



Experiment title:
Magnetic circular dichroism in soft x-ray Raman scattering in the transverse geometry from transition metal systems: a new approach

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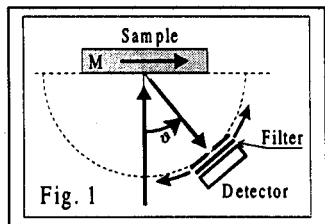
Report:

Integrated Resonant Raman Scattering

In 3d transition metals the radiative decay following a resonant excitation at the $L_{2,3}$ absorption edges shows two main high energy components. One leaving the system in a final state with valence excitations: $2p^63d^n \rightarrow 2p^53d^{n+1} \rightarrow 2p^63d^n$. And one characterised by a core hole in the final state: $2p^63d^n \rightarrow 2p^53d^{n+1} \rightarrow 2p^63s^13d^{n+1}$. Both emissions show the so-called resonant Raman behaviour when the incident photon energy is tuned across the absorption edges. In the soft x-ray regime the usual measurements are made by fixing the scattering angle and recording the emission spectra at fixed excitation energy. In this way the excitation energy is a parameter and one studies the evolution of the emission spectral shape.

We decided to adopt a different approach. Using a broad band pass photon analyser we can measure the integral intensity of the core-hole final state emission (inner shell emission). As the detector is small and the integral signal, though very weak, sufficiently intense it is possible in to scan the excitation energy and use the scattering angle as a parameter. We call this technique integrated resonant Raman scattering (IRRS).

With this connection we had previously measured, in the standard inner shell resonant Raman scattering, the magnetic circular dichroism in the so-called perpendicular geometry [1]. The sample is magnetised along the surface and orthogonal to the incident circularly polarised beam so that there is no absorption dichroism. Nevertheless this is not the case for the inner-shell Raman scattering, since the photon helicity, the magnetisation vector and the detection direction are in a non-symmetrical arrangement: reversing the magnetisation (or the photon handedness) can result in a difference in the emitted intensity.



We have investigated magnetic Cobalt in different conditions (polycrystalline Co metal thin film and Co in a CoFe_2O_4 ferrite single crystal film). The measurements were done with the experimental set-up sketched in figure 1. It is based on a large active area Si photodiode and a Fe film filter: the valence emission ($3d \rightarrow 2p$) is absorbed much more than the inner-shell emission ($3s \rightarrow 2p$). This is due to the fact that the $L_{2,3}$ absorption edges of Fe fall in between the two Co emissions.

Results and Discussion

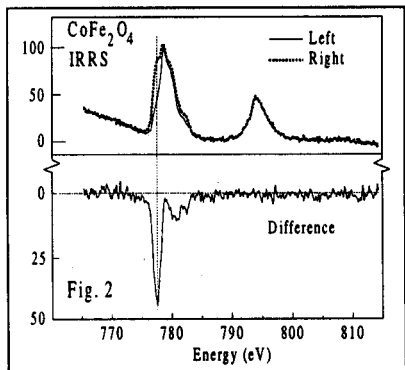
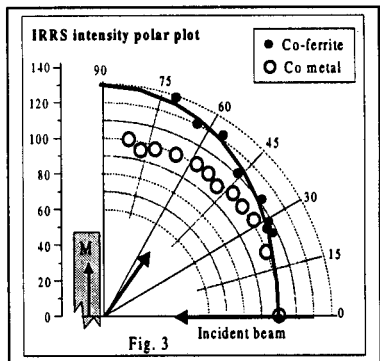


Figure 2 shows some typical Co-IRRS spectra of Co-ferrite taken at the emission angle $\theta=45$ degrees from the surface normal. The data show a rather strong dichroism in the L_3 region and zero dichroism in the L_2 region. In perpendicular geometry the dichroism at L_2 vanishes because if the core hole in the intermediate state has $j \leq 1/2$ the breaking of the mirror symmetry is then lost [2]. The absence of dichroism at L_2 is thus a support to the treatment given in ref. [2] and initially developed for resonant Auger emission. Accordingly to the theory the dichroism at L_3 can be traced back to the contribution of the quadrupolar terms in the multipole development of the ground state charge and spin wave functions. In figure 1 we restrict the scheme to a scattering in the horizontal plane, i.e. the plane defined by the incident beam and the sample magnetisation. The

combined action of the magnetisation and of the photon helicity makes the orientation of this quadrupolar distribution in the intermediate state non-symmetrical with respect to the mirror reflection by the vertical plane containing the incident beam. Thus the reversal of M (or of the helicity) gives origin to non equivalent reflected situations as seen by the detector when placed out of the mirror plane: a dichroism can be measured. Of course the displacement of the detector from the left to the right reverses the dichroism, giving a way of checking for instrumental asymmetries.

Extensive measurements have been made on Co ferrite and on Co metal, keeping the angle θ in the horizontal plane as a parameter. The main results are:

- (i) The dichroism is very much dependent on $h\nu_{in}$ at L_3 . The detailed calculations of this effect will be a very stringent test for any model used to describe resonant Raman scattering in magnetic systems.
- (ii) The angle dependence of the dichroism integrated over the L_3 edge follows a $\sin 2\theta$ law, in agreement with the sum rules developed in (2). This happens both in Co and in Co ferrite although the absolute values are different.



- (iii) The polar plot of the emitted intensity summed over the incident beam polarisations and over the whole L_3 peak is very different for the two samples. This is probably the most important and partly unexpected new result (see figure 3). The polar plot is basically isotropic in the metal and strongly anisotropic in the ferrite. The anisotropy can be ascribed to two distinct effects: the linear dichroism and the true quadrupolar moments of the ground state. The size of the asymmetry might suggest that the latter is really relevant in the Co ferrite. This result indicates that IRRS can be fruitfully used in the study of multipoles in the magnetic systems and in the comparison of localised vs. delocalised magnetic materials.

- References: [1] L. Braicovich *et al.* Phys. Rev. Lett. **82**, 1566 (1999).
 [2] G. van der Laan and B.T. Thole, J. Phys: Condens. Matter **7**, 9947 (1995).