Experimental report

21/10/2022

Proposal: 4-02-567 **Council:** 4/2019

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

Research area: Materials

This proposal is a continuation of 4-02-514

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Samples: YBa2Cu3O6.6

Instrument	Requested days	Allocated days	From	To
IN11	9	9	01/02/2020	10/02/2020

Abstract:

The phase diagram of high temperature superconductors is dominated by a pseudo-gap phase with highly unusual physical properties. Polarized neutron scattering experiments reported the appearance of an intre-unit cell magnetism when entering the PG state in three different cuprate families. However, other magnetic probes such as muons spin resonance (uSR) and nuclear magnetic resonance experiments could not see the static local fields expected for the magnetic order. Nevertheless, a recent uSR study reports a dynamic relaxation rate in longitudinal applied field in single crystals of YBa2Cu3O6+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 Fourier time varying in the range [0.01-20]ns. Our previous Neutron Backscattering measurements on IN16B also show the existence of quasi-elastic scattering possibly associated with such fluctuations. We ask for 9 days on the Neutron Spin Echo Spectrometer IN11 to confirm the magnetic nature of this scattering and determine the time-scale of the corresponding fluctuations.

ILL 4-02-567 : Time-scale of the intra-unit-cell magnetism in high-T_C cuprates

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1/ Motivations

The phase diagram of high temperature superconductors (HTSC) 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 concerning the nature of this state. One class of theories predicts broken time-reversal and inversion symmetry, due to ordered loop currents **[2-3]** or other similar intra-unit-cell (IUC) magnetic order **[4-5]**. Such an order is accessible to 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. In addition, resonant ultrasound spectroscopy measurements in $YBa_2Cu_3O_{6+x}$ provide strong indication that the PG state is a true symmetry-breaking phase below a temperature *T**, which depends on the doping as does the PG **[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 was reported in four different 2D high-TC cuprate families **[6-9]**, including $YBa_2Cu_3O_{6+x}$, and very recently in a quasi-1D cuprate **[11]**. This new magnetic phase can be described by a staggered orbital magnetism within the unit cell, either ***i)*** as proposed in the loop current (LC) model for the PG state **[8]**, ***ii)*** as a result of the intertwining between a topological order and discrete broken symmetries **[12]**, ***iii)*** a LC order emerging from a pair density wave state (PDW) **[14]**, or ***iv)*** a fractionalized PDW instability **[13]**. This implies that time reversal symmetry is broken in the PG state and the ordering temperature matches the hole doping dependence of the PG state and is likely to vanish around a quantum critical point close to $p \approx 0.2$ **[2]**.

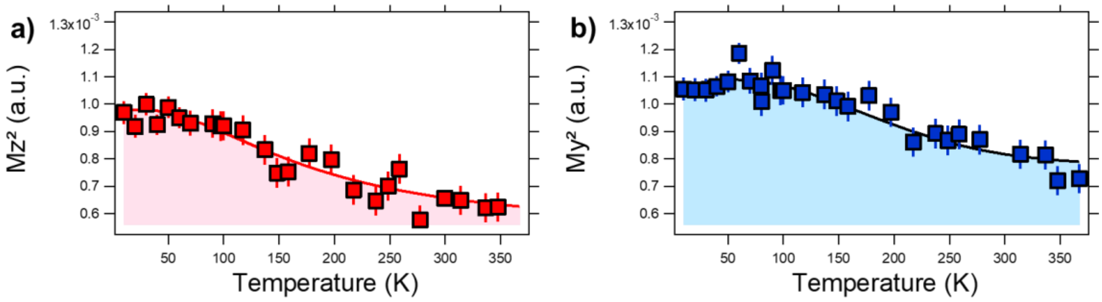
So far, the existence of the IUC magnetic order is well documented in YBa2Cu3O6+x over a broad hole doping range. However, other magnetic probes such as muon spin resonance (µSR) **[15]** and nuclear magnetic resonance (NMR) **[16]** experiments have failed to detect the static local fields expected for such a magnetic order. Nevertheless, we note that a recent µSR study has reported the discovery of low frequency magnetic fluctuations in the PG phase **[17]**. Interestingly, the amplitude of the fluctuating magnetic fields corresponds to the order of the magnitude deduced from PND. The quasi-static signal measured using PND (with a resolution of about 200 µeV) could then correspond to slow fluctuations associated with a temperature-dependent energy scale, varying in a range of ≈ 10 µeV, in agreement with µSR **[17]**. In order to check these assumptions, one needs to use advanced techniques, that are able to bridge the gap in terms of time-scale sensitivity between both kind of probes, such as neutron spin echo (NSE).

2/ Experiment

In order to access the timescale measured by µSR, we have recently performed a NSE experiment at the *IN11-C* spectrometer (Exp # 4-02-567). The study was focused on a YBa2Cu3O6.6 sample, with T^* = 250 K, and allowed us to evidence the following key properties:

1. The XYZ longitudinal polarization analysis unveils an elastic magnetic signal (*i.e.*, at a Fourier time τ = 0) at a *Q*-position of the form (1-δ,0,0) (namely, (0.71,0,0)) and, at a nearly equivalent position (0+δ,0,0) (namely, (0.24,0,0)). *The signal is found to be strictly along the a-axis and absent at equivalent Q-positions along the c-axis, in agreement with a LC like model.*
2. The in-plane and out-of-plane magnetic components could be extracted separately. Their temperature-dependence is shown in Fig. 1a,b. *Both components show a smooth enhancement upon cooling, with a quasi-saturation in the superconducting phase, i.e. below TC = 60 K.*

Interestingly, this magnetic signal appears in the same Q-range where a maximum of quasi-elastic scattering was observed using neutron backscattering on *IN16B* (**Exp # 4-02-514**). This signal is possibly related to slowly fluctuating uncorrelated loop currents domains [18]. Unfortunately, we were not able to determine the time-scale of the magnetic fluctuations on *IN11-C*, nor its evolution as a function of temperature, most likely due to the small amplitude of the associated magnetic moment. The observed enhancement of magnetic scattering with decreasing temperature could then either originate from a fully fluctuating state, or from mixed static and dynamic moment components (with further possible distinct temperature dependencies).



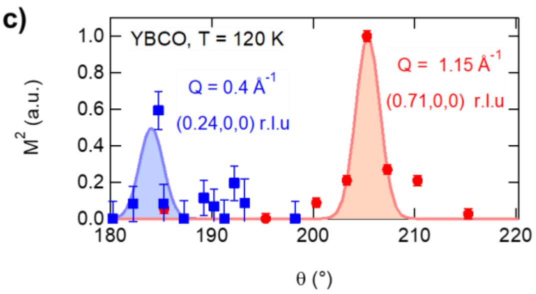


Figure 1: Temperature dependence of **(a)** the out-of-plane Mz^2 and **(b)** in-plane My^2 magnetic components as extracted from XYZ polarization analysis at τ = 0 ns. **(c)** Rocking scans around Q = (0.71,0,0) and (0.24,0,0) at T = 120 K. The data were collected on *IN11-C*.

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[18] See experimental report (**Exp #4-02-514**).