Experimental report

07/02/2021

Proposal: **EASY-517**

Council: 4/2019

Title: Determination of an anomalous optical like mode in the magnetic excitation of Pr3/2Sr1/2NiO4

Research area: Physics

This proposal is a new proposal

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Samples: Pr3/2Sr3/2NiO4

Instrument	Requested days	Allocated days	From	To
IN8	48	120	12/09/2020	17/09/2020

Abstract:

Investigation of spin stripes and their excitations in hole-doped 214-nickelates remains an important research field for the last two decades. Like La-based nickelates, the undoped Pr2NiO4 exhibits a long-range 3D magnetic ordering. Hole doping via Sr or O in Pr2-xSrxNiO4+d leads to the segregation of the holes in the form of stripes in the NiO2 layers. The coulomb frustration, induced by the holes, leads to the phase separation, plays an important role in the stacking of the NiO2 planes along the out-of-plane direction manifesting a competing interaction between in-plane and out-of-plane spin stripes influencing the overall characteristics of the magnetic excitations. From our previous study at thermal TAS PUMA@ FRM II, we have determined the spin-wave dispersion. The measured in-plane excitation shows a symmetrical shift of the dispersion from the AFM zone center in the energy range 35-45 meV, presenting a signature of the presence of an anomalous mode, most probably coming from the out-of-plane interaction which have been mostly neglected in the nickelates. In this proposal, we want to determine unambiguously the presence of this anomalous optical like mode.

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During this proposal time, we have started to explore magnetic excitations in the stripe phases of an **O-doped** Pr-based 214-type nickelates $Pr_2NiO_{4+\delta}$. We aim a complementary study in comparison to magnetic excitation studies that we have performed on Sr-doped samples $Pr_{2-x}Sr_xNiO_4$ [1,2]. Hole doping by Sr or O is electronically equivalent, though it is important to note that Sr doping is done at the expense of incorporating disorder as immobile Sr randomly occupies the Pr-sites without getting ordered in the lattice. In comparison, excess O occupies the interstitial sites and remains mobile in the lattice even below room temperature, causing a long-range O ordering along with the localization of induced holes which form stripes at low temperature. In O-doped samples stripes are expected to be long-range, hence can give very interesting and complementary results compared to Sr-doped samples in which the unavoidable disorder introduced by the Sr results into a short-range stripe correlation. We have chosen the Pr-based nickelates as they are excellent candidates for accommodating higher amount of interstitial oxygen with long-range ordering in comparison to the La-based nickelates.

The primary outcome of the EASY-517 proposal is significant as it gives many promising and complementary results compared to the studies of Sr-doped samples [1,2]. To start with, we have obtained the spin stripe incommensurability of epsilon $\varepsilon \approx 0.346$, which is interesting because it is slightly away from the commensurate 1/3 stripe, and stripe-discommensuration by a mixture of 1/3 stripe and checker board will give rise to a spin stripe model as in the Fig. 1. We have noticed a sharp rise in the spin ordering temperature Tso ≈ 220 K (Fig 1) in the O-doped sample, much higher compared to the Sr-doped samples (Tso ≈ 125 K). Most importantly, in our O-doped sample, we have obtained the spin stripe correlation of $\xi \approx 47$ Å, which is significantly higher compared to the Sr-doped samples (≈ 20 Å). This gives us an enormous opportunity to directly access the effect of long-range spin stripe ordering on the magnetic excitations of 214-type nickelates. We have done our primary linear spin wave theory based calculations in the stripe discommensurated spin stripe unit cell, similar to our previous studies [1,2], to get a sense of the expected magnetic excitations (in Fig. 2). To our surprise, we have noticed multiple equidistant side peaks of the magnetic satellites in the elastic scans (in Fig. 3), exactly as it appear in the simulation at zero energy transfer. Such experimental observation of multiple side peaks is not reported so far in the neutron scattering study of 214-type nickelates. Since the linear spin wave based calculations assume ideally a long range spin correlation, the interstitial oxygen induced long range spin stripe correlation plays a decisive role in the good agreement between calculation and experimental results. We confirm the multiple side peaks from the temperature dependence measurements as in Fig. 1. We have found out the distance between the peaks related to the ratio of the number of 1/3-rd stripe and checkerboard units. Nonetheless, from our preliminary measurements we have been able to confirm the top band of the magnetic excitation from the high-energy (> 60 meV) const-Q and cont-E scans at IN8. Furthermore, we have obtained the interstitial oxygen ordering correlation length (50 Å), almost same as the above mentioned spin stripe correlation length (47 Å), giving an indication that the oxygen ordering may have a strong influence on localizing the induced holes almost in the same length scale. This gives an opportunity, just by carefully tuning the O-content vs. Sr-content of the sample, to control the spin stripe correlation and magnetic excitations. Further experiments must be performed on different oxygen doped samples and co-dopes (Sr, O) samples to come to such conclusion.

