AIP Applied Physics Letters

Large electric field induced strains in ferroelectric islands

J. X. Zhang, G. Sheng, and L. Q. Chen

Citation: Appl. Phys. Lett. **96**, 132901 (2010); doi: 10.1063/1.3373915
View online: http://dx.doi.org/10.1063/1.3373915
View Table of Contents: http://apl.aip.org/resource/1/APPLAB/v96/i13
Published by the American Institute of Physics.

APPLIED PHYSICS LETTERS **96**, 132901 (2010)

Large electric field induced strains in ferroelectric islands

J. X. Zhang,[a)] G. Sheng, and L. Q. Chen
Department of Materials Science and Engineering, The Pennsylvania State University, University Park, Pennsylvania 16802, USA

(Received 4 February 2010; accepted 4 March 2010; published online 29 March 2010)

An effective mechanism that generates large recoverable electric field induced strains in ferroelectric islands is studied by phase-field modeling. The large strains originate from the reversible 90° domain switching between a_1 (a_2) domains and c domains, driven by an applied electric field and an internal stress field. The electric field induced strains could be effectively controlled by the magnitude of island-substrate misfit and the aspect ratio of islands. © *2010 American Institute of Physics*. [doi:10.1063/1.3373915]

Ferroelectric materials exhibit superior piezoelectric properties and are used in a wide range of applications such as actuators and sensors. The large piezoelectric response of these materials is due in part to intrinsic contribution (piezoelectric lattice strain) and in part due to extrinsic contribution, i.e., non-180° domain switching. The extrinsic contribution could be significantly enhanced through the control of domain switching. Large electric field induced strains were obtained in a single $\mathrm{BaTiO_3}$ crystal with the use of external compressive stress and oscillating electric field.[1,2] At zero applied electric field, the applied stress forces the polarization along the $[100]_p$ or $[010]_p$ direction, where the subscript p denotes pseudocubic indices. When an external electric field is applied along the $[001]_p$ direction, a large strain is achieved due to extensive 90° domain switching. More importantly, the strain is recoverable as the applied stress could drive the polarization back to the $[100]_p$ or $[010]_p$ direction after the applied electric field is removed. However, a complicated loading system is required to provide the constant stress in this approach, which limits its application in piezoelectric devices. To overcome such limitation, Ren and his colleagues[3,4] found a different mechanism for large electric field induced strains stemming from non-180° domain switching, where the interaction between polarization and point defects is the driving force for recovering the strains. A large recoverable strain of 0.75% at a low field generated from an aged $\mathrm{BaTiO_3}$ single crystal is reported.[3]

In this paper, we report an effective mechanism that generates large recoverable electric field induced strains in ferroelectric islands. The large strains originate from the reversible 90° domain switching between a_1 (a_2) domains and c domains, driven by the applied electric field as well as the internal stress field originating from the misfit between island and substrate. The electric field induced strains could be effectively controlled by the magnitude of island-substrate misfit and the aspect ratio of islands, which facilitates its potential applications.

A recently developed phase-field model of ferroelectric islands is employed in this work.[5] In the model, temporal domain structure evolution is described by the time-dependent Ginzburg–Landau equations,

$$\frac{\partial P_i}{\partial t} = -L\frac{\delta F}{\delta P_i}, \quad i=1,2,3, \tag{1}$$

where P_i is spontaneous polarization field, L is a kinetic coefficient that is related to the domain evolution, and F is the total free energy of the system,

$$F = F_{\mathrm{bulk}} + F_{\mathrm{wall}} + F_{\mathrm{elec}} + F_{\mathrm{elas}}, \tag{2}$$

where F_{bulk}, F_{wall}, F_{elec}, and F_{elas} are the bulk free energy, domain wall energy, electrostatic energy, and elastic energy, respectively. The mathematical expressions of these energy terms are given in Ref. 5.

