

ELNES spectrum unmixing and mapping for oxide/oxide interfaces.

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The two-dimensional electron gas (2DEG) formed at the interface between two insulating oxides shows many interesting and unexpected properties including conductivity, superconductivity, ferromagnetism and magnetoresistance, and has the potential for development of all-oxide electronic devices. Energy-loss near-edge structure (ELNES) is sensitive to the local electronic structure and has been used to investigate the 2DEG. However, the 2DEG signal is usually very weak and often mixed with other signals coming from the film and/or substrate. Here, we have applied a set of improved algorithms for spectrum unmixing and mapping onto the Ti-L and O-K edges at the LaTiO₃/SrTiO₃ and the γ -Al₂O₃/SrTiO₃ interfaces. Spectra from different phases and the region of the 2DEG were identified and extracted from the experimental data and 2D mapping was done using the extracted spectra. By analyzing the spectrum maps, the possible origin of the 2DEG has been revealed.

The EELS spectra were collected from 2D regions of the samples. Spatial and energy drift correction were applied and outlier data points were identified and removed. After background subtraction, the virtual dimensionality of the dataset was determined and the data dimensionality was reduced by an orthogonal transformation. Each data point was then weighted based on the signal-to-noise ratio (SNR). Pure spectra for each phase were identified and extracted using a noise-adaptive algorithm for mapping of the original 2D dataset. In each step, at least two different algorithms were tested and compared to ensure that no artifacts were present. Several algorithms were modified and improved so that they would work with typical SNR levels in ELNES signals.

Figure 1 shows results for a sample consisting of 1 unit cell of LaTiO₃ embedded between 25 unit cells of SrTiO₃. By comparing the Ti-L edge and O-K edge, it is clear that the combined effects of oxygen vacancies and the La layer contribute to reduction of the Ti⁴⁺. Further studies are ongoing to separate and understand the two effects. Figure 2 shows results for a γ -Al₂O₃/SrTiO₃ sample grown by atomic-layer deposition (ALD). The Ti-L edge shows reduction of Ti at the interface and the O-K edge shows corresponding oxygen vacancies at the same location. The extracted spectra match very well with reported spectra. These results indicate that the 2DEG present at the γ -Al₂O₃/SrTiO₃ interface is associated with the oxygen vacancies [1-3].

References:

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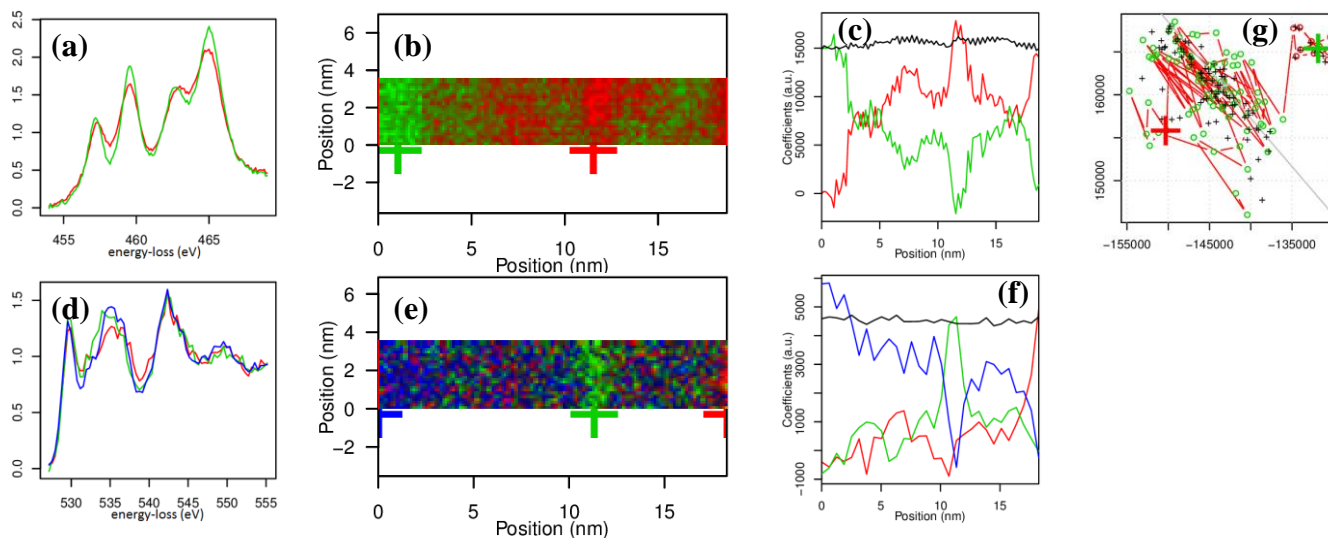


