PHYSICAL REVIEW B **81**, 064518 (2010)

Magnetic resonance in the model high-temperature superconductor $HgBa_2CuO_{4+\delta}$

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(Received 8 September 2009; published 26 February 2010)

Inelastic neutron-scattering measurements of single-CuO_2-layer $HgBa_2CuO_{4+\delta}$ reveal an antiferromagnetic resonance with energy $\omega_r=56$ meV ($\approx 6.8k_BT_c$) below the superconducting transition temperature $T_c \approx 96$ K. The resonance is energy-resolution limited and exhibits an intrinsic momentum width of about 0.2 Å^{-1}, consistent with prior work on several other cuprates. The rather large value of ω_r is identical to the characteristic energy of the electron-boson spectral density obtained from recent optical conductivity work, consistent with the notion that the charge carriers are strongly coupled to magnetic fluctuations.

DOI: 10.1103/PhysRevB.81.064518 PACS number(s): 74.72.−h, 74.25.Ha, 78.70.Nx

I. INTRODUCTION

A strong piece of evidence relating magnetic excitations to the superconductivity in the high-T_c cuprates comes from inelastic neutron-scattering (INS) experiments that reveal a well-defined antiferromagnetic (AF) excitation in $YBa_2Cu_3O_{6+\delta}$ (YBCO), $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi2212), and $Tl_2Ba_2CuO_{6+\delta}$ (Tl2201).[1–7] Near optimal doping, this "resonance" appears at an energy $\omega_r=5-6k_BT_c$, and its intensity follows a power-law behavior below T_c. However, the relevance of the magnetic fluctuations to the superconductivity has remained unclear. Photoemission,[8] optical conductivity,[9,10] and tunneling measurements[11,12] reveal an electron-boson coupling at energies similar to that of the resonance. Unfortunately, there is no consensus from these charge-sensitive measurements on whether the bosons involved are of phononic, magnetic or of some other origin.[11–15]

Detailed studies of the resonance have so far been limited to the structurally complicated double-CuO_2-layer compounds YBCO and Bi2212, for which the large single crystals required for state-of-the-art INS experiments have been available. The double-layer 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,[16–19] complicating the interpretation of the data and the comparison with charge-sensitive measurements. Results for single-layer compounds are potentially easier to interpret. Although no direct evidence for a resonance has been observed in $La_{2-x}Sr_xCuO_4$ (LSCO),[20,21] the result for Tl2201 suggests that the resonance may be a universal property of the hole-doped systems, regardless of the number of CuO_2 layers per unit cell.[7]

In this paper we present INS results for the magnetic resonance in single-layer $HgBa_2CuO_{4+\delta}$ (Hg1201) just below optimal doping (onset $T_c \approx 96$ K). Hg1201 exhibits the highest T_c at optimal doping (onset $T_c=97$ K) of all single-layer cuprates, possesses a simple tetragonal crystal structure with a large spacing between neighboring CuO_2 layers [Fig. 1(a)], and is thought to be free of substantial disorder effects.[22,23] Crystals with a mass of up to 1.2 g (Ref. 24) and tunable

