ESRF	**Experiment title:** Study of the ground state properties in strongly correlated systems: a new approach in Resonant Raman Scattering	**Experiment number**: HE1088
Beamline: ID08	**Date of experiment**: from: Sept 2001 to: Feb 2003	**Date of report**: 28/02/2003
Shifts: 60	**Local contact(s)**: N.B. Brookes	*Received at ESRF:*

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Report:

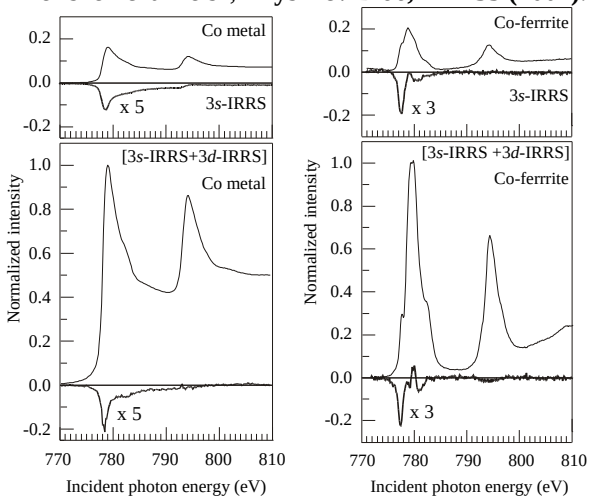
The long term proposal (LTP) HE1088 was aimed at studying of the electronic and magnetic properties of solid samples, especially those showing a strong electronic correlation by using methods and techniques based on the resonant inelastic scattering of soft x-rays. We proposed to make a step forward in exploiting the higher selectivity inherent to the scattering process (in comparison to the first order processes like x-ray absorption and x-ray photoemission), selectivity coming from the geometry of the scattering process. Moreover we suggested to make a full use of the capabilities of the newly constructed beam line ID08 in terms of photon polarisation control (circular or linear polarisation) and in terms of photon flux on the sample. In particular we proposed to perform new resonant Raman scattering (RRS; also resonant inelastic x-ray scattering, RIXS) experiments, at the $L_{2,3}$ edges of 3*d* transition metals and $M_{4,5}$ edges of the early lanthanides. Using circularly polarised incident photons for magnetic samples, we planned to measure magnetic circular dichroism (MCD) in the perpendicular geometry, mainly with the purpose of applying specific sum rules to the RRS spectra. Using linearly polarised photons for non ferromagnetic samples, we proposed to study the hybridisation properties of Cerium.

The reason for requesting a LTP was that some important instrumentation development was necessary to reach the scientific targets: a new instrument allowing the measurement of RRS spectra in the integrated mode at an arbitrary scattering angle had just been constructed two years ago; the design and construction another instrument for RRS in the integrated mode, using on a mulilayer-based broad band-pass monochromator was suggested as a first step towards the possibility of measuring the polarisation of the scattered photons; an important upgrade (new detector) of the AXES spectrometer for the energy resolved RRS measurement was expected, in order to improve both the energy resolution and the detection efficiency. We list below all the achievements obtained sofar (during the first 3 semesters of the LTP). The declared objectives of the LTP have mostly been achieved (in the first 3 semesters of the schedule) as we show below. We will follow the HE1088 proposal sequence, and we refer to the milestones declared therein.

