ESRF	**Experiment title:** Low dimensional magnetism and topological magnetic frustration in the quasi one-dimensional oxide $Ca_3Co_2O_6$	**Experiment number**: HE2160
Beamline: ID20	**Date of experiment**: from: 12/04/06 to: 18/04/06	**Date of report**: 25/07/06
Shifts: 18	**Local contact(s)**: C. Mazzoli	*Received at ESRF:*

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Report: $Ca_3Co_2O_6$ (see fig.1) can be indexed in the hexagonal setting of the R-3c space group N.167. The point group symmetries at the positions occupied by the two Co^{3+} ions are Co_1 in -3 and Co_2 in 32. These two different symmetries have led to a number of different hypothesis for the spins S and valences of the two ions, with the most widely accepted configuration being $S(Co_1)=0$, $S(Co_2)=2$, so only the prismatic Co would be magnetically active. This frustrated system exhibits a very complex phase diagram and an unusual behaviour of the magnetization curves [1,2]

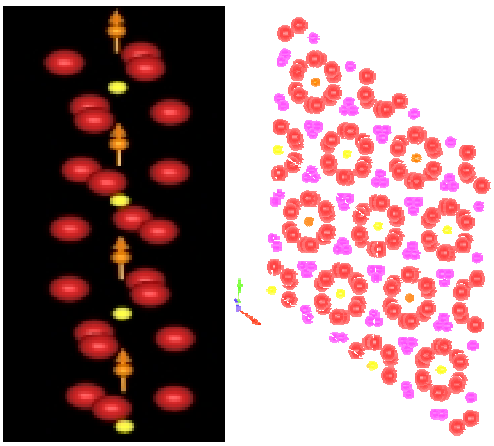

Fig.1: Two projections of $Ca_3Co_2O_6$: on the left the FM arrangement of the spins along the c direction, where the two Co ions are indicated in yellow and orange, and the O in red; on the right a view of the ab hexagonal planes where the triangular arrangement of the ions is evident.

The id20 beam line was aligned close to the Co K edge at 7.691 keV. The experiment was performed in the horizontal scattering geometry. In this experimental set-up the incident polarization is parallel to the scattering plane π. The polarization of the diffracted beam was linearly analyzed using the (222) reflection of a Au analyzer. The (110) diffraction surface of a 5x1x1 mm^3 single crystal sample of $Ca_3Co_2O_6$ was carefully prepared by polishing and etching the sample, which was then oriented with the hexagonal axis vertical, parallel to the magnetic field, and inserted in the id20 cryomagnet. In this configuration only the reflections with l=0 were accessible. The width of the charge reflections for all the investigated peaks was below 0.03 degrees underlining the excellent quality of the sample. In the magnetically ordered phase both in the resonant (E= 7.7028 keV) and the non-resonant condition, new reflections appear in the ππ channel, while no intensity was observed in πσ, violating the hexagonal extinction rules, and confirming the neutron experimental results [3]. The presence of non-resonant signal and the temperature dependence of the data make us confident that the signal is magnetic in origin. However, the huge resonant enhancement, unusually strong for a transition metal, suggests that there may also be a non-magnetic contribution to these reflections.

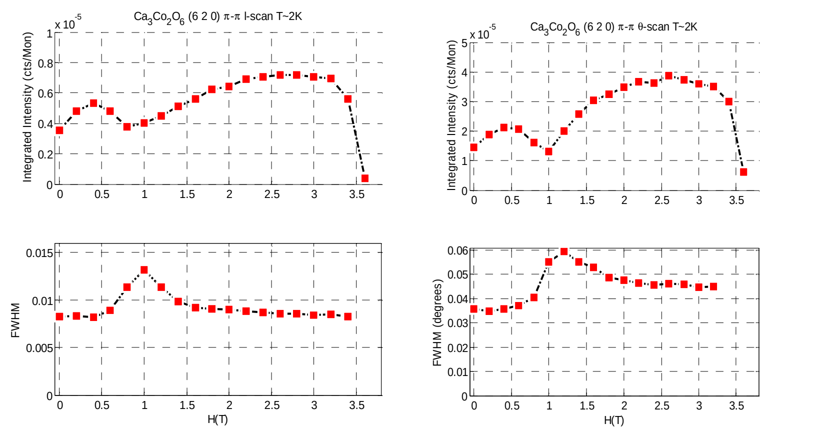

Fig.2:Dependence of the characteristics of the (6 2 0) reflection as a function of the applied magnetic field measured at base temperature, in the c direction (l-scan on the left) and in plane (θ-scan on the right, close to the (h –h 0) direction). At H=1T the magnetic correlation length decreases and the diffracted intensity goes through a minimum and starts to increase again as the frustration resulting from the in-plane interaction is compensated by the external field and the system gains energy aligning the moments along the field.

An intriguing aspect of the neutron experiment was the decrease in the integrated intensity of the magnetic peaks on cooling. We measured the same behaviour, but the higher Q resolution of the synchrotron light compared to neutrons, allowed us to explore the evolution of the FWHM of the forbidden reflections with temperature. Both the FWHM along the Ising chains and in the hexagonal plane increased on cooling; this points to an increase in the frustration driven magnetic disorder, rather than to the formation of several magnetic domains. The absence of any diffraction intensity in the πσ channel also strengthens the conclusion drawn from the neutron data that the moments point along the c-axis. On the other hand the measured FWHM is only slightly larger than the one measured on the charge peaks, pointing to a highly correlated system of spins both along the FM chains and in the hexagonal planes.

Applying a magnetic field to the sample produced no detectable change in the energy scans up to a field >3.5 T, where the sample entered the ferromagnetic state and the magnetic propagation vector changed. In contrast, the integrated intensity, the correlation lengths, and the centre of reflections depend on the magnetic field. The increase in the integrated intensity is consistent with the magnetisation measurements and with the reduction of the frustration due to the magnetic field.

Furthermore, at each temperature there seems to be a critical field value, H(T), where the centre of the peak starts to move in the l-direction and the FWHM goes through a maximum both in the l-direction and in the hexagonal planes, indicating a reduction in the length of the ferromagnetic chains and the size of the in plane correlations, before any spin reversal.

While the in-chain correlation length returns to the zero field value above H(T), the in-plane correlation lengths stays well above its initial value (see fig. 2). The effect on the position of the centre is also interesting as the centre position starts to move as the FWHM changes and then locks to a slightly different position after the FWHM change is complete. The evolution of the position of the centre of the peaks will require further investigation. In particular, in zero field we need to make sure that there is no incommensurability by measuring a large number of charge and magnetic reflections in a small number of applied fields. An lsr refinement of the UB matrix on several peaks might allow us to establish if a tiny displacement also occurs in the charge reflections, or if the features discussed above are purely associated with the magnetic signal.

This experiment has provided us with several new pieces of information about the magnetic moment behaviour in $Ca_3Co_2O_6$. However, this system is very complex. Due to the restrictions in the experimental geometry associated with the magnetic field, and to the limited amount of time available during one synchrotron experiment, we have not been able to collect all the information necessary to determine completely the origin of the effects observed. In conclusion, the study of the azimuthal dependencies of the XRS on some of the reflections measured in the present experiment, and access to a larger region of reciprocal space, on both charge and magnetic reflections, will open a new window onto this system complementing our preliminary results.

References

[1] S. Aasland et al., Solid State Comm. **101**, 187-192 (1997); [2] V. Hardy et al., Phys. Rev. B **70**, 64424 (2004) ; [3] O. A. Petrenko et al., Eur. Phys J. B **47**, 79-83 (2005).