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Probing Grain-boundary Structure and Electrostatic Characteristics in a $SrTiO_3$ Bi-crystal by 4D-STEM

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Grain boundaries (GB) play a critical role in the electrical performance of many electroceramic devices, e.g. varistors, resistors, and capacitors. It is well accepted that charged defects and space charges near the GB dominate its electrical behavior, which is often related to nonstoichiometry or segregation. The characterization of the atomic structure and electrostatic properties of a GB is therefore mandatory and has attracted attention for decades. For example, electron holography and differential phase-contrast imaging techniques have been used to measure the electrostatic potential of materials [1-4]. Recently, further progress on STEM detectors allows the detection of a full diffraction pattern for every probe position on the specimen, through which one can reconstruct electrostatic field maps at the nanometer and atomic scale [5, 6].

In this study [7], by employing four-dimensional scanning transmission electron microscopy (4D-STEM), we investigate the atomic structure and electrostatic properties of a GB in a $SrTiO_3$ bi-crystal. Figure 1a shows an overview high angular annular dark-field (HAADF)-STEM image of the GB structure of a Σ 5 (310) [001] bi-crystal. The 4D-STEM measurements were performed at the region marked with a white dashed box in Figure 1a. Figures 1(b), (c), (d), and (e) show the annular dark-field (ADF), annular bright-field (ABF), integrated differential phase contrast (iDPC), and ptychographic single-sideband (SSB) images, respectively, reconstructed from a 4D dataset. The oxygen positions at the GB are visible in the iDPC (Fig. 1d) and SSB (Fig. 1e) images, showing a better contrast than in the ABF (Fig. 1c) image. The distorted and incomplete oxygen octahedra in the GB core indicate a structural disorder along the z-direction.

Figure 2a-h shows the atomic electric fields in the GB under different focus conditions. The given intensity scale bars in the atomic electric-field maps indicate the relative intensity. The intensity of the electric field in the GB is different by changing the focus conditions. The structure model for the ADF- and ABF-image simulations displayed in Figure 2e is obtained from the proposed GB core structure in Figure 2a. Figures 2 (f), (g), and (c) represent the CoM images under the conditions that the electron probe is focused on the entrance surface, midplane, and exit surface of a 16 nm thick sample, respectively. The diffraction contrasts in the GB are different by changing the focus conditions. For example, the contrasts in the GB under the over-focus condition are brighter than in the bulk. However, an opposite situation is obtained in the under-focus condition. Therefore, it is hard to distinguish the purely geometrical and charge effects by atomic electric-field mapping in the GB. Furthermore, we will discuss geometrical and charge effects of atomic electric-field mapping with different sample thicknesses, and analyze the nanometer-scale electrostatic-field measurements across the GB [8].

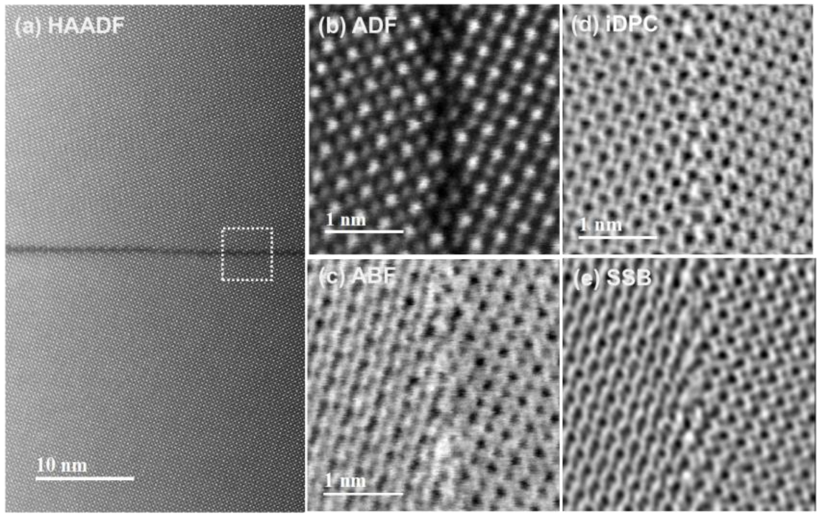

Figure 1. (a) An overview HAADF-STEM image of a Σ5 (310) [001] $SrTiO_3$ bi-crystal. Reconstructed (b) ADF, (c) ABF, (d) iDPC, and (e) SSB images from a 4D dataset.

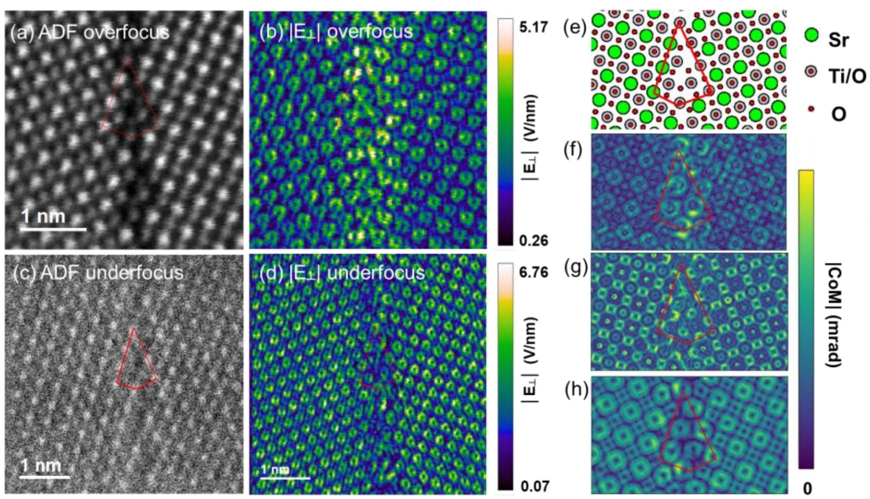

Figure 2. HAADF images in over-focus (a) and under-focus (c) imaging conditions. The corresponding reconstructed electric field maps in (b) and (d). (e) A supercell for image simulations was obtained from the proposed GB core structure in the HAADF image. The reconstructed CoM images under (f) over-focus, (g) in-focus, and (h) under-focus imaging conditions, respectively.

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