Experimental report

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Council: 4/2017

Title: Time-scale of the intra-unit-cell magnetism in high-Tc cuprates

Research area: Physics

This proposal is a new proposal

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Samples: YBCO6.6

Instrument	Requested days	Allocated days	From	To
IN16B	5	5	16/04/2018	18/04/2018
			20/04/2018	23/04/2018

Abstract:

The phase diagram of high temperature superconductors is dominated by a pseudo-gap phase with highly unusual physical properties. One theory predicted broken time-reversal and inversion symmetry due to ordered loop currents. Polarized neutron scattering experiments have reported the appearance of such magnetic order when entering the PG state in three different cuprate families. However, other magnetic probes such as muons spin resonance (µSR) and nuclear magnetic resonance experiments could not see the static local fields expected for the magnetic order. Recently, nevertheless, a µSR study reports a dynamic relaxation rate in longitudinal applied field in single crystals of $YBa_2Cu_3O_{6+x}$. The amplitude of the fluctuating magnetic fields is of the order of the magnitude deduced from polarized neutron diffraction. The magnetic correlations are fluctuating at about 10^8 Hz at low temperature, corresponding to an energy scale varying in the range 0.5-10 µeV. We then aimed to determine the finite time-scale of these magnetic correlations by using high energy resolution backscattering neutron spectroscopy. We asked for 5 days on IN16b to determine the magnetic fluctuations time-scale.

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Scientific case

The evidence for an Intra Unit Cell (IUC) magnetism, developing at the onset of the pseudo-gap (PG) state of high-T_c cuprates, was brought in several layered cuprates families, including YBCO, Hg-1201 and LSCO [1] using polarized neutron diffraction (PND). This IUC magnetism is associated with spontaneously circulating orbital currents within the CuO_2 planes and could be described by the staggered orbital magnetism within the unit cell as proposed in the loop current (LC) model for the PG state [8]. However, one major interrogation remaining, that is, why other magnetic probes such as muon spin resonance (µSR) [2] and nuclear magnetic resonance (NMR) [3] experiments fails to detect the static local fields generated by the IUC magnetism?

A recent µSR study reported the existence of magnetic fluctuations in single crystals of $YBa_2Cu_3O_{6+x}$ [4] with an amplitude of the fluctuating magnetic fields corresponding to the order of the magnitude of IUC ordered moments, deduced from PND [1]. Given the energy resolution of neutron TAS, which is of the order of 200µeV, the measured magnetic signal could then appear static in PND experiments while it can correspond to the low frequency dynamical fluctuations measured in µSR experiments.

The aim of this study was to measure the signature of such fluctuations owing to very high energy resolution of IN16B (0.8 µeV) and determine the finite time-scale of the magnetic correlations, which could help to bridge the gap between the static and dynamical probes.

Experimental details

Our experiment on IN16B (**Exp # 4-02-514**) was carried out on a twinned $YBa_2Cu_3O_{6.6}$ (T*=250K) sample with: a=b=3.85 Å; c=11.7Å. We used the Si (111) monochromator with a fixed incident wavelength λ=6.3 Å. The sample was aligned in the [h,0,0]/[0,0,l] scattering plane. The IUC magnetism can usually be observed at the Bragg reflection (10L) with integer L.

First, ω scans were performed in order to identify the (100) reflection, |Q|=1.63 Å^{-1}. Due to the bad Q-resolution, the Bragg peak intensity is spread over three detectors around the central value |Q|=1.63 Å^{-1} (or $2\theta=109°$). Indeed, IN16B possesses 18 detectors covering a 2θ range from 10.9° up to 142° with a step of 7°. The two first detectors, providing no relevant information are dismissed and our measurements rely on the 16 left detectors: (i) $D_<$: 10 detectors for $2\theta<2\theta_{100}$, (ii) D_0: 3 detectors with 2θ centered around the Bragg peak, (iii) $D_>$: 3 detectors with $2\theta>2\theta_{100}$

