Solid compounds of water and molecular hydrogen: from clathrate hydrate to filled ice

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Under high pressure (over 1 kbar) and low temperature (below 273 K), water and molecular hydrogen form several solid phases, namely (in order of increasing pressure) clathrate hydrate [1,2] and filled ices [3]. In these compounds, the guest molecule (H_2) is trapped in a water molecule skeleton without involving any chemical bond. These non-stoichiometric compounds have attracted some interest as materials for hydrogen storage, but they are interesting also because of the nano-confinement of the hydrogen molecule in the water structure.

For many of these phases the crystalline structure and the quantum dynamics of the H_2 molecule have been studied with different probes [3-5]. However some aspects like the nucleation and growth [6], the mechanism of diffusion of the guest molecules [7] and the stability of the empty host structure [8] are still today subject to debate. Furthermore only recently thermodynamic studies have demonstrated the presence of a new stable phase, named C_0, in the hydrogen-water system. Its structure is not completely determined [9,10] and until today there isn't a complete dynamic characterization.

Our experimental work aims to clarify these unresolved aspects about clathrate hydrate and filled ice, in particular for the poorly known C_0 phase. We synthesize these high pressure compounds in our laboratory and then we are able to perform *in situ* Raman spectroscopy or to recover them at ambient pressure for other type of analysis. The structure of our sample will be measured by means of neutron and X-ray diffraction, while an efficient study of molecular vibrations and diffusive motion of the guest molecule will be performed by means of Raman scattering and inelastic/quasi-elastic neutron scattering.

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