Nanoscale Origins of Ferroelastic Domain Wall Mobility in Ferroelectric Multilayers

Nagarajan Valanoor[1], Hsin-Hui Huang[1], Zijian Hong[2]‡, Huolin L. Xin [3]‡, Dong Su[3], Long-Qing Chen[2], Guanzhong Huang[3,4], and Paul R. Munroe[1]
1 School of Materials Science and Engineering, University of New South Wales, Sydney, NSW, 2052, Australia
2 Department of Materials Science and Engineering, Pennsylvania State University, University Park, Pennsylvania, 16802-5006, USA
3 Center for Functional Nanomaterials, Brookhaven National Laboratory, Upton, Brookhaven, New York, 11973, USA
4 Department of Materials Science and Engineering, Stony Brook University, Stony Brook, New York, 11794-3400, USA
Nagarajan Valanoor: nagarajan@unsw.edu.au

The nanoscale origins of ferroelastic domain wall motion in ferroelectric multilayer thin films that lead to giant electromechanical responses are investigated. We present direct evidence for complex underpinning factors that result in ferroelastic domain wall mobility using a combination of atomic-level aberration corrected scanning transmission electron microscopy and phase-field simulations in model epitaxial (001) tetragonal (T) $PbZr_xTi_{1-x}O_3$ (PZT)/rhombohedral (R) $PbZr_xTi_{1-x}O_3$ (PZT) bilayer heterostructures. The local electric dipole distribution is imaged on an atomic scale for a ferroelastic domain wall that nucleates in the R-layer and cuts through the composition breaking the T/R interface. Our studies reveal a highly complex polarization rotation domain structure that is nearly on the knife-edge at the vicinity of this wall. Induced phases, namely tetragonal-like and rhombohedral-like monoclinic were observed close to the interface, and exotic domain arrangements, such as a half-four-fold closure structure, are observed. Phase field simulations show this is due to the minimization of the excessive elastic and electrostatic energies driven by the enormous strain gradient present at the location of the ferroelastic domain walls. Thus, in response to an applied stimulus, such as an electric field, any polarization reorientation must minimize the elastic and electrostatic discontinuities due to this strain gradient, which would induce a dramatic rearrangement of the domain structure. This insight into the origins of ferroelastic domain wall motion will allow researchers to better "craft" such multilayered ferroelectric systems with precisely tailored domain wall functionality and enhanced sensitivity, which can be exploited for the next generation of integrated piezoelectric technologies. This work appears in
ACS Nano, 2016, **10** (11), pp 10126–10134.