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Vortex ferroelectric domains

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Abstract

We show experimental switching data on microscale capacitors of lead–zirconate–titanate (PZT), which reveal time-resolved domain behavior during switching on a 100 ns scale. For small circular capacitors, an unswitched domain remains in the center while complete switching is observed in square capacitors. The observed effect is attributed to the formation of a vortex domain during polarization switching in small circular capacitors. This dynamical behavior is modeled using the Landau–Lifshitz–Gilbert equations and found to be in agreement with experiment. This simulation implies rotational motion of polarization in the xy plane, a Heisenberg-like result supported by the recent model of Naumov and Fu (2007 *Phys. Rev. Lett.* **98** 077603), although not directly measurable by the present quasi-static measurements.

Over the past 60 years several physicists have considered the possibility that magnetic spins or electric polarization vectors might order not rectilinearly but in circles or toroids. These vortex domain structures (sometimes called “closure domains”) in ferroic materials were first predicted by Landau and Lifshitz in 1935 [1] and by Kittel in 1946 [2], who showed that formation of circular domains was likely in ferromagnetic nanodots due to the surface boundary conditions. In 1979, Mermin [3] analyzed their two-dimensional structures in terms of winding numbers, which was extended by Muxworthy *et al.* [4] to three-dimensional “vortex” domains. Such domain patterns are well known in nanoferromagnets, such as Fe/Ti ilmenite, and are best studied by electron holography [5], but have not been reported in ferroelectrics experimentally, although predicted very recently [6, 7]. The extension to ferroelectrics was made around 1984 by Ginzburg *et al.* [8] and was recently further developed using *ab initio* calculations by Naumov *et al.* [9]. We note that toroidal domains can arise for two unrelated physical reasons: (1) finite size effects and boundary conditions (present work) and (2) magnetoelectric coupling in multiferroic materials [7, 8, 10–12]. The present work does not involve magnetism.

Here we report the direct observation of a doughnut-shape domain pattern appearing during polarization reversal in micrometer-sized ferroelectric capacitors [13]. The

switching behavior of the out-of-plane polarization component P_z showing nucleation and wall propagation along the electrode edge, as well as evolution of the in-plane polarization components P_x and P_y , were simulated using the Landau–Lifshitz–Gilbert (LLG) equations. We show that the LLG model completely reproduces the P_z behavior, implying that the P_x and P_y switching kinetics are also adequately modeled. Detailed analysis of the simulated in-plane polarization pattern indicates the formation of the transient vortex domain during polarization reversal.

The switching experiments were carried out via ultra-fast stroboscopic piezoresponse force microscopy (PFM) where a conducting probing tip in contact with the top electrode was used for application of an electrical bias and detection of the domain patterns in the capacitors. A ferroelectric layer underneath the top electrode was partially switched by applying a short (less than total switching time) voltage pulse with amplitude above the threshold value. Then the entire capacitor was scanned in the PFM mode to visualize the resulting distribution of the out-of-plane, or vertical, P_z polarization component developed in the capacitor. The field was then applied for a longer time interval and the imaging PFM procedure repeated. Because domain nucleation is nearly 100% inhomogeneous in PZT, occurring at the same defect sites, it was then possible to superimpose the collection of images obtained for differ-

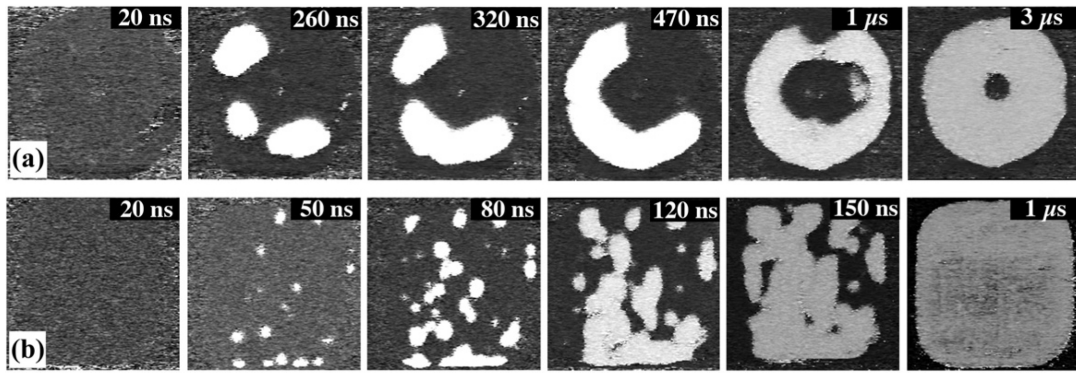


Figure 1. PFM images of instantaneous domain configurations with out-of-plane polarization component P_z developing at different stages of polarization reversal in (a) 1- μm -diameter circular capacitor and (b) 2.5 μm edge square capacitor. Applied electric field is 700 kV cm^{-1} . The total switching time for the domain in the center of the disc is about 1 s.

