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X-ray nanodiffraction of tilted domains in a poled epitaxial BiFeO$_3$ thin film

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We present measurements of crystalllographic domain tilts in a (001) BiFeO$_3$ thin film using focused beam x-ray nanodiffraction. Films were ferroelectrically pre-poled with an electric field orthogonal and parallel to as-grown tilt domain stripes. The tilt domains, associated with higher energy (010) vertical twin walls, displayed different nanostructural responses based on the poling orientation. Specifically, an electric field applied perpendicular to the as-grown domain stripe allowed the domain tilts and associated vertical twin walls to persist. The result demonstrates that thin film ferroelectric devices can be designed to maintain unexpected domain morphologies in working poled environments. © 2011 American Institute of Physics. [doi:10.1063/1.3665627]

Bismuth ferrite, BiFeO$_3$, has emerged as a model thin film multiferroic material exhibiting antiferromagnetic spin ordering in conjunction with strong ferroelectric polarization ($\sim$95 $\mu$C/cm$^2$).$^1$ The (pseudo)rhombohedral spontaneous strain of the BiFeO$_3$ unit cell couples to the polarization and is accommodated by structural twin walls that separate crystallographically distinct domains with coherent interfaces.$^2,4$ Of particular interest are domain walls that orient vertically in (001) epitaxial rhombohedral thin films. These vertical domain walls (VDWs) produce ferroelastic domains that are tilted away from the surface normal, requiring disclinations along each of the four cubic directions, two complementary domain variants (e.g., $r_1$ and $r_2$) populated the film.$^4,11$ These two ferroelastic domain variants can, in turn, exhibit four ferroelectric states ($r_{11}^-, r_{11}^+, r_{11}^+, r_{11}^-$). As shown in Figure 1, the as-grown sample exhibited (010) domain twin walls and was comprised of alternating tilted ferroelastic stripe domains. This domain structure allows ferroelectric 109$^0$ boundaries between $r_{11}^+ | r_{11}^-$ or $r_{11}^- | r_{11}^+$ at the (010) twin walls as well as 180$^0$ boundaries between $r_{11}^- | r_{11}^+$ and $r_{11}^+ | r_{11}^-$ within a single ferroelastic domain. These as-grown ferroelastic and ferroelectric states have been previously observed and verified with x-ray diffraction, transmission electron microscopy, and piezoresponse force microscopy in samples fabricated under the same condition.$^9,11$

To investigate the stability of the (010) VDWs in applied electric fields, gold interdigitated electrodes (IDEs) were patterned on the surface of the film in two orientations$^{12}$ and in-plane electric fields were applied perpendicular and parallel to the as-grown stripe direction (see Figure 1). The gap between IDE fingers was 6 $\mu$m, the IDE finger width was 9 $\mu$m, and the deposited gold thickness was $\sim$80 nm. Both electrodes were cycled with 1 ms pulses of $\pm$150 V until the film exhibited full saturation (10$^2$ cycles for $E_\perp$ and 10$^3$ for $E_{||}$). The hysteresis loops after cycling for each IDE are also shown in Figure 1.

The focused x-ray beam at the Hard X-ray Nanoprobe (operated by the Center for Nanoscale Materials at the Advanced Photon Source) was used to map local diffraction from ferroelastic domain distributions in the as-grown film and in the poled regions. Using an x-ray zone plate optic, 10 keV x-rays were focused to a 40 nm diameter spot that was broadened as needed by defocusing. As shown in Figure 1,
The incoming beam direction was parallel with the as-grown stripe direction and incident on the sample at an angle of 18.25° to satisfy the (002) Bragg peak condition. The ferroelectric hysteresis loops from cycling the two IDEs with 1 ms pulses are also shown. Focused beam specular nanodiffraction (shown schematically) was used to probe as-grown and poled regions of the film.

Schematic cross sections are shown in Figure 2 of left tilted (LT), right tilted (RT), and untilted domains (UT) illuminated by a nanofocused beam along with corresponding characteristic specular diffraction patterns. Domains with (001) lattice planes tilted away from the surface normal (LT) and (010) reciprocal space vector, whereas domains with untilted (001) lattice planes (UT) have no reciprocal space displacement. The film diffraction in this work could be quantified as some combination of these cases. Other domain displacement. The film diffraction in this work could be characterized by in-plane fields perpendicular and parallel to the as-grown VDWs. The ferroelectric hysteresis loops from cycling the two IDEs with 1 ms ± 150 V pulses are also shown. The nanoscale tilt domains in these regions were mapped in detail using diffraction from a focused 40 nm diameter beam (Figure 4). At this scale, the characteristic morphology of the as-grown tilt domains is unchanged after poling with a perpendicular applied field (Figures 4(a) and 4(c)), suggesting that ferroelectric alignment can occur in this geometry without removing the (010) twin walls (TWs). These nanodiffraction maps also confirm that the BiFeO3 film is predominantly untitled in regions poled parallel to the as-grown VDWs, consistent with slanted (110)-type twin platelets.
walls (dashed lines in Figure 4(b)) as have been seen experimentally in other studies.9,10

Domain walls in ferroelectric thin films interact with electric fields, strain, polarization, and charged defects.14,15 We found that maintaining a film nanostructure with out-of-plane domain tilts and vertical domain walls required a specific poling direction, a factor which is also correlated to the work needed to pole the film. Poling in the direction normal to the as-grown stripes (maintaining a tilted domain structure) required fewer cycles than were necessary to pole along the stripe direction (crystallographically reorienting the film). This asymmetry ($10^5$ cycles of $E_\parallel$ vs. $10^6$ of $E_\perp$) indicates a difference in the energy barriers associated with ferroelectric dipole alignment in the two directions and reflects changes in the ferroelastic domain structure and the associated coherency defects in the BiFeO$_3$ film. Additional contributions to the asymmetric poling behavior may be differences in the mobility of non-ferroelastic (180°) and ferroelastic (109° and 71°) walls in ferroelectric thin films16,17 as well as preferred polarization rotations of 71° in BiFeO$_3$.12,15 Future work is needed to isolate these processes and determine their relation to the observation of (010) TWs in poled BiFeO$_3$ thin films.

In summary, focused beam nanodiffraction was used to non-destructively compare domain characteristics in ferroelectrically poled epitaxial BiFeO$_3$ multiferroic films. It was determined that (010)-type twin walls present in the as-grown structure were maintained using a perpendicular electric field orientation. This result demonstrates that applied field orientation contributes to the stabilization of unexpected twin wall formations, having potential impacts on utilization of these nanostructures in functional devices.

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