

Direct observation of asymmetric domain wall motion in a ferroelectric capacitor

Ja Kyung Lee^{a,1}, Ga Young Shin^{a,1}, Kyung Song^a, Woo Seok Choi^b, Yoon Ah Shin^a,
Seong Yong Park^c, Jason Britson^d, Ye Cao^d, Long-Qing Chen^d,
Ho Nyung Lee^b, Sang Ho Oh^{a,*}

^a Department of Materials Science and Engineering, Pohang University of Science and Technology (POSTECH), San 31, Pohang 790-784, Republic of Korea

^b Materials Science and Technology Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831, USA

^c Analytical Science Group, Samsung Advanced Institute of Technology (SAIT), Yongin 446-712, Republic of Korea

^d Department of Materials Science and Engineering, The Pennsylvania State University, University Park, PA 16802, USA

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Abstract

We report in situ transmission electron microscopy observations of the 180° polarization switching process of a $\text{PbZr}_{0.2}\text{Ti}_{0.8}\text{O}_3$ (PZT) capacitor. The preferential, but asymmetric, nucleation and forward growth of switched *c*-domains were observed at the PZT/electrode interfaces, arising due to the built-in electric field induced at each interface. The subsequent sideways growth of the switched domains was inhibited by the depolarization field due to the imperfect charge compensation at the counter-electrode and also at the boundaries with preexisting *a*-domains, which contributed further to the asymmetric switching behavior. It was found that the preexisting *a*-domains split into fine *a*- and *c*-domains constituting a 90° stripe domain pattern during the 180° polarization switching process, revealing that these domains also actively participated in the out-of-plane polarization switching. The real-time observations uncovered the origin of the switching asymmetry and further clarified the importance of charged domain walls and the interfaces with electrodes in the ferroelectric switching processes.

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1. Introduction

The spontaneous polarization in ferroelectrics can be switched by applying an electric field. The polarization switching occurs through the nucleation and subsequent forward and sideways growth of the switched domains [1,2]. The reversible switching capability has been explored extensively for a wide range of applications, including transistors and memory devices. In the technically important $\text{Pb}(\text{Zr}_{1-x}\text{Ti}_x)\text{O}_3$ (PZT) thin film capacitors, switching

between two bistable polarization states that are related by a 180° rotation is implemented by applying a uniform electric field across the film. Generating a uniform field requires the use of two planar electrodes on the top and bottom surfaces of PZT thin film. It has been demonstrated that the nature of the ferroelectric/electrode interfaces has significant effects on the switching characteristics of a ferroelectric capacitor [3–6]. For example, practical ferroelectric/electrode interfaces allow polarization switching to occur at much lower coercive fields (E_C) than the theoretical predictions assuming ideal interfaces [7,8]. Moreover, the electrical boundary conditions of the top and bottom electrodes are often not identical, which lead to asymmetric switching between the two polarization states [5,9,10].

* Corresponding author. Tel.: +82 (0)54 279 2144; fax: +82 (0)54 279 2399.

E-mail address: shoh@postech.ac.kr (S.H. Oh).

¹ These authors contributed equally to this work.

In practical PZT thin film capacitors, domains with an unfavorably oriented polarization are frequently present, which further challenge understanding of the ferroelectric switching behavior. One notable example is the ferroelastic *a*-domain with a polarization rotated 90° from that of the active ferroelectric *c*-domains. It has been reported that *a*-domain walls are immobile [11–13] and act as nucleation centers for the switched *c*-domains [13,14]. Although the question whether these *a*-domains retain their in-plane polarization or undergo internal switching during the 180° switching of the out-of-plane polarization has been discussed recently, it has not been properly addressed due to the lack of convincing experimental observations.

Therefore, understanding the detailed influence of the internal boundaries in polarization switching, such as the electrode interfaces and the boundaries with unfavorably oriented domains, is critical for future engineering of novel ferroelectric devices. In this paper, in situ transmission electron microscopy (TEM) observations of the dynamic motion of domain walls during the 180° polarization switching of a ferroelectric PZT thin film capacitor with planar Ni (top) and SrRuO₃ (SRO; bottom) electrodes are presented. The asymmetric electrode configuration was employed in order to distinguish the characteristic roles of the metal and oxide electrodes in the nucleation and growth of the switched domains. A 110 nm thick PZT film was chosen for this study because at this thickness the film contains both the ferroelastic *a*-domains and dislocations that form to relax the misfit strain [15,16]. As these defects are ubiquitous in this type of ferroelectric capacitor, their influences on the domain wall motion have critical effects on the remnant polarization, switching speed and numerous reliability issues in ferroelectric thin film capacitors [11,17,18].

