Pseudogap in $YBa_2Cu_3O_{7-\delta}$ from NMR in high magnetic fields

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We report on ^{17}O(2,3) and ^{63}Cu(2) spin-lattice relaxation rates and the ^{17}O(2,3) spin-spin relaxation rate in different magnetic fields in $YBa_2Cu_3O_7$ near T_c. Together these measurements enable us to test the magnetic-field dependence of the pseudogap effect on the spin susceptibility in different regions of the Brillouin zone using the known form factors for different nuclei as filters. Thus, we study the momentum dispersion of the pseudogap behavior. We find that near the antiferromagnetic wave vector the pseudogap is insensitive to magnetic fields up to 15 T. In the remaining region, away from the (π,π) point, the pseudogap shows a magnetic-field dependence at fields less than 10 T. The first result is indicative of the opening of a spin pseudogap that suppresses antiferromagnetic correlations below a temperature T^*; whereas, the second result shows the effect of pairing fluctuations on the spin susceptibility as a precursory effect of superconductivity.

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I. INTRODUCTION

The nature of the onset of superconductivity in high-temperature superconductors (HTS) is of considerable interest since it reflects a complex interplay between magnetism and superconductivity that is not yet understood. Experiments show[1] that below a temperature T^*, higher than T_c, a gaplike structure appears in the electronic excitation spectrum. However, at present there is no consensus on either the origin of the pseudogap nor its relationship to superconductivity.[2–5,7] Photoemission and surface-tunneling studies suggest that the pseudogap above T_c evolves smoothly into the superconducting gap below T_c (Refs. 2 and 4) implying that the pseudogap originates from the pairing fluctuations as a precursory mechanism to superconductivity. In particular, angle-resolved photoemission studies[2,6] provide evidence for a highly anisotropic pseudogap, for $T_c < T < T^*$, which is similar in magnitude and angular dependence to the d-wave superconducting gap below T_c. In contrast, intrinsic tunneling studies,[3,8] which show that a distinct superconducting gap coexists with the pseudogap below T_c, argue against a superconducting pairing origin of the pseudogap. A crucial test for the latter is provided by investigating its sensitivity to magnetic field. This idea led to a series of nuclear magnetic resonance (NMR) spin-lattice relaxation rate measurements on the copper nucleus in high magnetic fields.[5,7,9,10] Recent neutron-scattering[11] and tunneling experiments[3,8,12] have also investigated magnetic-field effects in the normal state of HTS. A neutron-scattering experiment[11] up to 6.8 T suggests that the pseudogap is of pairing origin, while interlayer tunneling measurements[12] up to 60 T reveal that spin degrees of freedom play a predominant role in the formation of the pseudogap. NMR can be particularly useful for investigating the sensitivity of the pseudogap to magnetic field, but it is especially important since NMR can probe the q (momentum-transfer wave vector) dependence of the pseudogap in the spin excitation spectrum by taking advantage of known q dependent form factors that are different for various relaxation experiments involving different nuclei, specifically copper and oxygen.

In this work, we report a complete set of NMR relaxation measurements: ^{17}O(2,3) spin-lattice relaxation rate $^{17}T_1^{-1}$; ^{63}Cu(2), $^{63}T_1^{-1}$; and the ^{17}O(2,3) spin-spin relaxation rate $^{17}T_2^{-1}$, as a function of magnetic field near T_c, up to 23 T. These measurements reveal a field dependence of the dynamic spin susceptibility, $\chi(\mathbf{q},\omega\rightarrow 0)=\chi'+i\chi''$, that varies with $\mathbf{q}$. This indicates coexistence of pairing superconducting fluctuations and a spin pseudogap. Gorny *et al.*[7] pointed out that $\chi''(\mathbf{q},0)$ near $\mathbf{q}=(\pi,\pi)$ shows no major field dependence on the scale of 10 T based on ^{63}Cu NMR experiments. At this position in the Brillouin zone χ is strongly enhanced by antiferromagnetic (AF) spin fluctuations, and so this result suggests that the temperature dependence of $^{63}T_1^{-1}$ is not a manifestation of precursory superconductivity but is controlled by a much higher field scale possibly associated with a suppression of low-energy spin fluctuations. From our ^{17}O NMR relaxation measurements we find that $\chi''(\mathbf{q},0)$, away from the (π,π) point, is magnetic field dependent on the scale of 10 T. This field dependence can be explained in terms of superconducting fluctuations, or a pairing pseudogap that appears $\sim$20 K above T_c. We describe the experiment in Sec. II. In Sec. III, we discuss how NMR can be used to probe the q-dependent susceptibility. Results and discussion are presented in Secs. IV–VIII.[13,14]

II. EXPERIMENT

We have investigated a sample used in our previous work[5,15] on spin relaxation and Knight shift. It is a near-optimally doped $\sim 30-40\%$ ^{17}O-enriched, $YBa_2Cu_3O_{7-\delta}$ (YBCO), aligned powder sample, provided courtesy of P. C. Hammel at Los Alamos National Laboratory. The crystal $\hat{c}$ axis was aligned with the direction of the applied magnetic field, the z axis. Low-field magnetization data show a sharp transition at $T_c(0)=92.5$ K. This sample has a relatively

