



	<b>Experiment title:</b> <i>Study of the higher-order 5d-multipole moments in rare earths and transition metals using L<sub>2,3</sub> resonant Raman scattering sum rules</i>	<b>Experiment number:</b> HE1398
<b>Beamline:</b> ID24	<b>Date of experiment:</b> from: 4/12/2002 to: 15/12/2002	<b>Date of report:</b> 1/3/2004
<b>Shifts:</b> 27	<b>Local contact(s):</b> Olivier Mathon	<i>Received at ESRF:</i>
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## Report:

The experiment had an exploratory character. Starting from the experience accumulated on 3d transition metal systems in the soft x-rays (L<sub>2,3</sub> absorption edge) [1] we proposed to use magnetic circular dichroism (MCD) in the resonant Raman scattering (RRS) in the perpendicular geometry at the L<sub>3</sub> edge of rare earths. The ultimate goal was to use the special sum rules for RRS [1,2] to experimentally evaluate the magnetic and charge distribution multipolar moments of Gd and Ho. Working at the L<sub>3</sub> edge of rare earths allows to explore the ground state properties of the 5d valence states, that have a rather delocalised nature and hybridise both with the 4f states of the same atom and with the valence electrons of the neighbouring atoms. In case of fully successful measurements the comparison of results obtained on metallic (GdCo<sub>2</sub>, HoFe<sub>2</sub>) and ionic (Gd<sub>2</sub>Fe<sub>5</sub>O<sub>12</sub>, Ho<sub>2</sub>Fe<sub>5</sub>O<sub>12</sub> garnets) compounds could give interesting information on the magnetic moments and on the charge anisotropy of the 5d shell in two typical cases: Gd has a spherical 4f shell (4f<sup>7</sup>), Ho does not (4f<sup>10</sup>).

The novelty of the experiment required a long setting up preliminary phase: an electromagnet had to be modified to be mounted perpendicularly to the incident beam. The polar expansion allowed a magnetic field of 0.6 T in the typical measurement conditions. A Si photodiode was mounted on a rotary stage allowing 360 degree rotation around the incident beam axis. The diode could be as close as 20 mm from the sample. The scattering angle was fixed at the magic angle (56 deg), whilst the azimuth angle  $\beta$  could be changed with the motorised rotary stage. The circular polarisation of the incident beam was given by a  $\lambda/4$  diamond plate. The samples were in air at room temperature. An energy window of about 300 eV was chosen centred on the L<sub>1</sub> emission (3s $\rightarrow$ 2p transition), which gives a 10 times higher MCD signal in perpendicular geometry with respect to the L <sub>$\alpha$</sub>  (3d $\rightarrow$ 2p transition) [3], as we could verify in the case of Gd ourselves again.

We could measure with satisfactory data quality only two samples: GdCo<sub>2</sub> and HoFe<sub>2</sub>. The garnets could not be oriented in the correct way with the available electromagnet at room temperature: the experiment can be made only if the sample magnetisation is perfectly perpendicular to the incident beam, in order to eliminate all contributions coming from the absorption MCD (which is of the same order as the RRS MCD). The results are summarised in the figures 1 and 2. Gd and Ho show a 1% and 0.5% MCD signal at the energy of the absorption peak. The angular dependence in both cases is a cosine law, as expected. The sum spectra (left plus right circular polarisation) did not show an angular dependence big enough to be detected (i.e. if any this modulation, expected to follow a  $\sin^2\beta$  law, has an amplitude smaller than 0.1%). On the same sample we measured later (with the same magnetic field, in L <sub>$\alpha$</sub>  fluorescence yield) the absorption MCD. Gd and Ho gave 0.5% and 0.6% peak MCD respectively, with one important difference: for Gd the MCD spectral shape is very

similar in absorption and in RRS, for Ho the spectral shape is very different in the two techniques (see figure 3). In particular we can notice the in the RRS spectra the first MCD peak is absent: this peak is usually attributed mainly to electric quadrupole (E2) excitations in the absorption process [4]. This observation may indicate that in the RRS process the E2 component is negligible and the sum rule for RRS MCD could be applied, also considering that in RRS MCD is zero at  $L_2$  and the sum rule can be applied to the  $L_3$  spectrum alone. A more complete analysis of the experimental findings is going to be made based on numerical calculations and theoretical work.

