

Real-time x-ray interference studies of polarization reversal in ferroelectric thin films

C. Thompson^{a,b}, S.K. Strieffer^b, G.B. Stephenson^b, A. Munkholm^c, J.A. Eastman^b, O. Auciello^b, K. Ghosh^b, G.R. Bai^b, R. Rao^d, and C.B. Eom^d

^aDepartment of Physics, Northern Illinois University, DeKalb, IL 60115 USA

^bMaterials Science Division, Argonne National Laboratory, Argonne, IL 60439 USA

^cChemistry Division, Argonne National Laboratory, Argonne, IL 60439 USA

^dDepartment of Mechanical Engineering and Materials Science, Duke University, Durham, NC 27708 USA

Introduction

Microscopic domain dynamics control the macroscopic physical properties of ferroelectric systems. Therefore, we expect that nanoscale and microscopic structural information will have a significant impact on understanding fundamental mechanisms of ferroelectric response.

We studied the statics and dynamics of 180° domain response during switching in epitaxial films of perovskite-based oxide ferroelectrics by using surface x-ray scattering techniques. X-ray diffraction measures a variety of crystallographic parameters relevant to the structural properties of films. In addition, high-resolution x-ray scattering profiles are particularly sensitive to the ensemble of domain and polarization states in the ferroelectric film, including the distribution of parallel and antiparallel domains (180° domains). This is equivalent to distinguishing between unipolar and mixed domain structures formed of (001) and (00-1) orientations of noncentrosymmetric cells. The primary contrast mechanism in the crystal truncation rod profiles leading to this sensitivity is based on interference between the substrate and film rods [1]. This report outlines the first experiments using x-ray interference techniques to probe domain dynamics in real time (0.01 to 0.2 kHz).

Methods and Materials

Typical samples included 40 nm-thick $\text{Pb}(\text{Zr}_x\text{Ti}_{1-x})\text{O}_3$ (PZT) epitaxial films in the tetragonal phase deposited on SrRuO_3 (SRO) epitaxial electrode layers deposited on single-crystal (001) SrTiO_3 (STO) substrates. In PZT systems, the polarization axis of the film was oriented perpendicular to the surface. In experiments at BESSRC-CAT beamline 12-ID-D, we have developed electronics and data collection capability based on a multichannel scalar sampling technique to measure the evolution of the x-ray scattering profiles of a capacitor simultaneously with its macroscopic polarization response while under external electronic stimulation. The x-ray wavelength used was 14.5 keV, and the incident beam size was defined with slits as 0.030 mm x 0.200 mm. The capacitor was 0.25mm in diameter. We measured along the crystal truncation rods along the (001) Bragg peaks of the film (PZT), electrode layer (SRO), and substrate (STO).

Results and Discussion

X-ray scattering measurements performed synchronously with an ac electrical signal allow time-resolved observation of domain structure dynamics and piezoelectric activity in epitaxial ferroelectric thin films under approximately steady-

state conditions. Figure 1 illustrates the time evolution of the intensity profile across the (001) PZT and SrRuO_3 peaks at selected points in time during simultaneous 0.2 kHz excitation of the ferroelectric heterostructure. The (001) PZT peak position changes with voltage due to its piezoelectric response to the field. Note the shifts and appearances of new interference oscillations between the SRO and PZT Bragg peaks during the switching process. These are caused by the changing ensemble of parallel and antiparallel domains in the system as the ferroelectric film undergoes macroscopic switching.

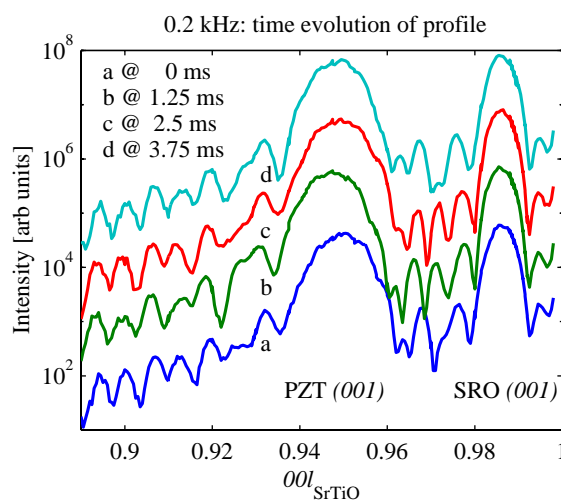


Figure 1: Time evolution of the intensity profile across the (001) PZT and SrRuO_3 peaks at selected points in time during simultaneous 0.2 kHz excitation of the ferroelectric heterostructure.

Acknowledgments

This work is supported through the State of Illinois under HECA, and the U.S. Department of Energy Basic Energy Sciences, Office of Science, under Contract No. W-31-109-Eng-38.

Reference

[1] C. Thompson, C.M. Foster, J.A. Eastman, and G.B. Stephenson, *Appl. Phys. Lett.* **71**, 3516 (1997).