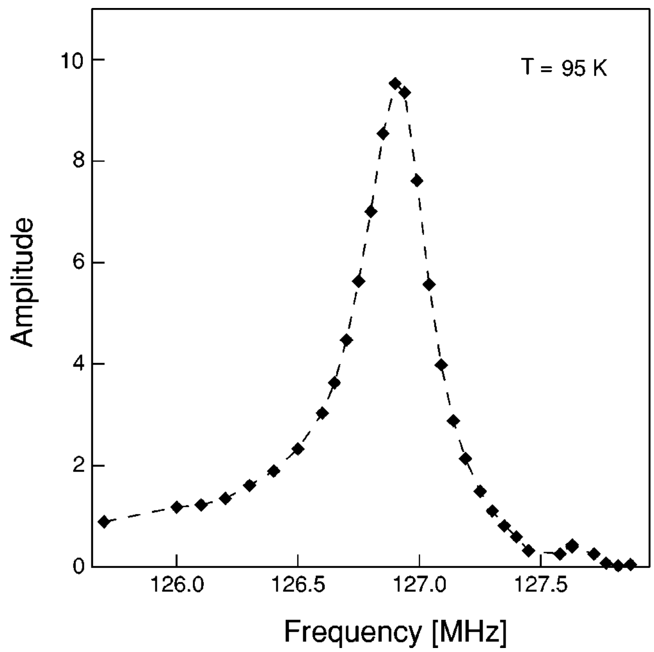

FIG. 1. The first high-frequency satellite, i.e., $\langle -\frac{3}{2} \leftrightarrow -\frac{1}{2} \rangle$ transition, spectrum of ^{65}Cu at 8 T and 95 K. The signal is essentially background free on the high-frequency side.

narrow nuclear quadrapole resonance (NQR) linewidth of ≈ 290 kHz and its NQR frequency is $^{63}\nu_{zz} = 31.5$ MHz. In Fig. 1, we show the first high-frequency satellite of the ^{65}Cu spectra at 8 T and 95 K. We have checked that the width of this spectrum is the same as the NQR linewidth. Our measurements were made at temperatures from 70 to 160 K and over a wide range of magnetic fields, from 1.1 T to 22.9 T. ^{17}O(2,3) NMR spin-spin relaxation was measured using a Hahn echo sequence: $\pi/2$-τ-π-acquire.

The spin-lattice relaxation rate was measured using the following sequence: $\pi/2$-τ_1-$\pi/2$-τ-π-acquire. $^{17}T_1^{-1}$ was measured on the first high-frequency satellite, i.e., $\langle -\frac{3}{2} \leftrightarrow -\frac{1}{2} \rangle$ Zeeman transition, of the O(2,3). To exclude the possibility of some field-dependent background contribution to the rate, we have compared T_1^{-1} values measured on that satellite to the rate measured at the $\langle \frac{3}{2} \leftrightarrow \frac{1}{2} \rangle$ transition. All $^{63}T_1^{-1}$ measurements were made on satellites, $\langle \pm\frac{3}{2} \leftrightarrow \pm\frac{1}{2} \rangle$. At low field, 1.1 and 2.4 T, the rate was measured on the high-frequency satellite of ^{63}Cu, which is the highest frequency Cu signal at that field, meaning that the high-frequency side of this transition is background free, following the approach suggested by Gorny *et al.*[7] Very good signal-to-noise ratio was obtained even at such low fields owing to the population difference enhancement by a strong quadrupolar interaction. At 8 T, T_1 was measured on the high-frequency satellite of ^{65}Cu, the highest frequency Cu signal at that field, whose spectrum is shown in Fig. 1. The rate of ^{63}Cu is then inferred from $^{65}T_1$ knowing that their ratios scale as the square of their gyromagnetic ratios γ, namely, $^{63}T_1 = {}^{65}T_1({}^{63}\gamma/{}^{65}\gamma)^2 = 0.8713\,{}^{65}T_1$. At 14.7 T, T_1 was measured on the low-frequency satellite of ^{63}Cu whose low-frequency side is background free. The rates were extracted by fitting to the appropriate recovery profiles, assuming a magnetic relaxation mechanism.

III. NMR TOOLS

In this section, we give a brief overview of how NMR is used to probe the q-dependent susceptibility. The spin-lattice relaxation rate is the rate at which the nuclear magnetization relaxes to its thermal equilibrium value in the external magnetic field. It can be conveniently expressed in terms of the generalized spin susceptibility $\chi(\mathbf{q},\omega)$ as,

$$\frac{1}{(T_1T)_\alpha} \propto \lim_{\omega_n \to 0} \sum_{\mathbf{q},\alpha' \neq \alpha} \left[|F_{\alpha'\alpha'}(\mathbf{q})|^2 \frac{\chi''_{\alpha\alpha}(\mathbf{q},\omega_n)}{\omega_n} \right], \quad (1)$$

where α is the direction of H_0; $F_{\alpha'\alpha'}(\mathbf{q})$, referred to as a form factor, is the Fourier transform of the hyperfine coupling between nuclei and electrons; and $\chi''_{\alpha'\alpha'}(\mathbf{q},\omega)$ is the imaginary part of the dynamic spin susceptibility for the wave vector $\mathbf{q}$ and nuclear Larmor frequency ω_n with the direction α' perpendicular to α.

