ESRF	**Experiment title:** **XMCD in hydrided multilayers $[La_{2+x}/Fe]\times n$ and $[Ce_{2+x}/Fe]\times n$: 5d magnetism and reversible hydrogen loading for $0\leq x<1$**	**Experiment number**: HE-502
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Report:

Rare-earth hydrides REH_x undergo a metal-to-semiconductor transition at $x\approx2.8$, with the accompanying change from high optical reflectivity to transparency for photon energies below ~2 eV. Between $x\approx2$ and $x\approx3$ the hydrogen concentration may reversibly be changed [1]. Progress in the theoretical understanding of these materials, has only recently been made[2]. The ground state of the trihydride ($x=3$) can be viewed as a band insulator due to strong electron correlations which open up a gap between the hydrogen-like valence band and the metal-like conduction band of primarily d symmetry. We have studied the changes of the electronic structure of La in multilayers $[LaH_{2+\delta}/Fe]\times n$ through the metal-to-semiconductor transition by X-ray absorption spectroscopy (XAS) and X-ray magnetic circular dichroism (XMCD) at the La-L_2 edge.

Preceeding experiments on LaH_2/Fe multilayers [3] had revealed that the 5d states of La in the interfacial region carry an ordered moment of essentially spin character; it is induced by hybridization with the 3d states of Fe. In the dihydride sublayers LaH_2, formed during preparation by reactive ion-beam sputtering, hydrogen is irreversibly bound. Additional H loading eventually leads to trihidrides with essentially no further change in the lattice parameter and is reversible. We have shown by neutron reflectometry that antiferromagnetic coupling of the Fe-layer magnetizations may be switched off and on by increasing the H content and removing it again [4]. This must be attributed to modifications of the band structure of the $LaH_{2+\delta}$ sublayers.

XAS and XMCD experiments were preformed in transmission mode at room temperature. Circularly polarized light was produced by a quarter wave plate. A magnetic field of up to 1kOe applied parallel to the beam and at glazing incidence with respect to the samples was sufficient to produce magnetic saturation. Equilibrium at a given pressure p_{H2} was reached in about 1 hour. Experience from the previous experience ascertains that at p_{H2} = 1 bar a composition $LaH_{>2.8}$ of the hydrided sublayers is reached [4]. The samples studied were $[LaH_{2+\delta}(26Å)/Fe(15Å)]\times n$ and $[LaH_{2+\delta}(100Å)/Fe(15Å)]\times n$ on Kapton foil substrates, both

with $\delta = 0$ prior to excess hydrogen charging. Besides, H charging was performed starting with an initially H-free multilayer [La(60Å)/Fe(30Å)]×n.

Below, we show the normalized XMCD signal of La at the L_2-edge in a multilayer (with initially LaH_2) exposed to various hydrogen pressures. A pronounced modification of the dichroic signal can be visualized: (i) The absolute value of the integrated signal deceases drastically at $p_{H2} = 1$ mbar and then recovers somewhat. (ii) With increasing H content the signal approaches a derivative-like shape. The same evolution has been observed for the multilayers with 100 Å LaH_x. Note that the isotropic absorption profile (not shown) reveals a slight increase of the white line intensity and a small shift (~0.1 eV) of the edge toward a lower binding energy.

Even if the interpretation of these observations must await a more detailed data analysis some conclusions can be drawn. It is difficult to associate the dispersive XMCD signal with the insulating $LaH_{>2.8}$ for which the 5d states should be unoccupied [2] in the ground state and hence the XMCD signal should vanish. Dispersionlike XMCD profiles have been observed at the $L_{2,3}$ edges of rare earths in insulating compounds [5], but in these materials the non-zero signals in spite of an empty 5d shell have been attributed to the exchange interaction between the excited 5d electron with the partly occupied 4f shell of the rare earths [6]. Obviously, for La this mechanism does not apply. Hence our observation is quite puzzling and deserves further investigation. Note that the beamtime allocated to us permitted only a preliminary study on a multilayer $[CeH_{2+\delta}/Fe]\times n$. It shows a similar evolution of the Ce-L_2 XMCD signal through the metal-semiconductor transition.

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