Experimental report

19/08/2022

Proposal: **DIR-253** **Council:** 4/2021

Title: Role of Discommensurations in the Magnetism of Charge-Stripe Ordered La2-xSrxNiO4

Research area: Physics

This proposal is a new proposal

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Samples: La1.63Sr0.37NIO4

Instrument	Requested days	Allocated days	From	To
PANTHER	7	7	15/09/2021	21/09/2021

Abstract:

In hole doped layered metal oxides there is a tendency for the charges to line up into semi-regularly spaced lines, charge-stripes. For charge-stripes that are pinned to the metal and/or oxygen sites the periodicity of charge-stripes needs to vary around the average periodicity for all but a few special commensurate doping levels. The variations in periodicity are called discommensurations. The role of charge-stripe discommensurations is now disputed in the charge-stripe ordered nickelates, along with a dispute on the possibility of significant inter-layer spin coupling. A resolution of the role of discommensurations in the magnetism of charge ordered phases of the nickelates is of great importance to aid our understanding of the role of charge order in high temperature cuprate superconductivity. This proposal will resolve the role of discommensurations in the magnetism of charge-stripe ordered La2-xSrxNiO4.

Role of Discommensurations in the Magnetism of Charge-Stripe Ordered $La_{2-x}Sr_xNiO_4$

Incommensurate charge ordering and associated spin correlations now appear ubiquitous to cuprate superconductors, yet there are many aspects of charge order that we do not understand, even in the perceived model charge-stripe ordered (CSO) systems such as non-superconducting $La_{2-x}Sr_xNiO_4$ (LSNO)[1-3]. It is common to describe charge-stripe order in LSNO as commensurate if the average charge-stripe spacing is a whole number of lattice spacings, and incommensurate if not. For incommensurate charge-stripe order in LSNO and the cuprates, we do not fully understand the effect on the magnetic interactions caused by the variation of charge-stripe spacing, discommensurations. Competing proposals on the effect of discommensurations exist for modelling LSNO and other nickelates [4-6], the results of this experiment will help resolve the role of discommensurations in LSNO, providing insights for our understanding of the cuprates.

In hole doped layered metal oxides there is a tendency for the charges to line up into semi-regularly spaced lines, charge-stripes. For charge-stripes that are pinned to the metal and/or oxygen sites the periodicity of charge-stripes needs to vary around the average periodicity for all but a few special commensurate doping levels, i.e. x = 1/3 in LSNO[7]. The variations in periodicity are called discommensurations. Charge-stripe order in LSNO produces magnetic Bragg peaks $((h \pm \varepsilon)/2,\ (k \pm \varepsilon)/2\ , l)$, where ε is known as the incommensurability that varies with the hole doping level $\varepsilon \sim n_h$, and the average periodicity is $1/\varepsilon$. The role of charge-stripe discommensurations is now disputed in the charge-stripe ordered LSNO. Discommensurations where first proposed to only spatially dampen magnetic excitations [2], with discommensurately CSO LSNO's spin wave dispersion matched that of the nearest commensurately CSO structure so that LSNO x = 0.37 should have a 90 meV bandwidth[4]. Alternatively, the discommensurations have been viewed as admixtures to the spin structure and the magnon bandwidth obtained from the combination of the different charge-stripe spacings[5]. This leads to a proposed bandwidth softening from 90 meV for x = 1/3 linearly to 55 meV for x = 0.5[5], so LSNO LSNO x = 0.37 should have an 80 meV bandwidth. Furthermore an ordered discommensuration spin stripe model has been proposed to model the magnetic excitations of $Pr_{3/2}\,Sr_{1/2}NiO_4$ (PSNO) with a significant out of plane spin interaction[6] that would produce dispersive excitations along (00l), unlike l independent models of spin excitations in LSNO [2,4,5]. By measuring the (h,k,l,ω) dependence of the complete magnon dispersion of LSNO x = 0.37 this experiment will resolve all of these proposals.

Using a high incident energy of $E_i = 112\ meV$ the full (h,k,l,ω) spin wave dispersion of LSNO x = 0.37 at a base temperature of 1.5 K over many zone centres. We have performed an initial analysis of this data. From this initial analysis it is clear that the magnetic excitations have no l-dependence, signifying that the out-of-NiO-plane spin interaction is insignificant in comparison to the intra-plane spin interaction in contrast to $Pr_{3/2}$ $Sr_{1/2}NiO_4$[6]. Determining the bandwidth of the magnetic excitations requires further analysis, in the energy range 80-90 meV excitations are observed in the data but the phonon bandwidth of LSNO is known to extend to 90 meV. Determination of the bandwidth of the magnetic excitations will therefore require studying the excitation spectrum across the many zone centres measured in the Panther data.

