

Experimental report

25/01/2024

Proposal: 4-01-1768

Council: 10/2022

Title: Spin and charge fluctuations in O-doped nickelates with hole concentration close to 0.5

Research area: Physics

This proposal is a new proposal

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Samples: Nd₂NiO_{4.23}

| Instrument | Requested days | Allocated days | From | To |
|---------------|----------------|----------------|------------|------------|
| ORIENTEXPRESS | 1 | 1 | 31/05/2023 | 01/06/2023 |
| IN8 | 6 | 11 | 12/06/2023 | 19/06/2023 |
| | | | 10/09/2023 | 13/09/2023 |
| | | | 08/12/2023 | 10/12/2023 |

Abstract:

The confirmation of stripes order in cuprates materials inspired speculations about the role fluctuating stripes in the superconducting pairing mechanism. Isostructural hole-doped nickelates, are a strongly correlated electron system that also exhibits charge order and in which both the spin and charge fluctuations could be detected. It has been shown recently the coexistence of oxygen and magnetic ordering below T_n in a Nd₂NiO_{4.1} with magnetic satellite reflections that adopt exactly the same modulation vectors as found for the oxygen ordering. Proving evidence of such coexistence for other oxygen doping might support the picture of pinning of magnetic order to oxygen order. In this, for the $n_h=1/2$ hole doping, the spin order is a 2D checkerboard pattern where all the nearest-neighbour sites of the spins are occupied by holes. It would then be interesting collecting the spin and charge order fluctuations in Nd₂NiO_{4.23} close to such ordering. In this way, we can unveil the behaviour of spin and charge fluctuation in the heavily over-doped region, much less explored, and correlate result with oxygen ordering and with the annexed lattice instabilities found in this composition.

4-01-1768. Spin and charge fluctuations in O-doped nickelates with hole concentration close to 0.5

Introduction. The experimental confirmation of stripes order in some cuprates materials inspired speculations about the role fluctuating stripes could have in the superconducting pairing mechanism [1]. This question remains matter of controversy. In the cuprates, static stripe order competes with superconductivity, while, fluctuating stripe order can coexist with stable high-temperature superconductivity. Investigate stripes phases in related compounds might reveal essential to unveil the possible role of the stripe instability in the physics of high-temperature superconductivity [2].

Hole-doped nickelates, isostructural to cuprates, are a strongly correlated electron system that also exhibits charge order. On doping layered transition-metal-oxide antiferromagnets with holes, it is possible for a stripe phase to develop along the diagonal direction with respect to the Ni–O bonds, with the holes segregating into charge stripes that form antiphase domain walls between antiferromagnetic domains. This hole doping can be made with introduction of cations like Sr (quenched disorder), by oxygen over-stoichiometry (annealed disorder) or both [3].

The interest in focusing on oxygen doped samples reside in the fact that it has been shown recently the coexistence of oxygen and magnetic ordering below $T_N = 48$ K in $Nd_2NiO_{4.1}$ with magnetic satellite reflections that adapt exactly the same modulation vectors as found for the oxygen ordering [4]. This point out a unique coexistence of 3D modulated ordering for spin and oxygen ordering. Proving such coexistence for other doping might support the picture of pinning of magnetic order to oxygen order in oxygen-doped samples. This would also imply that the quasi-2D nature of the spin correlations might not be the optimal model for oxygen-doped samples.

For a small hole concentration $n_h < 1/3$, the distance between the hole stripes is wide enough to accommodate three or more Ni chains in between, and this situation allows for Ni chains to form antiphase antiferromagnetic spin ordering across the hole stripes. In contrast, for the samples with larger hole doping n_h of $1/3 < n_h < 1/2$, the distance of the hole stripes is small, and only one or two Ni chains are accommodated between the hole stripes. Nevertheless well-developed stripe order is established within the NiO_2 planes up to $n_h = 0.5$ [5]. What is more interesting is that for the $n_h = 1/2$ ordering pattern, the spin order is actually a 2D checkerboard pattern and all the nearest-neighbour sites of the spins are occupied by holes. If this is accomplished and phase separation is not at play, in-plane AF interactions should result weakened. On the other side, the presence of a tighter 3D network due to interstitials, may might bring to increased out-of-plane magnetic interactions.

The behaviour of such fluctuations in the heavily over-doped region, which is much less explored, must be then explored in detail and correlated with oxygen ordering and the annexed lattice instabilities so far found in such composition [6].

