# Experimental report

13/02/2019

**Proposal:** **5-41-949** **Council:** 4/2017

**Title:** Charge correlations in cobaltates with hour-glass spectra

**Research area:** Physics

**This proposal is a resubmission of 5-41-915**

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**Experimental team:** Hanjie GUO

**Local contacts:** Andrea PIOVANO

**Samples:** La2-xSrxCoO4 (x=0.25, 1/3, 0.4)

| Instrument | Requested days | Allocated days | From | To |
|---|---|---|---|---|
| IN8 | 6 | 6 | 29/05/2018 | 04/06/2018 |
| | | | 04/06/2018 | 05/06/2018 |

**Abstract:**

Cobaltates have attracted enormous attention due to the occurence of hour-glass magnetic spectra in these systems. The origin of these spectra is highly discussed until now. Initially 100% charge stripe phases were expected to occur in these materials. Then, it was shown that only checkerboard charge order can be found. Very recently, it was guessed that 50% charge stripe and 50% checkerboard charge correlations appear. Now, we found that the fraction of charge stripes has to be by far much lower than 50%, if existent at all. However, the important question arises, whether even fractions of charge stripe phases are needed for hour-glass spectra or not. This was recently claimed. In order to answer this fundamental question about the origin of the famous hour-glass spectra we need to measure the fractions of all occuring phases accurately now. Hour-glass spectra of different shape were observed for x=0.25, 1/3 and 0.4 in the past. Hence, we need to measure the charge correlations in all of these materials in order to miss no system with charge stripe correlations and in order to derive conclusions from the hole-doping dependence in a systematic way.

## Charge correlations in cobaltates with hour-glass spectra

The hourglass shaped magnetic excitations have been observed recently in a copper-free insulating cobaltate system that is 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 significant volume fraction of charge stripe ordered phases. Thus, the hourglass shaped magnetic excitations were alternatively accounted for by a novel nanophase separation model which consists of undoped $La_2CoO_4$ and hole-rich $La_{1.5}Sr_{0.5}CoO_4$ nanometersized islands [2-3]. Within our nanophase separation scenario, the superexchange 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 region cannot follow that of the undoped region up to higher energies. 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 experiments were performed on the IN8 and IN12 spectrometer with unpolarized and polarized neutron analysis, respectively. Both the monochromator and analyzer were doubly focused. Figure 1 shows the elastic unpolarized neutron intensities along the *HH*7 direction at various temperatures. Several superlattice reflections, which can be indexed with propagation vectors $\mathbf{k_1}$ = (0.25 0.25 0), $\mathbf{k_2}$ = (~0.4 ~0.4 0) and $\mathbf{k_3}$ = (0.5 0.5 0), can be observed. The peak intensities at $\mathbf{k_1}$ and $\mathbf{k_3}$ decrease with increasing temperature, while it is almost temperature invariant for that associated with $\mathbf{k_2}$. Moreover, the peaks at $\mathbf{k_1}$ and $\mathbf{k_3}$ exhibit distinct transition temperatures, with $T_{N1}$ ~ 30 K and $T_{N2}$ ~ 70 K, as shown in Fig. 1(b). The nature of these peaks is revealed by polarized neutron measurements performed on the IN12 spectrometer. As shown in Fig. 2, the peaks at $\mathbf{k_1}$ and $\mathbf{k_3}$ positions can be observed both in the spin-flip (SF) and non-spin-flip (NSF) channel with neutron polarization along the *x*-direction, indicating that there are both nuclear and magnetic contributions. On the other hand, the peaks at $\mathbf{k_2}$ position are only observable in the $NSF_{xx}$ channel, indicative for a nuclear

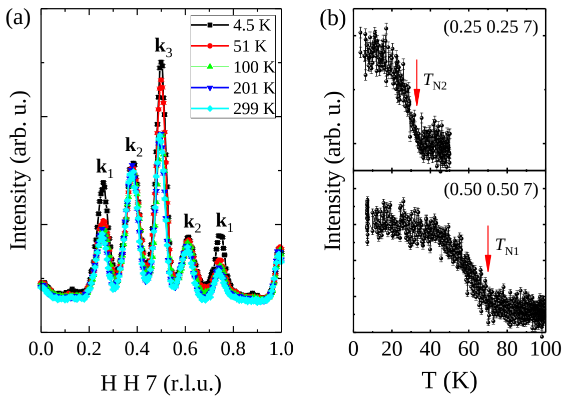


Fig. 1 Elastic neutron scattering measurements performed on the IN8 spectrometer.

origin of these reflections (like oxygen ordering).