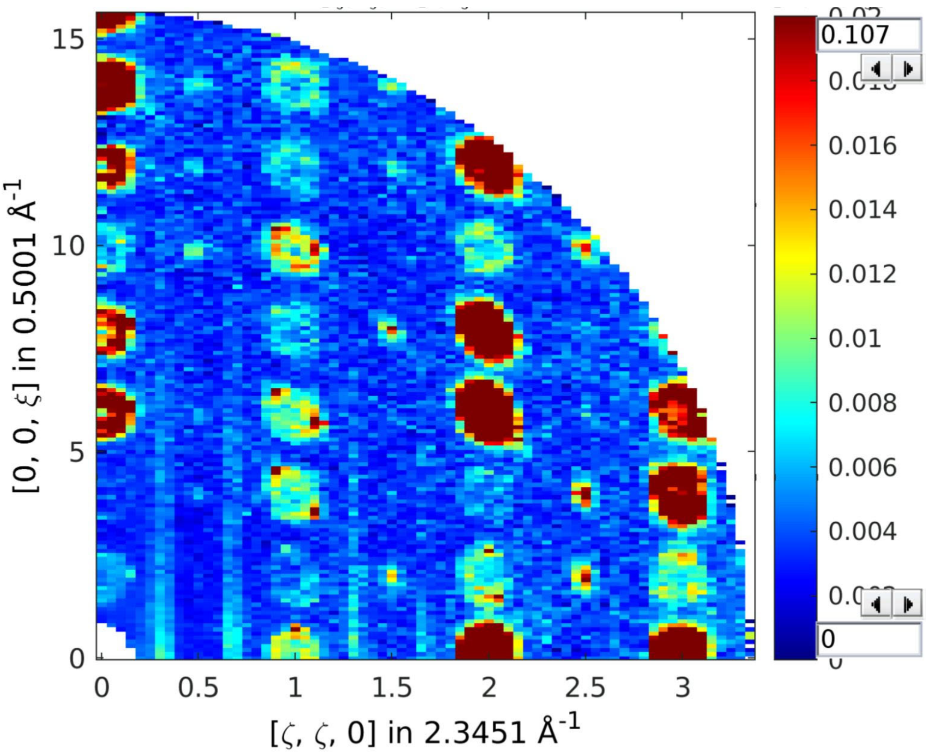

Figure 1; E = 6 -7 meV slice of the excitation spectrum of LSNO x = 0.37 taken with $E_i = 40\ meV$ at 2 K, showing the extensive Q-range accessible on Panther. Vertical streaks of magnetic excitations from the spin stripe order showing their lack of l dependence.

Additional data was taken at lower incident energies of 76.45 meV and 40 meV. The 76.45 meV will provides a higher resolution mapping of the spin wave dispersion in comparison to the $E_i = 112\ meV$ data, providing the most robust test of the standard spin wave modelling of the spin wave dispersion of the spin stripe order of LSNO[2,3]. Initial review of these data sets revealed the presence of a mode that to the best of our knowledge has not previously been observed, see figure 2. This mode's centring and dispersion suggests an antiferromagnetic origin, yet the mode strikingly has significant intensity at large integer values of l, and appears to have l dependence in contrast the known spin wave dispersion. This mode requires detailed analysis of across all zone centres, and potentially further study.

Recent studies of superconducting cuprates $La_{2-x}Sr_xCuO_4$ and $La_{2-x}Ba_xCuO_4$ have identified spin-phonon hybridisation of the excitation spectrum in the 16-19 meV energy range [7]. LSNO being pseudo-issostructural with these two cuprate materials, naively therefore should be expected to show a similar effect. In this energy range the spectral weight of magnetic excitations in LSNO is supressed [8], and transferred to higher energies [2,3,8,9]. Phonons in this energy range vibrate the in plane O atom positions, so are likely to change the bond angles that effect the superexchange interaction[8]. The sensitivity of the antiferromagnetism to bond angle means that the spin-phonon interaction that leads to hybridisation, could result in anti-hybridisation in LSNO[2,3,8,9]. Initial analysis of the data of this experiment indicate the potential of the data to extract this, although careful analysis is required as we do not have a magnetic blank to use to subtract the phonon background, unlike in the cuprates[7].