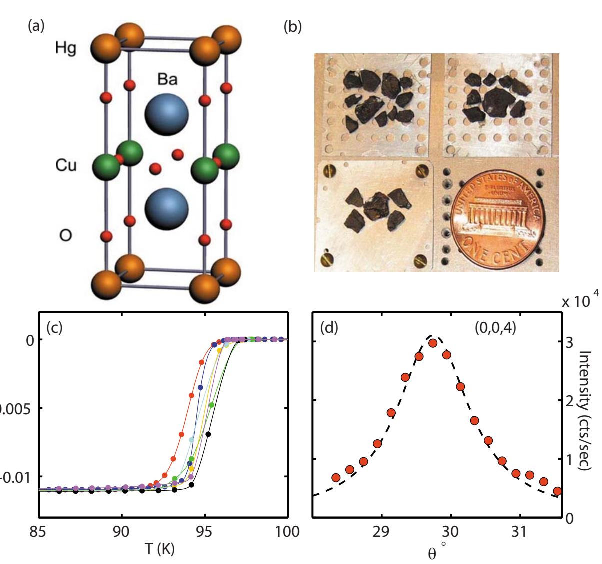

FIG. 1. (Color online) (a) Schematic tetragonal (space group $P4/mmm$) crystal structure of $HgBa_2CuO_4$ (without interstitial oxygen atom in the Hg layer). (b) Coaligned crystals. (c) Magnetic susceptibility for seven representative crystals. (d) Rocking scan through (004) Bragg peak.

over a wide doping range ($p \approx 0.08-0.22$ to date) (Ref. 25) have recently become available, enabling state-of-the-art neutron-scattering work.[26] Consequently, Hg1201 is the most promising cuprate for a quantitative experimental study of the magnetic resonance. We demonstrate the existence of the resonance, and hence provide further evidence that this magnetic mode is a universal feature, and that LSCO is not fully representative of the cuprates. The resonance in Hg1201 occurs at $\omega_r=56(2)$ meV, or $6.8(2)k_BT_c$, a value that is considerably larger than for YBCO, Bi2201, and Tl2212 near optimal doping, but that supports a universal linear relation between resonance energy and superconducting pairing gap.[27] The value of ω_r is furthermore identical to the "optical resonance" energy that has recently been found in slightly underdoped Hg1201 ($T_c=90$ K) via inversion of optical conductivity data.[10]

II. EXPERIMENTAL METHODS

The experiment was conducted on the thermal triple-axis spectrometer PUMA at the FRM-II reactor in Garching, Germany. The sample consists of 24 comounted crystals with a total mass of 1.2 g. The growth involves a two-step flux method which yields underdoped samples.[24] The crystals are subsequently annealed for about 2 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 is determined from dc susceptibility measurements [Fig. 1(c)]. Taken together, the sample is slightly underdoped with an average onset transition of approximately 96 K. The crystals are first comounted on three aluminum plates [Fig. 1(b)] and aligned using Laue x-ray diffraction. The aluminum plates are subsequently coaligned in the neutron beam such that the (110) and (001) nuclear reflections are 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 indicates that the [001] crystallographic axis is aligned within 1.4° [Fig. 1(d)]. The experiment uses pyrolytic graphite (002) for the double-focusing monochromator and analyzer, a fixed final neutron energy of 30.5 meV, and no collimations. The momentum resolution for this configuration is 0.1 r.l.u. full width at half maximum (FWHM), and the energy resolution is about 7–8 meV FWHM at energy transfers in the 50–60 meV range. Note that the thermal (Bose) population factor for $\omega=56$ meV is only 1.12 at $T=292$ K and thus has a negligible effect on the following analysis.

III. RESULTS

In Fig. 2 we reveal the existence of the magnetic resonance through the analysis of energy scans at the two-dimensional AF Brillouin-zone center $\mathbf{Q}_{AF}=(0.5,0.5,5.3)$ and at a background position $\mathbf{Q}_{BG}=(0.8,0.8,4.8)$. Note that we use $|\mathbf{Q}_{BG}|=|\mathbf{Q}_{AF}|$ to minimize changes in strongly Q-dependent multiphonon contributions; magnetic excitations in a single-layer system are expected to depend only weakly on Q_z. Raw data at 4 and 292 K are shown in Fig. 2(a); a peak is already discernible around 56 meV in the low-temperature spectrum at $\mathbf{Q}_{AF}$ and is absent in the high-temperature data. Taking the difference of these two scans reveals the sharp peak of the magnetic resonance, shown in Fig. 2(b). This panel also includes the results of performing the same subtraction on the data at $\mathbf{Q}_{BG}$. In order to remove the energy dependence seen at this background momentum and to isolate the magnetic intensity unique to $\mathbf{Q}_{AF}$, we perform a further subtraction between the data at the two momenta. The result of this double difference is shown in Fig. 2(c). We identify the resolution-limited peak at 56 meV in the double-difference data as the magnetic resonance.

