Collective control of a vortex array in a ferroelectric ultrathin film

Bo Ruan; Pengcheng Xiong; Qingyuan Liu; Ye Ji; Shuai Yuan

Appl. Phys. Lett. 124, 202901 (2024)
https://doi.org/10.1063/5.0204261
Collective control of a vortex array in a ferroelectric ultrathin film

Cite as: Appl. Phys. Lett. 124, 202901 (2024); doi: 10.1063/5.0204261
Submitted: 21 February 2024 · Accepted: 8 May 2024 · Published Online: 15 May 2024

Bo Ruan,1 Pengcheng Xiong,2 Qingyuan Liu,1 Ye Ji,3,a) and Shuai Yuan1,a)

AFFILIATIONS
1 School of Mechanical and Electrical Engineering, Guilin University of Electronic Technology, 541004 Guilin, China
2 Guangxi Key Laboratory of Functional Information Materials and Intelligent Information Processing, School of Physics & Electronics, Nanning Normal University, 530001 Nanning, China
3 College Physics Teaching and Experiment Center, Shenzhen Technology University, 518118 Shenzhen, China

a) Authors to whom correspondence should be addressed: silentmlight@gmail.com; xuanye69@guet.edu.cn; and yuanshuai2022@gmail.com

ABSTRACT
Recently, the observation of ferroelectric vortex arrays has triggered the investigation of topological domain structures and their characteristics. Vortices are typical topological domain structures with chirality in nanoscale ferroelectric materials. The chirality of a single vortex in a nanodot can be easily manipulated, but the collective control of a vortex array is exceptionally difficult and has not yet been realized. This Letter proposes an effective scheme for the collective control of a vortex array and investigates it via phase-field simulations. The results indicate that the collective control of a vortex array with bidirectional switching can be realized by introducing a bending film with periodic large curvatures under alternative electric fields. Furthermore, a general rule for determining the electrically controllable chirality of ferroelectric vortices is proposed. This Letter demonstrates the feasibility of the collective control of vortex arrays and provides insights for developing ferroelectric nano electronic devices based on vortex arrays.

Polar topologies with vortical configuration in low-dimensional ferroelectrics have attracted widespread attention owing to their application potential in nanoelectronic devices.1,2 Recently, various topological domain structures have been theoretically predicted and experimentally observed, such as flux-closure domain,3–5 vortex,6,7 skyrmion,8,9 meron,10,11 and hopfion.12 These structures exhibit fruitful emergent characteristics and topological phase transition behaviors. In vortical domain structures, chirality is a typical topological order parameter, describing the swirling characteristic of dipolar textures. Although clockwise (CW) and counterclockwise (CCW) vortices have opposite chirality, they degenerate in energy. Owing to the size effect and depolarization field influence, nanoscale vortex structures are robust and difficult to control. However, compared to the single vortex case, realizing the collective control of chirality in a vortex array comprising alternating CW and CCW vortices is more challenging.

Ferroelectric vortices were first predicted in nanodisks and nanorods, based on the first-principles-based effective Hamiltonian calculations.13,14 Thereafter, ferroelectric vortices have been explored in various nanomaterials under different conditions.15–18 The observation of vortex arrays in the [PbTiO3/SrTiO3] superlattice via dark-field transmission electron microscopy is particularly notable.7 Subsequently, studies have been conducted on the topological phase transition and performance control of vortex arrays.19–25 Several schemes have been proposed for effectively controlling the chirality switching of a single vortex, among which the electrically controllable ones include the introduction of an eccentric void,26 sweeping tip biases,27 dislocation pinning,28 distinct misfit strain,29 geometrical design,30 material distribution,31 and crystal orientation,32 and the mechanically controllable ones include the introduction of a centric void,33 twist force,34 and compositional gradient.35 However, the above-mentioned current schemes are ineffective for manipulating vortex arrays. In fact, the collective control of a vortex array is considerably more difficult. A vortex array is a winding chain comprising alternating CW and CCW vortices, where adjacent vortices cannot be decoupled and possess strong interdependence, similar to entanglement. Therefore, realizing the simultaneous switching of adjacent vortices with opposite chirality under the same electric field is difficult and necessitates the consideration of more complex asymmetric factors.

A single-crystalline ferroelectric ultrathin film can undergo a ~180° folding and has exceptional flexibility and super-elasticity when...
its thickness reaches nanoscale. Specifically, multidomain ferroelectric systems can be manipulated to induce bending films with periodic large curvatures. Furthermore, rippling can be generated in in-plane films via strain relaxation through the stretching and shrinking of the pre-stretched elastomeric substrate that is adhering to the film. Inspired by this, this study introduces periodic deflection deformation into a ferroelectric film, causing the symmetry-breaking directions of the adjacent vortices being in opposite directions. Consequently, CW and CCW vortex switching simultaneously achieved under the same external electric field, making the collective control of the chirality of a vortex array feasible.