CIRCULAR MAGNETIC DICHROISM IN RRS

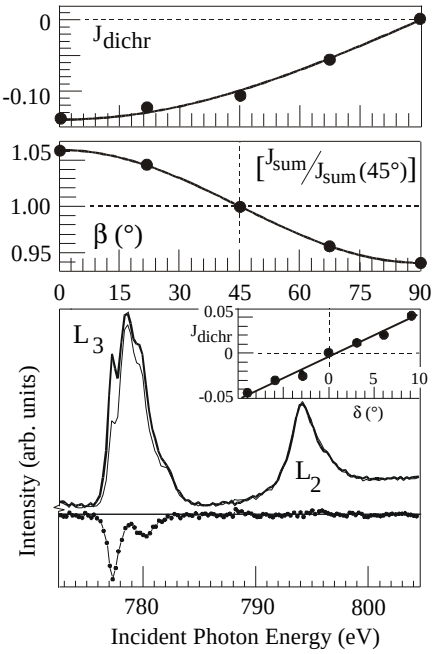
The **integrated RRS** machine based on filters was **successfully commissioned and used**. In particular integrated RRS (IRRS) MCD data were measured on Co metal polycrystalline, Co metal single crystal, Co^{2+} in the $CoFe_2O_4$ ferrite, Ni metal polycrystalline, Ni metal single crystal, Ni^{2+} in the $NiFe_2O_4$ ferrite (milestone 1). Those data have already been partially published. In particular in PUBLICATION 1 (see below) the experimental data served to confirm that the most advantageous decay channel in the RRS MCD in perpendicular geometry is the $3s \rightarrow 2p$ transition, as predicted by the theory. In PUBLICATION 2 the sum rule analysis for the RRS MCD was applied for the first time, allowing to estimate the values of the higher order charge and spin multipolar terms in the ground state of Co^{2+}, up to the 4^{th} order.

L. Braicovich, G. van der Laan, G. Ghiringhelli, A. Tagliaferri, N.B. Brookes, "Magnetic circular dichroism in x-Ray resonant Raman scattering in perpendicular geometry from $CoFe_2O_4$ and Co metal: comparison between valence and inner-shell channels", **Phys Rev B 66, 174435 (2002)**.



PUBLICATION 1 FIGURE 1

L. Braicovich, A. Tagliaferri, G. van der Laan, G. Ghiringhelli, and N.B. Brookes, "Sum rules in x-ray resonant Raman scattering: recovering Co ground state information in $CoFe_2O_4$ as a test case", **Phys Rev. Lett. in press**



PUBLICATION 2 FIGURE 2

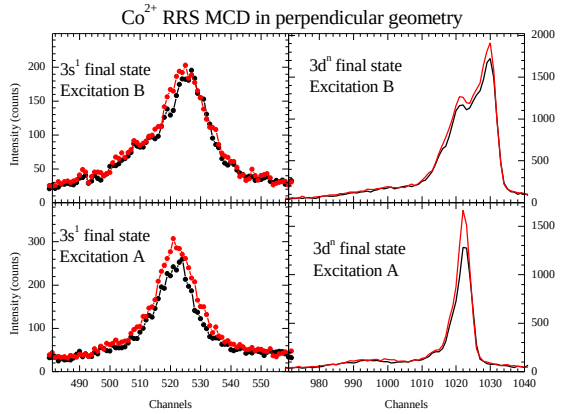


FIGURE 3

Moreover the comparison between Co^{2+} and Ni^{2+} shows a very different behaviour in the quenching of the orbital moment from the atomic value to the value found in the solid.

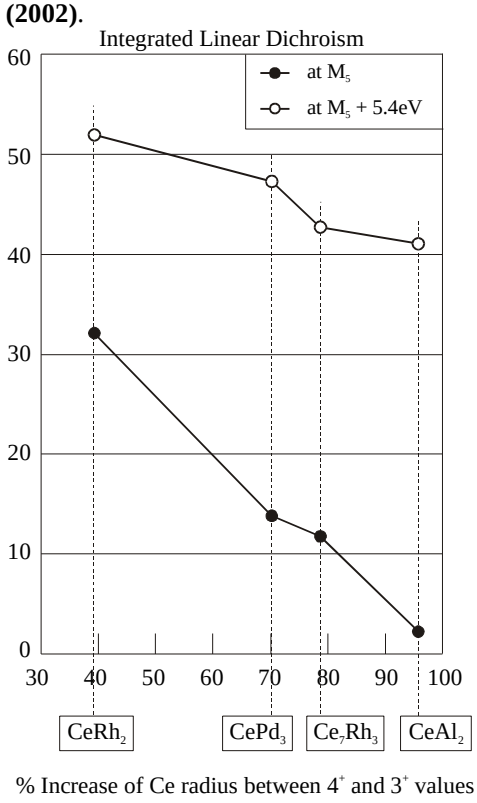
The **RRS MCD** was also measured at fixed scattering angle using the **AXES spectrometer**, i.e. taking full emission spectra when exciting at different energies across the L_3 absorption edge (milestone 1). In particular we collected data on Co metal polycrystalline and in Co^{2+} in the $CoFe_2O_4$ ferrite. An example of the data is shown in FIGURE 3. The data analysis is currently being made. This kind of measurements are potentially very informative, if an accurate comparison with model calculations is made. These spectra were take after the resolution of the emission spectrometer had been upgraded by replacing the two-dimensional position sensitive detector (see below).

