doi:10.1017/S1431927620014750
Microsc. Microanal. 26 (Suppl 2), 2020

Multiscale Electric Field Imaging of Vortices in $PbTiO_3$-$SrTiO_3$ Superlattice

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$PbTiO_3$-$SrTiO_3$ (PTO-STO) superlattice has been shown to host a variety of polarization domain structures depending on the periodicity of the superlattice. High periodicity superlattices favor a_1/a_2 domains, while low periodicity superlattices host flux closure domains and continuous vortices appear in between [1]. Four-dimensional scanning transmission electron microscopy (4D-STEM) and integrated differential phase contrast (iDPC) imaging have been applied extensively to study these polarization patterns [2,3]. For example, combined measurements of the polarization and electric field have shown that these vortices host regions of negative capacitance [2]. However, the atomic scale electric field in PTO-STO superlattices has yet to be investigated extensively.

Atomic electric fields were first imaged via DPC in STO in 2012 [4] and a method using 4D STEM has been developed as well [5]. Both techniques utilize a large convergence angle, resulting in a small probe size that provides sub-atomic resolution. Using this method, we have shown that the atomic electric field of ferroelectric $BiFeO_3$ is polarized as based on the displacive shift of ions in the atomic structure [6]. The electric field surrounding both Bi and Fe atoms showed a strong bias in the directions opposite to the atomic displacement. In other electro-magnetic field imaging methods that utilize 4D STEM [2], a small convergence angle is used, where the diffraction disks separate, offering mapping of large areas. Applying 4D STEM with both large and small convergence angle to the same materials can reveal how polarization patterns evolve from atomic scale to larger scale.

Here, in a 16x16 unit cell PTO-STO super lattice sample, we have first applied atomic electric field imaging using a 30mrad convergence angle; this results in a FWHM of about 0.6 Å for the electron probe. The atomic electric field in two vortices is shown in Fig. 1a. To reveal the polarization of the electric field, we have isolated areas around all the A-site atoms in the superlattice (Pb or Sr) and calculated the average electric field within them (as depicted in Fig1b). The average electric field is overlaid on a HAADF image which was collected simultaneously with the 4D STEM data in Fig. 1c. When compared with the polarization map based on atomic displacements of the same region shown in Fig. 1d, the atomic electric field shows the same vortex pattern as the atomic structure.

We have also measured the electric field with a 2.4 mrad convergence angle in the same sample; this results in a probe FWHM of 4 Å. A map of the electric field is shown in Fig. 2a and the electric field direction is shown in Fig. 2b. In Fig. 2b, the direction of the electric field is indicated by the inset color wheel. The electric field direction indicates that the electric field is primarily in-plane and does not follow the vortices observed at atomic scale. The difference in the atomic scale and large-scale atomic fields reveal how polarization patterns in long ranges emerge from the atomic scale [7].