ent pulse durations to form a slow-motion video of nucleation and domain wall motion during polarization reversal [13]. Domain imaging has been performed through the top electrode by applying an oscillating bias of 0.6 V (peak-to-peak) at 10 kHz. Samples used in this study were 50 nm thick epitaxial (001)-oriented $\text{PbZr}_{0.2}\text{Ti}_{0.8}\text{O}_3$ (PZT 20/80) capacitors with circular 1- μm -diameter and square 2.5 μm -edge Pt top electrodes fabricated by e-beam lithography and lift-off. PZT layers were fabricated by pulsed laser deposition on $\text{SrRuO}_3/\text{SrTiO}_3(001)$ substrates. Details on film fabrication and basic properties are given elsewhere [14].

The snapshots of instantaneous domain patterns corresponding to the vertical component of polarization P_z developing in circular and square PZT capacitors during polarization reversal are shown in Figure 1. It can be seen that in the circular capacitor domains nucleate at the electrode edge and propagate around the circumference forming a doughnut-type structure (with an inner circular region having polarization P_z pointing down and the outer ring region being oriented upward) in just a few microseconds (Figure 1(a)). This structure remains for >1 s, after which the unswitched central domain collapses to leave a uniformly polarized ground state. In contrast, the square-shaped capacitors never exhibit a doughnut-type domain structure (Figure 1(b)). Instead, switching proceeds via randomly distributed nucleation events with subsequent isotropic lateral domain growth. In general, the observed features of domain kinetics can be summarized as follows.

- (1) The doughnut-type domains occur only in small circular capacitors (<1 μm in diameter) and never in larger square-shaped capacitors.
- (2) There are always several nucleation sites and all the nucleation occurs near the walls, but none occur at the domain walls.
- (3) The doughnut-type domain structure develops within the time period of several microseconds, lasts for several seconds and then transforms to a single domain state.

The experimentally observed features of domain kinetics in circular capacitors lead us to assume that it might be not simply due to the flipping of polarization but is actually

a result of vortex state formation during switching. Vortex domains are well known in magnetism. Three-dimensional micromagnetic modeling by Muxworthy *et al.* [4] predicted that magnetostatic interactions can generate vortex states in submicrometer-sized grains which strongly depend on the grain size and anisotropy. The nucleation and evolution of these vortex domain states is unclear due to periodic boundary conditions imposed in the modeling, which obscures the inhomogeneous surface nucleation process in real materials. Note that, in ferroelectrics, polarization does not generally rotate continuously in space, so that the structures shown in [4] with magnetization vectors curling around a central core are only qualitative in the description of ferroelectrics. The ferroelectric case can be approximated in magnetic models by letting the anisotropy/exchange ratio become very large. The metastable nature of the vortex domain state was not emphasized by Muxworthy *et al.* [4] but is probably another reason why vortex domains have not been reported before in ferroelectrics: they are only metastable, lasting a few seconds. The collapse of the vortex state follows from Maxwell's equations, particularly

$$-\nabla \times D = dB/dt, \quad (1)$$

which implies that any vortex is unstable against decay unless $-\nabla \times D = 0$.

The simulation of ferroelectric vortex domain formation and switching has been performed using the LLG User Manual [15], which in turn is based upon the micromagnetic equations of motion for a Heisenberg magnet as developed by Scheinfein *et al.* [16]. The time evolution of a magnetization configuration described by the Landau-Lifshitz-Gilbert equation has been solved with the pseudo-spin configuration relaxed iteratively. For the application to ferroelectric PZT we adjusted the pseudo-spin parameters such that the anisotropy is larger than in ferromagnets and the exchange energy is smaller. The LLG simulation allowed us to reproduce all the details of the experimental data on domain switching (Figure 2).

- (1) The doughnut domains occur only in small circular-shaped and never in square-shaped capacitors.
- (2) There are always several nucleation sites.

