Experimental report

21/10/2022

Proposal: **5-53-286**

Council: 10/2018

Title: Search for loop currents in two-leg CuO ladders

Research area: Physics

This proposal is a continuation of 5-53-279

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Samples: $Sr_9Ca_5Cu_{24}O_{41}$
$Sr_6Ca_8Cu_{24}O_{41}$

Instrument	Requested days	Allocated days	From	To
IN3	1	1	19/09/2019	20/09/2019
D7	10	4	20/09/2019	24/09/2019

Abstract:

First proposed by C.M. Varma, at the end of the 90's, to account for anomalous electronic properties observed in the phase diagram of the superconducting layered cuprates, the loop current (LC) phase has attracted more attention since its magnetic hallmark has been detected by polarized neutron diffraction in the pseudogap phase of 4 layered cuprate families, but also of a layered iridate compound. In addition, LCs are expected to exist in two-leg CuO ladders. Search for LCs in this system using polarized neutron diffraction has never been carried out so far in the literature. Therefore, we propose to investigate the existence of LCs in series of $Sr_{(14-x)}Ca_xCu_{24}O_{41}$ compounds (with x=5 or 8) using polarized neutron scattering on D7.

Search for loop currents in a two leg ladder-cuprate

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 [1]. 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 holes carriers from the chains to the ladders subsystem [2]. Ca-doping results in a rich P-T phase diagram with various phases: spin liquid state, antiferromagnetic state, charge density wave, superconductivity under pressure [3]. A long-range ordered antiferromagnetic (AF-LRO) phase was also reported for x≥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 [4-5]. To account for such an AF state, one needs to elaborate a very complex magnetic pattern made of a large number of Cu spins.

