Magnetic resonance in the model high-temperature superconductor HgBa$_2$CuO$_{4+\delta}$

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(Dated: October 31, 2008)

We present an inelastic neutron scattering study of the structurally simple single-layer compound HgBa$_2$CuO$_{4+\delta}$ close to optimal doping ($T_c \approx 96$ K). A well-defined antiferromagnetic resonance with energy $\omega_r = 56$ meV ($\approx 6.8 k_B T_c$) is observed below the superconducting transition temperature $T_c$. The resonance mode is energy-resolution limited and exhibits an intrinsic momentum width of about 0.2 Å$^{-1}$, consistent with prior work on several other cuprates. However, the unusually large value of the mode energy implies a non-universal relationship between $\omega_r$ and $T_c$ across different families of cuprates.

PACS numbers: 74.72.Jt, 74.25.Ha,78.70.Nx

A strong piece of evidence relating magnetic excitations to the superconductivity in the high-$T_c$ cuprates comes from the analysis of inelastic neutron scattering (INS) experiments that reveal a well-defined antiferromagnetic (AF) excitation in YBa$_2$Cu$_3$O$_{6+\delta}$ (YBCO), Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ (Bi2212), and Tl$_2$Ba$_2$CuO$_{6+\delta}$ (Tl2201). Near optimal doping, this 'resonance' appears at $\omega_r = 5$-6 $k_B T_c$, and its intensity follows a power-law like behavior below $T_c$. The resonance has been associated with evidence of a bosonic coupling with the charge degrees of freedom: photoemission reveals a dispersion anomaly in the single-particle excitations and optical conductivity indicates the existence of a bosonic mode in the optical self-energy below $T_c$, both observed near the energy of the resonance.

There exist two classes of theoretical models for the resonance. Most theoretical approaches consider it a collective spin excitation in the presence of strong electronic correlations. These spin exciton models interpret the resonance as a $S = 1$ bound state below the threshold of the electron-hole excitation continuum. They give a good description of the unusual 'hour-glass' dispersion, centered at the energy $\omega_r$ and wavevector $Q_{AF} = (1/2,1/2)$, that was first established for YBCO and Tl2201. A second type of theoretical approach, motivated by an INS study that revealed an hour-glass dispersion in stripe-ordered La$_{1.875}$Ba$_{0.125}$CuO$_{4+\delta}$ (Hg1201), considers localized spin models that treat the resonance as a magnon-like excitation in a magnetically-ordered phase. In these models, the dispersion results from the spin-waves emerging from spin stripes and from one-dimensional triplet excitations within spin-ladders.

Detailed studies of the resonance have so far been limited to the structurally complicated bilayer compounds YBCO and Bi2212, for which the large single crystals required for state-of-the-art INS experiments have been available. The bilayer structure, with two closely spaced CuO$_2$ layers per unit cell, splits the resonance into two modes with odd and even symmetry under the exchange of the two layers, complicating the interpretation of the data. Results for single-layer compounds are potentially easier to interpret. Although no direct evidence for a resonance has been observed in La$_{2-x}$Sr$_x$CuO$_4$ (LSCO), the result for Tl2201 suggests that the resonance is a universal property of the hole-doped systems, regardless of the number of CuO$_2$ layers per unit cell. Unfortunately, for Tl2201 it has not been possible to reach the interesting underdoped regime and to prepare larger crystals, which has prevented a systematic INS study. It would therefore be highly desirable to investigate a single-layer system that (1) is structurally simple, (2) exhibits a high $T_c$, (3) is relatively free of disorder effects, (4) allows access over a wide range of doping, and (5) can be grown in form of sizable crystals.

In this Letter, we present INS results for the magnetic resonance of nearly optimally-doped HgBa$_2$CuO$_{4+\delta}$ (Hg1201). Hg1201 exhibits the highest $T_c$ at optimal doping (onset $T_c = 96-97$ K) of all single-layer cuprates, possesses a simple tetragonal crystal structure with a large spacing between neighboring CuO$_2$ layers (Fig.
nance energy is not universally related to $T_c$. Contrary to previous suggestions [31, 32, 33, 34], the resonance is rather large, and it has been suggested that for YBCO, Bi2212 and Tl2212 near optimal doping. This observation demonstrates that, contrary to previous suggestions [31, 32, 33, 34], the resonance energy is not universally related to $T_c$ across different families of materials.

The experiment was conducted on the thermal triple-axis spectrometer PUMA at the FRM-II reactor in Garching, Germany. The sample consisted of 24 co-mounted crystals with a total mass of about 1 g. The growth involved a two-step flux method which yields underdoped samples [28]. To achieve optimal doping, the crystals were subsequently annealed for about two months in air at 350°C to increase the oxygen concentration in the Hg layers, and hence the hole concentration in the Cu-O layers. The $T_c$ of individual crystals was determined from DC susceptibility measurements [Fig. 1(c)], and the entire sample had an average onset transition of about 96 K. Prior to the experiment, the crystals were co-mounted on three aluminum plates and aligned using Laue X-ray diffraction [Fig. 1(b)]. The aluminum plates were subsequently co-aligned in the neutron beam such that the (110) and (001) nuclear reflections were within the horizontal scattering plane (room temperature lattice constants: $a, b = 3.85$ Å and $c = 9.5$ Å). A mosaic scan at the (004) reflection indicated that the [001] crystallographic axes were aligned within 1.4° [Fig. 1(d)]. The experiment involved pyrolytic graphite (002) reflections for the double-focusing monochromator and analyzer, a fixed final neutron energy of 30.5 meV, and no horizontal collimations. This configuration resulted in a momentum resolution of 0.1 r.l.u. and an energy resolution of 8 meV at energy transfers in the 50-60 meV range.

