Hydrogen-bond symmetrization in methane and hydrogen hydrates in the Mbar range

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It is generally accepted that nuclear quantum effects play a crucial role in the transition to the nonmolecular form of ice by decreasing the transition pressure from 90 to approximately 60 GPa.[1] On the other hand, recent studies[2] on LiCl- and NaCl-doped ice have shown that the presence of salt inclusions in the ice lattice pushes the hydrogen-bond symmetrization to higher and higher pressures as the concentration of salt is increased. In other words, the incorporation of salt in high-pressure ice suppresses the quantum behaviour of protons.

This finding stimulates the investigation of similar effects in other water-based compounds, namely a field which remains largely unexplored. High-pressure studies on gas hydrates offer the possibility to explore nuclear quantum effects in network topologies for the water molecules which are different from that of ice VII. Some experiments have been performed in the past to identify signatures of the hydrogen-bond symmetrization in methane and hydrogen hydrates without reaching conclusive results.[3] We will present recent work on the hydrogen-bond symmetrization of methane and hydrogen hydrates using Raman scattering in the Mbar range and semiclassical simulations including nuclear quantum effects.

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3. Tanaka T. et al., *Phase changes of filled ice Ih methane hydrate under low temperature and high pressure*, J. Chem. Phys. 139, 104701, 2013 ; Hirai H. et al., *Structural changes of filled ice Ic hydrogen hydrate under low temperatures and high pressures from 5 to 50 GPa*, J. Chem. Phys. 137, 074505, 2012