Experimental report

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Proposal: **4-02-542** **Council:** 10/2018

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

Research area: Physics

This proposal is a continuation of 4-02-514

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Samples: $YBa_2Cu_3O_{6.75}$

Instrument	Requested days	Allocated days	From	To
IN16B	7	5	20/09/2019	25/09/2019

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 ask for 7 days on the High Energy Resolution Backscattering Spectrometer IN16B to measure these magnetic fluctuations and determine their time-scale.

Experimental Report # 4-02-542

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

• Scientific case

The phase diagram of high temperature superconductors is dominated by a pseudo-gap (PG) phase with highly unusual physical properties [1]. Many theories attribute its origin to the proximity of a competing state, but there is a wide disagreement about the nature of this state. One theory predicted broken time-reversal and inversion symmetry due to ordered loop currents [2-3] or other similar intra-unit-cell (IUC) magnetic order [4-5]. This is consistent with five different classes of symmetry-sensitive experiments: polarized neutron diffraction (PND) [6-9], optical birefringence, dichroic ARPES, second harmonic generation [10], and polar Kerr effect. Resonant ultrasound spectroscopy measurements in $YBa_2Cu_3O_{6+x}$ provide as well strong indication that the PG state is a true symmetry breaking phase below a temperature T^* which depends on the doping as does the pseudogap (see a review in [8]). In particular, polarized neutron scattering experiments reported the appearance of an IUC magnetic order when entering the PG state. This long-range magnetic order has been reported in three different cuprate families [6-9], including $YBa_2Cu_3O_{6+x}$. This new magnetic phase could be described by the staggered orbital magnetism within the unit cell as proposed in the loop current model for the PG state [8]. This IUC magnetic order indicates that time reversal symmetry is broken in the PG state, but translation invariance is preserved. The ordering temperature matches the hole doping dependence of the PG state and is likely to vanish around a quantum critical point close to p~0.2 [2]. So far, the existence of the IUC magnetic order is well documented in a wide hole-doping range. However, other magnetic probes such as muons spin resonance (µSR) [11] and nuclear magnetic resonance (NMR) [12] experiments could not see the static local fields expected for the magnetic order. Recently, nevertheless, a µSR study reports the discovery of low frequency magnetic fluctuations [13]. Interestingly, the amplitude of the fluctuating magnetic fields corresponds to the order of magnitude deduced from polarized neutron diffraction. The quasi-static signal measured in neutron diffraction experiments (with a resolution of about 200 µeV) could then correspond to dynamical fluctuations associated with an energy scale varying in a range of the order of 10 µeV depending on the temperature according to the recent µSR results [13].

Our previous experiment on IN16B (**Exp # 4-02-514**) on a $YBa_2Cu_3O_{6.6}$ (T*=250K) sample allowed us to evidence the onset of a quasi-elastic signal at (1 0 0) when entering the PG state at T*. The temperature dependence of the measured signal recalls the one associated to the IUC magnetism measured by PND [6].

On the other hand, for H < 1, a low Q quasi-elastic signal was shown to occur at high temperature and gradually vanish upon cooling through T*. The structure factor of this signal follows the one predicted for two loop currents on opposite sides of Cu-ions within the CuO_2 planes, suggesting the existence of isolated and uncorrelated loops triggering a broad Q signal at high T.

Thus, given the high Energy resolution of IN16B, which is not achievable on a TAS, the aim of this study was to investigate a compound with different doping (and thus T*). More precisely, we performed our measurements of T-dependence of the scattered quasi-elastic intensity in a second $YBa_2Cu_3O_{6.9}$ compound close to optimal doping (with Tc≈91.5K), in order to confirm that the observed take-off at (1 0 0) in $YBa_2Cu_3O_6$ occurs systematically at T*, which could help to bridge the gap between the static and dynamical probes.

• Experimental details

Our experiment on IN16B (**Exp # 4-02-542**) was carried out on a twinned $YBa_2Cu_3O_{6.9}$ (T*~200K) sample grown by the group of Pr. X. Yao at the Shangai University. The sample was free from the parasitic "green phase" usually encountered in YBCO samples and had the following lattice parameters: 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.

