ESRF	**Experiment title:** Spin and orbital moments in the spin chain compound $Ca_3Co_2O_6$	**Experiment number**: HE-2869
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Report:

Magnetic Compton scattering was used to investigate the size and origins of the spin magnetic moment in the geometrically frustrated compound $Ca_3Co_2O_6$. Experimental results from previous XMCD and theoretical calculations give differing values for the spin and orbital moments: the theoretical study [1] predicted significant Co orbital moments and O spin moments, and the experimental measurements using XMCD [2] seemed to suggest a magnetic moment larger than that found by bulk magnetometry. Our magnetic Compton scattering result, in conjunction with bulk magnetometry, shows unamibguously that the spin and orbital moments are (3.93 ± 0.04) μ_B and (1.3 ± 0.1) μ_B per formula unit.

$Ca_3Co_2O_6$ is a geometrically frustrated compound that exhibits a number of magnetisation steps [3,4], with associated hysteretic behaviour, as a result of its magnetic structure [5]. The favoured electronic configurations of the Co ions at the two sites (octahedron and prism) involves high-spin Co^{3+} prism sites and low spin Co^{3+} octahedra sites, although other configurations have also been predicted [6]. In a recent calculation using the local spin density approximation + Hubbard U (LSDA+U), and including spin-orbit coupling, Hua Wu *et al.* [1] predicted that there may be a considerable orbital moment on the Co site, and a spin moment on the oxygen sites of $0.13\mu_B$, or $0.78\mu_B$ per formula unit and a large $1.57\mu_B$ orbital moment on the prism sites. A recent x-ray magnetic circular dichroism (XMCD) experiment [2] probed the Co moment, and reported an orbital moment of either $1.2\mu_B$ or $1.7\mu_B$, using two methods of analysis, essentially consistent with that predicted by Hua Wu *et al*. The modelling used in [2] gave a Co moment of $5.3\mu_B$ per formula unit, somewhat larger than the total $4.8\mu_B$ obtained from magnetisation measurements (and the total Co moment predicted by Hua Wu [1]).

Spin polarised Compton scattering samples the spin-dependent electron momentum density through the use of circularly polarised synchrotron radiation. The technique involves high-energy inelastic scattering of a monochromatic beam of circularly polarised photons E_I=200-250keV. The energy dispersion of the scattered beam is directly related to the electron momentum distribution. In this case, an energy of ~215keV was used, with a scattering angle of 174 degrees, which gives the optimal resolution and countrate. In order to extract the spin polarised signal two measurements were made with parallel and antiparallel applied field directions with respect to the scattering vector. This experiment used the new 9T cryomagnet that has been installed by the Warwick group (long term proposal HE1675 [7], and recent publication [8]).

The spin moment determined from measurements made in applied magnetic fields of 2T, 5T and 7T are presented in figure 1, together with the total magnetic moment as measured in a SQUID magnetometer on the same sample. The data were obtained with a sample temperature of T = 1.5K. It can be seen that there is a significant orbital moment at all fields. The data taken at 5T and 7T agree, demonstrating that the magnetisation was saturated. The orbital moment, of $(1.3 \pm 0.1)\,\mu_B$ per formula unit, is given by difference between the spin moment and the SQUID result. Further analysis and interpretation is currently underway in order to determine which orbitals are responsible for the magnetism and whether there is a significant oxygen *2p* moment. A paper will be written for publication over the next ~6 months.

References:

[1] Hua Wu *et al.*, "Nature of magnetism in $Ca_3Co_2O_6$", *Phys. Rev. Lett.*, **95** 186401 (2005).

[2] T. Burnus et al., "Valence, spin and orbital state of Co ions in one dimensional $Ca_3Co_2O_6$: an x-ray absorption and magnetic circular dichroism study", *Phys. Rev. B* **74** 245111 (2006).

[3] V. Hardy *et al.*, "Temperature and time dependence of the field-driven magnetization steps in $Ca_3Co_2O_6$ single crystals", *Phys. Rev. B*, **70** 064424 (2004).

[4] Y.B. Kudasov, "Steplike magnetization in a spin-chain system: $Ca_3Co_2O_6$", *Phys. Rev. Lett.*, **96** 027212 (2006).

[5] Y.B. Kudasov, "Magnetic structure and phase diagram in a spin-chain system: $Ca_3Co_2O_6$", *Europhysics Lett.*, **78** 57005 (2007).

[6] see for example R. Vidya *et al.*, "Tailor-Made Electronic and Magnetic Properties in One-Dimensional Pure and Y-Substituted $Ca_3Co_2O_6$", *Phys. Rev. Lett.*, **91** 186404 (2003); V. Eyert *et al.*, "Extended moment formation and magnetic ordering in the trigonal chain compound $Ca_3Co_2O_6$", *Chem. Phys. Lett.*, **385** 249 (2004).

[7] Experimental report HE1675.

[8] C. Shenton-Taylor et al., J. Phys.: Condens. Matter **9** 186208 (2007).

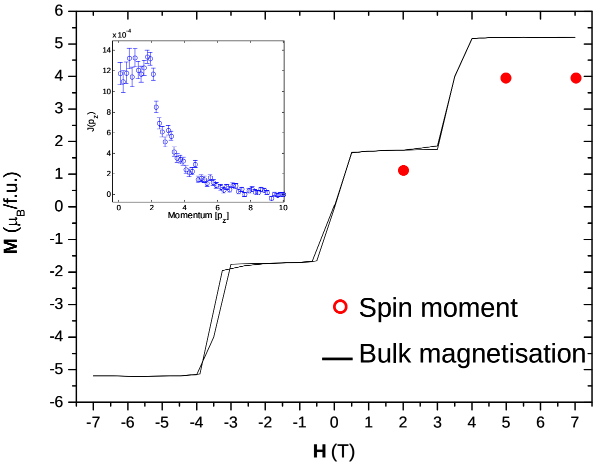

Figure 1. Experimental MCPs for $Sr_3Ru_2O_7$ measured in a field of 7T at 1.5K. Resolved along [100]: red (upper) data; resolved along [110]: blue (lower) data. Also shown are the LMTO calculations.

Moments in μ_B (f.u.)$^{-1}$:	Spin	Orbital	Total	Magnetometry
Theory [1]	Co 2.99 O 1.3	1.57 -	5.9	
XMCD (1) [2]		1.2		4.8
XMCD (2) [2]	3.6	1.7	5.3	4.8
Compton	3.93 ± 0.04	1.3 ± 0.1		5.2 ± 0.1