The q dependence of relevant form factors in this work, and the imaginary part of susceptibility dominated by AF spin fluctuations, are shown in Fig. 7 in the Appendix. We see from Eq. (1) that it is these form factors which enable us to probe $\chi(\mathbf{q},\omega_n)$ in different regions of the Brillouin zone, through the measurement of T_1. For ^{63}Cu(2) spin-lattice relaxation, the appropriate form factor has significant weight near $\mathbf{q} = (\pi,\pi)$, the AF wave vector. Since the imaginary part of the susceptibility is peaked at this wave vector the copper relaxation is dominated by AF spin fluctuations. In contrast for planar oxygen, ^{17}O(2,3), the spin-lattice relaxation in the normal state is mostly insensitive to AF fluctuations owing to its vanishingly small form factor at $\mathbf{q} = (\pi,\pi)$.

The nuclei also interact indirectly via conduction electrons[16] depending on the real part of their magnetic susceptibility. These indirect processes dominate the Cu spin-spin relaxation,[17] and are reduced for oxygen. However, an important part of T_{2G} of ^{17}O(2,3) still arises from Cu-O indirect coupling and can be written as

$$\left(\frac{1}{^{63-17}T_{2G}} \right)^2_{ind} \propto \sum_{\mathbf{q}} \left[{}^{17}F_{\alpha'}(\mathbf{q}) \cdot {}^{63}F_{\alpha'}(\mathbf{q}) \cdot \chi'(\mathbf{q},0) \right]^2, \quad (2)$$

where $^{17}F_{\alpha'}(\mathbf{q})$ and $^{63}F_{\alpha'}(\mathbf{q})$ are form factors of O and Cu, respectively, for $\alpha' = c$ for the case $\hat{c} || \hat{z}$. Unlike the case of $^{63}(T_{2G})_{ind}$ that arises from Cu-Cu indirect coupling and probes $\chi'(\mathbf{q},0)$ near (π,π), $^{63-17}(T_{2G})_{ind}$ arises from Cu-O coupling and probes $\chi'(\mathbf{q},0)$ in the intermediate region of the Brillouin zone between (π,π) and $(0,0)$. This relaxation experiment is complementary to the measurements of spin-lattice relaxation. Finally, the Knight shift probes the real part of static spin susceptibility at $\mathbf{q} = 0$, $\chi'(0,0)$ which we have reported earlier[15] using a wide range of magnetic fields.

To summarize, in order to characterize the dynamic spin susceptibility at different q, we have measured the following quantities: $^{17}T_1^{-1} \propto \chi''/\omega$ for $\mathbf{q}$ near (0,0); $^{63}T_1^{-1} \propto \chi''/\omega$ for $\mathbf{q}$

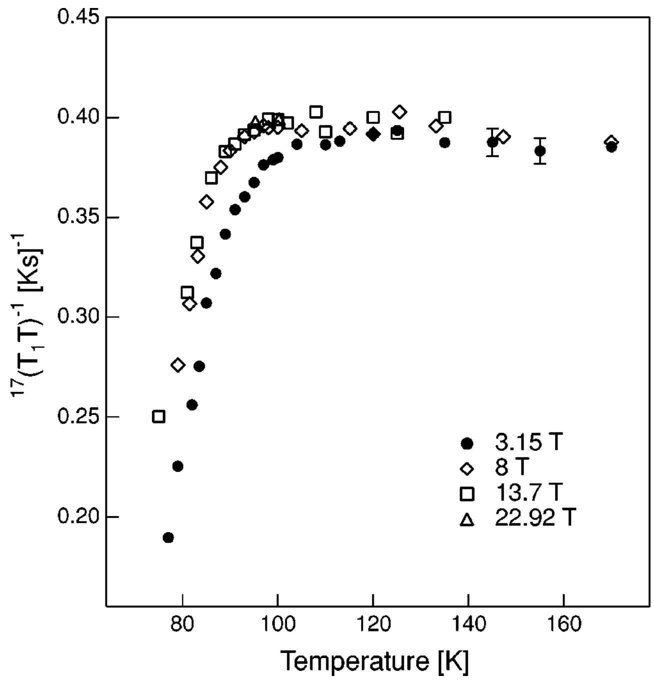

FIG. 2. Spin-lattice relaxation rate of $^{17}O(2,3)$ in YBCO as a function of temperature in the magnetic fields of 3.15, 8, 13.7, and 22.92 T. A typical value of the error bars at all applied fields is as shown at high temperatures, $T\sim140-160$ K, at 3.15 T.

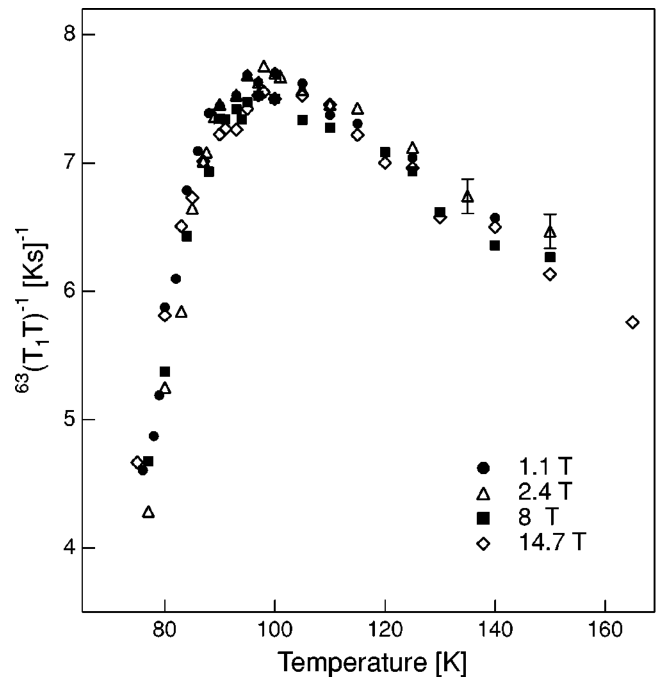

FIG. 3. Spin-lattice relaxation rate of $^{63}Cu(2)$ in YBCO as a function of temperature in the magnetic fields of 1.1, 2.4, 8, and 14.7 T. A typical value of the error bars at all applied fields is as shown at high temperatures at 2.4 T.

