ESRF	**Experiment title:** **X-ray magnetic circular dichroism in LaH_x/Fe and CeH_x/Fe multilayers: 5d magnetism across the metal-to-semiconductor transition in the hydrides**	**Experiment number**: HE-724
Beamline:	**Date of experiment****: from: 11. 09. 1999 to: 18. 09. 1999	**Date of report**: 27. 02. 2001
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*** Due to experimental problems encountered during the official beamtime (the cryostat of the beamline did not work) additional beamtime was granted within the in-house research period from 03. – 09.05.2000.*

Report:

The rare earth (RE) metals La and Ce form cubic hydrides REH_x. At $x \approx 2.8$ a metal-to-insulator transition occurs where the metal 5d-state-derived conduction band is depopulated in favour of low-lying hydrogen-derived valence bands and an energy gap of ~2 eV is formed. [1]. We have performed measurements of X-ray absorption (XA) together with X-ray magnetic circular dichroism (XMCD) at the $L_{2,3}$ edges of Ce and La to investigate the electronic and magnetic properties of metallic and insulating LaH_x and CeH_x thin films sandwiched with iron films in multilayers, $[LaH_x/Fe]\times n$ and $[CeH_x/Fe]\times n$. Previous experiments on such samples with the hydrides in the metallic phase ($x=2-\delta$) had shown that the hydrides are magnetically polarised by Fe near the interfaces [2]. Bulk metallic CeH_x orders magnetically only below 7 K, and LaH_x always remains paramagnetic. A priory, it is not clear if magnetic order persists in the insulating trihydride sublayers ($x \approx 3$), since in the bulk material the La or Ce 5d shell is empty in this phase [1].

In the multilayers examined here the RE 5d band states play a key role, by a similar mechanism generally accepted for RE compounds with transition-metals (TM) [3]: (i) 5d-magnetic order is induced near the interfaces by hybridisation with the spin-split 3d states of Fe. (ii) 5d-4f exchange interaction within the RE atom (in the case of Ce) couple the 4f moments to the neighbouring 3d orbitals of Fe. The photo-absorption process at the RE $L_{2,3}$ edges is dominated by the 2p-to-5d dipole transition. It does not simply reflect the difference in occupation of spin-split 5d bands in the ground state of the RE, but also the dependence of the 2p-5d radial matrix element on the relative orientation of the magnetic moments of the excited 5d photoelectron and the 4f electron [4]. Furthermore, band effects and atomic-like final-state interaction have to be included in the interpretation of the dichroic spectra. Model calculations [5] were successful in describing trends in the sign and the ratios of the dichroic signals observed [6,7] at the L_2 and L_3 edges of RE-TM compounds. However, details remain unexplained.

Multilayer samples $[Fe(15Å)/LaH_x(t)]\times n$ and $[Fe(16Å)/CeH_x(t)]\times n$ ($10\ Å \le t \le 100\ Å$) with metallic hydride sublayers ($x \approx 2$) were grown by reactive ion-beam sputtering in a H_2 atmosphere. Kapton-foil

substrates were used to permit XA measurements in the transmission mode. A Pd cap layer provided oxidation protection and a hydrogen window for additional hydrogen charging up to $REH_{\sim 3}$. The XA and XMCD spectra were measured between 10 and 300 K by reversing a magnetic field of ~100 Oe (sufficient for magnetic saturation) oriented along the propagation direction of the beam with fixed helicity.

Results

The isotropic $L_{2,3}$ XA spectra of La and Ce in the multilayers show a 'white line' profile near the edge. Its intensity grows significantly as the H concentration is increased to above x=2, which indicates a growing depletion of the 5d band states. For Ce this spectral shape reveals that the 4f states are localised in the hydride, as in the γ-phase of Ce metal.

It is deduced from the XMCD measurements that in the metallic phase of the hydride layers the ordered 5d magnetic moment of La and Ce induced by Fe at the interfaces is about a factor of 20 smaller than the Fe-3d moment determined previously [2] and is directed antiparallel to it. The most remarkable result is that the magnetic 5d polarisation does not vanish in the insulating state of the hydrides, $REH_{\sim 3}$. Roughly, the amplitude of the $L_{2,3}$ XMCD signals is decreased by a factor of two at room temperature. From the variation of the dichroic signals with the REH_x-layer thickness it is deduced that the 5d polarisation falls off exponentially into these layers with a decay length of about 10 Å, both for the metallic and insulating ReH_x phases. The finite 5d polarisation in the insulating hydrides points to the introduction of metal-like 5d states into their energy gap near the interfaces, similarly as in Fe/Ge/Fe and Fe/GaAs/Fe tunnelling structures [8].

The shape of the dichroic spectra is asymmetric, with positive and negative contributions. The asymmetry increases in the insulating phase of the hydrides. A dramatic increase of the XMCD intensity ratio, $I(L_2)/I(L_3)$, at low temperatures reflects the presence of a considerable 4f-5d exchange interaction [6]. It has a pronounced influence on the dichroic spectra of the CeH_x/Fe multilayers: it leads to a sign change between high and low temperature and results in a larger XMCD signal at 10 K for insulating than for metallic CeH_x.

We have analysed the shape of the XMCD spectra in a phenomenological two-parameter model [9] that is based on the ideas underlying the existing theoretical models [4,5]: (i) The parameter α combines the differences in the density of 5d hole states, $\Delta\rho=\rho^{\uparrow}-\rho^{\downarrow}$, and of the 2p-5d matrix elements, $\Delta M=M^{\uparrow}-M^{\downarrow}$, in an 'effective polarisation' $\alpha=(\Delta M/M+\Delta\rho/\rho)/2$ which may be positive or negative. The effect is that a transition to a band with fewer unoccupied states may result in a larger absorption due to a stronger transition probability. (ii) The parameter β represents a splitting in energy of the 5d majority and minority spin states due to the exchange interaction with the 4f states in the case of Ce, or with the Fe-3d states in the case of La. β explains the dispersive part of the XMCD line shape. For details and a thorough discussion of the erxperimental results the reader is referred to the thesis work of M. Muenzenberg [9].

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