Experimental Report

Proposal: 4-01-1271 Council: 10/2012

Title: Magnetic interactions in sodium cobaltate with square and striped superstructures

This proposal is continuation of: 4-01-1141

Researh Area: Physics

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Samples: Sodium cobaltate / Na_0.8CoO_2

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Instrument	Req. Day	s All. Days	From	To
IN20	14	14	03/04/2013	08/04/2013
			13/05/2013	27/05/2013
			31/05/2013	06/06/2013

Abstract:

Previous studies of the spin wave dispersion of sodium cobaltate suggested a rather surprising 3D exchange model. Our measurements using polarised neutrons on IN20 of Na_0.77CoO_2 have allowed us to separate the magnetic excitations from the phonon background. The results suggest a key role for the sodium superstructure, and we are developing a more physical exchange model with much smaller interplanar coupling so that the system is, in fact, quasi-2D. We now propose to perform an inelastic neutron scattering study of Na_0.8CoO_2 using IN20 in polarised setup. This sample forms square or striped superstructures, depending on cooling rate. Thus we propose to measure the spin wave dispersion for two different superstructures on the same crystal.

Strongly-interacting electron materials away from stoichiometric compositions have a strong tendency to form emergent superstructures on a nanometer scale, typically involving spin and charge density waves with accompanying lattice distortions. The character of these systems can be fundamentally altered by even very small changes in composition. For example, superconductivity in $La_{2-x}Ba_xCuO_4$ reaches a maximum transition temperature T_c of 30 K at x = 0.10 and 0.15, but is almost completely suppressed at x = 0.125. This effect is due to a static superstructure of spin and charge stripes which forms on the CuO layers [1].

The strong interplay between the magnetic and superconducting properties of Na_xCoO₂ has led to close comparisons with the physics of the superconducting copper oxides [2]. It is clear from the spin-wave dispersions from crystals of Na_{0.82}CoO₂ [3] and Na_{0.75}CoO₂ [4], that the magnetic exchange is very sensitive to composition. These results are very surprising, since the nearest-neighbour interplanar exchange is found to be comparable to the intraplanar coupling between much closer neighbours, so that the system is 3D.

We have grown large single crystals of Na_xCoO₂ using the floating zone technique at Royal Holloway. We have used single-crystal neutron Laue diffraction to determine the long-range 3D superstructures of Na_xCoO₂ [5,6]. We have now measured the magnetic excitations of large single crystal of composition Na_{0.8}CoO₂ in a well-defined superstructure, the so-called square phase [5]. Measurements were performed using IN20 with unpolarised neutrons to map out the dispersion, and polarised neutrons using the XYZ technique to isolate the magnetic signal.

The spin-wave dispersion in the [HH3] direction is presented in Fig. 1. Here the colour map is the unpolarised, inelastic signal obtained via constant energy Q-scans. The white dots are the centres of Gaussian fits to these scans, the white line is the expected spin wave dispersion [3,4], and finally the black dots are the pure magnetic signal as obtained from polarised measurements. In this figure we can see that the data agrees with theory below ~12 meV, however above ~12 meV the gradient of the dispersion becomes extremely steep. The intensity is very weak at $E \sim 13$ meV, precisely the energy of the rattling mode phonon of Na_{0.8}CoO₂ in the square phase [7].

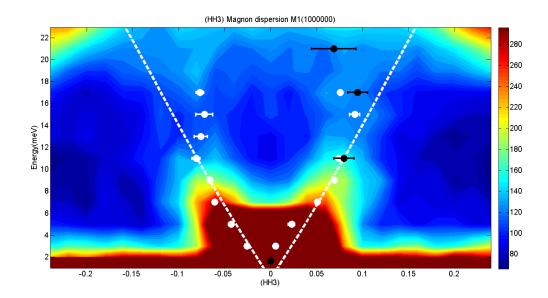


Fig. 1. Magnon dispersion for $Na_{0.8}CoO_2$ in the square phase. The colour map was obtained from unpolarised data, the white dashed line is the expected theoretical dispersion, the white points are fits to unpolarised data, and the black points are the fits to polarised data.

Scans were performed with polarised neutrons at a few selected points in order to confirm the magnetic nature of the scattering, see Fig. 2. These measurements demonstrate that the anomalously steep dispersion at high energies in Fig. 1 is magnetic. They also probe the spin-wave dispersion elsewhere in reciprocal space, and they allow the energy gap to be measured directly.

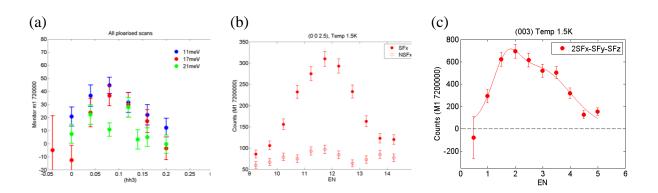


Fig. 2. Purely magnetic signal from $Na_{0.8}CoO_2$ in the square phase using polarised neutrons. (a) Q-scans along [HH3] at fixed energies 11, 17, and 21 meV, confirming the magnetic nature of the steep dispersion at high energies in Fig. 1. (b) Energy scan at the zone boundary along [00L]. (c) Energy scan at the zone centre, showing the energy gap.

We are currently modelling the data in order to understand the relationship between the magnetic excitations, the superstructure, and the phonon dispersion.

References

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