Ferroelastic domain wall dynamics in ferroelectric bilayers


Published in:
Acta Materialia

Document Version:
Early version, also known as pre-print

Queen's University Belfast - Research Portal:
Link to publication record in Queen's University Belfast Research Portal

General rights
Copyright for the publications made accessible via the Queen's University Belfast Research Portal is retained by the author(s) and / or other copyright owners and it is a condition of accessing these publications that users recognise and abide by the legal requirements associated with these rights.

Take down policy
The Research Portal is Queen's institutional repository that provides access to Queen's research output. Every effort has been made to ensure that content in the Research Portal does not infringe any person's rights, or applicable UK laws. If you discover content in the Research Portal that you believe breaches copyright or violates any law, please contact openaccess@qub.ac.uk.
Ferroelastic domain wall dynamics in ferroelectric bilayers

V. Anbusathaiah a, S. Jesse b, M.A. Arredondo a, F.C. Kartawidjaja c, O.S. Ovchinnikov d, J. Wang c, S.V. Kalinin b, V. Nagarajan a,∗

a School of Materials Science & Engineering, University of New South Wales, Sydney, NSW 2052, Australia
b The Centre for Nanophase Materials Sciences, Oak Ridge National Laboratory, Oak Ridge, TN 37922, USA
c Department of Materials Science and Engineering, National University of Singapore, Singapore 117576, Singapore
d Department of Physics and Astronomy, University of Tennessee, Knoxville, TN 37996, USA

Received 9 February 2010; received in revised form 16 May 2010; accepted 1 June 2010

Abstract

High-performance piezoelectric devices based on ferroelectric materials rely heavily on ferroelastic domain wall switching. Here we present visual evidence for the local mechanisms that underpin domain wall dynamics in ferroelastic nanodomains. State-of-the-art band excitation switching spectroscopy piezoforce microscopy (PFM) reveals distinct origins for the reversible and irreversible components of ferroelastic domain motion. Extrapolating the PFM images to case for uniform fields, we posit that, while reversible switching is essentially a linear motion of the ferroelastic domains, irreversible switching takes place via domain wall twists. Critically, real-time images of in situ domain dynamics under an external bias reveal that the reversible component leads to reduced coercive voltages. Finally, we show that junctions representing three-domain architecture represent facile interfaces for ferroelastic domain switching, and are likely responsible for irreversible processes in the uniform fields. The results presented here thus provide (hitherto missing) fundamental insight into the correlations between the physical mechanisms that govern ferroelastic domain behavior and the observed functional response in domain-engineered thin film ferroelectric devices.

© 2010 Acta Materialia Inc. Published by Elsevier Ltd. All rights reserved.

Keywords: Ferroelectrics; Piezoelectrics; Switching spectroscopy PFM; Band excitation piezoforce spectroscopy; Ferroelastic domains

1. Introduction

The coupling between strain and electric field is a fundamental physical phenomenon that underpins the electromechanical response in a broad range of devices, such as sensors, actuators and energy harvesting systems [1,2]. Since early applications of ferroelectric crystals to SONAR technology, significant attention has been paid to the development of ferroelectric material systems with enhanced electromechanical coefficients. The electromechanical response essentially comprises the intrinsic lattice contribution and an extrinsic contribution attributed to the mobility of ferroelastic domain walls.