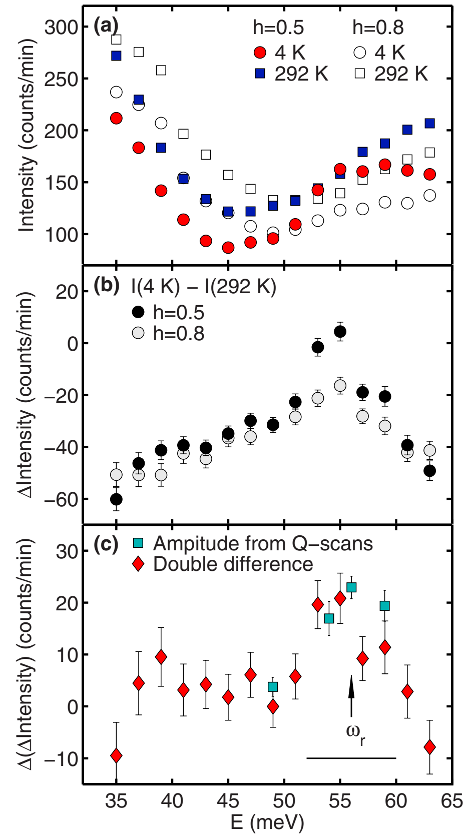

FIG. 2. (Color online) (a) Raw intensity as a function of energy at $\mathbf{Q}_{AF}=(0.5,0.5,5.3)$ (filled symbols) and $\mathbf{Q}_{BG}=(0.8,0.8,4.8)$ (open symbols), measured at 4 K (circles) and 292 K (squares). (b) Results of subtracting the 292 K intensity from the 4 K intensity for $\mathbf{Q}_{AF}$ (filled circles) and $\mathbf{Q}_{BG}$ (open circles). (c) Net intensity due to the magnetic resonance. The diamonds indicate the results of a double difference in temperature and momentum, i.e., the difference between the two data sets in (b). The squares indicate the amplitude from fitting the low-temperature momentum scans in Fig. 3(a). While the two data sets are not measurements of the same quantity, their energy dependence is similar. An analysis involving both data sets gives $\omega_r=56(2)$ meV. The horizontal bar indicates the FWHM energy resolution.

The analysis of momentum scans provides additional details of this magnetic resonance. In Fig. 3 we plot "rocking" scans in which the absolute value $|\mathbf{Q}|$ of the momentum is kept fixed. Note that these scans pass through $\mathbf{Q}_{AF}=(0.5,0.5,4)$, which has a different Q_z value from that in the energy scans. Figure 3(a) shows data at $T=4$ K for 49, 54, 56, and 59 meV. The peaks at 54 and 56 meV are sharp and

