Local Manipulation of Polar Skyrmions and Topological Phase Transitions

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Topological phases are a fertile playground emerging from within ferroelectric superlattices. Herein, using phase-field simulations, we demonstrate the local control of the skyrmion phase with applied electric field. Under a small electric field, the skyrmions inside the electrode can be erased and recovered reversibly. While a topological transition to labyrinth domains requires a high field, which can switch back to skyrmions with a relatively small electric field. It is shown that the shrinking and dissipation of the skyrmions leads to a large reduction in the dielectric permittivity with the magnitude of reduction depends on the electrode size.

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Complex topological patterns in ferroelectrics have garnered enormous interest recently, driven by the formation of a polar vortex [1-5], flux-closure domains [6, 7], polar spirals [8, 9], skyrmions [10-12] and merons [13], which has largely rejuvenated the understanding of the world of ferroelectrics and the associated theoretical frameworks [14]. These polar textures exhibit exotic physical phenomena such as negative capacitance [15-17], chirality [18-22], ultrafast light-induced supercrystal formation [23], and so on. In particular, the polar skyrmion, a nontrivial whirl-like structure with a topological charge of ±1, has been discovered in the PbTiO3/SrTiO3 (PTO/STO) superlattice system. It has triggered considerable attention for potential nanoelectronic device applications, such as in non-centrosymmetric ferromagnetic counterparts [24, 25]. For instance, previous studies have shown that the polar skyrmion in the PTO/STO system demonstrates topological protection under small electric stimuli, akin to those observed in the ferromagnetic systems [16]. Despite all these exciting discoveries, one intriguing question is yet to be answered: how can we locally move/manipulate the polar skyrmion? The dynamics of the skyrmion switching process might also induce transitions to other topological states of matter in a reversible fashion [11]. The reversible control of these transitions could give next level interest in technological applications such as ferroelectric racetrack memories or other polar skyrmion-based electronic devices.

In this letter, using phase-field simulations, we report the local electric erasure/recovery of polar skyrmions via an external electric field through a narrow top electrode contact. We discovered that under relatively small applied potential, the skyrmions can be fully erased and recovered which involves both skyrmion expansion and nucleation. When the applied potential is large, the skyrmions transform to labyrinthine domains near the electrode, which act as preferred nucleation sites at the recovery step. These labyrinthine domains can be erased with a smaller negative applied potential and form a stable skyrmion state again. The local dielectric properties are also investigated, showing a large reduction at the high field due to the erasure of the skyrmion state.

The details of the phase-field simulation method are given in the supplementary information (SI) section, following the protocols from previous studies [11, 26]. The simulation setup is described in Fig.1. As shown in Fig.1(a), PbTiO3/SrTiO3 (PTO/STO) superlattice is
periodically grown on top of a STO-(001) substrate, with a 3-dimensional mesh of 320×320×350, each grid representing 1 unit cell. A bottom electrode is introduced between the film and substrate, while a narrow electrode with designed width ($d_0= 8 \text{ nm}, 16 \text{ nm}, 32 \text{ nm}, 64 \text{ nm}, \text{ and } 128 \text{ nm}$) along the $X$-dimension is deposited on top of the film to generate a highly localized electric field. The top view shows the equilibrium polar skyrmion structure in the PTO layer (Fig.1b-1c) which is consistent with previous reports$^{[10,16]}$ and experimentally confirmed by scanning transmission electron microscopy (STEM, Fig. 1d-1e). Details of the sample growth and characterization process are given in SI. Fig.1(c) shows the magnified view of the in-plane vector mapping, which indicates the formation of a hedgehog-like skyrmion structure. The cross-section view of this sample is plotted in Fig. 1(e), showing the contrast of two different domains with straight 180° type perpendicular domain walls confirmed from both phase-field simulations and experimental observations. This demonstrates the 180° rotation of the polar vector through the skyrmion wall. An electric potential is then applied through the top electrode to the superlattice film, with the potential profile given in Fig.1(f).

Fig. 1 Initial setup and applied potential profile. (a) Schematics of the PTO/STO superlattice system. (b) Planar view of the in-plane polarization magnitude from phase-field, showing the formation of skyrmion bubbles. (c)
magnified view of the polar skyrmion bubble overlaid with the in-plane polar vector. (d) Experimental planar view from high angle annular dark field STEM, showing bubble like structure. (e) Cross-section view of the superlattice system from phase-field simulations (left) and experimental dark field TEM observation (right). (f) The applied voltage profile, the width of the electrode is $d_0$, while the magnitude of the voltage is $\varphi_0$.

