Experimental report

17/09/2018

Proposal:	**5-53-279**	**Council:**	4/2018
Title:	Search the the Hallmark of anapoles in CuO using polarized neutron Diffraction		
Research area:	Physics		

This proposal is a new proposal

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Samples: CuO

Instrument	Requested days	Allocated days	From	To
IN3	0	2	03/09/2018	05/09/2018
D7	10	6	04/09/2018	10/09/2018

Abstract:

The possible existence of an anapole order, breaking time-reversal symmetry, in the simplest cuprate compound CuO has attracted a lot of attention, as it could shed a new light on the nature of the ground state that governs the physics of the Cu-O based-compounds. In order to detect the hallmark of anapoles, S.W. Lovesey derived an expression that could be used to detect anapoles by means of polarized neutron diffraction. But, to date, no study has been yet attempted. Therefore, we propose to carry out a polarized neutron experiments on D7 to search for anapoles in CuO, following Lovesey's model.

Experimental Report # 5-53-279

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1. Initial proposal

The initially proposed experiment was to measure the signature of ordered anapoles accompanying the AF1 collinear order in tenorite CuO. The AF1 phase exhibits a propagation vector of the form [$\frac{1}{2}$,0,$-\frac{1}{2}$]. While the anapoles were expected to be confined in the [a,c] plane, the AF spin magnetic moments order along the b-axis. Such a study required to be performed in the temperature range $T<T_{N1}$ and with the sample aligned in the scattering plane [1,0,-1]/[0,1,0] such as wave vectors of the form [$\frac{1}{2}$+h,1,-$\frac{1}{2}$+l] would be accessible, allowing with the use of polarization analysis on D7 to access the anapole component of the measured magnetic signal as proposed by S.W.Lovesey [1].

The first encountered major problem is that the sample of CuO wasn't aligned in the right scattering plane (Fig 1.a). Attempts to perform the alignment using IN3 TAS and then Orientexpress came out to be complex owing to the monoclinic crystal structure of CuO and to the fact that no goniometer is available on D7 such that the sample had to be perfectly aligned in the corresponding scattering plane.

The first day of beam time was therefore use to perform various calibrations of the instrument, and after 24H of unsuccessful alignment, we decided to move to measuring a back-up sample in order not to waste the beam time, that is the spin ladders compound $Sr_6Ca_8Cu_{24}O_{41}$. Note that this study is the same as the one proposed by **Exp# 4-02-527** on IN12 TAS.

On the other hand, the CuO crystal was finally tightly aligned in the scattering plane of interest after the end of beamtime on D7 (Fig 1.b).

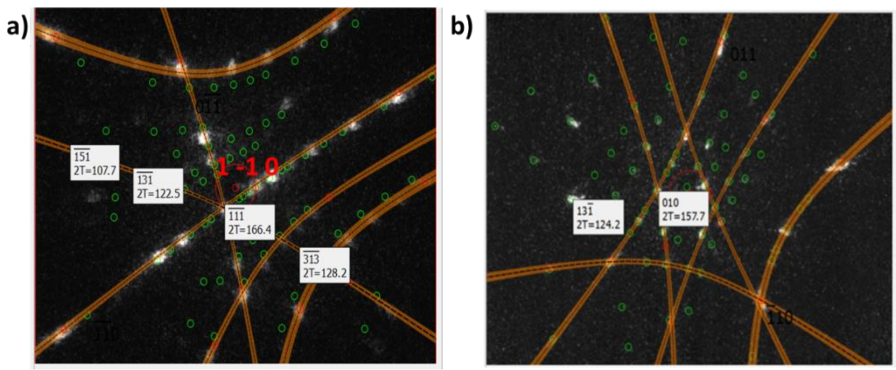

FIG. 1: LAUE pattern of CuO as determined by Orientexpress **(a)** At the beginning of the experiment, with [1 -1 0] in the scattering plane **(b)** tightly aligned with [1 0 -1]/[0 1 0] scattering plane.

2. Scientific case

The spin chains and spin ladders family $(Sr,Ca)_{14}Cu_{24}O_{41}$ exhibits pressure induced superconductivity upon Ca (hole) -doping. Its crystalline structure consists of two incommensurately interpenetrating chains and ladders subsystems. The ladders form arrays of CuO_2 units running along the c-axis. These (Fig 2.a) systems order antiferomagnetically with a decreasing T_N from 2 to 0.6K for x_{Ca}= 0 to 11.6 (Fig 2.c) [2].

