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Simultaneous Z-Contrast and Phase Contrast Imaging of Oxygen in Ceramic Interfaces

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Materials science often comes down to understanding the role of interfaces on material properties. Determination of the structure and composition of interfaces in oxides is complicated by the difficulty in imaging the oxygen sublattice. Professor Ruhle and others have recently shown imaging of oxygen in oxides is feasible.[1,2] However, here we have examined the oxygen sublattice at the $SrTiO_3/Al_2O_3$ interface using simultaneous coherent and incoherent imaging techniques. The instrument used in this work is the ORNL VG Microscopes HB603U scanning transmission electron microscope (STEM) operated at an accelerating voltage of 300 kV. This microscope has been fitted with a NION aberration corrector, which has improved resolution as measured using the intensity profile (FWHM) across single La atoms to 0.7Å. Aberration correction enables smaller, brighter probes with resulting improvements for Z-contrast imaging in resolution, image contrast and signal-to-noise ratio. In addition, via reciprocity, aberration corrected phase contrast images may be obtained simultaneously using a small axial detector.

Films of {111}-oriented $SrTiO_3$ grown on {0001}-oriented _-Al_2O_3 (sapphire) using pulsed laser deposition were examined in cross section. This heterostructure is complicated by the fact that multiple in-plane alignments can co-exist. Only by choosing appropriate deposition parameters (in particular, the growth rate and substrate temperature) was it possible to obtain single-phase epitaxial films. Figure 1 shows simultaneously collected atomic resolution annular dark field and bright field images of the [-110] projection of the $SrTiO_3/Al_2O_3$ interface. In this projection, {111} planes in $SrTiO_3$ have a ABAB stacking sequence, with A-type planes composed of SrO and O columns and B-type planes composed of Ti columns. The {0001} planes of Al_2O_3 in this projection are composed of alternating layers of Al and O. These images show that we can detect oxygen columns (Z=8) in both the Z-contrast and phase contrast images not only when embedded with relatively light Al columns (Z=13) but also in the presence of Ti (Z=22) and Sr (Z=38). The Z-contrast technique provides an incoherent image, thus, there is no phase problem for structure determination. The intensity of atom columns reflects their mean square atomic number (Z) and there are no contrast reversals with crystal thickness. In the phase contrast method of high resolution imaging, contrast is sensitively controlled by focus of the objective lens and specimen orientation, thickness and scattering power. This makes the images of unknown structures difficult to directly interpret. However, this sensitivity can be exploited to provide much greater contrast variations than can be obtained from the Z-contrast images of low atomic number elements. The simultaneous Z-contrast/phase contrast image pairs combine the advantages of both techniques and provide improved sensitivity to the oxygen sublattice in ceramic materials.

Simulations of both the coherent and incoherent scattering [3] will be used to demonstrate how the interfacial configurations affect both types of images and will be used to explore the possibility of quantifying oxygen content at ceramic interfaces.

References

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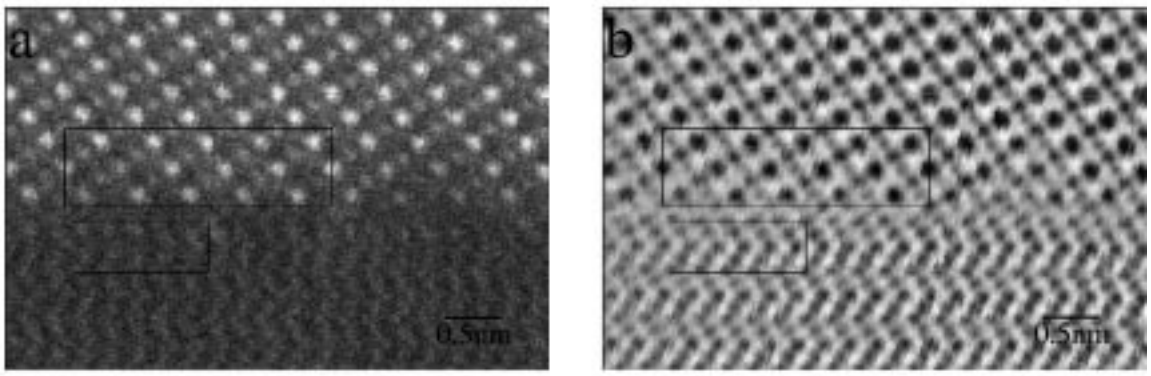

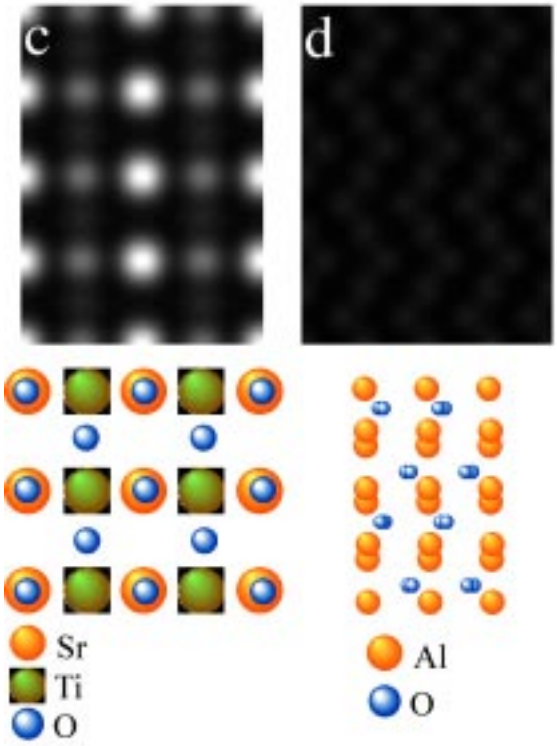

Simultaneous Z-contrast (a) and phase contrast (b) images (raw data) of the <-110> projection of {111} $SrTiO_3$ on (0001) Al_2O_3. The vertices of the larger box in the $SrTiO_3$ film are located on Ti columns while the vertices of the smaller box in the Al_2O_3 substrate are on O columns. It can be seen that the Ti plane of $SrTiO_3$ bonds to the oxygen plane of Al_2O_3. Simulations of the incoherent scattering from $SrTiO_3$ (c) and Al_2O_3 (d) match well with the experimental image and reveal the weak but detectable scattering from the oxygen sublattice of these materials.