Experimental Report

07/02/2014

Proposal: **4-01-1165** **Council:** 10/2011

Title: Study of the high-energy antiferromagnetic excitations in the under-doped high-temperature superconductor $HgBa_2CuO_{4+d}$

This proposal is a new proposal

Research Area: Physics

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Samples: $HgBa_2CuO_4$

Instrument	Req. Days	All. Days	From	To
IN1	8	8	30/11/2012	08/12/2012

Abstract:

One of the central issues in high-temperature superconductivity is to clarify the interplay between magnetism and superconductivity. Considerable work has been directed towards understanding the magnetic resonance and the seemingly universal hourglass-like magnetic dispersion near the antiferromagnetic (AF) wave-vector. The upper part of the hourglass is typically interpreted as due to AF correlations that persist from the undoped parent compounds up to optimal doping. This observation inspired numerous theoretical interpretations. Our ability to grow large Hg1201 single crystals has allowed us to study various magnetic excitations in the model-cuprate including the resonance, and the novel pseudogap state magnetic excitations. High energy AF correlations has never been studied in Hg1201 and the next-nearest-neighbor superexchange is unknown. Since Hg1201 has the highest TC of the single-layer cuprates, it is highly desirable to study AF correlations in this material. We here propose to take advantage of the high flux and hot beam available at IN1 in order to measure the high energy magnons, determine their dispersion, and extract the superexchange coefficient in underdoped Hg1201.

Our most recent inelastic neutron scattering results demonstrate the existence of novel magnetic excitations [1] that are likely related to the $\boldsymbol{q}=0$ pseudogap magnetic order [2-3]. Measurements of both under-doped ($T_c \approx 65\ K$; UD65) and nearly-optimally-doped ($T_c \approx 95\ K$; OP95) $HgBa_2CuO_{4+\delta}$ (Hg1201) reveal two weakly dispersive magnetic excitation branches throughout the entire Brillouin zone below T^* [7-8] (#4-01-887). Polarized neutron scattering measurements on IN20 proved the magnetic nature of both branches [1] (#4-01-817, #4-01-888, and #4-01-1005). Furthermore, the strength of the excitations is found to gradually decrease with increasing in-plane momentum transfer consistent with a magnetic form factor. However, our prior work, partly performed on IN8 (#4-01-951), showed that the dependence of the high-energy mode on *c*-axis momentum transfer is non-monotonic and hence cannot be explained by a conventional magnetic form factor. The goal of this new experiment was to confirm this unusual L dependence of the high-energy mode and to utilize the higher-energy neutrons at IN1 to explore the behavior at large momentum transfers.

The experiment was performed during 8 days. Approximately half a day was used to set up the spectrometer, and half a day was lost due to an unknown software error. We used a double-focusing Cu220 monochromator and vertical focusing Cu200 analyzer to increase energy resolution while maintaining sufficient flux. Measurements were performed on our nearly-optimally-doped Hg1201 sample (OP95) sample with increased sample mass (2.5 g).

Figure 1 shows both the previous work on IN8 and our new results on IN1. The high-energy mode is best measured by taking the temperature difference between energy scans at $T = 4K$ ($<< T^*$) and $T = 230\ K$ ($\approx T^*$), as shown in Fig. 1a. Figure 1c shows the L-dependence of the net intensity determined with three-point energy scans and from fitting the full energy scans at $\boldsymbol{Q} = (0,0,L)$ and $\boldsymbol{Q} = (1.3,1.3,L)$. The intensity of the high-energy mode is zero for $L = 0$, increases to a maximum intensity at $L \approx 8$, and decrease again at larger L (Figure 1). In order to check if the intensity of the mode recovers at even larger L, we performed similar scans with fixed energy k_f = 7.5 Å^{-1}. As shown in Fig. 1b, there is no apparent signal at $L = 20$.

During the remainder of the beamtime, we employed a more efficient technique of mapping out the $\boldsymbol{Q}$ dependence of the high energy-mode. Figure 2a shows temperature difference 180^o rocking scans centered around $\boldsymbol{Q} = (0,0,8.2)$ at the peak intensity (54 meV) and at the 'background' energies (63 and 46 meV). For these rocking scans, the end points correspond to $\boldsymbol{Q} = (+/-2.37,+/-2.37,0)$. The 54 meV rocking scan shows clear intensity enhancement for $L \neq 0$, while the 'background' scan is approximately flat. By taking advantage of the fact that the intensity of the mode is zero for $L = 0$, the net mode intensity of each point on the rocking scan can be determined by subtracting the average $L = 0$ value from it. We have found this method to be fully consistent with the results of Fig. 1, and a very efficient means of mapping out the $\boldsymbol{Q}$-dependence of the high-energy mode, as shown in Fig.2c. The global intensity map confirms and extends the results in Fig. 1.

A conventional magnetic form factor decreases with increasing Q, while the structure factor for phonons increases with increasing $\boldsymbol{Q}$. The non-monotonic L dependence is therefore highly unusual. We note that the sample was aligned in the (H,H,L) zone. The fact that the intensity is zero for $L = 0$ is also unusual for a magnetic excitation, since measurements at

(H,H,0) should be sensitive to magnetic fluctuations in both the *ab* plane and the along the *c*-axis. Based on these data, we conclude that the excitation exhibits a highly non-trivial magnetic structure factor and/or involves some admixture of lattice degrees of freedom. Considering our preliminary results with polarized neutrons on IN20 shown in Fig 1c (yellow triangles), the latter unconventional possibility appears likely and will be tested with spin-polarized neutrons in an upcoming experiment on IN20.