LINEAR POLARISATION EFFECTS IN NON-FERROMAGNETIC COMPOUNDS

The use of linear dichroism in RRS as a probe of the **hybridisation in the Ce intermetallics** has proved to be very effective (milestone 2). The first results were published (PUBLICATION 3) and reported already in the activity report of the experiment HE983 (experiment performed just before the start of the LTP). Those data were taken with the old AXES set-up: despite a partly unsatisfactory resolving power the integrated intensity of the spectra could be used as a reliable probe of the Ce 4*f* state population.

We also performed some measurements on **single crystalline Ce compounds**. An example of the data quality is shown in FIGURE 5.

C. Dallera, M. Marcon, G. Ghiringhelli, A. Tagliaferri, N. B. Brookes, G. L. Olcese, A. Palenzona, and L. Braicovich, "Study of bulk ground state properties of cerium intermetallics by linear dichroism in 4f resonant inelastic X-ray scattering," **Solid State Communications 121, 635 (2002)**.



PUBLICATION 3 FIGURE 4

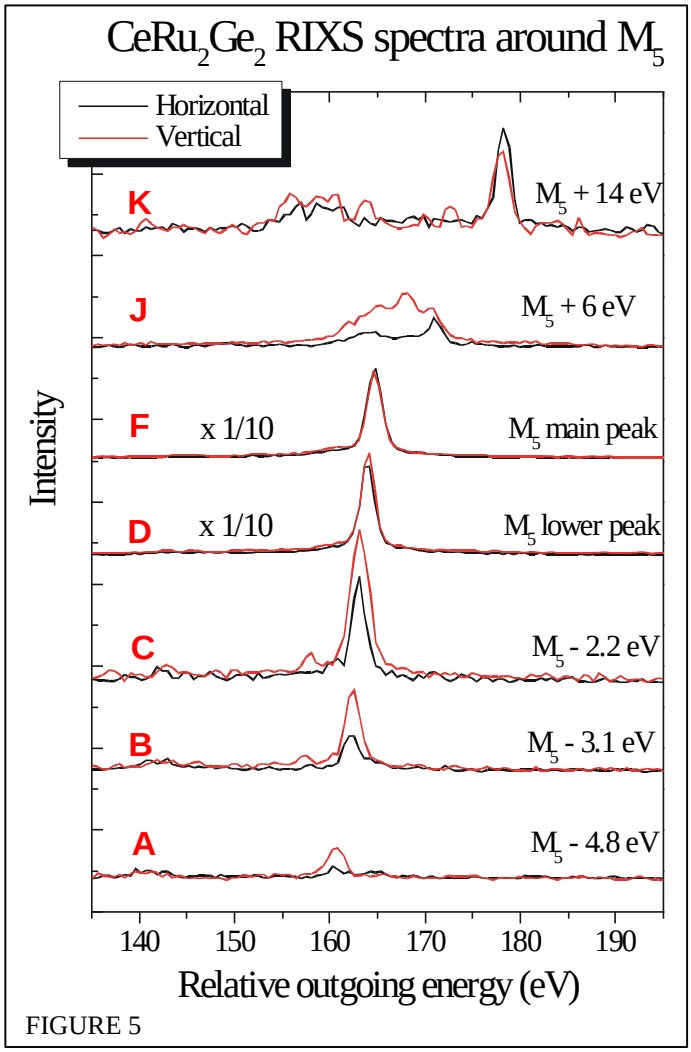


FIGURE 5

Linear dichroism in RRS was measured on a $Ce_{93}Sc_7$ polycrystalline sample as a function of the temperature, in order to study the α to γ phase transition of Ce (milestone 3). In this particular case the data appeared to be less sensitive than expected to the phase transition, and further investigations are needed on the sample quality and on the transition itself. In particular the transition has been recently studied using total electron yield and total fluorescence yield x-ray absorption spectroscopy. The phase transition appears very clearly both in the absorption and in the RIXS spectra.

