

# Experimental report

27/09/2023

**Proposal:** 4-02-623

**Council:** 10/2022

**Title:** c-axis correlations of the bi-axial magnetism in YBa<sub>2</sub>Cu<sub>3</sub>O<sub>6.6</sub>

**Research area:** Physics

**This proposal is a continuation of** 4-02-587

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**Samples:** YBa<sub>2</sub>Cu<sub>3</sub>O<sub>6.6</sub>

Instrument	Requested days	Allocated days	From	To
ORIENTEXPRESS	0	1	14/06/2023	15/06/2023
THALES	8	7	14/06/2023	21/06/2023
IN12	8	0		

## Abstract:

The understanding of the pseudo-gap (PG) phase of the superconducting cuprates is a major scientific challenge. While various symmetry breakings in the PG state have been discovered, including  $q=0$  magnetism that preserves the lattice translational (LT) symmetry and incipient charge-density-wave order that breaks the LT symmetry. However, none of these states can (alone) account for the PG, i.e., the partial gapping of the Fermi surface. Recently, we reported a hidden LT-breaking magnetism. Our polarized neutron diffraction measurements revealed magnetic correlations in underdoped bi-layer YBa<sub>2</sub>Cu<sub>3</sub>O<sub>6.6</sub> single crystals that emerge below the PG onset temperature with (i) a planar propagation wave vector  $(\zeta, 0)$   $(0, \zeta)$  yielding a doubling or quadrupling of the magnetic unit cell and (ii) magnetic moments mainly pointing perpendicular to the CuO<sub>2</sub> layers. The purpose of the experiment is to determine the evolution of the c-axis correlations of such magnetism as a function of temperature. We thus ask for 8 days on Thales/IN12 equipped with cryopad to carry out these investigations.

# Experiment report 4-02-624 on IN-12 : Search for bi-axial magnetism in single-layer Hg1201

## I. Sample

The sample was an assembly of 20 crystals of  $\text{HgBa}_2\text{CuO}_{4+\delta}$  (HBCO) coaligned on aluminium plates for a total mass of 0.7g. Positions of the form H0L were accessible with the 0K0 direction aligned along the vertical lab direction (Z axis).

## II. Experimental Setup

The experiment was conducted on the IN-12 cold triple-axis spectrometer with a Helmholtz setup. The beamline starts with a chopper to ensure the proper energy of incident neutrons and a polarizer, which direct the polarization of the incident neutron beam in the Z direction. A Heusler monochromator select only the neutrons polarized in the -Z direction and further set the wave vector of the incident beam to  $1.55 \text{ \AA}^{-1}$ . Then, a first flipper can be used to change the spin direction of the neutrons beam toward +/-Z, we stayed in -Z polarization of the incident beam for this experiment. The sample was placed in a cryo-furnace to perform measurement on a wide temperature range below and above ambient temperature. The cryo-furnace was in the middle of a Helmholtz coil to choose the direction of the incident neutron spins before interaction with the sample. Three orthogonal direction are available: X,Y and Z. Two slits placed along the incident and diffracted beam paths on both side of the Helmholtz coil reduce the effect of diffraction coming from the setup components near the sample. Another flipper was placed on the diffracted beam path to choose to look at spin flipped (SF) or not spin flipped (NSF) diffracted neutrons. Eventually, a Be filter cooled inside a cryostat added on the diffracted beam path completely absorb the  $\lambda/2$  photons. After that, another Heusler analyzer select both the diffracted neutrons energy and spin direction before they hit the detector.

