

**The interfacial structure of water droplets in a hydrophobic liquid**

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Nanoscale and microscopic water droplets and ice crystals embedded in liquid hydrophobic surroundings are key components of aerosols, rocks, oil fields and the human body. The chemical properties of such droplets critically depend on the interfacial structure of the water droplet. Here, we report the surface structure of 200 nm sized water droplets in mixtures of hydrophobic oils and surfactants as obtained from vibrational sum frequency scattering measurements. The interface of a water droplet shows significantly stronger hydrogen bonds than the air/water or hexane/water interface and previously reported planar liquid hydrophobic/water interfaces at room temperature. The observed spectral difference is similar to that of a planar air/water surface at ~50 K lower temperature. Supercooling the droplets to 263 K does not change the surface structure. Below the homogeneous ice nucleation temperature a single vibrational mode is present with a similar mean hydrogen bond strength as for a planar ice/air interface.

[1] Nikolay Smolentsev, Wilbert J. Smit, Huib J. Bakker, Sylvie Roke, *Nature Communications*, **2017**, 8, 15548.