Experimental report

03/02/2017

Proposal: **5-42-429** **Council:** 4/2016

Title: Probing the Charge Order Dimensionality in Hole-Doped Cobaltates

Research area: Physics

This proposal is a new proposal

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Samples: $La_{1.75}Sr_{0.25}CoO_4$
$La_{1.8}Sr_{0.2}CoO_4$

Instrument	Requested days	Allocated days	From	To
IN20	8	7	15/06/2016	22/06/2016

Abstract:

We propose to investigate the different types of short-range order (magnetic, structural and charge) present in $La_{2-x}Sr_xCoO_4$ with x = 0.20 and 0.25. Longitudinal polarized neutron diffraction will be used to separate the magnetic and non-magnetic diffuse scattering. Previous measurements performed on IN20 on a sample with x = 0.33 revealed a complex ground state in which incommensurate charge and magnetic stripe order coexists with commensurate checkerboard charge order. We aim to discover how the proportion of the incommensurate and commensurate ordered phases varies with doping. The results will clarify why the magnetic excitation spectrum has a characteristic hourglass shape similar to that observed in many copper oxide high temperature superconductors.

Probing the Charge Order Dimensionality in Hole Doped Cobaltates.

Understanding the origin of the universal "hourglass" magnetic excitation spectrum in cuprate superconductors is thought by many to be the key to determining the underlying magnetic interactions from which high temperature cuprate superconductivity is believed to originate [1]. The discovery that the isostructural antiferromagnetic insulator $La_{2-x}Sr_xCoO_4$ also has an hourglass shaped magnetic excitation spectrum established a solid empirical basis for theories of the hourglass spectrum based on short-range, quasi-static, stripe correlations [2]. Recent studies of $La_{2-x}Sr_xCoO_4$ for x = 1/3 and higher doping levels have however found that the charge order in $La_{2-x}Sr_xCoO_4$ is part checkerboard charge order, and part charge-stripe order [3,4]. The presence of two charge-stripe ordered phases has lead to a debate on the origin of the hourglass spectrum in $La_{2-x}Sr_xCoO_4$ [2-5], that is reminiscent of the present debate on the dimensionality of the charge order recently found in many different cuprate materials [6].

We have studied the phase diagram of $La_{2-x}Sr_xCoO_4$ over a wide doping range (0 < x < 0.9) by μSR and NMR techniques, and determined that there is a magnetically ordered phase below half doping that extends down to x = 0.2 [5,7]. In $La_{1.5}Sr_{0.5}CoO_4$, checkerboard charge ordered is believed to form a single ordered phase with a magnetic excitation spectrum that can fully be described within this model for spin-charge order [8]. At lower doping levels the checkboard charge order persists but there are insufficient holes in the material for checkerboard charge order to form a single phase[3]. Below half doping, on cooling the magnetic interactions gain strength as another charge-ordered phase develops in addition to the checkerboard charge ordered phase, and an apparent single magnetic phase is observed by μSR [4,5]. Questions to consider are whether the checkboard charge ordered phase can persist to x = 0.2, and whether a charge-stripe ordered phase predominates at low doping? Understanding the charge ordered structure at lower doping levels will allow a more definitive conclusion on the origin of the hourglass magnetic excitation spectrum in $La_{2-x}Sr_xCoO_4$ [2-5,9].

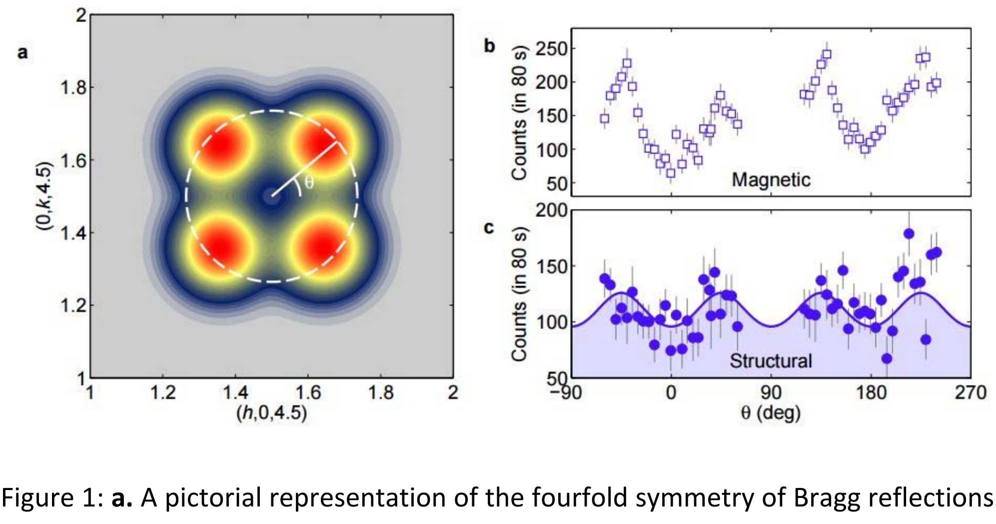
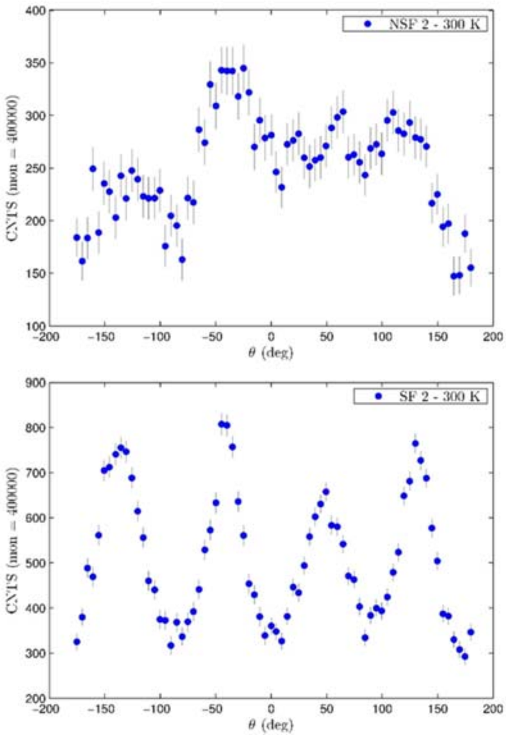


