Analyzing polarization and lattice strains at the interface of ferroelectric heterostructures on atomic scale using Cs-corrected scanning transmission electron microscopy (STEM)

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Ferroelectric thin films are attractive candidates for capacitors in random access memory (FeRAM) devices, in which a reversible spontaneous polarization is utilized to store information. However, below a critical thickness, the ferroelectric polarization usually disappears, since imperfections at the interface obstruct the spontaneous polarization of ferroelectric films. Therefore, an in-depth understanding of the chemical and structural information at the interface on the atomic scale is indispensable for processing high-quality epitaxial thin films.

The investigated thin film heterostructures were grown by RHEED-assisted pulsed laser deposition (PLD). The ultrathin epitaxial $BaTiO_3$ was grown on niobium doped $SrTiO_3$ and the top electrode $SrRuO_3$ was then deposited. One unit cell of $BaRuO_3$ is embedded between $BaTiO_3$ and $SrRuO_3$. Cross-section specimens were prepared by focused ion beam (FIB). To remove artifacts during FIB preparation, the specimen was finally thinned with a low acceleration voltage (2 kV).

To analyze the interface of ferroelectric heterostructures, the HAADF imaging technique is an ideal tool due to its high sensitivity to the atomic number Z. The use of the HAADF detector in STEM allows nearly a direct interpretation of the images unlike the ambiguities in high-resolution transmission electron microscopy (HRTEM).

To minimize the sample drift in STEM mode, a series of 30 HAADF images is acquired every second and the sample drift is corrected by the displacement correction with the *imtools* software package [1]. The drift corrected HAADF image shown in Figure 1 proves perfect epitaxial growth of the thin film heterostructures. The $BaTiO_3$ thin film is fully strained by the $SrTiO_3$ substrate because of the lattice parameter difference. The in-plane lattice parameters of the bulk form of $SrTiO_3$ and $BaTiO_3$ are 3.90 Å and 3.99 Å respectively [2]. For the identification of the tetragonal distortion of the $BaTiO_3$ film, the in-plane and out-of-plane lattice parameters are determined by analyzing the intensity profile (Table 1). As a result, the out-of-plane lattice parameter of 4.17 Å of the $BaTiO_3$ thin film exceeds considerably the value of 4.03 Å of the bulk form. The lattice parameters measured by HAADF are in good agreement with reported values from synchrotron x-ray diffraction and theoretical prediction [3].

The results shown in Figure 1 and Table 1 demonstrate the presence of the tetragonal distortion of the thin $BaTiO_3$ film strained by the $SrTiO_3$ substrate. Moreover, the small shift of Ti atomic column was visible in the HAADF image. However, local mistitling of $BaTiO_3$ film could lead to a misinterpretation. Therefore, to elucidate ferroelectric polarization the relative shift of Ti and O columns along the [110] direction should be studied by HRTEM in negative Cs imaging condition.

References

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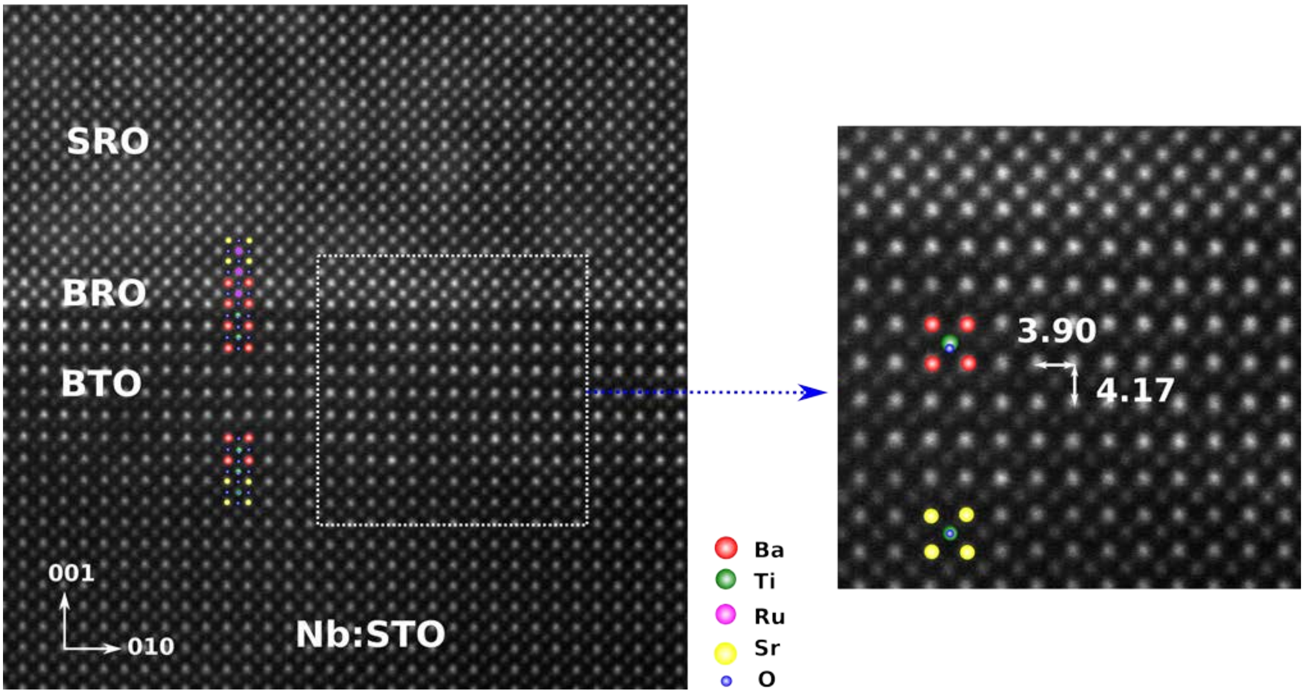

Figure 1. *The drift corrected HAADF image of ferroelectric heterostructures along the [100] direction. The image was acquired an aberration-corrected STEM at an acceleration voltage of 300 kV. The atomic column positions are indicated by different colours. The in-plane and out-of-plane lattice parameter of the $BaTiO_3$ are indicated and the small shift of Ti column is visible on the right image.*

Table 1. *The lattice parameters of ferroelectric heterostructures by analyzing the intensity profile.*

	in-plane (a) [Å]	out-of-plane (c) [Å]	c/a
SRO	3.90 ± 0.05	3.90 ± 0.05	1.00 ± 0.03
BTO	3.90 ± 0.05	**4.17 ± 0.05**	**1.07 ± 0.03**
STO	3.90 ± 0.05	3.93 ± 0.05	1.01 ± 0.03