ESRF	**Experiment title:** Study of ground state properties in strongly correlated systems: a new approach in Resonant Raman Scattering.	**Experiment number**: HE-1088 LTP
Beamline: ID08	**Date of experiment**: from: 05/09/2001 to: 13/09/2001	**Date of report**: 01/03/2002 *Received at ESRF:*
Shifts: 21	**Local contact(s)**: A. Tagliaferri	

Names and affiliations of applicants (* indicates experimentalists):

Prof. Lucio BRAICOVICH*
Dr. Nicholas BROOKES*
Dr Giacomo GHIRINGHELLI*
Dr. Philippe OHRESSER*
Dr. Alberto TAGLIAFERRI*
Prof. Gerrit VAN DER LAAN

Report:

The long term project (LTP) became operational in the second semester of 2001. Thus we used up to now only the first semester on the basis of 18 shifts. The project deals with the recovery of ground state properties in correlated systems by means of soft X-Ray resonant Raman scattering. In this connection the project contains two research lines:

A. the study of Cerium compounds with linear dichroism at the M5,4 edges in experiments resolved vs. the outgoing photon energy. This research line started before the LTP with the experiment HE-983 of June 8-12, 2001.

B. the study of ground state moments of magnetic systems with a new approach based on RRS sum rules applied for the first time to RRS (for this reason this will be called Integrated RRS i.e. IRRS). This research line started before the LTP with the experiment HE-884 of February 11-19, 2001 after some preceding exploratory work.

The research line A. has already given the first results (published in Solid State Communications). We have established the first correlation between the linear dichroism and the degree of hybridization on going from gamma-like to alpha-like Cerium compounds (see the activity report HE-983). Thus we report here the main results obtained in the field B where we have completed the study of Co-ferrite with the new IRRS method. We anticipate that we have been able to exploit the unique possibilities offered by a second order process as the Raman scattering to recover ground state multipolar moments up to order 4. That information is not accessible with first order spectroscopies, such as magnetic dichroism in absorption. This is the first implementation of this approach, which does not suffer of the limitations present the other typical second order process: the photoemission. Although in principle photoemission could provide this kind of information, photoelectron diffraction effects severely limit its exploitation to this purpose.

The method uses the angular dependence of the IRRS signal. As a qualitative presentation we consider the experimental geometry of Fig. 1 with circularly polarized light at normal incidence to the magnetization; **P** is the incident helicity vector, **M** the magnetization vector, and e the photon emission direction. Let J_+ and J_- be the IRRS signal measured with the two opposite polarisations of the incident beam and consider the b dependence along the cone in Fig. 1. Assume that the target atom has a quadrupolar charge distribution in the ground state as shown pictorially. The detection at the left and right hand side are not equivalent[i] so that a dichroism $J_{dichr} = J_+ - J_-$ is measured by reversing, in fixed geometry, either **P** or **M**. The dichroism can give information on ground state moments of odd order. Of course, J_{dichr} is zero in the symmetry plane *xy* (*b* = 90°, 270°) and is maximum in the *zx* plane. On the other hand, $J_{sum} = J_+ + J_-$ cannot distinguish between right and left and its angular dependence is expected to reflect the elongated shape of the target providing information on the ground state charge quadrupole. The feasibility of IRRS is supported by the observation of dichroism in conventional RRS from Ni ferrite and Co metal in perpendicular geometry[i] at a fixed scattering angle. We also measured XMCD and XMLD *in situ* using total electron yield. The experiments were done on beamline ID08 at the ESRF, which has complete polarization control and delivers ~100% circular polarization. The Co ferrite sample is crystalline grown epitaxially on MgO (3µ thick) oriented with the (100) direction along the field. The magnetic field, slightly below 1 T, is sufficient to magnetize the sample within 10% from saturation in plane as verified *in situ* with XMCD and with the symmetry properties of IRRS.

