Beamtime report of proposal HE- 3568

Temperature dependent investigation of the magnetic properties of LuFe2O4 by means of high field XMCD

The main aim of the proposal were:

1) To investigate the magnetic ground state of the of the magnetoelectric, ferroelectric compound LuFe2O4 by means of high field x-ray magnetic circular dichroism (XMCD).
2) To elucidate if also the Lu ions carry magnetic moment and its orientation with respect to the magnetization of the Fe ions

In order to magnetically saturate the multiferroic layered compound LuFe2O4 high magnetic fields of at least 15T are necessary at low temperatures (10 K). Therefore we wanted to investigate the internal magnetic properties of this compound by means of high field XMCD at the Le K and Lu L2,3 edges. Unfortunately the power supply of high field endstation (up to 18 T) available at ESRF ID12 failed during the first few measurements at the Fe K edge.

Nevertheless we were able to perform a number of valuable measurements using the second endstation in external fields up to 6T (Fig. 1). The upper panel displays the Fe K XAS (black) of LuFe2O4. The spectrum comprises to distinct features at the pre-edge located around 7112 eV and 7114 eV, followed by the main edge spanning the range from around 7117 eV and 7145 eV. The XMCD signals recorded at several temperatures are also shown in Fig. 1. The XMCD shows a rather sharp feature between 7112 eV and 7114 eV (pre-edge), followed by a smaller double peak with opposite sign located between 7115 and 7122 eV and a rather broad feature with positive sign spanning the range between 7122 eV and 7133 eV.

The lower panel of Fig. 1 shows our previous results recorded at the Fe L2,3 edges of LuFe2O4 for comparison [1] (left) and the results obtained during this beamtime at the Lu L2,3 edges (right). Both spectra were performed at T=125 K and B=6 T. Firstly, also the Lu ions carry a small magnetic moment, since a clear dichroic signal is obtained, although the size of the effect is only up to 0.4%. Whereas it is difficult to extract the magnetic moment quantitatively from the sum rules the overall shape of the XMCD allows us to conclude that the moments at the Lu ions are aligned in a similar way as those of the Fe ions, which built up a ferromagnetic ordering across the layers of the crystal [1]. Secondly a high orbital contribution to the magnetization can be expected also at the Lu ions (integral of the XMCD not shown here).

A deeper analysis including theoretical approaches, i.e. ab initio as well as charge transfer multiplet calculations is currently underway in order to clarify the following points:

1) Extract crystal field parameters and amount of charge transfer for the Lu ions by simulating the Lu L2,3 XAS and XMCD by means of charge transfer multiplet simulations. Here also complementary XAS and XMCD investigations at the Lu M4,5 edges are planned in order to get a complete “set” of XAS and XMCD at the Fe K, Fe L2,3, Lu L2,3 and Lu M4,5 edges
2) Describe the rather complex Fe K XAS and XMCD spectra. This is a bit challenging but also very interesting for this example of a mixed Fe2+ / Fe3+ compound. The main features of the XAS and XMCD are dominated by 1s -> 4p dipole transitions and can be described by band structure calculations already performed with the WIEN2kK package [1]. However, the pre edge comprise significant contributions of much more localized 1s -> 3d quadrupole transitions, these can be simulated within the multiplet approach. Whereas the quadrupole transition should dominate the pre-edge features stemming from the Fe2+ ions an admixture

of local 1s -> 3d quadrupole and non local 1s -> 4p dipole interactions can be expected [2]. Therefore a combination of the ab initio and multiplet simulations will be necessary to entirely understand the Fe K XAS and XMCD of LuFe2O4. With that approach we should be able to separate the localized and non localized features of the Fe K pre edge structure in the XAS and XMCD and to gain more insight into the complex internal magnetic properties of LuFe2O4 (including potential metal-metal or metal-oxygen interactions).