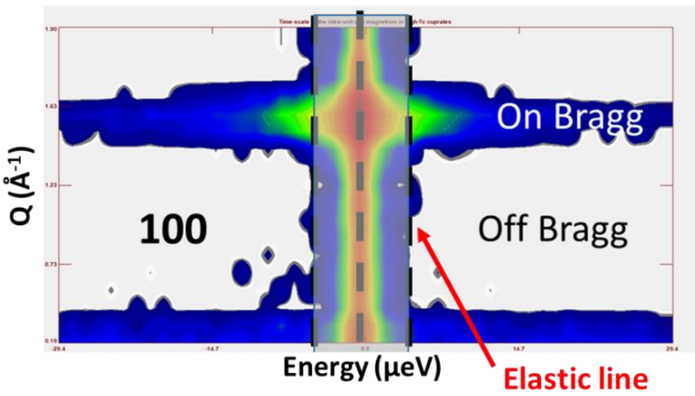

Fig. 1 Quasi-elastic map obtained for (100) at 2K. The "On Bragg" notation indicates the 3 detectors region corresponding to (100). The "Off Bragg" notation delimits the region corresponding to $2\theta_{Off\ Bragg\ peak}$

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 in our analysis, and our measurements rely on the 16 left detectors: (i) ***D<:*** 10 detectors for $2\theta<2\theta_{100}$, (ii) ***D0:*** 3 detectors with 2θ centered around the Bragg peak, (iii) ***D>:*** 3 detectors with $2\theta>2\theta_{100}$. After alignment, several kinds of scans were performed:

- **a.** Elastic fixed window scans (EFWS).
- **b.** Inelastic fixed window scans (IFWS) at 8 µeV.
- **c.** Quasi Elastic scans (QENS) in the range ±30µeV for several temperatures: 2, 50, 90, 150, 200, 250, 300K and 350 K.
- **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 (1,0,0) Bragg peak in the energy window [-30,30] µeV, measured at 2 and 350K (with the detector set $\mathbf{D_0}$). Each scan is the sum over the three aforementioned detectors around (1,0,0). The scans, at 2 K and 350 K consist in a central peak with a characteristic energy width of ~1.00 µeV (FWHM) and a quasi-elastic tail spread over the entire energy range [-30,30] µeV.

We integrated the scattered intensity in the energy range ±[5,30] µeV and checked its temperature dependence, looking at an anomalous behavior that could be hallmark of the IUC magnetic scattering. To highlight such a magnetic signal, we performed the integration for the scattered intensity in the $\mathbf{D_0}$ and **D<** regions. The full E-integrated data are reported as a function of temperature on **Fig. 2.d**.

Since the hallmark of IUC magnetism is expected to appear near the Bragg peak region ($\mathbf{D_0}$), we used the signal in the detector set **D<** as a reference.
In **Fig.2.d**, the sets of data corresponding to Q_{100} and $Q_{Off\text{- }Bragg} < Q_{100}$ overlap fairly well at the highest measured temperatures 300 and 350K. Below 300K, 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 a gradual increase of the until it saturates below 200K **(Fig.2e**).

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

Low-Q quasielastic signal off the Bragg peak: At high temperature, short ranged or almost incoherent IUC magnetism could exist, as a precursor. Following that idea, we analyzed the Q-dependence of the data corresponding to $2\theta<2\theta_{100}$, (detector set **D<**). Owing to the low Q resolution, we first summed the quasi-elastic scans over all the detectors in terms of 2θ corresponding to Q=0.5-1.4Å^{-1}. **Fig 2.b** shows the resulting Q-integrated quasi-elastic scans at 2 and 350K. The data exhibit a noticeable increase of the quasi-elastic intensity upon heating. Note that given the very narrow measured energy window [-30,30]µeV , this increase is beyond the Bose factor effect for phonons. This result suggests that there exists a quasi-elastic signal at $Q<Q_{100}$ at high temperatures, that vanishes at low-T. For comparison, the effect is even stronger than the one observed in $YBa_2Cu_3O_{6.6}$ (**Exp # 4-02-514**) shown on **Fig.2.c**.

For clarity, we also represent the Q–scans integrated between ±[5,30] µeV on **Fig.2.f**. The scans clearly show that the quasi-elastic scattering gradually decreases, upon cooling, starting from 350K. The difference between the 350K and 2K Q-scans is given on **Fig 2.g** and projected on (H 0 0). Indeed, as the sample is fixed to a constant ω 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 a peculiar form factor for the quasi-elastic scattering peaking at low-Q and that may be related to the presence of uncorrelated magnetic objects as seen in $YBa_2Cu_3O_{6.6}$.

