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Electric-field-induced domain switching and pinning state in lead-free ferroelectric BST 80/20 film

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ABSTRACT

Ferroelectric nanodomains were created in $Ba_{0.8}Sr_{0.2}TiO_3$ (BST 80/20) thin films by applying a voltage to a sharp conducting tip of a scanning probe microscope (SPM). The ferroelectric layer were grown on (100)-oriented silicon substrate by radio frequency magnetron sputtering. The surface of the sample shows small grains which diameter ranges from 50 nm to 75 nm and roughness is less than 5 nm. Using the piezoresponse mode of the SPM to detect the out-of-plane film polarization, the domain sizes were measured as a function of the applied writing voltage and the pulse time. It was found that the time dependence of the domain diameter in a 400 nm thick BST 80/20 film well described by logarithmic law observed earlier in $Pb(Zr_{0.2}Ti_{0.8})O_3$ (PZT) films. The dynamics of domain growth is analyzed theoretically taking into account the strong inhomogeneity of the external electric field in the film and the influence of the bottom electrode. Therefore, the BTS film with good polarization switching properties could act as a memory element in nonvolatile ferroelectric random access memory (NV-FRAM) devices.

Keywords: BST film, lead-free ferroelectrics, domain switching, piezoresponse force microscopy.

1. INTRODUCTION

Previous and today's dynamic random access memories (DRAMs) have been advanced by mainly focusing on how to make memory cells small to realize high density DRAMs. The most critical challenges in gigabit density DRAMs are yield loss due to large die size and small feature size, standby current failure caused by large chip size and small data retention times owing to reduced charge packet in the memory cell. In the recent years thin film perovskite materials with high dielectric constant such as PZT, $SrTiO_3$ and $(Ba,Sr)TiO_3$ (BST) have been investigated as dielectric materials for future DRAMs [1]. In order to analyze and understand domain structures and their domain growth dynamics in the ferroelectric films, piezoresponse force microscopy (PFM) has been widely used due to its high resolution, ambient measurement, and easy manipulation of polarization states [2]. The nanoscale studies of the ferroelectric domains still need to be performed to better understand the nanoscale domain growth dynamics and achieve high memory density near or beyond 1 $Tb \cdot in^{-2}$ because they are directly related to the nanoscale bit writing of the memory devices [3]. Recent nanoscale PFM-based studies have demonstrated that quenched defects inside the FE thin films play important roles in domain switching processes, including defect-mediated inhomogeneous nucleation, pinning-dominated nonlinear dynamics of domain walls, and many other intriguing phenomena [4,5]. The aim of this work is to investigate the local piezoelectric properties and nanoscale domain growth dynamics of nanostructured of lead-free ferroelectrics BST thin films deposited on a silicon substrate suitable for the silicon integrated circuit technologies. In addition, we observed the domain wall speeds of BST thin film by using (PFM), and there by demonstrated that the domain wall motion depends on the activation energy of their domain wall.

2. EXPERIMENTAL METHODS

The ferroelectric $Ba_{0.8}Sr_{0.2}TiO_3$ films with a thickness of 400 nm were prepared by the high-frequency reactive sputtering of a ceramic target in an oxygen atmosphere on the PLAZMA-50 SE setup as described elsewhere [6]. The crystallographic structure of the films obtained was investigated by X-ray diffraction as reported in the former paper [7],

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showing that the films possess pure perovskite structure. The piezoelectric response and domain switching effects were recorded in DART-PFM mode (MFP-3D Asylum Research, Oxford Instruments). For this, a conductive cantilever of spring constant 2 N/m and a Ti/Ir coated tip of radius 28 nm was used. All measurements were performed at ambient temperature.