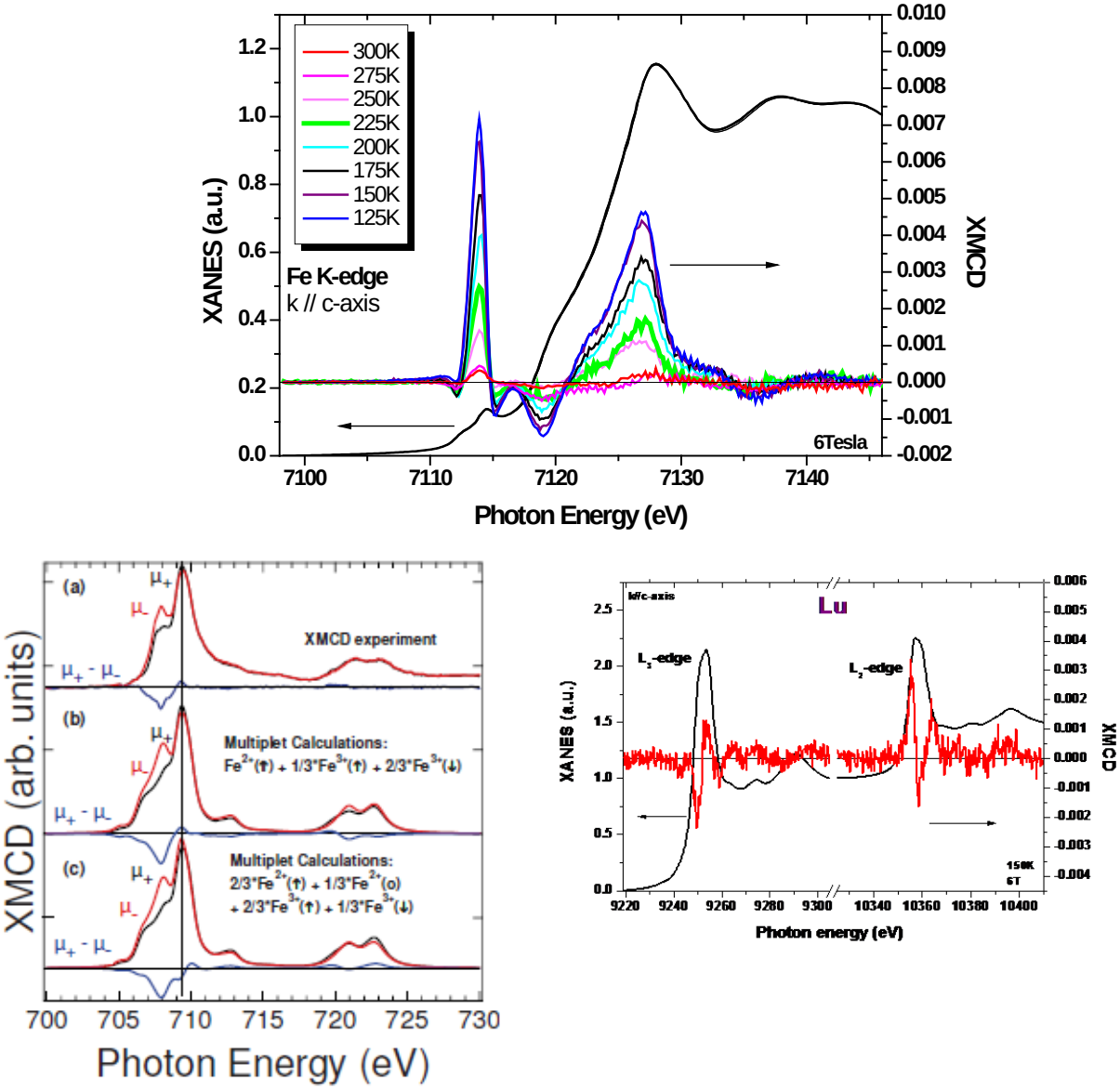

Fig. 1: Upper panel, XMCD spectra recorded at the Fe K edge as function of temperature. The isotropic Fe K XAS is also shown (black). All spectra have been taken under an external magnetic field of 6 T. Lower panel, Fe L2,3 XMCD recorded at T=150K, B =6T [kue09] (left), and Lu L2,3 XAS and the corresponding XMCD signal (red) taken at T=150K, B =6T (right).

References:

[1] K. Kuepper et al., Phys. Rev. B 80, 220409 (R) (2009).

[2] F. De Groot et al., J. Phys. Condens. Matter 21, 104207 (2009).