Measurements. $Nd_2NiO_{4.23}$ sample was measured on IN8 with the goal to point out *ab*-plane spin and charge dynamics (exp, 2023). We have measured excitation in the $Nd_2NiO_{4.23}$ sample close to the $n_h = 1/2$ aligned in $(hk0)$ scattering plane. The experiment allow collecting the dispersion of the spin fluctuations in this highly doped nickelate close to the checkerboard spin and charge order. Measurement were done in the spin order region at 1.5K and their persistence was analysed above T_n at 150K. Reference measurements were collected at 300K to evaluate contribution from magnetism and lattice dynamics. Furthermore, we looked for the charge order fluctuation with selected scans at higher Q .

The dispersion of the spin wave on $(hk0)$ plane dispersing from incommensurate magnetic peaks could be followed up to 60 meV along $(0k0)$ direction across 300 point (Figure 1, left panel). At 14meV the fit of the spin wave give an ϵ value of 0.38 r.l.u. corresponding to an overstoichiometry of $\delta = 0.23$. This spin wave does not disperse until 30 meV where an inward dispersion could be detected (black arrow), before dispersing out until 46 meV. A further dispersing signal could be detected that could be either phononic or magnetic, but originating from AF commensurate phase ($\delta = 0$). The top of the spin wave

dispersion could be estimated around 58 meV (see Figure 1b). At this energy a sharp signal at BZ boundaries is detected.

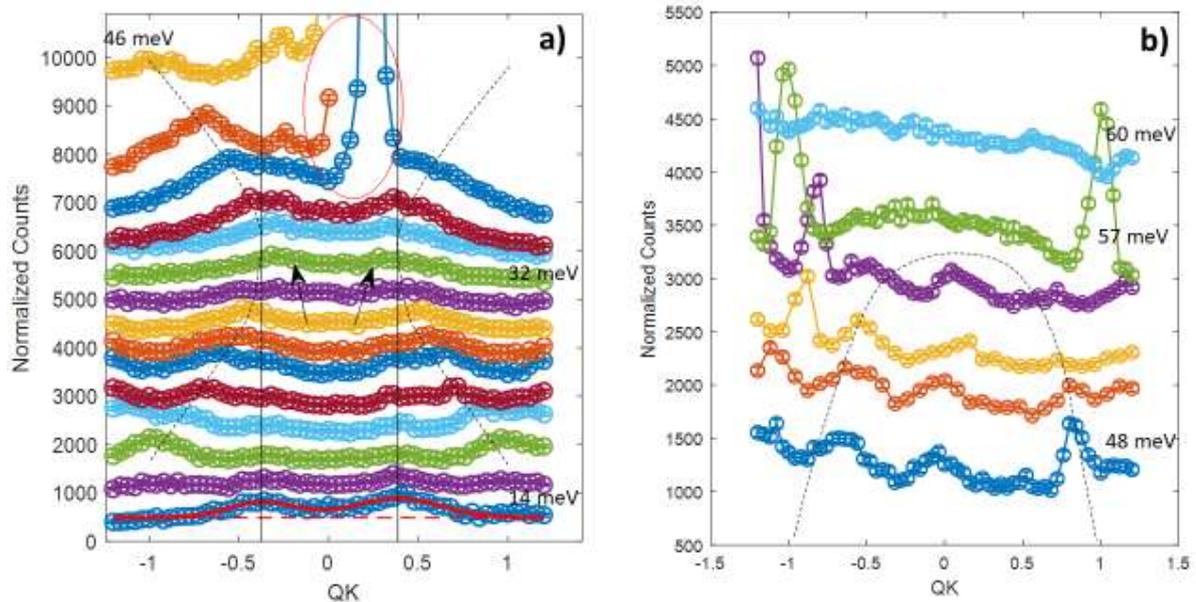


Figure 1. a): lower part of the spin wave dispersion relation ($0k0$) direction across 300 point. Scans were vertically sifted for clarity. At 14meV the fit of the spin wave give an incommensurability ε value of 0.38 r.l.u. well corresponding to an overstoichiometry of $\delta=0.23$. This spin wave does not disperse until 30 meV where an inward dispersion could be detected (black arrow), before dispersing out until 46 meV. A spurion is highlighted in red ellipse. B) upper part of spin wave dispersion reaching about 57 meV. At this energy a sharp signal at BZ boundaries is detected.

