

How does a large electric field change the structure and chemistry of an ionic liquid - solid interface?

Room temperature ionic liquids are widely used as solvents and electrolytes. Their low vapor pressure and excellent stability make them particularly attractive for electrolyte gating, a technique where the electronic properties of a surface can be modified by immersing it in electrolyte and applying a potential. Despite widespread use of ionic liquids for electrolyte gating in recent years, including to induce superconductivity in otherwise insulating materials and to tune across metal to insulator transitions, little is known about the structure and chemistry of the interface between the ionic liquid and the electrode surfaces.

By investigating electrolyte gated gold and strontium titanate surfaces, I found that 1) impurities in the ionic liquids can react to form passivating layers that are responsible for the change in electronic properties and 2) the ions in the liquid form a complex layered structure that changes with applied potential.

In gold films, electrolyte gating causes large changes in sheet resistance. The widely accepted mechanism for these changes is the formation of an electric double layer with a charged layer of ions in the liquid and accumulation or depletion of carriers in the thin film. Based on work at beamline 11-2, I demonstrated that a different mechanism is responsible. In particular, I used x-ray absorption near edge structure (XANES) to show that the previously reported large conductance modulation is due to reversible oxidation and reduction of the surface, rather than the charging of an electric double layer. These results represent a significant step towards understanding the mechanisms involved in electrolyte gating.

In strontium titanate, a band insulator, electrolyte gating induces a conductive surface layer. By measuring reflectivity and crystal truncation rods at beamline 7-2, I showed that the ions in the liquid form distinct, well-ordered layers near the crystal surface. With increasing applied potential, these layers extend further into the liquid and become more strongly polarized, the separation between layers increases (suggesting that the cations rotate), and excess cations accumulate at the surface.