Resonant X-ray Scattering Under Extreme Conditions on SmS and $CeFe_2$

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Introduction

Electron degrees of freedom promote structural modifications, transport effects, ferro-electricity, superconductivity, and magnetic phenomena such as spontaneous long-range magnetic order, magneto-resistivity, or magneto-electricity. Unravelling the ground state properties of these complex materials and their enigmatic modifications is advanced by the careful variation of the sample environment such as low temperatures, high magnetic fields and high pressures.
Resonant X-ray Scattering (RXS) is a site and shell selective technique and is based on the enhancement of the x-ray magnetic scattering cross-section close to an absorption edge, such as the K edges of transition metals, the $L_{2,3}$ edges of rare earths, or the $M_{4,5}$ edges of actinides. The resonant process involves the promotion of core electrons to empty intermediate states in the vicinity of the Fermi level and their subsequent decay accompanied by the emission of a elastic photon which reflects the electronic state of the magnetically ordered species. RXS thus represents a unique method to characterize magnetic interactions from the electronic point of view. Moreover, the resonant cross section has a characteristic polarization dependence, which brings further information on the magnetic moment direction and the space group of the ordered state.
This contribution is dedicated to the technical developments and experimental advances achieved for the realization of RXS under high pressure and high magnetic field measurements on the ID20 beamline at the European Synchrotron Radiation Facility (ESRF, Grenoble, France).

We present high pressure x-ray scattering experiments on single crystals of SmS, where the low temperature behavior is studied by fluorescence method, and Resonant Magnetic X-ray Scattering (RMXS) experiments on $Ce(Fe_{1-x}Co_x)_2$ where we show that there is a qualitative equivalence between the effects of pressure and cobalt doping in this system. In addition we show preliminary experimental results of the new high-magnetic field facility, which allows the application of a continuous magnetic field of 10 T at low temperature in the RXS regime.

Methods and Materials

RXS experiments were performed at the ID20 beamline (ESRF, Grenoble). Two undulators of period 35 mm provide photons with an high degree of horizontal linear polarization. A nitrogen-cooled Si(111) double monochromator inserted between two vertical focusing Si mirrors define a narrow energy window of 0.8 eV (FWHM) at 8 keV. The beam size at the sample position is 0.2x0.2 mm^2 for high pressure experiments (photon flux was about 10^{11} photons/sec), whereas for high magnetic field experiments the beam size is 0.5x0.5 mm^2. The energy was tuned to the Sm and Ce L_3 absorption edges for SmS and $Ce(Fe_{1-x}Co_x)_2$ experiments, respectively.

Hydrostatic pressure is created via a moissanite anvil cell allowing the possibility of high pressure in combination with large sample volumes. The scattering geometry for the pressure set-up on ID20 is unique with respect to other high pressure devices, since scattering occurs via the Be gasket which is transparent to x-rays in the energy range 3.5-12 keV, suitable for the RXS technique. The samples are cut to dimensions of 350x200x80 μm^3. Scattering is performed on a surface of 200x80 μm^2. In the case of $Ce(Fe_{1-x}Co_x)_2$, the crystal was cut with the scattering surface perpendicular to the <111> axis.
Pressure is transmitted via a hydraulic press, compatible with a standard orange cryostat via a guiding rod onto the pressure cell. The pressure medium most suitable for this set-up is liquid nitrogen which is loaded in a cryogenic dewar. The pressure is determined via the ruby fluorescence method with a fibre optic illuminating a ruby inside the anvil cell [1].
This device is well suited to perform a wide range of experiments requiring low temperature and low absorption, for example absorption and resonant spectroscopic techniques.
The application of a magnetic field is feasible in the second hutch EH2 of ID20, which accommodates the Huber 6-circles diffractometer, designed to support a superconducting split-pair magnet of 10T (Oxford Instrument Superconductivity). As well as the standard SIXC geometries, this diffractometer can be easily adapted to a wide variety of experimental geometries requiring heavy equipments such as cryostats or vacuum chambers for in-situ film grow.

Results

Both X-ray diffraction and fluorescence techniques where applied in the studies of structural and electronic transitions of SmS at low temperatures down to T = 4.5 K, and pressures up to 2.9 GPa. The application of pressure in the black phase of SmS has revealed a non-integer valence state for Sm with a linear pressure dependence and a value of 2.09 ± 0.02 just before the insulator/metal transition (Fig. 1).

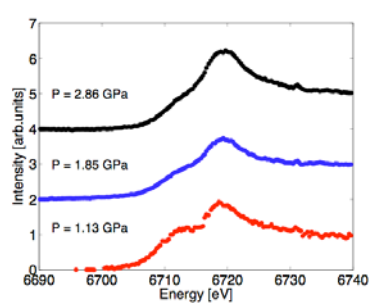

FIGURE 1: Fluorescence yield determined for various pressure in SmS [2].

A 13 % volume collapse at 1.13 GPa and 57 K leads to the gold phase where the application of pressure again linearly increases the valence state. These measurements are the first direct probe of the valence and structural state at the occurrence of the black to gold transition [2]. Of great interest is the observation of a non-trivalent Sm state above pressures where magnetically ordered SmS was determined.

