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Probing Stoichiometry in $LaAlO_3/SrTiO_3$ Interfaces by Aberration-Corrected STEM

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Interfaces between perovskite oxides provide one of the most exciting test beds for stabilizing exotic physical phases not present in bulk materials. The emergence of metallic conductivity, and low temperature superconductivity, between the two insulators $LaAlO_3$ (LAO) and $SrTiO_3$ (STO) is of particular interest; this conducting interface can be grown on silicon, providing a pathway to integration into modern electronic devices [1]. We have recently found the interface conducts when the La/Al ratio is at or below 0.97 ± 0.03 while samples that exceed this limit are insulating [2]. While density functional theory models have explained the conductivity of a simple stoichiometric LAO film with an electronic reconstruction at the interface, fully incorporating off-stoichiometry into the model requires a precise understanding of atomic positions, chemical intermixing and electronic structure as function of La-to-Al ratio. Here, we collect spectroscopic maps on a series of LAO/STO interfaces over a stoichiometry range to correlate the electronic structure information to the transport properties.

Scanning Transmission Electron Microscopy (STEM) in combination with Electron Energy Loss Spectroscopy (EELS) allows for spatially resolved, chemically sensitive investigations of oxide interfaces [3]. We used a Nion 5th-order aberration corrected 100 keV dedicated STEM (α_{max} = 33 mrad) to collect spectroscopic images of four LAO / STO interfaces with La/Al ratios ranging from 0.91 ± 0.02 (conducting) to 1.06 ± 0.02 (insulating). With both high spatial resolution (~1Å) and high usable collected beam current (nearly 300 pA), the two dimensional distribution of all atomic species—La, Al, O, Sr and Ti—can be extracted in minutes.

By directly comparing the spectroscopic maps of both insulating and conducting samples, we are uniquely poised to distill the microscopic characteristics that correlate with transport properties from those that do not. Notably, neither the conducting nor the insulating samples showed atomically abrupt interfaces, with interdiffusion over a few unit cells at the interface; the most diffuse interfaces showed a La concentration in the substrate dropping to < 1% by 5 u.c. and < 0.001% by 8 u.c. As shown in Fig. 1, there is no correlation between the degree of La interdiffusion at the interface and the transport properties. However, by collecting all atomic species present, we can determine the total concentration of the A-site cations, Sr and La, and the B-site cations, Ti and Al (Fig. 2) across the interface. We find a dip in the total B site cation concentration across the interface in the insulating samples not present in the conducting samples; neither series showed a dip in the total A-site cation concentration. This dip in the B-site cation concentration in insulating films can mitigate the polar catastrophe at the interface without a corresponding electronic reconstruction, and hence without free carriers. [4]

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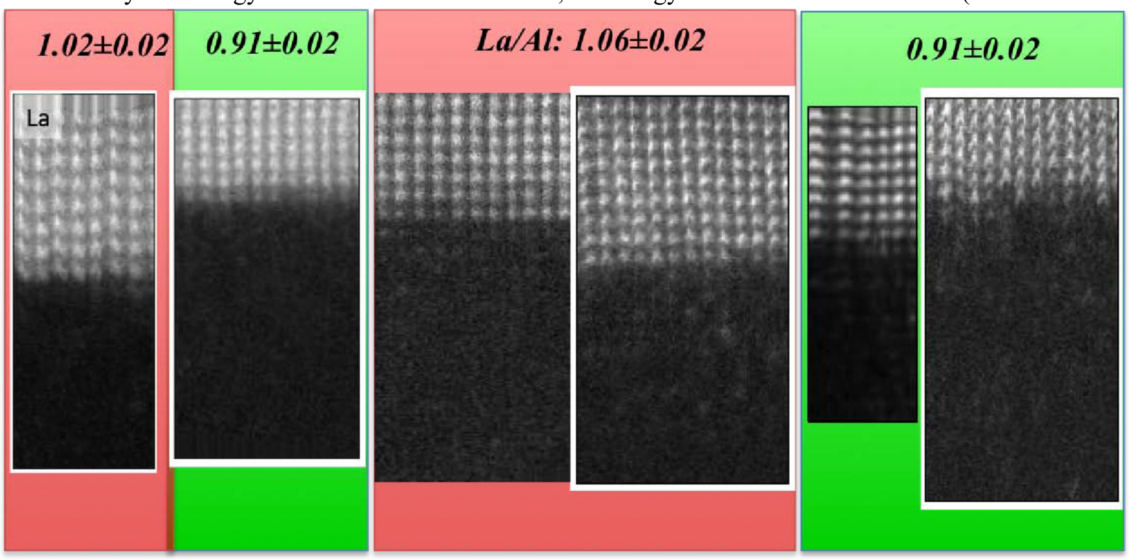

Figure 1. Spectroscopic maps of the La in $LaAlO_3/SrTiO_3$ interfaces. The maps are arranged in order of apparent La interdiffusion and labeled by the La/Al ratio. Samples with La/Al > 1 in red showed insulating interfaces while the others in green were conducting. As shown, there is no correlation between the La interdiffusion and the transport properties.

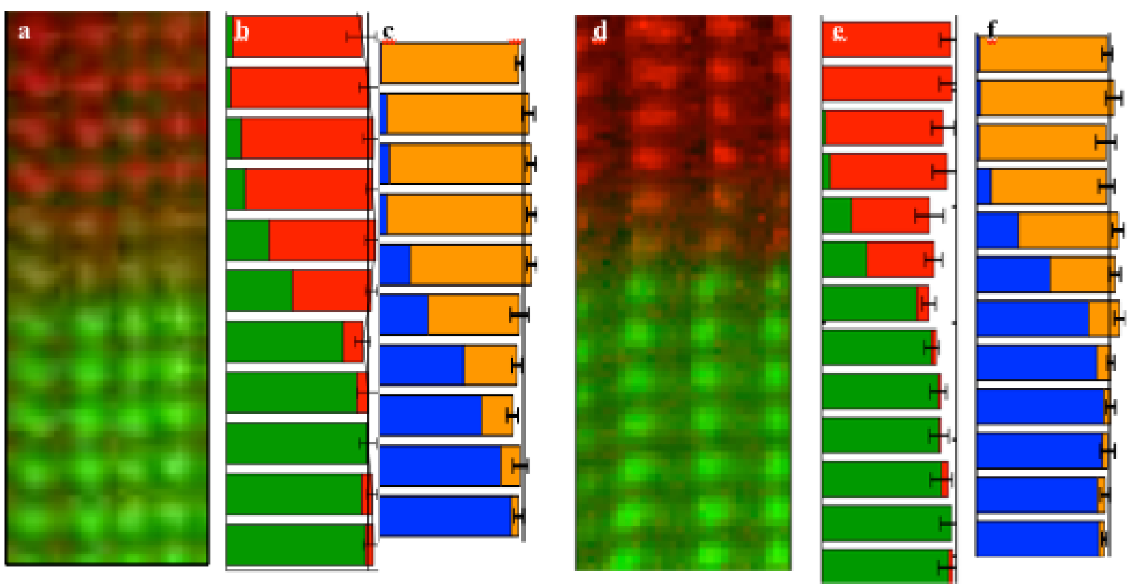

Figure 2. Comparison of the total A-site and B-site concentration a conducting (a-c) and insulating (d-f) sample. The spectroscopic maps (a and c) plot the B-site concentration Al in red and Ti in green. The bar plots (b and e) show the total concentration of the B-site cation at each atomic plane, with a dip in the concentration for only the insulating sample (e). The A-site concentration (c and f) shows the corresponding La concentration in gold and Sr in blue.

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