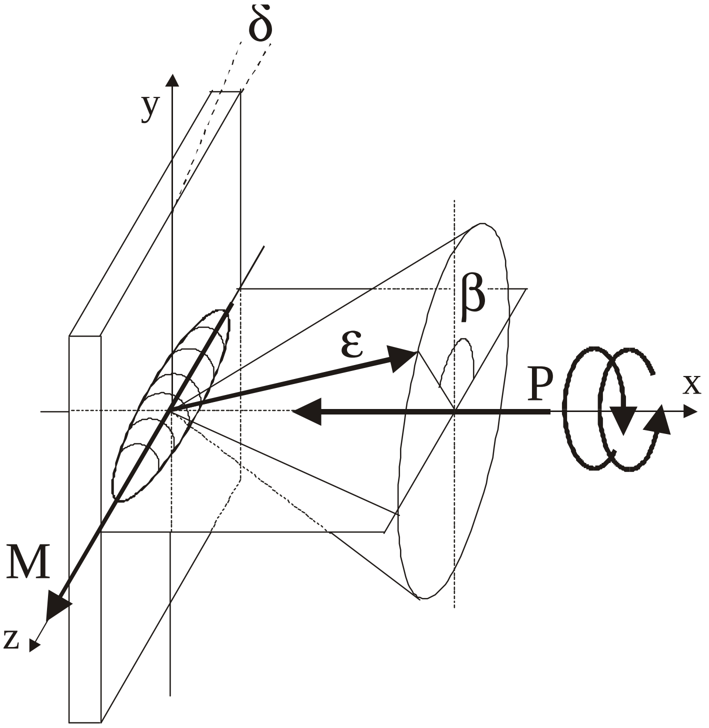

Fig. 1

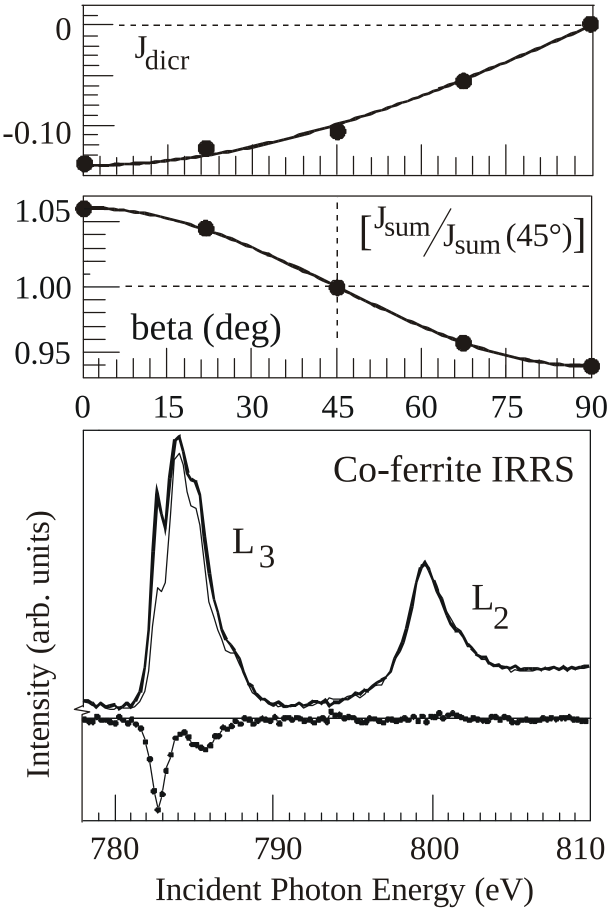

Fig. 2

The general idea is to take measurements in a suitable geometry allowing to gather some specific information on the multipole order. This idea has been presented qualitatively above in connection with quadrupoles where the basic information arises from the anisotropy of J_{sum} in a conical scan in perpendicular geometry (Fig. 1). Once the quadrupoles are known the hexadecapoles are obtained from the linear dichroism in IRRS at L_3 in the same geometry since the IRRS-MLD depends on hexadecapoles and quadrupoles. The first order multipoles (dipoles) come, as it is well known, from the XMCD sum rules (collinear absorption dichroism). Since the IRRS-MCD is sensitive also to order 3 the introduction of a collinear contribution in IRRS-MCD gives information on the octupoles. In this case one measures the IRRS-MCD in the symmetry plane (*ß* = 90°) by scanning the rotation *d* of the sample around the *y* axis (Fig. 1).

