Experimental Report

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Title: Sr-doping dependence of possible “hour-glass” magnetic excitations inLSNO

This proposal is a new proposal

Researh Area: Physics

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Samples: La2-xSrxNiO4

Instrument	Req. Days	All. Days	From	To
IN22	6	9	29/04/2013	07/05/2013
			18/07/2013	19/07/2013
IN8 Flatcone	5	5	04/04/2013	09/04/2013
IN3	0	5	03/04/2013	05/04/2013
			29/04/2013	30/04/2013

Abstract:

The recent observation of a “hour-glass” magnetic excitation spectrum in La2-xSrxCoO4 resembling on the famous observations in the isostructural HTSC cuprates has re-attracted enormous interest to the physics of charge stripe phases. For the isostructural nickelates no “hour-glass” dispersion was observed so far and even thought to be systematically absent due to a larger inter-stripe exchange coupling J’. However, we were able to observe the basic features of a “hour-glass” excitation spectrum in 14% hole-doped LSNO sample. Now, we would like to extend our studies to slightly higher Sr-doping (i) in order to separate the magnetic signals a little more from the central region for obtaining clearly separated magnon branches and (ii) in order to suppress the octahedral tilting reflection that appears between the magnetic signals (but vanishes for higher Sr-doping). Besides studying the most promising sample in great detail (by mapping the magnetic signals etc.) we would like to study also the Sr-doping dependence and the “nascency” and the vanishing of the “hour-glass” dispersion systematically as a function of Sr-doping by comparing at least three different LSNO samples.

Sr-doping dependence of possible "hour-glass" magnetic excitations in LSNO

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Charge stripes have been predicted as a combined charge and spin-density wave phenomenon [1-3] before being experimentally observed in $La_{2-x}Sr_xNiO_4$ (LSNO) [4] and in the Nd codoped $La_{2-x}Sr_xCuO_4$ [5]. However, the role of this charge stripe instability for the superconducting pairing mechanism still remains a matter of debate and it is still puzzling that both phenomena can coexist though charge stripes tend to supress superconductivity [6]. Remarkable attention has been re-attracted to the physics of stripe phases due to the recent discovery of a "hour-glass"-shaped magnetic spectrum in $La_{2-x}Sr_xCoO_4$ [7] resembling on the one in the superconducting cuprates [8]. Since this "hour-glass"-shape of magnetic correlations seems to be a unifying property of the cuprates, dynamic charge stripes might be essential for the understanding of these materials. However, this interpretation is still a matter of debate [9]. Apart from the cobaltates [7,9] also the nickelates could be a useful reference system since a rather stable diagonal charge stripe order has been observed in these systems [4]. However, so far mainly the higher Sr-doped or the oxygen-doped compounds have been studied and no "hour-glass"-dispersion has been found in the charge stripe ordered phases. But most studied systems with lower hole-doping suffer from being quite commensurate (the incommensurability is rather close to 1/3; compare **Fig. 1**). Especially the comparably high exchange coupling *J'* across the stripes and the long-range charge stripe correlations seems to be an obstacle for the observation of the so-called "hour-glass" dispersion in these materials [7,10]. Therefore, we were interested in studying charge stripes and magnetic excitations in low-doped LSNO with incommensurabilities $\varepsilon << 0.22$. Note that the commensurate order appears to be still the dominant ordering scheme in LSNO with x=0.275 [11] and that a high energy range of ~80 meV is needed for neutron spectroscopy of these samples (e.g. for x~0.31 [12]). So far, the study of lower Sr-doped samples might have been hampered by the difficulty of the growth of high-quality single crystals with correct oxygen content. Most studies of LSNO samples are done on samples with ε-values distinctly above 0.2 being still rather close to the commensurate value of 1/3 and having rather robust, long range ordered charge stripe phases. $La_{1.8}Sr_{0.2}NiO_{4-\delta}$ is one of the few samples where clearly incommensurate magnetic signals with small ε-value and smaller correlation lengths have been observed [13].
In contrast to that, we managed to grow also low Sr-doped LSNO materials without excess of oxygen and started studying first LSNO samples at the IN22 (and at the IN8, IN3) spectrometer in the past, see experimental reports in Refs. [14,15].
Whereas we focused on these extremely low hole-doped regime (*n*~12%-16%) in the past [14,15], we ***now*** tried to measure the Sr-doping dependence systematically and to measure also *slightly* higher hole-doped nickelate samples without exceeding the hole concentration *n* too much towards the commensurate regime around ~20%-25% of hole-doping. In this experiment we measured several samples and, then, focused on the most promising one (*ACK390*). This sample was studied with polarized neutrons at the IN22 spectrometer and with unpolarised neutrons at the IN8 spectrometer using the flatcone option. (Additionally, characterization experiments have been performed at the IN3 spectrometer.)

The measurement at the **IN8 spectrometer** with **flatcone option** reveals a nice overview of magnetic excitations of this nickelate which is shown in **Fig. 1** for various energy transfers. Apparently, the magnetic signal starting from the incommensurate magnetic peak positions exhibits a slight inwards-dispersion with increasing energy. Then, at higher energies around 20 meV a kind of resonance-like merging appears "quite suddenly". Finally, the outwards dispersing signal is observeable at higher energy tranfers (>> 20 meV). However, there is also sizeable signal from optical phonon modes especially in the interesting energy regime around 20 meV.

