Experimental report

17/02/2020

Proposal: **4-02-541** **Council:** 10/2018

Title: Search for low energy magnetic excitations in superconducting cuprates

Research area: Physics

This proposal is a continuation of 4-02-513

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Samples: YBa2Cu3O6.75

Instrument	Requested days	Allocated days	From	To
THALES	10	10	12/07/2019	22/07/2019

Abstract:

An intra-unit-cell (IUC) magnetism develops below the pseudo-gap (PG) temperature, T*, in the phase diagram of cuprate superconductors. Since the PG state is considered as being the mother-state out of which superconductivity emerges, it becomes crucial to understand the intrinsic nature of the IUC magnetic correlations and to search for the existence of related fluctuations that could contribute to generate anomalous electronic properties (marginal Fermi liquid, d-wave superconductivity). The present polarized neutron experiment is devoted to the search for new low energy magnetic fluctuations around the wave vector associated with the IUC magnetism reported by polarized neutron diffraction. The experiment will be performed on a new YBa2Cu3O6.75 single crystal, specifically grown for such an experiment.

TITLE: **Search for low energy magnetic excitations in superconducting cuprates**

INSTRUMENT : THALES + CRYOPAD
EXPERIMENT NUMBER : 4-02-541
USERS : D. BOUNOUA, L. MANGIN-THRO,Y. SIDIS, P. BOURGES
LOCAL CONTACT(S): M BOEHM, P. STEFFENS

We performed a polarized neutron scattering measurement on THALES equipped with its polarization set-up (Heusler monochromator and analyzer, and CRYOPAD). The final neutron wave vector was set to 1.5 $Å^{-1}$. A Be filter was installed on the scattered beam in order to remove higher order contaminations. The sample was installed inside a standard ILL orange cryostat. Note, here, that neither CRYOPAD, nor the orange cryostat, were part of the standard THALES sample environment. They were provided by the ILL sample environment department.

The sample was a $YBa_2Cu_3O_{7-\delta}$ single crystal, grown specifically for the experiment. At variance with regular $YBa_2Cu_3O_{7-\delta}$ large single crystals, our sample was green phase free. The green phase is usually used as a precursor in the sample synthesis, but about 15 % of the green phase powder are left in the final single crystal. The green phase orders magnetically at low temperature and gives a paramagnetic background at high temperature. Thanks to our new single crystal, we could overcome such a difficulty.

We used an optimally doped $YBa_2Cu_3O_{7-\delta}$ sample with a superconducting temperature of 92.7 K. In the phase diagram of underdoped cuprates, the superconducting phase develops at T_c deep inside the so-called for pseudo-gap state which shows up at T*. For our sample, the two phases merge into each other and T* becomes typically of the order of T_c.

The sample was aligned in (100)/(001) scattering plane, so that wave vector of the form (H,0,L) could be accessible. The wave vectors are given in reduced lattice units ($2\pi/a$, $2\pi/b$, $2\pi/c$) with a~b=3.85 Å (twinned sample), c=11.65 Å. An Intra-unit cell (IUC) magnetism - linked to existence of an orbital magnetism generated by (IUC) staggered loop currents - is expected in the vicinity of H=1. At very low energy, a dynamical and short ranged IUC magnetic response is expected well above T* ~Tc and could be redistributed in phase space below T*.

Based on a previous study in a different $YBa_2Cu_3O_{7-\delta}$ sample on IN16-B, we decided to move away from the (1,0,L) rod and to look at (0.75, 0, L). A quasi-elastic signal should be detected on elastic position with k_f=1.5 Å. If the magnitude of the signal was sufficient and counting time not prohibitive, its dynamical nature could be revealed by lowering k_f down to 1.1 Å. Finally, H and/E scan performed above and below T*~T_c should have indicated how the signal could shift in phase space.

We expected a magnetic signal of rather weak magnitude (a few percent of total scattering intensity). The use of THALES with its high neutron flux and its polarized set-up was essential to the success of the experiment. To extract the IUC magnetism, we had to perform a full polarization analysis with : (i) no magnetic field on the sample in order avoid the depolarization in the superconducting state, (ii) a high flipping ratio (FR) to minimize le leakage of the non-spin-flip (NSF) signal into the spin-flip (SF) channel, (iii) a very homogeneous polarization for the three polarizations, X,Y,Z, (IV) a rather uniform FR with 2θ, to check the existence/absence of the IUC magnetism at selected Q positions. Some of these criteria could be checked ONLY at the end of the experiment by replacing our sample with a reference Quartz sample.