Typical Co ferrite spectra are given in Fig. 2 (lower panel, raw data at ß? 0; upper panel: angular dependence of J_{sum} and of J_{dicr})). There is no dichroism at the L_2 edge as predicted by the theory in SO_2 since the core-hole angular momentum *j*=1/2 is too low to couple to quadrupolar and higher multipoles in the ground state. This supports the use of SO_2 expressions.

On this basis we were able to evaluate the ground state moments summarized in Table I. For space reasons we omit all technicalities in the derivation. The competition between quenching and preservation of atomic properties in the solid can be seen from the results of Table I where also the theoretical values for the isolated Hund’s rule Co^{2+} ion are given. The table shows at a glance the considerable residual memory of the atomic-like behavior seen up to high order multipoles. A further discussion takes advantage of the consideration of a known problem (typical of ferrites) i.e. the existence of a fraction of Co^{2+} going in the Fe site antiparallel to the majority Co^{2+} sublattice. This fraction is typically around 20% as supported also by our XMCD analysis of the Fe $L_{2,3}$. Obviously, the Co displacement reduces the odd moments without affecting the even moments. The corrected odd moments are in brackets in Table I. We have also calculated the Co^{2+} moments in an octahedral CoO_6 cluster with an exchange splitting of 80 meV at T=0 °K. A fine tuning of the calculation, including temperature dependence, is premature but the agreement with the experiment is very encouraging since the calculation gives a general picture of the quenching of the different quadrupoles in reasonable agreement with the experiment. A good example is the quadrupole quenching as shown e.g. by M_{quad}.

	Exp.	Th. cluster	Hund ion
$\underline{w}^{101}$ (orbital moment)	-0.21 [-0.35]	-0.73	-1.5
$\underline{w}^{011}$ (spin moment)		-2.64	-3
$\underline{w}^{211}$ (magnetic dipole)		0.13	0.5
$\underline{w}^{011} + 2\underline{w}^{211}$ ("effective" spin)	-1.34 [-2.23]	-2.38	-2
$\underline{w}^{202}$ (charge quadrupole)	-0.32	-0.29	-0.5
$\underline{w}^{112}$ (anisotropic spin-orbit)	0.56	0.52	1.5
$\underline{w}^{312}$ (quadr. from orbit oct. and spin)	-0.52	-0.92	-1
$M_{\text{quad}} = 2\underline{w}^{112} + \underline{w}^{202}$	0.81	0.75	2.5
$\underline{w}^{303}$ (charge octupole)		1.25	1
$\underline{w}^{213}$ (oct. from orbit quad. and spin)		0.45	0.5
$M_{\text{oct}} = 2\underline{w}^{213} + \underline{w}^{303}$	0.7 [1.17]	2.15	2
$\underline{w}^{404}$ (charge hexadecapole)		2.03	3
$\underline{w}^{314}$ (hexadec. from orbit oct. and spin)		-1.11	-1
$M_{\text{hex}} = 2\underline{w}^{314} + \underline{w}^{404}$	-0.25	-0.19	1

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[i] L. Braicovich, G. van der Laan, G. Ghiringhelli, A. Tagliaferri, M. A. van Veenendaal, N. B. Brookes, M. M. Chervinskii, C. Dallera, B. De Michelis, and H. A. Dürr, Phys. Rev. Lett. **82**, 1566 (1999).