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X-ray Absorption Spectroscopy and Electronic Structure Calculations of Water and Ice

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We have investigated the electronic structure of water and ice using a combination of experimental and theoretical techniques [1]. Measurements have been performed on the liquid using both X-ray Absorption (XAS) and X-ray Raman Spectroscopy. The spectrum of the liquid is distinctly different from that of tetrahedrally coordinated bulk ice, where the liquid shows a distinct pre-edge feature and a strong enhancement of the intensity at the edge. Through spectrum simulations and model experiments (bulk and surface of ice) we show that the specific features in the liquid spectrum are due exclusively to asymmetric configurations with only two strong hydrogen bonds: one donating and one accepting [1]. Here I will discuss the interpretation of the spectra on the basis of theoretical modeling using both cluster and periodic plane-waves techniques together with the various approximations involved. In [1] the appearance of the specific features in the liquid spectrum was shown to be represented by a cone-criterion for the donating H-bonds: one with the accepting molecule inside and the other outside the cone. This is a necessary, but not sufficient criterion to predict the pre-edge. From a very large number of models studied we can derive more stringent conditions on the local structure, which indicate more directly what type of configurations that are under-represented in dynamics simulations compared with experiment. Compton scattering measures the electronic momentum distribution. It is sensitive to intermolecular H-bond distances and the associated intramolecular O-H bond changes. I will discuss the theoretical implementation of this spectroscopy and show how it can be used to obtain further insight into the local structure of the liquid. [1] Wernet et al, Science **304**, 995-999 (2004)