

## ELNES analysis of $\gamma$ -Al<sub>2</sub>O<sub>3</sub>/SrTiO<sub>3</sub> and LaTiO<sub>3</sub>/SrTiO<sub>3</sub> interfaces.

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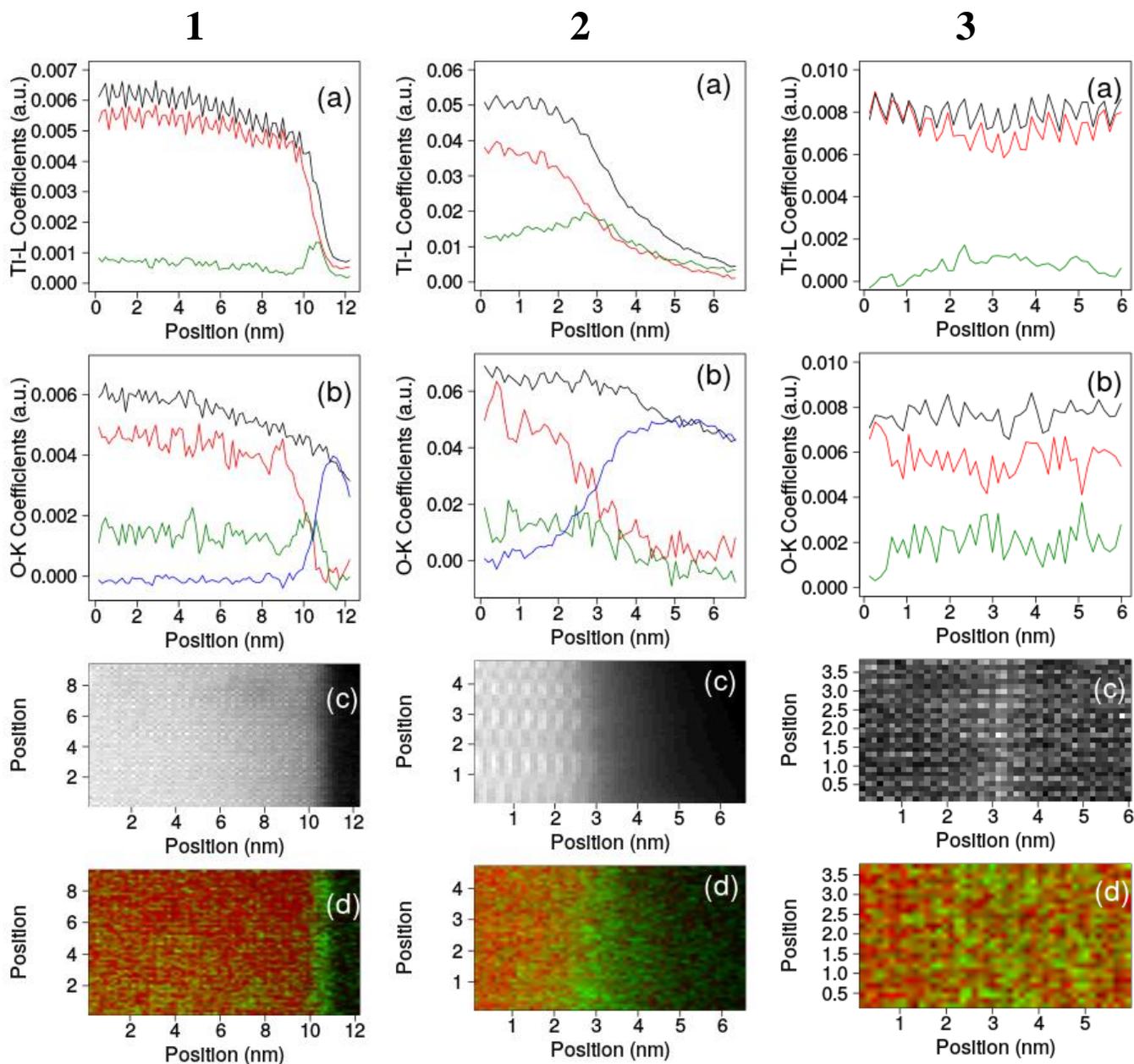
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Complex oxide-oxide interfaces are attracting attention because of many interesting and unexpected properties including conductivity, superconductivity, ferromagnetism and magnetoresistance. The two-dimensional electron gas (2DEG) formed at the interface between two insulating oxides shows potential for the development of all-oxide electronic devices. Here, we have investigated two SrTiO<sub>3</sub>-based systems with possible 2DEG based on  $\gamma$ -Al<sub>2</sub>O<sub>3</sub>/SrTiO<sub>3</sub> and LaTiO<sub>3</sub>/SrTiO<sub>3</sub>. 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  $\gamma$ -Al<sub>2</sub>O<sub>3</sub>/SrTiO<sub>3</sub> samples, grown by atomic-layer deposition (ALD) and molecular-beam epitaxy (MBE), respectively. The coefficients of SrTiO<sub>3</sub> are in red, the coefficients of SrTiO<sub>2.75</sub> (in which Ti<sup>4+</sup> is partially reduced to Ti<sup>3+</sup>) are in green, the fitting coefficients of  $\gamma$ -Al<sub>2</sub>O<sub>3</sub> 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<sub>g</sub> and t<sub>2g</sub> peaks will split in Ti<sup>4+</sup> but merge in Ti<sup>3+</sup>) 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  $\gamma$ -Al<sub>2</sub>O<sub>3</sub>/SrTiO<sub>3</sub> samples show a peak of SrTiO<sub>2.75</sub> at the interface. However, only samples with the 2DEG present show a peak for SrTiO<sub>2.75</sub> 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 SrTiO<sub>3</sub>/1 unit cell of LaTiO<sub>3</sub>/25 unit cells of SrTiO<sub>3</sub> on SrTiO<sub>3</sub> substrate. There is a broad peak of Ti<sup>3+</sup> at the SrTiO<sub>3</sub>/1uc LaTiO<sub>3</sub>/ SrTiO<sub>3</sub> interface in the Ti-L edge, which is consistent with theoretical prediction that a 2DEG will form when electrons from Ti<sup>3+</sup> in LaTiO<sub>3</sub> transfer into Ti<sup>4+</sup> in SrTiO<sub>3</sub> 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.**  $\gamma$ -Al<sub>2</sub>O<sub>3</sub>/SrTiO<sub>3</sub> sample grown by ALD at 345 °C with 2DEG density of  $6 \times 10^{13}$  cm<sup>-2</sup> 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.**  $\gamma$ -Al<sub>2</sub>O<sub>3</sub>/SrTiO<sub>3</sub> 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.** SrTiO<sub>3</sub>/1 $\mu$ c LaTiO<sub>3</sub>/SrTiO<sub>3</sub> 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.