This study employs the phase-field method to investigate the feasibility of the collective control of the chirality of a vortex array in a ferroelectric ultrathin film. Owing to the presence of the significant strain gradient between the concave and convex areas in the rippled film, the flexoelectric effect is considered in the phase-field model. The Helmholtz free energy $F$ is chosen as the total free energy of the system with the order parameters: the time-dependent Ginzburg-Landau (TDGL) equation expressed as $\frac{\partial \phi}{\partial t} = -\nabla \Delta \phi / \alpha - \nabla \cdot (\phi \nabla \phi)$; the equilibrium Gauss equation expressed as $D_{ij} = e_{ij} E_i + P_i = 0$; and the equilibrium mechanical equation expressed as $\sigma_{ij} = e_{ijkl} (\epsilon_{ijkl} - \epsilon_{ijkl}^0) = 0$.

Considering the extrapolation effect, the boundary conditions for the polarization field are $\frac{\partial P_i}{\partial r} + \eta_i G_{ijkl} P_j = 0$. For the electrostatic field, the ideal open-circuit boundary condition is set as $D_{r1} = 0$, and the ideal short-circuit one is set as zero potential. For the mechanical field, the stress boundary condition is $\sigma_{ij} n_j = 0$. By iteratively solving the third sets of equations and boundary conditions, stable system states are obtained.

Since the vortex array is a topological structure within the cross section of the thin film, a two-dimensional phase-field model is employed to investigate epitaxial PbTiO$_3$ ultrathin films. All the simulations are conducted considering a film with thickness of $h = 6$ nm and ambient temperature of 300 K, and the periodic boundary conditions are employed in the $x$ direction. The transversal film length within one period is denoted by $L$, where $L = 24$ nm in Figs. 2 and 3 and $L = 96$ nm in Fig. 4. The misfit strain subjected to the in-plane arising of the film stemming from the lattice mismatch between the film and the underlying substrate is set as $\varepsilon_m = -1.5\%$. The linear screening factor $\theta$ ($\theta \in [0, 1]$) is introduced to describe the electric boundary conditions of the top and bottom surfaces, with $\theta = 0$ denoting open circuit and $\theta = 1$ denoting short circuit. To form a stabilized CW–CCW vortex array and weaken the external electric field, $\theta$ is set as 0.6. Figure 1 exhibits the schematic of the feasibility of the collective control of the chirality of a vortex array in a ultrathin film with periodic large curvatures. The figure also shows that film rippling can be generated via strain relaxation from the stretching and shrinking of a pre-stressed substrate that is adhering to the film. For simplicity, the bottom surface is assumed to satisfy the displacement boundary condition and the top surface is assumed to be traction free. To describe the ripple morphology of the film, the periodic wrinkle displacement at the bottom surface is set to be a sinusoidal function $y = 0.1 \sin \frac{\pi}{L} \cdot x$. In Fig. 3, the amplitude $A_0$ and wavelength $\lambda$ are set as 0.1 and 12 nm, respectively. Generally, the $\lambda/h$ ratio in the rippled film observed in the experiment is large, e.g., $\lambda/h = 80$, indicating the presence of far more than two vortices in one ripple. However, to well elucidate the switching process of the vortex array, $\lambda/h = 2$ is chosen in Fig. 3, signifying the presence of two vortices (a pair of CW and CCW vortices) in one ripple. In fact, the vortex array switching is still guaranteed for the cases of multiple vortices in one ripple ($\lambda/h > 2$). Figure 4 demonstrates the influence of various factors including $\lambda/h$, $A_0$, and $L$. 

![Figure 1](https://example.com/figure1.png)
To characterize the vortex chirality, the toroidization $G$ is employed:

$$G = V^{-1} \int_0^L \mathbf{r} \times \mathbf{P} \, dV.$$ 

The lateral periodic boundary condition signifies the presence of an even number of vortices in the film (i.e., CW and CCW always appear in pairs); thus, the total toroidization of the vortex array is always zero. Therefore, local toroidization instead of total toroidization should be employed for characterizing the evolution process of the bidirectional switching of a vortex array under an alternating electric field. This study calculates the local toroidization based on the specimen of the right 1/4 part of the bending film.

