Experimental report

16/10/2023

Proposal: **CRG-2978**

Council: 10/2022

Title: Orbital magnetism in highly hole-doped two-leg ladder cuprates

Research area: Physics

This proposal is a new proposal

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Samples: $Sr_2Ca_{12}Cu_{24}O_{41}$

Instrument	Requested days	Allocated days	From	To
IN22	4	4	08/09/2023	13/09/2023

Abstract:

The abstract is in the scientific case pdf file.

Orbital magnetism in highly hole-doped two-leg ladder cuprates

Scientific Case: The family of the two-leg spin ½ ladder cuprates $Sr_{14-x}Ca_xCu_{24}O_{41}$ (hereafter: SCCO-x) has attracted a lot of interest, owing to the emergence of superconductivity upon substitution. The Ca-free compound $Sr_{14}Cu_{24}O_{41}$ is a quasi-1D system, which consists of two interpenetrating subsystems of CuO_2 chains and Cu_2O_3 two-leg ladders. It realizes an intrinsically hole-doped compound with an effective charge of +2.25 per Cu (mixed valence $Cu^{2+/3}$), where the holes are located within the chains subsystem. Substitution with Ca^{2+} on the Sr^{2+} site results in a transfer of the hole carriers from the chains to the ladders subsystem. Ca-doping results in a rich P-T phase diagram with various phases: spin liquid state, antiferromagnetic state, charge density wave and superconductivity under pressure. A long-range ordered antiferromagnetic (AF-LRO) phase was also reported for $x \geq 9$. The origin of the AF-LRO is however still unclear. Indeed, while it was attributed to AF ordering within the chains, it was also proposed to originate from AF spin ordering within the ladders. To account for such an AF state, one needs to elaborate a very complex magnetic pattern made of a large number of Cu spins.

We recently revisited the magnetic properties of two different SCCO single crystals with Ca doping levels x=5 and 8, using polarized neutron diffraction (PND) *[D. Bounoua et al, Commun. Phys.* ***3,*** *123 (2022)].* For both samples, PND measurements show the onset of a new magnetism. This magnetism is at short range and preserves the lattice translational invariance (q=0 magnetism). It further gives rise to scattering on top of Q-positions where no nuclear scattering is expected from space-group symmetry selection rules. The characteristic onset temperature for the magnetic correlations was found to be T_{mag}=50K and 80K for SCCO-5 and SCCO-8, respectively. The observed magnetism can be accounted for by an orbital magnetism produced by staggered loop currents (LCs) within the CuO_2 plaquettes of 2 leg-ladders. While LCs are expected to be absent in hole free ladders, they progressively develop upon hole doping. Interestingly, the increase of correlation lengths upon increasing the Ca-content goes along with the development of an AF-LRO at high Ca-doping. Besides, the LC-like q=0 magnetism appears on Q-positions where scattering from the AF-LRO was reported using PND. However, no AF-LRO is reported for SCCO-5 and 8, T_{mag}>>T_N . Considering that the reported spin models for AF-LRO are highly non-trivial, one may wonder whether the q=0 short range magnetism could not be a precursor phase to the LRO occurring at high Ca-doping.

A pilot PND experiment was carried out on the TAS IN22 (Exp CRG-2614), on a high quality SCCO-12 single crystal, where an AF-LRO occurs below T_N~2.7 K. This experiment revealed several features: (i) The orientation of the magnetic moments in the LRO phase is found in plane. (ii) A second magnetic phase compatible with a LC like phase is observed above T_N. The magnetic moment within this phase exhibits both an in-plane and out-of-plane components, in agreement with our previous reports in SCCO-5 and 8. (iii) The corresponding T_{mag} could not be accurately determined due to spurious scattering from (λ/2) contamination in the SF_X channel. A crude estimate T_{mag} gives ~65 K. (iv) The H-dependence of the magnetic scattering in the SF_X channel, in the tail of the λ/2 contamination suggested that the LC magnetism is at rather short range (see exp report CRG-2614). However, we were not able to determine its precise correlation length due to the tail of the λ/2 contamination. Thus, a better λ /2 filtration was mandatory to confirm this result.

A second experiment (Exp. #4-05-830) was performed on the IN22 TAS with the main aim of determining the correlation lengths and accurate temperature dependence of this q=0 magnetism. In order to get rid of the (λ/2) contamination, two PG filters were installed on k_f. However, the supplementary filtering and the lower reactor power (57 MW during CRG-2614 and 42 MW for Exp#4-05-830) yielded a strongly reduced magnetic intensity (for instance, on the magnetic Bragg (1,0,1) in the LRO phase, we find a ratio of 2.7 for the magnetic intensity) that prevented us from carrying our experimental plan.

Experimental Outcome: As a following up of those two experiments, we performed a third experiment on IN22 Exp CRG-2978, using the same sample. The experimental conditions were identical to the second experiment: Heusler/Heusler polarization set-up, Crypoad for the polarization analysis and 2 PG filters to remove higher harmonics on k_f (set to 2.662 $Å^{-1}$). The Reactor power was still lower than during the first experiment (43 MW instead of 57 MW).The flipping Ratio was about 18 in the three (XYZ) channels.

In the AF-LRO phase, the magnetic signal displays the same spin orientation as measured previously, but the magnetic intensity is only 1/5 of that of the first experiment and the signal/background ratio drops from 1/2 to 1/6. We noticed that certain nuclear Bragg reflections (+H,0,-L) and (+H, 0,-L) were actually missing at (+H,0,+L) and (-H, 0,-L), suggesting a sample misalignment. A surprising conclusion, keeping in mind that the sample alignment on its Al-holder was actually the same for all experiments. We failed to figure out the exact origin of such a problem.

