

Understanding Domain Structures in BiFeO₃ Thin Films by Combining Phase-Field Simulations, TEM, and PFM

G. Sheng, * J. X. Zhang, * B. Winchester, * P. P. Wu, * Y. L. Li, * C. T. Nelson, ** H. W. Jang, *** C. M. Folkman, *** R. J. Zeches, **** M. P. Chu, **** S. V. Kalinin, ***** D. G. Schlom, ***** R. Ramesh, **** C. B. Eom, *** X. Q. Pan, ** L. Q. Chen*

*Department of Materials Science & Engineering, Penn State University, University Park, PA 16802

** Department of Materials Science & Engineering, University of Michigan, Ann Arbor, MI 48109

*** Department of Materials Science & Engineering, University of Wisconsin–Madison, Madison, WI 53706

**** Department of Materials Science & Engineering, University of California, Berkeley, CA 94720

***** Center for Nanophase Materials Sciences, and Materials Sciences and Technology Division, Oak Ridge National Laboratory, Oak Ridge, TN 37922

***** Department of Materials Science & Engineering, Cornell University, Ithaca, NY 14853

BiFeO₃ is one of the most promising single-phase multiferroic systems for potential applications in multifunctional devices [2]. Its ferroelectric phase is a rhombohedral perovskite with spontaneous polarization along the pseudocubic <111> directions, and thus it possesses four possible ferroelastic variants (labeled as r_1 , r_2 , r_3 , r_4) and the eight polar domains (labeled as r_1^+ , r_2^+ , r_3^+ , r_4^+ , r_1^- , r_2^- , r_3^- , r_4^-). There are three possible types of domain walls across which polarization direction changes by 71°, 109° and 180°, respectively. A fundamental understanding of domain structures and their evolution under an electric field is critical for controlling their properties and thus for device applications of BiFeO₃.

We investigated the domain structures in epitaxial BiFeO₃ films using a combination of phase-field simulations [3], transmission electron microscope (TEM), and Piezoresponse Force Microscope (PFM). In particular, we studied the effect of epitaxial strain, substrate orientation, electric boundary conditions, and film thickness on the phase stability, domain wall orientations, and switching behavior. To graphically present the strain effect on domain structures, we constructed the temperature-strain phase diagram for BiFeO₃ thin films using phase-field simulations, which displays the stability regions for different polarization states as a function of temperature and biaxial strain. It is shown that if the strain can be made sufficiently large, it may lead to not only changes in domain structures but also strain-induced phase transitions [4].

It is found that one of the most efficiently ways to control domain structures is to employ substrate miscut. As an example, the domain structures of a (001) BiFeO₃ film on a (001) SrTiO₃ substrate and a (001) BiFeO₃ film on a (001) SrTiO₃ with a 4° miscut towards (100) with both top and bottom SrRuO₃ electrodes are shown in Fig 1. The (001) BiFeO₃ grown with bottom SrRuO₃ electrode on an exact (001) SrTiO₃ substrate display domain structures containing all four ferroelastic variants r_1 - r_4 with a mixture of 71° and 109° ferroelastic domain walls as shown in Fig. 1a, a cross-sectional dark-field TEM micrograph, along with a colorized PFM domain map (Fig. 1b). In contrast, the BiFeO₃ films grown with a bottom SrRuO₃ electrode on (001) SrTiO₃ with a 4° miscut along the [100] direction exhibit only two variants (Fig. 1c and d) and form a one-dimensional array of ferroelastic 71° degree domain walls perpendicular to the miscut direction [1]. As one can see in Fig. 2, the domain structures predicted by phase-field simulations are in remarkably good agreement

with experimental observations. In addition to substrate miscut, we also found that the relative fractions of 71° and 109° walls in (001) BiFeO₃ films can be controlled by changing the film thickness and electric boundary conditions.

The stability of a switched domain in BFO films was studied using a combination of PFM and phase-field simulations [5]. It was discovered that two parallel domain walls with the same orientation may have drastically different thermodynamic stability caused by the film-substrate misfit strain. It was also demonstrated that extended structural defects such as twin boundaries act as preferential nucleation sites during switching [6]. Therefore, the types of ferroelastic walls that exist in a domain structure are expected to influence the switching properties such as coercive field.

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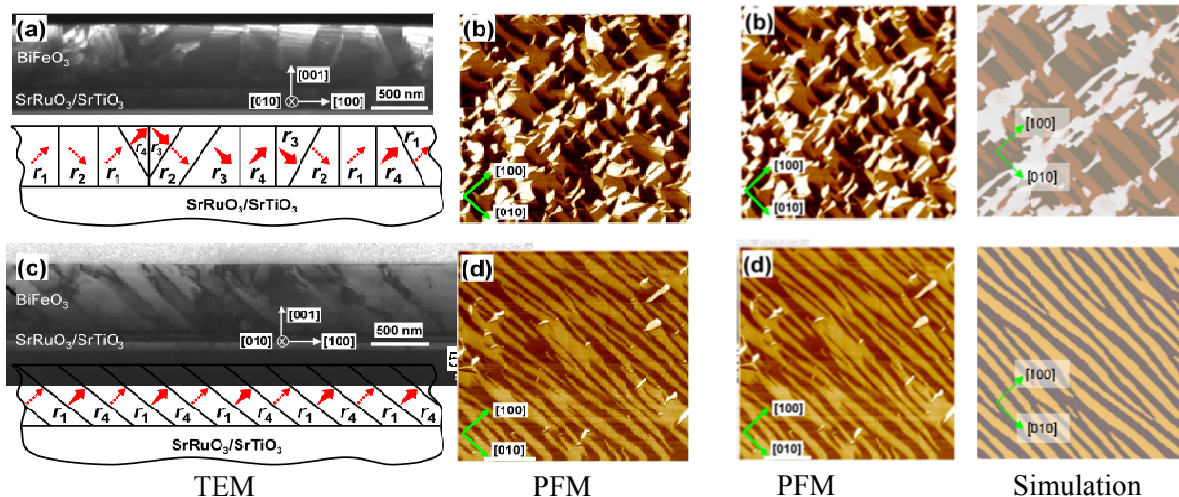


Fig. 1 Cross-sectional TEM images of BFO films (600nm) on (a) exact and (c) miscut STO. In-plane PFM images ($4\mu\text{m} \times 4\mu\text{m}$) of BFO films on (b) exact and (d) miscut STO[1].

Fig. 2. Comparison PFM images of 4- and 2-variant domain structures (left) [1] and those obtained from phase-field simulations (right, unpublished).