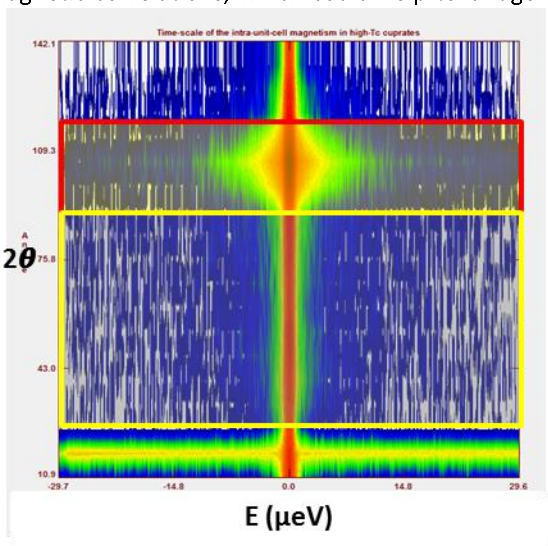

Fig. 1 *Quasi-elastic map obtained for (100) at 250K. The red rectangle delimits the 3 detectors corresponding to (100). The yellow rectangle delimits the region corresponding to* $2\theta_{\text{Off Bragg peak}}$

After alignment, several kinds of scans were performed:

a. Inelastic fixed window scans (IFWS) at various energy values: 2, 5 and 8 µeV. We counted 5 min per temperature for each IFWS.
b. Elastic fixed window scans (EFWS).
c. Quasi Elastic scans in the range ±30µeV: Counting 8 hours per scan for several temperatures: 2, 80, 150, 200, 250, 300K and 350 K (Fig 1).
d. Temperature dependencies on the Bragg peak (or EFWS) were also systematically performed during heating and cooling cycles.

Results

Quasi elastic signal at the (100) Bragg peak: Fig. 2.a. shows the quasi-elastic scans at the (100) Bragg peak in the energy window [-30,30] µeV, measured at 2 and 300K (with detector set D_0). Each scan is the sum over the three aforementioned detectors around (100). Both cans, at 2 K and 300 K, display the same quasi-elastic signal, made of a central peak with a characteristic energy width of 1.04 µeV (FWHM) and a quasi-elastic tail of the spread over the entire energy range [-10,10] µeV. According to polarized neutron diffraction (PND) experiments, the intensity of the IUC magnetism is 1000 times lower than the intensity of the Bragg peak.

Furthermore, the PND measurements were carried out on a cold TAS, for which the energy resolution was as PND experiments on TAS integrate could be at best tuned down to $\sim 200\mu eV$. In Fig.2.a, owing the huge intensity of the (100) nuclear Bragg reflection and its large (quasi-elastic) tail, no significant feature, that could have been ascribed to the IUC magnetism, could be observed upon heating (cooling). In order to get rid of the large elastic contribution that could mask the signal of interest, we zoomed on a selected ±[5,30] μeV. The scattered intensity was further integrated over that energy range in order to improve the statistic and enhance our threshold of detection of a tiny signal. We finally focused on the temperature dependence of that signal, looking of an anomalous behavior that could be hallmark of the IUC magnetic scattering. To highlight such a magnetic signal, one can carry out an independent measurement of the background on top of which it should develop using detector sets $\mathbf{D}_>$ and $\mathbf{D}_<$ (Fig. 1 and Fig.2 b).

The full E-integrated data are reported as a function of temperature on Fig. 2.c. The same integration was performed on the full detector set $\mathbf{D}_<$, not affected by the (100) Bragg scattering, and where, no magnetic signal is expected. This signal was used as a reference. In Fig.2.c, the sets of data corresponding to Q_{100} and $Q_{Off\ Bragg}$ <Q_{100} were shifted to match at the highest measured temperature 350K. Indeed, no IUC magnetic signal is expected at high temperature above T*, according to previous PND results. One can notice that, between 350 and 300K, the two datasets overlap fairly well. However, below T*=250K, the two datasets clearly split due to the enhancement of the quasi-elastic signal around (100). Their difference, i.e. I_{100}-$I_{Off\ Bragg\ peak}$, reveals the T-dependence that matches the one usually reported for the IUC magnetism [1] (Fig.2d).