Despite these degraded experimental conditions, we tried to measure the T-dependence of the magnetic signal at (-3,0,1). During the 1st experiment, the SF_X background was decreasing slowly on warming. At variance, in the present measurement, the background drops by 30 % between base temperature and 250 K. Owing to the weakness of the expected magnetic signal (~3 cnts/min) and the very poor signal/ background ratio (<1/30), we focused on the on a longitudinal polarization analysis at (-3,0,1) at fixed temperatures.

Fig. 1 a-c) report H-scans around (-3,0,1) in the SF_X channel at 10 K and 198 K. The magnetic signal left above T_N =2.5 K is not sizeable in the SF_X raw data. In order to estimate its magnitude, one needs to rely on a longitudinal polarization analysis. At 10 K, a weak magnetic signal of ~3 cnts / min is found at (-3,0,1) on top of an effective background. Such a background seems present at all temperatures (Fig. 1 c-e) and could be viewed as a threshold of detection of the magnetic signal (limited by the polarization accuracy and the low statistics). The temperature dependence, with polarization analysis, does not allow for an accurate determination of the onset temperature of the q=0 magnetism, T_{mag}. At best, our data suggest an upper value of T_{mag} ~30 K, which is even lower than the estimate we had from Exp CRG-2614.

If this trend is correct, this estimate would indicate that the q=0 magnetism starts decaying when the AF-LRO settles in at low temperature at large Ca content.

However, the need for additional filtering (to get rid of the λ/2 contamination) and the lower reactor power (43 MW) combined to the small magnetic signal amplitude, the small sample volume and possible misalignment, lead to only limited statistics preventing us from even reproducing the first data.

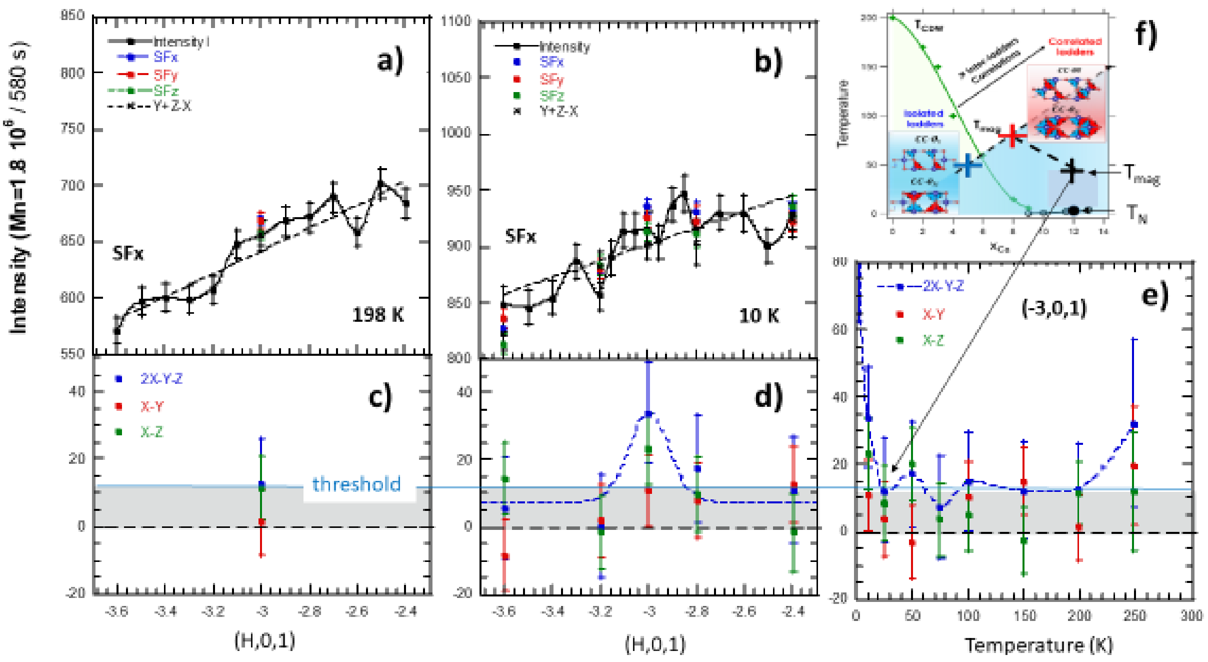

Figure 1 a-b) H-scans around (-3,0,1) in the SFx channel (black points): a) T=198 K, b) T=10 K. Data measured in the three polarization channels (SF_X, SF_Y, SF_Z) are reported in the H scan. c-d) H-scan around (-3,0,1) with XYZ polarization analysis, reporting the full magnetic intensity (2X-Y-Z) and the planar component along b (X-Z) and the complementary one (X-Y): c) at T=198 K, d) T=10 K. e) Temperature dependence of the polarization analysis at (-3,0,1). The magnetic intensity at 1.5 K (< T_n=2.5 K) is clearly visible. A weak magnetic signal is still observed at 10 K, but the signal left above 30 K does not display any T-dependence with error bars. The grey area in Fig. c-e suggests the existence of a threshold of detection of the magnetic intensity. Such a threshold may originate from the low statistics of our data and the experimental accuracy (stability of polarization in the XYZ channels) and. f) SCCO-x phase diagram.