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# Feedback Effect on High-Energy Magnetic Fluctuations in the Model High-Temperature Superconductor $\mathrm{HgBa_2CuO_{4+\delta}}$ Observed by Electronic Raman Scattering

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We use electronic Raman scattering to study the model single-layer cuprate superconductor $\mathrm{HgBa_2CuO_{4+\delta}}$. In an overdoped sample, we observe a pronounced amplitude enhancement of a high-energy peak related to two-magnon excitations in insulating cuprates upon cooling below the critical temperature $T_c$. This effect is accompanied by the appearance of the superconducting gap and a pairing peak above the gap in the Raman spectrum, and it can be understood as a hitherto-undetected feedback effect on the high-energy magnetic fluctuations due to the Cooper pairing interaction. This implies a direct involvement of the high-energy magnetic fluctuations in the pairing mechanism. All of these effects occur already above $T_c$ in two underdoped samples, demonstrating a related feedback mechanism associated with the pseudogap.

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High-temperature superconductivity in the cuprates arises from doping antiferromagnetic (AF) insulators. This has motivated intense research on the role of AF fluctuations in the mechanism of superconductivity [1]. Unlike phonons in conventional superconductors, AF excitations are generated by the same electrons that form the Cooper pairs. If such excitations act as the pairing bosons, their spectrum is hence expected to be strongly modified in the superconducting state. Such a "feedback effect" has indeed been observed by inelastic neutron scattering (INS) experiments, which have uncovered a pronounced superconductivity-induced spectral-weight redistribution of low-energy magnetic excitations into a "resonance" peak with energy 40–60 meV [2]. The magnetic resonance appears generic to superconductors near an AF instability, including the cuprates [2], the heavy-fermion compounds [3], and the iron-based superconductors [4], and its energy scales with the superconducting gap [5]. Based on these observations and on related anomalies in fermionic spectral functions, the resonance has been attributed to a feedback effect of the Cooper pairing interaction on low-energy spin fluctuations [6]. However, the spectral weight of these low-energy fluctuations appears insufficient to explain the large superconducting temperature $T_c$ in the cuprates [6]. Meanwhile, evidence from tunneling [7], photoemission [8], and optical [9] spectroscopies has indicated contributions from high-energy excitations to the pairing interaction.

Recent research has begun to explore the origin of this high-energy contribution. A strong magnetic response well above 100 meV has been found by INS in overdoped $\mathrm{La_{1.78}Sr_{0.22}CuO_4}$ [10] and by resonant inelastic x-ray scattering in various cuprates up to optimal doping [11]. These results demonstrate that high-energy fluctuations akin to magnons in the AF parent compounds are available as a possible resource for Cooper pairing deep in the superconducting regime of the phase diagram. However, it remains largely unknown whether this resource is actually utilized. To address this question, we have performed an accurate electronic Raman scattering (ERS) study of the model single-layer system $\mathrm{HgBa_2CuO_{4+\delta}}$ (Hg1201) [12]. Our results provide detailed information about the temperature evolution of the magnetic fluctuations that is difficult to obtain by INS and resonant inelastic x-ray scattering due to limited beam-time resources. With decreasing temperature, we observe an amplitude enhancement and an energy shift of a "two-magnon" peak attributable to high-energy magnetic fluctuations, which is accompanied by the opening of a gap and the appearance of a pairing peak above the gap. This effect occurs at $T_c$ in an overdoped sample and can hence be understood as a high-energy feedback effect analogous to the resonant mode observed by INS, indicating a contribution of the high-energy magnetic fluctuations to the pairing interaction. In underdoped samples, we observe the same phenomena at temperatures well above $T_c$. This suggests that a related feedback mechanism is operative in the pseudogap regime [13].

We studied three Hg1201 single crystals: strongly underdoped ($T_c = 77$ K, UD77), slightly underdoped ($T_c = 94$ K, UD94), and overdoped ($T_c = 90$ K, OV90), with estimated [12,14] hole concentrations of $p = 0.11$, 0.14, and 0.19, respectively. The crystals were grown by a self-flux method [15]. Sharp transitions at $T_c$, a large diamagnetic signal below $T_c$ in field-cooled measurements [12], and the observation of a long-range ordered magnetic vortex lattice in one of the samples (UD94) [16] demonstrate the high quality of our samples. Hg1201 is nearly ideal for ERS experiments because its simple tetragonal structure with only one $CuO_2$ plane per unit cell minimizes the number of Raman-active phonons and enables measurements in pure symmetry channels. Our ERS data, presented as the Raman susceptibility $\chi''$, were obtained in the $B_{1g}$ geometry, which is sensitive to electronic excitations from the antinodal regions of reciprocal space [17]. Details about the measurement condition can be found in the Supplemental Material [18].

