doi:10.1017/S143192762100283X
Microsc. Microanal. 27 (Suppl 1), 2021

Probing the dynamics of ferroelectric topological oscillators with the electron beam

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Skyrmions and merons in ferroic materials are topological structures with real-space swirling arrangements of order parameters. While current-driven motion of spin textures in magnets have attracted interest as nanoscale information carriers [1,2], very little has been demonstrated with their electric dipolar counterparts, which were only recently stabilized in ferroelectric heterostructures [3,4]. In part, this is because their motion does not couple to uniform external fields as for the magnetic case, but instead couples to field gradients. As the polar textures are intrinsically nm-scale, developing imaging and control methods for exploring the stability and response of such particle-like objects to external stimuli at these length scales is of importance for the realization of logic devices.

Here, we demonstrate that topological dipolar textures can be created and annihilated by using the electric-field gradient of a localized current, here provided by the electron beam of scanning transmission electron microscope (STEM) in the lifted-off [5], freestanding membrane of (SrTiO3)16/(PbTiO3)16/(SrTiO3)16 trilayer. To characterize the ferroelectric polarization, 4D-STEM was employed by recording polarity-sensitive Friedel pairs at every probe position using an electron microscopy pixel array detector (EMPAD) [6,7]. Figure 1 shows the detailed in-plane Bloch components in mixed phases including polar skyrmions and merons, characterized by topological charges of +1 and +1/2, respectively. By a continuous raster acquisition using STEM annular dark field (ADF), we can perturb and simultaneously capture snapshots of self-assembling labyrinthine patterns, where certain consecutive frames show the creation and annihilation of convex and concave disclinations oscillating analogous to a pendulum in phase space, (Figure 1d). The electron-beam driven phase transitions were further explored by parking a STEM probe for ~0.5 second followed by an ADF-STEM snapshot, repeated at multiple sites. Figure 2 shows the occurrence of phase transition was most frequent at parked sites, which were exposed to the STEM probe ~105 longer than rest of the labyrinthine domains. Monte Carlo and molecular dynamics effective Hamiltonian simulations [8,9] suggest that the electric-field gradient generated from the STEM probe couples with the concave and convex disclinations, leading to the dipoles reorienting into a different metastable state. Theoretical insights into this highly degenerate, frustrated system suggest that the transition among various metastable states, even at very low temperature and under no perturbing fields, originates from the intrinsic topological instabilities of Pontryagin charge.

While here we demonstrated that the electric-field gradient of the STEM probe can serve as a sub-nm in situ “switch”, allowing us to study the local response to external stimuli, similar control could be obtained in a device using current fab technology with a ~5nm ballistic point contact electrode [10].

