Experimental report

21/10/2022

Proposal:	**4-05-830**		**Council:**	10/2020

Title: Orbital magnetism in two-leg ladder cuprate

Research area: Physics

This proposal is a new proposal

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Samples: $Sr_2Ca_{12}Cu_{24}O_{41}$
$Sr_8Ca_6Cu_{24}O_{41}$

Instrument	Requested days	Allocated days	From	To
IN22	13	7	19/05/2021	26/05/2021

Abstract:

We discovered the existence of a new kind of short-range magnetism in the two-leg ladder $(Sr,Ca)_{14}Cu_{24}O_{41}$ family of cuprates [1]. Observed for the Ca contents x = 5 and 8, this magnetism develops within the two-leg ladders and exhibits increasing correlations with increasing hole-doping. Its magnetic scattering further displays a very specific momentum dependence, the hallmark of a theoretically predicted loop current magnetism in such a material [2, 3]. Our discovery brings a novel insight onto the nature of the magnetic properties of hole-doped two-leg ladder cuprates and suggests that a loop current phase could act as a precursor to the long range magnetic order state at larger Ca content. We propose to investigate the existence of possible magnetic fluctuations related to the LCs like phase for the x=8 and 12 Ca-doped samples using polarized neutrons To this aim, we ask for 13 days on IN22 equipped with CRYOPAD.

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

SCIENTIFIC CASE: The family of the two-leg spin ½ ladder cuprates $Sr_{(14-x)}CaxCu_{24}O_{41}$ (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 $x \geq 9$. 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 [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.

We recently 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. For both samples, PND measurements show the onset of a new magnetism, (i) at short range and (ii) preserving 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 : $\xi_c \sim 20$ 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 ($\xi c \sim 11$ Å and $\xi_a \sim 6$ Å). For both compounds, no magnetic correlations were found along the inter-plane direction (b-axis, Fig.1.d).

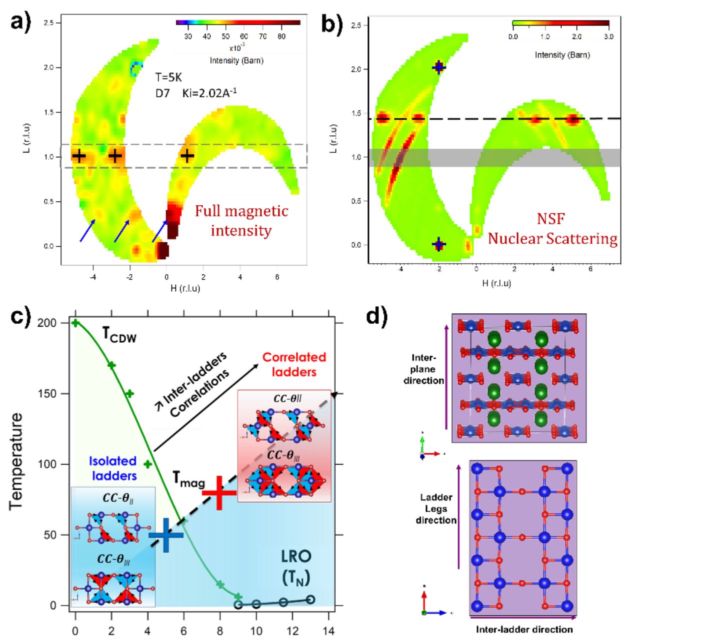

*FIG. 1: **(a) SCCO-8:** Mapping of the full magnetic scattering at 5K, deduced from XYZ-PA on D7. The map is given in r.l.u of the ladders subsystem and the intensities in mbarn. The area bounded by dashed lines indicates the ladder scattering ridge along (H,0,1) with magnetic spots located by crosses. The blue arrows show the satellite magnetic reflections. **(b) SCCO-8:** Mapping of nuclear intensity measured in the NSF_X 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 T_{mag} (crosses) increases. In heavily doped samples, an AFM-LRO further develops below T_N 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) [8]. (Down) CCθ-III model of LCs, as derived from a spin liquid initial state [9], 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.*