3. RESULTS AND DISCUSSIONS

A series of experiments were done to study the motion velocity of the domain walls in thin BST films. In this experiment the induced ferroelectric domains are formed by application of constant voltage pulses to a conducting cantilever (the cantilever in this case plays the role of the upper electrode), and the lateral size of a domain (radius) is measured as a function of a pulse duration. Figure 1a shows the PFM images of the array of 12 domains formed under action of the ±40 V voltage pulses of duration from 1 to 300 s. In order to exclude the effect of grain boundaries on the lateral growth of the created domain, the selected region was previously polarized by the voltage ±40 V. Before writing domains with negative voltage, rectangular shapes have been poled oppositely. Domain relaxation between writing and reading was not negligible. In both cases it is seen that the domain size depends strongly on duration of the voltage pulses applied between the tip and the bottom electrode.

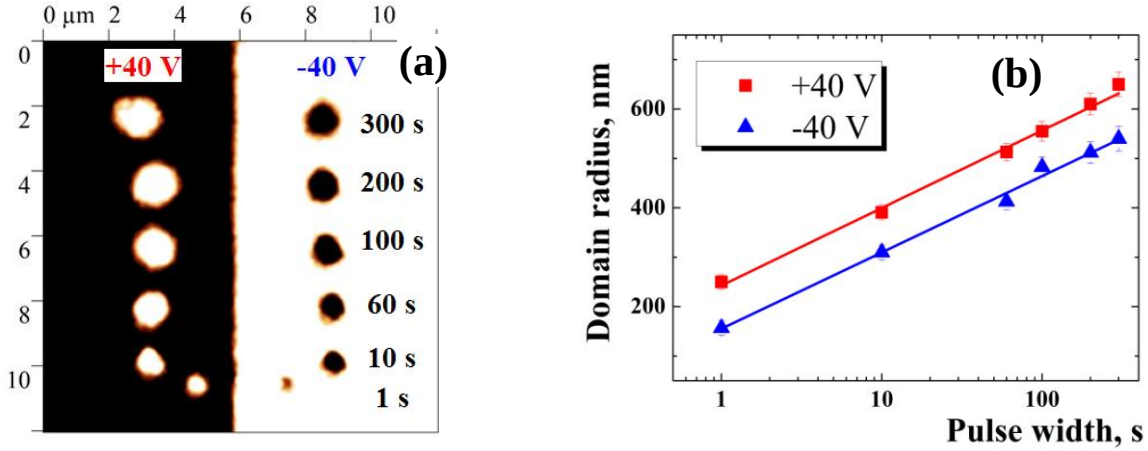

Figure 1. PFM images of dot patterned domains formed by positive and negative voltage pulses with different durations (a) and domain size dependence on the pulse conditions at negative and positive voltage pulses (b). Red and blue solid lines are fits to a logarithmic law.

As can seen from PFM image (figure 1a) the lateral size of the domains increases monotonously with the recording time (τ), which is 1, 10, 60, 100, 200, and 300 s. Figure 1b shows the dependences of the domain radius (r) on pulse duration τ at the constant voltage ±40 V. It is seen that growth of the domain goes fast at the short recording time $\tau < 60$ s and is considerably slowed down at $\tau > 100$ s. As shown in figure 1b, the domain size was linearly proportional to logarithmic value of the pulse width [3,8]. Also the difference in the radii of the induced domains depending on the sign of the polarizing voltage. For the same absolute applying voltage, the positive voltage pulses generate much larger domains compared to the negative voltage pulses as presented in figures 1a,b. This might be originated from the pinned region of BST 80/20 film. This negative pinning implies that if the modulus (amplitude) of the bias voltage is the same, easier switching/polarization switching of domains is reached at the positive voltage rather than at the negative. This pinning might come from the self-polarized region or the interface with nonuniform properties. The self-polarized regions can exist at ferroelectric/electrode interface. The approximation of the experimental data shown in Figure 1b by logarithmic law:

$$r(t) \approx r_0 \ln\left(\frac{t}{\tau}\right) + A \qquad (1)$$

gives the minimal size (radius r_0) of the induced domain and the pulse duration (τ) applied for creation of such a domain, A is constant. From fitting, we observed that for formation a minimal domain with radius 68 nm for negative voltage pulse is needed more time that for positive voltage: 0.1 s and 0.02 s, respectively.
For further understanding on the domain growth dynamics, the domain wall velocity was obtained from the growth of the lateral size. Figure 2 presents that the domain wall velocity of downward domains (positive voltage) was faster than that of upward domains (negative voltage).

