

Type of presentation: Poster

IT-11-P-2687 Direct Imaging of Two-Dimensional Electron Gas at Oxide Interfaces using Inline Electron Holography

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Recently, a variety of new physical properties and phenomena have been discovered to emerge at atomically engineered interfaces of complex oxide systems. One example is the two-dimensional electron gas (2-DEG) forming at the interface between two insulating perovskite oxides, LaAlO₃ (LAO) and SrTiO₃ (STO). Theoretically, the electron concentration at this atomically-controlled interface can be manipulated by means of the polarity-induced electric field, which is facilitated by simply changing the film thickness of LAO on a STO substrate. The resulting conducting "interface material" is known to be localized within a few nm from the interface. Although the existence of 2-DEG has been proved and utilized in many prototype devices, there are still compelling debates related to the origin, spatial distribution, and electrostatic compensation of this "interface material". Here, we directly visualize and quantify the 2-DEG forming at the interface of LAO/STO by using inline electron holography. By combining electron energy loss spectroscopy (EELS) and quantitative measurements of the atomic displacement of cations the possible origin of 2-DEG will be discussed.

A wide area 2-D charge density map with sub-nanometer resolution (~0.8 nm) was obtained by applying a Laplacian image filter to the electrostatic potential map as shown in Fig. 1. The electrostatic potential maps were retrieved by carefully calibrating the mean inner potentials and local thicknesses of LAO and STO. Whilst the charge density map obtained from the 3 unit cell (u. c.) sample, which is below the known critical thickness of 4 u. c., seems not to host any significant charge density near the interface (Fig. 1a), the 10 u. c. sample exhibits the negative charges beneath the interface (Fig. 1b). The width of 2-DEG, measured at full width at half maximum, is 0.82 ± 0.34 nm. In order to extract a correct density value of 2-DEG from the total charge density map, one has to take account of a change of the dielectric constant of STO near the interface due to a large intrinsic electric field. The calibration of the dielectric constant using Landau theory yields a 2-DEG density close to the theoretical expected value of $\sim 3.3 \times 10^{14}$ e cm⁻² corresponding to the transfer of 0.5 e per unit cell. The scanning transmission electron microscopy (STEM) analysis combined with EELS indicates that the origin of this interfacial 2-DEG is most likely related with the oxygen vacancies formed at the LAO surface (Fig. 3), which agrees well with the recent first principles calculations.

Acknowledgement: This works has been supported by the AFOSR under Grant numbers FA2386-13-1-4136 and FA9550-12-1-0342.

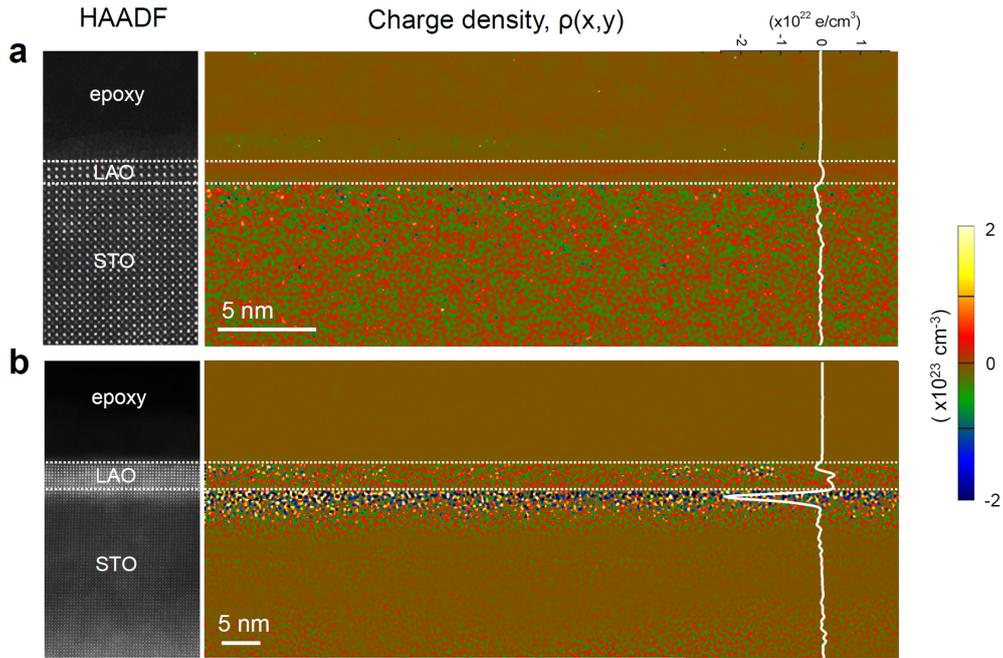


Fig. 1: Charge density maps across the $\text{LaAlO}_3/\text{SrTiO}_3$ interfaces obtained by using inline electron holography for: a. 3 u. c. and b. 10 u. c. LaAlO_3 samples.

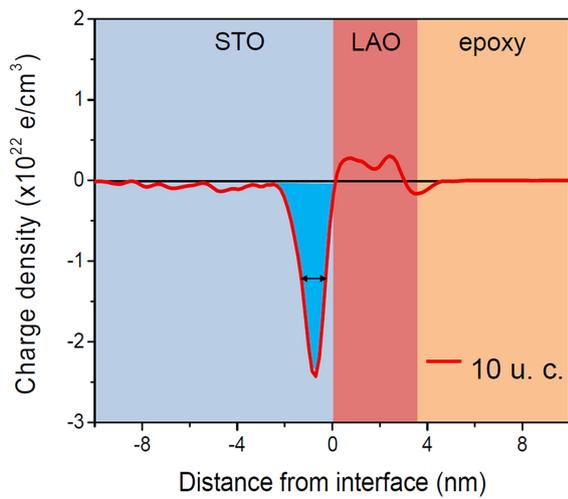


Fig. 2: Charge density profile extracted from the charge density map of 10 u. c. $\text{LaAlO}_3/\text{SrTiO}_3$ shown in Fig. 1b.

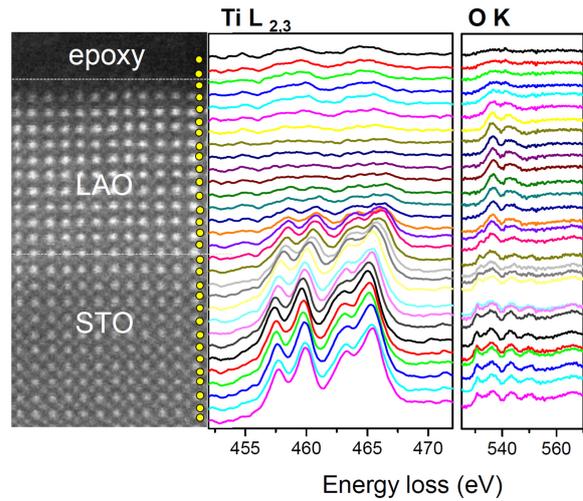


Fig. 3: High-angle annular dark field (HAADF) image of a 10 u. c. $\text{LaAlO}_3/\text{SrTiO}_3$ and EELS spectra of Ti- $L_{2,3}$ and O-K edges obtained from 2D line scans.