Experimental Report

Proposal: **5-53-229** **Council:** 4/2012

Title: Magnetic Short-Range order in beta-MnCo

This proposal is continuation of: 5-53-222

Researh Area: Physics

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Samples: Mn0.9Co0.1

Instrument	Req. Days	All. Days	From	To
D7	6	5	31/10/2012	05/11/2012

Abstract:

We propose to study the magnetic short-range order in single crystal beta-Mn0.9Co0.1 in order to compare the magnetic ground state with the previously measured high Co concentration, beta-Mn0.75Co0.25. Since - according to our model, the Co substituant atoms are non magnetic, and since Co doping doesn't change the lattice constant, we anticipate that the reduction in Co concentration will not affect the magnetic SRO greatly. This measurement will be crucial in confirming - or otherwise - this conjecture. We will need 6 days on D7 in order to measure 2 crystal orientations at low temperature.

β-Mn: ILL Annual Report

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At low enough temperature, most magnetic materials form a periodic magnetic order which minimises the energy of magnetic interactions. However, in so-called "frustrated" magnets, the geometry of the crystal structure prevents magnetic interactions from being fully satisfied—imagine three spins on a triangle, which can never be mutually antiparallel. Frustrated materials can remain in a paramagnetic phase down to very low temperatures. But these are not paramagnets in a conventional sense: spins remain correlated over short length scales, and these local correlations are thought to be implicated in phenomena as diverse as as emergent magnetic monopoles in spin ice and spin fluctuations in high-T_c superconductors [2].

We used the neutron instrumentation at the ILL to understand magnetic frustration in 20% Co-doped β-Mn. Pure β-Mn is of fundamental interest as the only element thought to exhibit a "quantum spin liquid" phase [3], but the availability of single-crystal samples makes closely-related β-MnCo a much better candidate for experiments. Unlike canonical frustrated magnets, β-MnCo is metallic, with a complex crystal structure but a simple chemical composition. Its crystal structure is primitive cubic with a unit cell containing 20 Mn atoms divided over two inequivalent crystallographic sites, only one of which is magnetic. Neutron diffraction measurements on diffractometer D15 at the ILL allowed us to determine the substitution of Co across the two Mn sites, revealing that Co substitutes essentially entirely onto the non-magnetic site. This result allowed us to simplify our analysis by considering only the magnetic lattice—a frustrated three-dimensional network of corner-sharing triangles.

Fundamental to our study was the polarized-neutron capability of spectrometer D7 at the ILL, which enables the magnetic scattering to be measured in isolation. Using the cryogenic facilities at the ILL we performed measurements at temperature $T = 0.05\,\mathrm{K}$. These measurements showed no sign of long-range magnetic order to this lowest accessible temperature, and the presence of highly-structured magnetic diffuse scattering patterns—shown in four reciprocal-space planes in Fig. 1—provides a key experimental signature of frustrated magnetism.

How, then, to interpret these data, and explain the lack of magnetic order in β-MnCo? The use of neutron polarization analysis allowed us to perform a fully quantitative analysis. We fitted the complete single crystal data using two techniques. First, reverse Monte Carlo (RMC) refinement, in which the orientations of 10^5 spins are iteratively refined to fit the experimental data, in order to identify the strongest magnetic correlations. Second, we used a mean-field approximation to fit a magnetic Hamiltonian to the data, identifying the strongest magnetic interactions. We found that the data can be well described using an effective Heisenberg model with two interaction parameters, antiferromagnetic nearest-neighbour J_1 and ferromagnetic 5th-neighbour J_5. The quality of fit to neutron scattering data obtained [Fig. 1 (bottom right panels)] is remarkable given the small number of parameters. This model also identifies a nearly-dispersionless branch in the Fourier transform of the magnetic interactions—a feature which corresponds to a large degeneracy of near-ground states, and therefore helps to explain the lack of magnetic order in β-MnCo.

We went on to ask how these interactions are manifest in the spin correlations obtained from RMC fitting. The relationship between J_1 and J_5 interactions is shown in Fig. 2. Each Mn atom is coupled by ferromagnetic J_5 interactions along helical chains [Fig. 2a]. Representing these chains as rods oriented along the helical axis maps a set of helices onto a triangular rod lattice. In this picture, the entire structure is described as four intersecting rod lattices. Mapping the β-Mn structure onto a rod

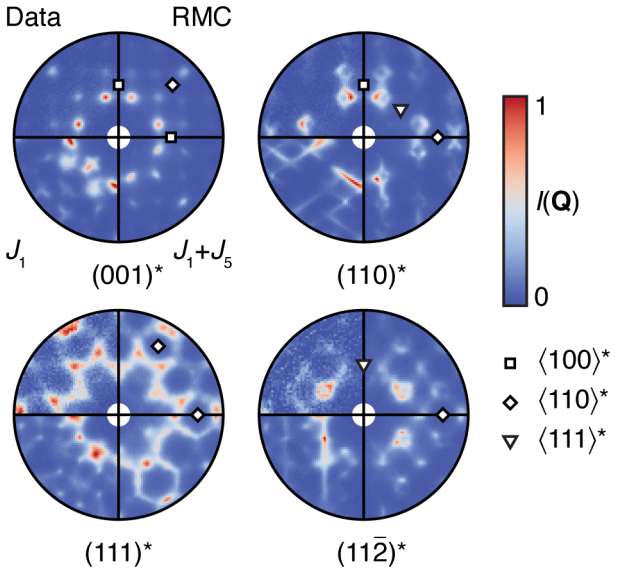

Figure 1: Magnetic diffuse scattering data and fits. Neutron scattering data were collected in four reciprocal space planes, clockwise from top left $(001)^*$, $(110)^*$, $(1\bar{1}2)^*$, and $(111)^*$. In each image, the top left panel shows data collected at 1.5 K, the top right panel shows the RMC fit to data, the bottom right panel shows the scattering for the J_1-J_5 model described in the text, and the bottom left panel shows the scattering assuming only antiferromagnetic J_1 interactions. Both J_1 and J_1-J_5 models are calculated using a mean field theory at simulation temperature $T = 1.01T_c^{MF}$, where T_c^{MF} is the mean field transition temperature. Whereas the J_1-only model is a very poor description of the data, both the RMC fit and the J_1-J_5 model give good agreement with experiment.