The complementary neutron scattering experiment at the **IN22 spectrometer** with **polarization analysis** yields important information about this 20 meV regime since magnetic and purely nuclear/structural scattering can be distinguished. The flipping ratio obtained in this polarized neutron experiment amounts to ~16 and the magnetic signal was always measured in the spin-flip channel ||x (with x||Q) . A setup with Helmholtz coils was implemented. As can be seen in **Fig. 2 a**, the energy scan at the planar antiferromagnetic wavevector not only reveals the optical phonon signal (around ~20 meV) but also strongly increased magnetic intensity around 20 meV. This corroborates that there is a resonance-like merging of magnetic intensity around 20 meV as was already indicated by the IN8 measurement. (The increased scattering in the spin-flip channel can not by explained by nuclear scattering arising from an imperfect polarization analysis since the flipping ratio amounts to ~16 and that increase of intensity is simply too massive for such an explanation. Nonetheless, it appears interesting that the nuclear scattering accidentally exhibits a very similar energy dependence at least below 35 meV (signal in the non-spin-flip channel is also shown in **Fig. 2 a**). Furthermore, the resonance-like increased magnetic intensity does not disappear steep again, but much slower and extends to much higher energies.
Complementary constant-Energy scans across the incommensurate peak positions (and across the planar antiferromagnetic wavevector) measured in the spin-flip channel ||x show that there appears “suddenly” additional magnetic intensity at 20 meV at the “magnon merging point” between both magnon branches, see **Fig. 2 b** (green data points). Around 30 meV this resonance-feature is already somewhat weakened and, thus, the outwards dispersion of the magnetic branches becomes visible, see **Fig. 2 b** (red data points). In contrast to most cuprates and cobaltates [9] the shape of this dispersion looks more like a “H”-shaped than like an “X”-shaped hour-glass dispersion, i.e. the branches are quite steep and “suddenly” there appears magnetic intensity around 20 meV that is located between these branches and that only vanishes very slowly with increasing energy. This resembles in some sense a little bit on the observations that have been made above T_C in certain YBCO samples in the past [18].

Our novel observations can be regarded as a major advance in the search for hour-glass-shaped magnetic excitations in the nickelates that were thought to be fully absent in all nickelates before [7].
Note, that our novel observations appear to be very different from the quite commensurate dispersion that we observed in slightly higher Sr-doped nickelate samples in the past – compare **Fig. 2 b** with “Figure 2 in experimental report of experiment N°-Ex : 4-01-763” [17]: there is clearly an outwards dispersing signal at 30 meV in *our new* sample ACK390 above the “resonance” feature but nothing like that can be found in an about 21% hole-doped (x=0.175) nickelate sample [17].
Unfortunately, we could not measure to higher energy transfers at the IN22 spectrometer (especially in **Fig. 2a**) and the outwards-dispersing high-energy branches are not resolved in an ideal manner in both measurements so far (also due to lack of time in both experiments and also due to the additional appearance of optical phonon modes in the IN8 measurement). Furthermore, in the IN22 measurement the polarization analysis was only performed in the spin-flip/non-splin-flip channels parallel to the scattering vector Q (x-channel) due to the lack of time. Therefore, it seems highly desirable now to study also (i) the high-energy regime at an instrument with polarizated neutrons and with a higher energy range and to study also (ii) the magnetic signal in the other spin-flip channels for further information about the nature of these magnetic excitations.

