

Sum Frequency Generation by Water on Supercooled H₂SO₄/H₂O Liquid Solutions at Stratospheric Temperature

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Sum frequency generation spectra indicate that the surface water of liquid sulfuric acid solutions varies as a function of concentration, but not with temperature. At 0.1x H₂SO₄ (where x = mole fraction, 38 wt%), subsurface ionic species orient surface water molecules. The surface of 0.2x H₂SO₄ (58 wt%) solution, however, features H₂SO₄/H₂O complexes both at 273 K and supercooled at 216 K. The results support a picture of stratospheric chemistry in which sulfuric acid aerosols are coated with hydrogen-bonded water/sulfuric acid complexes.