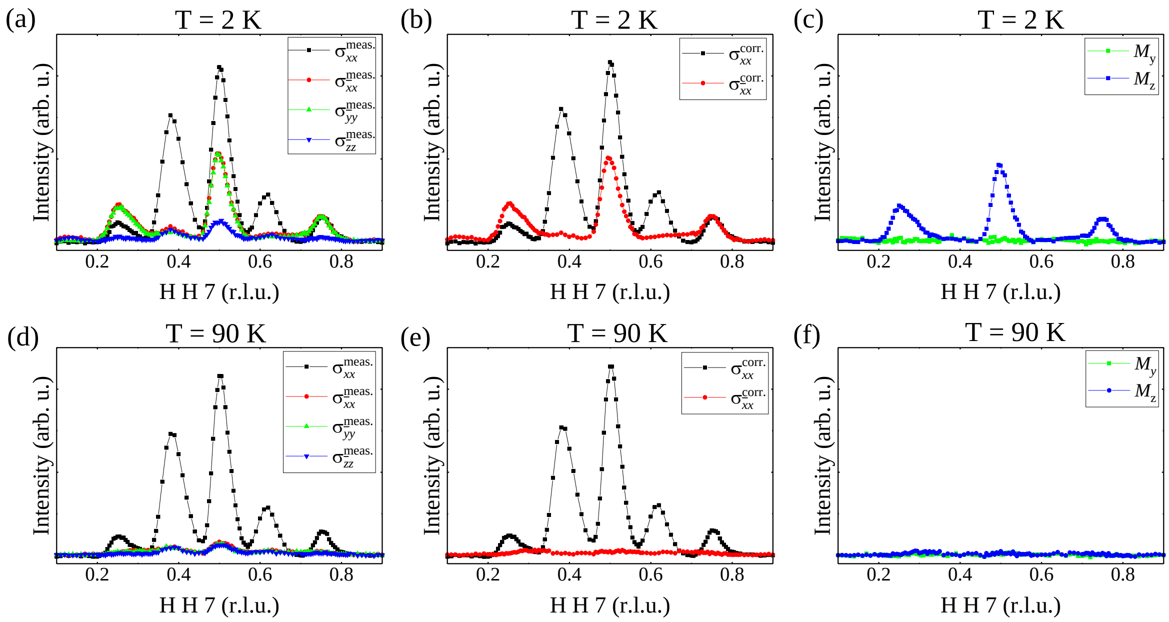


Fig. 2 Polarized neutron scattering measurements performed on the IN12 spectrometer.

The observation of two sets of entirely commensurate magnetic reflections at quarter- and half-integer positions in reciprocal space indicates a phase separation consisting of the checkerboard (CBCO) phase and phases with undoped $La_2CoO_4$-like islands. This is distinct from the Sr-doped cobaltates where only one set of incommensurate magnetic reflections was observed.

Fig. 3 shows the inelastic neutron scattering maps measured on the IN8 spectrometer. At low energies, the magnon dispersions disperses from the elastic **Q** positions associated with the CBCO phase. In contrast to a simple CBCO phase, the outwards dispersing branches are heavily suppressed. The inwards branches converge to the antiferromagentic zone center and disperse out again above ~16 meV. Thus, the shape of

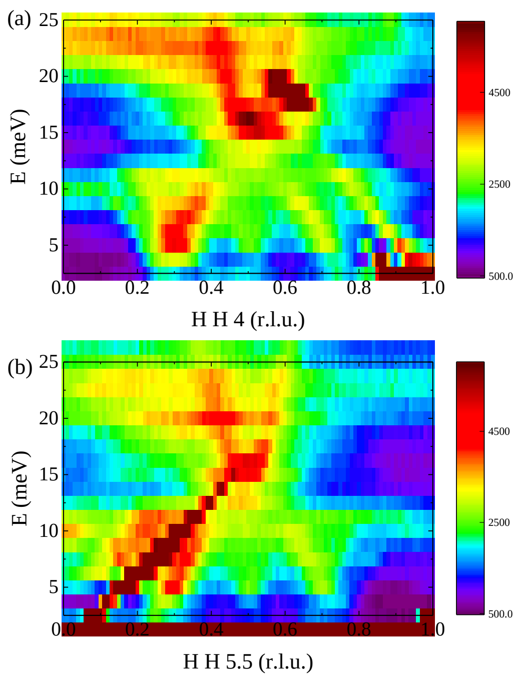


Fig.3 Inelastic neutron scattering intensity maps measured on the IN8 spectrometer at 2 K.

the magnetic excitations resembles the famous hourglass-shaped magnetic excitation spectrum of the cuprates that was also observed for the Sr-doped cobaltates $La_{2-x}Sr_xCoO_4$. **However, the observation of commensurate magnetic reflections at quarter- and half-integer positions in reciprocal space positions is different from these Sr-doped cobaltates.** And, this strongly supports the alternative stripe-free nanophase separation scenario as an explanation for the emergence of hourglass shaped magnetic excitation spectra, which is only based on undoped $La_2CoO_4$–like islands and checkerboard charge ordered regions.

**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).