Hourglass Dispersion and Resonance of Magnetic Excitations in the Superconducting State of the Single-Layer Cuprate HgBa$_2$CuO$_{4+\delta}$ Near Optimal Doping

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We use neutron scattering to study magnetic excitations near the antiferromagnetic wave vector in the underdoped single-layer cuprate HgBa$_2$CuO$_{4+\delta}$ (superconducting transition temperature $T_c \approx 88$ K, pseudogap temperature $T^* \approx 220$ K). The response is distinctly enhanced below $T^*$ and exhibits a Y-shaped dispersion in the pseudogap state, whereas the superconducting state features an X-shaped (hourglass) dispersion and a further resonance-like enhancement. A large spin gap of about 40 meV is observed in both states. This phenomenology is reminiscent of that exhibited by bilayer cuprates. The resonance spectral weight, irrespective of doping and compound, scales linearly with the putative binding energy of a spin-exciton described by an itinerant-spin formalism.

The dynamic magnetic susceptibility of the hole-doped cuprates exhibits a hourglass-shaped (or X-shaped, upon considering an energy-momentum slice through $q_{AF}$) spectrum centered at the two-dimensional antiferromagnetic (AF) wave-vector $q_{AF}$ [1–3]. The upper dispersive branch likely results from short-range AF correlations of local moments, but the cause of the downward dispersive branch, at energies below the neck of the hourglass, has remained unclear. Results for the two cuprates most widely studied via neutron scattering, (La,Nd)$_{2-x}$(Sr,Ba)$_x$CuO$_4$ (La214) and YBa$_2$Cu$_3$O$_{6+\delta}$ (Y123), support contradictory scenarios. For moderately- to overdoped Y123 (hole concentration $p \gtrsim 0.085$), the low-energy dispersion is accompanied by a magnetic resonance: an increase in scattering at $q_{AF}$ and energy $\omega_r$ [4, 5]. Both features appear in the superconducting (SC) state and can be understood, within an itinerant picture, as a dispersive spin exciton bound below the particle-hole continuum and associated with the d-wave SC gap [6, 7]. In contrast, La214 features an hourglass dispersion in both the SC and normal states, and no resonance in the SC state [2, 8, 9]. The discovery of static charge-spin ‘stripe’ order in La214 [10] motivated an interpretation in terms of fluctuating stripes [11]. Reconciliation of these discrepancies has been further complicated by the disparate crystal structures of Y123, a double-layer cuprate (two CuO$_2$ layers per primitive cell), and La214, a single-layer compound.

The observation of a magnetic resonance in single-layer Tl$_2$Ba$_2$Cu$_2$O$_{6+\delta}$ (Tl2201) [12] and HgBa$_2$CuO$_{4+\delta}$ (Hg1201) [13], which feature optimal $T_c$ values of nearly 100 K, more than twice that of La214, raised the prospect of a universal description of the magnetic response. However, detailed results have been difficult to obtain for these single-layer cuprates, and an hourglass dispersion has not been detected. Thus, a connection, or lack thereof, between the hourglass dispersion, the resonance, and superconductivity has not been universally established, rendering a satisfactory description of magnetic excitations of single- and double-layer cuprates elusive.

A recent study of underdoped Hg1201 (labeled HgUD71, $T_c = 71$ K) revealed a gapped Y-shaped spectrum both in the pseudogap (PG) and SC states, and no evidence for a resonance. The unusual response was attributed to strong competing PG order [14]. Since then, charge-density-wave (CDW) order in Hg1201 was found to be particularly pronounced at this doping level [15, 16]. Here we study a Hg1201 sample closer to optimal doping (HgUD88; $T_c = 88$ K), motivated by early work for optimally-doped Hg1201 that yielded initial evidence for a resonance [13]. First, we confirm the observation for HgUD71 [14] that the response is enhanced below $T^*$ and has a gapped, Y-shaped spectrum in the PG state. Whereas the large gap (about 40 meV) is unchanged in the SC state, the response of HgUD88 changes to a distinct hourglass topology and features a resonance-like enhancement at $\omega_r \approx 59$ meV. This is reminiscent of the phenomenology established for the bilayer cuprates [17, 18]. The characteristic resonance energy and spectral weight scale with the particle-hole Stoner continuum threshold energy in a manner consistent with results for other cuprates, and with expectations for a spin-exciton resulting from an itinerant spin formalism.

