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Blowing polar skyrmion bubbles in oxide superlattices

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ABSTRACT

Particle-like topological structures such as skyrmions and vortices have garnered ever-increasing interests due to their rich physical insights and potential broad applications in spintronics. Here we discover the reversible switching between polar skyrmion bubbles and ordered vortex arrays in ferroelectric superlattices under an electric field, reminiscent of the Plateau-Raleigh instability in fluid mechanics. An electric field phase diagram is constructed, showing a wide stability window for the observed polar skyrmions. A "volcano"-like pontryagin density distribution is formed, indicating the formation of a smooth circular skyrmion. The topological charge Q at different applied field is calculated, verifying the field-driven topological transition between Q = 0 and Q = ±1 states. This study is a demonstration for the computational design of field-induced topological phase transitions, giving promise for the design of next-generation nanoelectronic devices.

1. Introduction

A plethora of particle-like topological defects such as skyrmions with whirl-like non-trivial topological structures [1–5] and vortices with continuous rotating order parameter vectors [6–8] have been widely investigated in the ferromagnetic systems for decades. It is now well accepted that the antisymmetric chiral interactions such as Dzyaloshinskii-Moriya (DM) interaction [9,10] in the non-centrosymmetric ferromagnetic systems could give rise to the rotating spontaneous magnetization, benefiting from the spin-orbit coupling. Topological polar structures on the other hand are considered rare in the ferroelectric systems because such chiral interactions are absent since the fundamental origin of the ferroelectrics is different from the ferromagnetics [11,12].

The recent progresses in computational tools have enabled the predictive modeling of mesoscale polar topological transitions. One particular example is the state-of-art phase-field simulation [13], which is not only capable of simulating mesoscale microstructure evolution, but also allows predictions that can be validated by the experiments. For instance, a recent phase-field study has predicted the whole range of periodicity phase diagram for $(PbTiO_3)_n/(SrTiO_3)_n$ (PTO/STO in short) superlattice on a $DyScO_3$ substrate and in particular polar vortex lattice at intermediate periodicities due to the complex intimate energy competitions, which is beautifully confirmed by experimental observations [14,15].

The exciting discovery of polar vortex lattice has brought one key question as how to manipulate the ferroelectric topological structures via external stimuli. To date, the vortex switching and annihilation dynamics has been extensively investigated both experimentally and theoretically (e.g., micromagnetic simulations [16], phase-field simulations [17]) in ferromagnetics [18,19] and superconductors [20]. Some intriguing phenomena have been discovered, for example vortex-antivortex annihilation in ferromagnetics could not only reverse the polarity of the vortex core but also induce a burst of spin waves [18]. In the ferroelectric systems, the switching of the curl of the polar vortex has been studied in low dimensional ferroelectrics [21–23]. Here we predict the field-driven topological phase transitions between polar vortices and skyrmions.

2. Phase-field model

The switching of a polar vortex lattice under an electric field is studied via phase-field simulations. We employ the spontaneous polarization vector $\vec{P}(r) = (P_1(r), P_2(r), P_3(r))$ to describe a polar state, and the relative stability of a polar state is determined by its total free energy, F, which is a functional of $\vec{P}(r)$. The total free energy contains contributions from the chemical, polar exchange, elastic, and electrostatic energies under a constant temperature and an applied electric field, i.e.

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$$F(\overrightarrow{P}(r)) = \int \left[f_{chem}(\overrightarrow{P}(r)) + f_{grad}(\nabla\overrightarrow{P}(r)) + f_{elas}(\overrightarrow{P}(r)) + f_{elec}(\overrightarrow{P}(r))\right] dV \quad (1)$$

To obtain the stable polar state under a given temperature and electric field as well as the transition or switching from one polar state to another as an external electrical field is applied or the magnitude of the applied electric field is changed, we employ the time-dependent Ginzburg Landau (TDGL) equation,

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

where t is the evolution time and L is the “kinetic coefficient” for the polarization evolution.

More details on the calculations of different energy contributions to the total free energy as well as the numerical solutions to Equation (2) have been provided previously [24–26]. The model system chosen here is the $(PTO)_{16}/(STO)_{16}$ superlattice assumed to be fully coherent on a DSO $(110)_o$ substrate (the lattice constants to determine the effective substrate strain have been given in Ref. [15]). All the simulation parameters are adopted from the existing literature [15,24–28]. A thin film mechanical boundary condition is used, i.e. the top surface of the superlattice film is stress-free while in the substrate region far away from the film/substrate interface there is no elastic displacements [24]. In particular, the iterative perturbation method is employed to solve elasticity equations with different elastic constants for the PTO and STO layers [29]. The electric potential at the bottom of the superlattice is set to zero, and the top the applied voltage bias. A three-dimensional simulation system with a mesh size of $200 \times 200 \times 250$ is used with a grid spacing of 0.4 nm. Along the out-of-plane direction, the thickness of substrate, film and air are set as 30, 198 and 22 grids, respectively. Periodic boundary conditions are applied along the in-plane directions while a superposition method is used in the out-of-plane direction [30]. The simulations start from small random noise which resembles the thermal fluctuations during the film growth and quench process.

