Supplementary Information for Manuscript

No ice-like water at aqueous biological interfaces

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Supplementary Figure S1 shows that the double-peaked resonance disappears upon isotopic dilution also in the O-H stretch region. The reported spectra show the water response underneath a net neutral, zwitterionic 1,2-dipalmitoyl-*sn*-glycero-3-phosphocholine (DPPC) monolayer at a surface pressure around 25 mN/m. A more marked simplification of the vibrational response has recently been reported for a charged monolayer in Ref. [17].

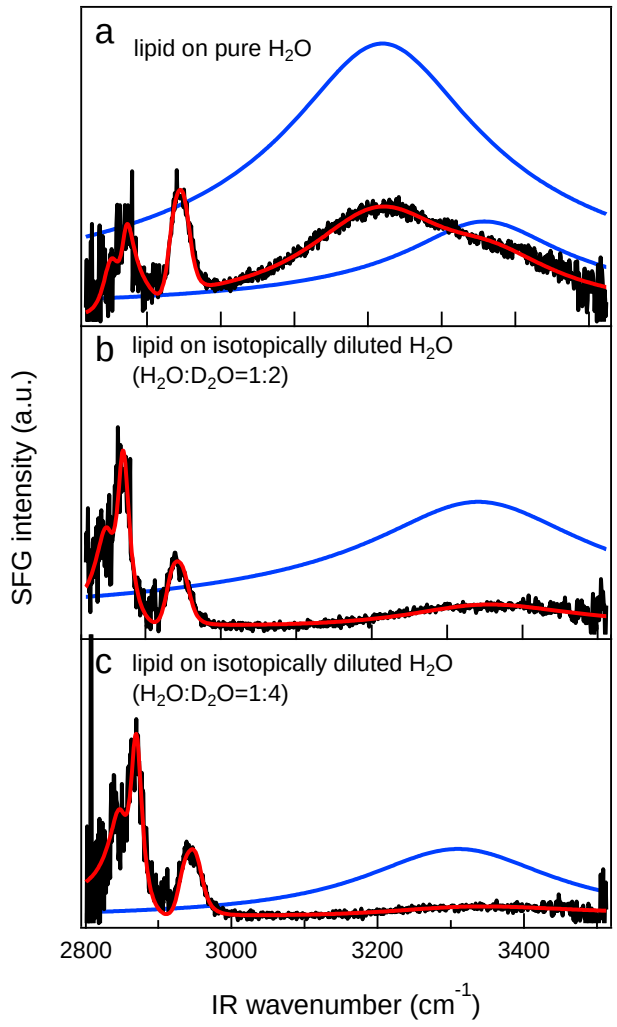

Figure S1: SFG spectra in the O-H (3100-3500 cm^{-1}) and C-H (2800-3000 cm^{-1}) stretch region of pure water H_2O (a) and isotopically diluted HDO (b,c) underneath a lipid (DPPC) monolayer. For the pure water case, a peak and a shoulder are observed at ~3200 and 3400 cm^{-1}, respectively. These peaks collapse onto one peak upon isotopic dilution. The red lines illustrate fits to the spectra, and the thin blue lines represent the different contributing fit components of the O-D stretch vibrations.