near (π,π); $^{63-17}(T_{2G})^{-1}_{ind}\propto\chi'$ for $\mathbf{q}$ between (0,0) and (π,π). Using these tools, we investigate the response of $\chi(\mathbf{q},0)$ to a magnetic field near T_c to determine which processes affect χ.

IV. $^{17}T_1$ RESULTS

As previously pointed out $^{17}(T_1)^{-1}$ probes the imaginary part of the electronic spin susceptibility, $\chi''(\mathbf{q},0)$, close to $\mathbf{q}=(0,0)$. In Fig. 2, we show the ^{17}O spin-lattice relaxation rate as a function of temperature in different magnetic fields. We find that the rate increases with increasing magnetic field, on the scale of 10 T, for $T<110$ K. At 95 K $^{17}(T_1T)^{-1}$ differs by $\sim7\%$ between 3.2 and 8 T. The departure of $^{17}(T_1T)^{-1}$ from the Korringa-like behavior, $(T_1T)^{-1}=$constant, shifts towards lower temperatures as the field increases and the rate drops sharply in the superconducting state, consistent with reduction of T_c by the field.[15] Thus, we can conclude that the pseudogap we observe here is tied, at least in part, to superconductivity. A simple shift in T_c is not enough to account for this field dependence above T_c because the curvature of the data changes with field. Our Knight-shift data[15] indicate a T_c shift of ~2 K from 3.2 to 8 T. However, $^{17}(T_1T)^{-1}$ has a value of 0.367 $(\mathrm{Ks})^{-1}$ at 3.2 T at 95 K and at 8 T the same value at 86 K. This shift of 9 K exceeds by far the shift of T_c with field. We can account for this behavior by *d*-wave density-of-states (DOS) pairing fluctuations following previously reported analysis.[5,15,28]

As the magnetic field increases it suppresses the negative DOS pairing fluctuation contribution to the rate causing the overall rate at a fixed temperature to increase with increasing field[28] as observed in Fig. 2. However, we observe that the field dependence of the rate saturates around 10 T and that at "high" field, $H\geqslant10$ T, it has well-defined field-independent curvature near T_c. The saturation of the field dependence on this low-field scale is not predicted by the calculations of superconducting fluctuation contributions to the NMR rate.[28] Thus, our observations indicate that DOS pairing fluctuations may not be the only process affecting $^{17}(T_1T)^{-1}$. In Sec. VII, we try to model the influence of a spin pseudogap on $^{63}(T_1T)^{-1}$ and, with the same parameters, estimate its effect on $^{17}(T_1T)^{-1}$.

V. $^{63}T_1$ RESULTS

In Fig. 3, we show $^{63}T_1$. We observe no discernible field dependence in the normal state within experimental accuracy of $\pm2\%$. This result is consistent with that reported by Gorny *et al.*[7] Above ~100 K, $(T_1T)^{-1}$ can be fitted to a Curie-Weiss-like relation, $(T_1T)^{-1}\propto T_x/(T+T_x)$, where we obtain $T_x=103$ K based on our 8 T data. This relation for $(T_1T)^{-1}$ is to be expected if it is dominated by AF spin fluctuations.[18] The peak in $^{63}(T_1T)^{-1}$ is observed at $T^*\sim100$ K. Reduction of $^{63}(T_1T)^{-1}$ below T^* has been associated with the loss of low-energy spectral weight[19] of the spin fluctuations, which is caused by the opening of a pseudogap. It is interesting to note that in spite of the fact that T_c decreases with increasing field, $^{63}(T_1T)^{-1}$ falls off independent of the magnetic field, indicating that down to ~80 K the zero-frequency limit of $\chi''(\mathbf{q},\omega)/\omega$ for $\mathbf{q}=(\pi,\pi)$ is not sensitive to superconductivity. This result implies that the suppression of $^{63}(T_1T)^{-1}$, and consequently the zero-frequency limit of $\chi''(\mathbf{q},\omega)/\omega$ for $\mathbf{q}=(\pi,\pi)$, is most likely due to the opening of a spin pseudogap, i.e., the loss of low-energy spin fluctuations.

Next we discuss the field dependence of the real part of the spin susceptibility, i.e., ^{17}O spin-echo decay arising from the indirect Cu-O coupling.

VI. Cu-O INDIRECT COUPLING

The main source of spin-echo decay of ^{17}O is the copper spin-lattice relaxation, as proposed by Walstedt and Cheong[20] and demonstrated by Recchia *et al.*[21] The *z*-component fluctuating fields from copper nuclear-spin flips are transferred to the oxygen nuclei by Cu-O nuclear dipolar interactions. An additional important contribution to the ^{17}O spin-echo decay is an indirect Cu-O nuclear coupling, denoted by k, mediated by the conduction electrons,[20,21] see Eq. (2).

