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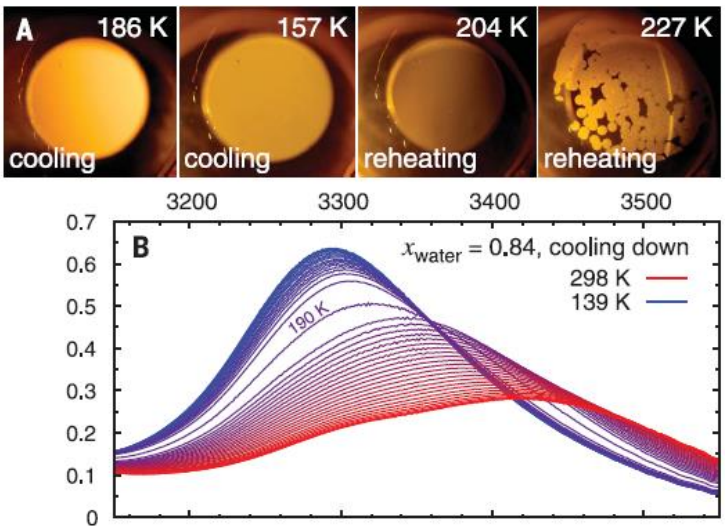
Is there a liquid-liquid transition in ideal aqueous solutions? Raman and x-ray study

An hypothesis proposed for water more than 20 years ago [1] is still highly debated [2]. Water might exist in two liquid states, which would differ by the local arrangement of the molecules. However, the two liquids would separate only at very low temperature, and thus, up to now, experiments were not able to detect this transition, despite record-low temperatures reached with liquid water (-43°C [3,4]).

A workaround strategy consists in mixing a solute with water to hinder crystallization and reach higher degrees of supercooling. However, the relevance of such studies to pure water is not straightforward because of the strong non-ideal interactions between water and solute. Recently, Austen Angell and his colleagues [5] have found a solute which makes ideal solutions with water, and allows them to cool all the way to 139 K without freezing. They have detected a heat capacity peak and an abrupt change (occurring within a few K) in the infrared spectra. They interpret these findings as a proof of a first-order transition between two liquid phases. Because of the ideal nature of the solutions, this would directly mirror the putative liquid-liquid transition in pure water.

Our project is to investigate further this possibility by complementary techniques. One is Raman spectroscopy, available in our laboratory, and the other is wide angle x-ray scattering, for which we have applied for beamtime in a synchrotron facility. We wish to check if the observed changes are not artifacts due to partial crystallization of the sample, and to obtain structural information on the phases which is currently missing.

This work can be extended during a PhD by investigating other phenomena in metastable liquids (at negative pressure, supercooled or supersaturated), in our lab and with our partners abroad.



Optical observation of the solutions and infrared spectra recorded during cooling [5].

[1] Poole *et al.*, *Nature* **360** 324 (1992)
[2] Gallo *et al.*, *Chem. Rev.* **116** 7463 (2016)
[3] Sellberg *et al.*, *Nature* **510** 381 (2014).
[4] Goy *et al.*, *Phys. Rev. Lett.* **120** 015501 (2018).
[5] Woutersen *et al.*, *Science* **359** 1127 (2018).