Figure 1(a) displays our results for UD77 over a wide energy ($\omega$) and temperature ($T$) range. The spectra show three key features, which we refer to using nomenclature consistent with the literature [17,19]: (1) the "pseudogap," which manifests itself at low temperatures as a depletion below 570 $\mathrm{cm}^{-1}$; (2) the "pairing peak" centered at 725 $\mathrm{cm}^{-1}$; and (3) the "two-magnon peak" at approximately 1700 $\mathrm{cm}^{-1}$. The energy of feature (1) is consistent with the pseudogap observed by angle-resolved photoemission near the antinodes of the superconducting gap function at comparable doping levels [20]. Feature (2) had long been associated with Cooper pair breaking [17]. Although recent results have cast some doubt on this interpretation [21], its temperature dependence (see below) indicates that it is closely related to superconductivity. The peak energy is consistent with the extrapolation of previous results for Hg1201 from higher doping [22] and with results for other cuprates at similar doping [21,23,24]. To the best of our knowledge, however, Fig. 1 contains the clearest observation of the pairing peak for a doping level as low as UD77 ($p = 0.11$). Feature (3) arises from high-energy electronic fluctuations that smoothly evolve with doping out of the two-magnon excitations in AF parent compounds [24,25]. Although additional quantum phases and correlations may play some role [26,27], the dominant character of these fluctuations thus appears to be closely related to high-energy magnons in the AF insulators.

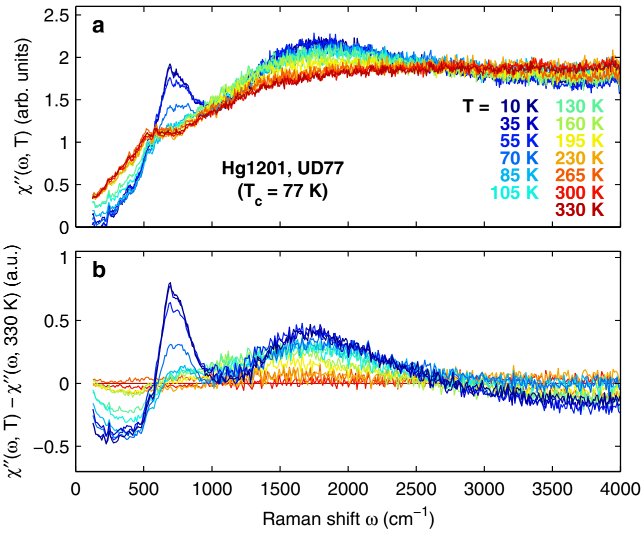

FIG. 1 (color online). (a) Raman spectra for sample UD77. (b) Differential spectra relative to 330 K.

We now discuss the evolution with temperature, which is best seen in the differential spectra $\Delta\chi''$ after subtracting the 330 K data [Fig. 1(b)]. In Fig. 2, the three key features are indicated by arrows color-coded with constant-energy plots in the insets. Our main finding pertains to the temperature dependence of the two-magnon peak (red arrow, circle) and its correlation with the other features. We begin our discussion with the overdoped sample, OV90. Upon cooling from 300 K, the two-magnon signal amplitude first increases linearly with decreasing $T$ [dashed line in the inset of Fig. 2(c)], indicating a slight reduction in thermal broadening. Then, near $T_c$, the signal increases rapidly, in concert with the development of the gap (blue arrow, diamond) and the pairing peak (green arrow, triangle). This $T$ dependence of the two-magnon peak is in fact strikingly similar to that of the low-energy resonance peak observed by INS [2], suggesting a related interpretation as a feedback effect of Cooper pairing on the magnetic fluctuation spectrum. The observation of such feedback for