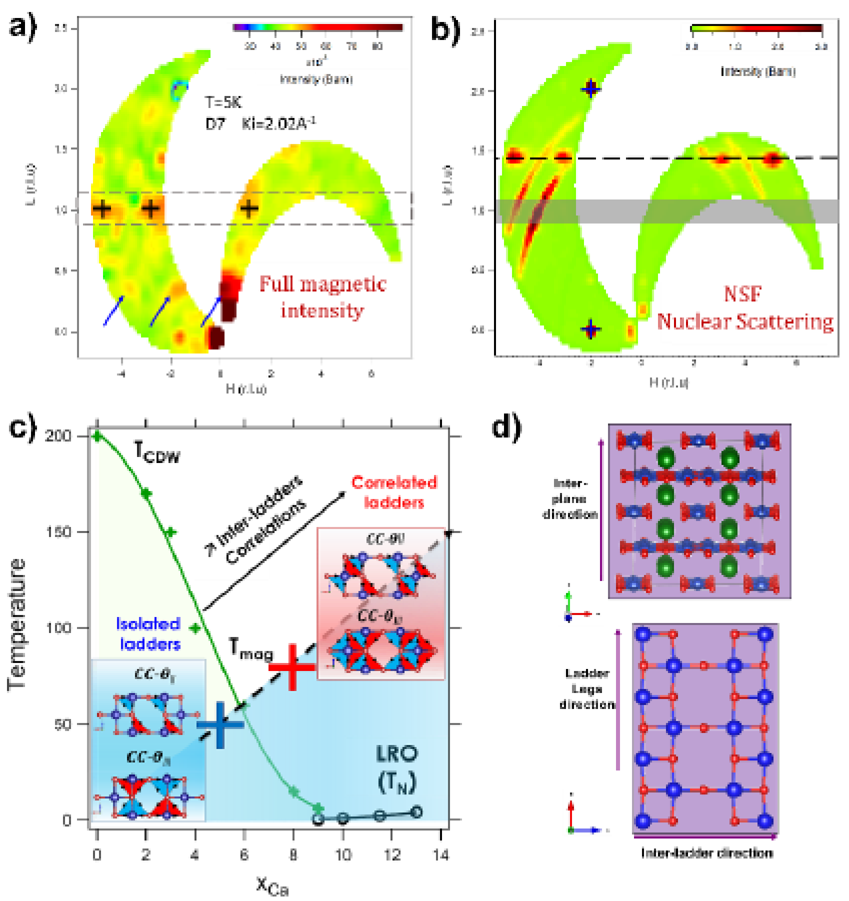

Figure 1 *(a) SCCO-8: Mapping of the full magnetic scattering at 5K, deduced from XYZ-PA on D7. The map is given in r.l.u of the ladders subsystem and the intensities in mbarn. The area bounded by dashed lines indicates the ladder scattering ridge along (H,0,1) with magnetic spots located by crosses. The blue arrows show the satellite magnetic reflections. (b) SCCO-8: Mapping of nuclear intensity measured in the NSFX channel. Spots at integer H and L values correspond to the nuclear scattering associated with the ladders, whereas the dashed lines are associated with the chains nuclear response. (c) Schematic phase diagram showing the evolution of the LC pattern as a function of the hole-doping (i.e. Ca-content). At large doping, inter-ladders correlations set-in and Tmag (crosses) increases. In heavily doped samples, an AFM-LRO further develops below TN of a few Kelvin. Insets: (Up) CCθ-II like model of LCs within one ladder unit cell with two staggered Cu-O orbital currents per Cu site flowing clockwise (red triangles) and anticlockwise (blue triangles). (Down) CCθ-III model of LCs, as derived from a spin liquid initial state [3], within the ladder cell consisting of two counter-propagating currents flowing between oxygen sites. (d) Up: Crystal structure of SCCO (Cu in blue, O in red and Sr in green). Down: [a,c] plane projection of the ladders planes. [6].*

We have revisited the magnetic properties of SCCO-x, using polarized neutron diffraction (PND) [6]. Our PND measurements in two different SCCO single crystals with Ca doping levels x=5 and 8, using two different instruments (The cold-TAS 4F1 and the multi-detector diffractometer D7), equipped with distinct neutron polarization set-ups and operating with 2 distinct neutron wavelengths (k_i=2.57Å^{-1} for 4F1 and k_i=2.02 Å^{-1} for D7). For both samples, PND measurements show the onset of a new magnetism. This one 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 [7] (Fig.1.a.b). The characteristic onset temperature for the magnetic correlations was found to be T_{mag}=50K and 80K for SCCO-5 and SCCO-8 (Fig.1.c), respectively. At low Ca content, only the magnetic response of one single isolated ladder is measured (SCCO-5 : ξ_c ~ 20 Å, along the ladders legs and no correlation along a, the ladders rungs , Fig.1.d), Increasing the Ca content, SCCO-8 exhibits finite correlations along both the a and c-axis (ξ_c ~11 Å and ξ_a ~ 6 Å). For both compounds, no magnetic correlations were found along the inter-plane direction (b-axis, Fig.1.d). The corresponding Q-dependence of the magnetic intensity along (H,0,1) can be accounted for by an orbital magnetism produced by staggered loop currents (LCs) within the CuO_2 plaquettes of 2 leg-ladders [8,9]. The result reminds the observation of loop currents confined in charged stripes ladders in lightly doped $La_{2-x}Sr_xCuO_4$ [10]. While LCs are expected to be absent in hole free ladders, they progressively develop upon hole doping [8,11]. Modeling our data by using two different patterns of LCs nicely captures the main features of our experimental results and give a very small amplitude for the corresponding magnetic moment (m ~0.05 μ_B). Our measurements further pinpoint the increase of correlation lengths upon increasing the Ca-content, going along with the development of a magnetic LRO at high Ca-doping. Besides, the LC-like q=0 magnetism appears on Q-positions where scattering from LRO was reported using PND [4-5]. However, no LRO is reported for SCCO-5 and 8, T_{mag}>>T_N (Fig.1.c).

Experiment # 5-53-286:

The objective of the experiment was to search for a possible magnetic response in the pure $Sr_{14}Cu_{24}O_{41}$. The sample consisted in four co-mounted single crystals of about 5 g total weight (4 times higher than $Sr_6Ca_8Cu_{24}O_{41}$ used in experiment #5-53-279). The sample was aligned in [a,c] plane of the ladders subsystem. A full rocking scan of 360° was first performed to check the alignment of the sample at 5K. Reflections of the form (H 0 0) and (0 0 $L_{C,L}$) where L_C and L_L correspond to the chains and ladders, respectively. The aim was to search for a possible magnetic signal occurring at positions of the form (H,0,1) with H=1 or 3 corresponding to the ladders subsystem and where a new magnetic signal was reported in Ca-doped samples.
Omega scans were performed using XYZ-PA first spanning a large portion of reciprocal space including both (1,0,1) and (3,0,1) Q-points at L_c=1 within the ladders scattering plane. The study was performed with a wavelength λ=3.1A and for two values of 2$\boldsymbol{\theta}$, namely, 75.5 and 80° in order to fully fill the dead zones corresponding to the hollow regions between detectors.

Results from NSF measurements at low temperature:

Figure 2 shows the NSFx map of the intensity at T=5K indicating nuclear scattering arising from the ladders subsystem (H,0,L) with L integer, the chains subsystem (H,0,1.43) and the characteristic charge order peaks [3] expected for both chains and ladders for this sample composition (free from Ca-doping).