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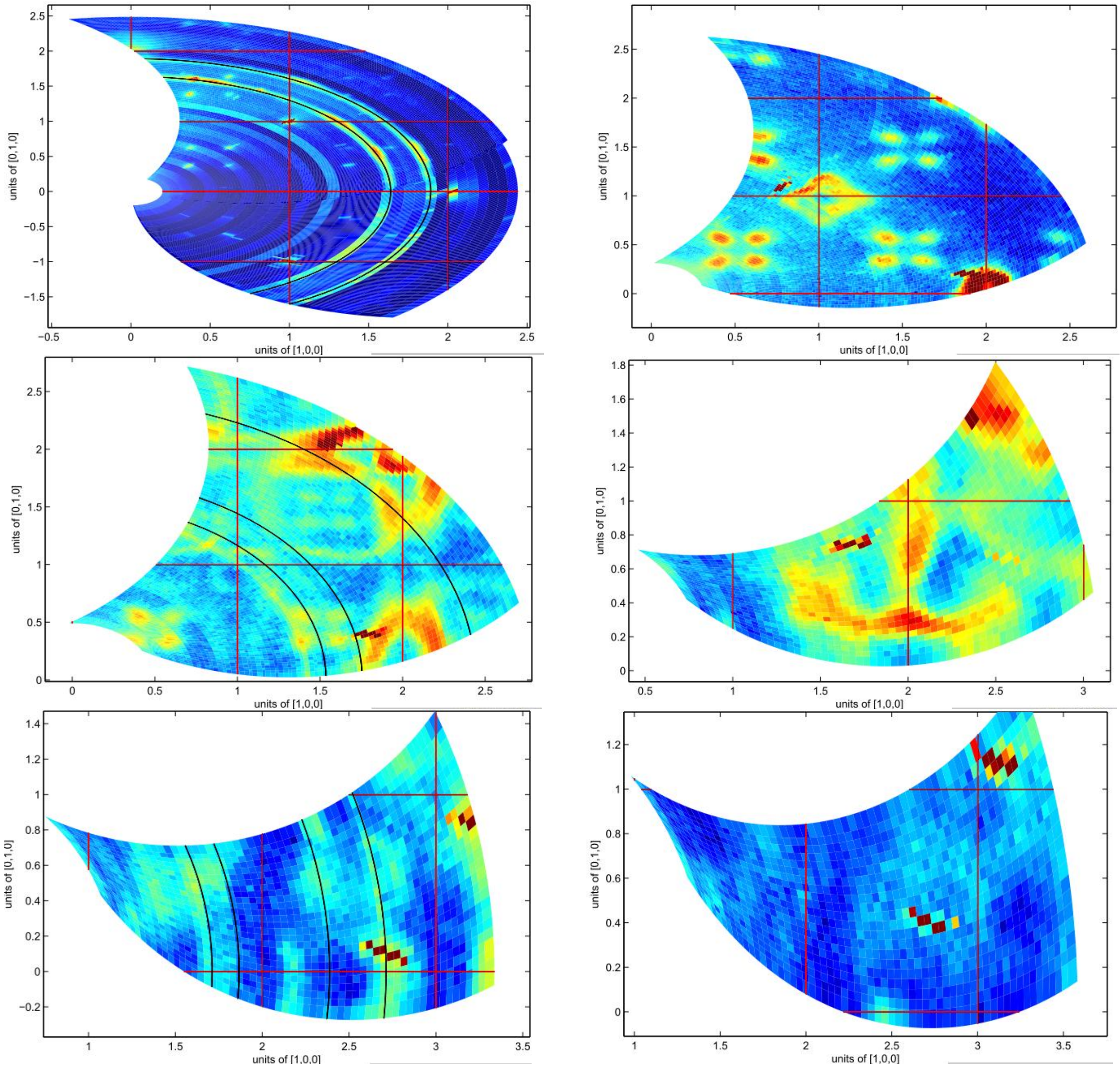

Figure 1: Neutron scattering intensities of *ACK390* measured at the **IN8 spectrometer with flatcone option** plotted as constant-energy maps within the reciprocal HK0 plane at energy transfers of 0 meV, 5 meV, 10 meV, 20 meV, 30 meV and 40 meV (from left to right first; then from top to bottom).

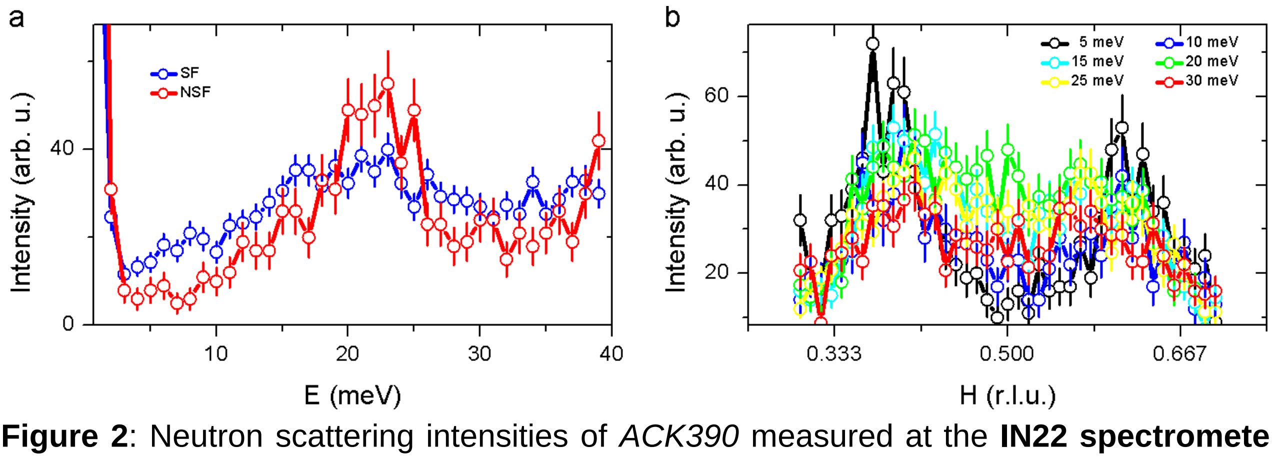

Figure 2: Neutron scattering intensities of *ACK390* measured at the **IN22 spectrometer** with polarization analysis. **a** Energy scans at (1/2 3/2 0) in spin-flip and non-spin-flip channels ||x. **b** Constant energy scans across (1/2 3/2 0) in the spin flip channel ||x.