3. Initial setup

The equilibrium polar vortex lattice is constructed for a $(PTO)_{16}/(STO)_{16}$ superlattice. As shown in Fig. 1(a), the polarization vectors rotate continuously inside the PTO layers, which agrees well with the structure from both theoretical calculations and experiment observations [14,15]. The neighboring vortices show the opposite vorticity (indicated by the blue and red regions, as calculated by $\nabla \times \overrightarrow{P}$), representing the clockwise-counter clockwise geometric arrangement of the vortices. While the planar view image cutting from the top of the PTO layer indicates the formation of long vortex lines (Fig. 1b), with one vortex line much thicker than the neighboring one with opposite in-plane polarization directions. The distance of the vortex cores in neighboring vortices is roughly 5–6 nm that scales with the PTO spacing of 16 unit cells [14,15and24]. The three-Dimensional (3D) structure of the polarization inside one PTO layer is further plotted in Fig. 1(c), where the long vortex tubes form perpendicular to the vortex plane. Meanwhile, this structure is highly asymmetric, forming an up-down zigzag configuration [31]. The out-of-plane field is then applied through a top capacitor electrode. The profile of time-dependent applied bias is given in Fig. 1(d), which increases 1.3 V every 400 time steps until reaching a maximum of 13 V, then decreases at the same rate to −13 V, and eventually recovers to 0 V again to form a complete switching circle.

4. Simulation results

The cross-section view of the entire switching circle is shown in Fig. 2(a)–(h). With the application of a positive bias (negative field), two neighboring vortices with opposite curls tend to move closer to form a close-pair structure, while the neighboring pairs are moving against each other, (Fig. 2b). The lateral movement of the vortex cores could reduce the area with upward polarization, thus reducing the electrostatic energy of the whole simulation system. This switching process demonstrates that the polar vortex lattice exhibit “dipole spring”-like behavior [32]. Increasing the applied electric field will lead to the destruction of vortex arrays as soon as two vortex cores reach the same lateral position (Fig. 2c). This process produces new *a*-domains, with the decrease in vorticity due to the annihilation of positive and negative vorticity regions. Consequently, at even higher biases, the full destruction of polar vortex lattice gives rise to regular *a*/*c*-twin domains.

Upon the gradual removal of the applied field, the reversible back-switching takes place. The *a*/*c* domain wall first decomposes into a pair of two vortices with opposite polarization vorticities. The two vortices then move laterally against each other to reduce the net polarization as to decrease the huge depolarization field. When applied field is removed, the ordered vortex array structure is restored, and zigzag vortex lattice pattern appears again (Fig. 2d).

A negative bias is gradually applied then. It is observed that the lateral motion direction of vortices is reversed (Fig. 2e) as compared to Fig. 2(b) with a positive bias. This could be understood since the preferred polarization direction flips with the change of direction of the applied field. An even higher bias led to the destruction of the vortex lattice again, forming *a*/*c*-twin domain structure (Fig. 2f). The orientation of the 90° domain wall switches as compared to Fig. 2(c) with a positive potential. As the applied bias is further increased, the bottoms of *a* domains shrink to needle-like configurations before they are eventually switched to *c* domains. Consequently, the removal of the applied field leads to the full recovery of the vortex structure again. More details are shown in the supplementary movie S1.

Previously, it is demonstrated through integrated micromagnetic simulations and experimental observations that switching of ferromagnetic nano-domains with wall junctions could induce a ferromagnetic vortex structure, resulting in a dimensionality crossover from 2-D domain wall to 1-D vortex [33]. In the current simulation, we found a similar dimensionality crossover in the polar vortex system where 1-D vortices can be reversibly switched to 2-D domain walls.

The planar view vector plot is shown in Fig. 3(a)–(f), corresponding to the structure of top PTO layer in Fig. 2(a)–(f). The initial structure in Fig. 3(a) indicate the formation of long vortex lines. Then, with the increase of applied bias, the size of both vortex lines shrinks, (see Fig. 3b), which is due to the switching of the *a*-like regions to *c*-like regions under an out-of-plane field. Simultaneously, the two vortex lines move towards each other to further reduce the area with opposite polarization directions. With higher bias (Fig. 3c), the thinner vortex lines “melt”, forming “bubble”-like polar pattern. This is in analogy to the Rayleigh-Plateau instability in fluid mechanics, where the lines of waterfall break into small water droplets to minimize the surface area. Previously, Scott et al. have discovered the Richtmyer–Meshkov type instabilities in $Pb_5Ge_3O_{11}$, where the application of an external electric field leads to the emission of a bubble-like ferroelectric domains due to the curving of the domain walls [34,35]. It is proposed that for some ferroelectric materials undergoing non-equilibrium processes, the