The sample, prepared following previously described procedures [14, 19, 20], consists of approximately 30 aligned single crystals with a total mass of 2.8 g with full-width-at-half-maximum (FWHM) mosaic of 1.5°. Similar to ref. [14], the value $T_c = 88$ K signifies the transition midpoint obtained by averaging uniform magnetic susceptibility data for the diamagnetic signal of the individual crystals. Measurements were performed on a time-of-flight (TOF) spectrometer [21], with the sam-
FIG. 1. (a)-(e) Constant-energy images of magnetic susceptibility at $T = 5$ K (left), 100 K (middle), and 250 K (right). Data within a 6 meV window centered at the indicated energies are averaged, except for $\omega = 23$ meV, where a 10 meV window was used. White dots (left-most panels): momentum resolution at each energy. (f)-(j) Corresponding constant-energy cuts averaged over $\{100\}$ and $\{010\}$ trajectories across $q_{AF}$. Solid lines: gaussian fits to data convolved with the momentum resolution. (k) Energy dependence of incommensurability $\delta$ at 5 K. Horizontal error bars: fit uncertainties for $\delta$. Filled black circles and open squares: data taken with incident energy $E_i = 100$ meV and 130 meV, respectively. Filled grey region: FWHM of the response. Hatched area: magnetic excitation gap. (l) Energy dependence of incommensurability $\delta$ at 100 K, with dispersion at 5 K (dotted line) shown for comparison. Horizontal black bar: experimental momentum resolution at $\omega = 40$ meV.

ple’s crystalline c-axis aligned along the incident beam, and incident neutron energies $E_i = 100$ meV (at 5 K, 100 K and 250 K) and 130 meV (5 K). The dynamic magnetic susceptibility, $\chi''(q,\omega)$, was determined from the scattering intensity, calibrated to a Vanadium standard, by normalizing by the anisotropic Cu$^{2+}$ form factor [22] and the Bose population factor. The temperature dependence was measured at $(q,\omega) = (q_{AF},60$ meV) at the Laboratoire Léon Brillouin, with the 2T triple-axes-spectrometer, with fixed final energy $E_f = 35$ meV. We quote the scattering wave-vector $Q = Ha^* + Kb^* + Lc^* \equiv (H,K,L)$ in reciprocal lattice units (r.l.u.), where $a^* = b^* = 1.62 \, \text{Å}^{-1}$ and $c^* = 0.66 \, \text{Å}^{-1}$ are the room-temperature values. Constant-$\omega$ data are fit to a gaussian, $\chi''(q,\omega) = \chi''_0 \exp\{-4ln2R/(2\kappa)^2\}$, convolved with the experimental momentum resolution, where $q$ is the reduced two-dimensional wave vector, $R = [((H-1/2)^2 + (K-1/2)^2)^{1/2} - \delta]^2$, $2\kappa$ the intrinsic FWHM momentum width, and $\delta$ the incommensurability away from $q_{AF}$; see ref. [14] for further details.

Figure 1a-j shows $\chi''(q)$ for select $\omega$ at $T = 5$, 100 and 250 K. At 5 K, the gapped spectrum evolves with increasing energy from an incommensurate ring that disperses toward $q_{AF}$ and then outward again, thus exhibiting an archetypical hourglass dispersion (Fig. 1k). At $T = 100$ K ($T_c + 12$ K), however, the low-energy response is commensurate with $q_{AF}$ (Figs. 1d,i), resulting in the Y-shaped dispersion (Fig. 1l) that is characteristic of the PG state [14, 17, 23]. Finally, at $T = 250$ K, just above $T^* \approx 220$ K [24], the response is considerably weaker than deep in the PG state.