While the intrinsic electromechanical response is well understood and is even amenable to first-principle calculations [3,4], the nanoscale mechanisms underpinning the extrinsic domain wall contribution remain largely unknown. Understanding the mechanisms that control the extrinsic domain wall motion is vital as it is the extrinsic component (rather than the intrinsic) that give rise to large electromechanical coefficients [5–7]. Thus these answers are crucial for establishing the size limit of ferroelectric microelectromechanical systems and developing the pathways for development of materials with improved properties. Early attempts to account for domain wall contributions were able to calculate the enhanced contribution from the ferroelastic domain walls [8]. The models considered motion of the domain walls along a single direction, where the entire domain moved in a reversible fashion under an applied stress or field. Whilst this mechanism can yield enhanced electro-
mechanical coefficients, it cannot account for the typically observed increased hysteresis. On the other hand, a number of studies [9,10] have addressed these behaviors in the context of Rayleigh [11] and Preisach [12,13] models of hysteretic behavior, allowing phenomenological description of the response mechanism. The corresponding microscopic mechanisms have been analyzed by several groups based on macroscopic measurements, including harmonic components of the response and nonlinear susceptibilities [9,14,15]. The contributions to extrinsic electromechanical response can originate from the motion of the non-180° ferroelastic domain walls, motion of the 180° ferroelectric domain walls in the presence of external fields (or in the presence of internal random fields), polarization enhancements at the ferroelastic domain wall [7,16] or switching of individual regions in the presence of strong disorder. However, these local mechanisms generally do not allow for hysteretic behavior, necessitating the consideration of pinning effects. Furthermore, recent studies of thickness dependence of non-linearities [17] and spatially resolved mapping of Rayleigh response [18,19] illustrate the presence of non-trivial length scales underpinning extrinsic response mechanisms. Overall, the relationship between the elementary mechanisms of domain structure motion and the emergence of complex hysteretic responses remains unresolved.

Here, we present a study that explores the local mechanisms involved in large electromechanical responses in dense (ferroelastic) domain structures. We exploit the interactions between two ferroelectric thin film bilayers that give rise to nanoscale ferroelastic domains that are labile under external bias. Previously we reported in this bilayered system a complex ferroelastic nanodomain microstructure with an extremely large grain size (see Fig. 1) that is present only in the Ti-rich top tetragonal layer [6]. This provides a facile platform for us to explore the local mechanisms in two ways. Firstly, the domains are not constrained to substrate and can therefore move under the application of an external electric field, leading to a giant electromechanical constant ($d_{33}(001)$) of $\sim 220$ pm V$^{-1}$, up to three times larger than what is normally observed in constrained single-layered PZT thin films. We have theoretically shown that the dense ferroelastic domain pattern can be attributed to the electromechanical coupling between the T and the R layers [20]. Secondly, the special arrangement of ferroelastic domains within a very large grain allows us to address the domain wall dynamics on the length scale above the single domain or domain wall size but well below the grain or device level. This means that the bilayered system proves to be an ideal case where we could probe the crucial coupling between single domain wall dynamics and collective and hysteretic behaviors that emerge in multiple domain wall systems, but without the influence of the grain boundary.

Our key finding is the direct observation of so-called domain wall twists as a mechanism for ferroelastic domain wall motion as well the observed hysteresis in the piezoelectric behavior.

2. Experimental procedure

A bilayered ferroelectric thin film heterostructure comprising a tetragonal (T) PbZr$_{0.3}$Ti$_{0.7}$O$_3$ film deposited on a rhombohedral (R) PbZr$_{0.7}$Ti$_{0.3}$O$_3$ film, on electrode-buffered Si substrates, has been chosen as a model system for the investigation presented here [21]. The films were deposited by a multistep sol-gel route assisted by a spin-coating method. X-ray diffraction measurement using a Philips X-pert MRD multi-purpose X-ray diffractometer verified that the structure of the PZT layers was polycrystalline, with a preferential orientation along the $\langle 0 0 1 \rangle / \langle 1 0 0 \rangle$ direction and a small fraction of $\langle 1 1 1 \rangle$ orientation. Cross-sectional electron microscopy and electron probe microanalysis confirmed that there is no intermixing between the functional bilayers. Further processing details and macroscopic ferroelectric properties are given elsewhere [21].

