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Polarization in Ferroelectric $BiFeO_3$ Imaged in 3D Using Four-dimensional Scanning Transmission Electron Microscopy

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Scanning transmission electron microscopy (STEM) has proven to be a highly effective tool for studying polarization in ferroelectric materials. In particular, atomic scale quantification of polarization by mapping the displacement of heavy atoms in high angle annular dark field (HAADF) STEM images has become a universally adopted method for visualizing the polarization in nanostructures such as domain walls [1], defects [2], and vortices [3]. Annular bright-field or integrated differential phase contrast imaging can be used to determine the polarization based on the positions of light elements as well to obtain a complete picture of the atomic structure [4,5]. Finally, recent development of four-dimensional STEM (4D-STEM), which combines the atomic structure with electric field and charge distribution, can reveal new properties of ferroelectric nanostructures [6]. However, in these studies, the polarization is generally only studied in the projected image in two dimensions because nature of STEM imaging. Measuring the polarization in three-dimensions is critical as most polarization patterns in materials have both in-plane and out-of-plane components. This is especially important to determine the exotic polarization topologies, such as skyrmions and other chiral states.

Here, to study the polarization of $BiFeO_3$ (BFO) in 3D, we have used a focused ion beam (FIB) to prepare atomically sharp needles from a 200nm epitaxial BFO film grown on a $TbScO_3$ substrate and attached them to a TEM grid. Needles are prepared such that the (110) plane is visible in TEM with no tilting. With the High-Angle Three-Axis (HATA) holder from Mel Build, we can tilt the sample by ±45 degrees to reach the (100) and (010) planes. To fully reconstruct the 3D structure, an extensive tilt series is usually required. However, due to the symmetry of BFO, projections along (110), (100) and (010) provide the most essential information. High quality data collected with these projections will reveal the detailed polarization states.

HAADF STEM images collected from a BFO needle are shown in Fig. 1. The polarization points towards [0-11] when the sample is tilted to (100) and then towards [-101] when tilted to (010). By combining these two projections, we know that the polarization points along [-1-11] in 3D. Fig. 2 shows images constructed from 4D datasets collected from the (100) and (010) projections. Fig. 2(ab) are annular dark field images collected simultaneously with the 4D datasets used to derive the corresponding charge density images in Fig. 2(cd). Results of the charge density images show reduced elongation of projected O columns in the (100) projection, indicating slight reduction of the polarization in this projection. The opportunities for 3D imaging offered by combining symmetry considerations, a specially prepared sample, and 4D STEM will be discussed [7].

