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ELNES spectrum unmixing and mapping for oxide/oxide interfaces.

Sirong Lu[1], Kristy J. Kormondy[2], Thong Q. Ngo[3], Elliott Ortmann[2], Toshihiro Aoki[4], Agham Posadas[2], John G. Ekerdt[3], Alexander A. Demkov[2], Martha R. McCartney[5], and David J. Smith[5]

1. School of Engineering for Matter, Transport and Energy, Arizona State University, Tempe, AZ 85287
2. Department of Physics, University of Texas at Austin, Austin, TX 78712
3. Department of Chemical Engineering, University of Texas at Austin, Austin, TX 78712
4. LeRoy Eyring Center for Solid State Science, Arizona State University, Tempe AZ 85287
5. Department of Physics, Arizona State University, Tempe, AZ 85287

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 $LaTiO_3/SrTiO_3$ and the γ-$Al_2O_3/SrTiO_3$ 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 $LaTiO_3$ embedded between 25 unit cells of $SrTiO_3$. 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 Ti^{4+}. Further studies are ongoing to separate and understand the two effects. Figure 2 shows results for a γ-$Al_2O_3/SrTiO_3$ 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-$Al_2O_3/SrTiO_3$ interface is associated with the oxygen vacancies [1-3].

References:

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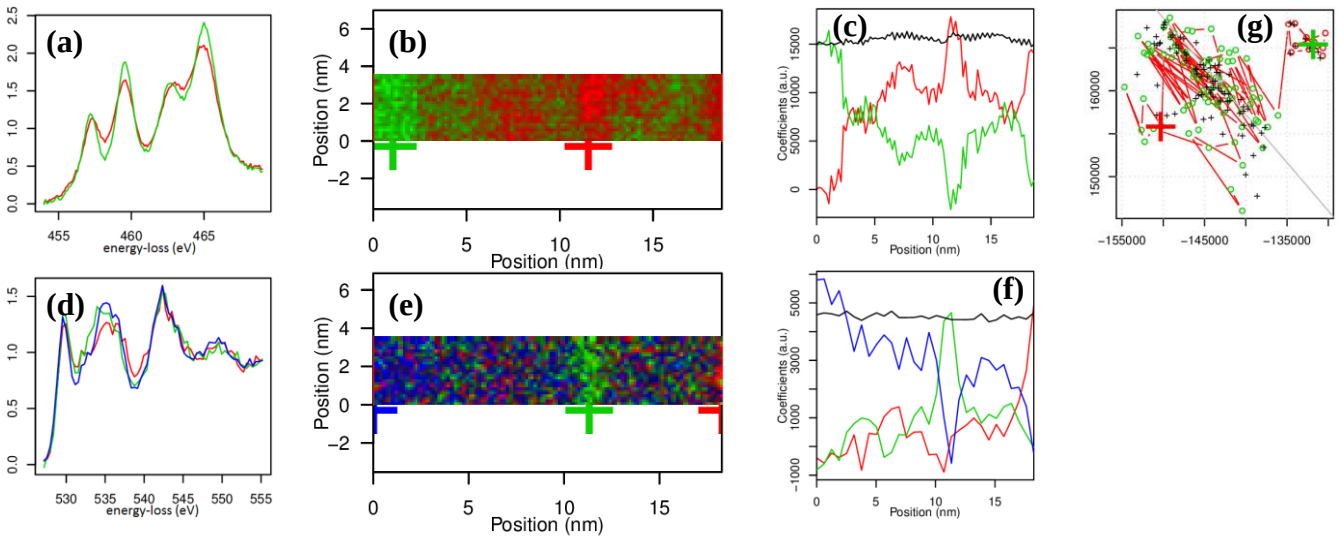

Figure 1. One unit cell of $LaTiO_3$ in between 25 unit cells of $SrTiO_3$ grown by MBE at 750 ℃ with 2DEG present. **(a)** Unmixed pure spectra of Ti-L edge. Red is reduced Ti. Green is $SrTiO_3$. **(b)** 2D mapping of spectral image; "+" symbols indicate approximate positions where pure spectra were 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).

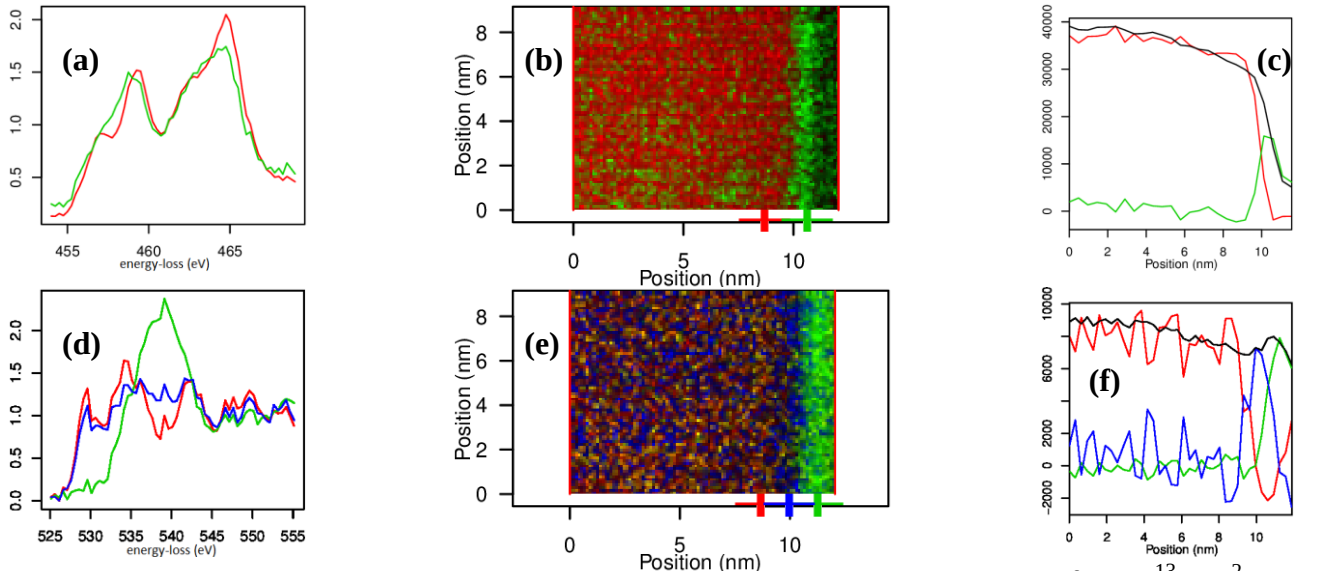

Figure 2. γ-Al_2O_3/$SrTiO_3$ sample grown by ALD at 345 ℃ with 2DEG density of 6×10^{13} cm^{-2} at room temperature. **(a)** Unmixed pure spectra of Ti-L edge. Red is $SrTiO_3$ (Ti^{4+}) and green is Ti at lower oxidation state. **(b)** 2D mapping of spectral image. Left part is $SrTiO_3$ and right part is γ-Al_2O_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 γ-Al_2O_3. Blue is $SrTiO_3$ with oxygen vacancies. **(e)** 2D mapping of spectral image. **(f)** Corresponding line-averaged profile of the two Ti species.

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