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Amorphous ices and liquid states

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The amorphous forms of water play an important role in the understanding of water's anomalous properties. Computer simulations suggest that the anomalous behaviour of ambient and supercooled water could be explained by a two state model of water [1]. Since the discovery of two distinct amorphous states of ice with different density (high- and low density amorphous ice, HDA and LDA) it has been lively debated whether and how this phenomenon of polyamorphism at high pressures is connected to the occurrence of two distinct liquid phases (HDL and LDL) [1]. Alternatively, amorphous ices have been suggested to be of nanocrystalline nature, unrelated to liquids.

The glass transition in both amorphous states, LDA and HDA, was investigated using different experimental techniques [2] and discussed controversially over the last years [3]. Our recent results using X-ray diffraction as well as X-ray correlation spectroscopy (XPCS) support the previous findings of HDA undergoing a glass-liquid transition at ambient pressure around 110 K and are consistent with the hypothesis of a liquid-liquid transition between HDL and LDL [4].

[1] P. Gallo, K. Amann-Winkel et al., Water a Tale of Two Liquids, Chem. Rev. 116, 7463-7500 (2016)

[2] K. Amann-Winkel, C. Gainaru et al., Water's second glass transition, PNAS 110, 17720 (2013)

[3] K. Amann-Winkel et al., Water's controversial glass transition, Rev. Mod. Phys. 88, 0110002 (2016)

[4] F. Perakis, K. Amann-Winkel et al., Diffusive dynamics during the high-to-low density transition in amorphous ice, PNAS 114, 8193 (2017)

Significance statement

A liquid-liquid transition in water terminating in a hypothesized second-critical point is discussed controversially. Our x-ray experiments on water and amorphous ice shed light on this important debate.

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