Experimental Report

29/03/2013

Proposal: **4-01-1196** **Council:** 4/2012

Title: Polarized-neutron study of novel magnetic excitations in underdoped high-temperature superconductor YBa2Cu3O6.6

This proposal is a new proposal

Research Area: Physics

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Samples: YBa2Cu3O6.6

Instrument	Req. Days	All. Days	From	To
IN20 CPA	10	12	14/03/2013	26/03/2013

Abstract:

The pseudogap phase in the underdoped part of the phase diagram of high-Tc cuprates continues to challenge condensed-matter physicists. A hidden magnetic order has been identified in the underdoped YBa(2)Cu(3)O(7-d) (YBCO) and HgBa(2)CuO(4+d) (Hg1201) systems. The discovery of unconventional magnetic excitations branches with almost no dispersion in underdoped Hg1201 on ILL instrumentation is the motivation for this proposal. These excitations are observed in the unexpected momentum transfer region around q~0, totally different from conventional spin fluctuations near (pi, pi) and it seems to be related to the magnetic order in the pseudogap phase. We propose to search for the counterpart of these excitations in an underdoped YBCO6.6 sample. Our previous attempt in a large sample (but slighly overdoped) did not evidence strong magnetic modes. Looking for underdoped YBCO6.6 sample is the necessary step in order to show whether these dispersionless modes in Hg1201 are a feature universal to cuprates.

Search for a new type of magnetic excitations with spin-polarized neutrons in high-temperature superconductor $YBa_2Cu_3O_{6.6}$

Scientific background Following the recent discovery of a new type of magnetic excitations in the single-CuO_2-layer high-temperature superconductor $HgBa_2CuO_{4+x}$ (Hg1201) [1], the main goal of our experiment is to search for similar excitations in the double- layer system $YBa_2Cu_3O_{6+x}$, in order to test the excitation's universality. The excitations in Hg1201 emanate from the 2D Brillouin zone center, and the signal appears below the pseudogap temperature T^*. Guided by those results, before the present experiment on IN20, we have performed an unpolarized inelastic neutron scattering measurement using a pure $YBCO_{6.6}$ sample, and observed a possible feature (Fig. 1a) from the intensity difference between energy scans taken at 4K and 230K (~T^*) at Q = (-0.05,-0.05,6.8). Our plan was to use spin-polarized neutrons to test whether this feature is magnetic and, more generally, to perform a thorough search for magnetic features far away from (pi,pi) over a wide energy range.

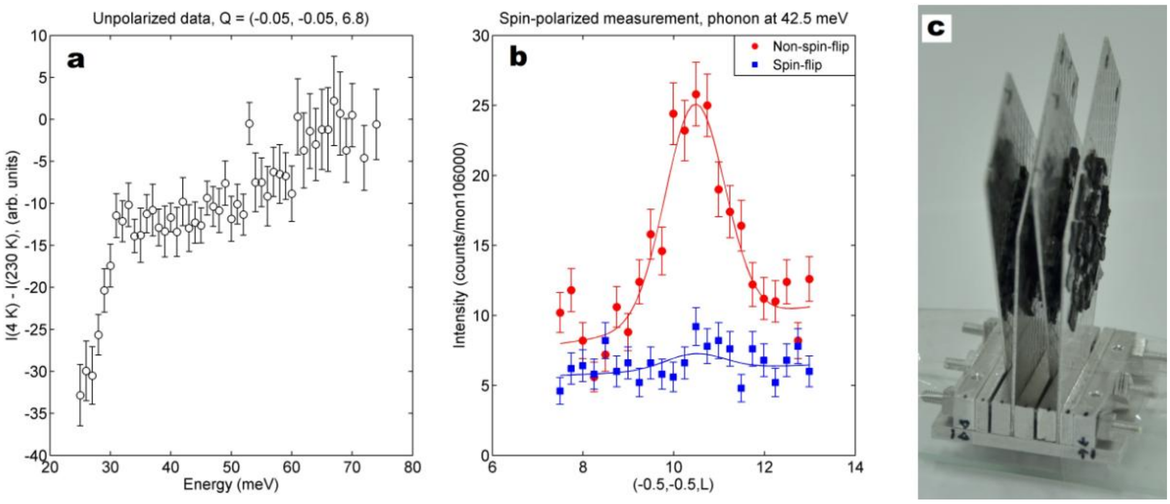

Fig. 1. (a) Intensity difference between 4 K and 230 K for energy scan at (-0.05,-0.05,6.8) measured on a detwinned (*m* ~ 2.5 g) $YBCO_{6.6}$ sample, using the unpolarized spectrometer PUMA at the FRM-II. (b) Test of flipping ratio on IN20 with our new large (*m* ~ 9.2 g) $YBCO_{6.6}$ sample using a phonon at 42.5 meV. (c) Picture of our new large sample.