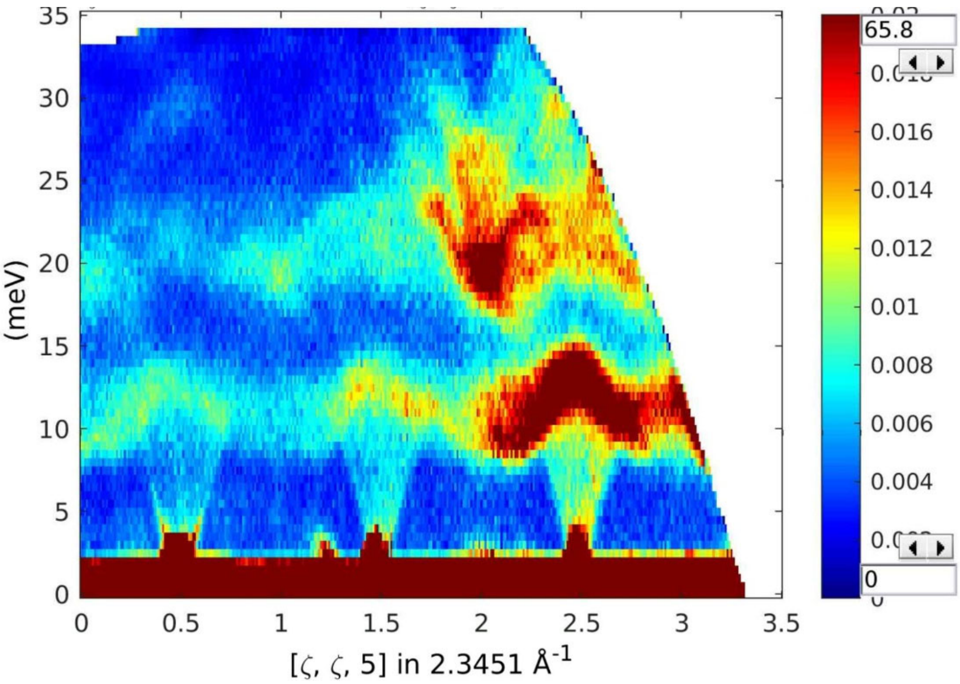

Figure 2: A slice of the excitation spectrum of LSNO x = 0.37 taken with Ei = 40 meV at T = 2 K. Additional new modes can be seen dispersing from positions such as (0.5, 0.5, 5), (1.5,1.5,5), etc., as well as at other l = odd integer positions.

The final aspect that this experiment will address is the nature of the magnetic excitations in the paramagnetic phase. In commensurate charge-stripe ordered PSNO the intensity of the magnetic excitations rapidly drops to zero in the paramagnetic phase[10], in apparent contrast to $La_2NiO_{4.11}$[11]. If magnetism plays a role in the formation of CSO paramagnons should be observed in the disordered phase, if they are not then the charge ordering drives the CSO in LSNO. Alternatively there may be different driving forces for commensurate and incommensurate CSO in LSNO, our study of the magnetic excitations at six temperatures 1.5-300 K along with further studies will help to resolve this question regarding CSO. Answering this questions is important to establish not just for LSNO, but to gain CSO understanding for the cuprate materials

[1] Yoshizawa et. al., Phys. Rev. B **61**, R854 (2000)
[2] H. Woo, et. al., Phys. Rev. B 72, 064437 (2005).
[3] A. T. Boothroyd, Phys. Rev. B **67**, 100407(R) (2003).
[4] P. G. Freeman, et. al., J. Korean Phys. Soc. **62,** 1453 (2013).
[5] G. Fabbris, et. al., Phys. Rev. Lett. **118**, 156402 (2017).
[6] A. Maity, et. al. Phys. Rev. Lett. **124,** 147202 (2020); Phys. Rev. B **103**, L100401 (2021).
[7] J. J. Wagman, et. al., Phys. Rev. B **91**, 224404 (2015), J. J. Wagman, et. al., Phys. Rev. B **91**, 224404 (2016), K. Ikeuchi, et. al., Phys. Rev. B **105**, 014508 (2022).
[8] P. G. Freeman, et. al., J. Phys.: Condens. Matt. 20 (10), 104229 (2008).
[9] A. T. Boothroyd, et. al., Physica B **345**, 1 (2004).
[10] A.Maity, et. al., Phys. Rev. B **106**, 024414 (2022).
[11] P. G. Freeman, et. al., Sci. Rep. **9**, 1 (2019).