Materials Science & Engineering Doctoral Defense

Transmission Electron Microscopy Study of the Two-Dimensional Electron Gas at SrTiO3-Based Oxide Interfaces

School for Engineering of Matter, Transport and Energy

Sirong Lu

Advisor: Dr. David J. Smith

abstract

The two-dimensional electron gas (2DEG) at SrTiO3-based oxide interfaces has been extensively studied recently for its high carrier density, high electron mobility, superconducting, ferromagnetic, ferroelectric and magnetoresistance properties, with possible application for all-oxide devices. Understanding the mechanisms behind the 2DEG formation and factors affecting its properties is the primary objective of this dissertation. Advanced electron microscopy techniques, including aberration-corrected electron microscopy and electron energy-loss spectroscopy (EELS) with energy-loss near-edge structure (ELNES) analysis, were used to characterize the interfaces. Image and spectrum data-processing algorithms, including subpixel atomic position measurement, and novel outlier detection by oversampling, subspace division based EELS background removal and bias-free endmember extraction algorithms for hyperspectral unmixing and mapping were heavily used. Results were compared with density functional theory (DFT) calculations for theoretical explanation. For the γ-Al2O3/SrTiO3 system, negative-Cs imaging confirmed the formation of crystalline γ-Al2O3. ELNES hyperspectral unmixing combined with DFT calculations revealed that oxygen vacancies, rather than polar discontinuity, were the key to the 2DEG formation. The critical thickness can be explained by shift of the Fermi level due to Ti out diffusion from the substrate to the film. At the LaTiO3/SrTiO3 interface, aberration-corrected imaging showed crystallinity deterioration in LaTiO3 films a few unit cells away from the interface. ELNES showed that oxygen annealing did not alter the crystallinity but converted Ti3+ near the interface into Ti4+, which explained disappearance of the conductivity. At the EuO/SrTiO3 interface, both high-resolution imaging and ELNES confirmed EuO formation. ELNES hyperspectral unmixing showed a Ti3+ layer confined to within several unit cells of the interface on the SrTiO3 side, confirming the presence of oxygen vacancies. At the BaTiO3/SrTiO3 interface, spontaneous polarization and local lattice parameters were measured directly in each unit cell column and compared with oxidation state mapping using ELNES with unit-cell resolution. The unusually large polarization near the interface and the polarization gradient were explained by oxygen vacancies and the piezoelectric effect due to epitaxial strain and strain gradient from relaxation.

April 13, 2018; 1:30 PM; PSF 306