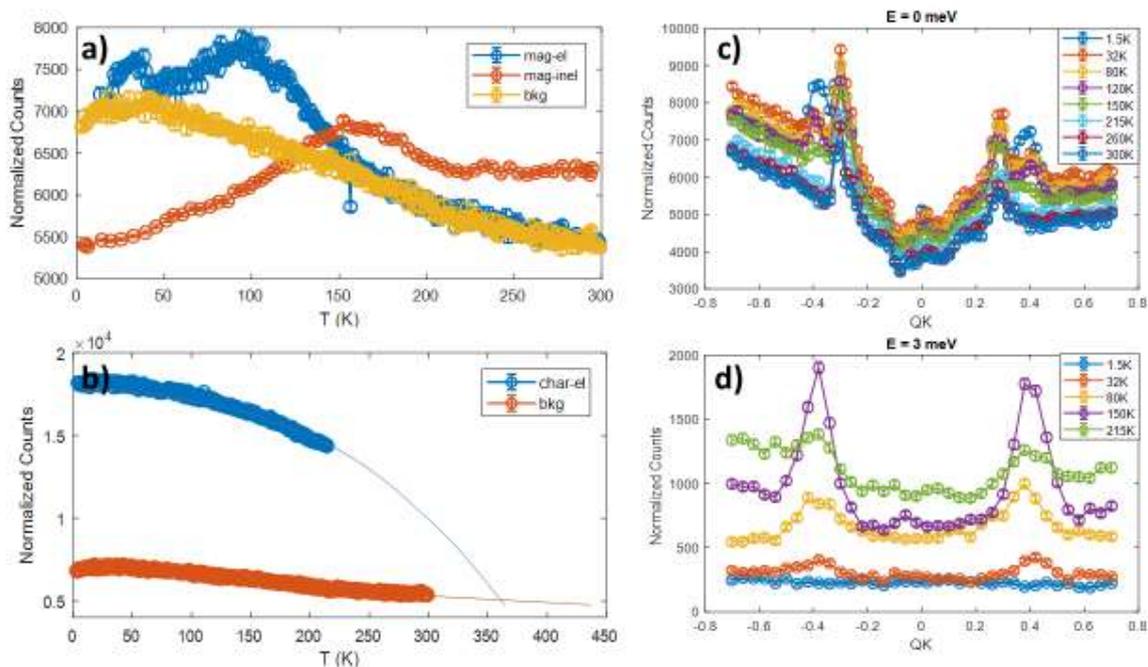


Figure 2. a): elastic and inelastic signal from magnetic order peak and its fluctuation at 3 meV, respectively. Magnetic order disappear around 150K, while magnetic fluctuations are gapped at lowest T, gap is filled at 150K and persist above 150K. b): intensity of a charge order peak with respect to background. Charge order persist well above 300K. Elastic c) and inelastic d) Q scans along ($0k0$) direction trough 100 point show magnetic order suppressed above 150K, while fluctuation result gapped at low temperature, reach their maximum intensity at 150K and persisting at 215K.

At the same time elastic and inelastic signal from a magnetic order peak (1 -0.38 0) and its fluctuation at 3 meV, have been followed during temperature ramps (Figure 2a). It shows that magnetic order disappear slightly above 150K (see also constant E scans in Figure 2c), while magnetic fluctuations are gapped at lowest T, then the gap is gradually filled up to 150K and finally decrease above 150K, yet persisting at least up to 220K (see also Figure 2d). Figure 1b, instead, shows the intensity of a charge order peak. Compared to background intensity, it looks that charge order persist well above 300K. At this point of the analysis we cannot yet be sure of the presence of signatures of charge fluctuations, but several features found in scan might be connected with this.

References. [1] S.A. Kivelson, I.P. Bindloss, E. Fradkin, V. Oganesyan, J.M. Tranquada, A. Kapitulnik, C. Howald, *Rev. Mod. Phys.* 75 (2003) 1201; [2] H. Ulbrich, M. Braden, *Physica C* 481 (2012) 31; [3] J. M. Tranquada, P. Wochner, D. J. Buttrey *Phys. Rev. Lett.* 79 (1997) 2133; [4] S. R. Maity, M. Ceretti, L. Keller, Jürg Schefer, Martin Meven, E. Pomjakushina, W. Paulus, *Phys. Rev. Mater.* 5 (2021) 014401; [5] H. Yoshizawa, T. Kakeshita, R. Kajimoto, T. Tanabe, T. Katsufuji,[†] and Y. Tokura, *Phys. Rev. B* 61 (2000) 854; [6] A. Perrichon, A. Piovano, M. Boehm, M. H. Lemée, M. Ceretti, and W. Paulus *Physical Review B* 107, 144303 (2023)