

Charge at ferroelectric interfaces

Scientific Achievement

Using *in situ* synchrotron x-ray scattering, we discovered that the equilibrium structure of ultrathin ferroelectric films involves nanoscale 180° stripe domains. This work, published in *Physical Review Letters* **89**, 067601 (2002), was the first observation of equilibrium stripe domains in epitaxial ferroelectric films. Three equilibrium domain arrangements were observed in epitaxially-strained films of PbTiO₃ on SrTiO₃, depending on film thickness and temperature: two stripe phases (F_{α} , F_{β}) having different periods in the 5 - 20 nm range, and a monodomain phase at lowest temperatures (F_{γ}). The formation of equilibrium stripes followed by a monodomain phase shows that the initially uncompensated interfaces of the ferroelectric film become compensated by free charge as the temperature is lowered. However, current theory does not explain compensation of the interface with the insulating substrate or of the free surface.

Significance

Changes in interfacial compensation with temperature suggest that we may be able to manipulate ferroelectricity using interface structure or chemistry, and vice versa. Using a combination of experiment and theory, we propose to understand and learn to control the compensation. Initial *ab initio* results suggest that ions can provide charge to compensate the free surface. Can we switch the adsorbed species at the surface by changing the polarization electrically? Can we switch the polarization of a film by changing the chemistry of its environment? We will develop predictions and investigate them using *in situ* x-ray scattering measurements. In addition, we have recently been able to image stripe domains by AFM. These images provide complementary information to scattering measurements, showing for example how the stripes can align with surface step edges. Understanding how to control stripe morphology and exchange of adsorbed ionic species will allow their use as templates for sub-lithographic-scale pattern formation.

Performers

D.D. Fong, R.-V. Wang, F. Jiang, P.H. Fuoss, S.K. Streiffer, J.A. Eastman, G.B. Stephenson (ANL-MSD)

K. Latifi, C. Thompson (Northern Illinois Univ.), A.M. Rappe (Univ. Pennsylvania), C.B. Eom (Univ. Wisconsin), K. R. Elder (Oakland Univ.)