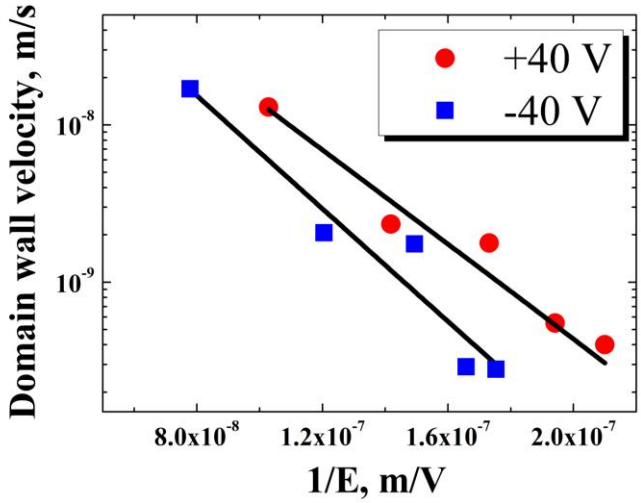

Figure 2. Electric field E dependence of domain wall velocity for the positive (red circles) and the negative (blue squares) voltage pulses. Black solid lines were approximated by Eq.2.

From the obtained values, domain wall velocity (υ) dependence on electric field (E) was described in figure 2 for each applying voltage pulse and they were approximated by the following equation [3,9]:

$$\upsilon = \upsilon_{\infty} \exp\left(-\frac{E_A}{E}\right), \tag{2}$$

where E_A is activation field of wall motion.

From the slope of the fitted lines in figure 2, activation fields of wall motion for positive and negative voltage pulses could be obtained to be 3.4×10^7 and 4.1×10^7 V/m, respectively. Activation field of positive voltage is smaller than that of negative voltage. This asymmetry in activation field is correlated with the pinning state of the films. The activation field was in the same order with those of other ferroelectric materials [10].
Figure 3a,b shows the PFM amplitude and phase hysteresis loops obtained on the as-grown BST 80/20 film. As expected, the BST film exhibited an asymmetric hysteresis behavior.

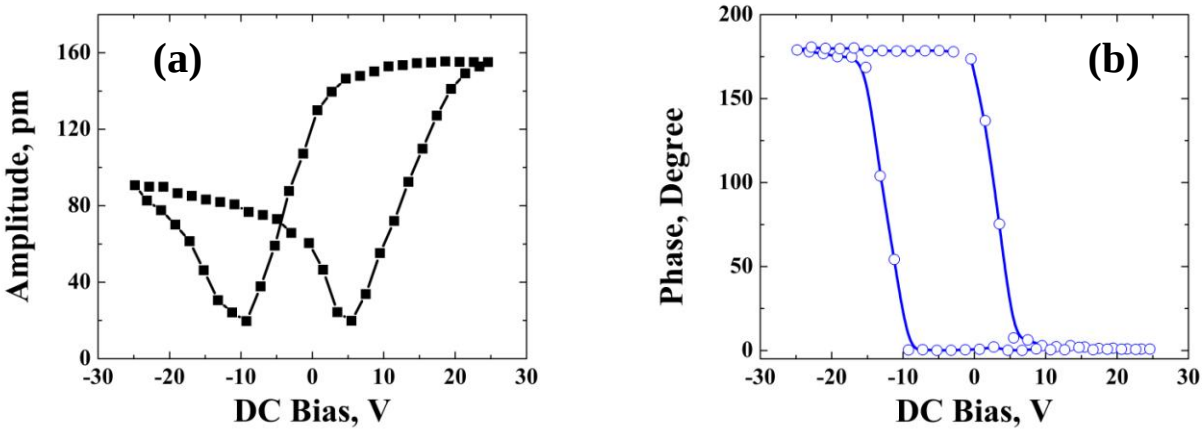

Figure 3. PFM amplitude (a) and PFM phase hysteresis (b) loops measured in BST film.

