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Magnetic circular dichroism in the $L_{2,3}$ x-ray fluorescence of Fe and Co

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Abstract: the $L_{2,3}$ x-ray fluorescence spectra of magnetized Fe and Co excited by circularly polarized synchrotron radiation exhibit magnetic circular dichroism. The results are compared to the energy resolved spin polarized densities of states given by first-principles calculations.

1. INTRODUCTION

High resolution x-ray fluorescence (XRF) spectroscopy consists in measuring inelastically scattered photons. The energy, direction and polarization of the primary radiation may be varied but essentially, as opposed to standard x-ray fluorescence analysis, the energy distribution of the scattered photons is measured. The excitation of a core level with well-defined angular momentum during the scattering process means that it is possible to probe states with a specific angular momentum at a specific type of site. This property is particularly useful when complex materials are being studied. Until recently, only the site and angular momentum selectivity have been exploited, though a few experiments on single crystals and layered materials have also made use of the polarization of the emitted photons to obtain more detailed information on the electronic structure [1]. Here we describe a new experiment in which the elliptical polarization of synchrotron radiation (off-orbit plane) is used to probe the spin polarization of a magnetically oriented ferromagnet. We show that magnetic circular dichroism (MCD) may be observed when we take the difference between spectra with the spin of the primary photons parallel or anti-parallel to the magnetization direction of the target. XRF MCD probes the spin polarization of the occupied states, whereas MCD in x-ray absorption experiments probes the empty conduction states.

2. EXPERIMENT

XRF experiments require a very high photon flux because fluorescence yield is generally low (a few percent for the $2p$ shell of the first transition metal series) and the fluorescence radiation must be energy dispersed with sufficient resolution (typically 5×10^{-4}). This implies detecting only a small solid angle of

the isotropically emitted fluorescence signal. To satisfy this requirement we used unmonochromatized white bending magnet radiation emitted 0.35 mrad above or below the orbit plane at LURE-Super-ACO. Under these conditions, the primary photons were roughly 50% circularly polarized at the $L_{2,3}$ threshold of the late 3d transition metals. Experiments were performed separately above and below the orbit plane to check for a reversal in the signal, but the dichroism was measured by changing the direction of magnetization (parallel or antiparallel) to photon helicity keeping the geometrical conditions rigorously unchanged. To be more exact the primary beam made an angle of 15° with the surface of the ferromagnet so as to intercept 0.2 mrad of the beam in the vertical plane. The magnetic field was produced by permanent magnets. XRF was observed almost perpendicularly to the sample surface to minimize self-absorption.

Our first results on iron [2] were obtained with a classic Johann-type spectrometer [3] using a step-by-step recording mode. The magnetic field was reversed at each energy step so that no normalization procedure needed to be applied before subtraction to obtain the dichroic signal. The data shown here used the same geometrical set-up but were taken with a new spectrometer equipped with a position sensitive detection device. In this case spectra were recorded over periods of about 10 min for each magnetization direction. Spectra were normalized to the same average count rate over the whole spectrum. This approximation was justified by the fact that the dichroic intensity was small and reversed for $L\alpha$ and $L\beta$ energy regions. An RbAP crystal bent to a radius of 0.5 m was used.

3. RESULTS and DISCUSSION

In our first experiment [2] we observed that when the magnetization direction in the Fe sample was reversed from antiparallel to parallel with respect to the helicity of the incoming photon, a positive difference was observed in the $L\alpha$ spectrum and a negative difference was observed in the $L\beta$ spectrum. $L\alpha$ had a marked dip in the middle of the dichroic signal but no equivalent structure was observed in the $L\beta$ region. These features are all clearly confirmed by the new experiments (Fig.1). Moreover the same overall dichroic signal is observed for cobalt (Fig. 2).

The experiment on iron was in good agreement with the spin-polarized band structure calculations [4] in which the fluorescence intensity I^λ (λ denotes the helicity of the photons) coming from the radiative decay of the core hole ε_c is given in terms of the unoccupied and occupied densities of states $n(\varepsilon_c + \omega_{in})$ and $n(\varepsilon_c + \omega_{out})$ and the relevant transition matrix elements M by :

$$I^\lambda \sim \sum_{\Lambda\Lambda'} n_\Lambda(\varepsilon_c + \omega_{in}) n_{\Lambda'}(\varepsilon_c + \omega_{out}) |M_{\Lambda\Lambda'}^\lambda|^2 |M_{\Lambda\Lambda'}^{\lambda'}|^2.$$

In the theory I^λ was evaluated from first-principles within the framework of a fully relativistic spin-polarized multiple scattering Korringa-Kohn-Rostoker approach.

The theoretical MCD signal was calculated for $\omega_{in}=1.5$ eV above the L_3 edge, but the XRF MCD signal depends on the probability of creating a majority or minority spin-polarized core hole. If we admit that the radiative decay of the core hole is independent of the primary photon energy, the shape of the MCD

fluorescence signal will reflect the spin polarization of the occupied states whatever the energy of the primary photons. Even so, the observation of fine structure though in apparent agreement the calculated spectrum was unexpected considering initial and final state lifetime broadening together with instrumental broadening.

Fig. 1. – Fe $L_{2,3}$ XRF signal and XRF MCD signal.

No calculation for the XRF anisotropy of Co is available at present but the augmented plane wave spin-polarized densities of states [5] suggest that no dip should be observed. It seems therefore that the dip observed in the $L\alpha$ dichroic signal is generic to the fluorescence spectrum.

Fig. 2. – Co $L_{2,3}$ XRF signal and XRF MCD signal.

From the photoemission experiments of Baumgarten *et al.* [6] and the calculated Fe 2*p* photoelectron spectrum due to Ebert *et al.* [7] we know that the binding energy of the magnetic field-split and spin orbit-split levels are different (0.5 eV for 2*p*_{3/2}, 0.3 eV for 2*p*_{1/2}) according to the helicity of the incoming photons. The dichroic signal in the fluorescence spectrum measures the difference between the majority spin band and the minority spin band the latter being shifted to a higher binding energy. In a simple Stoner-like model the majority band will be broader than the minority band so that the dip is largely a consequence of sensitivity to band width when the dichroic signal is small. A dip in the Lβ region is not observed because the anisotropy is larger and the shift due to the field-split 2*p*_{1/2} components is smaller. However, the more pronounced dip in the Fe spectrum, compared to Co, probably does indicate a contribution from the fine structure in the occupied density of states.

It should be underlined that the 2*p* binding energy dependence on helicity is fully included in the calculation but the full experimental broadening effects are not.

4. CONCLUSIONS

Despite the difficulties encountered in performing XRF MCD measurements, we consider that such studies must be developed further as they can be expected to provide information on the spin-resolved local density of occupied electronic states in multicomponent systems.

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