Resonant X-ray Microdiffraction Imaging of Polarization Switching in Ferroelectric $SrBi_2Ta_2O_9$

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Introduction

Much is known about the structure of polarization domains and domain walls in ferroelectric materials at scales as small the dimensions of a chemical unit cell. However, in many cases, the evolution of domains in operating devices at length scales that are orders of magnitude larger is unexplored. In addition to the fact that there is a fundamental interest in exploring the physics of domain nucleation and growth, there is a pressing need to do so because domains in ferroelectric materials have a critical role in devices for nonlinear optics, acoustics, and electronics.[1]. Operating thin film devices that use these materials have typically been studied by using electrical or electromechanical effects. However, spatially resolved measurements can be complicated by the metal film typically used to form a top electrode. Because the spontaneous polarization in ferroelectric materials is, in itself, a consequence of the material's unit cell symmetry, another approach is to pursue these problems with a direct structural tool. Here we present an x-ray resonant microdiffraction study of polarization reversal in a $SrBi_2Ta_2O_9$ (SBT) thin film in which the polarization switching is imaged with a scanned 0.7-μm-diameter x-ray beam.

Methods and Materials

An epitaxially grown, 250-nm-thick SBT film was produced by pulsed laser deposition onto a conducting Nb-doped $SrTiO_3$ substrate. In order to produce planar devices with a nonzero switchable ferroelectric polarization, a (011) substrate orientation was chosen so that the $SrBi_2Ta_2O_9$ film would grow along the [116] direction [2]. Because the SBT unit cell lacks a center of inversion symmetry, the intensities of x-ray reflections from regions of the film with different orientations of the ferroelectric **a** axis are not equal; Friedel's law does not apply. At x-ray energies of atomic resonances of the film's component atoms, this effect is especially large. In order to estimate the difference in intensity, we calculated the structure function of SBT near the Ta L_3 absorption edge as a function of the incident photon energy and the ferroelectric polarization of the film [Fig. 1(a)]. It is useful to express this difference as a contrast analogous to the "flipping ratio" used in neutron scattering [Fig. 1(b)]. At this resonance, there is a sharp maximum in the contrast between domains with different orientations of the SBT **a** axis relative to the sample surface normal. The structure factor plotted in Fig. 1(b) is calculated from tabulated atomic scattering factors for Sr, Bi, and O and from near-resonance optical constants for Ta obtained from absorption measurements on a Ta metal foil. Atomic positions within the SBT unit cell were taken from powder diffraction measurements by Shimakawa et al. [3].

X-ray microdiffraction was used to spatially resolve the switching of these SBT devices. The radiation from an undulator insertion device at the Michigan-Howard-Lucent Technologies-Bell Labs Collaborative Access Team (MHATT-CAT) sector was focused by using a Fresnel zone plate with a 10-cm focal length. Radiation focused into orders other than the first was rejected by using a 20-μm aperture in a Pt-Ir disk. The focal spot size was limited by ambient vibrations to a diameter of 0.7 μm full width at half maximum (FWHM). The negligible absorption of hard x-rays in thin films is essential for these measurements; there was insignificant (~10%) attenuation of the signal due to absorption in the Pt top electrode.

Results

Figure 2(a) shows images formed by rastering the sample through a 100 × 100-μm region, with the sample

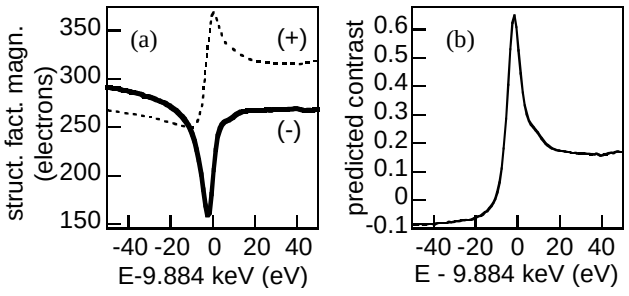

FIG. 1. (a) Computed structure factors for $SrBi_2Ta_2O_9$ unit cell orientations resulting from positive and negative pulses. (b) The normalized contrast [defined as $(I_+ - I_-)/(I_+ + I_-)$] for {2212} reflections computed by using (a).

goniometer set for SBT {2212} type reflections and the incident beam energy set at 9.884 keV. The two images in Fig. 2(a) were acquired after positive and negative voltage pulses to the top electrode. Fig. 2(b) shows the difference between the images of Fig. 2(a) and reveals the regions in which the remnant polarization remained switched. Only a few regions of the sample show a measurable change in polarization during the voltage pulse cycle. It is possible that this inhomogeneity is an artifact of the process of fabricating the electrodes of these devices. Images of the same area (not shown) formed with incident beam energy detuned to 20 eV below the Ta L_3 resonance showed no features related to the domain structure. The contrast predicted by using Fig. 1(b) reaches 60%, while the measurements show a maximum flipping ratio of only 15%. Origins of this discrepancy may include uneven switching within the x-ray spot size and uncertainty associated with the optical constants used to calculate Fig. 1(b).