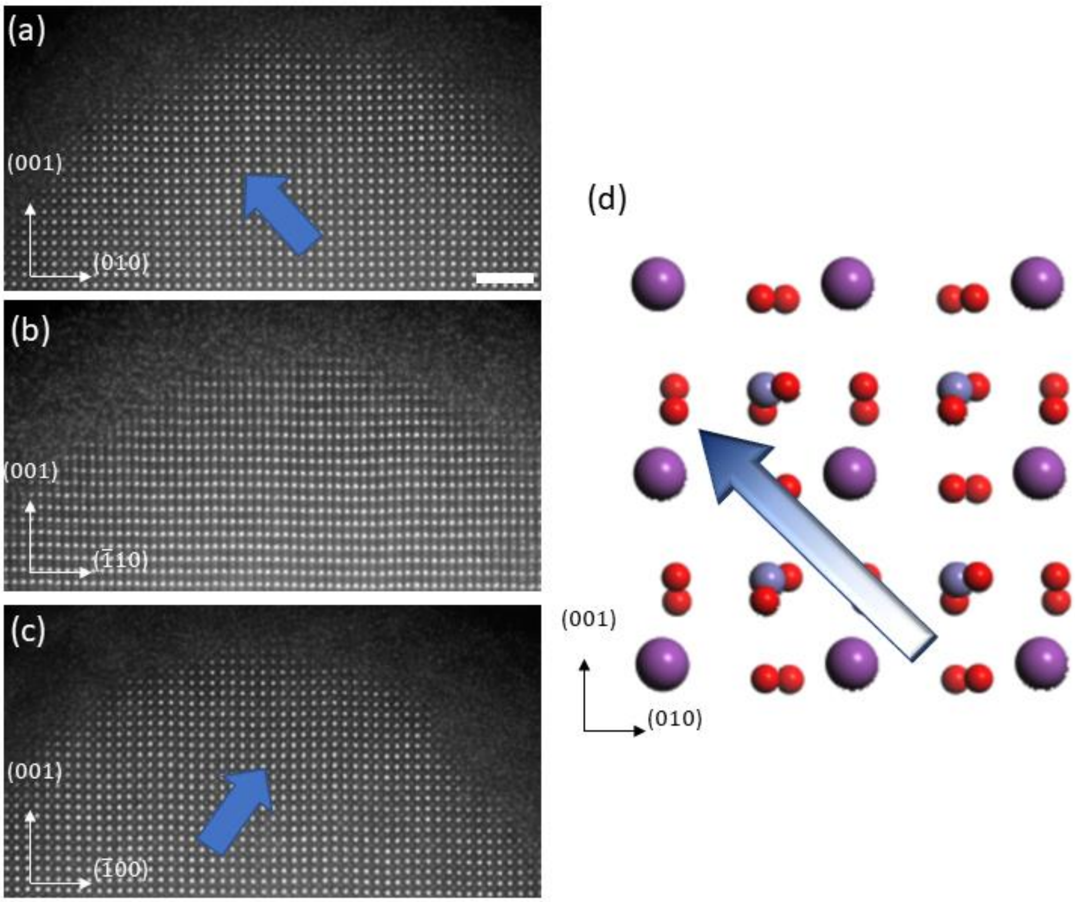

Figure 1. (a-c) HAADF images of the tip of the BFO needle imaged from the (100), (110) and (010) projections, respectively. (d) A structure model of BiFeO3 shown along the (100) projection with the polarization direction indicated. The polarization points from bottom right to top left, front to back. Scale bar is 2 nm

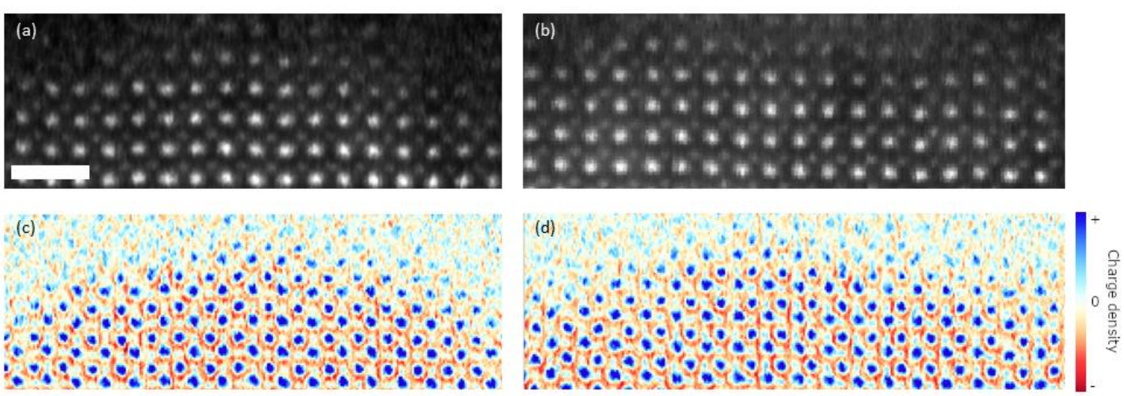

Figure 2. (a, b) HAADF images of the needle tip taken simultaneously with 4D STEM data from the (100) and (010) projections, respectively. (c, d). The charge density calculated from 4D STEM. Positive charge is shown in blue, negative in red. Oxygen columns show increased elongation in the (010) projection. Scale bar is 1nm.

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