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Reconstruction of Polarization Vortices by Diffraction Mapping of Ferroelectric $PbTiO_3$ / $SrTiO_3$ Superlattice Using a High Dynamic Range Pixelated Detector

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Ferroelectric polarization vortices realized in the superlattice of complex oxides [1] are emerging phenomena that can be used for low-power memory storage. These complex oxides display local polarizations on the nanometer to atomic scale, requiring characterization techniques sensitive to these effects. Direct measurements of local electric and magnetic fields have been investigated by scanning transmission electron microscopy (STEM) techniques using segmented detectors such as differential phase contrast (DPC) imaging [2], where atomic-resolution reconstruction of the nuclear electric fields have been attempted [3], and direct electron detectors for diffraction imaging showing strain maps calculated by recording the full convergent beam electron diffraction (CBED) pattern [4]. Here, we apply a high dynamic range, high-speed electron microscopy pixel array detector (EMPAD) developed at Cornell specifically for STEM imaging where the full CBED pattern is recorded at an acquisition rate of 1 ms [5]. By careful consideration of symmetry relations of the diffracted disks, quantitative polarization vortices can be reconstructed at nanometer resolution and information from polarity, local specimen tilts, thicknesses, dislocations and strains can be extracted simultaneously.

To investigate the small polarization effects, a high signal to noise ratio (SNR) is required. The EMPAD has exceptionally high dynamic range - a full well of $>10^6$ primary electrons per pixel per image, and single electron sensitivity at 1162 frames per second, on the Tecnai F20 at 200 keV and 10 pA. By collecting the full CBED, we have multiple signal detection capabilities; ADF image of $PbTiO_3$/$SrTiO_3$ superlattice (Figure 1a) down the $[010]_{pc}$ zone axis and changes in the scattered beam's momentum from individual diffracted disk, in this case, the (020) for $<P_x>$ and (200) for $<P_y>$ (Figure 1 b,c), are directly measured and taken simultaneously. Owing to the high dynamic range of the EMPAD, we can analyze shifts in center of mass (COM) of individual diffracted spots, equivalent to having a DPC detector at each diffracted spot. Simple *post hoc* vectorization of $<P_x>$ and $<P_y>$ reconstructs the polarization vortices (Figure 2d) at 0.5-1 nm nanometer resolution, but over an arbitrarily large field of view, distinct from traditional ADF-STEM imaging which requires every atomic position to be measured to picometer precision, limiting both precision and sample size. The EMPAD approach also greatly reduces sensitivity to scan distortions and artifacts from image-enhancing filters.

In plan-view down the $[001]_{pc}$ zone axis, $PbTiO_3$/$SrTiO_3$ superlattice display stripe domains running perpendicular and parallel (Figure 2a) to the surface from the reconstruction of the summed diffracted disk at (320). Fluctuations in contrast from adjacent stripes (Figure 2b) depend on the intensity of the (300) spot and its conjugate; one region displays enhanced intensity for the (300) (Figure 2c) while the other (-300) dims, and vice-versa (Figure 2d). By analyzing the (300) and (-300) spots, we observe the breaking of Friedel's rule, a direct observation of the polarity in our specimen expected for the ferroelectric state.

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The EMPAD allows investigation of non-centrosymmetric variations in the CBED patterns, giving further insight on the Friedel's rule and the nature of our specimen. However, quantitative reconstruction of polarization and fields can only be allowed if specimen tilt, strains, thickness and dislocations are considered. Therefore, careful inspection and analysis of the full CBED pattern and its symmetries can further our quantitative understanding of the polarization and ferroelectric fields in $PbTiO_3/SrTiO_3$ superlattice. [6]