packing suggests considering the spin correlations in two different orientations: along the J_5 rods, and in the plane perpendicular to them. Spin correlations along J_5 rods are always ferromagnetic, with a simple exponential decay as a function of distance along the rod [Fig. 2b]. This simple distance dependence means that it is possible to interpret a rod as behaving like a single collective object, and to calculate the summed spin correlation value along each rod. These summed correlations alternate throughout the triangular rod packing in a pattern which strongly resembles an antiferromagnetic Heisenberg model on the simple triangular lattice [Fig. 2d (left panel)]. We therefore suggest that the comparable strength of J_1 and J_5 leads, over short length scales, to frustration of multi-spin rods on a triangular lattice. This emergent behaviour contrasts with the frustration of single spins on a hyperkagome lattice which would result from a J_1-only model [1].

The identification of strong magnetic frustration and emergent spin structures in such a chemically-simple system provides a valuable experimental reference point against which to benchmark developments in the theory of unconventional metals. An obvious avenue for future research is to identify nesting vectors of the Fermi surface in order to determine the origin of the interactions we have identified empirically. However, perhaps our key result is therefore to show how typical behaviour—the emergence of complexity from simple building blocks—can be reversed in a structurally-complex system such as β-MnCo. There may be hints here of a more

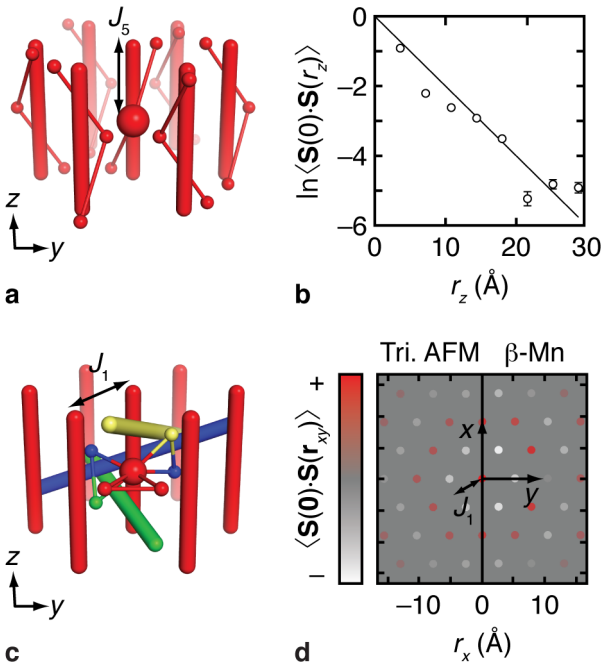

Figure 2: Structural relationship between J_1 and J_5 interactions, and impact on spin correlations. (a) Ferromagnetic J_5 interactions couple spins along helical chains, shown as thin red lines. The direction of propagation of each chain is shown as a thick red rod. The complete crystal structure can be described as four rod sublattices, each of which describes a triangular rod lattice. For clarity, only a single rod sublattice is shown. (b) Spin correlations $\langle \mathbf{S}(0) \cdot \mathbf{S}(r_z) \rangle$ along J_5 rods are always ferromagnetic, and decay exponentially with distance r_z along the rod. (c) Antiferromagnetic J_1 interactions, shown as thin lines, couple spins from adjacent rods. An arbitrary reference Mn atom, shown as a large red sphere, is coupled to two atoms within the same rod sublattice (coloured red); its four other nearest neighbours belong to the other three rod sublattices (coloured green, blue and yellow). (d) The right panel shows spin correlations, $\langle \mathbf{S}(\mathbf{0}) \cdot \mathbf{S}(\mathbf{r}_{xy}) \rangle$, perpendicular to J_5 rods. Values shown represent a sum along each rod, as described in the text. The spin correlations alternate between ferromagnetic and antiferromagnetic throughout the triangular rod lattice. The left panel shows spin correlations, $\langle \mathbf{S}(\mathbf{0}) \cdot \mathbf{S}(\mathbf{r}) \rangle$, for a frustrated antiferromagnetic Heisenberg model on the simple triangular lattice.

general phenomenon: the emergence of simple collective states within complex networks has also been observed in fields as diverse as neural signalling and the dynamics of amorphous materials. Hence a focus on structural complexity may prove an important experimental strategy for realising novel states of condensed matter.

[1] B Canals and C Lacroix. Mean-field study of the disordered ground state in the β-Mn lattice. *Phys. Rev. B*, 61:11251–11254, 2000.

[2] C Lacroix, P Mendels, and F Mila, editors. *Introduction to Frustrated Magnetism*, volume 164 of *Springer Series in Solid State Sciences*. Springer, 2011.

[3] H. Nakamura, K. Yoshimoto, M. Shiga, M. Nishi, and K. Kakurai. Strong antiferromagnetic spin fluctuations and the quantum spin liquid state in geometrically frustrated β-Mn, and the transition to a spin-glass state caused by non-magnetic impurity. *J. Phys.: Condens. Matter*, 9:4701–4728, 1997.