ESRF	**Experiment title:** **MCD and core hole screening dynamics in $CeFe_2$**	**Experiment number**: HE-3015
Beamline: ID12	**Date of experiment**: from: 15-07-2009 to: 21-07-2009	**Date of report**: 20-AUG-2009
Shifts: 18	**Local contact(s)**: **Dr Andrei ROGALEV**	*Received at ESRF:*

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The goal of this experiment was twofold. First to measure the x-ray magnetic circular dichroism (XMCD) at the Ce L3 x-ray absorption edge in $CeFe_2$ using partial fluorescence yield at the 3d to 2p decay channel to reduce final state lifetime broadening from 4 eV to approximately 0.7 eV. Secondly, to record the dichroism in the emission process only, to look for dynamic spin-dependent screening processes along the lines of experiments suggested by Braicovich and co-workers in the soft x-rays range [1].The narrow energy band of photons selected by the analyzing spectrometer to make the experiment possible (the resolving power was 5000) made for low count rates so very careful optimization had to be applied to detect the relatively small XMCD signal (< 1%) we therefore focused on the first part of the proposal, and simply checked on the feasibility of the second part of the proposal which will be resubmitted.

Overview

As demonstrated by K. Hämäläinen and co-workers [2], the significant lifetime broadening in x-ray absorption spectra due to the short-lived inner shell core hole excitation can be eliminated by recording partial fluorescence spectra. In fact, by setting up specific geometrical conditions it has been predicted that the RIXS XMCD is a two-step process so that the XMCD observed can be related to the Ce L_3 XMCD XAS signal (see [4] and references therein). The advantage now is that the spectral broadening is then determined by the convolution of the smaller lifetime broadening of the shallower core hole final state of the RIXS process and the overall experimental energy resolution (x-ray monochromaticity and spectrometer resolution). These partial fluorescence yield spectra can therefore have a higher energy resolution and therefore reveal more detail than a standard XAS spectrum. For completeness we underline that the result is not simply a high resolution image of XMCD signals and care must be taken to interpret the result but the technique can uniquely reveal fine detail in 4f excitation spectra not observable in the normal way. (See Carra et al [3] for a detailed discussion concerning such partial fluorescence yield data)

We chose as a "textbook" example $CeFe_2$. Impurity Anderson model (IAM) calculations and local spin density approximation (LSDA) calculations show that the Ce L_3 absorption edge (~5723 eV), which exhibits a ~1% XMCD effect can only be explained by the presence of localized and delocalized 4f states as indeed suggested by the magnetic properties below the Curie temperature. There is debate however as to whether the intrinsic resolution at the Ce L_3 edge is sufficient to reveal sufficient detail to allow for a simple interpretation [4,5]. Indeed in the XAS lifetime broadening of the Ce $2p^{-1}$ final core hole state of the absorption process is ≈ 4eV while monitoring the ground state (GS) - $2p^{-1}5d$ - $3d^{-1}5d$ RIXS channel with high energy resolution, we expect to more clearly observe this contribution to the XAS spectrum as well as its proper magnetic dichroism.

X-ray Emission results

The pre-requisite for recording of XAS spectra with sub-lifetime broadening is an experimental resolution, which is better than the inner shell core hole lifetime. This implies that both, the x-ray beam monochromaticity as well as the energy resolution of the x-ray spectrometer, have to be optimized. By carefully tuning the beam line monochromator and optimizing our custom-built x-ray spectrometer, we achieved the anticipated combined energy resolution of ~1.1 eV, which is significantly smaller than the inherent Ce $2p^{-1}$ lifetime (4 eV). This resolution is demonstrated by recording an elastic peak obtained by setting the beamline energy to 4836.7eV (i.e. at approximately the Ce Lα emission energy) as shown in Figure 1. Figure 2 shows also typical RIXS spectra record for several excitation energies through the Ce L_3 absorption edge.

