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In situ spectroscopy of shallow absorption edges using hard x-rays

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X-ray absorption spectroscopy (XAS) has proven a powerful tool to study the local atomic and electronic structure of matter, especially in disordered systems where the information given by other structural probes such as x-ray diffraction is limited. As such, XAS has found its way into the different scientific communities from physics, chemistry, materials science, to the life- and geosciences. When studying absorption edges in the keV regime, *in situ* experiments in extreme conditions can be carried out using conventional XAS. This, however, is difficult for elements with absorption edges in the soft x-ray regime.

Here, we present how inelastic scattering from core electrons can be used to study shallow absorption edges under extreme conditions. Non-resonant x-ray Raman scattering (XRS), often also referred to as non-resonant inelastic x-ray scattering (NIXS), is an energy loss technique which enables one to acquire bulk sensitive information from soft x-ray absorption edges using hard x-rays. [1]

As an example, we will demonstrate how the study of the oxygen K-edge of water under high temperature and high pressure can advance the understanding of this highly relevant substance under conditions prevalent in the deep Earth. Besides the general scientific interest in the structure of water across its extensive phase diagram, water plays a key role in heat and mass transfer and in element fractionation processes in the Earth's lithosphere, such as volcanism and ore deposit formation. [2] This is largely due to its polar character and great tendency to dissociate, which makes it a substantially more powerful solvent than other volatiles. [3] The solvent properties of H_2O depend not only on density and dissociation, but also on structural parameters. However, this is poorly understood, particularly at the supercritical conditions within the Earth's interior.

Supercritical conditions were achieved using a hydrothermal diamond anvil cell (HDAC). The combination of a HDAC and XRS is unique as it allows parallel optical monitoring of the sample volume during data acquisition. Changes in the shape of the oxygen K-edge with p and T are interpreted as reflecting changes in the bonding and the local structure. The open question of the role of hydrogen-bonding in supercritical water is addressed by comparison of the measured spectra with results of state of the art *ab initio* calculations from molecular dynamics simulations.

[1] W. Schülke, Oxford University Press (2007). [2] H. Keppler, Nature 380 (1996) 237. [3] C.E. Manning, Earth Planet. Sci. Lett. 223 (2004) 1.