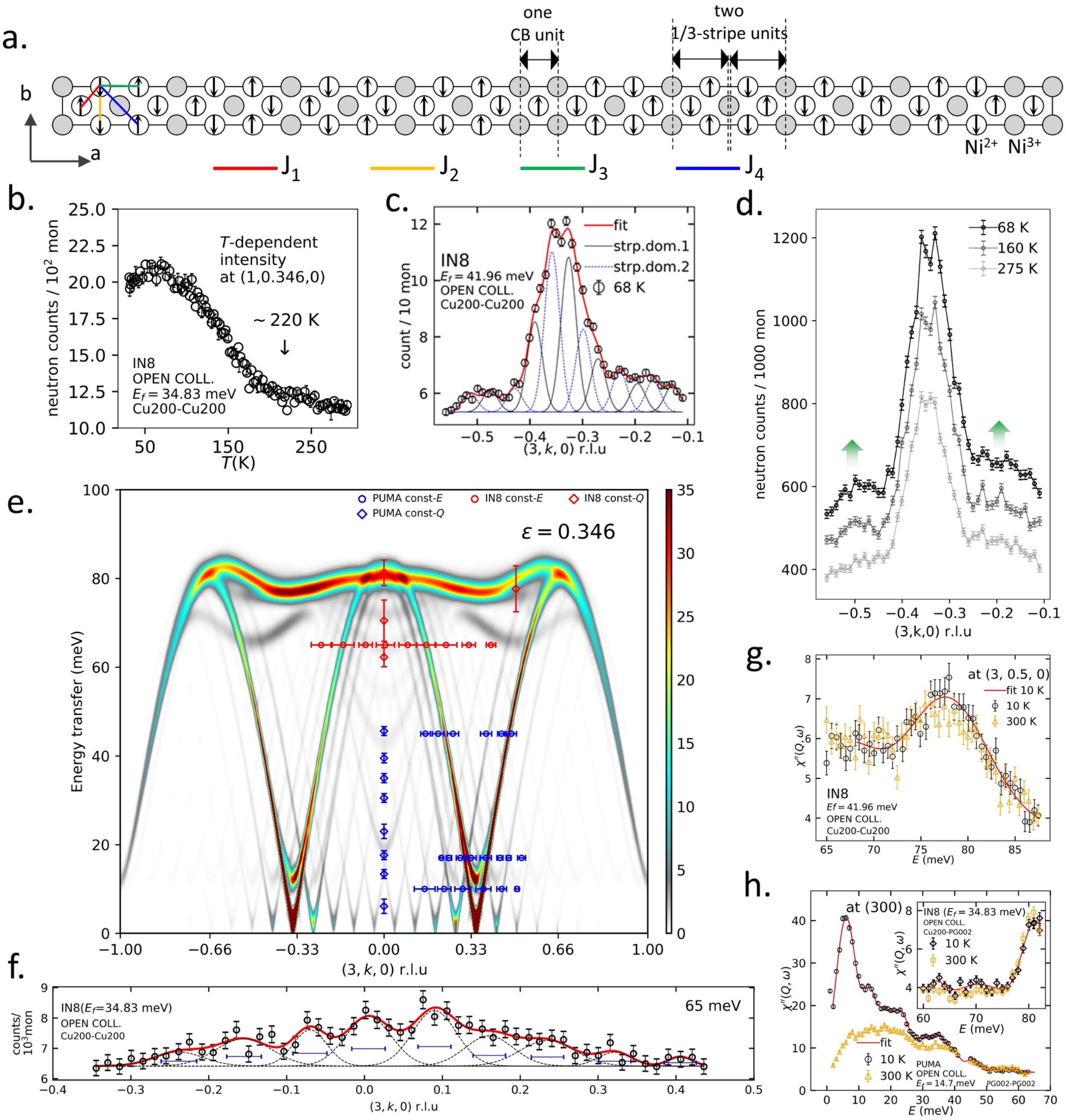

Figure 1: (a) DCSS unit along *a*-axis corresponding to the ε = 0.346 presented for a single twin domain. For the other orthogonal twin domain, the DCSS unit is the same but rotated by 90° around the *c*-axis. Solid lines in different colors indicate Exchange interactions. (b) *T*-dependent magnetic intensity of the spin stripe order satellite at (1, 0.346, 0). (c) Elastic scan through the magnetic satellite at (3, -0.346, 0). Multiple magnetic satellites in the elastic line separated by Δq_m = 0.076 r.l.u. (d) Temperature dependence of the scan, with decreasing temperature the intensity of side magnetic peaks increase along with the peak at the magnetic modulation. (e) Spin wave dispersion calculated from SpinW. The measured peak positions from the inelastic scans have been overlaid on the calculated dispersion. Horizontal and vertical bars represent the FWHMs of the peaks in Q and E respectively. (f) Constant-E scan performed at 65 meV on both sides of the AFM zone centers. (g,h) Bose corrected Constant-Q scans at (3, 0.5, 0) and at (3, 0, 0).

[1] A. Maity et al., Phys. Rev. Lett. 124, 147202 (2020).
[2] R. Dutta et al., Phys. Rev. B 102, 165130 (2020).