Mitrović *et al.*[5] extracted $^{63}T_1$ from a fit of the $^{17}T_2$ data near T_c choosing k to match the high-temperature results for $^{63}T_1$ well above T_c. This procedure is incorrect, since the wrong field dependence of $^{63}T_1$ is inferred as compared with direct measurements presented in Sec. V. We suspect that the parameter k, that describes Cu-O indirect coupling, is a temperature- and field-dependent quantity, contrary to the assumption made in the work of Recchia *et al.*[21] and Mitrović *et al.*[5]

We now examine in more detail the relaxation described by the parameter k. We can extract the part of the spin-spin relaxation due to Cu-O *indirect* coupling from our $^{17}T_2$ data by dividing our measured signal M by that calculated for *direct* dipolar coupling. We take into account relaxation from the direct dipolar interaction between copper and oxygen spin flips using our direct measurements of $^{63}T_1^{-1}$. We then fit the residual decay to a Gaussian function of time and show the resulting relaxation times in Fig. 4 vs temperature for magnetic fields from 2.1 to 22.8 T. The $^{63-17}(T_{2G})^{-1}_{ind}$ data is extracted with a typical accuracy of $\pm 5\%$.[22,23]

In the high-temperature region, $T>100$ K, we observe that the rate decreases with decreasing temperature and shows no discernible field dependence. For lower temperatures there is a small well-defined field dependence for the $^{63-17}(T_{2G})^{-1}_{ind}$ data as reported earlier by Mitrović *et al.*[5] In Fig. 4, this is evident in the normal state for temperatures $T_c(H)<T<100$ K where $T_c(H)$ is denoted by the intercept between the data and the dashed line shown in the figure. The relatively low-field scale for this dependence, in contrast to $^{63}(T_1T)^{-1}$ in Fig. 3, suggests a connection to superconductivity, most likely from pairing fluctuations. Below T_c, the rate shifts to lower temperatures as the field increases, consistent with reduction of T_c by the field, indicating that this lower temperature behavior is also connected to superconductivity. For example, at the applied field of 3.2 T and at the temperature $T=0.9T_c(H)$ the rate drops by $\sim 20\%$ from its value at $T_c(H)$. For higher applied fields the decrease is smaller.

Superconductivity can affect T_2 in two ways: through vortex vibrations, whose precise contribution to the rate is not known; and, through the suppression of χ' due to pair formation. Vortex vibrations might be responsible for the observations below $T_c(H)$. We have performed a random-phase approximation (RPA) calculation[26] of the temperature and field dependence of both χ' and $^{63-17}(T_{2G})^{-1}_{ind}$ due to pair formation, similar to Bulut and Scalapino,[24] and found that the calculated temperature dependence is too small to account for our observations near T_c.[25]

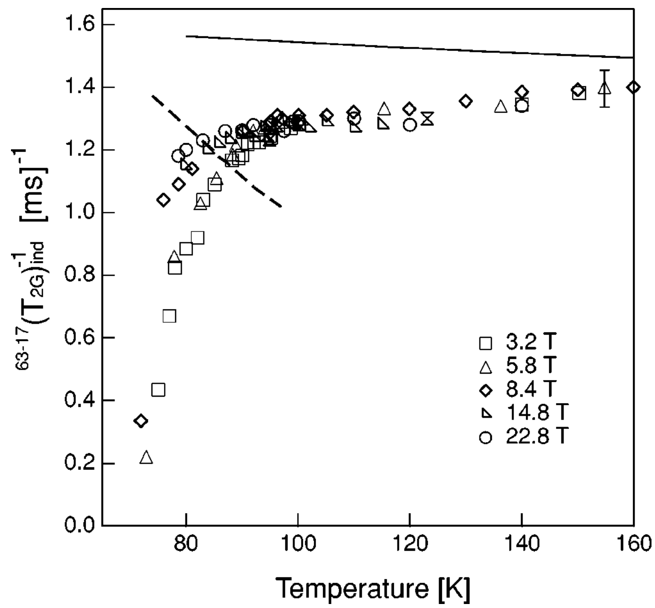

FIG. 4. Spin-spin relaxation rate of ^{17}O(2,3) after dividing out the part of the relaxation coming from the direct Cu-O dipolar interaction as described in the text. The data at temperatures higher than indicated by the dashed line are above $T_c(H)$ (Ref. 15) and in the normal state. The solid line is the calculated spin-spin relaxation from Cu-O indirect coupling using the same susceptibility parameters as we used to calculate $^{17}(T_1T)^{-1}$ and $^{63}(T_1T)^{-1}$ for $T>120$ K (Ref. 26).

It is possible that the relaxation described by k, does not arise only from Cu-O indirect coupling but comes rather from an additional relaxation mechanism that is highly sensitive to superconductivity and associated only with oxygen. A possible candidate for this extra relaxation component is the low frequency mostly oxygen charge fluctuations discussed by Suter *et al.*[27] They showed in $YBa_2Cu_4O_8$ that there is a significant contribution from quadrupolar fluctuations, i.e., low-frequency charge fluctuations, to $^{17}T_1$ in addition to the dominant contribution from magnetic fluctuations that onsets at $T\approx 200$ K.