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]. While LCs are expected to be absent in hole free ladders, they progressively develop upon hole doping [8,10]. Modeling our data by using two different patterns of LCs nicely captures the main features of our experimental results. Our measurements further pinpoint the increase of correlation lengths upon increasing the Ca-content. Interestingly, this 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 [4-5]. However, no AF-LRO is reported for SCCO-5 and 8, $T_{mag} >> T_N$ (**Fig.1.c**). Additionally, considering a phase of AF interacting spins would give to scattering on top of half integer positions of the form $(\frac{H}{2}, K, \frac{3L}{2})$. Instead, the reported spin models for AF-LRO [4-5] are highly non-trivial which raises questions about the nature of the interactions within this phase (spins? orbitals? LCs?). Within this picture, the **q**=0 short range order could naturally be a precursor phase to the LRO occurring at high Ca-doping.

Polarized neutron Diffraction in SCCO-12

Exp CRG-2614 : A pilot PND experiment was carried out on the TAS IN22 (see experimental report CRG-2614), on a high quality SCCO-12 single crystal, where an AF-LRO occurs below $T_N \sim 2.7K$ as confirmed by our measurements. This experiment revealed several features: i) The orientation of the magnetic moments in the LRO phase is found to be in plane according to polarization analysis (XYZ-PA), consistent with earlier reports [3] ii) A second magnetic phase compatible with a LC like phase is found above T_N. The magnetic moment within this phase exhibits both an in-plane and out-of-plane components . These observations are consistent 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, which did not allow us to perform a proper temperature dependency. However, an estimate of about $T_{mag} = 65K$ can be extracted from the fit to the XYZ-PA results at different temperatures. iv) The H-dependency of the magnetic scattering in the SF_X channel, in the tail of the λ/2 contamination is consistent with a typical correlation length of ~8 Å, suggesting that the LC magnetism is still at rather short range. This result should be confirmed by further measurements with a better λ/2 filtration.

Exp #4-05-830: this second experiment 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 graphite filters were installed on k_f. However, the supplementary filtering and the lower power reactor (57 MW during CRG-2614 and 42 MW for Exp#4-05-830) yielded a strongly reduced magnetic intensity. For instance, on Bragg (1,0,1) in the LRO phase, we find a ratio of 2.7 for the magnetic intensity. Meanwhile, the spin flip background increased by 36 %. These effects, all together, prevented us from carrying our experimental plan, owing the degraded signal/background ratio and time required to get a decent statistics for a full XYZ polarization analysis. The few data collected did improve our understanding of the q=0 magnetism in SCCO-12. The H- and T-dependences of the magnetic signal remain to be measured accurately with optimal experimental conditions.

References

[1] M.Uehara et al., *J. Phys Soc Jpn*, **65**(9), 2764 (1996).
[2] Y. Gotoh et al., *Phys. Rev. B* **68**, 224108 (2003).
[3] T. Vuletic, et al., *J. Phys.Rep* **428**(4), 169–258 (2006).
[4] M.Isobe, M et al., *Phys. Rev. B* **59**(13), 8703 (1999).
[5] G.Deng, et al., *Phys. Rev. B* **88**(17), 174424 (2013).
[6] D. Bounoua et al Commun Phys 3, 123 (2020).
[7] $Sr_{14}Cu_{24}O_{41}$ crystallizes in an orthorhombic structure (Fig.1.e) The chains and ladders subsystems are described by the orthorhombic space groups *Amma* and *Fmmm*, respectively, that interpenetrate incommensurately along the c-axis with 10*cChains=7*cLadders. Upon Ca doping, the chains sub-space group changes from *Amma* to *Fmmm* such that the whole structure was described by Deng *et al.,* as belonging to *Xmmm(00g)ss0* superspace group.
[8] C. M. Varma. Phys. Rev. B 73, 155113 (2006).
[9] S. Scheurer et al., Phys. Rev. B 98(23), 235126 (2018).
[10] P. Chudzinski et al., Phys. Rev. B 76, 161101(R) (2007); Phys. Rev. B 78, 075124 (2008); and Phys. Rev. B 81, 165402 (2010).