Experimental report

13/10/2022

Proposal: **4-02-588** **Council:** 4/2020

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: $YBa_2Cu_3O_{6.6}$
YBCO6.9

Instrument	Requested days	Allocated days	From	To
WASP	10	5	10/02/2021	15/02/2021

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 intra-unit cell magnetism when entering the PG state in four 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 $YBa_2Cu_3O_{6+x}$. 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 Fourier time varying in the range [0.01-20]ns. Our previous neutron spin echo experiment shows the existence of a magnetic scattering possibly associated with such fluctuations. We ask for 10 days on the Neutron Wide Angle Spin Echo Spectrometer WASP to further study this magnetism and determine the time-scale of the possibly associated fluctuations.

ILL 4-02-588 : Time-scale of the intra-unit-cell magnetism in high-T_C cuprates

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1/ Motivations

This experiment aimed at characterizing the dynamics of the so-called intra-unit-cell (iUC) order within the pseudo-gap phase of the high-T_C cuprate $YBa_2Cu_3O_{6.6}$. The iUC order was initially discovered using polarized neutron diffraction (PND) and assumed as *static* on the ps-time scale[1]. However, a recent µSR study rather suggests that it involves moments which are *slowly fluctuating* (over of a few ns time-scale) but with the same magnitude as that deduced from PND[2]. It therefore seems of prime interest to tackle this issue using Q-resolved neutron spectroscopy, and especially neutron spin echo (NSE) which offers a built-in polarization analysis capability *and* the required time-resolution. Following this idea, we have performed a first experiment on IN11C[3] but were unable to obtain a reliable estimate of the time-scale of the spin fluctuations due to the rather low beam polarization and counting statistics. This strongly motivated us to apply for a follow-up experiment on the new WASP instrument, granting access to a larger Q-range and offering a much larger flux at the sample position.

2/ Experiment

The experiment started with a check of the instrument's tuning. The resolution curve obtained on a standard TiZr sample yielded reasonable values (**Fig. 1a**). The situation was however suboptimal in the case of paramagnetic scattering by $HoTi_2O_7$, in both "nuclear" (with π-flipper on) and "magnetic" (with π-flipper off) modes (**Fig. 1b**). *Note that the latter configuration - showing polarization rates lower than 10 %- was the one we planned to use to study the dynamics of the iUC magnetism.*

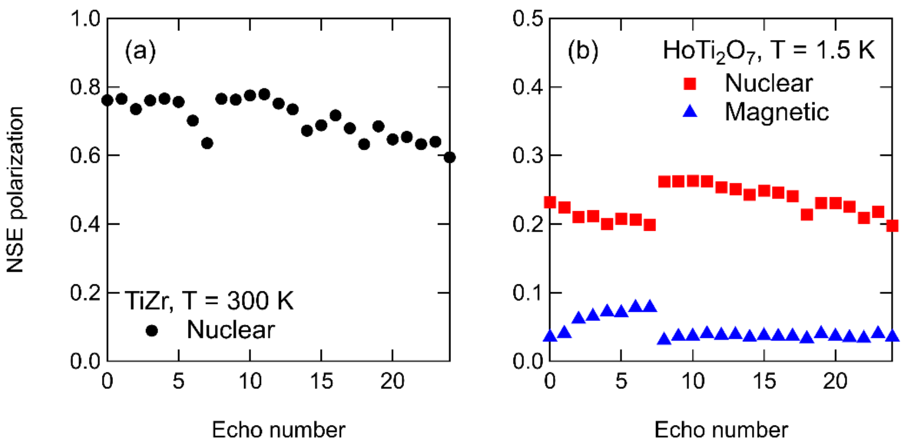

Fig. 1 – NSE resolution curves measured at the beginning of the experiment **(a)** on TiZr at T = 300 K (maximum expected value = 1) and **(b)** on $HoTi_2O_7$ at T = 1.5 K (maximum expected value = 0.5).