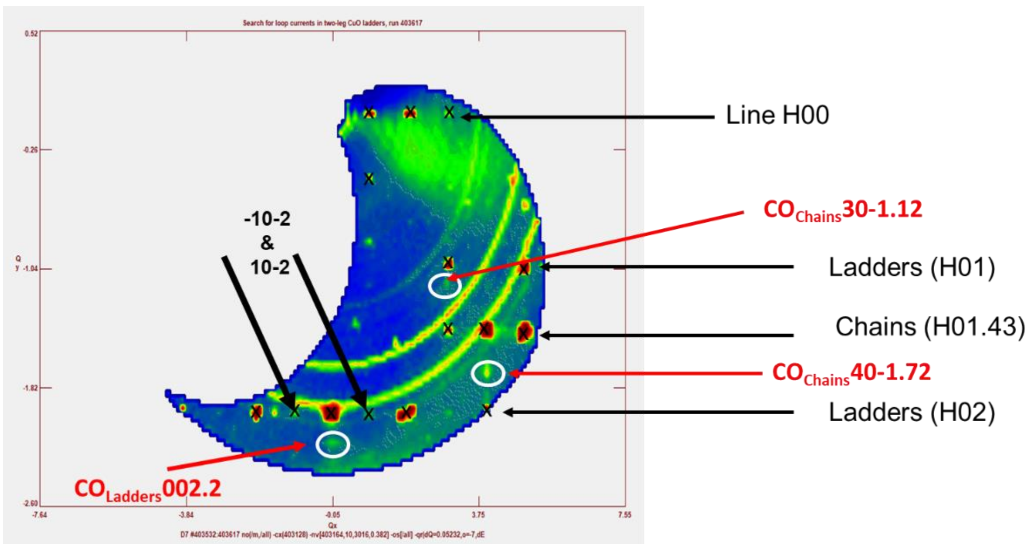

Figure 2: NSFx map collected at T=5K

Results from XYZ-PA measurements at low temperature:

Figure 3 shows the resulting magnetic intensity at T=5K (2*SFx-SFy-SFz, corrected by an angle α=41.6° corresponding to the Scharpf angle between X and ki). The map was obtained first by collecting data at a large number of omega values (86 in total) to span both (1,0,1) and (3,0,1). Owing to the presence of nuclear scattering at (3,0,1) arising from a change of the space group selection rules where moving from Ca-doped samples to undoped ones, we then focused on the region around (1,0,1) performing 20 omega scans around that Q-position to enhance the data quality and draw a conclusion. The map shown on Fig.3.a is a combination of both datasets and shows no feature of magnetism occurring at either ladders Q-positions but scattering at (4,0,2) and (0,0,2) most probably arising from polarization leakage at these strong nuclear Bragg peaks positions. The polarization imperfections were corrected using a quartz sample and the intensity converted to absolute units using a vanadium reference sample. Fig.3.b shows an H-scan across (H,0,1) showing the absence of magnetic scattering or any peculiar structure within an error bar of ∓10 mbarns.

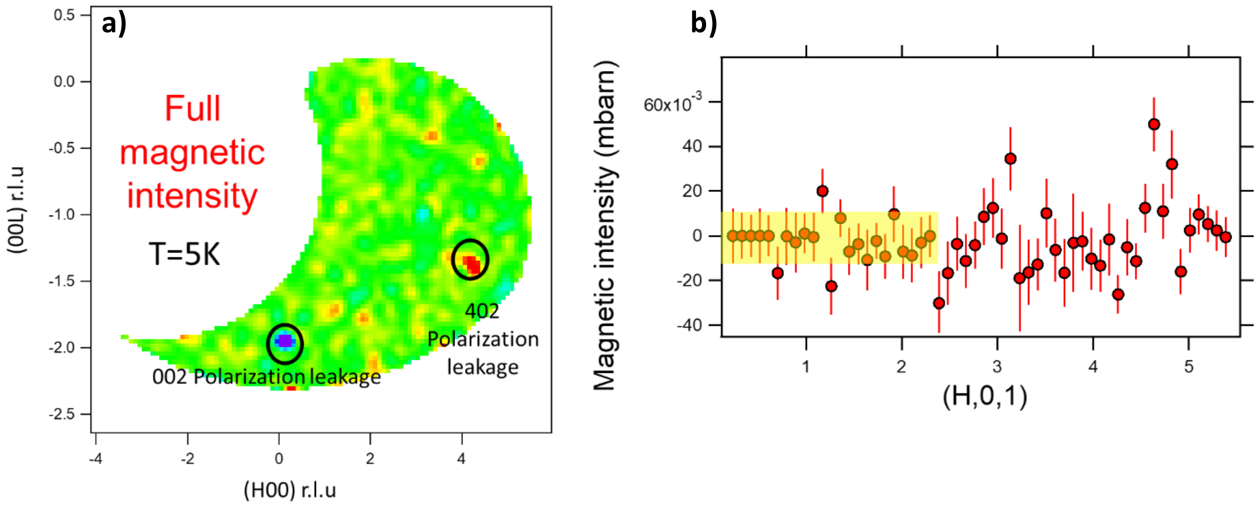

Figure 3: a) Full magnetic intensity as extracted from XYZ-PA at T=5K. **b)** H-scan along (H,0,1) extracted at L=3 from **a**.

References

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