Owing to the long counting time and the need to perform measurements in both SF and NSF channels for 3 polarization, it took us very long time (about 6 days) before understanding that something went wrong in our measurements. Fig. 1 shows the NSFx scattering at (0.75,0,0) at different times of the measurements. The scattering intensity is time dependent. This is due to a leak on the orange cryostat. The scattering by nitrogen gas increased with time. The scattering vanishes below 77 K (close to T_c !). The scattering of the neutron by a gas being damped upon increasing temperature, such a parasitic quasi-elastic scattering goes away from the elastic line on increasing temperature.

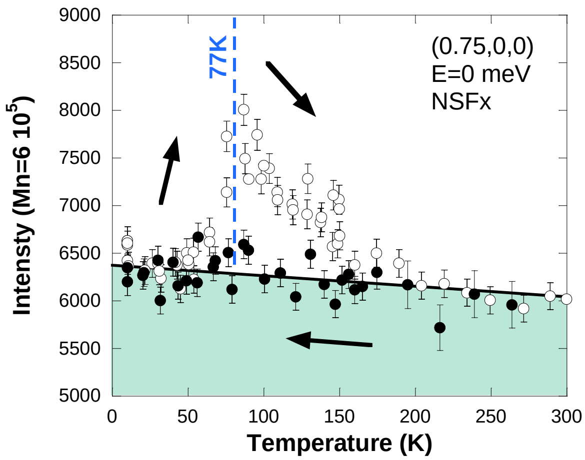


Fig. 1 : Scattered intensity in the NSFx channel as a function of temperature. The full symbols were measurements on cooling down after pumping the sample chamber. The open symbols were measured on warming up after few tens of hours. The contamination with nitrogen gas is straightforward owing to the marked change at 77 K (liquid/gas transition).

In order to save our neutron beamtime, we pumped continuously the sample chamber for measurements and used exchange gas only for changing temperature. Raw data in each channels were useless, but using a full polarization analysis one should be able to extract the IUC magnetism, under the assumption that FRz~FRy~FRz were large enough. The Quartz measurement at the end of the measurement confirm that the 3 flipping ration at (0.75, 0, 0) were quite homogeneous and as high as 30. Fig.2 show full magnetic scattering at (0.75,0,0) which drops down below T*~Tc. The magnetic signal appears on a "negative background due to the fact that the 3 flipping ratios FRx,Fry,FRz are close each other but not strictly identical. In order to estimate the background level, we rotated Q to (0,0,L) while keeping |Q| constant. At (0,0,L), the magnetic structure factor of the IUC magnetism is always null. Measurements at (0,0,L) can be used to estimate a background reference. The point (0,0,2.3) is reported in Fig. 2 and it materializes the negative background level.

In theory the "negative background level" could be also cross-checked by a measurement at (H,0,0) by moving away from H=0.75. Our measurement on Quartz revealed that when moving from H=0.75 down to H=0.5, the magnitude of FR drop from ~30 drawn to ~9. That is not acceptable: the flipping ration with CRYOPAD should uniform, i.e A4 independent. This problem should be fixed.

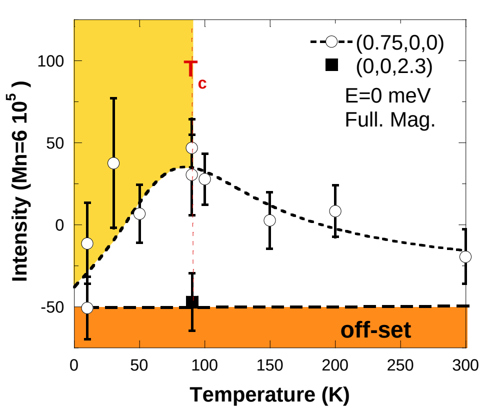


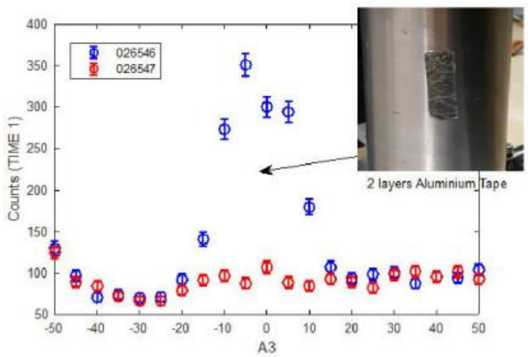
Fig.2 : Full scattered magnetic intensity at (0.75,0,0) obtained after full polarization analysis. The reference for the background level is given by a measurement at (0,0,2.3), which has the same [Q|, but for which the magnetic structure factor of the IUC magnetic response should cancel out..