In the first study, $+6 \text{ V}$ is applied through an 8 nm wide top electrode. The kinetic evolution pathway of the polar skyrmions is shown in **Fig.2 (a)-(e)**. The skyrmions inside the electrode region shrink first after $10^2$ timesteps (with each timestep in the order of tens of fs$^{[27]}$), and then gradually disappear. In the neighboring regions, the skyrmions are gradually pushed away from the electrode. They become asymmetric where it is darker near the electrode side and brighter away from the electrode and eventually, all the skyrmions inside the electrode region are erased.

The detailed evolution pathway is presented in the supplementary video **S1**. The polar structure of the asymmetric skyrmion in the vicinity of the electrode is highlighted in **Fig. 2(d)**, in which a large in-plane polarization is observed, pointing away from the planar electrode. Close to the electrode side, the in-plane polarization pointing towards the electrode becomes much smaller in magnitude. To understand this phenomenon, the electric distribution of the top PTO layer is plotted (**Fig. 2e and 2f**). It is shown that the electric field is predominantly pointing downwards inside the electrode region, which triggers the burst of the skyrmions to form a simple $c$' domain inside the electrode region. In the neighboring regions, a large in-plane electric field is observed due to the local electric potential transition from $+6 \text{ V}$ to $0 \text{ V}$, driving the asymmetric skyrmion transition and pushes them away from the electrode. Subsequently, the applied electric field is removed (**Fig. 2g-2i**). The asymmetric skyrmions first expand and relax to a more symmetric skyrmion, then the nucleation and growth of new skyrmions can be observed to fill in the regions inside the original electrode. After $10^4$ timesteps, the system reverts to a fully skyrmionic state. Thus, we have demonstrated the ability to locally erase and reset the skyrmion state, which is electrically controllable and reversible.
Fig. 2 Kinetics of the skyrmion switching and recovery under a small, applied potential of 6 V and narrow electrode of 8 nm. (a)-(c) The kinetic evolution pathway after $10^2$, $10^3$ and $10^4$ timesteps, respectively. The white dashed region is the electrode. (d) magnified view of the asymmetric skyrmion overlaid with the inplane polar vector. (e) 3-dimensional plot of the inplane electric field $E_x$ on top of the PTO layer. (f) Line plot of the local electric field and polarization distribution cut through the center of the electrode. The electric field is overwhelmingly downwards inside the electrode while large inplane components are observed away from the electrode, which drives the formation of inplane polarization components on the two sides of the electrode. (g)-(i) The relaxation kinetics after the field is removed, after $10^2$, $4\times10^2$ and $10^4$ timesteps, respectively. The arrows indicate the nucleation of new skyrmions.

On the other hand, when the applied bias is large (e.g., +9 V, Fig. 3), a skyrmion shrinking and dissipation process is also observed inside the electrode region, similar to the previous case (see
Fig. 2). Whereas in the neighboring regions, the skyrmions “melt” and merge to form long stripe domains along the Y direction. After $10^4$ timesteps, all the skyrmions inside the electrode regions are erased, while the neighboring regions are filled with long stripes (Fig. 3c). These stripe domains have very large unidirectional in-plane polarization pointing away from the electrode region (Fig. 3d). This can be rationalized since under +9 V, the in-plane field is much higher as compared to the +6 V case, which is sufficient to switch all the skyrmions to unidirectional in-plane polarization and connect them (Fig. 3e and 3f). Interestingly, when the applied potential is removed, the stripe domains are expanding towards the electrode region, forming labyrinthine domains to reduce the depolarization field in this system. Unlike the unidirectional stripes formed in the initial switching process (Fig.3c), these labyrinthine domains have alternating in-plane polarization components to minimize the depolarization field. Whereas new stripes are nucleated and grown inside the electrode region, from the vicinity of the old stripes to the center of the electrode region. The detailed switching and recovery process is further shown in supplementary videos S2. Previously, topological transitions from vortex to skyrmions have been widely reported, by thermal or electrical driving forces $^{[11,28,29]}$. Here in this study, we demonstrated an electric field-driven localized topological transition from skyrmions to labyrinthine domains, with both states stable at room temperature and zero electric field.
Fig. 3 Kinetics of the skyrmion switching and recovery under high potential of +9 V and wide electrode of 32 nm. (a)-(c) The kinetic evolution after 100, 400 and 10000 timesteps, respectively. The white dashed region is the electrode. (d) magnified view of the stripe region overlaid with the inplane polar vector. (e) 3D plot of the inplane electric field Ex on the top PTO layer. (f) comparison of the inplane field under +9 V and +6 V used in the last case. (g)-(i) The relaxation kinetics after the field is removed, after 100, 1000 and 10000 timesteps, respectively. The formation of stable long stripes is observed.