Figure 1: **a.** A pictorial representation of the fourfold symmetry of Bragg reflections from short range charge-stripe order in $La_{5/3}Sr_{1/3}CoO_4$ measured around (h,k,4.5). The path of a novel θ scan is indicated, this scan was achieved by performing an out-of-scattering plane scan by tilting the cryostat. The θ scan was performed using polarized neutron diffraction so that the structure of the **b**. magnetic Bragg reflections, and **c.** additional charge-stripe order Bragg peaks are separated. Both the magnetic and charge peaks have a fourfold symmetry consistent with charge-stripe order.

In our previous study of $La_{2-x}Sr_xCoO_4$ x = 1/3 we used polarized neutron scattering to separate the magnetic and structural diffraction signals in the (HHL) scattering plane of $La_{5/3}Sr_{x1/3}CoO_4$. Longitudinal polarization analysis was employed with the neutron polarization, **P**, parallel to the scattering wavevector, **Q**, so that elastic spin-flip scattering is from magnetic order and non-spin flip scattering from structural order (after corrections for imperfect neutron polarization are applied). Consistent with published results on the charge ordered structure of $La_{1.6}Sr_{0.4}CoO_4$, we observed checkerboard charge order at 300 K [8]. We repeated the same scans of the charge order structure at 2 K, then performed a data subtraction of the 300 K data from the 2 K data, and this revealed two incommensurate structural peaks consistent with charge-stripe order. In figure 1 we show a circularscan we performed out of the (HHL) scattering plane that confirmed a fourfold symmetry of the magnetic order and the additional charge ordering signal observed at low temperature/. The fourfold symmetry is consistent with charge- stripes running parallel to [110] and [-110].

In the new experiment we studied the magnetism and charge order of $La_{2-x}Sr_xCoO_4$ x = 0.25, to understand how the order develops well away from the single phase checkerboard charge order of the x = 0.5 [8]. The sample we studied is the same crystal as used in our previous study of the magnetic excitations, for which studies of the low energy excitations were restricted by 4 meV FWHM resolution [9]. Again we used polarized neutron diffraction to separate the magnetic and structural signal, and subtracted the 300 K data from the 2 K data. From these scans we observed a signal consistent with charge-stripe order in several different Brilliouin zones. In figure 2 we show the circular scan out of the (HHL) scattering plane of the fourfold symmetry of both the magnetic order and charge-stripe order. We also observed evidence for charge-stripe order in scans parallel to the (HH0) direction.



a

b

Figure 2: $La_{2-x}Sr_xCoO_4$ x = 0.25 a θ-scan around (0.5,0.5,6.6) where the 300 K data have been subtracted from the 2 K data in **a.** NSF, **b.** SF (bottom) channels. The NSF and SF scattering show a fourfold symmetry consistent with charge-stripe order, where the charge order is observed in the NSF channel, and the magnetic order is observed in the SF channel.

Additional to the study of the ordering in this material, a short survey of the lowest energy magnetic excitations was performed to complete measurements of the magnetic spectrum reported at higher energies in reference [9].

The results of this proposal will allow a comparison of the charge-stripe order variation in $La_{2-x}Sr_xCoO_4$ between the x = 1/3 and 0.25, and suppression of the checkerboard charge order. Initial results indicate that there is distinct differences in the charge ordering and accompanying magnetic order of these two materials. Preliminary analysis reveals that the charge-stripe scattering of the x = 0.25 sample to be of a more diffuse nature than that at x = 1/3, and charge order begins to develop at a temperature which is ~50 K lower in the x = 0.25 material compared with x = 1/3. A detailed of analysis of the these results is required before any conclusion can be drawn on which model of the magnetic excitations in $La_{2-x}Sr_xCoO_4$ is more appropriate for describing the origin of the hourglass shaped magnetic excitation spectrum in this material.

References;

[1] S. M. Hayden *et al*., Nature **429**, 531 (2004); J. M. Tranquada, *et al*., Nature **429**, 534 (2004); + many other papers.

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