Conventional piezoforce microscopy (PFM) implemented in a Veeco multimode atomic force microscope (AFM; Nanoscope IIIa controller) with a Pt/Ir-coated cantilever (with a typical tip radius of 7 nm, a force constant of 0.2 N m$^{-1}$, and a resonance frequency of 13 kHz) was employed at a scan rate of 0.8 Hz for the visualization of
the domain structure. An AC signal $V_{ac} = V_o \sin(\omega t)$ with an amplitude of 1.5 V and a frequency of 7 kHz was applied between the AFM tip (movable top electrode) and the bottom electrode of the sample to acquire the piezoresponse images of the out-of-plane and in-plane components with the aid of two lock-in amplifiers. The as-grown (virgin) domain structure was visualized by monitoring the product of first harmonic amplitude and cosine of the phase, i.e. the $(R \cdot \cos \theta)$ component, which is the real part of the complex piezoresponse image. Then, through the same AFM tip, DC bias was applied locally to a grain of interest to perform the domain-switching experiment. Cross-sectional transmission electron microscopy (TEM) analysis was carried out using a JEOL 3000F transmission electron microscope operated at an accelerating voltage of 300 kV.

Band excitation piezoforce switching spectroscopy (BEPS) was implemented on a Veeco Dimension AFM equipped with a Nanoman V controller. The data acquisition system was developed in a MATLAB/LABVIEW environment. The measurements were performed in the point-by-point mode using Cr–Au-coated tips (Micromasch CSC-37 C, resonance frequency $\sim$28 kHz, spring constant $k \sim 0.35$ N m$^{-1}$). The complete waveform 2–4 million points, 1–2 s, was generated using a National Instruments’ PXI-6115 Multifunction DAQ system. Finally, the simple harmonic oscillator (SHO) fitted using the MatLab GUI program produced four-dimensional (4-D) maps of the amplitude $A(\omega)$, phase $\theta(\omega)$ and frequency responses spatially as a function of the DC bias $V_{dc}$.

3. Results and discussion

Although we found that the ferroelastic domains are never eliminated, under electrical bias these domains reconstruct into various arrangements different from the virgin state. This gives rise to several scientific questions, such as the role of the local coercivity field, the influence of the electrostatic boundary conditions and the local crystalline properties, which are best answered by switching spectroscopy PFM (SSPFM) and vector PFM because they give both high spatial and time resolution. We discuss below results obtained from each of the PFM techniques mentioned above.

3.1. Dense domain switching and wall twisting

The surface morphology and the dense domain structure of this bilayered ferroelectric thin film are presented in Fig. 1. The $3 \times 3$ $\mu$m$^2$ topography scan (Fig. 1a) shows that the grain size of the film [6,21] is very large, of the order of a few microns. The corresponding vertical piezoforce microscopy (VPFM) (Fig. 1b) and lateral piezoforce microscopy (LPFM) (Fig. 1c) images captured in its virgin state reveal the dense domain structure of the film, notably with varying domain periodicity between the grains.

Qualitative understanding of the role of applied DC bias is presented in Figs. 2 and 3. The series of vertical and lateral PFM images in Fig. 2a–j, which were subjected to a DC bias experiment as a function of the applied electric field ($-1$ to $-4$ V), show that the dense domains move in a fashion such that they lack any irreversible domain structure up to a critical bias; in other words, they relax swiftly after removal of the DC switching bias in this conventional PFM experiment (note that the reversible switching is verified by the abnormally large electromechanical responses in the proximity of the coercive bias, as shown later in the hysteresis experiments). Above a critical bias the domain structure undergoes dramatic in-plane reorientation (Fig. 3). Fig. 3 is a panel of vertical and lateral PFM images that was subjected to $-5$ V DC switching experiments. Fig. 3a and b are the vertical PFM images captured at the virgin and after applying $-5$ V DC bias at the centre region respectively. Fig. 3c is

