Nanoscale Bubble Domains in Ultrathin Ferroelectric Films

Nagarajan Valanoor[1,*], Qi Zhang[1], Lin Xie[2,3], Guangqing Liu[1], Sergei Prokhorenko[4], Yousra Nahas[4], Xiaoqing Pan[5], Laurent Bellaiche[4], and Alexei Gruverman[6]

[1] School of Materials Science and Engineering, The University of New South Wales, Sydney, New South Wales 2052, Australia

[2] National Laboratory of Solid State Microstructures and Department of Materials Science and Engineering, Nanjing University, Nanjing, 210093, P. R. China

[3] Department of Chemical Engineering and Materials Science, University of California, Irvine, CA 92697, USA

[4] Physics Department and Institute for Nanoscience and Engineering, University of Arkansas, Fayetteville, AR 72701, USA

[5] Department of Physics and Astronomy, University of California, Irvine, CA 92697, USA

[6] Department of Physics and Astronomy, University of Nebraska, Lincoln, NE 68588, USA

*Nagarajan Valanoor: nagarajan@unsw.edu.au

We report nanoscale "bubble domains" in ultrathin epitaxial $PbZr_{0.2}Ti_{0.8}O_3/SrTiO_3/PbZr_{0.2}Ti_{0.8}O_3$ ferroelectric sandwich structures. The formation of bubble domains is attributed to this particular sandwich heterostructure which drives a competition between incomplete polarization screening (electrostatic) and compressive mechanical strain (elastic) parameters. These nano-domains are revealed by high-resolution piezoresponse force microscopy (PFM) and are further corroborated by aberration-corrected atomic-resolution scanning transmission electron microscopy mapping of the polarization displacements. Using above techniques, it is confirmed that the bubble domain are laterally confined spheroids of sub-10 nm size with local dipoles opposite to the macroscopic polarization of their surrounding ferroelectric matrix. An incommensurate phase and symmetry breaking is found within these domains, resulting in local polarization rotation thus imparting a mixed Néel-Bloch-like character to the bubble domain walls. Time-dependent piezoresponse force microscopy studies show that the bubble domains transform to a labyrinthine state under the repeated scanning with AC bias. This is in agreement with first principles-based effective Hamiltonian simulations which reveal that bubble domains represent a transitional topological state at the interface between the two stable topological phases. Moreover, local PFM spectroscopic testing of the bubble domains reveals an electromechanical response twice as large as that of a reference PZT film without bubble domains. Our observations are in agreement with ab-initio-based calculations, which reveal that the bubble domains can be stabilized only within a very narrow range of the electric and elastic boundary conditions. The findings highlight the richness of polar topologies that may develop in ultrathin ferroelectric structures and bring forward the prospect of emergent electronic functionalities due to topological transitions.