excitation piezoresponse force microscopy, switching spectroscopy, and non-linearity studies have been applied. These studies reveal both unexpected crystal and domain structures can be stabilized in these heterostructures and exotic low- and high-field responses can be obtained. Of particular interest will be the results of temperature-dependent probes of susceptibility which reveal large, nearly temperature-independent properties from 25-500C and the observation of highly-mobile ferroelastic domain wall structures which can give rise to local enhancement of susceptibilities. These observations could represent a ground-breaking advance in the performance of these materials.