Mesoscale Grand Challenge or Priority Research Direction

Ionic Liquid at Interfaces and Under Confinement

Opportunity

Ionic liquids (IL) are composed of two ionic molecules of opposite charge where Coulombic and van der Waals interactions are both significant. ILs possess high ionic conductivity, thermal stability, and a wide electrochemical window and are expected to replace conventional electrolytes in many applications. Their potential-dependent mesoscale structural properties, are important to advances in the field.

Approach

A successful approach to understanding *surface-induced* mesoscale and intermolecular structure in Ionic Liquids requires both atomically flat interfaces and patterned substrates. The latter will *increase* the surface to bulk scattering, thus enabling reliable measurements on the potential-induced order for the first time. These structural and dynamics studies demand synchrotron and neutron-based scattering methods.

Meso Challenge

A key challenge is to understand how interface and nano-confinement affects the mesoscale and intermolecular order of ILs under potential control and the influence of the molecule's internal structure including the specific choice of the ion or alkyl ligands.

Impact

Understanding the mesoscale structure and the physics of confinement will allow scientists to tailor the design of IL's for efficient conversion and storage of electric energy in devices such as batteries, photovoltaics and supercapacitors.

References: Themed issue on "Interfaces of Ionic Liquids," *Phys. Chem. Chem. Phys.* **14**, 4993-5308 (2012).
E. W. Castner and J. F. Wishart, Spotlight on ionic liquids. *J. Chem. Phys.* **132**, 120901 (2010).
M. Mezger, H. Schröder, H. Reichert, S. Schramm, J. S. Okasinski, S. Schöder, V. Honkimäki, M. Deutsch, B. M. Ocko, J. Ralston, M. Rohwerder, M. Stratmann & H. Dosch, Molecular Layering of Fluorinated Ionic Liquids at a Charged Sapphire (0001) Surface. *Science* **322**, 424-428 (2008).

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Ionic Liquid at Interfaces and Under Confinement (Ben Ocko)

Possible figure for the report, additional text can be provided by request

Although the molecular size of each ionic liquids molecule is Å in size, the subsequent interfacial ordering extends over the mesoscale and is expected - currently no experimental data - to depend on the applied potential. In (A) a sketch of the ionic liquid [bmpy]+[FAP]− , cation (red) and the anion (blue) is shown. X-ray reflectivity (XR) measurements, carried out at the solid/liquid interface with sapphire at 72.5 keV, show that the interfacial structure more closely resembles the double layer model (B) rather than an alternative, checkerboard model. The best fit to the XR reveals surface induced, layered density profile, where the electron densities are shown in (C) for the cations (red), anions (blue), and total (black) electron. From M. Mezger, H. Schröder, H. Reichert, S. Schramm, J. S. Okasinski, S. Schöder, V. Honkimäki, M. Deutsch, B. M. Ocko, J. Ralston, M. Rohwerder, M. Stratmann & H. Dosch, Molecular Layering of Fluorinated Ionic Liquids at a Charged Sapphire (0001) Surface. *Science* **322**, 424-428 (2008).