The cycling test is further performed (Fig. 4) to showcase the controllable reverse transition, starting from the state after poling by +9 V and relaxed to zero after $10^4$ timesteps (e.g., state at Fig. 3i). An opposite bias of -6 V is applied later (Fig. 4b), which melts the vortex and labyrinthine domain to skyrmion bubbles through a Rayleigh-Plateau mechanism (see supplementary video S3 for details), as has been reported previously [11]. After the field is
removed, these residual small bubbles expand to become normal skyrmions, while the nucleation of skyrmions in other regions is also observed (Fig. 4c). Whereas on the two sides nearby the electrode, the labyrinthine domains remain since the inplane field generated under -6V is not large enough to erase them. Then, in the second cycle, +9 V is applied again to erase the skyrmion bubbles (Fig. 4d) with purely c-domain inside the electrode region, which switch back to labyrinthine state again after the field is removed (Fig. 4e). An applied potential of -6 V is applied again to start the second cycle, leading to the topological transition between labyrinthine domains and skyrmions (Fig. 4f). And the skyrmion state is stable when the applied field is removed (Fig. 4g). Thus, we realized the localized reversible transition between skyrmions and labyrinthine domains through control of the magnitude of the applied voltage.

To further understand the properties accompanying the topological transitions, the dielectric properties underneath the electrode regions of the different states are further plotted in Fig.4(h). The initial dielectric constant of the skyrmion bubble is ~650, which agrees with the previous experimental measurements and theoretical calculations\cite{16}. It can be observed that with the increasing of the applied electric field, the dielectric permittivity decreases. This is largely due to the shrinking and switching of the skyrmions, which decreases the area with negative permittivity inside the PTO layers, consistent with the previous studies\cite{13, 14}. Notably, under the same applied voltage, generally the wider the electrode, the larger the dielectric permittivity. This can be understood since under the same nominal out-of-plane electric field, the remaining skyrmion density/area is much higher/larger for the case with a wider electrode than a narrower electrode, as evidenced by Fig. 4(i) and 4(j). This will contribute to extra negative capacitance regions that will ultimately increase the overall dielectric property. When the electrode is thin (e.g., 8 nm), with a large nominal applied field (i.e., +800 kV/cm), the dielectric constant underneath the electrode can be reduced to ~100, showing a large reduction of the dielectric property (over 80%) due to the erasure of the polar skyrmions with
large negative capacitance regions.

**Fig. 4 Cycling test and reversible transition between labyrinthine state and skyrmion.** (a)-(d) The kinetic evolution of the first cycle, after applying -6 V to erase the labyrinthine domains to form stable skyrmion when field is removed, while applying +9 V could erase the skyrmions and form labyrinthine state. (e)-(g) The second cycle from cortex to skyrmion after applying -6 V. (h) local dielectric permittivity as a function of nominal applied electric field through a narrow top electrode. (i) and (j) Spatially resolved local permittivity of the top PTO layer with 551 kV/cm electric field, 8 nm and 32 nm wide electrode, respectively. For wide electrode (32 nm), the residual skyrmions can be observed with negative permittivity, while the narrow electrode (8 nm), all the skyrmions have been erased.

In conclusion, we report the local reversible control of polar skyrmions through a top electrode in the PTO/STO superlattice system. Under the small applied potential with a narrow electrode geometry, the skyrmions inside the electrode can be locally erased while the skyrmions in the neighboring regions are repelled by the electrode, giving rise to asymmetric
shapes with in-plane polarization mainly pointing away from the electrode. The skyrmion state is recovered when the potential is removed. Meanwhile, with a large applied potential and a wide electrode, labyrinthine domains form on the two sides of the electrode, which remain stable even when the applied potential is removed. The labyrinthine states can be switched and transform back to the skyrmion state after applying a relatively smaller potential. Thus, we realized a reversible transition between skyrmion and labyrinthine states through controlling the distribution of the applied potential. The dielectric permittivity is further calculated, which shows a large reduction under a high field. Interestingly, generally, the wider the electrode, the larger the average dielectric response under the same applied potential. This can be attributed to the higher retention of the skyrmion regions with negative permittivity. We envision this work to spur further interests in the skyrmion physics in ferroelectric system as well as the potential applications towards device applications in memory, transistor, or dielectric devices.

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