![Fig. 2. Reversible dense domains. (a, c, e, g, i) Vertical PFM images from the virgin state and as a function of DC switching biases ($-1$, $-2$, $-3$ and $-4$ V), respectively. (b, d, f, h, j) Simultaneously captured lateral PFM images.](image-url)
the lateral PFM image captured simultaneously with Fig. 3b after the application of the DC bias. As seen in Fig. 3b and c, the domain structure for the region under bias has undergone significant changes. The simple lamellar order is now transformed and several banded-like domain structures with varying in-plane orientations have been created. We refer to this structure as in-plane twist domains and propose that this twisting process is one of the central mechanisms responsible for the observed hysteresis. If the twist motion is confined to a small area, the process is reversible and can be observed only in hysteresis measurements. If the size of the twisted region exceeds the domain spacing, the twist is irreversible and transformation to a different texture occurs. This process is schematically illustrated in Fig. 3d. Note that these measurements (imaging following bias pulse application) can illustrate only irreversible twists associated with large-scale changes in the domain structure. In the following we argue that small bias pulses may result in smaller reversible twists, as explored below using a novel spectroscopic approach.

3.2. Reversible dense domain behavior

To obtain quantitative information on local domain dynamics, we utilized band excitation switching spectroscopy piezoresponse force microscopy (BE-SSPFM)\cite{22} implemented in the vertical and lateral and on-field and off-field modes. In conventional SSPFM\cite{23,24}, the hysteresis loops are measured over a spatially resolved 2-D grid on the sample surface. The individual loop represents a change in electromechanical response measured with a $V_{ac} = 0.5-3 \ V_{pp}$ signal at a frequency of $\sim 10-100 \ kHz$ to a slowly ($\sim 0.01-1 \ Hz$) changing triangular signal with a

---

Please cite this article in press as: Anbusathaiah V et al. Ferroelastic domain wall dynamics in ferroelectric bilayers. Acta Mater (2010), doi:10.1016/j.actamat.2010.06.004
magnitude of \( \sim 10 \ V_{pp} \). The use of a large \( V_{ac} \), which is necessary to improve the signal/noise ratio, effectively smoothes the hysteresis loop. At the same time, the use of resonance enhancement in PFM (by a factor of \( Q \), where \( Q \) is of the order of \( \sim 100 \) for contact resonances) is limited by the strong position dependence of contact resonance [25,26]. Here, we utilize the recently developed band excitation method [27] as applied to PFM spectroscopy [22]. In BE, the tip is excited by a digitally synthesized signal containing a range of frequencies encompassing the resonance, at each voltage and space-step. The measured 4-D data set of response vs. frequency, voltage and position is analyzed using direct integration or SHO fit to yield the response vs. bias, i.e. the hysteresis loop.

The BE-SSPFM was performed at the centre of the region (inside the white square) described in Fig. 4a (topography), and the subsequent VPFM amplitude and phase images after switching are shown in Fig. 4b and c, respectively. The switching region is clearly identified by contrast variation in the phase image. Fig. 4d-f shows the thus-obtained \( 80 \times 80 \) pixel (1.6 x 1.6 \( \mu \)m) vertical BEPS spatial maps of the ferroelectric parameters, viz. the positive remnant (\( R^+ \)) (Fig. 4d), negative remnant (\( R^- \)) (Fig. 4e) and switchable polarization (\( SP \)) (Fig. 4f) respectively. The ferroelastic dense domain with sub-10 nm width still appears even after the application of a strong band-excitation signal. Comparisons of these spatial maps clearly indicate the variation in the remnant state between the grains. For instance, on a larger macroscopic scale, the grain indicated by “X” possesses a lower \( R^+ \) compared to grain “Y”, which has a relatively large \( R^+ \) value, whereas the \( SP \) is presumably uniform irrespective of the region. In contrast, inside an individual grain, the fine structure of the dense domain architecture shows distinct variation in the

Fig. 4. BEPS maps of the dense domains. (a) Topography. Simultaneously captured VPFM amplitude (b) and phase images (c) after the BE-SSPFM measurement. The white square in the topography image denotes the BEPS run region. (d) Positive remnant (\( R^+ \)), (e) negative remnant (\( R^- \)) and (f) switchable polarization spatial maps of the dense domains from the vertical BEPS response. (g-i) Corresponding BEPS response from the lateral direction.
remnant state as well as in the SP. This is attributed to the difference in $c$ and $a$ domain orientation. The SP is also maximum at the ferroelastic domain wall boundary ($c$–$a$ domain junction). However, the simultaneously acquired lateral BEPS maps (Fig. 4g–i) shows very small spatial variation in the ferroelectric parameters. This could be due to the off-contact resonance of the tip-surface interaction, where the lateral responses were captured at the same bandwidth of frequency as the vertical BEPS.

