Experimental report

12/09/2019

Proposal: **4-02-538**

Council: 4/2018

Title: Intermediate spin phase of charge-stripe ordered La1.63Sr0.37NiO4.

Research area: Physics

This proposal is a new proposal

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Samples: La1.63Sr0.37NiO4

Instrument	Requested days	Allocated days	From	To
IN12	7	7	17/10/2018	24/10/2018

Abstract:

Striking features of the temperature evolution of the magnetism of charge-stripe ordered La2NiO4.11 have been revealed by high resolution cold neutron scattering. A wave vector offset that is not understood by theory, and a kink in the magnetic excitation dispersion near the spin ordering temperature. We argue that our findings indicate an intermediate spin phase, that will be effected by the changing symmetry observed in the magnetism of La2-xSrxNiO4 above one third doping. The offset between magnetic order and magnetic excitations is highly similar to that reported recently in an underdoped La-based cuprate[3]. We propose to use the changing symmetry of the temperature dependence above one third doping in charge-stripe ordered La2-xSrxNiO4 to further illuminate the ordering process of this charge-stripe ordered material.

We undertook an investigation of the response of the magnetic order and magnetic excitations to the changing periodicity of charge-stripe order (CSO) with temperature in $La_{1.67}Sr_{0.37}NiO_4$. In $La_{2-x}Sr_xNiO_4$ holes doped into the material by substituting La by Sr, go into the Ni oxide layer and form lines of charges at 45 degrees to the Ni-O bonds, charge-stripes, when sufficient holes are doped into the material. The remain Ni sites order antiferromagnetically between the charge-stripes that act as anti-phase domain walls, producing magnetic peaks centred at $((h\pm\varepsilon)/2$, $((k\pm\varepsilon)/2$, l) where ε is known as the incommensurability. The periodicity of the charge order is $1/\varepsilon$. It has been found that there is an important variation of the expected doping dependence $\varepsilon = x$, with ε observed to be systematically shift towards $\varepsilon = 1/3$ for all doping levels with CSO[1]. Furthermore as the material is warmed towards the charge ordering temperature ε tends towards a value of 1/3[2]. This experiment was undertaken to determine if the magnetism evolves with this changing periodicity of the CSO above one third doping in the same fashion as below one third doping.

In figure 1 are scans through the magnetic Brag reflections and associated magnetic excitations of LSNO x = 0.37. We established in similar scans that by 60 K gapped magnetic excitations from the charge-stripe electrons have been thermally wiped out, consistent with earlier studies[3]. Unlike in CSO $La_2NiO_{4.11}$ we observe in figure 1 the magnetic excitations to be centred on the magnetic Bragg peaks[4]. We are currently discussing the implications of this null result for the cause of the offset of the magnetic excitations in $La_2NiO_{4.11}$, possibly LSNO x = 0.275[5] and in an oxygen doped La cuprate[6].

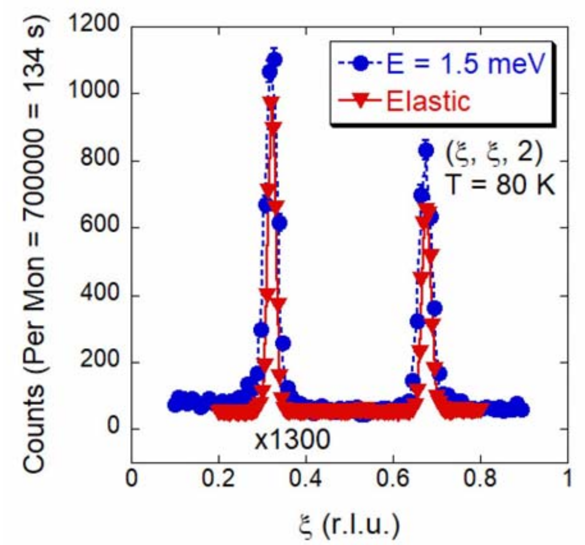

Figure 1: Scans parallel to the (hh0) direction of the magnetic Bragg peaks and magnetic excitations of CSO LSNO x = 0.37

We established the temperature dependence of ε for the order of the x = 0.37. In figure 2 we plot the temperature evolution of the modulus of the shift of ε for x = 0.275 where ε increases with increasing temperature, and for x = 0.37 where ε decreases with increasing temperature. The transition from low temperatures where ε is approximately constant with increasing temperature, to high temperature where ε changes with temperature, is abrupt for x = 0.275 but gradual for x = 0.37. For x = 0.37 a corner temperature for the crossover was determined to be 160 ± 5 K from this data.