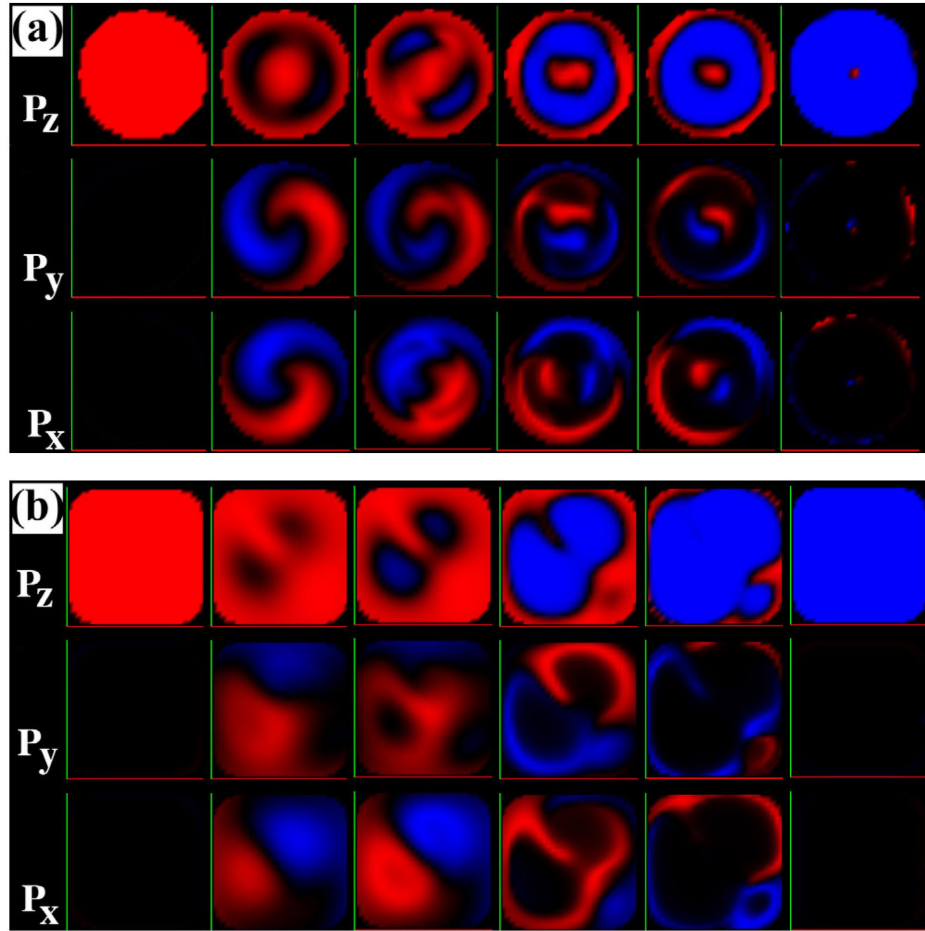


Figure 2. Simulation of domain switching kinetics in: (a) 1-μm-diameter circular capacitor and (b) 2.5 μm edge square capacitor. The top trace shows the time evolution of out-of-plane polarization component P_z ; lower traces illustrate time evolution of in-plane polarization components P_x and P_y , which cannot be measured with the present PFM technique. Red corresponds to positive polarization directions: P_z —out-of-plane, P_y —upward, P_x —to the right; blue indicates negative directions.

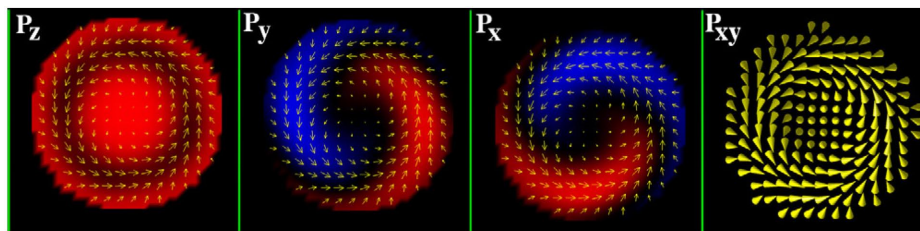


Figure 3. Two-dimensional maps of polarization components P_z , P_y , and P_x and in-plane polarization P_{xy} corresponding to an instantaneous domain pattern in the 1-μm-diameter circular capacitor during switching; the in-plane polarization map P_{xy} illustrates a vortex structure similar to the one in [9].

- (3) All the nucleation occurs near the circular capacitor edge, but none occur at the edge.
- (4) The switching occurs in approximately 100 time steps and the doughnut-type center remains for approximately 5×10^6 time steps and then it vanishes. Our time steps thus each represent ~ 200 ns in the experiments.

