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Flicker vortex structures in multiferroic materials

Z. Zhao,1,2 X. Ding,2 and E. K. H. Salje1,a)
1Department of Earth Sciences, University of Cambridge, Cambridge CB2 3EQ, United Kingdom
2State Key Laboratory for Mechanical Behavior of Materials, Xi’an Jiaotong University, Xi’an 710049, People’s Republic of China

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Computer simulation of ferroelastic materials reveals dynamic polar vortex structures related to flexo-electricity between cation and anion lattices. At finite temperatures, the vortices are found to flicker in time and space. Widely spaced ferroelastic twin boundaries nucleate vortices while dense twin boundaries suppress them. The time averaged number of vortices at any site decays exponentially, indicating the highly mobile dynamics of the vortex lattice. Applied electric fields break the rotational symmetry of vortices and finally destroy them. The total number density of vortices follows a field and temperature dependence as \( N(E) = N_0 / [1 + A \exp(E / k(T - T_{\text{VF}}))] \) with \( T_{\text{VF}} < 0 \). The observed vortex structures are akin to those observed in magnetic and superconducting disordered vortex lattices. © 2014 AIP Publishing LLC.

Evans et al.1 investigated the magnetic switching of ferroelectric domains in multiferroic PZTFT at room temperature and questioned the coupling mechanism between electric and magnetic degrees of freedom. They enumerated several aspects of their experimental observations, which are suggestive of a strain-mediated coupling: first, the magnetic fields cause changes in the ferroelastic, and not simply the ferroelectric, domain components of the microstructure; second, some amount of reversibility in domain reorientation has been observed while the magnetic fields often cause sporadic and unpredicted changes in domain states, which are commensurate with sudden releases of elastic energy; third, the order of magnitude of the coupling coefficient is commensurate with sudden releases of elastic energy; fourth, some amount of reversibility in domain reorientation has been observed while the magnetic fields often cause sporadic and unpredicted changes in domain states, which are commensurate with sudden releases of elastic energy; fifth, the interatomic interaction is not simply the direct electrostatic interaction between charged sublattices; sixth, the interatomic interaction is not simply the direct electrostatic interaction between charged sublattices; seventh, the interatomic interaction is not simply the direct electrostatic interaction between charged sublattices; eighth, the correlation between the magnetic and ferroelectric components of the microstructure is not simply the direct electrostatic interaction between charged sublattices.

The generic model consists of two sublattices with positive and negative charges (Fig. 1). The atoms of the ferroelectric sublattice have negative elementary charges \((-1.602 \times 10^{-19} \text{C})\) while the second sublattice has positive elementary charges. Both sublattices have identical numbers of atoms to ensure electric neutrality. Long range Coulomb interactions and short range interatomic potentials govern the interaction between the two sublattices. We calculate the Coulomb interactions in the Ewald construction with a dielectric constant of 1000, which is typical for the incipient ferroelectric materials. Short range interatomic potentials are designed for the two sublattices separately, and they mimic the chemical bonds. The interactions of ferroelastic sublattice consist of three components: (1) harmonic nearest neighbor interactions, \( U(r) = 20(r - 1)^2 \), (black springs in Fig. 1), (2) double well potentials for next

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FIG. 1. Model with two charged sublattices. Coulomb interactions and interatomic interactions are combined in the model. The interatomic interactions between the nearest-neighbors are harmonic (shown by springs). Non-convex interactions (gray sticks) along diagonals in the ferroelastic sublattice lead to the formation of twin structures.

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a)Author to whom correspondence should be addressed. Electronic mail: ekhard@esc.cam.ac.uk
The macroscopic spontaneous strain is determined by the macroscopic shear angle of the bulk after annealing. It decreases with increasing temperature and shows a ferroelastic phase transition at $T_C = 100\,\text{K}$ (Fig. 2). The ferroelastic twin walls remain at their initial positions after annealing at low temperatures. Spontaneous nucleation of additional twin boundaries occurs near the transition point. During heating, these additional twin boundaries appear at $0.9\,T_C$ (Fig. 3); the microstructure then evolves into cross-hatch tweed structures\textsuperscript{14} at temperatures above $T_C$.