As an example, $(001)_p$ oriented $\mathrm{PbZr_{0.2}Ti_{0.8}O_3}$ (PZT) islands grown on a cubic substrate are studied in this paper. We employ a model of $128\Delta\times128\Delta\times36\Delta$, and the thickness of the substrate is taken as $h_s=16\Delta$. The Landau free energy coefficients, electrostrictive coefficients, and elastic constants of $\mathrm{PbZr_{0.2}Ti_{0.8}O_3}$ are collected from literature:[6] $\alpha_1=3.44(T\text{-}456.4)\times10^5$ $(\mathrm{C^{-2}\,m^2\,N})$, $\alpha_{11}=-3.05\times10^7$ $(\mathrm{C^{-4}\,m^6\,N})$, $\alpha_{12}=6.32\times10^8$ $(\mathrm{C^{-4}\,m^6\,N})$, $\alpha_{111}=2.48\times10^8$ $(\mathrm{C^{-6}\,m^{10}\,N})$, $\alpha_{112}=9.68\times10^8$ $(\mathrm{C^{-6}\,m^{10}\,N})$, $\alpha_{123}=-4.90\times10^9$ $(\mathrm{C^{-6}\,m^{10}\,N})$, $Q_{11}=0.081$ $(\mathrm{C^{-2}\,m^4})$, $Q_{12}=-0.024$ $(\mathrm{C^{-2}\,m^4})$, $Q_{44}=0.064$ $(\mathrm{C^{-2}\,m^4})$, $c_{11}=1.75\times10^{11}$ $(\mathrm{N\,m^{-2}})$, $c_{12}=0.794\times10^{11}$ $(\mathrm{N\,m^{-2}})$, and $c_{44}=1.11\times10^{11}$ $(\mathrm{N\,m^{-2}})$, and $T=25$ °C. The cell size in real space is chosen to be $\Delta=l_0$, where $l_0=\sqrt{G_{110}/\alpha_0}$ and $\alpha_0=|\alpha_1|_{T=25\,°\mathrm{C}}$. We choose the gradient energy coefficients as $G_{11}/G_{110}=0.6$. If $l_0=1.0$ nm, $G_{110}=1.73\times10^{-10}$ $(\mathrm{C^{-2}\,m^4\,N})$, and the corresponding domain wall thickness is about 1.5 nm.

For $\mathrm{PbZr_{0.2}Ti_{0.8}O_3}$, there are six possible orientation variants at room temperature with the polarization along the $[100]_p$, $[\bar{1}00]_p$, $[010]_p$, $[0\bar{1}0]_p$, $[001]_p$, and $[00\bar{1}]_p$ directions

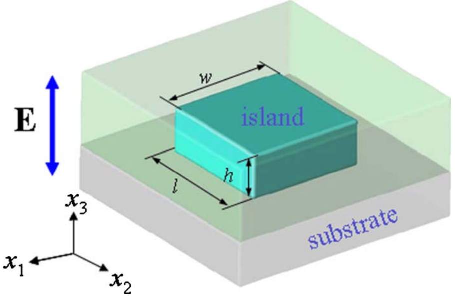

FIG. 1. (Color online) Schematic illustration of a ferroelectric island.

[a)] Current address: Research and Development, Carpenter Technology Corporation. Reading, PA 19601. Electronic mail: jxianzhang@hotmail.com.

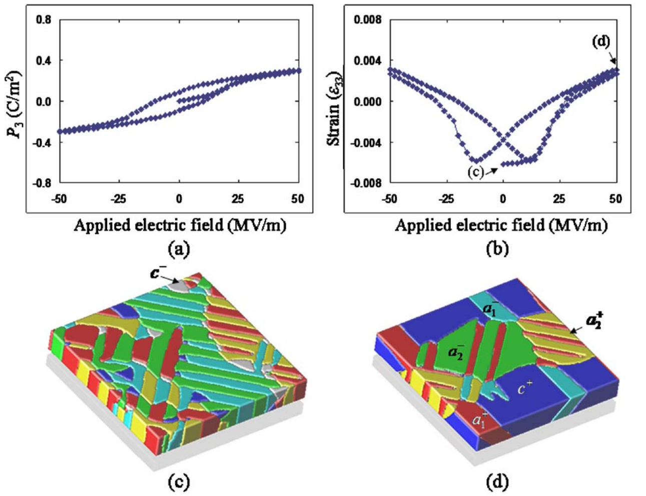

FIG. 2. (Color online) (a) P_3 vs applied electric field and (b) strain ε_{33} vs applied electric field curves of a continuous PZT thin film ($h=16$ nm). The corresponding domain structures at (c) 0 MV/m and (d) 50 MV/m) (substrate induced strain = 0.8%). Each color represents one type of domain. The corresponding domain is indicated in the figures.

