Experimental report

14/02/2018

Proposal:	**5-15-614**	**Council:**	4/2016
Title:	Charge correlations and Phase Diagram of La2CoO4+d		
Research area:	Physics		

This proposal is a new proposal

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Samples: La2CoO4.1, ... , La2CoO4.2

Instrument	Requested days	Allocated days	From	To
IN3	3	3	28/02/2017	03/03/2017
IN8	5	5	03/03/2017	08/03/2017

Abstract:

Recently, we were able to develop a new explanation for the emergence of hour-glass magentic spectra in cobalt oxides and to discard an alternative charge stripe scenario. In our nano phase separation scenario, the hour-glass dispersion arises from strongly decoupled excitations within undoped and hole-rich checkerboard charge ordered regions of nanometer size.
Besides the Sr-doped cobaltates La2-xSrxCoO4 we also started studying the oxygen doped counterpart La2CoO4+d now in order to test our nano phase separation scenario for a different system. Indeed, the exchange couplings are strongly reduced in the oxygen doped cobaltates. Surprisingly the magnetic excitations are also different from the ones in La2-xSrxCoO4. Therefore, La2CoO4+d is an important system to get further insight in our nanophase separation scenario with very different input parameters. So far, nothing is known about the charge correlations in La2CoO4+d, which is an important part (input parameter) of our nano phase separation scenario. Therefore, we propose to study the unknown La2CoO4+d phase diagram in detail now, especially with focus on the charge correlations.

Charge and magnetic correlations in oxygen doped cobaltates

The hourglass shaped magnetic excitation spectrum which is a ubiquitous property of the high temperature superconducting cuprates has been observed for the first time in a copper-free isostructural reference system recently - the insulating cobaltates $La_{2-x}Sr_xCoO_4$ [1]. The microscopic origin of these spectra has been attributed to disordered charge stripe phases [1]. However, our subsequent neutron and x-ray scatterings studies show that there is essentially no detectable volume fraction of a charge stripe ordered phase. Thus, the hourglass shaped magnetic excitations can be alternatively accounted for by a novel nanophase separation model based on nanometersized undoped La_2CoO_4–like and half-doped $La_{1.5}Sr_{0.5}CoO_4$–like islands [2-3]. Within our nanophase separation scenario, the exchange interaction *J'* in the hole-rich region is much smaller than the superexchange interaction *J* in the undoped region. Hence, the excitations in the hole-rich regions cannot follow that of the undoped ones up to higher energies. In order to gain a better understanding of the nanophase separation model, we extended our studies to the oxygen doped cobaltates, i.e. to $La_2CoO_{4+\delta}$. Our previous studies have shown that the superexchange interaction *J'* in the oxygen doped samples is smaller than that of the Sr-doped ones. Hence, the oxygen doped cobaltates give rise to the study of the magnetic excitations within our nano phase separation scenario for different values of the exchange interactions. *However, unfortunately, no hour-glass magnetic excitation spectrum has been discovered in any oxygen doped cobaltate so far.*

Here, we have synthesized various $La_2CoO_{4+\delta}$ samples with different oxygen contents that were characterized with different techinques including X-ray absorption spectroscopy measurements at the synchrotron. We studied the *charge and magnetic correlations* within these samples at the IN8 spectrometer with, both, the monochromator and analyzer being doubly focused and with two PG filters mounted after the cryostat. One of all studied samples turned out to be highly interesting:

Figure 1 shows first of all the elastic scans for that $La_2CoO_{4+\delta}$ sample along diagonal direction. As can be seen, several superlattice reflections can be observed at 300 K, thus, indicating more complicated structural properties than found in the $La_{2-x}Sr_xCoO_4$ system (with space group *I4/mmm*). With decreasing temperature, these peak intensities increase except for H = 0.38 and 0.62. Moreover, additional peaks centered at H = 0.3 and 0.7 appear. The increase of peak intensities and the appearance of additional peaks below ~30 K could be related to the magnetic correlations within this compound. In order to understand the origin of all these peaks, polarized neutron scattering experiments are needed.

Figure 2 shows the exciting outcome for the magnetic excitations in that $La_2CoO_{4+\delta}$ sample – the first observation of an hour-glass magnetic spectrum in $La_2CoO_{4+\delta}$. The outwards dispersing branches are heavily suppressed at lower energies, and, an outwards-dispersion can be observed again above ~15 meV. **Figure 3** shows an energy scan that is indicating a merging point at ~15 meV. In order to clarify the in-plane or out-of-plane nature of these excitations future measurements with polarized neutrons are necessary. Measurements up to higher energies are also needed for a comparison with the Sr-doped cobaltates which exhibit anomalous out of plane excitations at about 40 meV [3].

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).

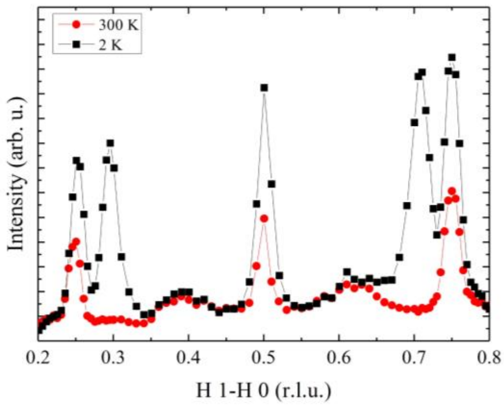

Fig. 1 Elastic neutrons scattering intensities along the diagonal direction at 300 K (red) and at 2 K (black).

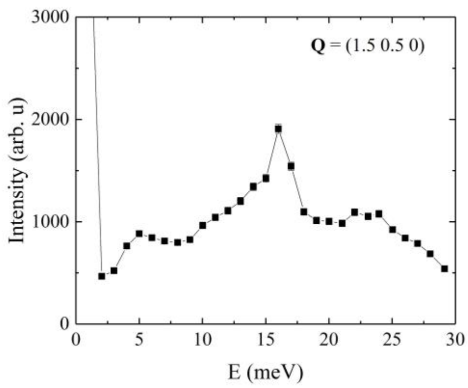

Fig. 3 Energy scans at constant momentum transfer **Q** = (1.5 0.5 0).

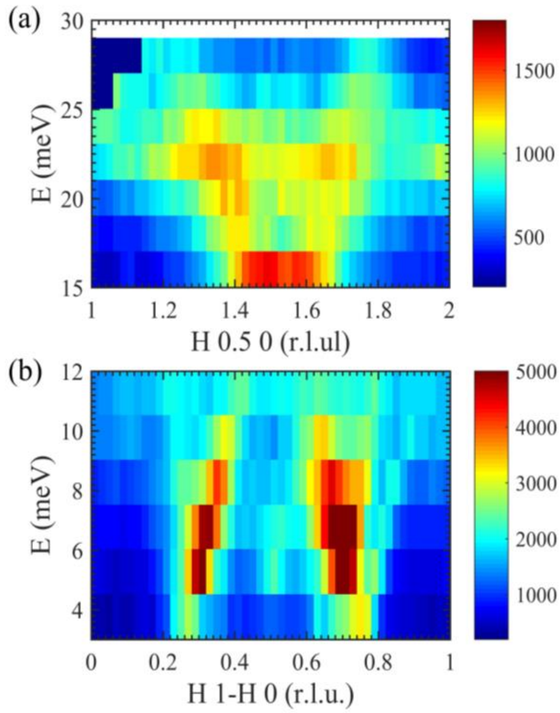

Fig. 2 Inelastic neutron scattering intensities at (a) high and (b) low energies.