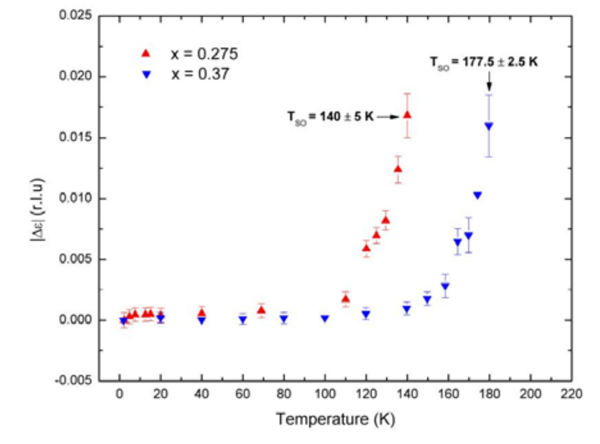

Figure 2: Temperature evolution of the change in ε determined from the magnetic Bragg peaks in LSNO x = 0.275 and 1/3.

In LSNO x = 1/3 when the magnetic order occurs the out of Ni-O plane charge-stripe correlation length reduces[7], an inverse order by disorder transition due to the competing charge and spin interactions. A reduction of the out of plane magnetic order correlation length in the x = 0.275 occurs below 110K most prominently for l = even magnetic Bragg reflections[8], possibly indicating that the reduction in out-of-plane correlation length occurs when ε transitions to its base temperature value. In x = 0.37 we however observe a slightly different effect.

In figure 3 we show the temperature dependence of the out-of-plane peak width for x = 0.37, only the l = even peaks show any sign of broadening on cooling below 150 K. There is a 10 K temperature lack between the transition into the constant ε regime and the broadening of magnetic Bragg peaks in x = 0.37. Additionally in figure 4 we see that in scans parallel to (hh0) both the l = even and odd peaks broaden on cooling in the in-plane direction. In the x = 0.275 an in-plane broadening was thought of as an artefact of the larger out-plane broadening, but our results of x = 0.37 show there is an in plane effect. Furthermore we observed a temperature history effect of the broadening of magnetic Bragg reflections in scans took to improve statistics of figure 3.

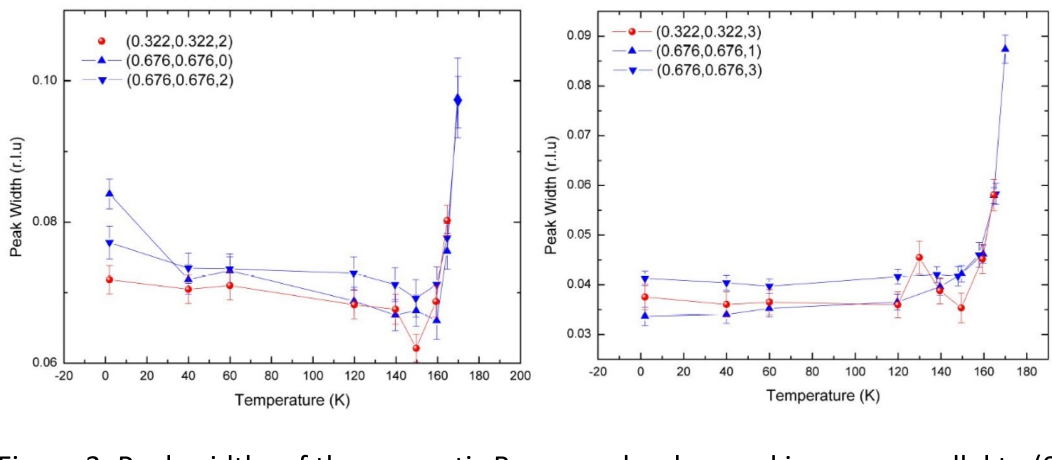

Figure 3: Peak widths of the magnetic Bragg peaks observed in scans parallel to (00l), left for l = even, right for l = odd. For l = even a small broadening of the magnetic peaks is observed below 150 K, but not for l = off peaks.

The magnetic excitations were also studied at 120 K and just above the magnetic ordering temperature, 177.5±2.5K, at 180 K. The significantly reduced reactor power during this experiment, limited the scope of these measurements during this experiment, but clear results were obtained. Consistent with unpublished data on $La_2NiO_{4.11}$, we observe only the lowest energy magnetic excitations respond to the changing periodicity of ε.