Regardless of the precise origin of the relaxation mechanism described by k, we see that it depends on temperature and magnetic field, in contrast with previous assumptions.[21]

VII. PSEUDOGAP MODEL

In the following, we try to model the influence of a spin pseudogap on $^{63}(T_1T)^{-1}$ and, with the same parameters, estimate the effect on $^{17}(T_1T)^{-1}$.

We take the Millis, Monien, and Pines (MMP) (Ref. 18) phenomenological expression for the dynamical susceptibility, altered so as to include the incommensurations in the susceptibility peaks at $\mathbf{Q}_i=(\pi\pm\delta,\pi\pm\delta)$ AF wave vectors,[29]

$$\chi(\mathbf{q},\omega)=\chi_{AF}+\chi_{FL}=\frac{1}{4}\sum_j\frac{\alpha\xi^2\mu_B^2}{1+\xi^2(\mathbf{q}-\mathbf{Q}_j)^2-i\omega/\omega_{SF}}+\frac{\chi_0}{1-i\pi\omega/\Gamma}, \tag{3}$$

where ξ is the spin-fluctuation correlation length in units of the lattice constant a, α is a scaling factor, ω_{SF} the frequency of spin fluctuations, and χ_0 and Γ are terms added to describe the Fermi-liquid (FL) background for AF fluctuations. The rate divided by the temperatures, for $H||\hat{c}$ is then evaluated by summing the product of the form factor and imaginary part of $\chi(\mathbf{q},\omega)$ divided by frequency in the limit of $\omega\rightarrow 0$ over all $\mathbf{q}$,

$$\frac{1}{T_1T}=\frac{k_B}{2\mu_B^2\hbar^2}\sum_q F_c(\mathbf{q})\left[\frac{1}{4}\sum_j\frac{\alpha\xi(T)^2\mu_B^2/\omega_{SF}}{[1+\xi(T)^2(\mathbf{q}-\mathbf{Q}_j)^2]^2}+\frac{\chi_0\pi}{\Gamma}\right]. \tag{4}$$

We take Shastry-Mila-Rice[30] form factors given in Eq. (A1). In addition, ω_{SF} is assumed to be proportional to $\xi(T)^{-2}$ and that $\xi(T)=\xi_0[T_x/(T_x+T)]^{1/2}$. Temperature dependence of $\xi(T)$, ω_{SF} and other parameters were determined so that both calculated $^{63}(T_1T)^{-1}$ and $^{17}(T_1T)^{-1}$ coincide with our data. Assuming that $\mathbf{Q}_{AF}=(\pi\pm 0.1,\pi\pm 0.1)$, we find the following values for the parameters used to calculate $(T_1T)^{-1}$ s: $\xi(T)=3.07[114\ \mathrm{K}/(114\ \mathrm{K}+T)]^{1/2}$, $\omega_{SF}=6.09\xi(T)^{-2}$ meV, $\alpha=14.8\ (\mathrm{eV})^{-1}$, and for the Fermi-liquid part, $\chi_0\pi/\mu_B^2\hbar\Gamma=8.885\ \mathrm{eV}^{-2}$.[31]

We obtain values of $(T_1T)^{-1}$, for both ^{17}O and ^{63}Cu, shown as the solid curve (extending to dashed below 120 K) in Fig. 5. We notice that $^{17}(T_1T)^{-1}$ increases slightly with decreasing temperature similar to $^{63}(T_1T)^{-1}$ due to the increasing correlation length for spin fluctuations, indicating that ^{17}O is not completely shielded from the AF spin fluctuations by its form factor as shown in the Appendix.

We then model the opening of the pseudogap by assuming that it only affects ω_{SF}. We take the following phenomenological form for ω_{SF}:

$$\omega_{SF}^{-1}\propto\frac{\{\tanh[(T-T_p)/c_1]\}}{[\xi_0^{-2}\xi(T)^2]}, \tag{5}$$

where $c_1=14.5$ K and $T_p=70$ K are parameters chosen with the sole purpose to allow a fit to the measured $^{63}(T_1T)^{-1}$, giving the solid curve in Fig. 5(b) below $T\approx 100$ K. Using exactly the same pseudogap parametrization, we calculate $^{17}(T_1T)^{-1}$ giving the corresponding solid curve in Fig. 5(a). We clearly see that the suppression of $^{63}(T_1T)^{-1}$ due to the opening of the spin pseudogap, as modeled here, will also cause a small suppression of

