ESRF	**Experiment title:** Low dimensional magnetism and topological magnetic frustration in the quasi one-dimensional oxide $Ca_3Co_2O_6$	**Experiment number**: HE2409
Beamline: ID20	**Date of experiment**: from: 25/04/07 to: 1/05/07	**Date of report**: 27/08/08
Shifts: 18	**Local contact(s)**: C. Mazzoli	*Received at ESRF:*

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Report:

The spin chain system $Ca_3Co_2O_6$, due to its combination of low dimensionality and geometrical frustration, has very complex magnetic properties and therefore it has attracted considerable attention in the past ten years [1].

The new findings of the experiment here reported show that this material is very interesting also for another peculiar feature: the possibility to observe a pure dipolar-quadrupolar E1E2 scattering signal. The present results will soon appear in ref. [2].

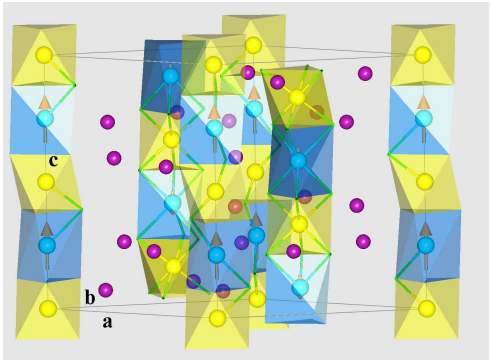

Fig.1: The magnetic and crystal structure of $Ca_3Co_2O_6$. The distorted octahedra are in yellow (light grey) and the trigonal prisms, are in light blue (dark grey).

The system, shown in Fig. 1, consists of chains made up of alternating distorted octahedra and trigonal CoO_6 prisms sharing faces, running along the hexagonal *c* axis and arranged in a triangular pattern within the *ab* plane [3]. The different local environments leave the Co^{3+} ions on the octahedral site (CoI) in a low-spin (S = 0) state, and those on trigonal prism (CoII) sites in the high-spin (S = 2) state. The local anisotropy of the trigonal prism forces the magnetic moments to point along the *c* axis [4].

$Ca_3Co_2O_6$, until very recently, has been described in terms of ferromagnetic chains coupled antiferromagnetically on a triangular lattice.

Our previous experiment performed on ID20 has led to the discovery of a small incommensurability in the magnetic propagation vector along the c-axis [5], bringing to a full re-evaluation of the role of the local exchange integrals in the description of the system [4]. Moreover, a new series of weak magnetic reflections with the same propagation vector but in position where the main magnetic contribution was expected to be zero, has been found.

The main aim of the present experiment was to carry out an azimuthal dependence study in order to determine the origin of the observed RXS signals.

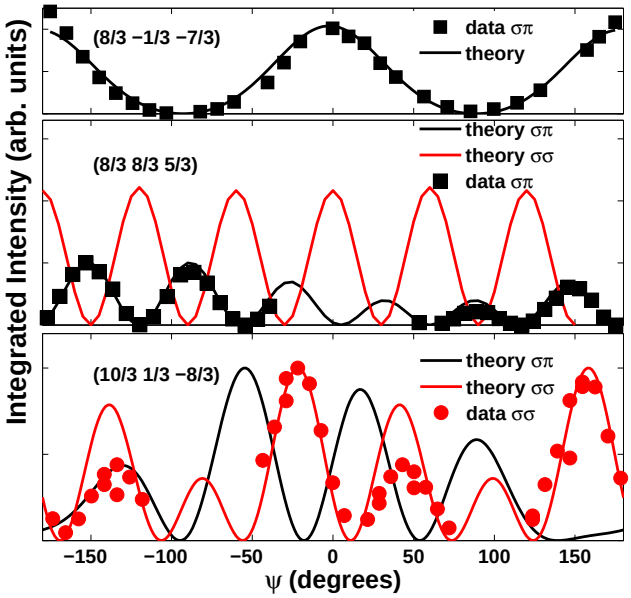

fig. 2 Theoretical and experimental azimuthal dependence of three magnetic reflections at the Co K-absorption edge. In the first panel an E1E1 reflection is reported, whereas the central and bottom panels show reflections resulting from the E1E2 interference together with the model predictions. The amplitude is the only free parameter in the fit.

$Ca_3Co_2O_6$ is usually described in the hexagonal setting of the *R-3c* space group, as this representation allows one to immediately identify the triangular arrangement of the CoO_6 chains within the *ab* planes. However, as this setting is non primitive, it makes both the description of the magnetic structure and the analysis of the symmetry of the tensors more difficult. For these reasons and with the advantage of naturally describing the incommensurability in the proper reciprocal lattice cell, in this report the rhombohedral axes are used throughout.

Single crystals of $Ca_3Co_2O_6$ were grown by a flux method. Two crystals with different orientations were used for the RXS experiments. The first crystal was 5x2x1 mm^3 with the largest natural face perpendicular to the (1,0,-1) reflection. The second crystal was polished to have a diffraction face perpendicular to the (1,1,1) direction. The beamline optics were optimized at 7.7 keV, close to the Co K-edge.

The sample was mounted in a displex cryostat. The diffractometer was operated in the vertical plane scattering mode with an azimuth set-up to allow for a sample rotation about the scattering vector. Hence, the natural polarization of the incident beam was perpendicular to the scattering plane. The integrated intensity of the reflections was measured using a photon counting avalanche photodiode detector. The polarization of the reflected beam was linearly analyzed by rotating the scattering plane of a highly oriented <00L> pyrolitic graphite plate.

The very different azimuthal dependences of the main magnetic reflection (8/3,-1/3,-7/3) and two representatives, (10/3,1/3,-8/3) and (8/3,8/3,5/3), of the new E1E2 reflections are reported in Fig. 2. The lines are fits to the data. In the (8/3,-1/3,-7/3) case only the time odd parity even tensors can contribute, whereas for the new E1E2 reflections only the time odd parity odd tensors contribute (for a detailed explanation see [2]).

Therefore the new reflections allow direct access to the dipolar-quadrupolar E1E2 scattering channel and are characterized by completely different energy and azimuthal (fig.2) dependences. The theoretical possibility of observing isolated E1E2 electromagnetic multipoles has attracted a lot of interest in the recent years. Unfortunately, in many system of interest parity even and parity odd tensor contributions occur at the same positions in reciprocal space. As evidenced by the results of the present work, in $Ca_3Co_2O_6$ it is possible to completely separate the parity even from the parity odd terms and measure for the first time a pure parity and time odd tensorial contribution from a E1E2 signal.

References

[1] Hardy V. et al, Phys. Rev. B **70**, 064424 (2004) and references therein.

[2] Bombardi A *et al*., to appear in Phys. Rev. B 78 Rapid Comm. (2008).

[3] S. Aasland et al., Solid State Comm. **101**, 187-192 (1997)

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[5] Agrestini S *et al*., Phys Rev B 77, 140403(R) (2008)