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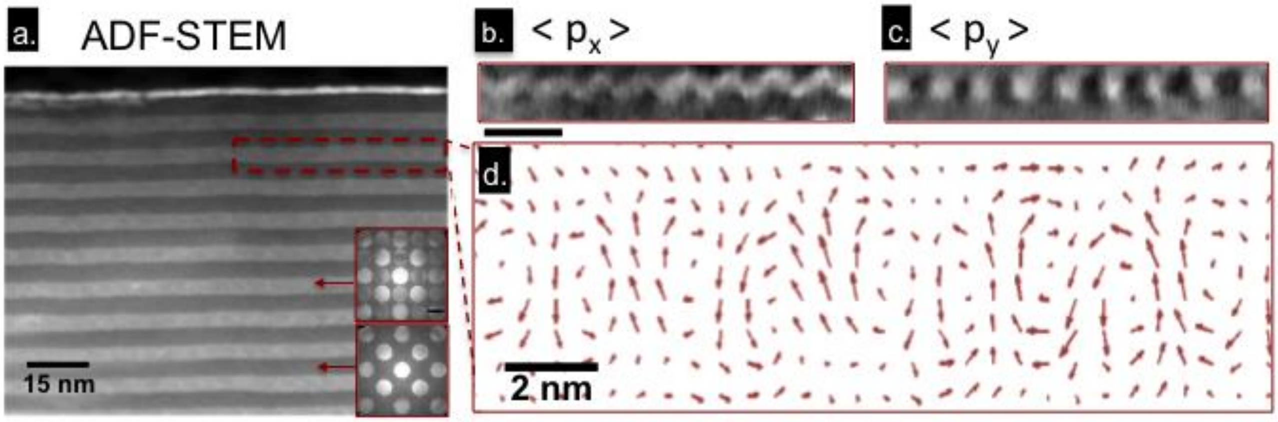

Figure 1. a) ADF image of $PbTiO_3/SrTiO_3$ superlattice along $[010]_{pc}$ (cross section view). CBED pattern of $PbTiO_3$ (top) and $SrTiO_3$ (bottom) are shown, black bar inset on $PbTiO_3$ displays a size scale of 6.4 mrad. b) $\langle p_x \rangle$, momentum shifts of the (020) in x. c) $\langle p_y \rangle$, momentum shifts of the (200) in y. Black bar on lower left of b represent a scale of 10 nm. d) Polarization vortices reconstructed using $\langle p_x \rangle$ and $\langle p_y \rangle$ from region inside of dashed line in (a).

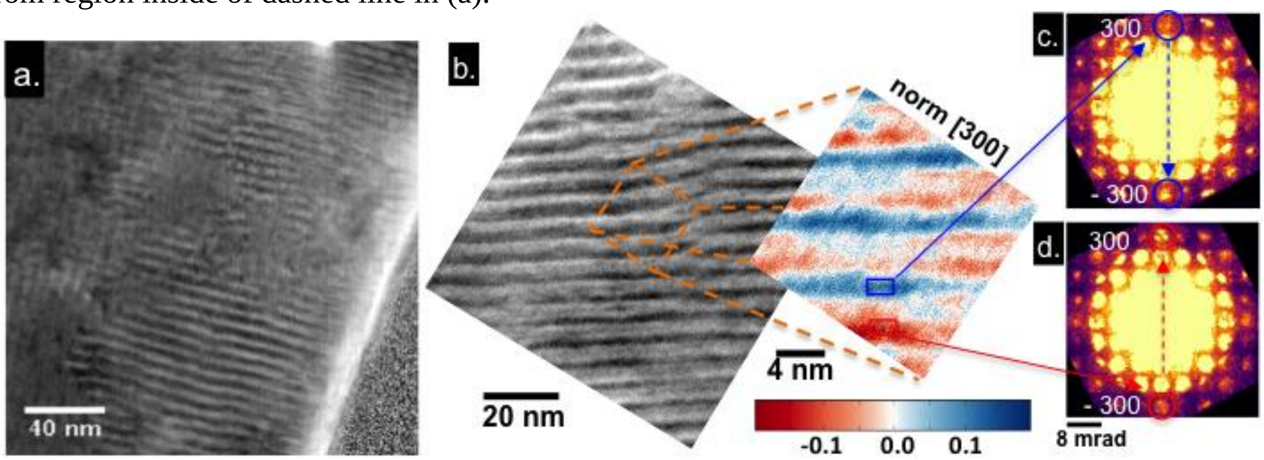

Figure 2. a) Summed (320) diffracted disk image displays stripes running along the horizontal and vertical direction of the specimen in plan-view $[001]_{pc}$. b) Analysis of the normalized (300) disks showing stripe domains at low magnification and high magnification. Contrast arises from the symmetry breaking in the alternating intensities of the (300), (-300) pair of disks. c) CBED of region showing reduced 300 and enhanced -300 contrast, d) vice-versa.

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