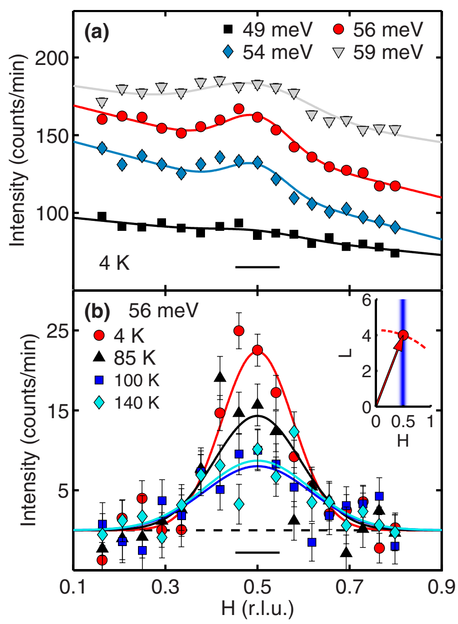

FIG. 3. (Color online) (a) Momentum scans across (0.5,0.5,4) at 4 K at energy transfers of 49, 54, 56, and 59 meV, offset for clarity. The scan trajectory is indicated in the inset. (b) Momentum scans at four temperatures at the resonance energy $\omega_r=56$ meV after subtracting a linear (sloping) background. The curves in (a) and (b) are the results of Gaussian fits. The horizontal bars indicate the FWHM momentum resolution.

strong, whereas the signal at 59 meV is weaker and broad and the signal at 49 meV is much weaker. While the extent of these data is limited, the resonance energy (where the signal amplitude reaches a maximum) seems to be the energy where the momentum width is a minimum. The energy dependence of the amplitude of these momentum scans is plotted in Fig. 2(c). While this is not the same physical quantity as the double-difference data, the two agree quite well with each other. Taking into account both sets of data we conclude that the resonance energy is $\omega_r=56(2)$ meV.

Note that in Fig. 2(c) the resonance peak is energy-resolution limited. This is consistent with the magnetic resonance in optimally doped YBCO (Ref. 28) and Tl2201.[7] In Bi2212 the energy 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.[5,6,29] The energy-resolution-limited resonance in Hg1201 is thus consistent with expectations that disorder effects are weak.[22,23,25]

Figure 4 summarizes the temperature dependence of the scattering at $\omega=\omega_r$ and $\mathbf{Q}=\mathbf{Q}_{AF}$ obtained from Gaussian fits to the momentum scans shown in Fig. 3(b). As in optimally doped YBCO, the intensity increases rapidly below T_c. Using a heuristic fit of the peak amplitude to a power-law form, we find that the onset of the enhancement occurs at 95 K, consistent with the average T_c value of our sample. The peak at $\mathbf{Q}_{AF}$ does not vanish completely above T_c: the normal-state response remains centered at $\mathbf{Q}=\mathbf{Q}_{AF}$ and is about one third of that at low temperature. In YBCO the existence of a normal-state response depends on the doping level. It is absent in optimally doped YBCO,[4] whereas a sample of

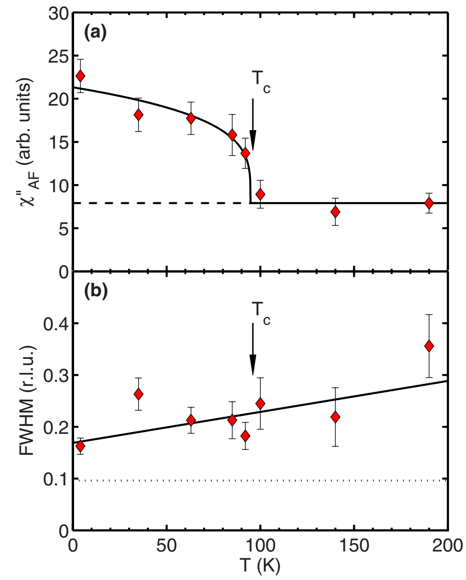

FIG. 4. (Color online) (a) Temperature dependence of the peak amplitude obtained from Gaussian fits of rocking scans [Fig. 3(b)] at $\omega_r=56$ meV. A fit to the heuristic power-law form $(T_r-T)^{2\beta}$ gives $T_r=95$ K, which is consistent with $T_c=96(1)$ K, and $\beta=0.26(7)$. (b) Temperature dependence of the momentum width (FWHM) from the same rocking scans. The width at 4 K of 0.16(2) r.l.u. corresponds to an intrinsic width of about 0.2 Å^{-1}. The solid line is the result of a linear fit, and the dotted line indicates the instrument resolution.

slightly underdoped YBCO was found to exhibit a normal-state peak.[30]

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 superconducting state at 4 K. This value is consistent with the prior results for optimally doped YBCO (Refs. 1–4) and for Tl2201.[7] Figure 4(b) shows that the width obtained from Gaussian fits of the momentum scans increases with temperature and, within the errors, does not exhibit an anomaly across T_c.

