



Raman spectroscopy as a probe for the quasi-liquid layer at the ice surface

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Raman spectra of the water OH-stretch were acquired at air-ice and air-water interfaces using a novel glancing-angle technique. The shapes of the OH-stretching bands at solid and liquid aqueous interfaces indicate that water molecules at the air-ice interface are less strongly hydrogen bonded than they are in either bulk ice or liquid water, and that hydrogen bonding of water molecules at the air-water interface is weaker than that at the air-ice interface. Spectra measured at the surface of dodecane at high and low relative humidities indicate that this method is sensitive to fewer than fifty monolayers of water.

Modifications to the local environment of the surficial water molecules are manifested as changes in the shape of the OH-stretching band, namely shifts in the wavelength of the peak intensity and changes in the FWHM. Hydrogen bonding at the ice surface is more sensitive to temperature than it is either in supercooled water or in bulk ice. The presence of sodium chloride, which disrupts hydrogen bonding in liquid water, shows the same effect at the air-ice interface. The presence of hydrochloric acid, however, shows the opposite effect: It appears to increase the extent of hydrogen bonding at the ice surface. The presence of nitric acid and ammonium hydroxide do not affect the shape of the OH-stretching band.