Experimental Report

06/02/2014

Proposal:	**4-01-1142**	**Council:**	10/2011	

Title: Magnetic and non-magnetic contributions to the novel pseudogap excitations in the high-Tc superconductorHgBa2CuO4+d

This proposal is a new proposal

Research Area: Physics

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Samples: HgBa2CuO4

Instrument	Req. Days	All. Days	From	To
IN20	14	8	19/11/2012	27/11/2012

Abstract:

Our recent inelastic neutron scattering (INS) studies of the model high-Tc superconductor HgBa2CuO4+d (Hg1201) led to the
observation of two novel magnetic excitation branches that are different from the universal hourglass-like dispersion in other hole-doped cuprates (ILL news 17.12.2010: High-Temperature Superconductors reveal part of their mystery!). These excitations appear below the pseudogap temperature T* and likely originate from the novel magnetic order that has recently been established to be universally present in the cuprates. Recently we have observed momentum transfer dependence of the high energy mode to be inconsistent with traditional magnetic or structural form factors. This, in combination with preliminary polarized inelastic neutron scattering results, indicate possible magnetic and structural characteristics of the novel excitations at intermediate and large momentum transfers. We propose to study the novel excitation with polarized neutrons in both the spin-flip and non-spin-flip channels in order to determine its true magnetic and structural momentum transfer dependence.

Magnetic and non-magnetic contributions to the novel pseudogap excitat ions in the high-T_c superconductor $HgBa_2CuO_{4+d}$

Following the discovery of weakly-dispersing Ising-like magnetic excitations in the pseudogap phase in nearly-optimally-doped (T_c = 95 K; T^* ≈ 210 K; sample labeled as OP95) and underdoped ($T_c = 65$ K; $T^* \approx 330$ K; sample labeled as UD65) $HgBa_2CuO_{4+x}$ (Hg1201) in the 50–60 meV range [1], a second branch of similar excitations at lower energy (25–40 meV) was observed (#4-01-887 and #4-01-951)) [2]. For longitudinal polarization analysis, it is convenient to define the coordinate system based on the relative orientations of the neutron spin polarization (**P**) at the sample, the scattering wave-vector **Q**, and the scattering plane that contains **Q**: the three orthogonal axes are defined by **P**||**Q** and **P**⊥**Q** in the scattering plane, and **P**||**z**, where **z** is the direction perpendicular to the scattering plane. In OP95, the intensity difference between low and high temperature, measured in the spin-flip (SF) with **P**||**Q** scattering geometry, shows a peak at ≈ 54 meV near the 2D zone center consistent with the unpolarized results (Fig. 1). Since no prominent nuclear scattering feature is observed in the non-spin-flip geometry (NSF) channel, the experiment's high flipping ratio of about 10 ensured that the observed spin-flip (SF) signal is not due to polarization leakage. Polarization analysis with SF measurements in the 3 spin geometries defined above was consistent with the presence of magnetic scattering, albeit with lower statistics[1,2]. Curiously, with unpolarized neutrons we have found that the intensity of the high-energy mode intensity is highly non-monotonic in the c-axis momentum transfer L (see Fig. 1). It increases with increasing L, as demonstrated by the full energy scans measured at IN8 (report #4-01-887) in Fig. 1a, peaks at L~6.5, and decreases again at larger L. This behavior cannot be easily explained by a magnetic form factor. This raises the distinct possibility that at intermediate and larger momentum transfers both magnetic and non-magnetic scattering contribute to the pseudogap excitations observed with unpolarized neutrons. The goal of the present experiment was to disentangle magnetic from structural contributions to the scattering.