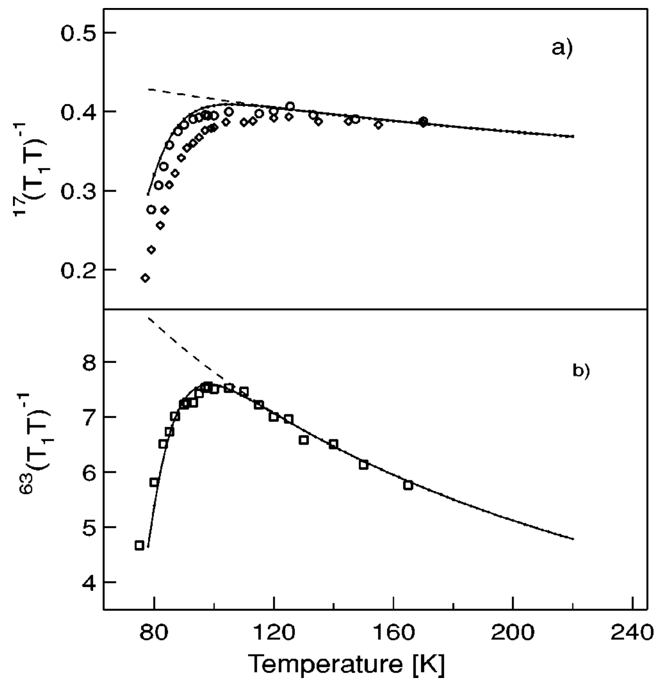

FIG. 5. (a) Spin-lattice relaxation rate of ^{17}O(2,3) at 3.15 T (open diamonds) and 8 T (open circles); and (b) Spin-lattice relaxation rate of ^{63}Cu(2) at 8 T (open squares) as a function of temperature. Solid and dashed lines are calculated as explained in the text.

$^{17}(T_1T)^{-1}$ that reproduces the observed curvature of the high-field $^{17}(T_1T)^{-1}$ data near T_c. From our simplistic model, we have shown phemonenologically that $^{17}(T_1T)^{-1}$ is affected by the opening of a spin pseudogap and that it is this process that dominates the oxygen spin-lattice relaxation rate at fields above 10 T near T_c, adding to the effects of superconducting pair fluctuations that give field dependence at low field.

We point out that it is possible that pairing fluctuations might also affect $\chi''(\mathbf{q},0)$ near $\mathbf{q}=(\pi,\pi)$.[28] However, it is not observed. The observed pairing fluctuation contribution to $^{17}(T_1T)^{-1}$ from the Fermi-liquid susceptibility near T_c at small wave vectors $\mathbf{q}$ would change $^{63}(T_1T)^{-1}$ by less then a percent, making it impossible to discern in our experiment.

VIII. FIELD DEPENDENCE OF NMR RATES

We summarize our relaxation experiments by showing the relative effect of magnetic field on $\chi(\mathbf{q},0)$. This can be conveniently represented by $R(H)$ defined as $R(H)=[(T_{1,2})_{tot}^{-1}-(T_{1,2})_n^{-1}]/(T_{1,2})_n^{-1}$, where the normal-state rate, $(T_{1,2})_n^{-1}$, is a fit to the field-independent high-temperature behavior ($T>120$ K) of the appropriate rate, 1 or 2. The results at $T=95$ K are given in Fig. 6. The two upper graphs indicate that both real and imaginary parts of the spin susceptibility away from $\mathbf{q}=(\pi,\pi)$ have magnetic-field dependence above T_c on the scale of 10 T. The field dependence of χ'' is likely caused by superconducting pair fluctuations; specifically, there is a field-induced suppression of the negative contribution to the rate from the density of states fluctuations.[28]

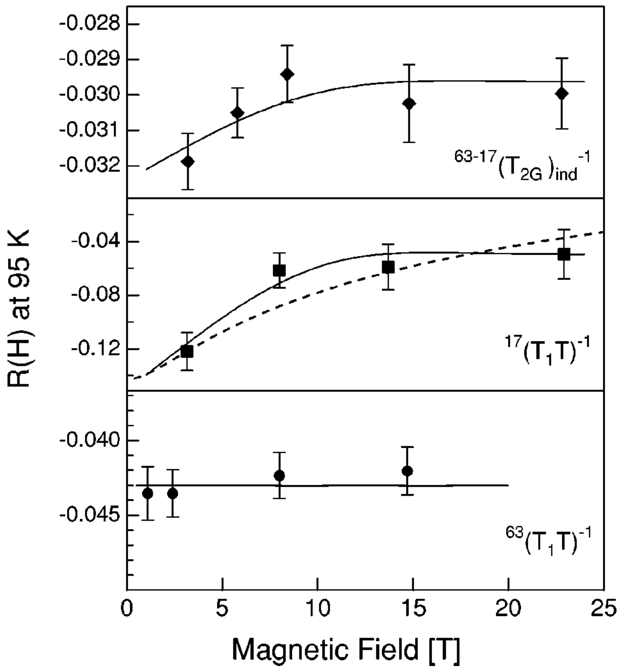

FIG. 6. Field dependence of the relaxation $R(H)=[(T_{1,2})^{-1}_{tot}-(T_{1,2})^{-1}_{n}]/(T_{1,2})^{-1}_{n}$, at 95 K. The normal-state rate, $(T_2)^{-1}_n$, of $^{63-17}(T_{2G})^{-1}_{ind}$ was defined as $(T_2)^{-1}_n=1.127\ (\mathrm{ms})^{-1}+T0.001\,76\ (\mathrm{K\,ms})^{-1}$; for $^{17}(T_1)^{-1}$ we take $(T_1)^{-1}_n=\mathrm{const}=0.395\ \mathrm{s}^{-1}$; and, for $^{63}(T_1)^{-1}$ we take $(T_1)^{-1}_n=15.08\ \mathrm{s}^{-1}[104\ \mathrm{K}/(104\ \mathrm{K}+T)]$. The dashed line is a *d*-wave calculation of the fluctuation contribution to the rate (Refs. 5 and 28). The solid curves are guides to the eye.

