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Meeting-report

Disentangling Tilt and Polarization Measurements in 4D-STEM Measurements of a Multilayer by Inversion of a Stacked Bloch Wave Model

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Engineered oxide superlattices have been shown to exhibit a number novel polar textures at the nanoscale as a result of complex interplay between elastic, electrostatic, and dipole energies. Visualization and quantification of these polar textures has been achieved with many techniques, such as x-ray scattering and second harmonic generation. The most highly resolved snapshots of these textures typically are obtained using transmission electron microscopy. Electron imaging of these structures has conventionally been performed using darkfield imaging [1] and recently by using four-dimensional STEM (4D-STEM). These approaches rely on the change in the intensity of certain Bragg reflections due to the distortion of the structure that accompanies electrical polarization. In darkfield imaging this is accomplished by imaging one such reflection, while in 4D-STEM additional robustness is obtained by combining virtual darkfield images from a Friedel pair of reflections or by observing features in Kikuchi bands.

Unfortunately, these approaches to imaging polarization structures are challenging when examining a multilayer sample. The intensity of the low-order Bragg reflections used for mapping the polar texture are also strongly impacted by a number of other effects. Slight tilts of the foil across a large field of view also cause intensity differences between Friedel pairs of reflections, and this can cause the apparent polarization to be strongly suppressed or even reversed. This is illustrated in Figure 1, which shows the polarization measured from the symmetry breaking of the (200) Friedel pair for a fully x-polarized PTO layer in a multilayer sample as the foil is tilted away from the perfect zone axis orientation. When tilted as by only 5 mrad (about 0.3°) in the direction of the polarization, the polarization measurement is fully inverted. Because the polarization textures examined here are only present in multilayer samples, multiple scattering effects strongly affect the diffraction intensities and lead to a complicated and nonlinear relationship between the local polarization and diffracted intensities.

In this work, we develop a dynamical inversion procedure for disentangling the local tilt of the crystal from the local polarization and accounting for multiple scattering effects. We model the electron diffraction process using a stacked Bloch wave model [2], evaluated as the product of multiple scattering matrices corresponding to each layer of the material. Similar to the PRIMES family of methods [3], we parameterize the model using the structural distortions and experimental artifacts that are relevant to our measurement, namely local polarization and tilt, and use an optimization routine to determine these parameters from the measured diffraction intensities. As the diffraction calculations used in the model are relatively computationally expensive, we use a semi-analytical method for finding the gradients of the diffracted intensities with respect to the structural parameters in order to efficiently perform regularized gradient descent.

Experimental 4D-STEM data were obtained from a STO/PTO/STO trilayer sample (where each layer is 16 unit cells thick) imaged in nanodiffraction mode in plan-view and using an energy filtered direct electron detector. The polarization map obtained from fitting to the stacked Bloch wave model is shown in Figure 2, which shows the texture of the polar vortices. By matching the model to all of the diffracted intensities measured in a 4D-STEM dataset, rather than one or two Bragg reflections, we are able to extract a more robust measurement and separate the effects of mistilt and polarization. We will also discuss the potential of using this approach to determine the chirality of the vortices by recovering third-dimension information from the stacked Bloch wave model [4].

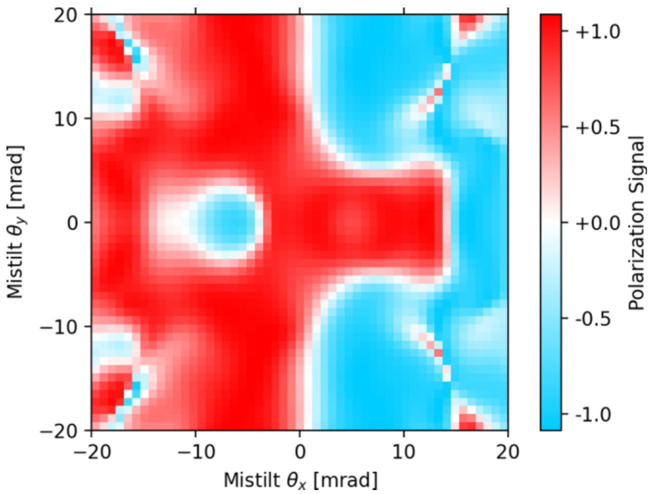

Fig. 1. Apparent polarization of the PTO layer (color axis) in a fully x-polarized STO/PTO/STO trilayer as measured by the asymmetry of the (200) Friedel pair as a function of mistilt from the perfect zone axis orientation. Tilts of about 5 mrad away from the x-axis cause a reversal of the apparent polarization.

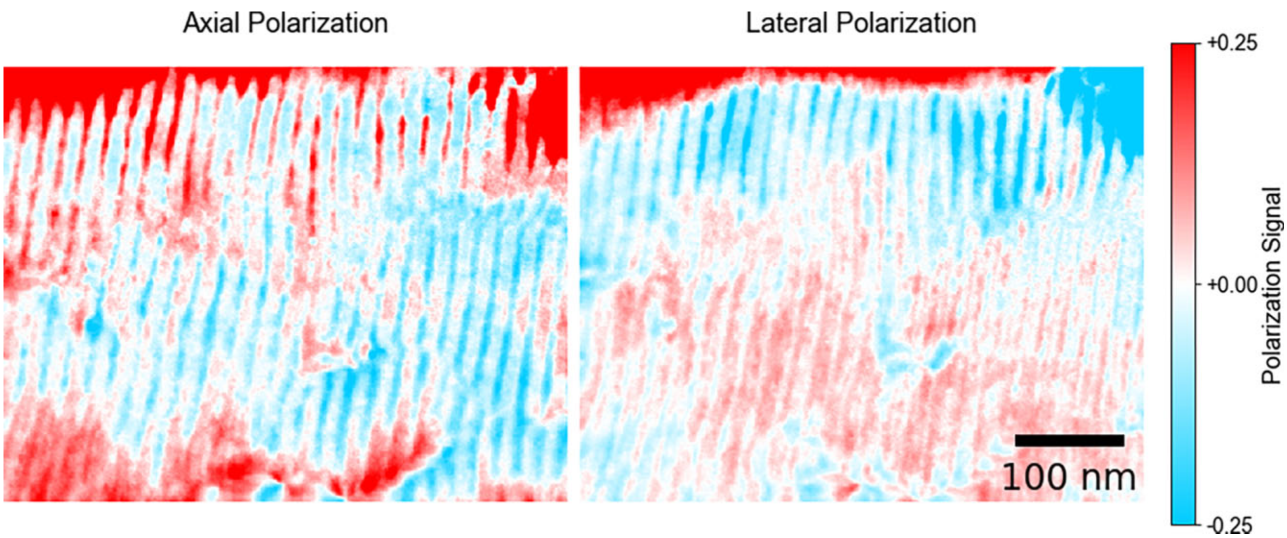

Fig. 2. Polarization texture of the PTO layer in a STO/PTO/STO trilayer sample recovered by stacked Bloch wave model inversion, exhibiting polar vortices.

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