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Polarization switching in quasiplanar BiFeO₃ capacitors

Lu You,¹ Elvin Liang,¹ Rui Guo,¹ Di Wu,² Kui Yao,³ Lang Chen,¹ and Junling Wang^{1,a)}

¹*School of Materials Science and Engineering, Nanyang Technological University, Singapore 639798*

²*Department of Materials Science and Engineering and National Laboratory of Solid State Microstructures, Nanjing University, Nanjing, Jiangsu 210093, People's Republic of China*

³*Institute of Materials Research and Engineering, Agency for Science, Technology, and Research (A*STAR), 3 Research Link, Singapore 117602*

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Polarization switching in multiferroic BiFeO₃ is studied using a quasiplanar capacitor geometry. Macroscopic quantitative hysteresis measurements using single-pair electrodes yield results that agree well with the theoretical predictions. Nanoscale ferroelectric domain analyses reveal that highly aligned 71° stripe domains are created upon electrical switching. Careful reconstruction of the polarization configuration demonstrates that in-plane polarization reversal is achieved by a coherent 71° switching mechanism, consistent with the macroscopic measurement results. Such control of polarization switching in quasiplanar BiFeO₃ capacitors is crucial for the electrical control of the multifunctionality of BiFeO₃. © 2010 American Institute of Physics. [doi:10.1063/1.3479911]

Multiferroic BiFeO₃ (BFO) has attracted much research work in recent years,¹ not only because of the coupling between ferroelectric and antiferromagnetic order parameters for magnetoelectric applications² but also due to the discovery of unique properties related to the ferroelectric domain walls.³ Because of the structural discontinuity, domain wall may exhibit dramatically distinct properties from those of the bulk material. Through the spin-charge-lattice coupling, unusual phenomena including electrical conduction,⁴ photovoltaic effect,⁵ and magnetic coupling⁶ have been observed at the ferroelectric domain walls of BFO thin films. To study domain wall functionalities in BFO requires the capability to control ferroelectric switching and observe the process *in situ*, which can not be done in conventional vertically sandwiched capacitors. To solve these issues, planar-electrode structure has been developed.⁷ Ferroelectric domains can be studied *in situ* by piezoelectric force microscopy (PFM) under this horizontal geometry. The ideal layout of a planar-electrode structure should be two electrodes embedded in the BFO film, as demonstrated in Ref. 9. But electrodes patterned directly on top of the BFO film have also been used, and ferroelectric switching can still be realized under this quasiplanar electrode geometry.⁵ However, quantitative study of the polarization switching and corresponding domain evolution remain unclear.

To shed more light on the in-plane (IP) polarization switching in multiferroic BFO, quasiplanar capacitors of Pt/BFO/Pt with different channel widths have been fabricated and characterized. 60 nm BFO film was epitaxially grown on SrTiO₃ (STO) (001) single crystal substrate by pulsed laser deposition. The growth temperature and oxygen partial pressure were 700 °C and 100 mTorr, respectively. Following the BFO deposition, 40 nm Pt planar electrodes were patterned on top via a standard lift-off process. The edge of the electrode is parallel to the [010] direction of the STO substrate. The device structure is schematically illustrated in Fig. 1(a). The spatial distribution of electric field was mapped out

using a commercial finite element simulation software (COMSOL INC.). Figure 1(b) depicts only the cross-sectional IP electric field distribution. Despite the highly nonuniform field close to the electrode edges, majority of the BFO film within the gap between the two electrodes experiences almost constant IP electric field. Ferroelectric properties were measured by Precision LC ferroelectric tester (Radiant Technologies). However, due to the greatly reduced area of quasiplanar capacitor, capacitances which result from both the connection wires and stray field in air and STO substrate [Fig. 1(b)] become dominating compared to the ferroelectric

FIG. 1. (Color online) (a) Schematic drawing of device structure and experiment setup for quasiplanar-electrode BFO capacitor. (b) Corresponding IP electric field distribution on the cross section of a 1.4 μm capacitor, simulated by finite element modeling. The relative dielectric constants used in the simulation for BFO and STO are 80 and 300, respectively.

^{a)} Author to whom correspondence should be addressed. Electronic mail: jlwang@ntu.edu.sg.

FIG. 2. (Color online) (a) Diagram of the voltage profiles used for reconstruction of remanent hysteresis. (b) Example of the deduction in remanent hysteresis for 5.3 micron quasiplanar capacitor. (c) Voltage dependence of remanent hysteresis for 5.3 micron quasiplanar capacitor measured with a frequency of 1 kHz. (d) PUND measurement for capacitors with different channel widths using 0.1 ms voltage pulse.

polarization, as shown in Fig. 2(b) (black curve). Fortunately, these linear parasitic elements are nonremanent in nature. As a result, by applying a second voltage cycle, the nonremanent polarization loop can be obtained, which can be subsequently subtracted from the initial result containing both remanent and nonremanent components [Figs. 2(a) and 2(b)]. Using this method, highly square hysteresis loops were obtained, indicating narrow coercive field (E_c) distribution and coherent switching of polarization vectors [Fig. 2(c)]. Here the nominal electric field is calculated using voltage divided by the channel width without considering the nonuniformity of the field. Positive-up-negative-down (PUND) pulse measurements were carried out with 0.1 ms pulse width to provide accurate remanent polarization (P_r) values for different channel widths [Fig. 2(d)].