Beyond conventional magnetism carried by S= ½ Cu spin, another form of magnetism has been proposed in this family of cuprate: Chudzinski et al., [3] predicted the onset of LCs phase within the Cu-O planes of the two-leg ladders, following the pattern proposed by Varma. The LC pattern is expected to be analogous to the one measured in $La_{2-x}Sr_xCuO_4$ (x=8.5%) [4], where LCs are likely to be confined within charge rich ribbons (stripes) within CuO2 planes. The ribbons would correspond to the two-legs ladders in $(Sr,Ca)_{14}Cu_{24}O_{41}$, where the short correlation lengths along the ladder rungs

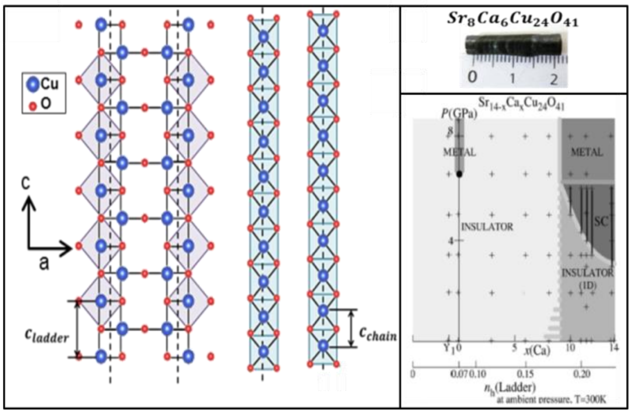

FIG. 2: ***(a)*** *Projection on the [a,c] plane of the edge sharing CuO_2 chains (right) and Cu_2O_3 ladders (left). The Cu ions are bridged at 90° by oxygen. The inter- and intra-ladders Cu-O-Cu bridges form binding angles of 180°. The figure shows 1 ladder+ 2 inter-ladder spacing as referred to in the text. Within the ladder, each Cu is surrounded by 4 O atom .* ***(b)*** *Single crystal of $Sr_6Ca_8Cu_{24}O_{41}$. The single crystal was grown by the travelling solvent floating zone method using an image furnace at SP2M-ICMMO.* ***(c)*** *Phase diagram of $Sr_{(14-x)}Ca_xCu_{24}O_{41}$ at 300K versus pressure and Ca concentration (x). At x=0, $Sr_{14}Cu_{24}O_{41}$ undergoes an insulator-metal transition above P= 6GPa. For $0<x\leq 8$ the compounds remain insulating upon increasing x or P. For $x\geq 10$ the insulating state undergoes a crossover to 1D insulator. Upon increasing P, superconductivity (SC) emerges and vanishes above 5.5 GPa to a metallic behaviour [2].*

are intended to result in a diffuse magnetic signal along the a-axis, while larger correlation lengths would set in along the legs c-axis. Moreover, an earlier polarized neutrons scattering study of the ladder spin dynamics in the pure $Sr_{14}Cu_{24}O_{41}$, reported by Boullier *et al.,* [6] reveals a strongly anisotropic dynamical magnetic structure factor assigned to a possible signature of circulating orbital currents.

3. Experiment

The sample was aligned in [a,c] plane of the ladders subsystem. A full rocking scan of 360° was first performed to check the alignement 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. Note that only reflections (H K L) satisfying H+K even and H or L even for (H 0 0) and (0 0 L) are allowed.

The aim was to evidence a magnetic signal at (1 0 1) where no nuclear scattering was present. Thus, omega scans of ±21°, around the expected (1 0 1) position, were performed with a full polarization analysis, as follow:

- A time basis of (2x70+30)sec was used for counting 1:8 NSF:SF signal in the three polarizations. In the SF channel for instance, we counted 170x8 sec per point $\sim 23\,min$ multiplied by 3 polarizations and 21° omega rotation with a step of 1° leading to 24h total time. The previous time was further doubled as the measurements were performed 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.
- The same measurement was done in the NSF channel which leads to a total measuring time of 48+6 ➔ 54hours on (101) at 5K.
- The measurement was repeated on (-1 0 0) to take a low temperature reference where no magnetic signal is present. A single value of $2\boldsymbol{\theta}$ was used, that is 75.5°. We counted in total during 17 h on this position.
- We then counted (101) again at 300K, with half statistics than 5K, leading to 26 supplementary counting hours.
- The remaining time was dedicated to temperature stabilization, instrument motion, and alignement check.
- The study was performed with a wavelength λ=3.1Å

4. Results

The key results arising from our measurements are the following:

- The measurements realized on (101) at 5K revealed a clear difference between the SF_y and $SF_{X,Z}$ signals (Fig 3.a). The longitudinal polarization analysis allowed to unambiguously evidence the magnetic nature of the signal (Fig 3.b). The measured signal is broad along a* as expected from short correlation lengths. The measured FWHM corresponds to $\frac{2}{3}\,a$, namely $\sim$1 ladder + 2 inter-ladder spacing (Fig 1.a).