One obvious limitation of the present PFM experiments and modeling should be mentioned here. In the LLG simu-

lation, we watch the actual dynamical switching process in detail, while the domain walls are moving. This process involves not only a simple Ising-like up and down flipping of polarization but also a development of the in-plane component of polarization, which arises prior to the complete polarization reversal. However, it is difficult to verify this via direct PFM experiment for at least two reasons. First, the actual domain switching measurements using stroboscopic PFM are quasistatic. The PZT capacitors are par-

tially switched and allowed to remain in this state for a time required for PFM domain image acquisition, which is much longer than the switching time. On the other hand, the vortex-type domains represent non-steady polarization state with the in-plane components appearing only during dynamic switching. Second, during PFM measurements through the top electrodes, the lateral PFM signal is inevitably affected by the contribution of d_{31} and d_{32} constants of the piezoelectric tensor of tetragonal PZT, which makes unambiguous detection of the in-plane polarization difficult [17].

The line of inference is that, since the numerical simulation very closely replicates experimental evolution of the P_z polarization as a function of time, the evolution of polarization components P_x and P_y obtained in the numerical simulation should also adequately reflect the actual kinetics of the in-plane polarization during switching. In Figure 3, a two-dimensional map of the in-plane polarization distribution P_{xy} , obtained by combining the simulated instantaneous maps of P_x and P_y components, shows the vortex structure consistent with the pattern modeled by Naumov and Fu [7] and with recent results by Junquera [18].

A simpler but analogous description for the domain patterns observed in Figures 1 and 2 can be done by means of Bessel functions, which are used for description of the propagation of electric currents in cylindrical waveguides [19–21]. Let us consider the circular PZT capacitors to be cylindrical waveguides. In this case, the lower TEM modes are well known and they look like the patterns we see. So we might treat the domain wall nucleation and growth in PZT capacitors as occurring at the high-field regions of a circular waveguide of radius B . The doughnut-shape domain can be described by an internal radius a and an external radius b , with $b < B$. One finds that the impedance Z of the transmission “line” is the ratio of voltage between the inside and outside to the axial current flowing normal to the disc. In our case the axial current J is not ohmic but instead a displacement current, so borrowing the waveguide results [22] we can write

$$dP/dt = J = (2n/c)V(\mu/\epsilon)^{1/2} \ln(b/a) \quad (2)$$

where ϵ is the dc current $\epsilon(0)$ and not $\epsilon(\infty) = n^2$. Although this approach is only qualitative (it yields only order-of-magnitude agreement with the measured values of dP/dt), the idea of replacing real current $J(t)$ with displacement current $i(t) = dP/dt$ in ferroelectric nanorods during switching merits further consideration.

Figure 2 shows both radial and bending mode Bessel function distribution of polarization. To understand the dynamic response of P_x and P_y components, one must Fourier-transform the spatially uniform applied field $E(t)$ to $E(f)$. A pulsed field has two main Fourier components: f_1 at the reciprocal of the rise time and f_2 at the reciprocal of the pulse width. Using the known values of Young’s modulus Y , density ρ , and Poisson’s ratio ν , the lowest radial frequency for a thin PZT disc of thickness $d = 50$ nm can be estimated as

$$f_1 = (2.09/2\pi B)(Y/(\rho(1 - \nu^2)))^{1/2} = 218 \text{ MHz}$$

and the next-lowest bending mode as

$$f_2 = (38.5d/2\pi B^2)(Y/(12\rho(1 - \nu^2)))^{1/2} = 13.2 \text{ MHz.}$$

Hence, applied voltage pulses with a $\tau_r = 5$ ns rise time will resonate strongly with the radial Bessel function at approximately 200 MHz and the 100 ns wide pulses will resonate strongly with the bending mode. Hence, in our experiment, both bending and radial modes are resonantly driven, suggesting a possibility of moving 90° domains. This also explains why such vortex domains were never observed with slow rise time or sinusoidal voltages. It is worthwhile emphasizing that similar vortex domain structures in ferroelectric cylinders were recently predicted in Luk’yanchuk’s theoretical paper [23], which shows analytically that the stable domain configuration in a thin PZT disc under an applied field is a second-order Bessel function appearing with an unswitched edge and a “hole” in the middle.

In summary, we report observation of the instantaneous P_z domain configurations during switching in micron-sized ferroelectric capacitors on a 100 ns timescale. Formation of the doughnut-shape P_z domain pattern with an unswitched circular domain at its center is attributed to a vortex domain structure developing during dynamic switching. This conclusion is made based on numerical switching simulations using the Landau–Lifshitz–Gilbert equations which agree very closely with experimental data for both circular and square-shaped capacitors. The simulations predict a significant in-plane polarization during switching in circular capacitors, which agrees with the model of Naumov and Fu [7] but is beyond present experimental detection.

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