Experimental report

17/09/2018

Proposal:	**4-02-513**	**Council:**	4/2017
Title:	Search for orbital magnetic fluctuations in YBa2Cu3O7		
Research area:	Physics		

This proposal is a new proposal

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Samples: YBa2Cu3O7

Instrument	Requested days	Allocated days	From	To
IN12	10	0		
THALES	0	7	14/03/2018	21/03/2018

Abstract:

In unconventional superconductors such as cuprates, the mechanism leading to high-temperature superconductivity has been challenging to understand. Although numerous physicists have concluded that quantum criticality of pseudo-gap phase plays a role in mediating the pairing glue, there is a longstanding disagreement on the real order parameter: either the antiferromagnetic order or another novel magnetic order, such as loop currents. Using polarized neutron diffraction, we have established an intra-unit-cell magnetic order in four different cuprate families. The associated magnetic fluctuations would be, according to the theory, directly related to the mechanism for the high-temperature superconductivity. In slightly overdoped YBa2Cu3O7, a ridge of orbital magnetic fluctuations is expected at low energy. We already have evidenced a peak in H-scan at 3 meV in this composition, using unpolarized neutrons. We here propose polarized inelastic neutron measurements to prove if the peak corresponds to the orbital magnetic fluctuations, and to follow it to higher energy. We request 10 days on IN12. IN12 is suited to achieve the best compromise of high flux and low background.

Experimental report - # 4-02-513 // THALES

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Scientific case: 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.

Experimental results: Using a $YBa_2Cu_3O_{6.75}$ single crystal (T_c=78 K, T*~200 K, hole doping p=0.14), we carried out a study of magnetic properties at finite energy around the momentum **Q**=(1,0,L), associated with the observed IUC magnetism in the PG state. Measurements were performed on THALES (k_f=1.5 Å^{-1}) equipped with its full polarized set-up (Heusler monocromator and analyzer, CRYOPAD). In addition to the speed selector on the incoming neutron beam, a Be filter was inserted in the scattered beam. The sample was aligned in (100)/(001) scattering plane and mounted inside a standard orange cryostat.

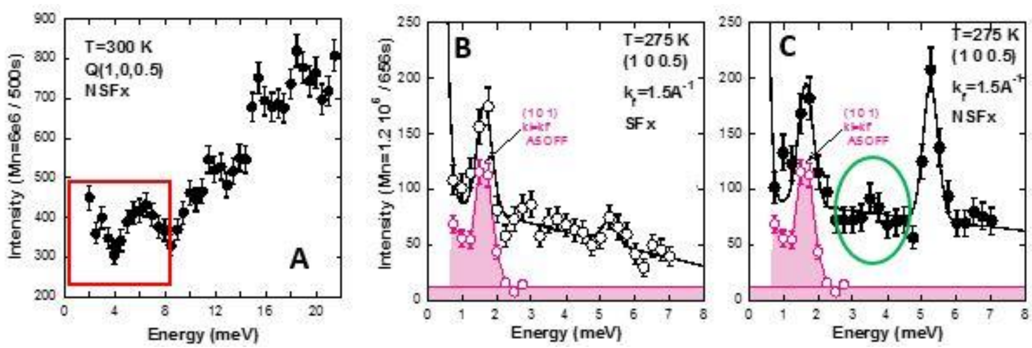

*Figure 1 Energy scans at **Q**=(1,0,0.5): A/ at 300 K in the NSF channel on IN20 (k_f=2.662 Å^{-1}) ,B,C) at 275 K (k_f=1.5 Å^{-1}) on THALES in the SF and NSF channels , respectively. Magenta symbols correspond to measurements performed A5.*

In a previous experiment (#4-02-472, see Exp. Report), the same sample was studied on IN20. The energy scan at (1,0,0.5) measured at 300 K in the non spin flip (NSF) channel is reported in Fig.1-A. On THALES, we focused on the energy range 0-8 meV, with a much better energy resolution. The phonon at 5.5 meV in Fig.1-A (IN20) is clearly visible on THALES in the NSF channel at T=275 K (Fig.1-C). In the spin flip (SF) channel, with the X polarization, its magnitude drops down by a factor ~10, corresponding to the typical flipping ratio (FR) available for inelastic measurements on the instrument. Prior to the study on THALES, a pilot experiment was performed using unpolarized neutrons (k_f=1.55 Å^{-1}) on the cold TAS 4F2 (LLB/Orphée). This study clearly showed that the energy scan at (1,0,0.5) was extremely clean and featureless in between the incoherent scattering (E<0.2 meV) and the lowest energy phonon (E~5.5 meV). Fig.1-B-C gives a different picture. A spurious scattering can be observed at ~1.5 meV. As demonstrated by measurements with the analyzer OFF (A5-OFF), this spurious scattering is not polarized. It corresponds to the (1,0,1) Bragg reflection through an incoherent scattering process on the analyzer. This leads to a first remark : the Be filter and the speed selector can improve the quality of the data, but do not totally eliminate the spurious scattering involving incoherent scatterings on the analyzer, but also on the monochromator. The efficiency of the Be filter is ~10^{-4} for k_f >1.55 Å^{-1} and the speed selector has an efficiency of ~10^{-5} outside a wave vector window of +/-25 % of k_i .The second remark is related to the unpolarized background level (A5-OFF) which remains quite significant, with ~1 count/min. Focusing now on the energy range between 2 meV and 3 meV, one easily identifies that the NSF signal is not featureless, a point that becomes clearer when performing constant E-scans.