[1] P. Bourges *et al.*, C.R. Phys. **22**, 55, 7-31 (2021)
[2] J. Zhang *et al.*, Sci. Adv. **4**, 1, eaao5235 (2018)
[3] Experiment ILL 4-02-567.

We then searched for the Q = (1-δ, 0, 0) magnetic signal previously studied on IN11C. We have explored a broad range of neutron wavelengths (3.2 → 7.2 Å), temperatures (100 → 300 K) and sample rocking angles but systematically failed to find it. This is surprising, given that *(i)* the sample was the same as for the IN11C experiment and *(ii)* we have carefully checked its orientation within the scattering plane.

In order to avoid wasting our remaining beam time, we have then turned our attention to the Q = (1/2, 0, 1/2) signal reported by Bounoua *et al.*[4]. The latter was easily found using XYZ polarization analysis (XYZ-PA). Interestingly, WASP yields a scattered intensity ≈ 18 times larger than on the three-axis spectrometer 4F1 (Orphée reactor), where this signal was first evidenced (**Fig. 2a**). We have checked that it was not due to powder lines of an impurity phase by performing a rocking scan (**Fig. 2b**) and measured the temperature-dependence of its integrated intensity in the 100 – 340 K range (**Fig. 2c**).

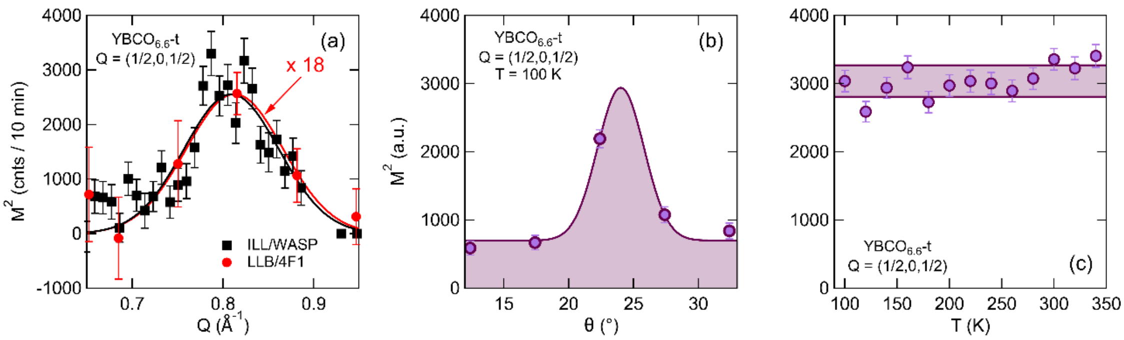

Fig. 2 – (a) Magnetic signal at Q = (1/2, 0, 1/2) obtained using XYZ-PA. **(b)** Rocking scan demonstrating its intrinsic nature. **(c)** Temperature-dependence of its integrated intensity.

Although not being the initial focus of the experiment, we have decided to study its dynamic properties. We have spent some time scanning the current of various power supplies in order to try improving the "magnetic" resolution curve (**Fig. 1b**), but our efforts turned out being unsuccessful. *It was only discovered on the last day that the power supply of the outer π/2 flipper was down (although we had not received any warning from the control software), likely explaining the low NSE polarizations.* This, however, did not affect the XYZ-PA measurements (*i.e.*, the results displayed in **Fig. 2**).

3/ Conclusions

As illustrated by this report, the experiment turned out being both frustrating *and* promising. On the one hand, we could neither find the magnetic signal at Q = (1-δ, 0, 0) (*for unknown reasons*) nor perform actual NSE measurements (*due to technical issues*). On the other hand, the rather high count rate of the Q = (1/2, 0, 1/2) magnetic signal opens very interesting perspectives for its future study by NSE, in $YBCO_{6.6}$ and similar systems.

[4] D. Bounoua *et al.*, arXiv:2111.00525v2 (2022), to appear in Communications Physics.