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Study of the Ultrathin Ferroelectric $BaTiO_3$ Film using Scanning Transmission Electron Microscopy

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Ferroelectric materials can be used for the ferroelectric random access memory (FeRAM) For the miniaturization the thin film structures are used in general. However, there is the critical thickness of ferroelectric materials, below which ferroelectricity abruptly disappears. Besides, the depolarization field, the epitaxial strain, the termination of the ferroelectric film, and the electric filed affect the ferroelectric properties.

Due to the complex of the influences on the ferroelectric properties, the critical thickness of the ferroelectric materials could not be clearly explained. In this study, the ferroelectric heterostructure of $SrTiO_3$ (STO)/ $BaTiO_3$ (BTO)/ $BaRuO_3$ (BRO)/ $SrRuO_3$ (SRO) is grown by pulsed laser deposition (PLD). The top termination of the BTO thin film was altered by the deposition of the additional BRO layer. The cross-sectional TEM specimen was prepared by the focused ion beam (FIB) workstation. After the FIB milling, the specimen was cleaned by Ar^+ ion milling with the low energy and incident angle.

The ferroelectric heterostrcuture was investigated using scanning transmission electron microscopy (STEM). High angle annular dark filed (HAADF) imaging was performed to obtain the local structural distortions at the atomic level. In addition, the atomic columns can be clearly identified by the Z-contrast in the HAADF image. To precisely measure the atomic column positions, a fast sequential imaging was applied. The specimen drifts between image frames were corrected by the cross-correlation algorithm and then the corrected frames were averaged out. By applying this technique, the specimen drift could be significantly reduced with a reasonable signal-to-noise ratio. Thus, the atomic column positions and the corresponding intensities were quantitatively analyzed.

In STEM, a spatially resolved electron energy-loss spectroscopy (EELS) is applicable to access information on the local electronic structures. In particular, the fine structures on the Ti-L23 edge are sensitive to the neighboring atomic environment and closely related to the ferroelectric properties [1]. To obtain EEL spectra at the unit cell level StripeSTEM [2] was performed and the fine structures of the Ti-L_{23} edges through the interfaces were analyzed in terms of the crystal field splitting (CFS) effect and the valence states.

By applying drift-corrected HAADF imaging the tetragonal distortion of the ultrathin BTO film strained by STO was confirmed. A relative shift of the Ti-columns with respect to the center of the perovskite structure was observed. In the BTO thin film, the reduction in the crystal field splitting energy and an asymmetric broadening of the e_g peak were observed. These results indicate that the ultrathin BTO film (7 unit cells) strained by STO exhibit the ferroelectric polarization along the [001] direction.

References:
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[2] Heidelmann, M., Barthel, J. and Houben, L., Ultramicroscopy **109** (2009), p. 1447–1452. [3] The authors acknowledge support by the German research foundation (DFG) through projects MA1280/32-1 and Di919/3-1k.

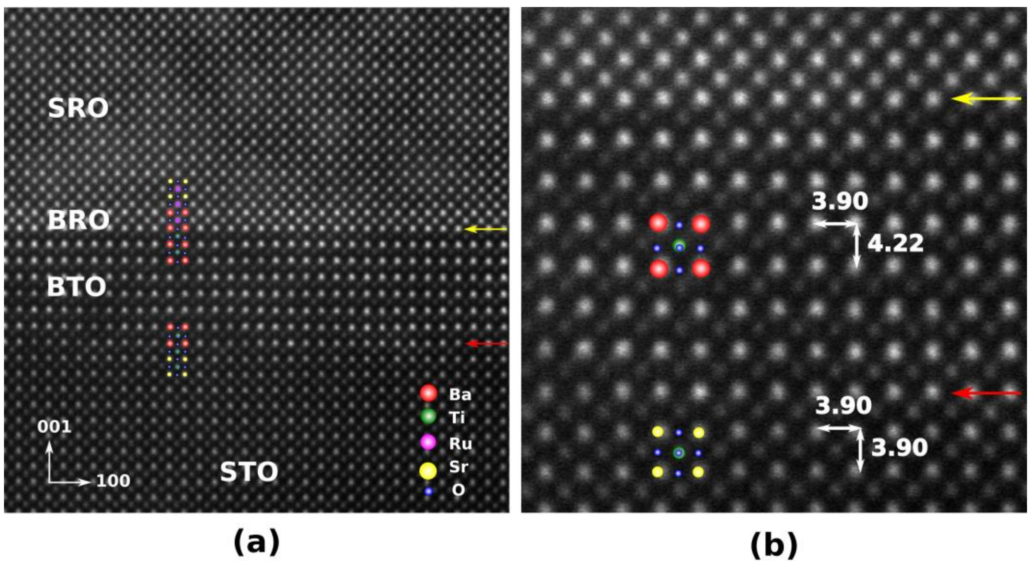

Figure 1. The drift-corrected HAADF image of the STO/ BTO/ BRO/ SRO heterostructure projected along the [010] direction.

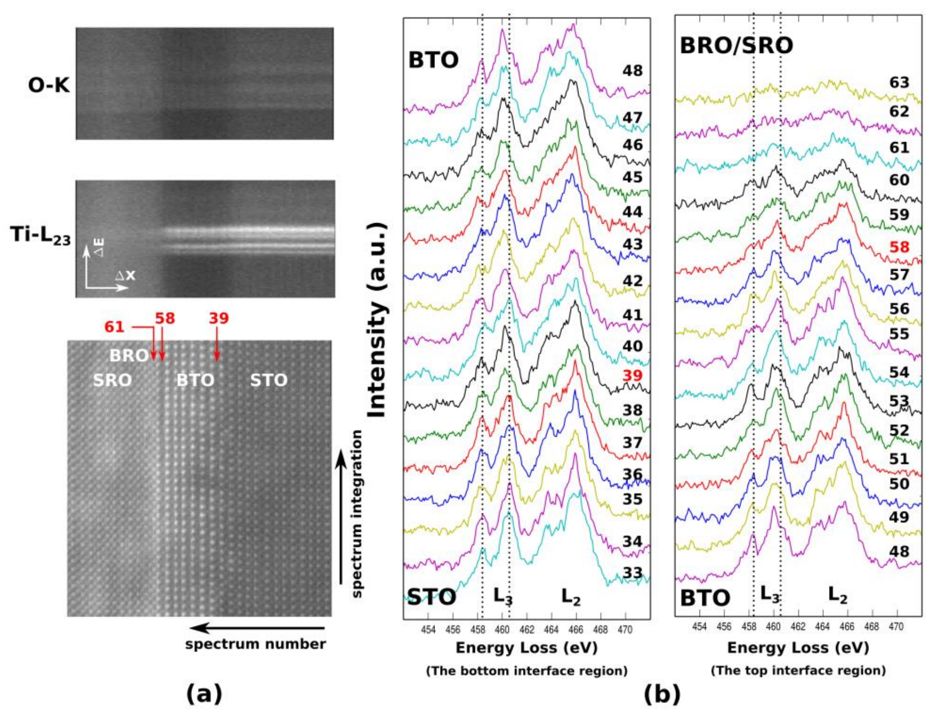

Figure 2. (a) StripeSTEM. (b) A series of EEL spectra of the Ti-L_{23} edge which covers the range from the bottom and top interface.

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