The hysteresis loops are off-centered toward the negative direction, indicating the large polarization imprint. In addition, the upward polarization state (at negative bias) was hardly detectable because the retention time was too short.

Figure 4 shown the PFM image of dot patterned domains on as-grown BST 80/20 film applying negative voltages to the tip of SPM. For the investigation of written domains on as-grown surface of the BST film the negative voltage pulses were applied to fixed locations within this area, thus we have an array of 23 stable domains created by applying voltage pulses of fixed height (V_{tip}=-20 V ÷ -60 V with step 10V) and various durations ranging from 1 to 100 s. The dynamics of domain growth is analyzed experimentally taking into account the strong inhomogeneity of the external electric field in the film.

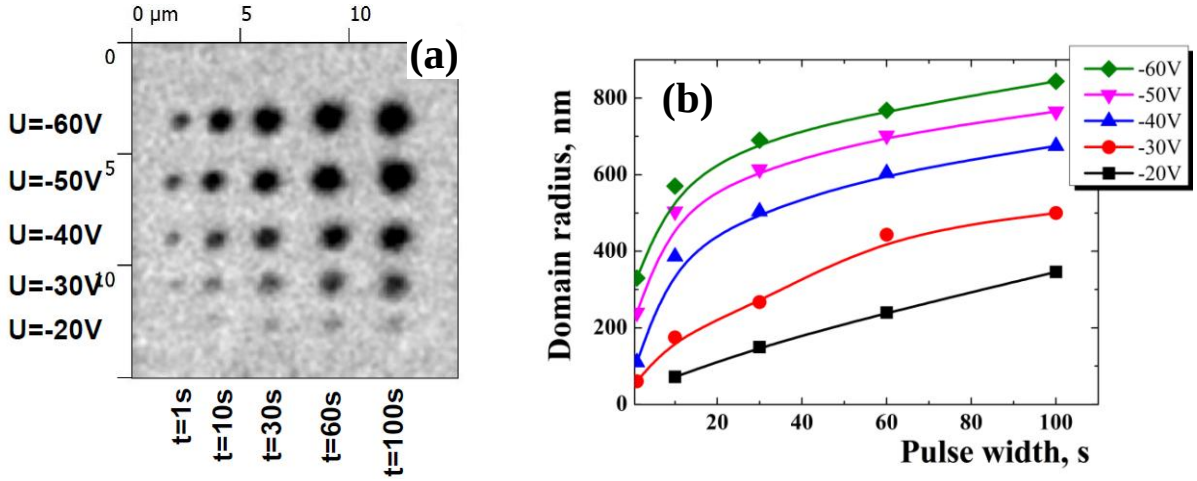

Figure 4. PFM image of dot patterned domains formed by negative voltage (a) and domain size dependence on the pulse conditions (b). Solid lines are guide lines for eye.

The dot patterned domains of figure 4 show two separate domain growth steps [11]. First, they show nucleation and forward domain growth. Then, they undergo lateral domain growth from the nucleation site. For higher DC voltage pulses (≥ -30 V) there is a nonlinear growth of domains (exponential law) while for smaller voltages (DC= -20 V) there is a linear growth. Thus, under higher voltage pulses, such as ≥ -40V, the domain diameter first increased steeply with the duration time and then grew slowly when the domain approached a stable size. The domain growth behaviors $r(t)$

under relatively high and low voltage pulses followed the nonactivated and activated motion as suggested by Molotskii [12] respectively. When the driving energy from the external field exceeded the periodic barriers, the domain wall performed a nonactivated motion as in a viscous medium. By comparison, when the driving energy was smaller than the barriers, the domain wall performed the activated motion by crossing the barriers with thermal fluctuations.