To gain further insight into these nanodomain behaviors, we performed high-resolution BEPS studies within a single grain (500 $\times$ 500 nm), as shown in Fig. 5. To avoid the cross-talk between the vertical and the lateral BEPS at this resolution, both vertical and lateral BEPS responses were acquired at their respective contact resonance frequencies. The topography of the film surface (1.5 $\mu$m) and the simultaneously captured amplitude and phase images of the vertical PFM where the BEPS is performed can be seen in Fig. 5a–c. The contrast variations in the phase image after switching spectroscopy identify the BEPS region. The 40 $\times$ 40 pixel vertical and lateral BEPS spatial maps for the switchable polarization are shown in Fig. 5d and e. These images clearly demonstrate the variability of switching behavior in ferroelastic domain switching, manifested through varying (bright, dark and gray) contrasts. The bright region in the vertical map should be the $c$ domain as it shows an enhanced switching signature; correspondingly, the dark contrast in the vertical maps must originate from the $a$ domains. The region in-between these $c$ and $a$ domains (grain in contrast) are the ferroelastic domain walls. The same region (ferroelastic domain wall) in the lateral map shows a very bright contrast. Bright contrast regions here indicate high switch-

![Image](image_url)

Fig. 5. High-resolution BEPS maps of ferroelastic nanodomains. (a) Topography. Simultaneously captured VPFM amplitude (b) and phase images (c) after the high-resolution BEPS measurement. (d) Vertical and (e) lateral switchable polarization spatial maps of the dense domains acquired in series at their respective contact resonance frequencies. (f) Average vertical and (g) average lateral local hysteresis loops respectively from the $c$ and $a$ domains and at the domain wall region.

Please cite this article in press as: Anbusathaiah V et al. Ferroelastic domain wall dynamics in ferroelectric bilayers. Acta Mater (2010), doi:10.1016/j.actamat.2010.06.004
able polarization in the plane of the substrate, suggesting that the ferroelastic domain wall boundary could be a source of enhanced nucleation activity.

To quantify the observed effect, the local average hysteresis loops at the c (bright) and a (dark) domains and at the domain wall using the vertical and lateral BEPS maps were measured and are shown in Fig. 5f and g, respectively. The exact regions of the local hysteresis loops analysis are shown by red (c domain), green (a domain) and white (domain wall) small squares in Fig. 5d. Comparison of these local loops reveals that the remnant state of the a domain in the vertical direction is large, as these domains require a large depolarizing field to switch back or relax. On the other hand, the domain wall in the lateral loops shows significantly higher remnant polarization than the bright and dark regions, signaling that the ferroelastic domain wall movement in the lateral direction is predominant. However, we do not see any reorientation of the domain walls before and after the BEPS operation and hence conclude that this is a signature of reversible ferroelastic domain wall motion.