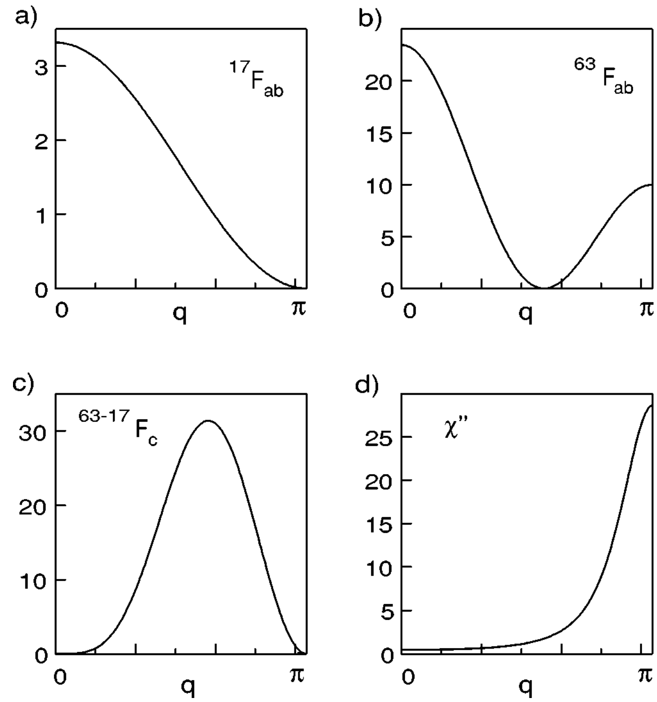

FIG. 7. Form factors (Ref. 30) in units of B^2, as defined in Eq. (A1) (solid curves): (a) Form factor that determines $^{17}T_1$ for a magnetic field $H_0||\hat{c}$. (b) Form factor that determines $^{63}T_1$ for $H_0||\hat{c}$. (c) Form factor that determines $(^{63-17}T_2)_{ind}$ for $H_0||\hat{c}$. (d) χ'' dominated by antiferromagnetic-spin fluctuations (Ref. 18) plus a small Fermi-liquid background.

The dashed line in Fig. 6 shows the fit to the theoretical calculation for the field dependence of the fluctuation corrections for *d*-wave pairing assuming that the $^{17}T_1^{-1}$ data are not influenced by a spin pseudogap. The temperature scale for this calculation is set by the zero-field transition temperature. The curvature of the field dependence is dictated by the mean-field transition temperature in a field, determined by the divergence of the pair fluctuations and obtained from our fit to the spin susceptibility.[15] There is only one fitting parameter for the overall scale of the fluctuation contributions to $(T_1T)^{-1}$. The fit is not perfect; however, the order of magnitude of the calculated field dependence, below 15 T, agrees quantitatively with the experimental data at $T=95$ K and provides evidence for the existence of *d*-wave pairing fluctuations.

IX. SUMMARY

Our measurements show that near T_c the electronic spin susceptibility responds to a magnetic field differently in different parts of the Brillouin zone. This result implies that the spin susceptibility is affected by different physical processes. Near $\mathbf{q}=(\pi,\pi)$ antiferromagnetic spin fluctuations dominate the spin susceptibility, which is insensitive to superconducting fluctuations. In the region away from $\mathbf{q}=(\pi,\pi)$ the influence of superconducting fluctuations on the susceptibility is evident. This is consistent with a Fermi-liquid-like behavior in which the susceptibility is suppressed by superconducting fluctuations for $\mathbf{q}$ less than the inverse of the superconducting coherence length. The magnetic-field behavior of $\chi(\mathbf{q},0)$ indicates the coexistence of two pseudogaps of different origins. One pseudogap dominating $\chi(\mathbf{q},0)$ near $\mathbf{q}=(\pi,\pi)$ is insensitive to magnetic fields in our experimental range $\geqslant 15$ T. This insensitivity indicates that this pseudogap is not intimately tied to superconductivity and that its possible origin is the opening of the spin pseudogap, i.e., loss of the low-frequency spin fluctuations. The second pseudogap, evident in $\chi(\mathbf{q},0)$ away from $\mathbf{q}\sim(\pi,\pi)$ has a low-field scale of <10 T and likely originates from superconducting fluctuations as a precursory effect of superconductivity. The latter can be expected since the appropriate field scale in this case is determined[28] by the thermodynamic critical field, ≈ 5 T.

Finally, we emphasize that in the high-field limit, the temperature dependence of all the rates we have measured, changes markedly above T_c, around ~ 100–110 K, indicating sensitivity to opening of the spin pseudogap.

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APPENDIX: FORM FACTORS

The form factors, relevant for this work, are Shastry-Mila-Rice[30] form factors given by

$$^{63}F_c=[A_{ab}+2B(\cos q_xa+\cos q_ya)]^2,$$

$$^{63}F_{eff}=[A_c+2B(\cos q_xa+\cos q_ya)]^2,$$

$$^{17}F_ab=2C^2[\cos(q_xa/2)^2+\cos(q_ya/2)^2],$$

$$^{17-63}F_c={}^{63}F_{eff}^{17}F_ab, \tag{A1}$$

where $A_{ab}=0.84B$, $A_c=-4B$, $C=0.91B$, and $B=3.82\times10^{-7}$ eV. Their q dependence is shown in Fig. 7 along with the imaginary part of the susceptibility that is dominated by AF spin fluctuations.

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