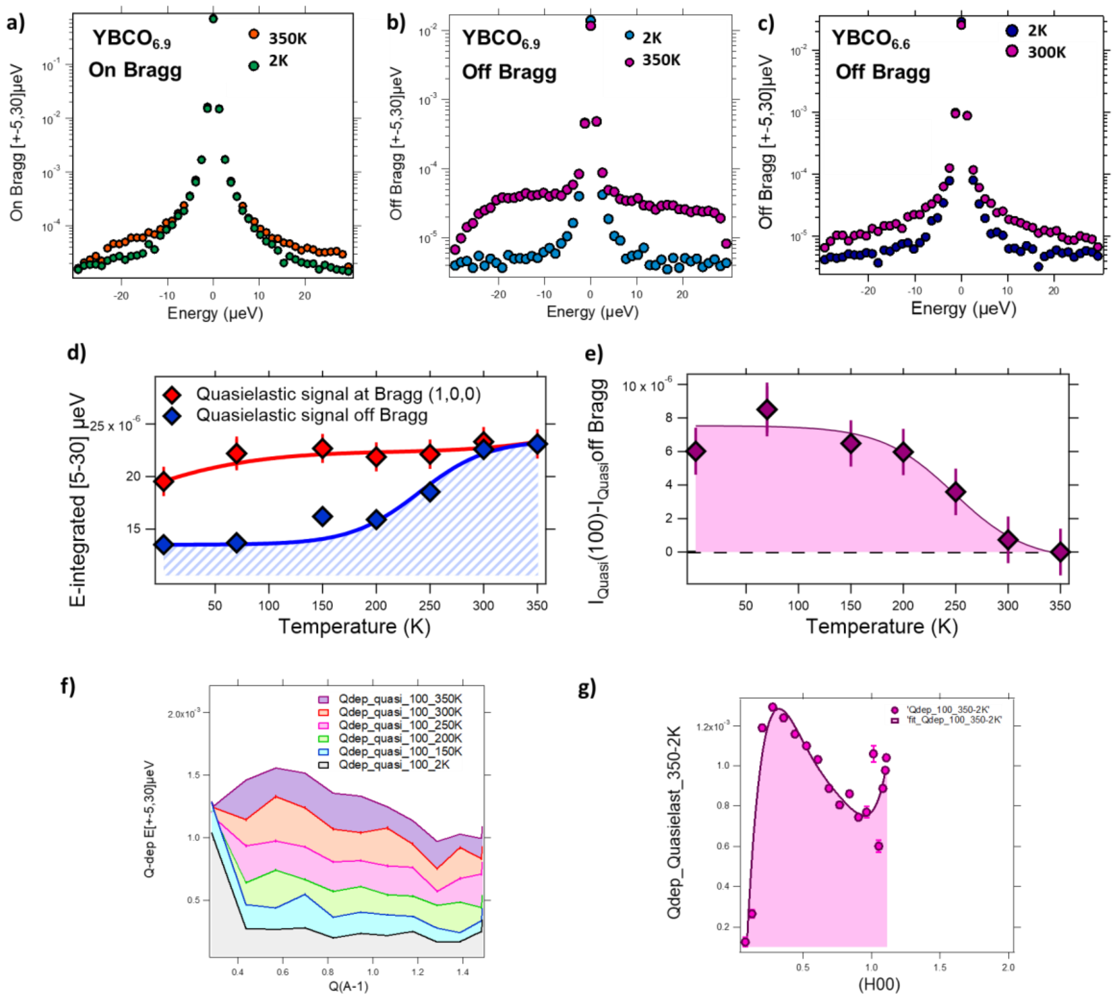

***Fig.2**: (a) Quasi elastic signal at (1 0 0) measured at 2 and 350K, represented in log scale for the intensity. Quasi elastic scan integrated over $Q_{Off\ Bragg\ peak}$=[0.5-1.4]$Å^{-1}$ (b) at 2 and 350K in $YBCO_{6.9}$ (c) at 2 and 300K in $YBCO_{6.6}$. A clear extra signal can be seen at 350K (a-b) and 300K (c). The data are represented in log scale for the intensity (d) Temperature dependence of the Q_{100} and $Q_{Off\ Bragg\ peak}$ integrated in the range E=[±5,±30]. The Q_{100} data shows a take-off from the low-Q data at T*. (e) Difference between I_{100} and $I_{Off\ Bragg\ peak}$ vs temperature. The figure shows the saturation of the quasi-elastic signal at (100) below 200K. (f) Temperature dependence of 2θ (Q)-scans. The figure shows a gradual decrease of the scattered intensity off the Bragg peak starting from 2 to 350K. (g) Projection of the difference between Q scans at 350 and 2K on (H,0,0) showing the existence of a broad-Q signal at high temperature.*

Summary:

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

1. A quasi elastic scattering at (100) that seems to saturate below 200K (~T*) as seen from the difference I_{100}-$I_{Off\ Bragg\ peak}$.
2. The existence of a quasi-elastic scattering spread over Q-space, for Q<Q_{100} at high temperature, that vanishes upon cooling.

In the picture where the measured quasi-elastic signal is related to the IUC-magnetism, finite size magnetic objects 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 gradually decreases in favor of a signal located at Q (at 100) as measured from PND experiments. In order to confirm the magnetic nature of this signal, further neutron spin-echo measurements should be carried out.

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