In Situ Spectroscopic Identification of Molecules at Aerosol Interfaces

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The interfacial roles of aerosols in chemical processes in the atmosphere have long been recognized. However, *in situ* interface-specific spectroscopy is lacking. Based on our recent success in *in situ* second harmonic scattering, we continue to develop an interface- specific electronic sum frequency scattering to directly probe electronic spectra of molecules at laboratory-generated aerosol interfaces. We successfully probed interfacial electronic spectra of organic molecules on aerosols floating in the gas, which are different than those in bulk solution. We found that the surface driving force for the curved aerosol surfaces is less than that for the planar air/water interface. These results challenge the long-standing wisdom that aerosol surfaces facilitate stronger surface binding of molecules than the air-water interface. Furthermore, polarized electronic sum frequency scattering experiments show orientational configurations of organic molecules at the aerosol interfaces. Our findings would open a new avenue for the studies of chemical and physical properties of environmental aerosols.