



	Experiment title: <i>Measuring the ground state multipolar moments of magnetic systems with integrated resonant Raman scattering.</i>	Experiment number: HE884
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Report

In Resonant Raman Scattering (RRS) the cross section is a function of the incident and outgoing photon energies, $h\nu_{in}$ and of $h\nu_{out}$. In this field we have introduced a new method: we call IRRS (Integrated RRS) in which we measure *directly* the integral of the energy distribution of the emitted photons obtaining a spectrum which is a function of $h\nu_{in}$ only. This gives a signal with sufficient intensity to be mapped in a reasonable time over the scattering angles in convenient geometrical arrangements. This method is suited to study the angle dependence and to apply sum rules after a subsequent integration over $h\nu_{in}$ at a given excitation edge. In this way the sum rules approach is extended to RRS i.e. far beyond the traditional absorption spectroscopy.

The aim of the experiment was to apply this concept to magnetic systems based on Co by measuring IRRS spectra coming from the reaction $2p^63d^n \rightarrow 2p^53d^{n+1} \rightarrow 2p^63d^{n+1}3s^1$ excited with circularly polarised light in the geometry shown in the box A of the figure. The spectra are measured vs. the angle β around the cone with half-aperture $\text{acos}(1/\sqrt{3})$ ("magic angle"). In this geometry the β dependence is not affected by changes in self-absorption and saturation, thus solving the main difficulties of the previous experiments. *The present results are the first with the conical scan.* We show, as an example, in box B the $\beta = 0$ spectra in Co-metal and in Co-ferrite (CoFe_2O_4) i.e. $I_{\text{dichr}} = [I^+(h\nu_{in}) - I^-(h\nu_{in})]$ and $I_{\text{sum}} = [I^+(h\nu_{in}) + I^-(h\nu_{in})]$ where the index refers to opposite helicities of the incident beam. In this geometry there is no absorption dichroism but there is a RRS dichroism shown in the figure by the heavy solid lines. Note that the IRRS dichroism at L_2 must be zero apart from small effects due to the crystal field. The β dependence of the dichroism is of the type $(\sin\beta)$ as predicted by the theory (panel C where we give as an example the value of the peak dichroism (a) in ferrite). This agreement encourages a deeper use of the theory in order to recover ground state information. This

requires also the β dependence of I_{sum} since the theory shows that I_{sum} can be β dependent. The anisotropy of I_{sum} is described by a factor ($\cos(2\beta)$) weighted with a coefficient depending linearly on the ground state quadrupoles of the system (charge and magnetic quadrupolar moments). In a system fully adapted in symmetry to a high symmetry crystal the quadrupolar moments are obviously zero. Thus the presence of quadrupoles shows that the ion/atom is not fully adapted in symmetry and retains some memory of the isolated ion/atom. This crucial aspect in the description of the system can hardly be obtained with other methods and is one of the main goals of this new research. In effect I_{sum} is strongly anisotropic in Co-ferrite as shown by the ($\cos(2\beta)$) dependence of the peak (a) (box D). The anisotropy is very strong while a small but visible effect is seen also in the Co-metal (peak (a)). The quality of the measurements allows also the small effects in the metal to be seen, as shown by the upper panel of box D giving the metal data expanded by a factor of 10. The main results are the following:

(i) the conical scan works well and puts in evidence the existence quadrupolar moments of the ground state. This shows the potential of the new apparatus designed to this purpose (at present based on filters to define the bandpass)

(ii) the anisotropy of I_{sum} is very much dependent on the nature of the sample and correlates with the different degrees of symmetry adaptation. The effect is small in itinerant Co metal and is much stronger in ionic Co in ferrite. This opens a whole range of possibilities in the study of systems intermediate between the atomic-like and the band-like behaviour.

(iii) the results are the ideal basis for the use of sum rules allowing the determination of the higher order multipoles not accessible to absorption. This requires the angular dependence of the integrals of I_{dier} and of I_{sum} over the L_3 edge since the quadrupole moments come quantitatively from the β dependence of these integrated values. In Co-ferrite the theory given in Ref.^(1, 2) is appropriate and will give higher order multipole moments with a good degree of confidence. This requires also the knowledge of the traditional absorption MCD in the same experimental conditions as in the Integrated RRS. These measurements has been done during the present experiment. The sum rule analysis is in progress.

See figures on next page

¹ G. van der Laan, and B.T. Thole, J. Phys.: Condens. Matter **7**, 9947 (1995).

² M. van Veenendaal, P. Carra, and B.T. Thole, Phys. Rev. B **54**, 16010 (1996).