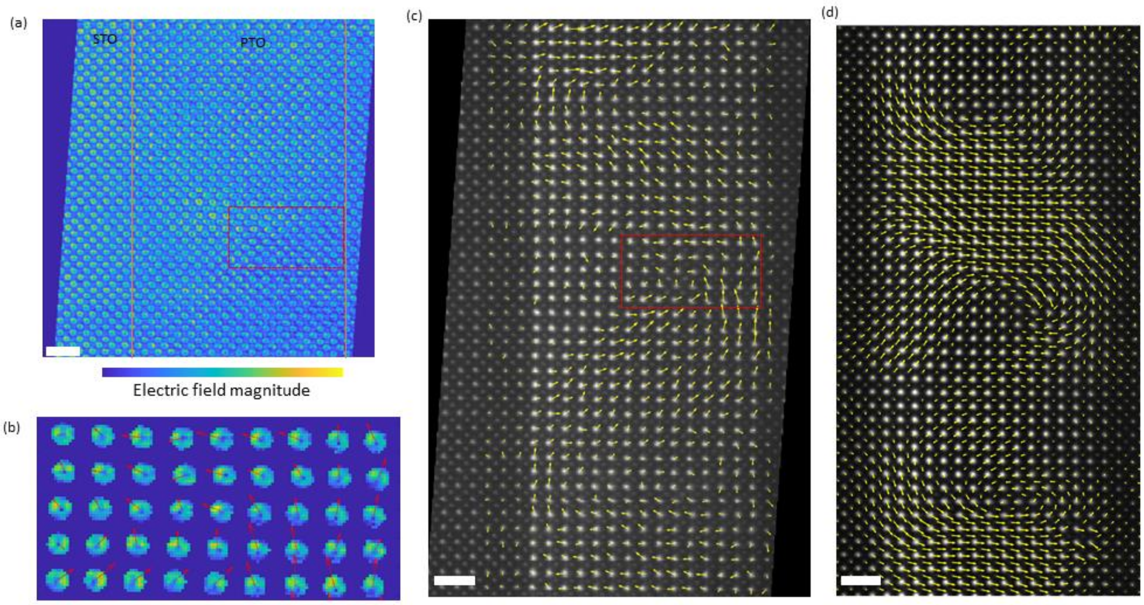

Figure 1. (a) The local electric field of two polarization vortices in one layer of the 16x16 PTO-STO superlattice. The A-site atom locations were segmented from the electric field map in (b); the red arrows indicate the average electric field vector for each Pb atom. This same averaging method is applied to the entire data set in (c) and overlaid with the HAADF image. (d) shows the conventional polarization map from this same region. The average electric field map and the conventional polarization map show nearly the same pattern. The scale bars in (a), (c), and (d) are all one 1 nm.

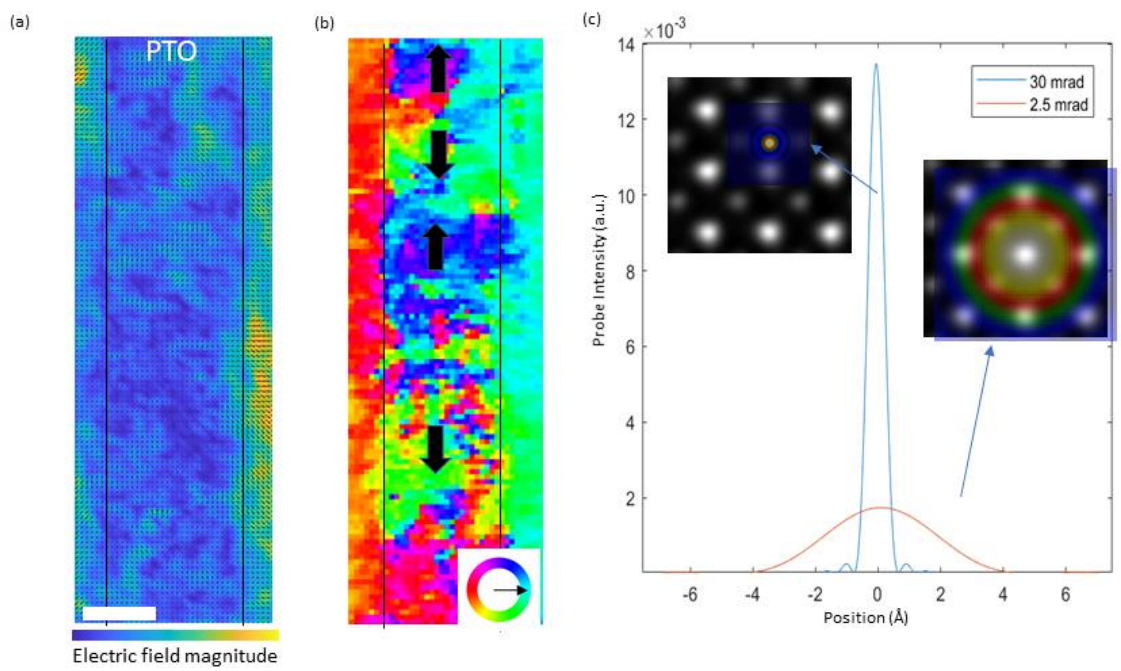

Figure 2. A mapping of the electric field using a low convergence angle electron probe. The magnitude of the electric field vector is shown in the color scale in (a) while the direction is shown in (b). The scale bar is 3 nm. (c) A comparison of the electron probe sizes when using a 30 mrad convergence angle (atomic

https://doi.org/10.1017/S1431927620014750 Published online by Cambridge University Press

resolution) and 2.5 mrad convergence angle (low resolution). The in-sets show the 2D probe intensity overlaid on an STO lattice to show the relative size.

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[7] This work was supported by the Department of Energy (DOE) under Grant DE-SC0014430. TEM experiments was conducted using the facilities in the Irvine Materials Research Institute (IMRI) at the University of California-Irvine.

https://doi.org/10.1017/S1431927620014750 Published online by Cambridge University Press