The sample measured was OP95, with an increased mass (2.5g). Figure 2a shows energy scans at **Q** = (.05,.05,-8.7) in both the SF and NSF channel with P||Q showing the peak at 54.5 meV, which is the higher energy Ising-like excitation. Interestingly the same peak is observed in both SF and NSF channels with the relative fitted intensity $I^{SF}_{P||Q}$=1.2±0.2 counts and $I^{NSF}_{P||Q}$=0.7±0.2 counts. The flipping ratio was approximately 13, so the peak in $I^{SF}_{P||Q}$ cannot be due to leakage from a coherent structural contribution in the NSF channel, consistent with prior results. Fig. 2(b) shows the same scans at T=230K>T*. The peak is essentially gone, thus confirming the identification of this peak as the temperature dependent Ising-like mode previously observed[1-2]. To further ascertain the nature of this mode, we perform polarization analysis measurements on one background energy and the peak energy in both SF and NSF geometries. The result is shown in Fig. 3a and 3b. Surprisingly, the intensities of all 3 polarizations are consistent with each other. This is contrary to what is expected for magnetic scattering. Since the system is tetragonal, and the scattering geometry is such that **Q**//c*, at the peak, $I^{SF}_{P||Q}$ should exhibit twice the magnetic signal of **P**⊥**Q** and **P**||**z**. Likewise in the NSF geometry $I^{NSF}_{P||Q}$ should show no magnetic signal while $I^{NSF}_{P\perp Q}$ and $I^{NSF}_{P||z}$ should show half the magnetic signal. It is clear that the results are inconsistent with either simple coherent magnetic or structural (i.e. phononic) scattering(considering the large flipping ratio coherent structural scattering would not have been detectable in the SF channel). As a check of instrument stability and consistency, we plot $I^{SF}+I^{NSF}$ in Fig. 3c. LPA dictates that this value for all three polarization configurations have to equal each other, and they do for the peak and “background” energies, thus demonstrating the stability of our result. We additionally find that the SF scattering in all 3 spin channels is suppressed at 230K at the peak energy (crosses in Fig. 3a) indicating that the temperature dependent behavior is present regardless of neutron spin polarization.

Since magnetic scattering should be strongest at low **Q**, we attempted the polarization analysis at a much lower **Q**. However, since we were short of time, we could only attempt SF in 3 spin channels at the putative peak energy of 54.5 meV. Figure 4 shows the result, which again does not show magnetic scattering within the statistical uncertainty. Based on prior measurements, we conservatively estimate an expected intensity of 1 count in the scale used here. The baseline is indicated by the dotted line. If 1 count of pure magnetic intensity is present $I^{SF}_{\|x}$ and $I^{SF}_{\perp Q}$ should lie on the dashed line.

The current results are rather surprising. The lack of sensitivity to polarization direction and the ratio of $I^{SF}_{\|Q}$ to $I^{NSF}_{\|Q}$ seems to be most consistent with incoherent nuclear spin scattering. However, it is an odd type of scattering that seems to be dependent on **Q** and temperature. Further study to clarify the situation at low **Q** is necessary.

[1] Y. Li *et al.*, Nature **468** 283 (2010).
[2] Y. Li *et al.*, Nature Phys. **8**, 404 (2012).

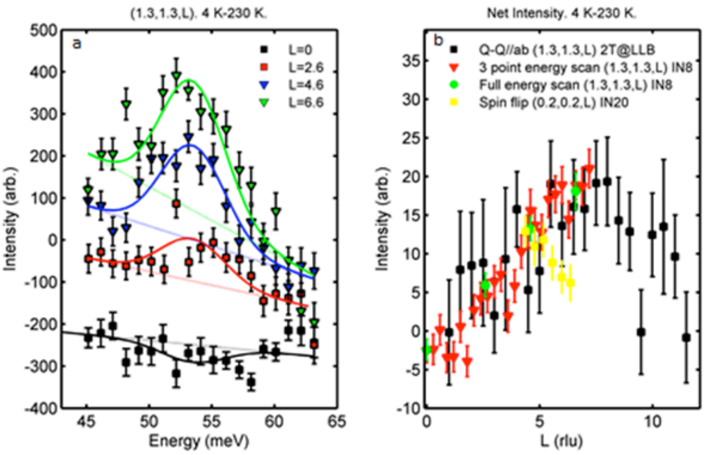

Figure 1. Prior unpolarized data (a) Intensity difference between 4 K and 230 K for OP95 showing evolution of the high-energy mode with increasing *L* (IN8). (b) Net intensity (scaled for comparison between data taken at different spectrometers) of the high-energy mode for OP95 as a function of *L* determined with three different techniques, all of which are consistent with each other. Yellow squares are obtained by taking the temperature difference of the intensity in the SF channel at the excitation peak
energy.

