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In situ Electric Field Manipulation of Ferroelectric Vortices

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Topological defects in ferroic order offer both localized non-bulk properties and spatial control within a chemically homogeneous medium, making them attractive as dynamic, low-dimensional, functional elements such as high-density memory "bits." Ferroelectric vortices (Fig. 1), which form spontaneously within a certain size regime of $PbTiO_3/SrTiO_3$ superlattices [1], exhibit non-bulk properties including significant Néel rotational character [2], emergent chirality [3], and local negative capacitance [4]. The vortices measure ~5nm, making scanning/transmission electron microscopy S/TEM the principal experimental tool for extracting their local structure and properties [2,4]. In this work, we explore spatial control of these unique defects using *in situ* TEM. In the case of ferroelectrics, the electrical polar order is nominally controllable via electric fields, with reorientation of the polarization occurring in either a growth or a nucleation and growth mechanism. Vortices present several complicating factors underscoring the need for *in situ* study, namely large competing internal depolarization fields and a rotational symmetry, which imply that these defects cannot be externally stabilized using simple long-range (i.e., unidirectional) fields.

In situ characterization was performed using dark-field (DF) diffraction contrast TEM on a JEOL 3010 at 300kV. This method provides rapid acquisition (30ms frame time) and sensitivity to the polarization direction from dynamic scattering arising from the non-centrosymmetry of the crystal along the selected Bragg peak. For this work the $[00\bar{2}]_{pseudocubic}$ film normal Bragg peak was selected, providing bright/dark contrast corresponding to the projected polarization to the [001] axis (Fig. 1). Electric fields are applied via a freely positioned tungsten probe at the superlattice surface biased to a bottom electrode (Fig. 2A).

Two vortex variants are explored that correspond to different superlattice layer thicknesses. The first, 16 unit cells, exhibits a vortex structure as a very stable ground state. The response to an applied electric field, shown as a DF-TEM time series in Figure 2A, is an induced asymmetry of the vortices within the $PbTiO_3$ layer. In this case the vortex structures respond to the applied fields via short-range small domain wall translations without the need of nucleation events. At high fields, some regions transition from the vortex structure into a/c or in-plane a-domains. The latter is the case depicted for the sub-region shown in Figure 2A at +15V. Despite a large field response, this system exhibits very little hysteresis (Fig 2B), depolarizing fields render the induced asymmetries metastable upon field removal. In the second variant, 8 unit cell superlattice layers, the vortices are close in energy to an alternative in-plane a_1/a_2 domain configuration. In this case, unlike the previous case, the induced in-plane transition can remain stable upon removal of the external field. This is likely to be the mechanism for similar hysteretical switching between these two configurations demonstrated in macroscale surface probe microscopy [5,6].

References:

[1] Z Hong et al., Nano Lett. **17** (2017), p. 2246.
[2] A Yadav et al., Nature **530** (2016) p. 198.
[3] P Shafer et al., PNAS (2018). DOI: 10.1073/pnas.1711652115.
[4] A Yadav et al., Nature **565** (2019), p. 468.
[5] A Damodaran et al., Nat. Mater. **16** (2017), p. 1003.
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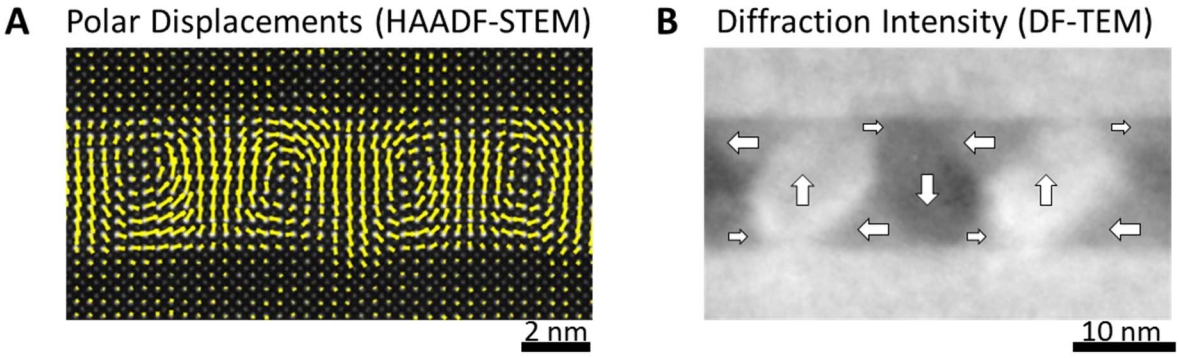

Figure 1. Vortex structure within ferroelectric $PbTiO_3$ layer. **A)** High angle annular dark field (HAADF)-STEM image from $PbTiO_3$(10u.c.)/$SrTiO_3$(10u.c.) superlattice depicting local cation sublattice noncentrosymmetry as with vectors. **B)** DF-TEM image formed from $[00\bar{2}]_{pc}$ reflection tilted to a strong diffraction condition from $PbTiO_3$(16u.c.)/$SrTiO_3$(16u.c.) superlattice. Polarization directions corresponding with image contrast are shown by arrow overlays.

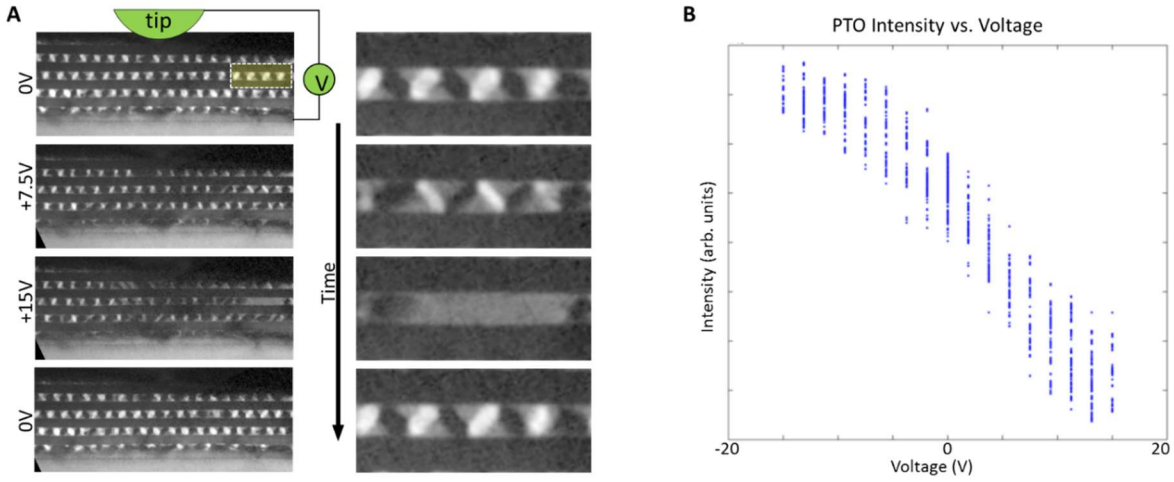

Figure 2. *In situ* electric field applied to $PbTiO_3$ (16u.c.)/$SrTiO_3$ (16u.c.) superlattice. **A)** Time/voltage DF-TEM series from initial ramp from 0V to +15V back to 0V with highlighted region expanded at right. **B)** $PbTiO_3$ layer diffraction net intensity vs. applied voltage.

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