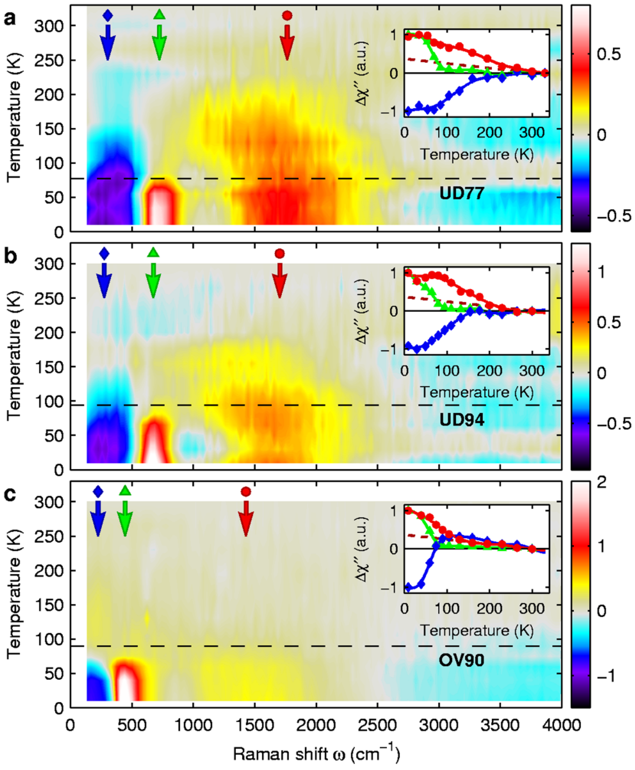

FIG. 2 (color online). Main panels: $\Delta\chi''$ relative to the highest temperatures. The dashed lines indicate $T_c$. Insets: $\Delta\chi''$ at energies indicated by the color- and symbol-coded arrows, normalized to the lowest temperature. The solid curves are guides to the eye. The dashed lines (identical in all insets) describe the high-temperature behavior of the two-magnon amplitude in OV90.

energies far above the superconducting gap is new and surprising [28].

We now turn to the underdoped samples UD77 and UD94 [Figs. 2(a) and 2(b)], where the pseudogap opens up at a characteristic temperature $T_{\mathrm{gap}}$ well above $T_c$ and evolves smoothly through $T_c$. The same trend is observed for the anomaly in the $T$-dependent intensity of the two-magnon peak, which shifts to progressively higher temperatures with decreasing doping. The highly accurate data on the two-magnon peaks in UD94 and UD77 also reveal a slight increase of its energy below $T_{\mathrm{gap}}$ ("banana shape" in the color plots). This further confirms the correlation between these features and demonstrates that a feedback mechanism akin to the one observed in OV90 is also present in the pseudogap regime.

The pairing peak continues to exhibit a strong anomaly at $T_c$ in the underdoped samples [Figs. 2(a) and 2(b)]. However, close inspection of our data (Fig. 3) reveals remnant signals at the pairing peak energy up to 130 K in UD77 and 110 K in UD94. This has not been observed in previous ERS studies on underdoped cuprates, probably due to the peak's weak intensity in underdoped systems [17,22,24] and/or the presence of impurities and strains [21]. These difficulties have been overcome in our study. The onset temperatures of the pairing peaks in UD77 and UD94 are well above $T_c$ and not far from $T_{\mathrm{gap}}$, as can be seen from the tails of the green curves (with triangular symbols) in the insets of Figs. 2(a) and 2(b). In contrast, no extra intensity can be detected already at $T_c = 90$ K in OV90 [Fig. 3(c)]. Despite some quantitative differences in the onset temperatures of the three ERS features in UD77 and UD94 that presumably reflect their different energy scales, the correlation among their doping dependences is a very robust result.

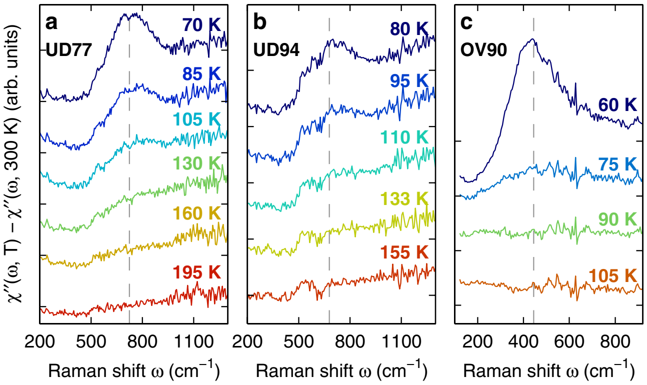

FIG. 3 (color online). Differential spectra relative to 300 K near the pairing peak. Phonon peaks in UD94 and OV90 were removed prior to the subtraction. The data are offset for clarity. The dashed lines indicate the peak positions determined at 10 K.