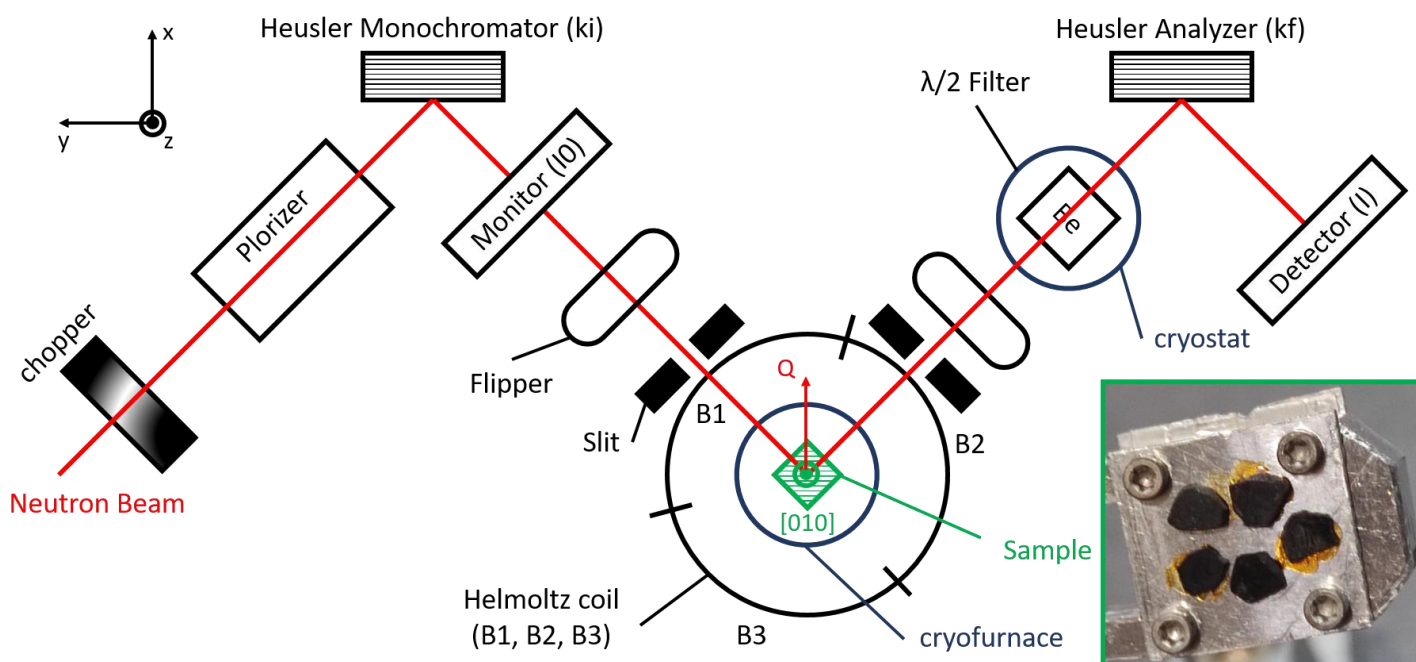


Fig 1. Scheme of In-12 Helmholtz setup used for measurements. Inset shows a picture of the montage with co-aligned  $\text{HgBa}_2\text{CuO}_{4+\delta}$  crystals glue on aluminum plates.

## III. Protocol

The experiment mainly consisted of polarization analysis on points of the reciprocal space along the H00 direction in SF configuration. Points with  $H = 0.2, 0.25, 0.3, 0.4, 0.45, 0.5, 0.55, 0.6, 0.7, 0.8, 1.2, 1.3, 1.4, 1.5$  was studied at 100K and points 0.2, 0.3, 0.5, 0.6 at 400K.

## IV. Results

A first polarization analysis has been initially performed in the H00 direction with a default Helmholtz coil orientation noted as  $\text{Helm}=0^\circ$  (**Fig 1-a**). From this analysis, the magnetic intensity along the Y and Z directions corresponding respectively to the a and b lattice directions inside the sample were extracted and plotted in Fig 1-b. The total magnetic intensity was also extracted too and plotted. The extraction has been done with the

usual formulas of polarization analysis assuming that both background and flipping ratio independent are of the channel:

$$I_{SFY} = I_{m,Y} + I_{m,Z} + I_{Bg} + \frac{I_{NSFY}}{FR}$$

$$I_{SFZ} = I_{m,Z} + I_{Bg} + \frac{I_{NSFZ}}{FR}$$

$$I_{SFZ} = I_{m,Y} + I_{Bg} + \frac{I_{NSFZ}}{FR}$$

with the flipping ratio defined on a Bragg peak as :