The response at $\omega = 60$ meV (Fig. 1c,h), where the upward dispersion begins (Fig. 1g,k,l), is significantly larger at 5 K than at 100 K. This is reflected in a sharp peak at 5 K in the energy dependence of $\chi''_{AF}$ (Fig. 2a). Detailed measurement of the temperature dependence of $\chi''_{AF}$ at 60 meV (Fig. 3b) shows a distinct increase of scattering below $T^*$, consistent with the result for HgUD71 [14]. However, contrary to HgUD71 (Fig. 2a, 3b), this is followed by a further increase below $T_c$. We identify this feature below $T_c$ in HgUD88 as the resonance [4, 13, 25].

The resonance shows a distinct enhancement in mag-
FIG. 2. (a) Energy dependence of magnetic susceptibility at $q_{AF}$, $\chi''_{AF}$, determined from fits to data such as those in Fig. 1 (see text). Filled circles: $E_i = 100$ meV; the data around $\omega = 30$ meV are contaminated by phonon scattering [14] and indicated by open circles. Open squares: $E_i = 130$ meV. Solid lines: guides to the eye. Horizontal bars represent the energy binning (only shown for 5 K). A large difference in $\chi''_{loc}$ is observed across $T_c$ (see text). Filled circles: $\gamma$ and orange lines in a)

(b) Energy dependence of local susceptibility, $HgUD71$ [14] at 5 K and 85 K (grey and orange lines in a) and b)). (b) Energy dependence of local susceptibility, $\chi''_{loc}$. For both $HgUD88$ and $HgUD71$ [14], the magnetic response exhibits large gaps in the PG and SC states.

magnetic scattering below $T_c$ in optimally- and over-doped cuprates [3, 13, 26]. However, it is harder to discern in underdoped samples, which already exhibit significant magnetic scattering in the normal state [14, 23, 26], because the instrumental energy resolution is large compared to the resonance width. For $HgUD88$, where $\omega_r$ and hence the FWHM energy resolution of the triple-axis spectrometer are particularly large, the temperature dependence is considerably smoothed (Fig. 3b).

The resonance is better revealed as a peak in $\Delta \chi''_{AF} = \chi''_{AF}(5 K) - \chi''_{AF}(100 K)$ (Fig. 3a) at $\omega_r = 59(1)$ meV, with a width that is not much larger than the resolution of the TOF spectrometer (about 5 meV FWHM). The ratio $\omega_r/(k_B T_c) = 7.9$ is the largest value reported for the cuprates [27, 28]. Using $\Delta_{SC} \approx 42(2)$ meV [29, 30] for the SC gap amplitude, the ratio $\omega_r/\Delta_{SC} \approx 0.70(3)$ is consistent with the value 0.64(4) established for unconventional superconductors [28].

The present result for $HgUD88$ bears a striking resemblance to observations for bilayer Y123 [17, 23]. The hourglass dispersion, particularly the dispersive low-energy branch, is present only below $T_c$, and thus a characteristic of the SC state. Above $T_c$, both the resonance and its downward dispersive branch disappear, yielding a Y-shaped spectrum [31]. However, for $HgUD88$ the neck of the hourglass at 5 K is somewhat extended compared to other cuprates (Fig. 1f). Furthermore, the upper dispersion branch extends to slightly lower energy at 100 K than at 5 K. These subtle features, established in the TOF experiment, in combination with the coarse triple-axis energy resolution used to measure the temperature dependence, might further obscure a distinct enhancement of $\chi''_{AF}(\omega_r)$ at $T_c$ (Fig. 3b).

Considering the spectral weight of the resonance, $W_r = \int d\omega \Delta \chi''_{AF}$, we find $W_r = 0.54(4)$ $\mu_B^2$ Cu upon integrating from 51 to 64 meV. $W_r$ can be related to $\omega_r$. Within the itinerant picture, the interacting spin susceptibility is computed using the random phase approximation. In the SC state, the resonance at $q_{AF}$ is part of a spin exciton, i.e., a spin-triplet collective mode bound below the threshold of the Stoner continuum, $w_c$ [7, 32]. The resonance weight is linearly related to the reduced binding energy, $(\omega_r - \omega_c)/\omega_c$, where $V$ is the planar interaction that enhances the bare susceptibility and $g = 2$ is the Landé factor. The quantities $\omega_r$ and $\beta$ are related to the hot-spots (hs), defined as Fermi-surface points connected by $\Delta_{LS}$, where $\Delta_{LS}$ is the planar interaction that enhances the bare susceptibility and $g$ is the Landé factor.

