Experimental report

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Proposal: **4-02-491** **Council:** 10/2016

Title: Magnetic excitations in oxygen doped cobaltates

Research area: Physics

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Samples: La2CoO4+d

Instrument	Requested days	Allocated days	From	To
IN3	0	2	10/04/2018	12/04/2018
IN8	6	6	12/04/2018	17/04/2018
			28/05/2018	29/05/2018
IN12	0	1	19/06/2018	20/06/2018

Abstract:

Recently, we were able to develop a new explanation for the emergence of hour-glass magentic spectra in cobalt oxides. In our nano phase separation scenario, the hour-glass dispersion arises from strongly decoupled excitations within undoped and hole-rich regions of nanometer size. Whereas Co ions within the nanometer-sized hole-rich regions are coupled with weak exchange interactions J', the Co ions within the nanoscopic undoped islands are coupled with stronger exchange interactions J. In order to distinguish between two possible sub-scenarios of our nano phase separation model, we started to study oxygen-doped cobaltates. Compared to La2-xSrxCoO4 distinctly smaller exchange interactions J' appear in the La2CoO4+delta system which is a promising property. Our main goal is to find out whether the crossing of the hour-glass spectrum originates from an in-plane anisotropy gap within the undoped islands or from a crossing of dispersions. Due to the smaller exchange interactions in oxygen doped cobaltates this difference will become apparent in the magnetic excitation spectra which will enable us to answer this fundamental question of the origin of hour-glass spectra.

Magnetic excitations in oxygen doped cobaltates

An hourglass shaped magnetic excitation spectrum has recently been observed in the copper-free cobaltates that are isostructural to the high temperature superconducting cuprates. The microscopic origin of the suppression of the outwards dispersion was first explained by a scenario based on disordered charge stripe ordering [1]. However, our studies by means of neutron and x-ray scatterings show that there is no detectable volume fraction of the charge stripe ordering. Thus, the hourglass shaped magnetic excitations were alternatively accounted for by a novel nanophase separation model which consists of undoped La_2CoO_4–like and hole-rich $La_{1.5}Sr_{0.5}CoO_4$–like nanometer-sized islands [2-3]. Within our nanophase separation scenario, the superexchange interaction *J'* in the hole-rich regions is much smaller than the superexchange interaction *J* in the undoped region. In order to have a better understanding of the nanophase separation model, we extended our studies to the oxygen doped cobaltates $La_2CoO_{4+\delta}$. Our previous studies have shown that the superexchange interaction *J'* in the oxygen doped sample is smaller than that of the Sr-doped compound, while the *J* is presumably supposed to be unchanged with different doping levels. We can therefore study the magnetic excitations by tuning the *J'* systematically with different oxygen doping levels.

Our experiment was performed on the IN8 spectrometer equipped with an orange cryostat. Both, the monochromator and analyzer were doubly focused, and two PG filters were mounted behind the sample. Several superlattice reflections have been observed at 300 K at (H 1-H 0) with H = 1/4, ~0.4, 1/2, ~0.6 and 3/4, indicating a complicated structure. With decreasing temperature, these peak intensities increase - except the ones at H = ~0.4 and ~0.6. For an unambiguous clarification of the nature of these peaks, i.e., either magnetic or nuclear origin, future polarized neutron scattering measurements are needed.

Figure 1 shows the magnetic excitations measured at 2 K. The constant-energy Q scans were performed along the diagonal direction for the energies below 12.0 meV and along the (H 0.5 0) direction for energies above 15.0 meV. As can be seen, basic features of the hourglass shaped magnetic excitations can been observed. The outwards dispersions are suppressed at low energies, and disperse out again above ~15 meV. Figure 2 shows the energy scans at constant Q = (1.5 0.5 0). The resonance-like increase of intensity (magnon merging point) could be observed at E = 16 meV. In order to clarify the nature of these excitations, future measurements with polarized neutrons are also necessary in order to determine whether these excitations are in plane or out of plane excitations etc or magnetic at all. Measurements up to higher energies are also needed in order to compare with the Sr-doped cobaltates which show an anomalous out of plane excitations at about 40 meV.

References:

[1] A. T. Boothroyd, P. Babkevich, D. Prabhakaran, and P. G. Freeman, Nature **471**, 341 (2011).
[2] Y. Drees, D. Lamago, A. Piovano, and A. C. Komarek, Nature Commun. **4**, 2449 (2013).
[3] Y. Drees, Z. W. Li, A. Ricci, M. Rotter, W. Schmidt, D. Lamago, O. Sobolev, U. Rütt, O. Gutowski, M. Sprung, A. Piovano, J. P. Castellan, and A. C. Komarek, Nature Commun. **5**, 5731 (2014).

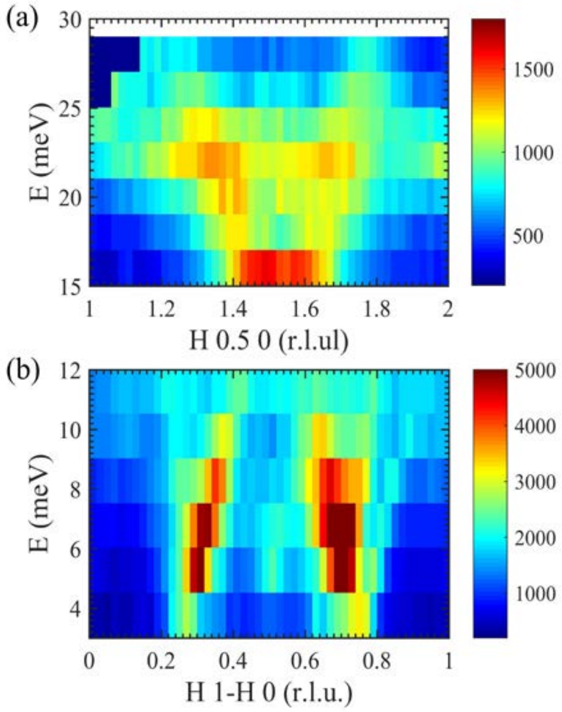

Fig.1 Inelastic neutron scattering intensities at (a) high and (b) low energies.

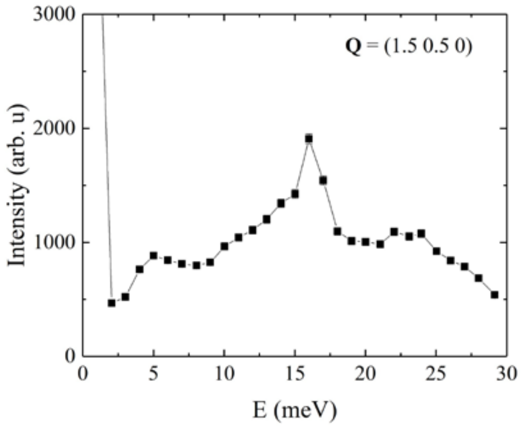

Fig. 2 Energy scans at constant momentum transfer **Q** = (1.5 0.5 0).