$$FR = \frac{I_{NSF}}{I_{SF}}$$

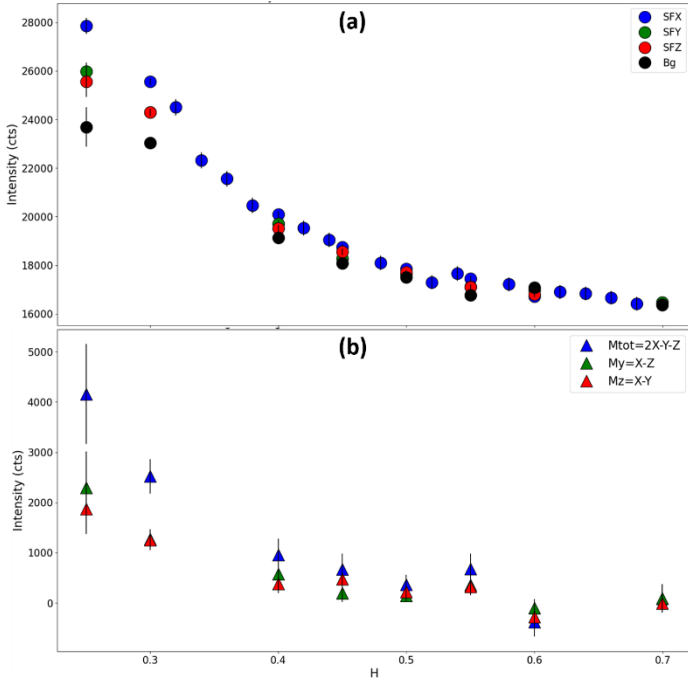


Fig 2. (a) Measured SF intensity along H00 in all X (blue), Y (green) and Z (red) channels at 100K with Helm=0°. Background has been extracted assuming it is the same for the three channels and added as black points. Total magnetic Intensity and magnetic intensity in both Y and Z directions were extracted from the SF intensity along H00.

We see that almost every point were a polarization analysis has been performed along H00 have some magnetic intensity except for H=0.6 and 0.7. This magnetic intensity is maximum at H=0.2 and decreases when H increases. Such a magnetic signal is too intense and inconsistent with every previous results on magnetism in High-Tc cuprates. We have determined it mainly comes from the diffracted beam coming too close to the Helmholtz coil pillars when approaching H=0. This cause inhomogeneous depolarization of the beam between the channels causing this artefact.

To get rid of this pillar artifact, we performed the same measurement but with the Helmholtz coil rotated by a certain angle noted Helm. In total, the position of the Helmholtz coil was rotated several time during the experiment. Helm=15° allows us to measure the H00 points up to 0.7. The coil was then tuned to Helm=-5° to measure the points beyond 100. The flipping ratio was measured on the main 002 Bragg peak for each of the Helmholtz coil positions. The results can be find in the following table:

Helmoltz, T \ Channel	0°, 100K	15°, 100K	-5°, 100K	15°, 400K
X	28.0	25.8	25.7	28.0
Y	27.0	26.5	26.2	27.3
Z	28.7	26.8	26.8	27.9

