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Size Effect on Spontaneous Flux-closure Domains in $BiFeO_3$ Thin Films

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In ferroic materials, polarization and magnetization vectors may rotate around a point and form flux-closure vortex domains. These vortex domains are intriguing in terms of both fundamental research and application interest due to the exotic polarization configuration and the switchability that could give rise to an unusually high density of bits for storage. Although the vortex domains are well-known to ferromagnetic materials, they have only recently been found in ferroelectric nanostructures and thin films. Here, we report direct observation of ferroelectric flux-closure vortex domains at the $BiFeO_3/TbScO_3$ hetero-interfaces using aberration-correction scanning transmission electron microscopy (STEM). We show that while in thicker films (~20 nm) the flux-closure quadrants are composed of shape-conserving nano-domains with regular 109 ° and 180 ° domain walls, the polarization vortex in thinner films (~5 nm) involves dipole rotations in a more continuous manner without formation of apparent nano-domains at the interfaces. These results indicate a size effect on the spontaneous flux-closure domains in ferroelectric thin films, revealing a method to tune the polarization vortex structures for practical devices.

$BiFeO_3$ is a room-temperature multiferroic exhibiting coupled ferroelectric (T_C ~ 1,103 K) and antiferromagnetic (T_N ~ 650 K) order. In its pseudocubic unit cells (Figure 1a), the oxygen octahedra and the central Fe cation are displaced from their respective positions at the face and body centers, giving rise to a large spontaneous polarization (~100 μC cm^{-2}) along the <111> directions. Figure 1b shows a high-angle annular dark-field (HAADF) STEM image of a $BiFeO_3$ thin film in [100] pseudocubic orientation, where the Bi columns appear as the brighter dots, the Fe columns show weaker contrast, and the oxygen atoms are not visible. In the HAADF image, we can define a vector, $\boldsymbol{D_{FB}}$, which is the atomic displacement in the image plane of the Fe cation from the center of the unit cell formed by its four Bi neighbors; and directly measure the $\boldsymbol{D_{FB}}$ vectors by fitting the atom columns as two-dimensional (2D) Gaussian peaks. This $\boldsymbol{D_{FB}}$ vector, as the dominant manifestation of the ferroelectric polarization in $BiFeO_3$, points toward the center of the negative oxygen charges, and thus is opposite to the polarization vector in the image plane. Using HAADF imaging, we imaged the spatial distribution of the $-\boldsymbol{D_{FB}}$ vectors of the $BiFeO_3/TbScO_3$ interface to determine the polarization distribution (Figure 2). The polarization maps show that, in both the thicker (20 nm) and thinner (5 nm) films, terminations of the 109 ° domain walls at $BiFeO_3/TbScO_3$ interface involve the formation of localized vortex domain structures. In the 20 nm $BiFeO_3$ film (Figure 2a), the vortex is accompanied by triangular domains consist of a mirrored pair of inclined 180 ° domain walls in conjunction with the previously existing 109 ° domain wall [1]. In the 5 nm $BiFeO_3$ film (Figure 2b), however, no obvious

triangular nano-domain patterns are observed; and instead, the polarization vectors rotates smoothly to form the vortex core.

In conclusion, using atomic resolution aberration-correction scanning transmission electron microscopy, we have shown that a size effect on the spontaneous flux-closure domains can lead to formation of smoothly continuously rotated polarization patterns in ultrathin ferroelectrics. This is in contrast to the vortex-like structures observed in most previous studies, where the polarization flux-closure patterns are generally composed of shape-conserving domains separated by regular domain walls. As the observed continuous polarization rotation pattern can minimize the crosstalk between neighboring storage bits, our findings provide new insights into the design of ferroelectric devices based on the exotic polar vortex structures.

References:

[1] C. T. Nelson, etc., Nano Letters **11** (2011), 828-834.
[2] The authors gratefully acknowledge the financial support through DOE grant DoE/BES DE-SC0014430.

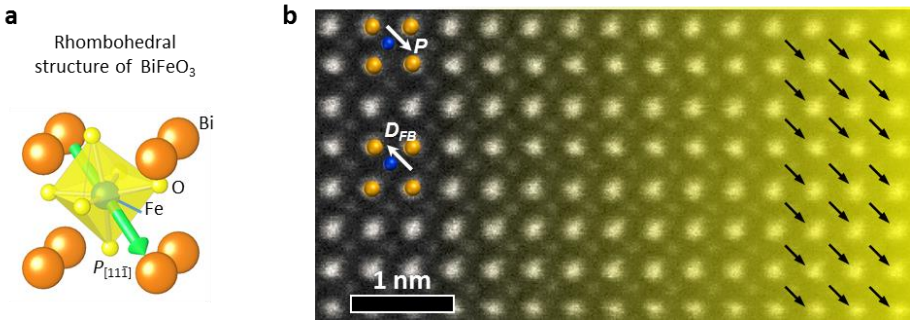

Figure 1. (a) Atomic models of the rhombohedral structure $BiFeO_3$. (b) A HAADF STEM image of $BiFeO_3$ atomic structures, where the $BiFeO_3$ atomic models and polarization map are overlaid.

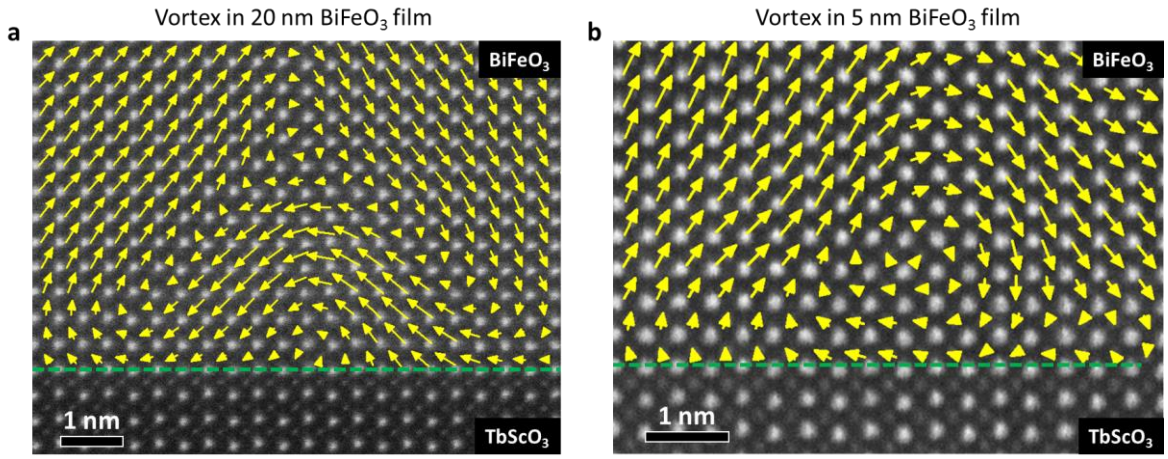

Figure 2. (a) A HAADF STEM image of 20 nm $BiFeO_3$ film grown on $TbScO_3$ substrate, where the mapping of polarization vectors are overlaid. (b) A HAADF STEM image of 5 nm $BiFeO_3$ film grown on $TbScO_3$ substrate, where the mapping of polarization vectors are overlaid.