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Molecular Beam Studies of Gas Transport and Reactions at the Surface of Surfactant-Coated Sulfuric Acid

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Recent studies have demonstrated that reactivity and microscopic structure of liquid surfaces can be explored by proper modifications of the conventional techniques employed in solid-state surface chemistry. In the first part of this talk, I will briefly introduce some important examples of these studies. Then, in the main part, I will focus on a recent study of molecular beam scattering at surfaces of supercooled sulfuric acid. Gas-liquid scattering experiments under high vacuum condition offer the opportunity to investigate heterogeneous reactions that alter ozone levels in the lower stratosphere and upper troposphere. These reactions are catalyzed by supercooled sulfuric acid aerosols, which provide a medium for the trapping, protonation, and dissociation of ambient gases such as H_2O, HCl, and HBr. Field studies suggest that aerosol particles just below the stratosphere are often coated with organic molecules. We investigated the effects of these surface films on gas transport and interfacial reactions by directing a beam of HX molecules at surfactant-coated D_2SO_4/D_2O solutions in vacuum. The fraction of molecules undergoing H/D exchange and the solvation times of the desorbing HX and DX species are monitored by time-of-flight and pulsed-beam measurements. These experiments reveal the pathways and time scales for gas permeation through the surfactant and for reactions with the surfactant head groups.