

# Controlling Emergent Structures and Properties in Epitaxial Ferroelectric Films

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Modern approaches to epitaxial thin-film growth have enabled unprecedented control of ferroelectric materials including the realization of enhanced  $P_S$  and  $T_C$ , production of ordered domain structures, and improved properties. Today, however, we are looking beyond simple lattice mismatch control for new ways to manipulate and control ferroic response and to produce unexpected emergent effects. In this talk, we will investigate a number of observations of such emergent or unexpected properties in epitaxial thin films made possible via innovative synthesis and processing methodologies. In our recent work, we have observed a range of interesting effects in single-layer films, superlattices, and compositionally-graded versions of materials. For example, recent progress in the use of *in situ* and *ex situ* defect introduction has been observed to be effective in controlling defect-induced electronic states that can drive dramatic changes in leakage currents and improve ferroelectric response. Among the potential topics to be explore are further advances and understanding of the mechanism behind these effects and routes to create novel switching phenomena in ferroelectrics. Yet another example is the observation of epitaxial-strain-induced phase-separation in ferroelectric materials where, via a spinodal-like instability, self-assembled hierarchical domain structures are created under mild tensile strains. In turn, long-range elastic and electrostatic coupling can give rise to deterministic three-state switching and corresponding large electromechanical responses in these materials. Among the potential topics to be explored are new “big data” analyses of band-excitation switching spectroscopy data wherein novel data analysis approaches are used to extract additional details about the nature of this multi-state switching process. Still another example is work on ferroelectric superlattices, wherein emergent ferroelectric vortex phases have been observed. Among the potential topics to be explored is the recent observation of phase coexistence between this emergent electric toroidal phase and a classical  $a_1/a_2$  ferroic phase and the potential for field-driven susceptibilities in this system. Finally, in (111)-oriented versions of tetragonal ferroelectrics, we have identified unexpected thickness dependence of the coercive field. The role of complex, electrostatic- and elastically-frustrated domain structures in moderating this non-traditional response will also potentially be discussed. All told, the focus will be on highlighting novel approaches to the synthesis of these complex materials to enable new properties and effects in these systems.