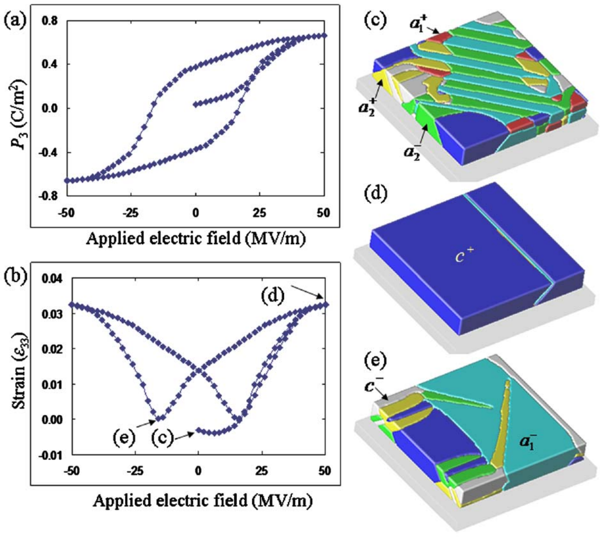

FIG. 3. (Color online) (a) P_3 vs applied electric field and (b) strain ε_{33} vs applied electric field curves of a PZT island ($w=l=112$ nm, $h=16$ nm, and substrate induced strain=0.8%). The corresponding domain structures at (c) 0 MV/m, (d) 50 MV/m, and (e) −16 MV/m. Each color represents one type of domain. The corresponding domain is indicated in the figures.

of cubic paraelectric phase. They are labeled as a_1^+, a_1^-, a_2^+, a_2^-, c^+, and c^- domain, respectively. Under an applied electric field or stress, the polarization could switch among these orientations, i.e., 90° domain switching and 180° domain switching. In this work, an external electric field ($\mathbf{E}$) is applied perpendicular to the film surface as shown in Fig. 1, and a single c^+ (or c^-) domain structure is expected with a large enough electric field. Therefore, to maximize 90° domain switching and hence the electric field induced strain, a_1 and a_2 domains are preferred at zero electric field.

It has been well studied that the volume fraction of various domains in ferroelectric PZT films could be effectively controlled by substrate induced strains.[7–9] Large compressive strains prefer c domains while large tensile strains result in a mixture of a_1 and a_2 domains. In between, the coexistence of a_1, a_2, and c domains is expected. As an example, Fig. 2(c) shows the domain structure of a PZT film with a 0.8% tensile strain, and it consists of 97% a_1 (a_2) domains and 3% c domains. When an external electric field is applied along the $[001]_p$ direction, the electrostatic energy drives the switching of a_1 and a_2 domains to c^+ domains. However, there is a large elastic energy penalty for such switching process caused by substrate constraint since elastic energy was minimized with the original volume fractions of various domains. As shown in Fig. 2(d), there are still a large amount of a_1 (a_2) domains left (39%) even with a large electric field, so 90° domain switching is limited.

Previous studies demonstrate that[5,10–12] by cutting a continuous film into isolated islands, the substrate constraint could be partially removed due to the free surfaces. Thus the 90° domain switching from a_1 and a_2 domains to c^+ domains is easier than the continuous film, and a nearly pure c^+ domain is obtained with an electric field of 50 MV/m, as shown in Fig. 3(d). However, to obtain a recoverable electric field induced strain, the a_1 and a_2 domains need to be restored when the applied electric field is removed. As studied previously,[5] inside the island, there exists internal in-plane stress generated by the misfit between the island and substrate, which could be used as the restoring driving force for a_1 and a_2 domains. The sign and magnitude of the internal stress depend on island-substrate misfit and the aspect ratio of island, i.e., lateral dimension over island thickness $w(l)/h$. A tensile in-plane stress was generated under a tensile substrate induced strain (0.8%), favoring a_1 and a_2 domains. As a result, with the decrease of the applied electric field, c^+ domains switch back to a_1/a_2 domains gradually. Eventually, as much as 77% a_1 (a_2) domains have been restored as