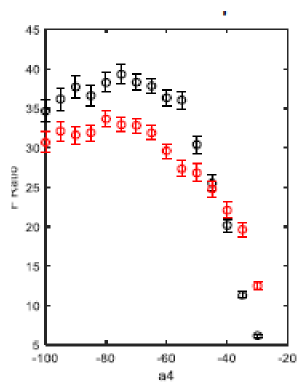
To conclude, we can consider our measurement as a "pilot" experiment, which provides indications that an IUC magnetism could be detected in $YBa_2Cu_3O_{7-\delta}$. The experiment is time consuming but is technically feasible…. at least with an orange cryostat and CRYOPAD working as they should. Despite of the strong support provided by our local contacts, we could not run the experiment in the proper conditions: the above-mentioned imperfections of the experiment set-up will be used to cast some doubt on the validity of our observation.

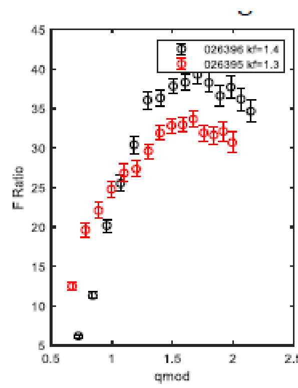
<u>Origin of the technical issues in Exp. : 4-02-541</u>

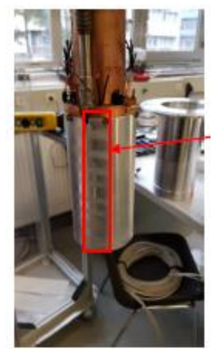
During Exp. 4-02-541, with the Cryopad on Thales, two issues were pointed out:
- a problem with the cryostat
- a variation of the flipping ratio with a4

The cryostat problem has been understood during the experiment and could be fixed after some time.
The polarization problem has been more puzzling and harder to understand. That is why the beam line scientist did few more tests, with a quartz sample. They repeated the tests several times and in several configurations, and they failed to reproduce a significant variation of the polarization with a4.
Only recently, they finally understood the origin of the reported problem. For very specific values of a3, they found an increase of background at low a4. With this, they were able to reproduce the mysterious observation. That means although that the polarization was constant, a higher background makes the flipping ratio worse. The scan on quartz illustrate that.









The cause of the problem is a piece of tape that was left inside the cryostat on a heat shield outside the calorimeter during maintenance work in the beginning of the year, and which creates strong incoherent scattering. On behalf of the ILL, we apologize for that. The tape has now been removed and the problem is fixed.
In particular, one can see that the effect of piece of tape shows up for a small a3 range only (when the tape is in the incident beam). This is the reason why it was not detected earlier. Also, this scattering is seen only at small a4.

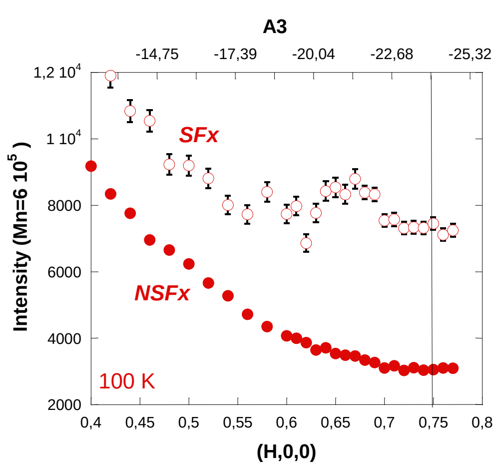


***Fig.3** : H scan across (0.5,0,0) in the SF and NSF channel for polarization X. data were measured at T=100 K.*

One clearly observe a net increase of the intensity in both SF and NSF channels at low A4, which is extremely surprising for this A4 area.

What is the consequence on data in Exp-4-02-541 ? For measurements at (0.75,0,0), a3 was -24. At a3=-15 the problem related to the Al place appeared. The a3-width of the contamination is about 30 degrees, so although we do not know for sure, it is possible that at a3=-24 the beam hit the tape. If so, the effect was constant in time. Fortunately, the a4 was almost all the time at -48 or below. Thus, one may think that the measurements was free from the contamination from the AL tape. Unfortunately as show in the H scan (Fig. 3), there is nevertheless a net increase in the data at small A3 and A4 (2xA3) in the data, which is clearly visible in data measured with a long counting time as ours. This up-turn of the scattered intensity in both SF and NSF channel was absent in the pilot experiment carried out on cold TAS 4F1 at Orphée/LLB. This demonstrates that the portion of the momentum space was outside the main contamination of the AF tape, but its parasitic scattering tail still affect the data. The up-turn of the intensity does come the sample.