- The measurement carried out on (-1 0 0), another extinction from the lattice, and didn't reveal such a signal (Fig 3.c).
- With the actual data statistics, it's not clear whether a magnetic signal persists on (101) at 300K or if the observed intensity is just resulting from an increase of the background at high temperature (Fig 3.d). Further analysis of the data needs to be performed at this point.

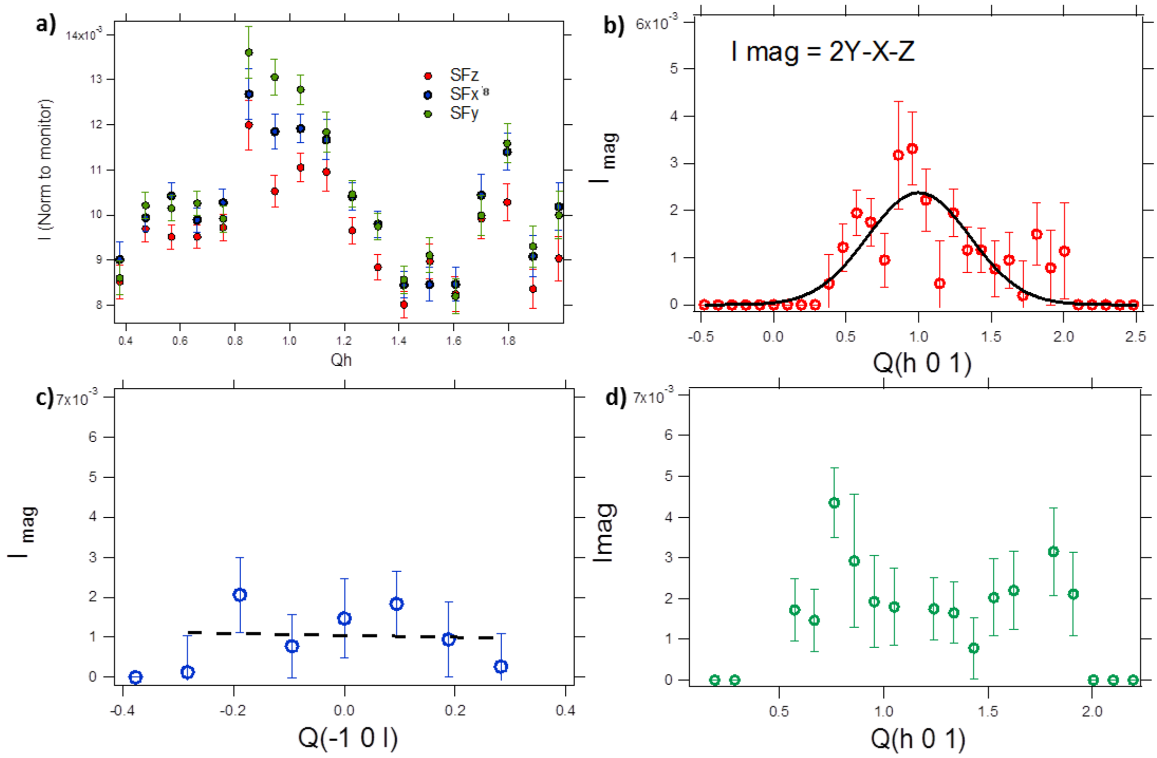

*FIG. 3:. **(a)** Measured signal in the SF (X, Y and Z) channels around (1 01), where SFy is enhanced. **(b)** Longitudinal polarization analysis result on the signal measured at (1 0 1) at 5K on D7, in $Sr_6Ca_8Cu_{24}O_{41}$. Note that in the used configuration for D7, the Q vector (1 0 1) is parallel to Y giving the polarization sum rule: I_{mag}=2Y-XZ- **(c)** Longitudinal polarization analysis on another extinction, namely (- 1 0 0), showing the absence of a magnetic signal on this position at 5Kn. (**b)** Longitudinal polarization analysis result on the signal measured at (1 0 1) at 300K. The data were corrected for detectors efficiencies (Vanadium) and polarization (Quartz).*

5. Conclusion

Our resulting measurement on D7 on the spin chains and spin ladders system $Sr_8Ca_6Cu_{24}O_{41}$ shows the development of a magnetic signal on (1 0 1) within the ladder subsystem, equivalent to (1 1 0) in YBCO [5], where the observed magnetic signal was assigned to the onset of and Intra Unit Cell magnetism originating from circulating orbital currents within the CuO_2 planes. This study will surely motivate further investigations on D7. Especially, the observed diffuse signal strongly indicates weak correlations along the ladders rungs (along a), which makes D7 the most suitable instrument for such studies.

6. References

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[3] P. Chudzinski et al., *Phys. Rev. B* 76, 161101(R) (2007); *Phys. Rev. B* 78, 075124 (2008);
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[5] L. Mangin-Thro et al., Nat. Comm. 6, 7705 (2015) (and references therein).