Experimental setup We co-mounted more than 80 pure $YBCO_{6.6}$ single crystals to reach a total sample mass of more than 9 grams (Fig. 1). The sample was mounted in an orange cryostat inside the CryoPAD, with reciprocal-lattice vectors (H, H, L) in the horizontal scattering plane. Sample mosaic as seen from rocking scans through nuclear Bragg peaks is about 1.5 degrees. We used Heusler (111) monochromator and analyzer with $k_f = 4.1$ A^{-1} for the entire measurement. A good flipping ratio of around 11 was obtained for both elastic and inelastic configurations (Fig. 1b).

Main conclusions Figure 2 shows our main result related to the originally proposed goal. After discovering a spurion problem from the CryoPAD set up and finding a temporary solution for it (see below), we managed to take reliable polarization analysis data for the Q position (-0.05,-0.05,6.8), which was previously investigated in our unpolarized measurement. Our data indicate no detectable magnetic signal in the range of 25-65 meV, and therefore the feature in the unpolarized data (Fig. 1a) has to be due to some phonon anomaly. With our precise measurement of the odd-parity spin fluctuations at Q=(-0.5,-0.5,5.1), which was made possible by the excellent signal-to-background ratio of our sample, we can put a rather tight intensity upper bound to show that there is no strong magnetic excitation in YBCO at Q=(-0.05,-0.05,6.8). While this implies a clear difference between the magnetic excitation spectra in YBCO and Hg1201, since YBCO is a double-layer system, a thorough comparison with single-layer Hg1201 would require additional measurements in YBCO at momentum transfers along the *c*-axis that corresponds to odd parity upon exchanging the two nearest CuO_2 sheets, as well as along the *ab*-plane as has been done for Hg1201 [1]. We did not have enough time to perform such measurements because more than half of our beam time was lost due to the spurion problem which will be detailed below.

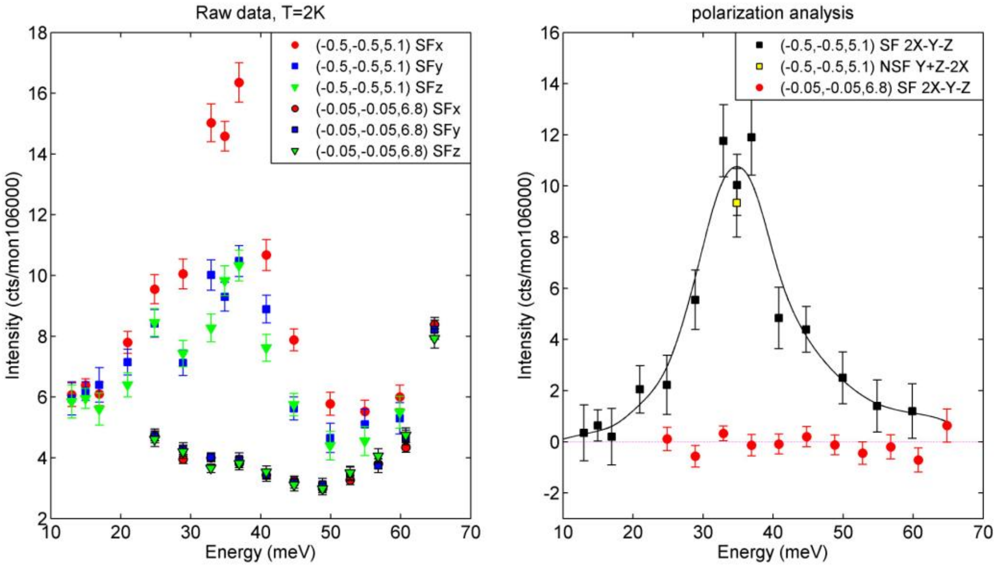

Fig. 2. Magnetic signal extracted from spin-polarization analysis at the antiferromagnetic wave vector for L=5.1 (odd parity) and near the 2D Brillouin zone center for L=6.8 (even parity) in the superconducting state.

Spurious effect from CryoPAD spin nutator N2 Starting from the third day of our beam time, we had been misled by a spurious effect related to CryoPAD for the subsequent 7 days. Here we describe the problem in some detail, in order for the instrument team to find a permanent fix for the problem.

As our main goal was to detect magnetic signals at momentum positions far away from (pi,pi), where no magnetic signal is generally expected, we relied on the use of longitudinal polarization analysis, in which magnetic signals should be extractable by the intensity combination 2X-Y-Z from the spin-flip channels. As shown in the right panel of Fig. 3, our initial data (black squares) taken in this way indicated the presence of a positive feature in the 45-60 meV range. The width of the feature is not much broader than the instrument's energy resolution in this energy range, and we even observed some variation in the intensity between 2K and 300K. All of these seemed to suggest that there is a $\mathbf{q}_{2D}$=0 magnetic signal at energies similar to those of the new excitations in Hg1201.