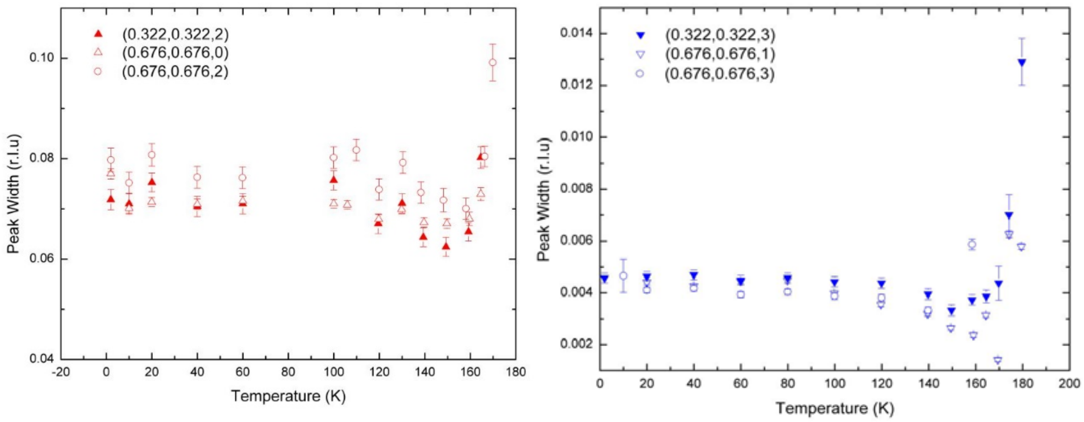

Figure 4: Peak widths of the magnetic Bragg peaks observed in scans parallel to (hh0), left for l = even, right for l = odd. For l = even a small broadening of the magnetic peaks is observed below 150 K, but not for l = off peaks.

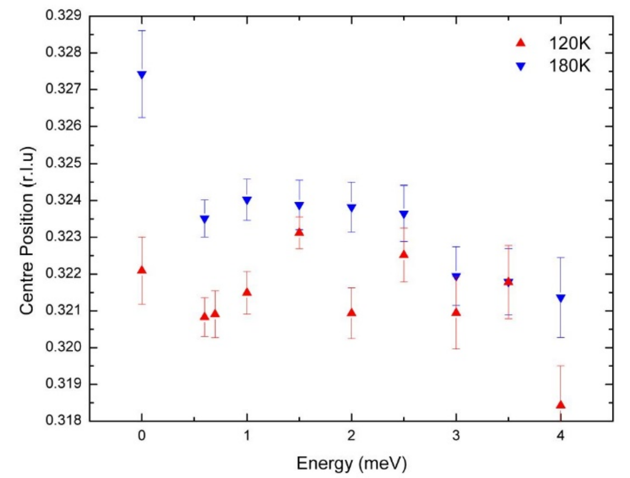

Figure 5: The energy evolution of the h centre of magnetic excitations in LSNO x = 0.37 measured at two temperatures at (hh2). By 180 K the CSO periodicity has increased significantly, and remnant critical scattering is observed to be centred near positions equivalent to (0.327, 0.327, 2). The magnetic excitations above ~3 meV do not respond to the changing periodicity of ε.

This study of the evolution of the magnetism in response to the changing periodicity of the CSO of LSNO x = 0.37 has subtle, yet distinct differences to those observed for CSO doping level of LSNO below one third doping. We are currently considering the significance of these differences, alongside the published findings of others, and the relevance of these findings for furthering our understanding of charge order in the hole doped cuprates.

References:

[1] H. Yoshizawa, et. al., Phys. Rev. B61, R854 (2000)
[2] K. Ishizaka et. al., Phys. Rev. Lett.92, 196404 (2004)
[3] P. G. Freeman, et. al., J. Phys.: Condens. Matt.20, 104229(2008).
[4] P. G. Freeman, S. R. Giblin, et. al., accepted for publication.
[5] H. Woo, et al., Phys. Rev. B72, 064437(2005)
[6] H. Jacobsen, et. al., Phys. Rev. Lett. 120, 037003 (2018)
[7] Shu-Han Le, et. al., Phys. Rev. B 92, 205114 (2015).
[8] P. G. Freeman, et. al., J. Phys.: conf. ser.340, 012060 (2012)