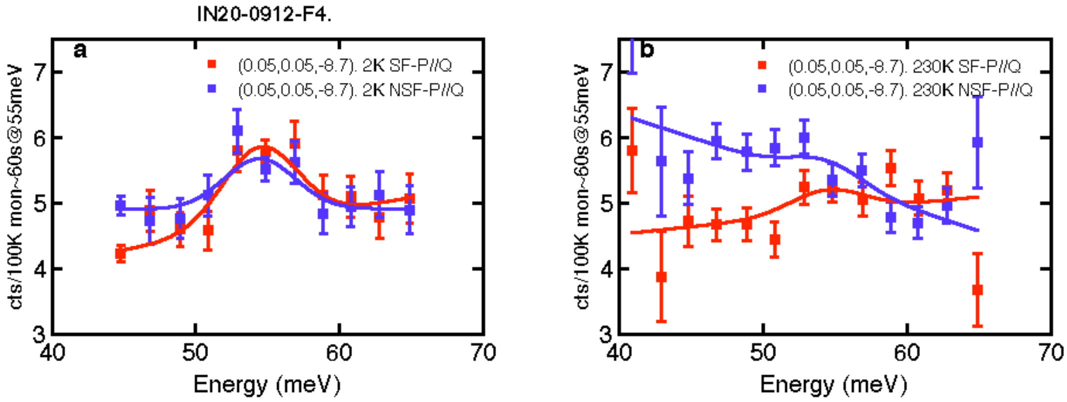

Figure 2 (a) Spin-flip (SF) and non-spin-flip (NSF) spectra at **Q** = (0.05, 0.05, -8.7) for OP95. Filled symbols are measured with the initial neutron spin polarization (**P**) parallel to **Q**, a geometry in which all magnetic fluctuations are probed in NSF and no magnetic fluctuations are probed in NSF. Lines are Gaussian fits with a linear background. T= 2K (b) The corresponding spectra at T=230 K, which is slightly higher than T*≈210 K.

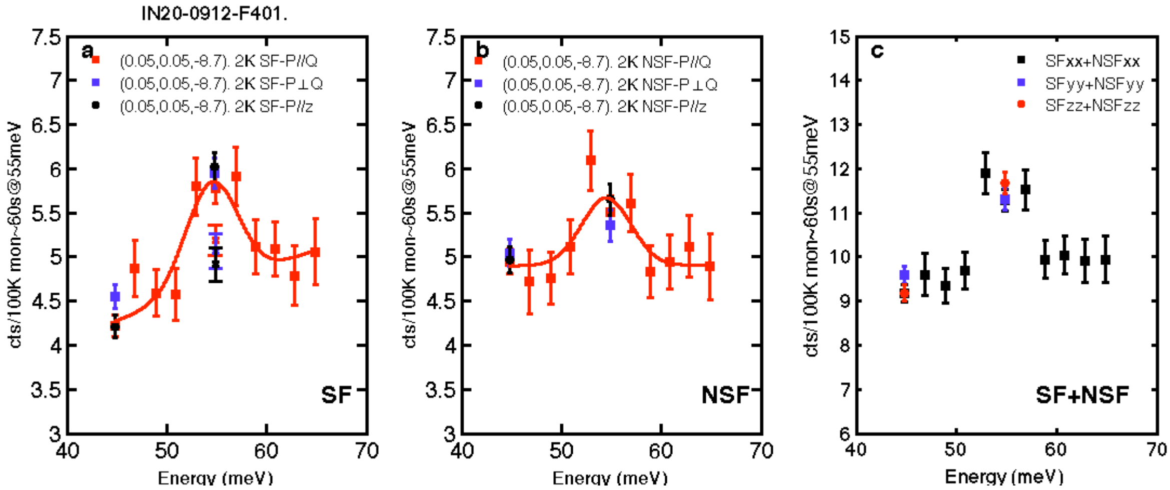

Figure 3 (a) SF spectra at 2K (solid symbols) and 230 K(crosses, not shown in legend for clarity) in all three spin polarization configurations. (b) SF spectra at 2K (solid symbols) in all three spin polarization configurations. (c) Sum of SF and NSF intensities at 2K.

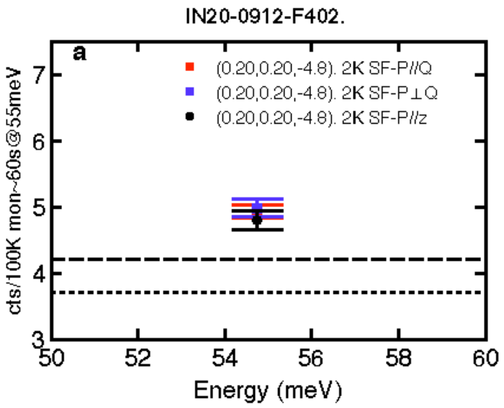

Figure 4. SF spectra at **Q** = (0.2, 0.2, -4.8) at the putative peak energy of 54.5 meV at 2K. We estimate that the signal is approximately 1 count in the scale used here, which would put the baseline at the dotted black line. If the full signal is magnetic, we would expect $I^{SF}_{F\|z}$ and $I^{SF}_{F\perp Q}$ to fall on the level indicated by the dashed line.