Figure 1. One unit cell of LaTiO_3 in between 25 unit cells of SrTiO_3 grown by MBE at 750°C with 2DEG present. (a) Unmixed pure spectra of Ti-L edge. Red is reduced Ti. Green is SrTiO_3 . (b) 2D mapping of spectral image; “+” symbols indicate approximate positions where pure spectra were extracted. (c) Corresponding line-averaged profile. (d) Unmixed pure spectra of O-K edge. Red is SrTiO_3 with oxygen vacancies. Green is LaTiO_3 . Blue is SrTiO_3 . (e) 2D mapping of spectral image. (f) Corresponding line-averaged profile of the two Ti species. (g) Schematic plot illustrating how the spectra extraction algorithms work. Circular points are data points projected in low dimensional space. Small black “+” are data points after weighting by SNR. Large “+” are “pure” spectra identified by a projection to a hyperplane (not shown here) perpendicular to line from origin to the average (gray line).

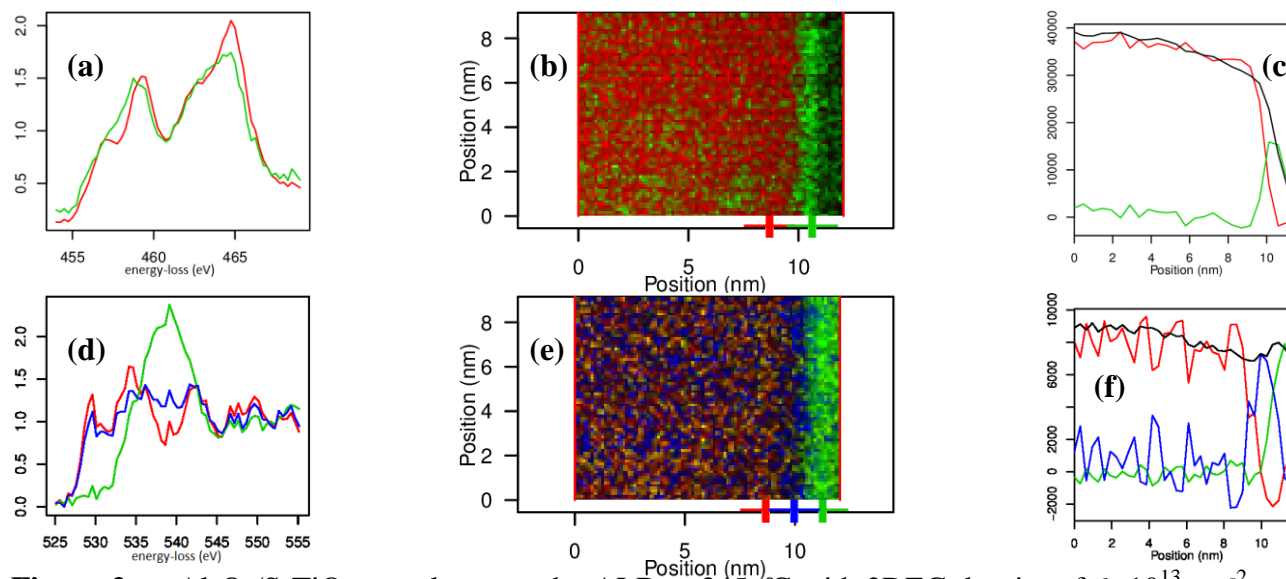


Figure 2. $\gamma\text{-Al}_2\text{O}_3/\text{SrTiO}_3$ sample grown by ALD at 345°C with 2DEG density of $6 \times 10^{13} \text{ cm}^{-2}$ at room temperature. (a) Unmixed pure spectra of Ti-L edge. Red is SrTiO_3 (Ti^{4+}) and green is Ti at lower oxidation state. (b) 2D mapping of spectral image. Left part is SrTiO_3 and right part is $\gamma\text{-Al}_2\text{O}_3$. The interface is at around 10nm. “+” symbols indicate approximately where the pure spectra were extracted. (c) Corresponding line-averaged profile of the two Ti species. Black line is the sum. (d) Unmixed pure spectra of O-K edge. Red is SrTiO_3 (Ti^{4+}). Green is $\gamma\text{-Al}_2\text{O}_3$. Blue is SrTiO_3 with oxygen vacancies. (e) 2D mapping of spectral image. (f) Corresponding line-averaged profile of the two Ti species.