Chemical interfaces contain unique molecular architectures that contribute to important processes including surfactancy, self-assembly, and heterogeneous reactivity. However, specific relationships between intermolecular interactions and the observed interfacial behaviors remain poorly defined. This is particularly true of soft, high vapor pressure, dynamic interfaces which are challenging or impossible to investigate with ultra-high vacuum techniques. Research in the Shaw group focuses on making analytical measurements at these chemical interfaces that are relevant to energy, medical, and environmental fields. This talk will introduce our approach to studying interfacial measurements, describe spectroscopic data showing unique, long-range ordering behaviors of ‘interfacial’ molecular and ionic liquids, and present results from electrochemical studies as a method to perturb and control the observed interfacial behaviors.