3.3. On-field vs. off-field behavior

All of the above discussions were based on BE-SSPFM measured with off-field bias, which has some time lag between the write bias and the read bias. This time could indeed be enough for an untethered domain wall to relax back to its virgin state and would thus inhibit the understanding of the local switching behavior in real time. To overcome this issue, the switching behaviors of these dense domains were performed under switching bias (on-field) and without switching bias (off-field) in a confined region inside a grain. The pulse profile, which is used to study the on-field vs. off-field behavior, is shown in Fig. 6. In this profile the triangular switching waveform is used for the write operation and the superimposed band-excitation signal (BE waveform) is used for the read operation. It is also noted that the band-excitation signal is present in both the DC-on and -off steps when performing the on-field vs. off-field BEPS measurements. A vertical PFM image of the dense domain structure studied and a corresponding BEPS spatial map (30 × 30 pixel) of the positive remnant state (R+) used for the local hysteresis measurement are shown in Fig. 7a and b, respectively. The local average amplitude and phase of the cantilever responses in the switching region for the on-field and off-field BE signals as a function of the triangular switching waveform are shown in Fig. 7c-f. The switching events can be clearly seen as straight vertical lines in these images where the amplitude of the response goes effectively to the noise value while the phase changes by π. Further, the contact resonance frequency lies nearly at the same value between the on-field and off-field BE signals. However, the switching sequence varies markedly between the two measurements. The quantitative amplitude and phase responses obtained from this local region both on-field and off-field are plotted together in Fig. 7g and h. From these plots, it is noted that the on-field amplitude loop opens more and shows strong enhancement near the coercive bias (i.e. where the switching is happening). Furthermore, the corresponding on-field phase loop shows distinctly reduced coercive bias compared to the off-field phase loop. This could be due to the (within resolution) instantaneous relaxation of the ferroelastic domains back to their virgin state after the removal of the external electric field. This type of effect is only possible when the domains are unpinned and free to move reversibly under an external electric field [6,28], thus reinforcing our observations in Figs. 5 and 7.

3.4. Dense domain dynamics at the junction

We conclude with a quantitative demonstration of the enhanced ferroelastic switching activity at the so-called cellular or three-domain wall junctions. Cross-sectional TEM (Fig. 8a and b) and vertical PFM images (Fig. 8c) reveal that the domain arrangement in this bilayer thin film is far from a simple two-domain (c/a/c/a) structure; rather, it has a complex three-domain (c/a/c/a) architecture with a cellular arrangement as described by Roytburd et al. [29] (see the 3-D schematic in Fig. 8d). In order to investigate the switching behavior at a ferroelastic domain wall junction and to understand the underlying mechanism of wall twisting under an applied electric field [6], BE-SSPFM was performed inside the region marked by the square in Fig. 9a. The resultant BE spatial map (30 × 30 pixel) of the SP illustrated in Fig. 9b clearly shows the junction region marked inside the rectangular box. A careful and more precise measurement of the local coercive field (positive, negative and mean) along a domain towards the junction and away from the junction is plotted as a function of spatial position (Fig. 9c). The same plot also shows the coercive field (mean values) from the edge region of the domains. Interestingly, one can find a nearly 50% decrease in the value of the coercive field for regions close to the junction region, whereas the coercive fields at the edges remain constant irrespective of their...
domain alignment. This proves that the energy landscape for switching in the proximity of a ferroelastic domain junction is significantly different from the simple ferroelectric or even two-domain ferroelastic switching; the reduced coercive fields reveal that a cellular ferroelastic domain wall boundary has enhanced nucleation activity at the center. We believe that this behavior is indicative of the relative ease with which the junction between two orthogonal domain textures can shift, compared to the wall twist within the texture.

3.5. Discussion

The spatially resolved mapping of hysteresis behavior in the dense domain films above illustrate that the strong electromechanical response is closely linked to the ferroelastic...
domain wall dynamics, as previously known. However, three key experimental observations can be enumerated. First, the wall motion on the small scales is generally reversible, are not coupled with hysteretic behavior and do not contribute to the off-field response, as shown in Figs. 2 and 3. Secondly, wall twists can demonstrate both reversible (for small fields – Figs. 5 and 7) and irreversible dynamics associate with significant rearrangement of domain structure. Finally, the junctions between the domain textures (e.g. \(a_1-c\) and \(a_2-c\)) are extremely labile (Fig. 9). Below, we consider the role of these phenomena in the context of reversible and irreversible dynamics and enhanced responses in the ferroelectrics.

