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Quadrupolar transitions by MCXD at L edges? Search of evidence

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Abstract

To explain the low-energy features of magnetic circular X-ray dichroism (MCXD) at L edges of rare earth compounds, quadrupolar electric transitions (E2) from 2p to 4f levels have been invoked. Such transitions should be distinguished from dipolar electric (E1) ones by looking at the different behaviour of the E1 and E2 MCXD cross sections as a function of the temperature and the angle between the quantization axis and the photon wave vector. However, convincing experimental proof of such transitions has not yet been given. A good candidate for these key tests is Yb, where the E2 transitions are allowed at the L_3 edge and forbidden at the L_2 edge. In $YbFe_2$, we observe an important change in the shape of the spectrum with temperature at the L_3 edge of Yb.

1. Introduction

The magnetic circular X-ray dichroism (MCXD) signal is the difference between the absorption cross sections of X-rays polarised circularly left and right. Originally, MCXD at $L_{2,3}$ edges of rare earths (RE) was interpreted by taking into account only dipolar electric transitions (E1): $2p_{1/2,3/2} \rightarrow 5d$. In our systematic study of $L_{2,3}$ edges of RE in $REFe_2$ compounds (RE = Sm, Gd, Dy, Ho, Er, Tm, Yb and Lu) [1], we have found that, as soon as the 4f shell is incomplete, the L_3-edge MCXD signal presents two structures with opposite sign, unlike $LuFe_2$ which has only one peak. At the L_2 edge, the MCXD signal can present several structures, whose relative intensities change along the RE series and with temperature. This is not the behaviour expected for the 5d magnetic moment in these systems [2].

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2. Quadrupolar electric transitions

To explain the low-energy features observed at the L edges of RE, quadrupolar electric transitions (E2) [$2p_{1/2,3/2} \rightarrow 4f$] have been invoked [3]. The extra Coulomb interaction between the 2p core hole and the photoelectron, which is stronger when the photoelectron has an f symmetry (E2) than a d symmetry (E1), shifts the E2 transitions by about 7–9 eV towards low energy in relation to E1 transitions. High resolution X-ray absorption measurement at the L_3 edge of Dy [4] has shown a pre-edge feature 7 eV lower than the edge (E1), which has been interpreted with E2 transitions. The calculated [3] E2 MCXD cross section [$\mu^C(E2)$] fits the low energy structure observed at the L_3 edge in heavy RE compounds [5].

Carra et al. [3] also calculated the dependence of $\mu^C(E1)$ and $\mu^C(E2)$ on the angle α between the incident

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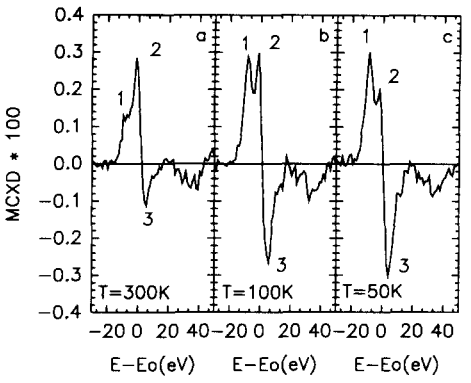

Fig. 1. MCXD at L_3 edge of Yb in $YbFe_2$ for $\alpha = 0^\circ$ at (a) $T = 300$ K, (b) $T = 100$ K, (c) $T = 50$ K.

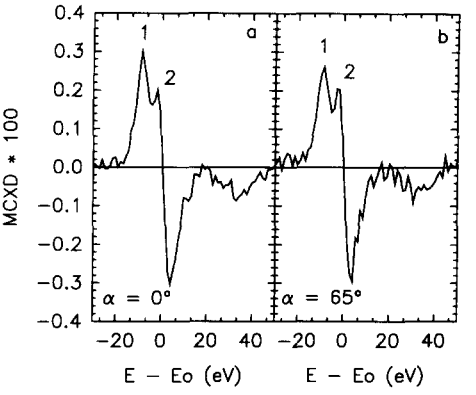

Fig. 2. MCXD at L_3 edge of $YbFe_2$ at $T = 50$ K, (a) $\alpha = 0^\circ$, (b) $\alpha = 65^\circ$ [normalised by cos (65°)].

photon wave vector and the quantization axis of the system. The E1 contribution is proportional to $\cos\alpha$ whilst the E2 contribution can be divided into two terms: one proportional to $\cos\alpha$, as for the $\mu^C(E1)$, and the second to $\cos^3\alpha$: $\mu^C(E2) = A_1 \cos\alpha + A_3 \cos^3\alpha$. Any deviation from the pure $\cos\alpha$ dependence should be a signature of E2 contributions.

3. Results

Ytterbium appears to be a good candidate to test the E2 contribution, because E2 transitions are allowed at the L_3 edge and forbidden at the L_2 edge. We measured the MCXD spectra at the L_3 edge of Yb in $YbFe_2$ at several temperatures for $\alpha = 0^\circ$ and $\alpha = 65^\circ$.

The spectra are mainly composed of two structures, positive at lower energy and negative at higher energy. The low-energy peak can be decomposed into two peaks (1 and 2 in the figures), which is not the case for the other rare earth compounds.

The contrast between the two peaks of the low-energy feature changes with temperature. We observe that for $\alpha = 0^\circ$ (Fig. 1) as well as for $\alpha = 65^\circ$, peak 1 decreases, whilst peak 2 increases with increasing temperature. Peak 3, which is expected to be due to E1 transitions only, has the same evolution as peak 1. This has not yet been correlated with the variation of E1 and E2 MCXD cross sections with temperature.

In the peculiar case of Yb, the calculated A_1 and A_3 terms of $\mu^C(E2)$ are opposite at low temperature. Thus, $\mu^C(E2)$ should be zero for $\alpha = 0^\circ$ and positive for $\alpha = 65^\circ$.

We do not observe any significant deviation from the $\cos\alpha$ dependence at $T = 50$ K (Fig. 2) and $T = 100$ K. This means that either $T = 50$ K is already too high or the low energy feature contains E1 transitions.

4. Conclusion

The MCXD signal at the L_3 edge of Yb has a low-energy structure composed of two peaks whose temperature evolution is different. The angular variation does not show the deviation from $\cos\alpha$ dependence expected for the E2 contributions. Then the low-energy feature could contain E1 transitions.

References

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