Our previous polarized neutron study on IN20 suggested the existence of a tiny magnetic scattering at 7 meV and above T*. In addition, our unpolarized neutron scattering study, performed on the cold TAS 4F2 highlighted a signal at 3meV above T* (Fig.2). In Fig.2, the IUC magnetic fluctuation seems to vanish when the IUC magnetic order settles in. The signal appears on top of a nuclear background and a magnetic background. The later comes from the paramagnetic scattering of the so-called green phase, $Y_2BaCuO5$, which undergoes an antiferromagnetic transition at 28 K. About 15% of green phase powder are usually present in large $YBa_2Cu_3O_{6+x}$, as this phase is used as precursor in the sample growth method. The steep decrease of the scattered intensity in Fig. 2 at low temperature actually corresponds to disappearance of that paramagnetic background when entering the antiferromagnetic state of the green phase.

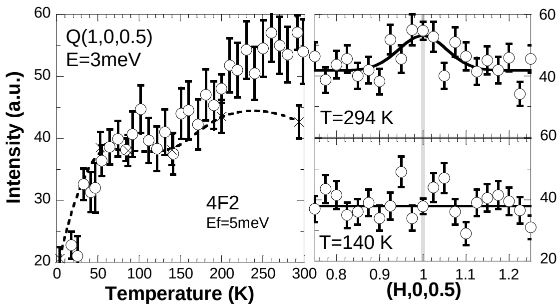

Figure 2 Unpolarized neutron data from the cold TAS 4F2 (k_f=1.55 Å^{-1}, double PG monochromator, PG analyzer, Be filter) on the same sample. Temperature dependence at Q=(1,0,0.5) and 3meV and constant E-scan at 294 K and 140 K. The dashed line indicates the position of a q-independent background.

Fig.3 shows a series of constant E-scans at 3 meV and at 3 different temperatures (2K, 140K and 275 K). Both NSF and SF scans display a spurious peak located at H=1.2. Since that spurious signal was absent in the unpolarized measurements from 4F2 (Fig.2), we believe it could be related to an incoherent scattering process on the monochromator (likely filtrated by the double-monochromator on 4F2). While Fig.1 shows that a measurement at 3 meV is far below the phonon with the lowest energy 5.5 meV, there is still a tiny signal centred at H=1 in the NSF channel. The magnitude of the signal, as a function of temperature, seems to be controlled by the detailed balance factor. This phononic-like signal is actually quite mysterious, because it shows up in an energy range free from phonons and, furthermore, was absent on unpolarised data (see Fig.2 at 140 K). This signal seems to be an artefact, whose origin remains unclear.

The scans reported in Fig.3 can be depicted as follows: The signal appears on top of the extrinsic background (dark grey) of ~1 count/ min. On top of it, one finds a featureless intrinsic background which hardly varies from the SF to NSF channel and is then characterized by an effective flipping ratio of ~1 (dark grey in SF and light orange). In the NSF channel, one observes the ghost-phonon contribution centred at H=1 (dark orange). Above 28 K, the SF channel is fully dominated by the paramagnetic scattering of the green phase (light green).

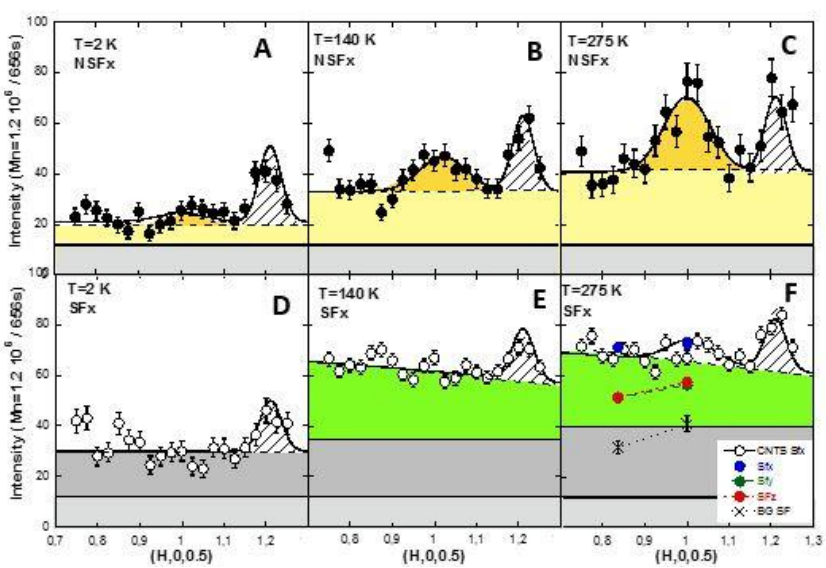

*Figure 3 Constant Energy scans around **Q**=(1,0,0.5) for neutron spin polarization X , in the NSF (A-C) and SF (D-F) channels. From left to right, the temperature are: T=2 K (A,D), T=140 K (B,E), T=275 K (C,F). The magnitude of extrinsic background measured with analyzer OFF, is indicated in light grey color. In F, the X,Y and Z polarization were measured at H=0.5 and H=0.6 and are reported in blue, green and red, respectively. The crosses indicate the SF background, determined from full polarization analysis.*