Alternatively, the resonance has been attributed to a redistribution of spectral weight of local spin fluctuations from energies below to energies above a spin gap that appears in the SC state [11, 35]. The gap in $HgUD88$ is apparent from the lack of low-energy magnetic scattering (Figs. 1e,j). To better determine the gap size, we examine the local susceptibility, $\chi''_{loc}(\omega) = \int \chi''(Q, \omega) d^2q \int d^2q$ (integration over the AF Brillouin zone). As seen from Fig. 2b, $HgUD88$ features a particularly large gap of about 40 meV in both the PG and SC states. With increasing temperature, the strength of magnetic excitations decreases, yet the gap does not close. Consistent with the result for $HgUD71$ [14], the gap thus is a property of the PG and not the SC state [26, 35]. We thus cannot attribute the resonance to a spectral weight redistribution due to the opening of a gap. Although prior neutron scattering work yielded evidence for a “spin-pseudogap” [4, 14, 27, 36], the present result constitutes the clearest and largest manifestation of such a gap.

The spin-exciton scenario can semi-quantitatively account for (i) the magnitude of the resonance and (ii) its connection to a downward dispersing mode in the SC state of $HgUD88$. However, it fails to explain the absence of both features in $HgUD71$ [14]. It is interesting to
Our results establish that excitations across the Fermi-surface in the presence of either SC and/or PG order should be considered in accounting for the magnetic spectrum in both double- and single-layer cuprates. Recent transport measurements indicate Fermi-liquid behavior in the PG state [24, 39, 40], which adds further support for the need to pursue such formulations. However, the spin-exiton scenario can in principle only generate a single pole (below the Stoner continuum) at each \( \mathbf{Q} \), and thus this scenario cannot account for both the downward and upward dispersive branches. In addition to the pair-breaking peak in the ERS \( B_{1g} \) channel, a two-magnon peak is observed [30], which indicates the persistence of short-range local-moment AF correlations, likely associated with the upward dispersive part of the spectrum. A theoretical approach that incorporates both itinerant and local spins, such as in ref. [41], might thus be necessary. Understanding the Y-shaped spectrum and the large spin gap will likely require the consideration of the relationship between the magnetic degrees of freedom and the experimentally-detected broken symmetry states [16, 42–46] in the PG state.

It has been proposed that spin-fluctuation mediated pairing is the common thread linking a broad class of unconventional superconductors [47]. In the case of the cuprates, AF correlations have been argued to be the cause not only of the \( d \)-wave superconductivity, but also of the PG phenomena [48–50]. We have established a phenomenology of the magnetic response in the PG and SC states that is common to single- and double-layer compounds, namely a Y-shaped PG spectrum that evolves into an X-shaped (hourglass) response accompanied by a resonance in the SC state. Based on general considerations, the magnetic resonance is associated with a SC gap function that undergoes a sign change, which is naturally the case in a Fermi-liquid picture for the \( d \)-wave cuprates [28]. In the La-based cuprates, the magnetic response does not undergo a sudden change from \( X \)- to Y-shaped at \( T_c \). However, the low-energy incommensurability was found to decrease slowly with increasing temperature, e.g., from \( \delta \approx 0.12 \) at 8 K to 0.08 at 200 K in \( La_{1.875}Ba_{0.125}CuO_4 \) [51]. It is tempting to attribute this to pre-formed SC pairs, which have been argued to appear at high temperatures in the La-based cuprates [52]. However, the lack of a \( Q_{AF} \) resonance, the prominence of stripe correlations [11] and recent experiments indicating a narrow SC fluctuation range above \( T_c \) [53–55] indicate the proximity of a stripe instability in this particular cuprate family as the dominating factor determining their low-energy magnetic spectrum.

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In the normal state of Y123, the low-energy response is only commensurate along $b^*$. The response along $a^*$ is still incommensurate, albeit with an incommensurability that is smaller than in the SC state [23]. This is in contrast to the low-energy response in HgUD71 [14] and HgUD88, which is commensurate and isotropic.

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