The most well-understood mechanism of hysteresis behavior in ferroics is local wall pinning. Such pinning is possible only on a relatively long length scale above the Larkin length of the domain wall [30], above which the walls develop characteristic irregular fractal geometry [31]. These mechanisms have been observed for ferroelectric walls in several previous works [31–34]. However, for ferroelastic walls, the strong elastic effects associated with the twin interface and wall–wall repulsion result in an increase in the Larkin length that in our case exceeds lateral domain sizes. This is also demonstrated by DC bias switching images (Fig. 3b and c), where (within instrument resolution) no fractal bending of domain walls was observed upon bias. Instead of such roughening, we reported clear reorientation of the domain walls along the in-plane directions. Hence, while we see enhancement of the response at the walls, confirming the model of Pertsev and Emelyanov [35], there is no hysteresis here (only on-field response).

On the other hand, an interesting physical mechanism we observe as a ubiquitous aspect here is the twist of these ferroelastic domains under bias. We argue that such small twists...
(on the scale of less than the domain wall period) are reversible, and probably explain the on-field and off-field loops in Fig. 7. On the other hand, large twists are associated with large changes in domain structures and are irreversible, as seen in the DC bias switching experiments. For such twists to occur on a reversible basis, the underlying crystallographic framework must be amenable to local variations in the polarization directions (and correspondingly the crystallographic directions). The domain wall junctions between two textures allow the irreversible motion (at essentially zero restoring forces), acting as a lubricant between the two mechanical interfaces. Correspondingly, combined with the well-known evolution of domain structures in polycrystalline ferroelectrics (no domain for small grains, a single texture in larger grains, multiple textures in large grains), this domain wall twist–junction motion mechanism of hysteresis and enhanced responses may offer an alternative insight into the emergent collective phenomena in ferroelectric materials. Additionally, these studies can rationalize previous observations that have studied nonlinearity as a function of grain size. Large grains sizes would support families of lamellar domains. This will have potential for junction motion and hence nonlinearity. On the other hand, if grains are small enough to have only one (or no) texture, there is no small-field nonlinearity and only large-field switching.

4. Conclusions

In summary, the reversible and irreversible domain dynamics in polydomain ferroelectric films are studied on the inter- and intragranular levels. Using innovative spectroscopic PFM experiments, we demonstrate the twist mechanism for domain reorientation in uniform domain textures. Using on- and off-field spectroscopy, we demonstrate that for small bias amplitudes the twist is reversible, and leads to significant enhancement of the on-field electromechanical response. The comparison of vertical and lateral PFM spectra further validates this model. Finally, we demonstrate that the junctions between the domain textures can act as facile interfaces to enable the twist mechanism in uniform fields. These observations provide a direct fundamental insight into the mesoscopic mechanisms of nonlinear and hysteretic behavior in polycrystalline ferroelectrics and, furthermore, outline the physical domain phenomena that may be desired in new and improved (nano)electromechanical devices. It should be noted that the observations above have been specifically made for the case of a bilayered film system. Similar findings for bulk polycrystalline ferroelectrics are needed to confirm whether this mechanism is an universal phenomenon.

Acknowledgements

The work at UNSW was supported by ARC Discovery and LIEF Grants. V.A. acknowledges the ARCNN overseas travel grant to visit Oak Ridge National Laboratory (ORNL). A portion of this research at the Center for Nano-phase Materials Sciences (CNMS), ORNL (under user proposal CNMS2008-263) was sponsored by the Scientific User Facilities Division, Office of Basic Energy Sciences, US Department of Energy. The work was supported in part (S.J. and S.V.K.) by the division of Scientific User Facilities, US Department of Energy, through CNMS. F.K. and J.W. acknowledge the support of the Science and Engineering Research Council – A*Star, Singapore, under Grant No. 052 101 0047, and the National University of Singapore.

References


Please cite this article in press as: Anbusathaiah V et al. Ferroelastic domain wall dynamics in ferroelectric bilayers. Acta Mater (2010), doi:10.1016/j.actamat.2010.06.004