In the SF channel, at T=275 K (Fig.3-F), the scattered intensity is reported for the X,Y,Z polarizations. From a full paralysation analysis, one can determine the SF background (crosses given by Iy+Iz-Ix). Using a full polarization analysis, we found that the in-plane and out-of plane magnetic scatterings (given by Ix-Iz, Ix-Iy) are strictly equivalent, since the scattering intensities in Y and Z are equal and half of the X scattering. That is what one expects for a paramagnetic scattering. The full polarization analysis further reveals a slow decay of the magnetic scattering with increasing |Q|, consistent with the decrease of the green phase scattering controlled by the magnetic form factor of copper. Such a decay is also clearly visible in the raw data (Fig. 3-E-F). Since the SF channel is widely dominated by the paramagnetic scattering of the green phase (Fig.3 E-F) that vanishes at low temperature, the systematic use of the full polarization cannot be a relevant method to detect a tiny magnetic signal. On the contrary, focussing on the constant energy scans in the SF channel with the X polarization, there could be a rather weak signal peaked at H=1 and T=275 K (Fig. 3-E), that could be absent at T=140 K. To this respect, polarized neutron data on THALES (Fig.3 E-F) and unpolarised data on 4F2, seem to rather consistent and both data sets suggest the existence of a new type of magnetic fluctuations.

In conclusion, our study shows that our polarized neutron data suffer from 3 major drawbacks: (i) the paramagnetic scattering from an impurity phase (the green phase), (ii) incoherent scattering processes which are still present, (iii) a ghost-phonon contribution induced close to 3 meV, while the lowest phonon contribution is located at 5.5 meV. In spite of these experimental complications, our data in the SF channel suggest the existence of an intrinsic magnetic signal of 1 count / min, centered at (1,0,0.5). and a T evolution consistent with previous unpolarized results, supporting the existence of IUC magnetic fluctuations above the pseudo-gap temperature T*.

Thanks to the present experiment, we made significant progress in understanding how to refine the method and experimental procedure to prove or disprove the existence of the magnetic scattering associated with the IUC magnetism bound to the PG physics of superconducting cuprates. To make progress, it becomes crucial to grow $YBa_2Cu_3O_{6+x}$ without green phase. Once, such a sample will be available, one should be able to perform of similar type of measurements, using systematically the full polarization analysis to extract the magnetic signal, without the screening of impurity phase paramagnetic scatterings. Finally, changing the sample and perhaps changing a bit the neutron wave length and/or **Q**=(1,0,L) would contribute to weaken or remove the spurious scatterings.

Supplementary information: Once the green phase becomes antiferromagnetic, its paramagnetic fluctuations are replaced by a sharp excitation at 7 meV (Fig.4). This excitation is visible in the SF channel at 16 K and at **Q**=(1,0,0.9). Owing to the rather good flipping ratio of the spectrometer, its leakage in the NSF channel is hard to detect (Fig.4). This confirms that the polarization set-up is performant enough to disentangle the nuclear and magnetic scatterings. For sake of comparison, Fig.4-B shows the same measurement on 4F2. One can notice that in the SF channel on Fig.4-A, the signal/background ratio is ~20% better than for the unpolarized data. Combining the SF+NSF data and removing the extrinsic signal (A5-OFF), in order to avoid counting it twice, one can estimate the corresponding signal background ratio for an unpolarized measurement. It should be 20 % worse than on 4F2. This indicates, that one should be able to improve the experimental condition and further reduce the background.

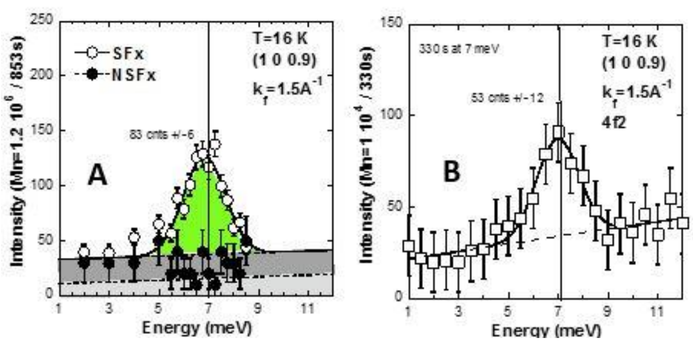

Figure 4 Energy scans at 16 K and Q=(1,0,0.9): A/ polarized neutron data on THALES (k_f=1.5 Å^{-1}) in the SF (open symbols) and NSF channel (full symbols), B/ Unpolarized neutron data on 4F2 (k_f=1.55 Å^{-1})