Title: Electronic and magnetic properties of iron hydride under pressure using XAS and XMCD at the Fe *K*-edge: a combined theoretical and experimental study

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Interest for iron hydrides has grown recently because they are model objects for transition metal hydrides - which are promising candidates to address the problem of hydrogen storage[1] - and because hydrogen has become one of the major candidates for the light element present in the Earth's core -with the observation of its solubility in Fe under high pressure conditions. [2]

Iron hydride FeH can be synthetized by application of a 3.5 GPa pressure on Fe in a H_2 environment.[3] We have investigated the transition from *bcc*-Fe to double hexagonal (*dhcp*) FeH and the compression of FeH using a combination of X-ray Absorption Spectroscopy (XAS) and X-ray Magnetic Circular Dichroism (XMCD) at the Fe *K*-edge. XMCD is the difference, for a ferromagnetic material, between the absorption of left and right circularly polarized X-rays. XAS provides element-specific information on the local structure while XMCD gives information on the magnetic properties of the sample. As XAS and XMCD are recorded simultaneously, it allows obtaining the structural and magnetic properties in the same conditions, in particular at the very same pressure, which is a valuable advantage considering the difficulties to know precisely and to reproduce pressure conditions.

Performing the experiments with hard X-rays is mandatory to probe samples at high pressure in Diamond Anvil Cells. Nevertheless, unlike the $L_{2,3}$ absorption edges for which well-established sum rules allow to extract spin and orbital contributions, the quantitative analysis of *K*-edge XMCD spectra is far from straightforward. The crucial support of theoretical interpretations is then required to interpret the experimental results.

We have computed XAS and XMCD spectra under pressure in *bcc*-Fe and *dhcp*-FeH within a monoelectronic framework. Our approach[4] is based on a semi-relativistic DFT calculation of the electron density in the presence of a core hole using plane-waves and pseudo-potentials. The semi relativistic absorption cross-section[5] is calculated with a Lanczos algorithm from the all-electron wave functions reconstructed using PAW projectors.

Our method has been successful to reproduce the experimental spectra and to investigate the magnetic structure of FeH. Moreover, we have identified a transition around 28 GPa, which is a purely magnetic transition from a ferromagnetic state to paramagnetic state.

This combined theoretical and experimental study gives a unique insight into the effect of hydrogen on the electronic and magnetic structure of iron and also on the physical content of XMCD at the *K*-edge. We will discuss these two aspects in detail.

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