Hydrogen bonding configurations in water and aqueous solutions from X-ray spectroscopy

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The question of the structure of the hydrogen bonding network in water has been discussed intensively for over 100 years and has not yet been resolved. Due to the lack of detailed experimental information many theories exist, and in particular results from theoretical molecular dynamics (MD) simulations have dominated the field in the last 20 years. In order to gain new unique information regarding the internal structure of the H-bond network in water, it is essential to develop new techniques, both experimental and theoretical.

We have recently devoted a major effort to the development of x-ray spectroscopy measurements of water in the different aggregation forms and adsorbed on surfaces. Using x-ray absorption spectroscopy (XAS) and x-ray Raman scattering (XRS) together with density functional theory (DFT) calculations we have demonstrated the appearance of specific spectral features that can be related to asymmetric H-bond configurations[1,2]. These can be seen at the surface of ice, as water clusters adsorbed on metal surfaces and in the liquid phase showing the existence of local structures with broken H-bonds. The surprising result for the liquid phase is the large number of broken or weak H-bond species compared with the established wisdom based on MD simulations which challenges many of the existing water theories. It has generated a large interest world wide with many news articles in the public domain and has caused a considerable debate in the physical chemistry community. In the present contribution new results will shown on X-ray Raman measurements of heavy water, EXAFS of water using X-ray Raman and various measurements of water in different model systems. The proposed understanding of water based on XAS-XRS will be discussed in the light of results using other techniques.

1. *The Structure of the First Coordination Shell in Liquid Water*
Ph. Wernet, D. Nordlund, U. Bergmann, H. Ogasawara, M. Cavalleri, L.Å. Näslund, T. K. Hirsch, L. Ojamäe, P. Glatzel, M. Odelius, L.G.M. Pettersson, and A. Nilsson
Science **304** (2004) 995.

2. *The X-ray Absorption Spectrum of Liquid Water from Molecular Dynamics Simulations: Asymmetrical Model*
M. Odelius, M. Cavalleri, A. Nilsson and L. G. M. Pettersson
Phys. Rev. B **73** (2006) 024205.