Experimental report

03/09/2019

Proposal:	**4-02-539**	**Council:**	4/2018
Title:	Temperature Evolution of the Magnetism of Charge Ordered La2-xSrxCoO4x = 0.25.		
Research area:	Physics		

This proposal is a continuation of 5-42-429

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Samples: La1.75Sr0.25CoO4

Instrument	Requested days	Allocated days	From	To
IN8	4	4	07/09/2018	11/09/2018

Abstract:

The occurrence of charge-stripe order and checkerboard charge order in La2-xSrxCoO4 has lead to a stripes versus checkerboard charge order-Neel antiferromagnet nano-phase separation debate on the origin of the hourglass shaped magnetic excitation in this material[1-3]. Resolving this dispute may prove crucial for understating the universal hourglass shaped magnetic excitation spectrum of hole doped cuprate superconductors. The key to resolving our understanding of La2-xSrxCoO4 is to resolve the structure of the charge ordered phase to lower doping levels, further away from the pure checkerboard charge order at half doping.

Nearly 25 years after the discovery of charge-stripes in a hole doped cuprate by J. M. Tranquada et. al., the importance of charge order, the dimensionality of this order and how it effects the magnetic interactions in the cuprates remains intensely debated. The observation of an hourglass shaped magnetic excitation spectrum in insulating charge ordered La2-xSrxCoO4 (LSCoO) was thought to be a breakthrough in understanding the universal hourglass magnetic excitation spectrum in cuprate high temperature superconductors [1]. Later studies of the charge ordering in LSCO revealed an apparent competition between charge-stripe and checkerboard charge order in LSCoO for x ≥ 1/3[2,3]. This has led to a vibrant debate on the origin of the hourglass magnetic excitation spectrum in LSCoO as due to either spin –stripe interactions or nanophase separation between checkerboard and antiferromagnetic nanophases[2,3]. We undertook this study of LSCoO x = 0.25 to further enlighten our understanding of the magnetism of charge ordered LSCoO.

Undoped La2CoO4 is a Mott insulating antiferromagnet with Co2+ in a high S = 3/2 spin state. In LSCoO substitution of La by Sr dopes holes into the Co-O layers resulting in Co3+ in a low spin state S = 0. When there is sufficient holes doped into the material the Co3+ form charge ordered structures[2,3]. Lines of regularly spaced charges at 45 degrees to Co-O bonds; charge stripes, or charge order where every other site is part of a checkerboard like arrangement; checkerboard charge order. At lower temperature the S = 3/2 spins of the remaining Co2+ antiferromagnetically order, the question of how the interactions between these ordered spins produce the hourglass magnetic excitation is the question of debate in LSCoO.

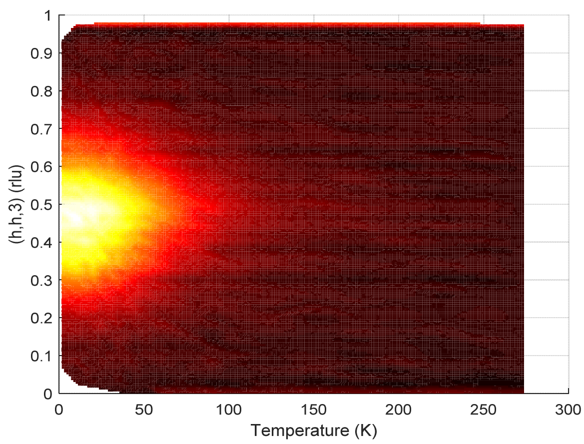

Figure 1: Scans of the antiferromagnetic magnetic peak in LSCoO x =0.25 versus temperature.

For LSCoO x = 0.25 our IN20 study found charge-stripe order with a stripe spacing expected at one third doping and checkerboard charge order as expected at half doping, with both charge orders having a similar temperature dependence as observed in the x = 1/3[3]. Strikingly the magnetic order was observed to be located at the commensurate antiferromagnetic wave vector rather than at incommensurate positions expected for charge ordered phases, although the magnetic excitation spectrum is reported to have an hourglass shaped excitation spectrum [4]. The spin polarization of this order and the accompanying magnetic excitations were observed to be constant across the whole broad peak. To determine whether the broad magnetic order peak is from a single ordered magnetic phase, or not, we studied the temperature dependence of the magnetic excitations across the magnetically ordered phase to approximately three times the magnetic ordering temperature.

In Figure 1 we show a surface plot created by scans in (hh0) direction at l = 3 of the magnetic Bragg peak in the x = 0.25. The transition to the unordered phase is gradual, with an ordering temperature of approximately 100 K with critical scattering surviving to over 150 K. Detailed analysis of the temperature evolution of this Bragg peak will be performed to identify any significant peak shape change. The transition from the Low Temperature Orthorhombic (LTO) phase into the high temperature phase on increasing temperature is also observed to be ill defined. In figure 2 we can see that the LTO structural distortion peak gradual disappears in the 150-200 K temperature range.

