

Domain scaling and coupling of structural distortions in tensile-strained PbTiO_3 heterostructures

Céline Lichtensteiger, Marios Hadjimichael, and Jean-Marc Triscone

DQMP - University of Geneva

In ferroelectric thin films, the complex interplay between mechanical and electrostatic boundary conditions allows for the formation of a large variety of domain structures with fascinating properties. Heterostructuring and careful tuning of the epitaxial strain allow for precise control of these boundary conditions, leading to the formation of novel domain structures such as polar merons in tensile-strained PbTiO_3 films and skyrmions in $\text{PbTiO}_3/\text{SrTiO}_3$ superlattices. The structural coupling across PbTiO_3 layers in these systems can also lead to the formation of complex three-dimensionally ordered supercrystal structures, recently observed in tensile-strained $\text{PbTiO}_3/\text{SrTiO}_3$ and $\text{PbTiO}_3/\text{SrRuO}_3$ superlattices. Domain structures in ferroelectric systems not only change the properties of the ferroelectric itself, but can also be used to change the properties of other materials through electrostatic and structural coupling.

Here, we study heterostructures of PbTiO_3 under tensile strain, deposited using off-axis RF magnetron sputtering. The films are epitaxially grown on (110)-oriented DyScO_3 substrates and are sandwiched between top and bottom 55 unit cell-thick SrRuO_3 layers. The tensile strain imposed by the substrate favours a ferroelastic domain structure, with PbTiO_3 adopting both in-plane and out-of-plane polarization orientations. Using a combination of x-ray diffraction (XRD) and atomic force microscopy (AFM), we study the domain structure in these systems as a function of PbTiO_3 layer thickness. We find that the anisotropic strain imposed by the orthorhombic substrate creates a large asymmetry in the domain configuration, with domain walls macroscopically aligned along one of the two in-plane directions. We show that the periodicity estimated by XRD as a function of the film thickness deviates from the Kittel law. Above a certain critical thickness, the large structural distortions associated with the ferroelastic domains propagate through the top SrRuO_3 layer, creating a modulated structure that extends beyond the ferroelectric layer thickness, with signatures observed both in XRD and AFM.

Our results shine light on the complexity of ferroelastic domain structures in PbTiO_3 -based multilayers and their sensitivity to both electrostatic and mechanical boundary conditions.