However, after further testing the feature at several additional **Q** positions (e.g., (0.3,0.3,6.8), (0.7,0.7,6.8), (0,0,10.2) etc.) and also performing polarization analysis in the non-spin-flip channels (each of these tests took more than 10 hours to obtain sufficient statistics), we gradually realized that the feature could be related to a spurious effect from the instrument. A closer inspection of the instrument confirmed the spurious origin of the effect: we found that when the A4 angle is in the range between 27 and 36 degrees, and in particular when one measures in the X polarization (p1=p2=(1,0,0)), a cable connecting the second nutator N2 buckles, and that part of it enters the direct beam, as shown in Fig. 3, left panel. This produced strong diffuse scattering in all directions, and some of the scattered neutrons found their way to the detector. Since the buckling was strongest in the X channel, and because only for certain A4 range the cable was best illuminated by the direct beam, it made our 2X-Y-Z analysis positive for the 45-60meV range in the energy scan. The temperature dependence of intensity may be explained by the fact that the movement of the cable is not 100% reproducible - the measurement performed at 2K might have encountered a more serious contamination than then one at 300K, leading to a peak in the intensity difference. After we installed additional (temporary) B_4C shielding to hide the buckled cable from the direct beam, the positive feature in the 2X-Y-Z analysis disappeared (Fig. 3, right panel, red circles).

The presence of the contamination was in principle detectable by the M2 monitor, which also showed a peak in the above-mentioned energy range (Fig. 3 right panel, grey diamonds). In fact, we noticed the unusual M2 reading shortly after we started to see the effect in our polarization analysis. Together with our local contact Elisa Wheeler, we inspected our data and the instrument for possible contaminations. Unfortunately, when we did so, we were mainly focusing on spurious effects that can typically occur on a 3-axis spectrometer, such as incoherent analyzer scattering, powder lines from sample and/or holder etc., and when we checked the instrument configuration, our scan ended with the Z polarization, where the cable was not strongly buckled. Coincidently, we also found the same M2 anomaly in our energy scan at (-0.5,-0.5,5.1), where we knew there is a genuine magnetic signal. The fact that M2 reading was unusual there as well made us think that the reading of M2 was over-sensitive to the neutron intensity coming from the sample, and hence might not necessarily indicate a problem. All of these delayed our discovery of the contamination until the last 36 hours of our beam time. In total, we lost more than 6 days of beam time, both for trying to get very high statistics to confirm the effect in our polarization analysis data, and for performing the various tests to eventually prove that the effect was spurious.

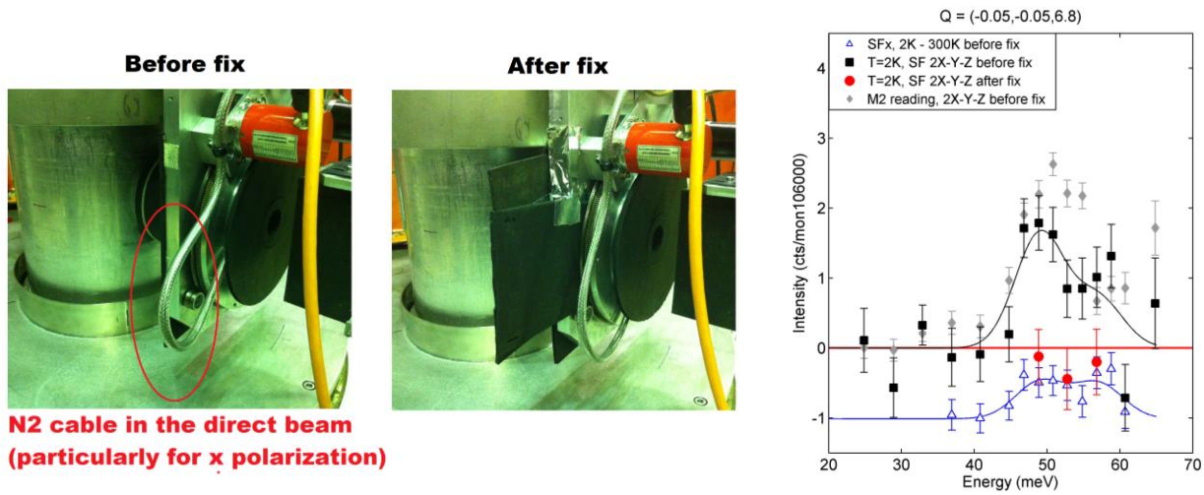

Fig. 3. Pictures showing that a buckled part of the N2 cable enters the direct beam and how our fixed the problem. The contamination leads to a peak in the spin-flip 2X-Y-Z analysis, which disappears after the cable is hidden from the beam.

References:

[1] Y. Li *et al.*, *Nature* **468**, 283 (2010).