4. CONCLUSIONS

Summarizing our research, $Ba_{0.8}Sr_{0.2}TiO_3$ thin films with a thickness 400 nm were prepared on Si substrates by the RF magnetron sputtering. We have investigated the nanoscale domain growth dynamics of ferroelectric BST thin films using PFM on both as-grown and prepoling surface. The nanoscale domains of dot patterns were formed by applying voltage pulses to the tip of SPM. The domain radius was linearly proportional to logarithmic value of the pulse width. The observed difference between the linear sizes of the domains formed at different signs of electric field is associated with the influence of interface effects on propagation of polarization under the cantilever. Also, the obtained activation field was in the same order with ferroelectric oxides. Therefore, the BTS film with good polarization switching properties could act as a memory element in nonvolatile ferroelectric random access memory (NV-FRAM) devices.

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REFERENCES

[1] Scott, J.F, “Ferroelectric Memories”, Springer, 255 (2000).
[2] Kholkin, A. L., Shvartsman, V. V., Kiselev, D. A. and Bdikin, I. K. “Nanoscale characterization of ferroelectric materials for piezoelectric applications,” Ferroelectrics. 341(1), 3-19 (2006).
[3] Tybell, T., Paruch, P., Giamarchi, T. and Triscone, J. M. “Domain wall creep in epitaxial ferroelectric $Pb(Zr_{0.2}Ti_{0.8})O_3$ thin films,” Physical Review Letters, 89(9), 097601 (2002).
[4] Yang, S. M., Yoon, J. G. and Noh, T. W. “Nanoscale studies of defect-mediated polarization switching dynamics in ferroelectric thin film capacitors,” Current Applied Physics. 11(5), 1111-1125 (2011).
[5] Kalinin, S. V., Morozovska, A. N., Chen, L. Q. and Rodriguez, B. J. “Local polarization dynamics in ferroelectric materials,” Reports on Progress in Physics. 73(5), 056502 (2010).
[6] Afanasiev, M.S., Kiselev, D.A., Levashov, S.A., Luzanov, V.A., Nabiyev, A.A., Naryshkina, V.G., Sivov, A.A. and Chucheva, G.V. “The Influence of the substrate material on the structure and electrophysical properties of $Ba_xSr_{1-x}TiO_3$ thin films,” Phys. Solid State. 60(5), 954-957 (2018).
[7] Afanasev, M.S., Byrov, A.V., Egorov, V.K., Lychnikov, P.A. and Chucheva G.V., “Technological control of element structure perovskite layer of planar nano structures deposited in HF-plasma,” Science Intensive Technologies. 13(5), 53–57 (2012).
[8] Guo, E. J., Roth, R., Herklotz, A., Hesse, D. and Dörr, K. “Ferroelectric 180 domain wall motion controlled by biaxial strain,” Advan. Mater. 27(9), 1615-1618 (2015).
[9] Kim, Y., Kim, W., Choi, H., Hong, S., Ko, H., Lee, H. and No, K. “Nanoscale domain growth dynamics of ferroelectric poly (vinylidene fluoride-co-trifluoroethylene) thin films,” Appl. Phys. Lett., 96(1), 012908 (2010).
[10] Chen, Y. C., Lin, Q. R. and Chu, Y. H., “Domain growth dynamics in single-domain-like $BiFeO_3$ thin films,” Appl. Phys. Lett. 94(12), 122908 (2009).
[11] Gruverman, A., Rodriguez, B. J., Dehoff, C., Waldrep, J. D., Kingon, A. I., Nemanich, R. J. and Cross, J. S., “Direct studies of domain switching dynamics in thin film ferroelectric capacitors,” Appl. Phys. Lett. 87(8), 082902 (2005).
[12] Molotskii, M. “Generation of ferroelectric domains in films using atomic force microscope,” Jour. App. Phys. 97(1), 014109 (2005).