ION ORGANIZATION AND REVERSED ELECTRIC FIELD AT AIR/AQUEOUS INTERFACES REVEALED BY HETERODYNE-DETECTED SUM FREQUENCY GENERATION SPECTROSCOPY

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Sum frequency generation (SFG) is a second order optical spectroscopy that probes regions of non centrosymmetry, interfaces, and allows for the understanding of molecular organization at air/aqueous interfaces. An overview of our work in this area is presented with emphasis on phase-sensitive SFG (PS-SFG) spectroscopy. PS-SFG is a variant of SFG and is used in our laboratory to investigate the average direction of the transition dipole of interfacial water molecules. The orientation of water at air/aqueous inorganic salts interfaces of $CaCl_2$, NaCl, Na_2SO_4, $(NH_4)_2SO_4$, and Na_2CO_3 is inferred from the direct measurement of the transition dipole moment. We find that charge separation at the air/water interface is most obvious for the aqueous ammonium sulfate solution where the local electric field has a greater magnitude at this interface relative to the other salt solutions. 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.