Figure 2(a) shows energy scans at 4 K and 292 K at the two-dimensional AF Brillouin zone center $Q_{AF} = (0.5, 0.5, 5.3)$ and at a background position, $Q_{BG} = (0.8, 0.8, 4.8)$. The latter was chosen so that $|Q_{BG}| = |Q_{AF}|$ to minimize changes in phonon contributions, which have a relatively strong $Q$-dependence. The main background contributions are then largely removed by taking the difference of intensities at these two wavevectors. Figure 2(b) reveals that the background-subtracted response near 56 meV is enhanced at low temperature. The underlying change upon cooling into the SC state is more clearly evident from Fig. 2(c), which shows the double difference of the scattering intensity and reveals a resolution-limited peak at 56(1) meV. This intensity enhancement at low temperature and $Q = Q_{AF}$ is the magnetic resonance.

Further evidence for a well-defined excitation at $Q = Q_{AF}$ and $\omega_r = 56$ meV comes from constant-$Q$ scans such as those at 48 and 56 meV shown in Fig. 3(a). These ‘rocking’ scans are scans in $Q$-space through the two-dimensional AF zone center at fixed momentum transfer $|Q| = |Q_{AF}|$, which minimizes the slope of the background scattering. As can be seen from Fig. 3(c), the amplitude obtained from Gaussian fits to such scans agrees well with the double-difference obtained from energy scans. The latter effectively removes phonon contributions as well as any additional weakly temperature-dependent magnetic contributions. We note that the thermal (Bose) population factor for $\omega = 56$ meV is only 1.12 at $T = 292$ K and can thus be neglected in the above analysis.

For Bi2212, the energy (and momentum) width of the resonance is rather large, and it has been suggested that this may be the result of the relatively large amount of quenched disorder present in this compound [5, 6, 35]. On the other hand, as for optimally-doped YBCO [30] and Tl2201 [7], we find that the resonance in Hg1201 is energy-resolution-limited [see Fig. 2(c)], consistent with expectations that disorder effects are weak [26, 27]. As shown in Fig. 3(b), the response at $\omega_r = 56$ meV remains commensurate at higher temperatures. As for optimally-doped YBCO, the measured intensity increases quickly below $T_c$. Figure 4 summarizes the temperature dependence of the magnetic response.

![Image](https://example.com/image.png)
(a) Energy scans at $Q_{AF} = (0.5, 0.5, 5.3)$ and $Q_{BG} = (0.8, 0.8, 4.8)$, measured at 4 K and 292 K. (b) The difference between the intensities at $Q_{AF}$ and $Q_{BG}$ shown in (a) reveals a relative enhancement of scattering at 4 K. (c) The double difference reveals a well-defined (resolution-limited) enhancement of scattering at $Q_{AF}$ upon cooling into the SC state. Phonon scattering, unlike magnetic scattering due to the resonance, is expected to decrease, not increase upon cooling. The square symbols are amplitude values at the two-dimensional zone center obtained from rocking scans at 4 K [see Fig. 3(a)]. The continuous line is the result of a fit to a resolution-limited Gaussian (FWHM of 8 meV) centered at $\omega = 56(1)$ meV.

The response does not vanish completely above $T_c$. About one third of the low-temperature intensity remains in the normal state, without any further temperature dependence up to 190 K. While this is different from prior results for YBCO [4], a non-zero intensity centered at $Q = Q_{AF}$ is not unexpected. Even in the absence of the resonance for temperatures above $T_c$, the non-zero energy and momentum resolution of the experiment results in an overlap with the expected normal state magnetic excitations. Furthermore, part of the remaining signal could be due to phonons.

The response at $\omega = \omega_r$ is broader than the momentum resolution, with an intrinsic width of approximately 0.2 Å$^{-1}$ (FWHM) deep in the SC state at 4 K. This value is consistent with the prior results for optimally-doped YBCO [1, 2, 3, 4] and Tl2201 [7]. Figure 4(b) shows that the width obtained from Gaussian fits of the rocking scans does not exhibit an anomaly across $T_c$, consistent with a lack of a significant increase of damping in the normal state, in contrast to predictions based on localized spin models [37]. Nevertheless, our result is consistent with a monotonic increase of the momentum width with increasing temperature. The scattering near $Q = Q_{AF}$ may contain two distinct contributions in the SC state: (1) a broad and (nearly) temperature independent part already present in the normal state, and (2) the resonance, which is only present in the SC state.

Contrary to prior suggestions [31, 32, 33, 34], we con-
The characteristic temperature distribution of the magnetic resonance does not scale universally with the superconducting transition temperature. The present work lays the foundation for the systematic study of the doping dependence of the magnetic resonance and, more generally, of the full dynamic magnetic susceptibility in this model single-layer copper-oxide superconductor.

This work was supported by the DOE under Contract No. DE-AC02-76SF00515 and by the NSF under Grant No. DMR-0705086.

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