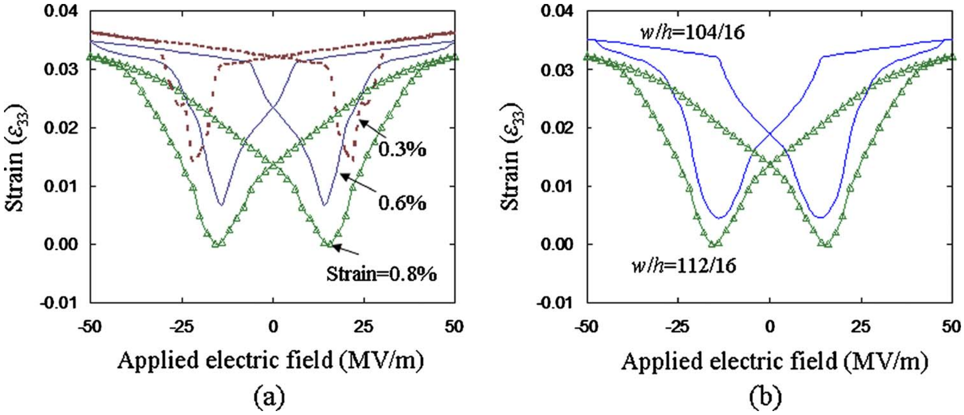

FIG. 4. (Color online) Strain ε_{33} vs applied electric field curves as a function of (a) substrate induced strains ($w=l=112$ nm, $h=16$ nm) and (b) aspect ratio w/h. ($h=16$ nm, substrate induced strain=0.8%).

shown in Fig. 3(e). As a result, a large recoverable electric-field induced strain of ~3% was obtained [Fig. 3(b)], which is about three times larger than that of the continuous film under the same electric field [Fig. 2(b)].

By controlling the substrate induced strains, the driving force for restoring a_1/a_2 domain could be effectively changed. As shown in Fig. 4(a), when the tensile substrate induced strains decrease, a much smaller recoverable electric field induced strain was obtained since only a small fraction of a_1/a_2 could be restored. The maximal ε_{33} achieved (deformation at electric field of 50 MV/m), however, increases with the decrease of substrate induced strains. It should be noted that large compressive substrate induced strains prefer *c* domains and 180° domain switching. For such a case, the intrinsic contribution dominates and a ferroelectric island behaves as a single domain single crystal, as shown in experimental observations.[13] On the other hand, the internal stress inside islands also depends on the aspect ratio of islands. As shown in the previous study,[5] the smaller the aspect ratio, the smaller the average in-plane stress. Therefore, the electric field induced strains could also be controlled by the aspect ratio of islands as shown in Fig. 4(b).

In summary, we report an effective mechanism that generates large reversible electric field induced strains in ferroelectric islands without using complicate loading systems. The mechanism takes advantage of the misfit between the islands and substrate to recover the strain. The electric-field induced strains could be effectively controlled by the magnitude of island-substrate misfit and the aspect ratio of islands, which further facilitate its potential applications.

The authors are grateful for the financial support from the National Science Foundation through Grant No. ECCS-0708759.

[1] E. Burcsu, G. Ravichandran, and K. Bhattacharya, Appl. Phys. Lett. **77**, 1698 (2000).
[2] E. Burcsu, G. Ravichandran, and K. Bhattacharya, J. Mech. Phys. Solids **52**, 823 (2004).
[3] X. Ren, Nature Mater. **3**, 91 (2004).
[4] L. X. Zhang, W. Chen, and X. Ren, Appl. Phys. Lett. **85**, 5658 (2004).
[5] J. X. Zhang, R. Wu, S. Choudhury, Y. L. Li, S. Y. Hu, and L. Q. Chen, Appl. Phys. Lett. **92**, 122906 (2008).
[6] M. J. Haun, E. Furman, S. J. Jang, H. A. Mckinstry, and L. E. Cross, J. Appl. Phys. **62**, 3331 (1987).
[7] Y. L. Li, S. Choudhury, Z. K. Liu, and L. Q. Chen, Appl. Phys. Lett. **83**, 1608 (2003).
[8] Y. L. Li, S. Y. Hu, and L. Q. Chen, J. Appl. Phys. **97**, 034112 (2005).
[9] K. S. Lee, J. H. Choi, J. Y. Lee, and S. Bark, J. Appl. Phys. **90**, 4095 (2001).
[10] S. Bühlmann, B. Dwir, J. Baborowski, and P. Muralt, Appl. Phys. Lett. **80**, 3195 (2002).
[11] V. Nagarajan, A. Stanishevsky, L. Chen, T. Zhao, B. T. Liu, J. Melngailis, A. L. Roytburd, and R. Ramesh, Appl. Phys. Lett. **81**, 4215 (2002).
[12] Z. Ma, F. Zavaliche, L. Chen, J. Ouyang, J. Melngailis, A. L. Royburd, V. Vaithyanathan, D. G. Schlom, T. Zhao, and R. Ramesh, Appl. Phys. Lett. **87**, 072907 (2005).
[13] V. Nagarajan, A. Roytburd, A. Stranishevsky, S. Prasertchoung, T. Zhao, L. Chen, J. Melngailis, O. Auciello, and R. Ramesh, Nature Mater. **2**, 43 (2003).