[1] L. Braicovich, A. Tagliaferri, G. van der Laan, G. Ghiringhelli, and N. B. Brookes, Phys. Rev. Lett. 90, 117401 (2003)

[2] F. Borgatti, G. Ghiringhelli et al, Phys. Rev. B, accepted

[3] L. Braicovich, G. van der Laan, G. Ghiringhelli, A. Tagliaferri, and N. B. Brookes, Phys. Rev. B 66, 174435 (2002)

[4] F. Bartolomé, J.M. Tonnerre, L. Sève, D. Raoux, J. Chaboy, L.M. Garcia, M. Krisch, and C.C. Kao, Phys. Rev. Lett. 79, 3775 (1997)

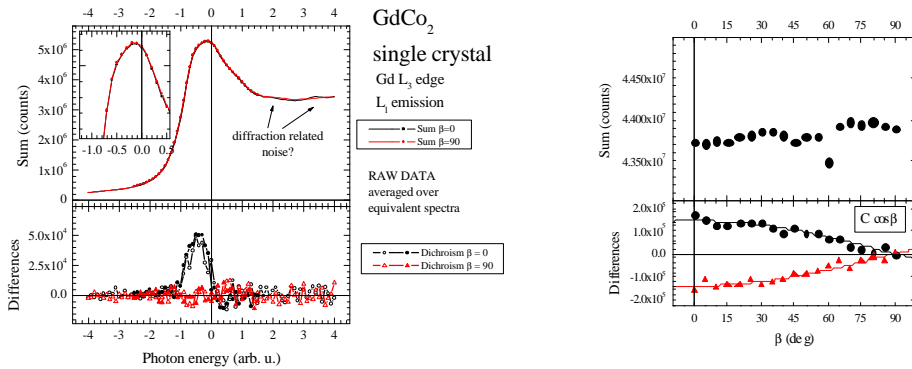


FIGURE 1. GdCo<sub>2</sub>. Left panel: IRRS spectra at L<sub>3</sub>. Right panel: the Sum and dichroism signals plotted as functions of the azimuthal angle  $\beta$ .

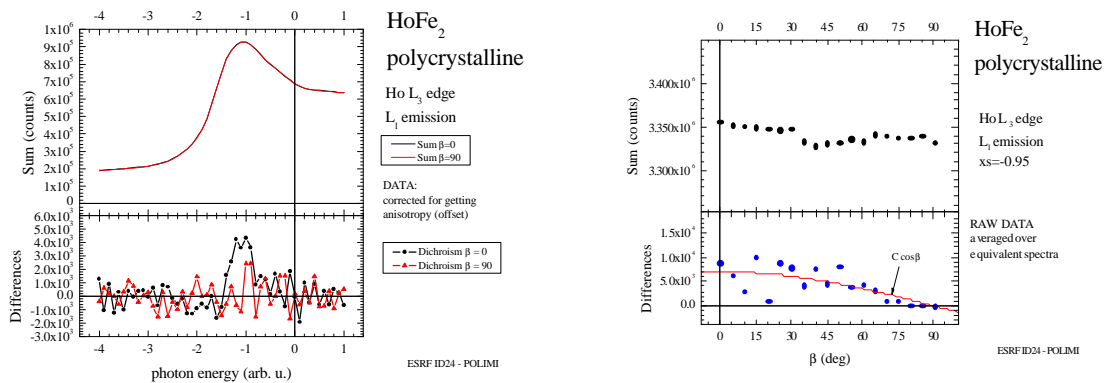


FIGURE 2 HoFe<sub>2</sub>. Left panel: IRRS spectra at L<sub>3</sub>. Right panel: the Sum and dichroism signals plotted as functions of the azimuthal angle  $\beta$ .

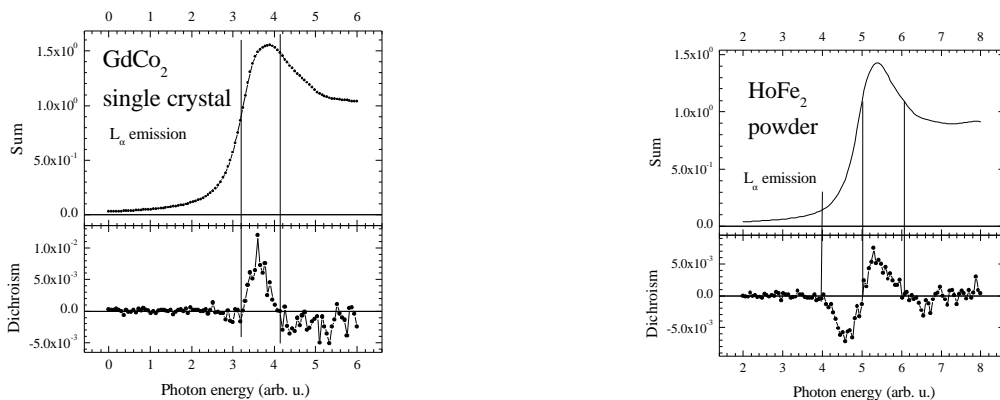


FIGURE 3. Absorption MCD at L<sub>3</sub> measured in fluorescence yield mode (L<sub>α</sub> emission)