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Organization at the Air-Aqueous Interface by Heterodyne-detected Phase-Sensitive Sum Frequency Spectroscopy[1]

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Water and ions organize at the air – aqueous interface and the ion distributions within this region give rise to interfacial electric double layers. Geochemical solid-aqueous and atmospheric aerosol relevant air-aqueous interfaces were studied using vibrational sum frequency generation (VSFG), and in some cases, heterodyne-detected VSFG spectroscopy. Solid-aqueous and air-liquid interfaces were also investigated using total internal reflection Raman spectroscopy, infrared reflection absorption spectroscopy (IRRAS), and Brewster angle microscopy (BAM), respectively. Here, we show results from aqueous solutions containing salts such as sulfates and chlorides where surface adsorption and electric field direction reversal was observed. Of the salts studied at the air-aqueous interface with heterodyne-detected VSFG, the magnitude of the electric field in the surface extending to the subsurface regions decreases in the order: $(NH_4)_2SO_4 > Na_2SO_4 > Na_2CO_3 \geq CaCl_2 > NaCl$; the electric field is opposite in direction for the sulfates and carbonate relative to the chloride salts.

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