First, the evolution behaviors of a vortex array in a non-curvature thin film under an in-plane alternating electric field are analyzed. The loaded alternating electric field is segmented as follows: $E_x = 0$ when $t^* = 0$–100, 200–300, and 400–500, $E_x = 0.8$ MV/cm when $t^* = 100$–200, and $E_x = −0.8$ MV/cm when $t^* = 300$–400. Figure 2(a) presents a series of snapshots of the domain structure evolution, and Fig. 2(b) depicts the evolution curve of the local toroidization $G$ of the vortex within the dashed box in (a). In the initial state, for a stable state A with $t^* = 90$, the vortex array comprises four vortices, i.e., CCW−CW−CCW−CW, and the toroidization $G$ of the initial CW vortex within the dashed box is $−0.46$ e/Å. Upon the application of an electric field $E_x = 0.8$ MV/cm along the $x$-axis ($t^* = 100$–200), the cores of the CCW vortices in the vortex array are gradually pushed upward, while those of the CW vortices are pushed downward ($A \rightarrow B \rightarrow C \rightarrow D$), resulting in the gradual decrease in the absolute value of $G$. Consequently, owing to the polarization enhancement under an electric field, a phase transition occurs from a vortex array into a stable-state single domain. When $t^* \approx 135$, $G$ decreases to zero; thus, the stable state E at $t^* = 190$ becomes a single domain state. When the electric field is withdrawn ($t^* = 200$–300), the polarization undergoes a re-nucleation process ($F \rightarrow G \rightarrow H \rightarrow I$) that is similar to high-temperature annealing and then evolves back into a vortex array as the initial one (state J). In the dashed box in the figure, a CCW vortex gradually forms and turns stablizes at $t^* \approx 270$; consequently, $G$ of state J becomes $−0.46$ e/Å at $t^* = 290$. Thereafter, an electric field $E_x = −0.8$ MV/cm is loaded ($t^* = 300$–400). The cores of the CCW vortices in the vortex array are gradually pushed downward, while those of the CW vortices are pushed upward ($K \rightarrow L \rightarrow M$), causing $G$ to decrease to zero at $t^* \approx 335$. Similarly, the stable state N at $t^* = 390$ becomes a single domain state. When the electric field is withdrawn again ($t^* = 400$–500), after experiencing a re-nucleation process ($O \rightarrow P \rightarrow Q \rightarrow R$), the vortex array finally evolves back to the initial configuration, with $G$ of state S relaxing to $−0.46$ e/Å at $t^* = 490$. The energy of state E or N as a single domain is higher than that of state J or S as a vortex array, owing to the former being additionally affected by an external electric field. The evolution of the single domain back to the initial vortex array configuration after the removal of the electric field may stem from the incomplete formatting of the initial domain configuration by the relatively small electric field. In fact, when the electric field is sufficiently large for completely formatting the initial domain configuration, the evolution of the chirality of the vortex array from a single domain state is uncertain. The results indicate that the effective collective control of the chirality of a vortex array in a curvature-free ultrathin film cannot be deterministically achieved.

However, the collective reversal of the chirality of a vortex array becomes feasible when deflection deformation with a periodic sinusoidal curvature is introduced into the film. In this process, the applied alternating electric field is as the same as that in Fig. 2. Figure 3(a) exhibits the vortex switching snapshots, and Fig. 3(b) displays the evolution curve of the toroidization $G$ of the vortex within the dashed box in (a). Similar to earlier, the initial vortex array comprises four vortices, i.e., CCW−CW−CCW−CW, and the $G$ of the initial CW vortex within the dashed box is $−0.46$ e/Å at state A with $t^* = 90$. When an electric field $E_x = 0.8$ MV/cm is applied along the $x$-axis ($t^* = 100$–200), the cores of the CCW vortices are gradually pushed upward, while those of the CW vortices are pushed downward ($A \rightarrow B \rightarrow C$). As the vortex cores move to the right as a whole ($C \rightarrow D \rightarrow E \rightarrow F$), four vortex cores with chirality opposite to the previous ones appear near the surfaces. In the dashed box, a CW vortex is reversed to a
CCW vortex, and the value of $G$ sharply changes from negative to positive, signifying vortex chirality switching. In state G at $t^* = 190$, $G$ of the CCW vortex reaches 0.43 e/Å. Therefore, the polarization spiral direction reversal from state B to state G under the induction of film bending with a sinusoidal curvature is crucial for realizing the collective control of a vortex array. After the electric field is removed ($t^* = 300$–3000), the vortex cores in state G gradually return from the surfaces to the vicinity of the neutral layer of the bending film, finally relaxing to the stable state J (G → H → J). In state J at $t^* = 290$, $G$ of the relaxed CCW vortex is 0.46 e/Å. When the electric field is reversed ($t^* = 300$–4000), the cores of the CCW vortices in the vortex array are gradually pushed downward, while those of the CW vortices are pushed upward (K → L → M). Owing to the crucial phase transition from state M to state O, the collective switching of the polarization spiral direction in the vortex array is again achieved at $t^* ≈ 325$ (state O). In state P at $t^* = 390$, $G$ is $-0.43$ e/Å, indicating vortex switching within the dashed box from CCW to CW. When the electric field is withdrawn ($t^* = 400$–5000), the vortex cores in state P gradually return from the surfaces to the vicinity of the neutral layer of the bending film, finally relaxing to the stable state S (P → Q → R → S). In state S at $t^* = 490$, G of the relaxed CW vortex arrives to $-0.46$ e/Å. This demonstrates that the collective control of the bidirectional switching for a vortex array can be realized in films with periodic sinusoidal curvatures under alternating electric fields.