Our result for Hg1201 provides further evidence that the resonance is already present in single-layer compounds, and that the lack of this feature in LSCO may be material specific. LSCO exhibits a rather low value of T_c, in part due to the considerable quenched disorder in close proximity to the CuO_2 layers,[23] and hence may not be fully representative of the cuprates.

We note that the resonance energy in Hg1201 is significantly higher than in other hole-doped cuprates. Contrary to prior suggestions,[31–36] our findings imply that ω_r does not universally depend on T_c. In overdoped YBCO ($T_c=75$ K) (Ref. 18) and Bi2212 ($T_c=70$ K) (Ref. 19), the two mode energies were found to be equal within error: $\omega_r=34-35$ meV, or about $5.5k_BT_c$. Closer to optimal doping, the two mode energies begin to differ from each other in these double-layer compounds. Considering the average mode energy, one finds $\omega_r\approx5.8k_BT_c$ near optimal doping.[16–19] For optimally doped Tl2201 ($T_c=92.5$ K), $\omega_r=47$ meV, or $5.9k_BT_c$.[7] While these values are consistent with each other, they are considerably smaller than our result of $\omega_r = 6.8(2) k_B T_c$ for (nearly) optimally doped Hg1201.

For Hg1201, the bosonic spectrum coupled to the conduction electrons has so far only been estimated from optical spectroscopy for a somewhat underdoped sample.[10] Remarkably, the peak energy of the bosonic spectrum (56 meV) is identical to ω_r, consistent with the notion that the bosonic spectrum is of magnetic origin. Optical conductivity is an inherently momentum-integrated probe, yet the optical response could be dominated by magnetic contributions near $\mathbf{Q}_{AF}$.[35] Similar evidence for a close correspondence between charge and magnetic energy scales was previously found for Tl2201, for which analysis of optical data of a somewhat overdoped sample ($T_c = 90$ K) resulted in a characteristic energy scale of about 43 meV (Ref. 37) that compares quite well with $\omega_r = 47$ meV for an optimally doped sample ($T_c = 92.5$ K) measured with INS.[7] Results for overdoped LSCO indicate a peak at about 15 meV in the bosonic spectrum for a sample with $T_c = 31$ K,[38] which is similar to the characteristic energy of about 10 meV of the local (momentum-averaged) magnetic susceptibility for a sample with $T_c = 26$ K.[39] For the double-layer compounds YBCO and Bi2212, both the even and the odd resonance modes contribute to the local magnetic susceptibility. As a result, prior comparison of the bosonic mode energy from optical conductivity with the odd-parity resonance mode energy is less adequate.[9,40]

IV. CONCLUSIONS

In summary, we have performed a neutron-scattering study of the magnetic resonance in Hg1201 close to optimal doping. Overall, the signatures of the resonance are remarkably similar to double-layer YBCO and single-layer Tl2201, although the observed resonance energy is rather large. The latter is identical to the "optical resonance" energy obtained for Hg1201, which strongly suggests a significant coupling of the charge carriers to the magnetic fluctuations. The observed high value of ω_r / T_c has served as motivation for a separate effort that lead to the insight that the resonance energy and superconducting gap are universally related.[27] The present work lays the foundation for a systematic study of the doping dependence of the magnetic resonance and, more generally, of the full dynamic magnetic susceptibility in this model copper-oxide superconductor.

ACKNOWLEDGMENTS

We thank A. V. Chubukov for valuable comments. This work was supported by the DOE under Contract No. DE-AC02-76SF00515 and by the NSF under Grant No. DMR-0705086. N.B. acknowledges the Alexander von Humboldt Foundation.

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