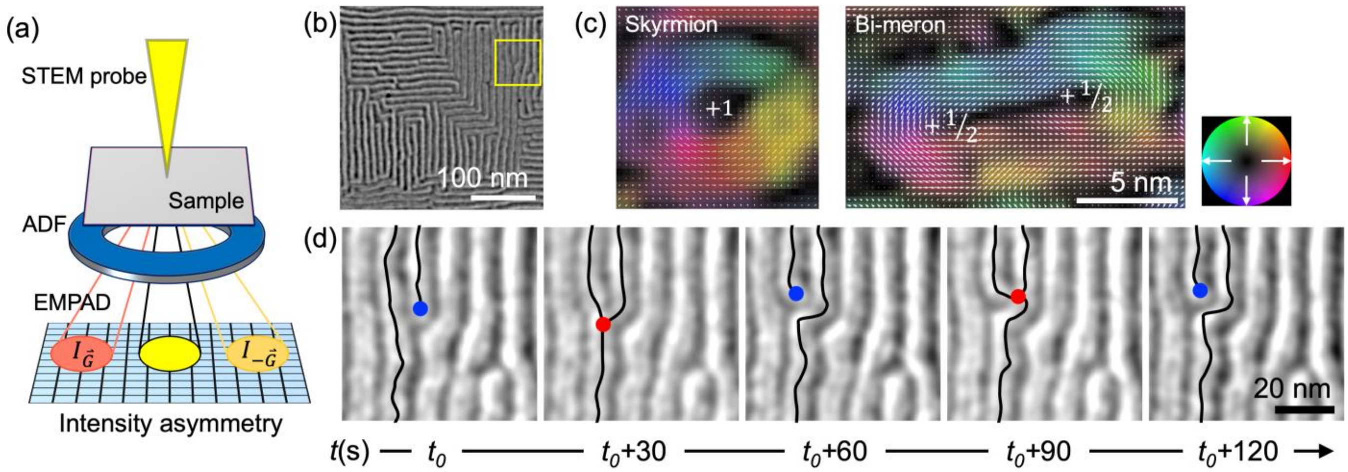

Figure 1. Perturbing a ferroelectric topological oscillator in the electron microscope. (a). Schematic of ADF-STEM and 4D-STEM using an EMPAD. (b). ADF-STEM image of the labyrinthine phase in the freestanding STO/PTO/STO trilayer. (c). In-plane Bloch components of the polar skyrmion (left) and bi-meron (right) imaged by 4D-STEM. (d). Continuous ADF-STEM raster acquisition showing the oscillating creation and annihilation of convex (blue) and concave (red) disclinations analogous to a pendulum. Blue and red dots denote the meron and antimeron with topological charges of +1/2 and -1/2, respectively.

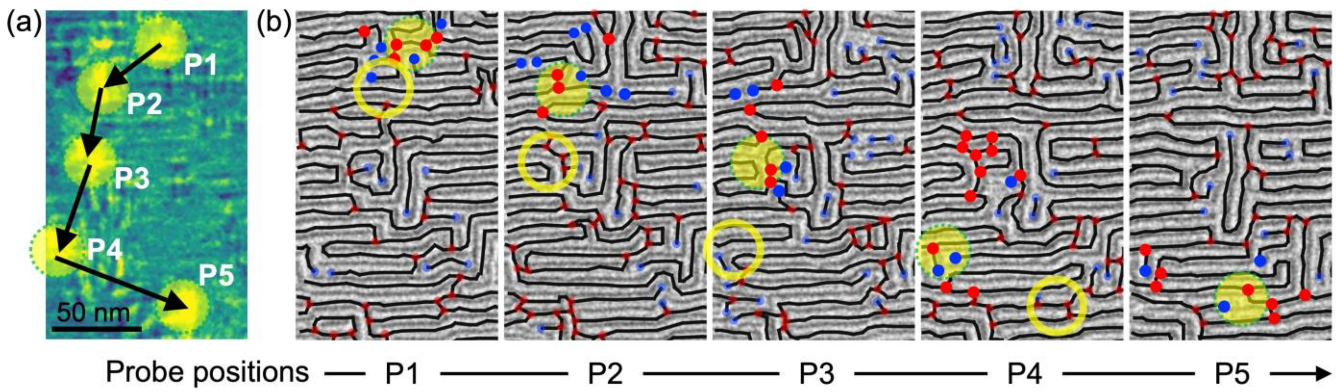

Figure 2. Electric-field gradient control of polar merons and antimerons. (a). Sum of difference in neighboring frames. The white arrows and yellow circles show the trajectories of successive electron beam parking. (b). ADF-STEM snapshots of the electron-beam driven reorientation of the labyrinthine phase. The convex (blue) and concave (red) disclinations near the beam parking positions were marked by filled circles, whereas the rest were marked by translucent circles. The filled and unfilled yellow circles indicate the current and previous beam parking position, respectively.

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[10] This work is supported by the AFOSR Hybrid Materials MURI, award # FA9550-18-1-0480. Facilities support from the National Science Foundation (DMR-1429155, DMR-1539918, DMR-1719875). Computations support from the DARPA Grant No. HR0011727183-D18AP00010 (TEE Program).