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Maxima in supercooled water's thermodynamic response and correlation functions using x-ray free electron laser

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Water is one of the most critical substances on earth for life. Many important biological and geological reactions important for life take place in water. It has anomalous properties like increased specific heat capacity and isothermal compressibility on cooling at ambient conditions. These become more pronounced on supercooling. These thermodynamic properties are a result of the unique H-bonding structure in liquid water. Despite its importance, there is still debate about the intermolecular structure of water. The experimental challenge to supercool water below its homogenous freezing limit (≈233 K at 1 atm) has meant that there is little data at such supercooled conditions. Ours was the first research groups to cool bulk water below its homogenous freezing limit. We have an experimental set-up where we evaporate ≈15 µm diameter water droplets in vacuum. The resulting evaporative cooling enables us to reach temperature as low as ≈227 K with still around 1% of droplets remaining unfrozen. We study these droplets with femtosecond x-ray pulses and measure the scattering pattern by 'diffract before destroy' technique. In this study, we measure the small angle x-ray scattering (SAXS) of water where the structure factor of water, S(q) shows an anomalous increase as the scattering vector, q ⟶0. This behavior becomes more pronounced on supercooling. The isothermal compressibility is directly proportional to S(q=0). The correlation length in liquids is the damping factor in the asymptotic decay in the pair-correlation function. According the Ornstein-Zernike theory, the correlation length is proportional to the slope of S(q) in the SAXS region. Thus, we can measure both the isothermal compressibility and the correlation length from SAXS. As we supercool water from ambient conditions, we observe that the isothermal compressibility and correlation length increase with decreasing temperature until it reaches a maxima around 229 K and then decreases with decreasing temperature. A similar behavior was observed for D_2O where we observe the maxima around 233 K. In addition to these observations, we also observe that the liquids undergo an accelerated increase in the tetrahedral structures with decreasing temperature and this rate (with respect to temperature) reaches a maximum at similar temperatures. All these observations indicate to the first experimental evidence of a Widom line which is defined as the locus of points in the Pressure-Temperature surface where correlation length reaches a maximum. The difference between the maximum values for H_2O and D_2O indicate the importance of nuclear quantum effects.

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