The channel width here is equivalent to the film thickness in vertically sandwiched structures. As expected, capacitors with different channel widths show almost constant P_r values, which should be the projection of the IP polarization vector along [100] direction under our electrode configuration. This polarization component (P_1) can be calculated through the following equation:⁸

$$P_1 = \sqrt{P_s^2 - P_3^2} / \sqrt{2},$$

where P_s is the intrinsic spontaneous polarization in BFO, P_3 is the out-of-plane (OP) polarization component. Given that $P_s = 102 \mu\text{C}/\text{cm}^2$ and $P_3 = 75 \mu\text{C}/\text{cm}^2$ in fully strained

BFO on STO substrate,⁸ we can estimate the P_1 to be $\sim 48.9 \mu\text{C}/\text{cm}^2$, in perfect agreement with the experiment result of $48 \pm 3 \mu\text{C}/\text{cm}^2$. From here, it can be inferred that the measured polarization originates from only IP component. The nominal coercive field (E_{cn}) decreases with increasing channel width as expected. Ideally, the thickness dependence of E_c in ferroelectric films should follow the semiempirical scaling law, $E_c \propto d^{-2/3}$.⁹ However, in such a quasiplanar structure, electric field is highly nonuniform. Thus, it would be more reasonable to make some corrections to the E_{cn} . Accordingly, average electric field in the BFO film between two electrodes was calculated as the effective coercive field (E_{ce}) based on the finite element modeling results. By fitting the E_{ce} obtained in this study and data from the literature,^{8,10-14} we can get a slope $k = -0.62 \pm 0.03$ [Fig. 3(b)], in good agreement with the scaling factor of 2/3.

An important advantage of the planar-electrode geometry as compared to the vertical one is the ability of *in situ* ferroelectric domain imaging. Well-established PFM techniques allow us to fully reconstruct the polarization configuration in three dimensions by combining IP and OP PFM images.¹⁵ Figure 4(a) shows the IP PFM image of the as-grown BFO film with three different colors while no contrast can be observed in the OP PFM image, indicating that the OP polarization component is pointing out of the paper [Fig. 4(a), upper right inset]. Based on the IP and OP images, it can be concluded that three out of eight possible ferroelectric variants coexist in the as-grown BFO film, creating two stripe domains (purple and yellow) interlacing with each other. The configurations of IP polarization vectors are schematically depicted in the inset of Fig. 4(a), featured by “head-to-tail” zigzag patterns. Upon application of electric field (30 V and 1 ms voltage pulse), the original two sets of stripe domains change to highly ordered single stripe domain (purple) with only two variants [Fig. 4(c)]. The average domain size increases from ~ 100 to 170 nm. If electric field with opposite polarity (-30 V and 1 ms voltage pulse) is applied, the stripe domain pattern maintains, however, the contrast changes (yellow) due to the rotation of the IP polarization vectors [Fig. 4(d)]. It should be noted that there is no change in the OP PFM image during the electrical switching [Figs. 4(c) and 4(d), bottom right insets], suggesting that only IP polarization rotations contribute to the polarization switching. Interestingly, the domain switching is accomplished by merely 90° rotation of the IP polarization vector in each stripe domain without creating extra domain walls. The marking in the red circles [Figs. 4(c) and 4(d)] serves as an aid for the readers. It can be seen that purple domain in Fig. 4(c) changes to brown in Fig. 4(d) while the brown one in Fig. 4(c) is replaced by yellow domain in Fig. 4(d), which

FIG. 3. (Color online) (a) Channel width dependence of the remanent polarization P_r and nominal coercive field E_{cn} . (b) Corrected effective coercive field E_{ce} according to finite element simulation vs. capacitor thickness (channel width).

FIG. 4. (Color online) IP PFM images for the quasiplanar BFO capacitor (a) in the as-grown state ($5\ \mu\text{m} \times 5\ \mu\text{m}$), [(c) and (d)] after electrical switching with opposite polarities ($8\ \mu\text{m} \times 8\ \mu\text{m}$), and (b) in the intermediate stage during the switching process ($8\ \mu\text{m} \times 8\ \mu\text{m}$). The inset shows the corresponding domain configurations schematically. The OP PFM images are also shown in the inset of (a), (c), and (d). In all images, the cantilever is scanning along $[110]$ direction.

corresponds to a 71° switching, resulting in a net polarization along the electric field direction. Domain merging also takes places, as denoted in the blue circle of Figs. 4(c) and 4(d). Such a coherent rotation behavior of the stripe domains is consistent with the macroscopic hysteresis measurement results. Moreover, in the intermediate stage of polarization switching (28 V and 1 ms voltage pulse), coexistence of two sets of stripe domains with opposite net polarization direction occurs, separated by a boundary perpendicular to the electrode edge [Fig. 4(b)]. As illustrated in the inset of Fig. 4(b), it is found that the domain boundary is comprised of small 71° domain walls, connecting each stripe domain. The preference of 71° polarization switching rather than 109° one can be explained by the different activation energies of different switching routes.¹⁶ Under the IP electric field, 71° polarization rotation should have less energy barrier than 109° one due to the shorter switching path and less strain accompanied.

In conclusion, ferroelectric polarization switching in quasiplanar BFO capacitors is studied both macroscopically and microscopically. Well-defined hysteresis loops are obtained, with the P_r and E_c values matching the theoretical calculation. The domain switching process can be described by a coherent 71° rotation of polarization vectors. Through intimate spin-charge-lattice coupling, the order parameters in multiferroic BFO can be controlled using electric field, which makes an important step for further applications.

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