INSTUMENTATION

IRRS instrumentation: The machine allowing the exploration of any scattering angle, based on filters, has been commissioned and satisfactorily used. The new machine based on a multilayer mirror as polychromator has been designed and is under construction (milestone 4). It is planned to be used during the fourth semester of the LTP (June and July 2003). The chosen multilayer will provide polarimetric capabilities over an energy range including Fe, Co, Ni $L_{2,3}$ and La and Ce $M_{4,5}$. We hope be able to explore this option already in the first days of operation (milestone 5).

RRS instrumentation (AXES with dedicated monochromator): As scheduled, in April 2002 the two-dimensional position sensitive detector of our emission spectrometer was replaced: instead of a micro-channel plate electron multiplier coupled to a resistive anode encoder (nominal spatial resolution > 50 microns) we are now using a back-illuminated CCD detector, Peltier cooled, with 20 micron pixel size. The new detector was previously characterised in Milano using a Cu-anode x-ray tube. The properties of the new detector required to set up a new procedure for extracting the spectra from the collected two-dimensional images. Moreover the data acquisition procedure was made fully controlled via the "SPEC" language used at ESRF: since then AXES has been **totally integrated in the beam line** and it is run as a standard beam line instrument, allowing the automation of the main alignment and acquisition procedures. **The gain in resolving power** has been dramatic (at least a factor of 3), as well as the increase in detection efficiency (higher than a factor of 5 if compared to the best performances achievable with the previous detector). See FIGURE 6 for a comparison of the LaF_3 RIXS spectrum taken with the two detectors. We have also succeeded in using the two undulators of ID08 together, with a further gain of 2.5 in intensity and no apparent degradation of the monochromator performances. Nevertheless at present the limiting factor is the dedicated monochromator, which had been designed to optimise the intensity and not the energy resolution. An upgrade of the monochromator is foreseen in the coming year.

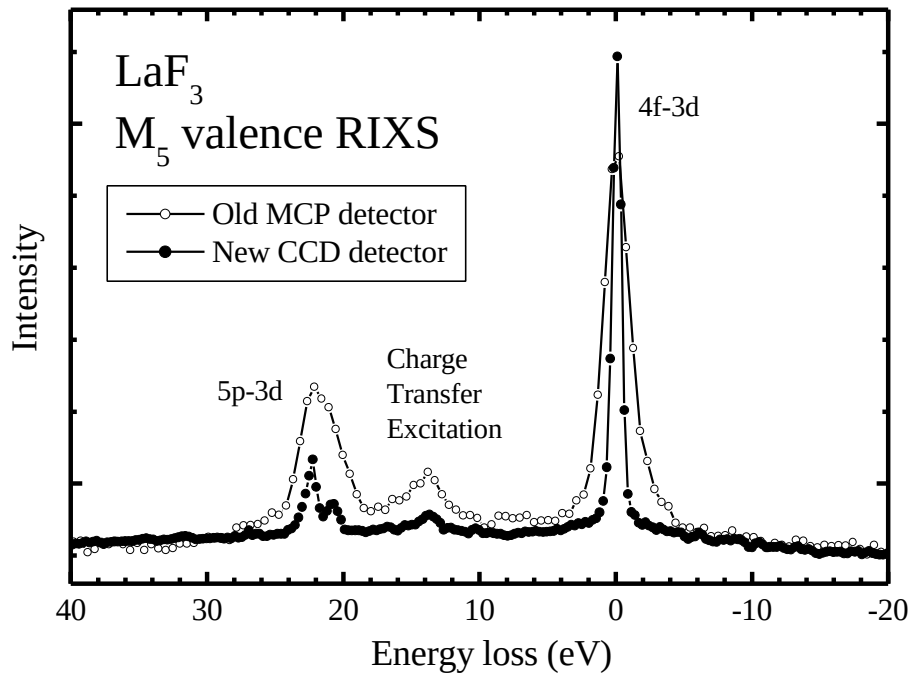


FIGURE 6