Intriguingly, we observe an almost opposite behavior for the low-energy mode. As shown in Fig. 2c (#4-01-951), the low-energy mode is strongest when ***Q*** has a zero or small *c*-axis component, and weaker when ***Q****//c*. The ***Q***-dependence of the low-energy mode is less well studied because unpolarized neutron measurements are typically obscured by large phonon contributions in the energy range in question (25-35 meV) ,and a significant effort is required to find a 'clean' ***Q*** position to measure at. A definite determination of the *Q*-dependence of the low energy mode will require a dedicated experiment with polarized neutrons.

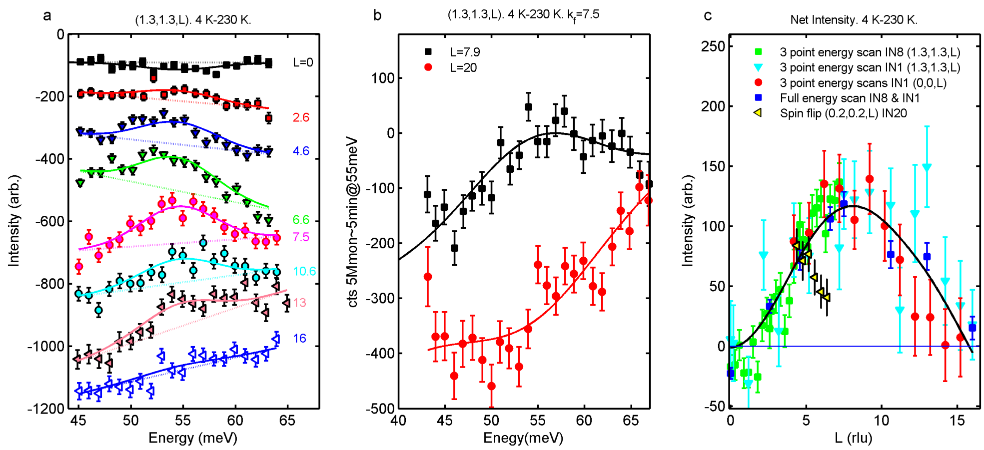

Figure 1: *L* dependence of the high-energy pseudogap excitation. (a) Intensity difference between energy scans taken at $T = 4\ K$ ($<< T^*$) and $T = 230\ K$ ($\approx T^*$) reveal the high-energy excitation for various L. $L = 0$ to $L=6.6$ were measured previously on IN8 (#4-01-951). $L=7.5$ to $L=16$ were measured on IN1 during this experiment, with $k_f = 5.67$ Å^{-1}. The data obtained at different spectrometers are suitably normalized for comparison on the same scale. (b) Intensity difference between energy scans with the larger value $k_f = 7.5$ Å^{-1}, in order to access larger L. (c) L dependence of the net intensity of the high-energy excitation. Green and cayan symbols: determined by subtracting the average of scans along *(1.3,1.3,L)* at two background energies (46 meV and 63 meV) from a scan at the peak energy (54 meV). Red symbols: the net intensity along *(0,0,L)* is determined in a similar fashion. Blue symbols: fitted peak intensities from full energy scans shown in (a). Yellow points are preliminary spin-flip data taken at 53 meV on IN20 with polarized neutrons.

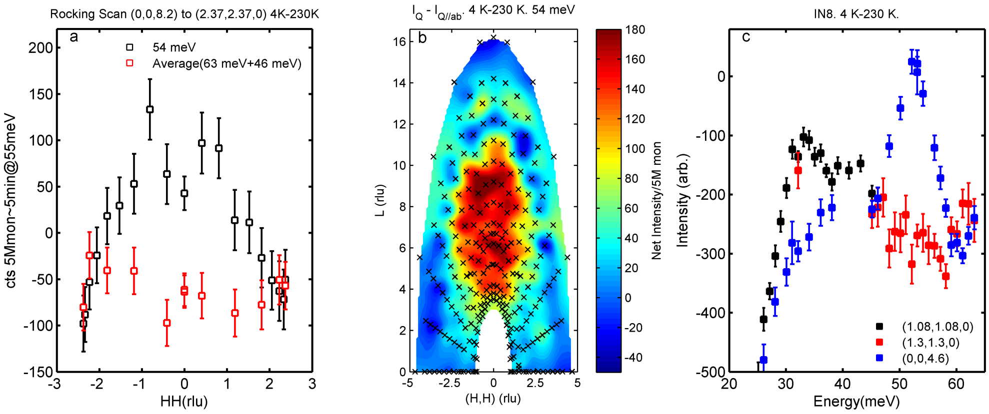

Figure 2: Net intensity of the high-energy excitation determined from rocking scans. (a) Intensity difference between *4 K* and *230 K* of 180° rocking scans about $\boldsymbol{Q}$ = *(0,0,8.2)* at 54 meV and the average of the background at 46 and 63 meV. (b) Net intensity map of the high-energy mode throughout $\boldsymbol{Q}$-space determined as described in the text. Black crosses indicate positions at which measurements were made. (c) Intensity difference between 4 K and 230 K at the indicated $\boldsymbol{Q}$ positions contrasting the behavior of the low- and high-energy modes with respect to the direction of Q along the *c*-axis or in the *ab*-plane.

[1] Y. Li *et al.*, Nature **468** 283 (2010); Y. Li *et al.*, Nature Phys. **8**, 404 (2012).
[2] B. Fauqué *et al.*, Phys. Rev. Lett. **96**, 197001 (2006).
[3] Y. Li *et al.*, Nature **455**, 372 (2008); Y. Li *et al.*, Phys. Rev. B **84**, 224508 (2011).