In order to put the spectral features' characteristic energies on a quantitative footing, we have performed model calculations based on the $t$-$t'$-$J$ Hamiltonian, $H = H_{t,t'} + H_J$. We calculate the spectral response, including both the pairing peak (following [30] and using an *ab initio* tight-binding energy dispersion given in [31]) and the two-magnon peak. The intensity of the latter [32,33] is proportional to $-\mathrm{Im}\{R(\omega)[1 + (1/Sz\alpha)R(\omega)]^{-1}\}$, with

$$R(\omega) = 4\sum_{\mathbf{k}} f_k^2 \frac{\omega_k + \Sigma(\mathbf{k}, \omega)}{\omega^2 - 4[\omega_k + \Sigma(\mathbf{k}, \omega)]^2}. \qquad (1)$$

Here, $f_k$ is the $B_{1g}$ symmetry factor, $\Sigma(\mathbf{k}, \omega) = \Sigma^0 - i\Gamma$ is the self-energy of the one-magnon Green's function (treated as a phenomenological parameter), and $\omega_k = \frac{J^* Sz}{\hbar}\sqrt{1 - \gamma_k^2}$ with $\gamma_k = [\cos(k_x a) + \cos(k_y a)]/2$ is the magnon dispersion. $\alpha = 1.158$ is a numerical constant [33], and $S = 1/2$, $z = 4(1 - p)$, and $a$ are the quantum number of spin, the average number of nearest neighbors, and the in-plane lattice spacing, respectively. In general, our analysis of $H_J$ is valid up to the energy of undamped magnon excitations $\omega \simeq 4J^*$, where $J^*$ is an effective doping-dependent exchange parameter. Interference effects between $H_{t,t'}$ and $H_J$ are neglected. Additional phonon peaks in UD94 and OV90 (but not in UD77), possibly due to oxygen superstructures [34], are not considered. Using AF interaction and gap parameters $J^* = 548$, 516, and 460 cm$^{-1}$ and $\Delta = 379$, 347, and 234 cm$^{-1}$ for UD77, UD94, and OV90, respectively, we find reasonable agreement between the calculation and the experiment [Fig. 4(a)].

Figures 4(c) and 4(d) present the outcome of this analysis, along with our empirical estimates of the characteristic energies. Based on $\Delta\chi''$ in Fig. 4(b), we identify four energies, from low to high, as summarized in Fig. 4(c): (1) the onset of the gap (where $\Delta\chi''$ crosses zero), $\omega_{\mathrm{gap}}$; (2) the center of the pairing peak, $\omega_{\mathrm{pair}}$; (3) the center of the two-magnon peak, $\omega_{\mathrm{2mag}}^{\mathrm{peak}}$; and (4) the high-energy leading edge of the two-magnon peak (half-maximum position), $\omega_{\mathrm{2mag}}^{\mathrm{edge}}$. We make the following observations.

First, while all energies decrease with doping [Fig. 4(c)], $\omega_{\mathrm{2mag}}^{\mathrm{edge}}$ varies only slightly and appears to set an upper bound for $\omega_{\mathrm{2mag}}^{\mathrm{peak}}$ in the extrapolation to zero doping. This is consistent with previous results [24,25]. However, in the undoped limit (which is not accessible in Hg1201), $\omega_{\mathrm{2mag}}^{\mathrm{peak}}$ and $\omega_{\mathrm{2mag}}^{\mathrm{edge}}$ are typically found in the 2800–4000 cm$^{-1}$ range, larger than our extrapolated values [Fig. 4(c)]. [For convenience, in Fig. 4(d), we use the definition of $J^* = \omega_{\mathrm{2mag}}^{\mathrm{peak}}/3$, the same as in [24], which gives $J^*$ as slightly larger than in our model calculation.] We speculate that $\omega_{\mathrm{2mag}}^{\mathrm{edge}}$ is related to the bare AF exchange interaction $J$, which shows only weak doping dependence in other cuprates [11].

