By exploiting the strong UV charge-transfer-to-solvent (CTTS) resonances of selected anions in aqueous electrolytes, their interfacial adsorption properties are measured by UV-SHG spectroscopy. Temperature and concentration dependences are determined, with the goal of establishing a molecular description of selective ion adsorption. A study of prototypical chaotrope thiocyanate reveals that its strong adsorption is driven by enthalpic forces and impeded by entropy. A study of nitrite indicates even stronger adsorption as an ion pair with sodium. Evaporation rates are measured by combining liquid microjet technology and Raman thermometry. The relationship between surface propensities of ions and evaporation rates is investigated. A detailed molecular mechanism for aqueous evaporation is sought.