Discussion

A previous strategy to obtain the response of small areas of ferroelectric devices has been to fabricate devices of small lateral dimensions [4] or to apply voltages with a scanned probe. [5] The microdiffraction technique discussed here is a new approach for studying thin film ferroelectric devices that will complement these techniques by allowing ferroelectric devices to be studied simultaneously by structural and electrical probes at the length scale of important switching phenomena.

Acknowledgments

The authors acknowledge helpful discussions with B. Lai, Z. Cai, and P. Littlewood. Operation of the MHATT-CAT beamlines at the APS was supported by U.S. Department of Energy (DOE) Grant No. DE-FG02-99ER45743. Use of the APS was supported by the DOE Office of Science, Office of Basic Energy Sciences, under Contract No. W-31-109-ENG-38.

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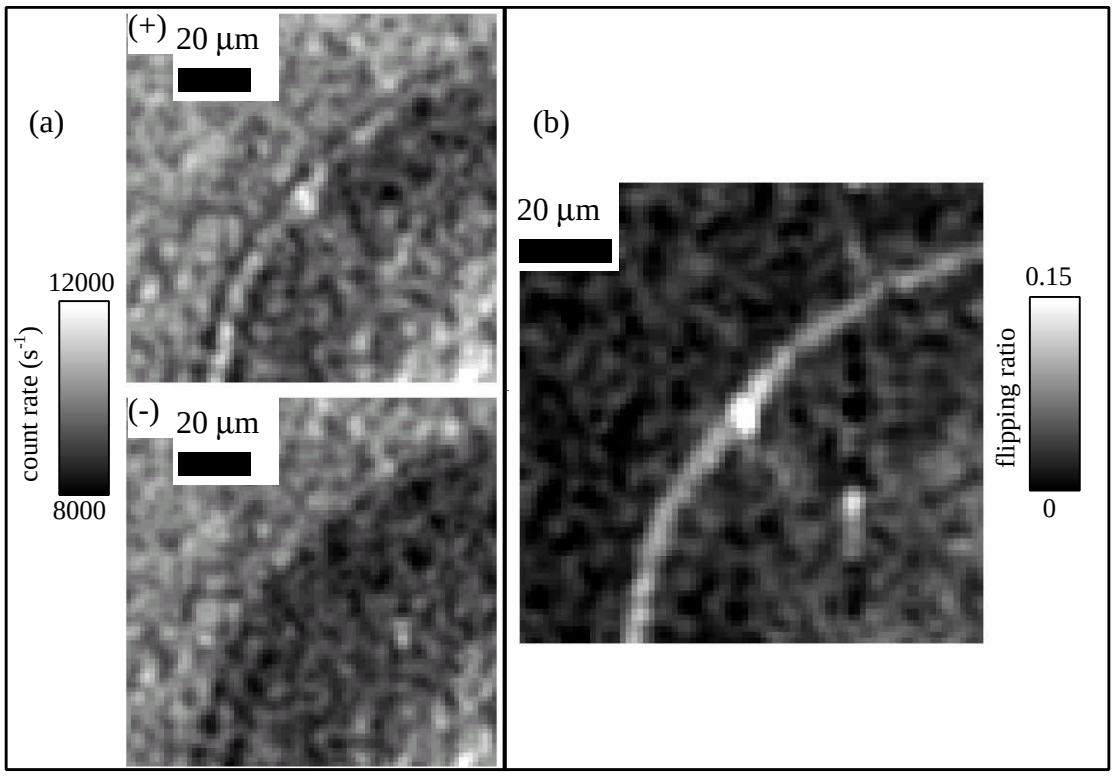

FIG. 2. (a) Images of the intensity of the {2212} reflections of the SBT film in a 100 ´ 100-mm region following positive and negative polarity voltage pulses with peak fields of 320 kV cm^{-1}. The ferroelectric capacitor device is formed by a circular Pt contact covering the portion of the image inside the arc from lower left to upper right. (b) The "flipping ratio" for the two images that appear in (a). The area in which the remnant ferroelectric polarization has switched appears as a bright streak through the image.

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