The cubic Laves phase $CeFe_2$ is an itinerant system with a nominal ferromagnetic ground state and a Curie temperature of T_C = 230K. Inelastic neutron scattering experiments revealed the existence of antiferromagnetic (AF) fluctuations indicating that the system is close to an AF instability [3]. In fact the cobalt doped material $Ce(Fe_{1-x}Co_x)_2$ shows a discontinuous transition into an AF state, with a rhombohedral lattice distortion [4]. The Néel temperature of 69K for 7% Co doping, reaches a maximum of over 100K for the 15% doped compound, before decreasing. High pressure results, together with measurements of resistivity under pressure, show that high pressure is analogous to Co doping in this system. On the 7% doped system T_N is enhanced by applying pressure reaching about 130K at 2GPa, whereas in the 10% doped system T_N is rapidly depressed to about 37K at 1.6 GPa.

Fig. 2 shows recent experimental achievements in the detection of the small resonant magnetic signal at the Ce L_3 edge in 10 % doped $CeFe_2$.

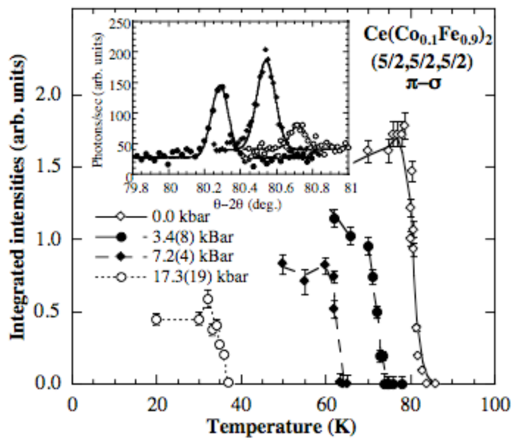

FIGURE 2: Temperature dependence of AF reflection (5/2,5/2,5/2) of 10% Co doped crystal of $CeFe_2$ for different applied pressures. The inset shows the corresponding θ–2θ scan taken at low temperature.

The application of the pressure on the 10% doped sample decreases the Néel temperature T_N from 81K (at P=0) to 38K (P=17.3 kbar), whereas for the 7% doped sample an increase of T_N from 70K at 0 pressure to 89K at 9.5 kbar. These results suggest the analogy between the pressure and Co-doping [5].

Fig.3 shows RXMS under high magnetic field performed on the two oriented crystals of $Ce(Fe_{1-x}Co_x)_2$ (7% and 10% Co doped) with the [1-10] axis along the magnetic field and the [111] axis in the horizontal scattering plane. The magnetic signal, as for the high pressure studies, was analyzed with a LiF(004) crystal and the magnetic intensities collected in the π–σ polarization channel. The magnetic phase diagram was studied for different applied magnetic fields, as shown in the inset of Fig.3. The results can be compared with the previous magnetization measurements of Ref. [7], in which some differencies have been found. Notice that in this case only the sublattice magnetization due to the Ce 5d electrons is probed, and that in general by RXMS technique it is possible to study the difference between sublattice magnetizations, for example by tuning the incident energy around the K-absorption edge of the Fe in order to probe the 3d states, exploiting thus the chemical and shell selectivity of this technique.

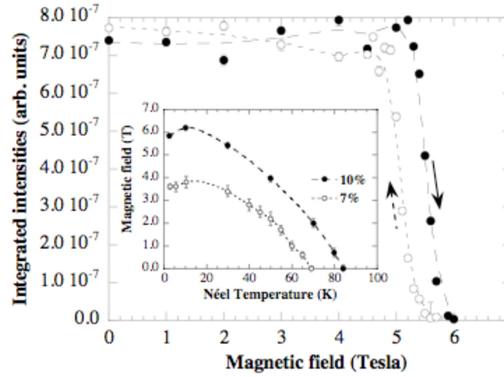

FIGURE 3: Field dependence of RXMS at Ce L_3 edge (E=5.721 keV) taken at T=2K on the same sample and of Fig. 2. The inset shows the magnetic phase diagram determined at for the 7% (open circles) and 10% (closed circles) Co-doped samples.

Conclusions

Combining extreme conditions such as high pressure, low temperatures and high magnetic fields with the RXS technique opens new possibilities to investigate and single out structural, magnetic and anomalous scattering. New exciting fields of research like the orbitally ordered systems, multiferroics, actinides, charge ordered systems can benefit of the use of these unique sample environment conditions.

These first results on the high pressure are encouraging and show clearly the feasibility of this technique, thus opening a possible large field of applications where low-temperatures and low photon energies are required. However there are still a few issues to resolve before we can provide it as a reliable user facility. In particular the anvil cell design and its coupling with the Be gasket should be improved in order to achieve reliable and higher pressures.

Further improvements of the focusing and stability properties of the beamline optics certainly will allow a future extensive use of high pressure technique in this domain, and a possible combination of both high magnetic field and high pressure setups.

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