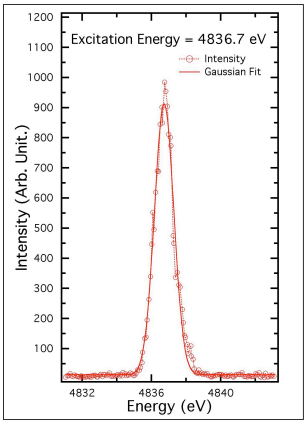


Fig. 1.: Elastic Peak

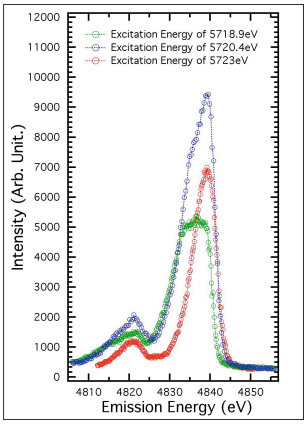


Fig. 2.: Typical Ce 2p3d RIXS spectra excited at several eneries given in the legend.

X-ray Absorption in PFY results

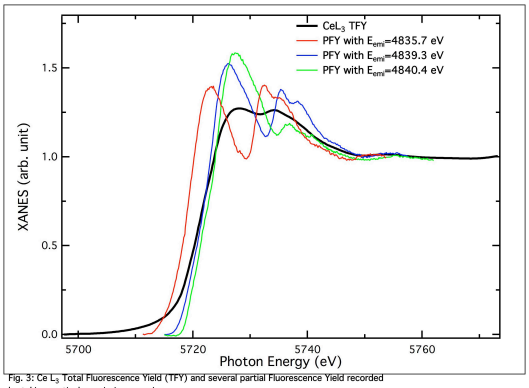
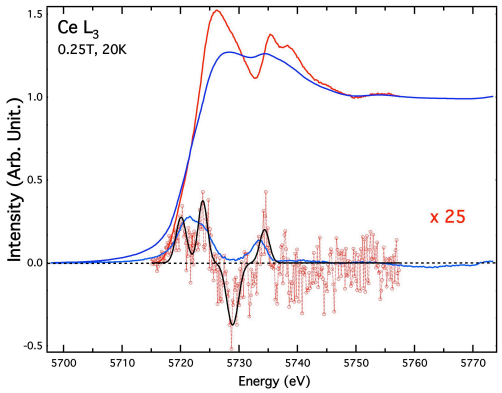


Fig. 3: Ce L_3 Total Fluorescence Yield (TFY) and several partial Fluorescence Yield recorded by taking particular emission energies.

We successfully recorded the Ce L_3 x-ray absorption spectrum of $CeFe_2$ in total (TFY) and partial (PFY) fluorescence yield showing, respectively, the normal absorption spectrum and the sub-lifetime broadened data reproduced in Fig. 3. As expected the quality of these spectra is excellent and more important the effect of reduced spectral broadening is obvious as illustrated by the appearance of two clear fine structures in the second peak of the main edge. Nevertheless there is no clear evidence of fine structure at the onset of the absorption edge.

RIXS-assisted high-resolution XMCD



Finally we report that we succeeded to record a high resolution XMCD signal at the Ce L_3 edge. This challenging experiment was only possible thanks to the high stability of the ID12 beamline and required 24h minimum to record one spectrum. Data were collected by reversing the magnetic field and the helicity of the incoming beam. In figure 4 we display data taken with a narrow spectrometer window centered at 4839.3eV together with normal TFY- XMCD under optimized conditions.

As underlined by a fit (black line), one can clearly distinguish several well separated peaks in the RIXS MCD not resolved in the XAS MCD (blue line). This is a clear indication that the final state must be understood in terms of a mixed valency with both localized and delocalized 4f states. Though it is generally accepted that the magnetic properties of $CeFe_2$ cannot be interpreted in terms of itinerant 4f electrons only, these results provide the clearest proof that XAS spectra calculated on the basis of LSDA [5] are far from satisfactory since they miss the presence of the important features. However, there is good qualitative agreement between an XAS calculation based on the impurity Anderson model and our results.

Despite the difficulty of the experiment which requires exceptionally stable experimental conditions to be able to extract valid XMCD signals, we believe we have demonstrated the potential for making further progress in the study of the magnetic properties of highly correlated systems.

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