These results suggest that the quasi-elastic signal measured for (100) is similar to the one measured by PND highlighting the possible dynamical nature of the magnetic signal as seen from μSR experiments.

Low-Q quasielastic signal off the Bragg peak: It is likely the IUC magnetic developing in the PG state takes the form of finite size magnetic domains, slowly fluctuating. At high temperature, short ranged or almost incoherent IUC magnetism could exist, as a precursor. Following that idea, we analyzed with scrutiny data corresponding to $2\boldsymbol{\theta}<2\boldsymbol{\theta}_{100}$, (detector set $\mathbf{D}_<$). Owing to the low Q resolution and as no signal was expected in this region, we first summed the quasi-elastic scans over all the detectors in terms of $2\boldsymbol{\theta}$ corresponding to Q=0.5-1.4Å^{-1}. Fig 2.b shows the resulting Q-integrated quasi elastic scans at 2 and 300K. The data exhibit a noticeable increase of the quasi-elastic intensity upon heating (Fig.2.b). Note that given the very narrow measured energy window [-30,30]μeV , this increase is beyond the Bose factor effect for phonons as it gives rise to a 50% extra intensity at 20μeV for instance, instead of 7% for simple thermal effects of the lattice. This result suggests that there exists a quasi-elastic signal at Q<Q_{100} at high temperatures, that vanishes at low-T.

For clarity, we also represent the Q–scans integrated between ±[5,30] μeV on Fig.2.d. The scans clearly show that the quasi-elastic scattering gradually decreases, upon cooling, starting from 250K (Fig 2.e). The difference between the 300K and 2K Q-scans is given on Fig 2.f and projected on (H 0 0). Indeed, as the sample is fixed to a constant $\boldsymbol{\omega}$ value, the recorded intensity on the multi-detector corresponds to Q-positions of the form (H,0,L) mixing a* and c*, except for |Q|=1.63 Å^{-1} where the response is strictly due to (100) reflection.

The figure shows the perfect agreement with the computed magnetic structure factors for a loop current (LC) pattern (Eq.1), one of the most popular interpretations of the IUC magnetism. This result suggests the existence of quasi-elastic scattering related to magnetic scattering from isolated loops at high temperature. Note that the calculated structure factor plotted on Fig.2.f is further multiplied by the form factor of oxygen.

$$|F_{loops}|^2 = \left(1 - \frac{(\sin(\pi\,\mathrm{H}))}{(\pi\,\mathrm{H})}\right)^2 / Q^2 \qquad \text{(Eq.1)}$$

Summary:

Putting together data from sections 1 & 2, two conclusions can be drawn:

1. The development of quasi elastic scattering at (100) at the onset temperature of the PG state (T*), following the usual T-dependence for the IUC-magnetism related to LCs.
2. The existence of a quasi-elastic scattering spread over Q-space, for Q<Q_{100} at high temperature, that vanishes upon cooling through T*, following the structure factor expected from LCs.

In the picture where the measured quasi-elastic signal is related to the IUC-magnetism, finite size magnetic domains with fluctuating magnetic moments would exist at high temperature, giving rise to the broad-Q measured signal at high –T. When crossing T*, the signal related to this finite size domains vanishes in favor of a signal localized in Q (at 100) as measured from PND experiments. In order to confirm such a picture, the measurement of a second YBCO sample with another level of doping is to be done in order to confirm the link between the enhancement of the quasi-elastic signal on 100 and T*.