Dipoles form spontaneously and organise themselves on a mesoscopic scale. The most common pattern involves vortices, which are recognised by the collective rotation of dipoles on a scale of some 1 nm. In the middle of a vortex is a core with a diameter of 5 lattice constants. The vortex strength is calculated as the integral of the absolute dipole displacement inside the core. Vortices do not form static lattices in our simulations but are highly mobile (Fig. 3(a)) and "flicker" in the same way as tweed nanostructures flicker in ferroelastic structures.\textsuperscript{14} The total number of vortices also fluctuates with time; its number density is Gaussian.
distributed. The system contains very few vortices at very low temperatures where thermal excitation is weak (Fig. 3(b)). With increasing temperature, the number of vortices increases dramatically and reaches saturation. A singularity was found in the phase transition temperature interval (Fig. 3(b)). Widely spaced twin boundaries nucleate vortices while high dense twin boundaries suppress vortices. The averaged vortex strength is dominated by thermal excitation but is enhanced in the transition temperature interval when the dipoles inside the twin boundaries contribute to vortices (Fig. 3(b)).

The number of vortices decays exponentially with time at any site (Fig. 4(a)) when the dipoles are time-averaged because vortices nucleate, move, and decay. The decay exponent is constant at low temperatures and reaches a minimum near the transition temperature because the polarity in the additional twin boundaries increase thermal disorder and hence increase the flicker of the vortices (Fig. 4(b)). The number of vortices decays exponentially when the dipole movements are averaged over time. The decay follows $N \sim \exp(-t/\theta_0)$. Data at $0.3^\circ T_C$ (black triangles), $0.9^\circ T_C$ (dark blue circles), $0.96^\circ T_C$ (green triangles), $0.99^\circ T_C$ (purple squares), $1.1^\circ T_C$ (orange diamonds), $1.2^\circ T_C$ (light blue circles) and $1.5^\circ T_C$ (red squares) are shown in the graph. (b) The decay parameter $1/\theta_0$ is constant at low temperatures and reaches a minimum in the transition temperature interval. The number of vortices is almost unchanged at temperatures above $T_C$ where dynamical tweed structures are formed.

The field dependence of the vortex structure was measured using electric fields perpendicular to the twin walls (Y direction in Fig. 3(a)). The fields break the rotational symmetry of vortices and finally destroy them. The number of vortices decays with increasing electric field strength $E$ (shown in Fig. 5). This decay follows $N = N_0/[1 + A\exp(E/E_0)]$, where $N_0$ is the number of vortices without electric field and $E_0$ is proportional to $T - T_{VF}$ with $T_{VF} = -0.45^\circ T_C$. This leads to an effective incipient Vogel-Fulcher behaviour where the energy $E_0$ reaches zero at negative temperatures. Such behavior is not unusual for the transition of incipient ferroelectrics such as SrTiO$_3$ where the extrapolated ferroelectric transition occurs at negative temperatures and where the glass freezing temperature is also negative.\(^{15}\)

The simulated vortex structures explain avalanche behaviour (jerks) by jamming\(^{12}\) and show that strain induced coupling with polar properties is rather straightforward for twinned microstructures. The jerks are the same as simulated already for the ferroelastic lattice under strain deformation. In addition, the rapid movement and the field induced destruction of vortices also couple to the strain variable and can be observed by acoustic emission and dielectric measurements. Skyrmion lattices with magnetic vortex structures\(^{16}\) are similar to our polar vortex structures so that coupling between dipolar vortices and magnetic vortices becomes possible and may explain the origin of electric-magnetic coupling via a common strain. Superconducting vortex lattices follow a similar pattern with no long range order\(^{17}\) and also show jerky large scale oscillations under external fields.\(^{18}\) Direct observations of vortices in multiferroic materials are not uncommon (e.g., Y MnO$_3$ in Ref. 19) so that we believe that the gradient (flexo-) effects discussed in this paper are universal and a rather common phenomenon in multiferroic materials while their direct observation still remains elusive.

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FIG. 4. (a) The number of vortices decays exponentially when the dipole movements are averaged over time. The decay follows $N \sim \exp(-t/\theta_0)$. Data at $0.3^\circ T_C$ (black triangles), $0.9^\circ T_C$ (dark blue circles), $0.96^\circ T_C$ (green triangles), $0.99^\circ T_C$ (purple squares), $1.1^\circ T_C$ (orange diamonds), $1.2^\circ T_C$ (light blue circles) and $1.5^\circ T_C$ (red squares) are shown in the graph. (b) The decay parameter $1/\theta_0$ is constant at low temperatures and reaches a minimum in the transition temperature interval. The number of vortices is almost unchanged at temperatures above $T_C$ with $1/\theta_0 \approx 0$.

FIG. 5. The number of vortices decays with increasing electric field $E$. The decay follows Vogel-Fulcher type statistics $N_0/[1 + A\exp(E/E_0)]$ where $E_0 \sim T - T_{VF}$ shown in the inset graph. $N_0$ is the number of vortices without electric field and the extrapolated temperature $T_{VF} = -0.45^\circ T_C$.\(^{15}\)
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