2. Experimental procedures

2.1. Preparation of PZT capacitor with planar metallic electrodes

A 110 nm thick epitaxial Pb(Zr_{0.2}Ti_{0.8})O₃ film was grown epitaxially on a 20 nm thick SRO buffered Nb-doped SrTiO₃ (001) (Nb:STO) substrate by pulsed laser deposition [16]. The use of the conductive Nb:STO (001) substrate was considered to make it easy for the SRO bottom electrode to be connected to an electrically grounded Cu support grid. The Ni and Ti bilayer thin films were deposited on top of the PZT film by e-beam evaporation to a total thickness of ~350 nm. While the Ni thin film served as the top electrode constituting a metal–ferroelectric–metal (MFM) structure, the Ti thin film served as a contact pad to a Pt–Ir surface probe (see Fig. 1a). The bilayer electrode design minimizes the mechanical strain imposed on the PZT film upon contact of the surface probe. This configuration also allows an electric field to be applied homogeneously across the whole active area of the PZT film under the Ni electrode.

It is emphasized that the double planar electrode configuration adopted in this study offers unprecedented advantages over the configurations in previous studies using a tip-based point electrode in piezoresponse force microscopy [19,20] and recent in situ TEM experiments [17,18,21,22]. First, the dynamic motion of the switched *c*-domains can be examined under a uniform electric field over a large active area ($0.1 \times 1.2 \mu\text{m}^2$ in lateral size). Second, the additional piezoelectric and flexoelectric coupling effects on the spontaneous polarization that may arise from the forced contact of the probe against the film surface can be avoided [23,24]. Third, but most importantly, the use of top and bottom electrodes and the current flowing between these two electrodes described using the *I*–*V* characteristics of ideal back-to-back Schottky diodes (shown in Fig. 1a and b and discussed in Section 3.1) ensure that the electrical boundary conditions closely resemble those in an actual ferroelectric capacitor [1].

2.2. TEM sample preparation and in situ TEM

A cross-sectional specimen of the MFM capacitor for in situ TEM was prepared by utilizing a focused ion beam (FIB, Helios NanoLab™, FEI) lift-off technique. The Nb:STO substrate of the cross-sectional TEM lamellar specimen was attached to one of the sample mounting posts of the Cu support grid by depositing a layer of conductive Pt using the FIB. The lamellar specimen was isolated into several parts by cutting vertical trenches to suppress possible leakage current of the MFM capacitor. At the final stage of milling, a low energy Ga⁺ ion beam at 1 kV was used. The prepared TEM specimen was mounted on a Au wire using Ag epoxy and then the sample assembly was fixed to the piezo-stage of a STM-TEM holder (Nanofactory™). While the SRO bottom electrode of the MFM capacitor was electrically grounded through the conductive Nb:STO and the Cu support grid, the Ti/Ni top electrode was positioned to make contact with a Pt–Ir surface probe installed in the TEM holder for applying a bias. A field-emission TEM operated at 200 kV (JEM-2100F, JEOL) was used in this experiment. Real-time movies were acquired with a CCD camera (ORION 200D, Gatan) at 25 frames s^{−1}.

2.3. Diffraction contrast TEM imaging of switched *c*-domains

Conventional diffraction contrast TEM in bright-field (BF) mode was used to distinguish the *c*-domains with opposite polarization. In the case of a (001)-oriented tetragonal PZT film, the ferroelectric domains with antiparallel polarization orientations, i.e. **P**_[001] and **P**_[00 $\bar{1}$], could be distinguished easily under the diffraction contrast owing to their different diffraction intensities. In general, for a centrosymmetric crystal structure, Friedel's law dictates that the intensities of two reflections with the reciprocal lattice vectors of **g** and $-\mathbf{g}$, *I*_g and *I* _{$-\mathbf{g}$} , respectively, are the

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