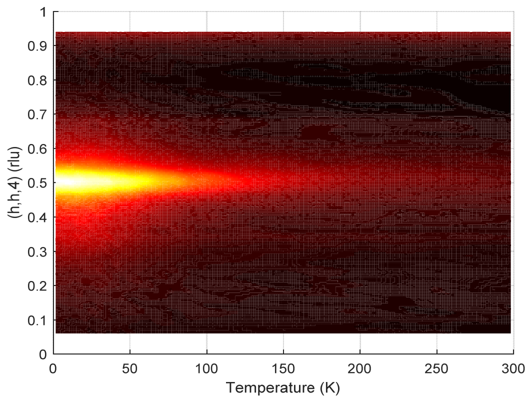

Figure 2: Scans of the Low Temperature Orthorhombic Distortion peak of LSCoO x = 0.25 measured at different temperatures.

After establishing the detailed temperature evolution of the magnetic order and the structure of the material we undertook measurements of the temperature dependence of the magnetic excitations of the x = 0.25. The temperature dependence of scans parallel to (hh0) of the magnetic excitations at different points in the hourglass magnetic excitation spectrum were performed, selected from our knowledge of the magnetic excitation spectrum [4]. In figure 3(a) we show the scans of the magnetic excitations at an energy of 7 meV. With increasing energy transfer the magnetic excitations are observed to anomalously sharpen with increasing temperature until approximately 150 K, where the excitations broaden with increasing temperature as expected for paramagnons in the magnetically unordered phase. From fits of the magnetic excitations with a Gaussian line shape on a tilted background the temperature evolution of the width of the magnetic excitations was obtain, as shown in Fig. 3(b). The magnetic excitations from an anitferromagnet should slowly broaden on warming towards the spin ordering temperature, above which they should be observed to broaden significantly. The broadening of the magnetic excitations above 150 K in LSCoO x = 0.25 behave as expected, but not the sharpening of these excitations on warming towards 150 K.

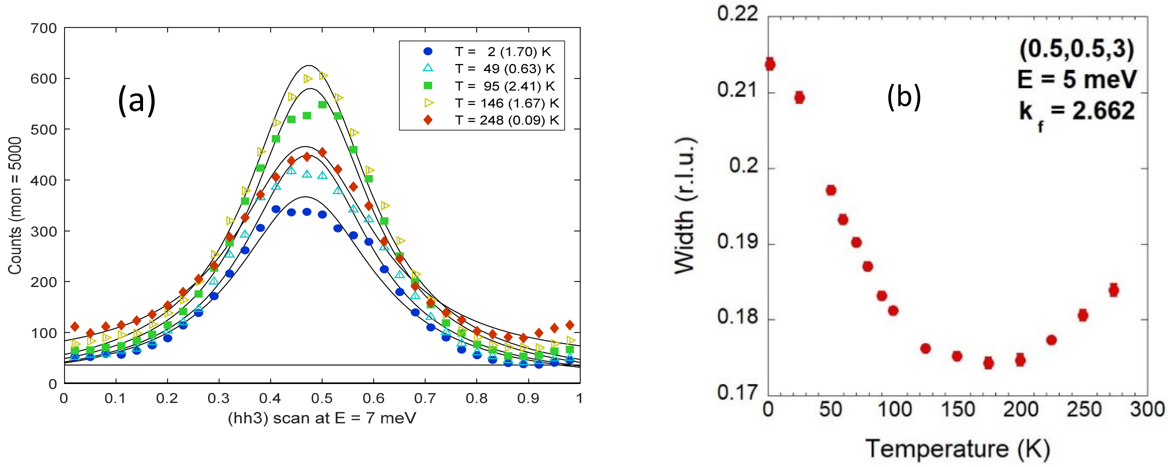

Figure 3: (a) Scans of the antiferromagnetic magnetic excitations at E = 7 meV of LSCoO x = 0.25 measured at different temperatures. (b) The temperature evolution of the Gaussian width of the magnetic excitations at E = 5 meV of LSCoO x = 0.25.

The results of our measurements of LSCoO x = 0.25 via polarized and unpolarised neutron scattering reveal the charge and spin ordering process of this material to be complicated. We are presently performing detailed analysis of the results of these studies, and considering theoretical models that can provide an understanding of charge and spin processes occur in these materials.

References:
[1] A. T. Boothroyd, P. Babkevich, D. Prabhakaran and P. G. Freeman, Nature **471**, 341 (2011)
[2] Y. Drees et al., Nature Commun. **4**, 2449 (2013)
[3] P. Babkevich, P. G. Freeman, M. Enderle, D. Prabhakaran, and A. T. Boothroyd, Nature Commun. **7**, 11632 (2016).
[4] S. Gaw, et. al., Phys. Rev. B **88**, 165121 (2013).