To explore the effectiveness of the proposed scheme, Fig. 4 investigates the influences of various factors, including screening factor $\theta$, wave amplitude $A_0$ (transversal displacement undulation), and wavelength $\lambda$ (longitudinal displacement undulation), on the feasibility of deterministic bidirectional switching of a vortex array in a bending film with sinusoidal ripple. The figure shows that the proposed scheme has more widespread feasibility than previously anticipated. In this section, the following default conditions are assumed: the wavelength $\lambda$ of the ripple is 24 nm, the amplitude $A_0$ is 0.1 nm, and the screening factor $\theta$ is 0.6. When analyzing the influence of a factor, default values are assumed for the other factors.

The switching processes of vortex arrays in thin films under different wavelengths, amplitudes, and screening factors are depicted in Figs. 4(a)–4(c). The wavelength $\lambda$ as the transversal displacement undulation is a significant parameter in the vortex array switching process. Figures 4(a-i)–4(a-iv) display the snapshots of the domain structure evolution during the bidirectional switching at wavelengths of 24, 32, 48, and 96 nm. Although multiple vortices (more than two) appear in one ripple, the chirality switching of the vortex array is still guaranteed. As the longitudinal displacement undulation, amplitude $A_0$ is another important parameter influencing vortex array switching. Figures 4(b-i)–4(b-iv) exhibit the snapshots of the vortex array switching under sinusoidal deflection with amplitudes of 0.05, 0.2, 0.3, and 0.4 nm. The figure denotes that even a small wave amplitude can induce the collective switching behavior of a vortex array. Moreover, as a crucial vortex array generation parameter, the screening factor $\theta$ plays an important role in chirality switching behaviors. Figures 4(c-i)–4(c-iv) display the snapshots of the collective chirality switching of vortex arrays at screening factors of 0.0, 0.8, 0.85, and 1. When $\theta < 0.8$, the vortex morphology is hardly unaffected and vortex array switching is realized. When $\theta > 0.85$, the number of vortices in the vortex array sharply decreases, but the collective switching is still feasible. When $\theta = 1$ (short circuit), although an Ising-type $180^\circ$ domain wall forms instead of a vortex, polarization switching is still feasible. In other words, the influence of the linear screening factor is nonlinear, which is consistent with previous studies.46,47 The study results show that the proposed scheme for vortex array switching has universal feasibility and may provide inspiration for future experiments.

Figures 3 and 4 imply that the opposite curvatures at the concave and convex areas generated from sinusoidal deflection in an ultrathin film are essential for the simultaneous reversal of adjacent vortex pairs in a vortex array. Figure 5(a) summarizes the deterministic effect of the bending curvature and external electric field on the vortex chirality. Specifically, under an external electric field along the x-axis (−x-axis), a CW (CCW) vortex is favored for convex bending and a CCW (CW) vortex is favored for concave bending. Figure 5(b) proposes a general
right-hand spiral rule for electrically controllable vortex chirality. This rule is as follows: assuming that the unit vector in the symmetry-breaking direction is $\mathbf{f}$ and that in the external electric field loading direction is $\mathbf{e}$, the unit vector $\mathbf{g}$ in the direction of chirality of a favorable vortex can be obtained by the cross product of $\mathbf{e}$ and $\mathbf{f}$: $\mathbf{g} = \mathbf{e} \times \mathbf{f}$. The angle $\theta$ between $\mathbf{f}$ and $\mathbf{e}$ ranges from 0 to $\pi$, i.e., $\theta \in [0, \pi]$. Specifically, the symmetry-breaking directions of the concave and convex bending in the film are exactly opposite and point toward their respective curvature centers. Furthermore, previous studies on vortex switching via external electric fields also obey this rule,$^{26,28,35}$ where the key to feasible control is the construction of a symmetry-breaking direction. According to this rule, vortices with certain chirality can be written and erased as needed.
This study investigated the feasibility of the collective control of the chirality of a vortex array in a ferroelectric ultrathin film using phase-field simulations. By introducing deflection deformation with a periodic large curvature into a film, a periodic symmetry-breaking feature was constructed, which realized the simultaneous switching of adjacent vortices under an alternating electric field. This ultimately enabled the electrical control of the chirality of a vortex array. Furthermore, a general right-hand rule was proposed for determining electrically controllable chirality of ferroelectric vortices. The results provide a feasible scenario for the collective control of chirality of a ferroelectric vortex array and may be conducive for facilitating the development of high-density nonvolatile random-access memories based on ferroelectric vortex arrays.

