

ELNES analysis of γ -Al₂O₃/SrTiO₃ and LaTiO₃/SrTiO₃ interfaces.

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Complex oxide-oxide interfaces are attracting attention because of many interesting and unexpected properties including conductivity, superconductivity, ferromagnetism and magnetoresistance. The two-dimensional electron gas (2DEG) formed at the interface between two insulating oxides shows potential for the development of all-oxide electronic devices. Here, we have investigated two SrTiO₃-based systems with possible 2DEG based on γ -Al₂O₃/SrTiO₃ and LaTiO₃/SrTiO₃. Energy-loss near-edge fine structure (ELNES) analysis using aberration-corrected STEM can provide rich and detailed information about local electronic structure with a spatial resolution of better than one unit cell.

The EELS spectra were collected over a 2D area, and then averaged along the interface to reduce the impact of electron-beam damage during data acquisition. The ELNES signals were extracted by removing the background with power law fitting, and the ELNES were then fitted to known spectra in the literature. The fitting coefficients of each species are directly related to the area density. Figures 1 and 2 show the fitting coefficients of two different γ -Al₂O₃/SrTiO₃ samples, grown by atomic-layer deposition (ALD) and molecular-beam epitaxy (MBE), respectively. The coefficients of SrTiO₃ are in red, the coefficients of SrTiO_{2.75} (in which Ti⁴⁺ is partially reduced to Ti³⁺) are in green, the fitting coefficients of γ -Al₂O₃ are in blue, and the black lines are the sum of coefficients from different species. The Ti-L edge is sensitive to the oxidation state of Ti species (e_g and t_{2g} peaks will split in Ti⁴⁺ but merge in Ti³⁺) and the O-K edge is sensitive to any order or disorder of the oxygen sub-lattice. From the Ti-L edge both of the γ -Al₂O₃/SrTiO₃ samples show a peak of SrTiO_{2.75} at the interface. However, only samples with the 2DEG present show a peak for SrTiO_{2.75} from the O-K edge at the interface, suggesting that oxygen vacancies in STO near the interface could be the key to the 2DEG [3]. Figure 3 shows analysis for a sample grown by MBE with 25 unit cells of SrTiO₃/1 unit cell of LaTiO₃/25 unit cells of SrTiO₃ on SrTiO₃ substrate. There is a broad peak of Ti³⁺ at the SrTiO₃/1uc LaTiO₃/ SrTiO₃ interface in the Ti-L edge, which is consistent with theoretical prediction that a 2DEG will form when electrons from Ti³⁺ in LaTiO₃ transfer into Ti⁴⁺ in SrTiO₃ near the interface. Future work will include improving the signal to noise ratio by an order of magnitude by using lower dose, noise subtraction and averaging methods, and then quantitatively comparing the ELNES results with DFT calculations.

References:

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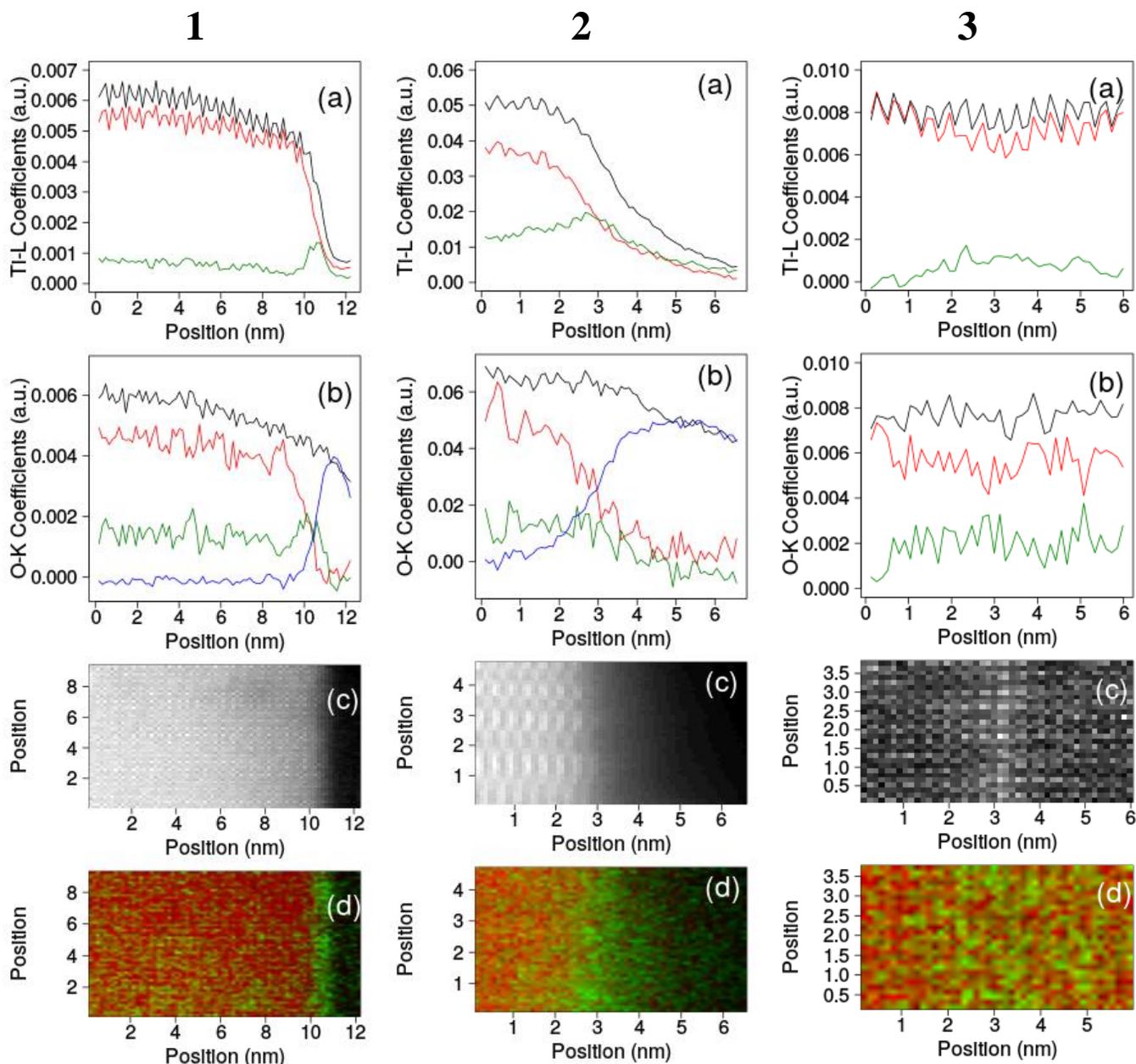


Figure 1. γ -Al₂O₃/SrTiO₃ sample grown by ALD at 345 °C with 2DEG density of 6×10^{13} cm⁻² at room temperature. (a) Fitting coefficients from Ti-L edge. (b) Fitting coefficients from O-K edge. (c) Corresponding HAADF image. (d) False color map for coefficients of different species from Ti-L edge.

Figure 2. γ -Al₂O₃/SrTiO₃ sample grown by MBE at 600 °C then annealed in air at 400 °C. No 2DEG is present. (a) Fitting coefficients from Ti-L edge. (b) Fitting coefficients from O-K edge. (c) Corresponding HAADF image. (d) False color map for coefficients of different species from Ti-L edge.

Figure 3. SrTiO₃/1 μ c LaTiO₃/SrTiO₃ interface. (a) Fitting coefficients from Ti-L edge. (b) Fitting coefficients from O-K edge. (c) Corresponding HAADF image. (d) False color map for coefficients of different species from Ti-L edge.