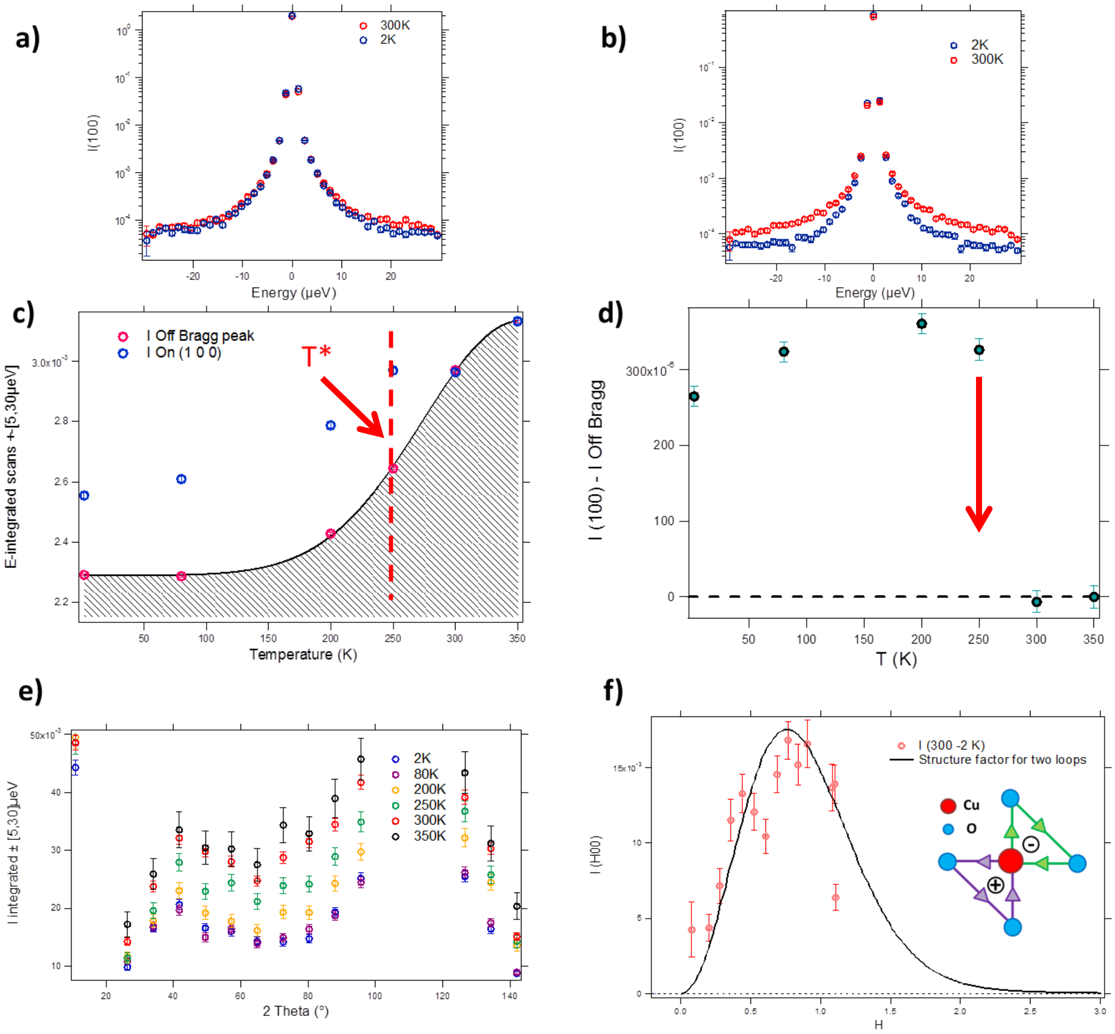

Fig.2: (a) Quasi elastic signal at (1 0 0) measured at 2 and 300K, represented in log scale for the intensity. (b) Quasi elastic scan integrated over $Q_{Off\ Bragg\ peak}$=[0.5-1.4]$\AA^{-1}$ at 2 and 300K, a clear extra signal can be seen at 300K, represented in log scale for the intensity (c) Temperature dependence of the Q_{100} and $Q_{Off\ Bragg\ peak}$ integrated in the range E=[±5,±30]. The Q_{100} data clearly shows a take-off from the low-Q data at T. The data were shifted such as to overlap for T=350K where no magnetic signal is expected. (d) Difference between I_{100} and $I_{Off\ Bragg\ peak}$ vs temperature. The figure shows the onset of quasi-elastic signal on (100) below T*. (e) Temperature dependence of 2θ (Q)-scans. The figure shows a gradual decrease of the scattered intensity off the Bragg peak starting from 250K. (f) Projection of the difference between Q scans at 300 and 2K following the structure factor of loop currents. Insert: cartoon of 2 loop currents tuning clockwise and anti-clockwise with the planar CuO_2 unit cell.*

References:

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