S.Y. acknowledges the financial support from the National Natural Science Foundation of China (No. 12302430) and the Special Project of Central Government for Local Science and Technology Development of Guangxi Province (No. ZY23055043). Y. J. acknowledges the financial support from the National Natural Science Foundation of China (No. 12302430). Q. L. acknowledges the financial support from the National Natural Science Foundation of China (No. 12302430). Q. L. acknowledges the financial support from the National Natural Science Foundation of China (No. 12302430). Q. L. acknowledges the financial support from the National Natural Science Foundation of China (No. 12302430). Q. L. acknowledges the financial support from the National Natural Science Foundation of China (No. 12302430). Q. L. acknowledges the financial support from the National Natural Science Foundation of China (No. 12302430). Q. L. acknowledges the financial support from the National Natural Science Foundation of China (No. 12302430). Q. L. acknowledges the financial support from the National Natural Science Foundation of China (No. 12302430). Q. L. acknowledges the financial support from the National Natural Science Foundation of China (No. 12302430). Q. L. acknowledges the financial support from the National Natural Science Foundation of China (No. 12302430). Q. L. acknowledges the financial support from the National Natural Science Foundation of China (No. 12302430). Q. L. acknowledges the financial support from the National Natural Science Foundation of China (No. 12302430). Q. L. acknowledges the financial support from the National Natural Science Foundation of China (No. 12302430). Q. L. acknowledges the financial support from the National Natural Science Foundation of China (No. 12302430). Q. L. acknowledges the financial support from the National Natural Science Foundation of China (No. 12302430). Q. L. acknowledges the financial support from the National Natural Science Foundation of China (No. 12302430). Q. L. acknowledges the financial support from the National Natural Science Foundation of China (No. 12302430). Q. L. acknowledges the financial support from the National Natural Science Foundation of China (No. 12302430). Q. L. acknowledges the financial support from the National Natural Science Foundation of China (No. 12302430). Q. L. acknowledges the financial support from the National Natural Science Foundation of China (No. 12302430). Q. L. acknowledges the financial support from the National Natural Science Foundation of China (No. 12302430). Q. L. acknowledges the financial support from the National Natural Science Foundation of China (No. 12302430). Q. L. acknowledges the financial support from the National Natural Science Foundation of China (No. 12302430). Q. L. acknowledges the financial support from the National Natural Science Foundation of China (No. 12302430). Q. L. acknowledges the financial support from the National Natural Science Foundation of China (No. 12302430). Q. L. acknowledges the financial support from the National Natural Science Foundation of China (No. 12302430). Q. L. acknowledges the financial support from the National Natural Science Foundation of China (No. 12302430). Q. L. acknowledges the financial support from the National Natural Science Foundation of China (No. 12302430). Q. L. acknowledges the financial support from the National Natural Science Foundation of China (No. 12302430). Q. L. acknowledges the financial support from the National Natural Science Foundation of China (No. 12302430). Q. L. acknowledges the financial support from the National Natural Science Foundation of China (No. 12302430). Q. L. acknowledges the financial support from the National Natural Science Foundation of China (No. 12302430). Q. L. acknowledges the financial support from the National Natural Science Foundation of China (No. 12302430). Q. L. acknowledges the financial support from the National Natural Science Foundation of China (No. 12302430). Q. L. acknowledges the financial support from the National Natural Science Foundation of China (No. 12302430). Q. L. acknowledges the financial support from the National Natural Science Foundation of China (No. 12302430).

REFERENCES


FIG. 5. Determination of vortex chirality. (a) Influence of film curvature and electric field direction on the formation of a vortex. (b) A general right-hand rule of determining electrically controllable chirality of ferroelectric vortices.