Nature of Amorphous Polymorphism of Ice

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The phenomenon of amorphous polymorphism, i.e. the existence of at least two different disordered states in a single substance, has been observed for the first time in solid water, where a high-density amorphous (HDA; m/V_{H2O} = 1.17 g/cm^3) and a low-density amorphous (LDA; m/V_{H2O} = 0.93 g/cm^3) phase can be prepared at T = 77 K [1]. Despite a remarkable progress comprising the comprehension of properties of this phenomenon the most essential question, namely 'What is the origin of amorphous polymorphism in water?', could not be answered unequivocally, yet.

The complexity of the amorphous polymorphism has been enhanced recently by the detection of a third distinct amorphous structure called very high-density amorphous (vHDA) ice with a mass density of m/V_{H2O} = 1.25 g/cm^3 [2]. Amazingly, the three amorphous phases vHDA, HDA and LDA can be reversibly converted into each other depending on the thermodynamic conditions. This behaviour indicates that, despite the apparent metastable character of the amorphous structures, their existence must be based on a generic thermodynamic principle.

In this talk, we focus on the relationship between vHDA, HDA and LDA. Based on results from elastic and inelastic neutron and x-ray scattering experiments we show that in terms of thermodynamics only two amorphous states exist in water. The transformation between these two states can be interpreted as a first order transition with an activation energy of about 40 kJ/mol, the energy of two H-bonds [3].

References

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