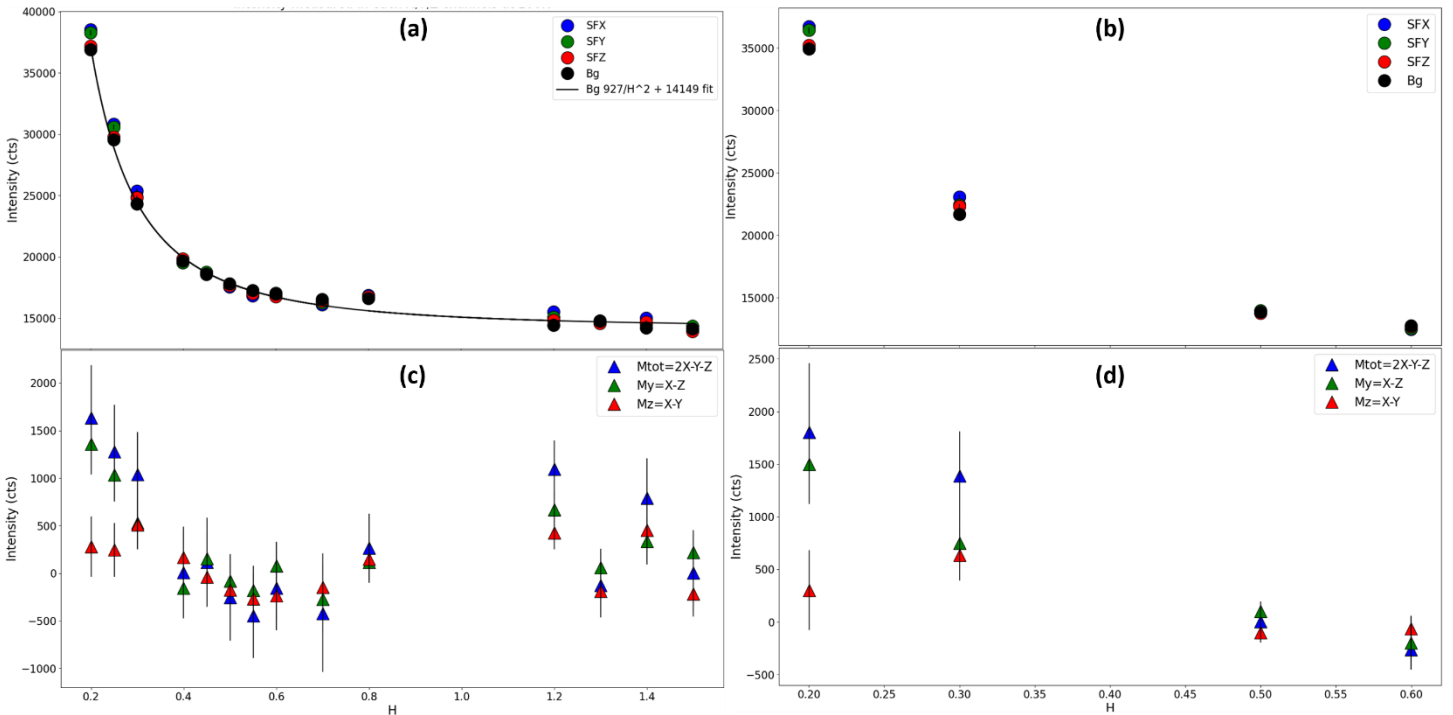


Fig 3. Measured SF intensity along H00 in all X (bleu),Y (green) and Z (red) channels at 100K (a) and 400K (b) with Helm=15° for H below unity and Helm=5° beyond. Background have been extracted assuming it is the same for the three channel and added as black points. Total magnetic Intensity and magnetic intensity in both Y and Z direction extracted from the SF intensity along H00. The black curve in (a) is a 2<sup>nd</sup> order inverse fit of the background.

We see that the flipping ratio is consistent regardless of the Helmholtz configuration and varies between 25.8 and 28.7, which gives a polarization leak in the SF channel of 3.5% to 4% of the NSF intensity.

In this second dataset with Helm at  $-15^\circ$ , the pillar artifact is no longer present. As expected the magnetic Intensity drops for every point (**Fig 2-a**). A small magnetic Intensity could be seen between  $H=0.2$  and  $0.3$  decreasing with  $H$  (**Fig 2-b**). We associate this with  $H$  getting to close to the direction of the incident beam as it is shown by the background increase at low  $H$ . For  $H=0.4$  to  $0.7$  the magnetic intensity reaches 0 with several points where the SFX and SFY channel show higher intensity than the SFX channel. This should be impossible if the background is the same in the 3 channels but is likely just caused by statistical fluctuation of the intensity. On the  $[0.5\ 0\ 0]$  where we were looking for a magnetic intensity the cumulated error bars on the magnetic signal is of 900 counts for a background of 17800. It correspond to an error of 3% of the background. Any signal smaller than that would be invisible with current counting time and setup. The  $q=1/2$  magnetic signal observed in  $\text{YBa}_2\text{Cu}_3\text{O}_{6.6}$  (YBCO) was an intensity of 2% of the general background [1]. On top of that our sample mass was of 0.7g when the sample YBCO sample weight 2g and HBCO have only one  $\text{CuO}_2$  plan by elementary lattice when YBCO has tow. It means statistical errors of the background intensity are largely enough to hide the signal we are looking for if it have the same relative intensity as in YBCO. Another difficulty with HBCO is that Hg atom is a strong incoherent scatterer, causing a large elastic background. Eventually, At 400K the analysis of the data lead to the same results as at 100K with the background increasing strongly at low  $H$  with a small magnetic intensity emerging. At  $H=0.5$  and  $H=0.6$  as expected, we measure no magnetic signal (**Fig 2-c,d**).

## V. Conclusion

With the current experimental setup it would have been impossible to observe a signal as small as the  $q=1/2$  magnetic signal in  $\text{YBa}_2\text{Cu}_3\text{O}_{6.6}$ . Solutions need to be found to reduce the background or enhance the potential signal we look for. Taking a higher mass of HBCO sample is an option assuming the incoherent scattering coming from it does not also enhance the background.

## VI. References

[1] D. Bounoua et al., Comm.Phys 5, 268 (2022); D .Bounoua et al., arXiv:2302.01870 (under review in Phys Rev B).