Second, with increasing doping, both the pairing peak and the pseudogap increase in signal amplitude [Fig. 4(b)] and the values of $\omega_{\mathrm{gap}}$ and $\omega_{\mathrm{pair}}$ track each other. This implies that the ERS pseudogap is connected to the pairing peak, even though our data do not conclusively show

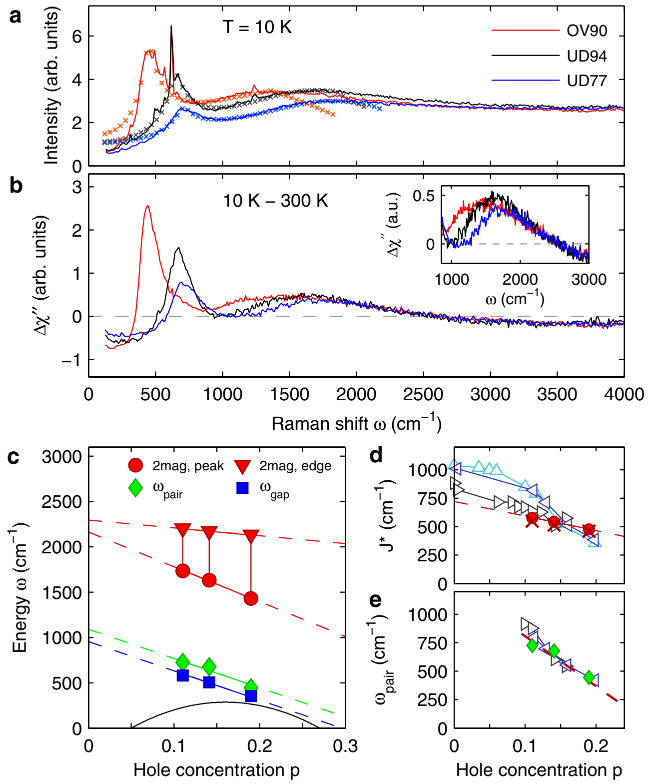

FIG. 4 (color online). (a) Solid curves represent raw data at 10 K. The cross symbols represent model calculations for $\omega$ up to $4J^*$. (b) $\Delta\chi''$ between 10 and 300 K. Phonon peaks in UD94 and OV90 are removed prior to the subtraction. The inset shows an enlarged view of the two-magnon peak. (c) Characteristic energies in Hg1201 (see text). The dome-shaped curve represents $4.28k_BT_c$. (d) Values of $J^* = \omega_{2\mathrm{mag}}^{\mathrm{peak}}/3$ [except for the cross symbols, which are from the calculations in (a)] and (e) $\omega_{\mathrm{pair}}$, in $La_{2-x}Sr_xCuO_4$ (upward triangles), $YBa_2Cu_3O_{6+\delta}$ (rightward triangles), $Bi_{2.1}Sr_{1.9}Ca_{1-x}Y_xCu_2O_{8+\delta}$ (leftward triangles) [24], and Hg1201 (filled symbols, this work). The dashed line in (e) summarizes the $\omega_{\mathrm{pair}}$ for Hg1201 reported in Ref. [22].

whether they have the same onset temperature. Since the onset temperature of the pairing peak is highest in the most underdoped sample UD77 (Fig. 3), this temperature (possibly identical to $T_{\mathrm{gap}}$) might indicate the mean-field $T_c$ [35] and be related to the values of $\omega_{\mathrm{gap}}$ and $\omega_{\mathrm{pair}}$. The characteristic temperatures $T_{\mathrm{gap}}$ as defined by the 10% depletion are considerably lower than the pseudogap temperature $T^*$ determined from, e.g., in-plane resistivity and NMR: for doping levels similar to UD94 and UD77, $T^*$ is approximately 200 K [36,37] and above 250 K [12,37], respectively. This difference may be related to the presence of multiple characteristic temperatures above $T_c$ [36,38], which might further depend on the time scale of the probe.

Finally, we find no clear correlation between $J^*$ and $T_c$ near optimal doping in a comparison with other compounds [Fig. 4(d)], including $La_{2-x}Sr_xCuO_4$, which has a relatively low $T_c^{\max} < 40$ K. All of them have nearly the same $J^*$ for $p \sim 0.16$. This implies that other factors affect the attainable $T_c^{\max}$, as has been suggested by other authors [25,39].

To conclude, we have observed a correlation among the temperature dependences of the two-magnon peak, the pseudogap, and the pairing peak in a model cuprate high-$T_c$ superconductor. In the overdoped regime, this correlation can be attributed to a feedback effect of Cooper pairing on high-energy magnetic excitations, analogous to the low-energy resonant mode observed by INS [2]. This is consistent with anomalies observed in various fermionic spectral functions [8,9] and directly supports prior indications of a substantial contribution of high-energy magnetic fluctuations to the pairing interaction [11,40]. The observation of a closely similar feedback effect in the pseudogap regime is consistent with prior reports of superconducting correlations above $T_c$ [36,38,41], although other ordering phenomena [42] and excitations [43] may also contribute to this effect in the underdoped samples.

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