The Pseudogap in YBa$_2$Cu$_3$O$_{7-\delta}$ from NMR in High Magnetic Fields

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(Version March 22, 2022)

We report $^{17}$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$_2$Cu$_3$O$_{7}$ 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 $(\pi, \pi)$ 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.

PACS numbers: 74.25.Nf, 74.40.+k, 74.72.Bk

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 that below a temperature $T^*$ that is higher than $T_c$, a gaplike structure appears in the electronic excitation spectrum. However, at present there is no consensus concerning the relationship between this pseudogap and superconductivity. There are several possibilities which can be crudely divided into two groups: pairing correlations above $T_c$, with a relevant energy scale $k_B(T-T_c)$, and high energy mechanisms, on the scale $k_BT^*$, such as charge or spin gaps. Measurements under strong magnetic fields may help to discriminate between these and to find correlations between them and superconductivity. This idea lead to a series of NMR spin-lattice relaxation rate measurements in high magnetic fields. Recent neutron scattering experiments have also investigated magnetic field effects in the normal state of HTS. NMR may be particularly useful for investigating the energy scale of the pseudogap if performed over a wide range of magnetic fields. In addition, we find that 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 with the different nuclei, copper and oxygen.

Recent NMR experiments that investigate the effect of magnetic field on the pseudogap in nearly optimally doped YBCO include measurement of the $^{63}$Cu(2) NMR spin lattice relaxation rate, $T_1^{-1}$, by Mitrović et al. and Gorny et al. The results of these two papers on $^{63}$Cu(2) $T_1^{-1}$ are contradictory. Gorny et al. reported that the spin-lattice relaxation rate in YBCO$_{7-\delta}$ is magnetic field independent indicative of the opening of a spin-pseudogap. These authors point out that their results are inconsistent with other reports, which concluded that there is a small but significant magnetic field dependence to $(T/T_c)^{-1}$ near $T_c$ in optimally doped material with an interpretation in terms of pairing fluctuations. In the present work we confirm the results of Gorny et al. for copper relaxation and extend this to $^{17}$O(2,3) experiments that give additional insight regarding the onset of superconductivity.

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, $\chi(q, \omega \rightarrow 0) = \chi' + i \chi''$, that varies with q. This indicates that multiple processes of different origin affect $\chi(q,0)$. Based on $^{63}$Cu NMR experiments Gorny et al. pointed out that $\chi''(q,0)$ near $q = (\pi, \pi)$ shows no major field dependence on the scale of 10 T. At this position in the Brillouin zone $\chi$ is strongly enhanced by antiferromagnetic (AF) spin fluctuations, and so this result suggests that the temperature dependence they observe is controlled by a much higher field scale possibly associated with a spin-pseudogap. Our experiments reach similar conclusions. In addition we find from $^{17}$O NMR that $\chi''(q,0)$, away from the $(\pi, \pi)$ point, is magnetic field dependent on the scale of 10 T. Whereas such behavior might be expected near $q = (0,0)$, it is less clear for momenta in the intermediate region between $q = (0,0)$ and $q = (\pi, \pi)$. This field dependence can be explained in terms of superconducting fluctuations, or a pairing pseudogap that opens up $\sim 20$ K above $T_c$. The existence of the pairing pseudogap is compatible with a Fermi-liquid like contribution to the susceptibility. We describe the
II. EXPERIMENT

We have investigated two samples. The first sample, A, has been used in our previous work on spin relaxation and Knight shift. It is a near-optimally doped \( \sim 30-40\% \) \(^{17}\)O-enriched, YBa\(_2\)Cu\(_3\)O\(_{7-\delta}\), aligned powder sample. This sample has a relatively narrow NQR line width of \( \approx 290 \) kHz and was provided courtesy of P. C. Hammel at Los Alamos National Laboratory. Its NQR frequency is \(^{63}\nu_{zz} = 31.5\) MHz. The second sample, B, is a \( \sim 60\% \) \(^{17}\)O-enriched, YBa\(_2\)Cu\(_3\)O\(_{7-\delta}\), aligned powder sample whose NQR line width is \( \approx 450 \) kHz and \(^{63}\nu_{zz} = 31.2\) MHz. After \(^{17}\)O exchange at 550 \(^\circ\)C, this sample was annealed at 390 \(^\circ\)C for a week and consequently might be slightly overdoped. The crystal \( \hat{c} \)-axis of both samples were aligned with the direction of the applied magnetic field, the \( z \)-axis. In Fig. 1 we show the first high frequency satellite of the \(^{65}\)Cu spectra for each of the two samples at 8 T and 95 K. We have checked that the width of these spectra is the same as the NQR linewidth. Low-field magnetization data, for both samples, show a sharp transition at \( T_c(0) = 92.5 \) K. 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: \( \pi/2-\tau-\pi \)-acquire. Our typical \( \pi/2 \) pulse lengths were 1.5 \( \mu \)s, except at 2.1 T where pulse lengths were 2.5 \( \mu \)s, giving us a bandwidth \( > 100 \) kHz.

The spin-lattice relaxation rate was measured using the following sequence: \( \pi/2-\tau_1-\pi/2-\tau-\pi \)-acquire. \( ^{17}T_1^{-1} \) was measured on the first high frequency satellite, i.e. \( (\frac{3}{2} \leftrightarrow \frac{1}{2}) \) 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 \( (\frac{3}{2} \leftrightarrow \frac{1}{2}) \) transition. \( ^{63}T_1^{-1} \) was measured using the same sequence as for measurement of the \(^{17}T_1^{-1} \) except that typical \( \pi/2 \) pulse lengths were 3 \( \mu \)s. All \(^{63}T_1^{-1} \) measurements were made on satellites, \( (\pm \frac{5}{2} \leftrightarrow \pm \frac{3}{2}) \). 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. 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 \(^{65}\)Cu is then inferred from \(^{65}T_1 \) knowing that their ratios scale as the square of their gyromagnetic ratios (\( \gamma \)), namely \( ^{63}T_1 = ^{63}T_1 \ast (^{63}\gamma/^{65}\gamma)^2 = 0.8713 \ast ^{65}T_1 \). 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, listed in Table I, assuming a magnetic relaxation mechanism (only \( \Delta m = \pm 1 \) transitions are allowed).

| Spin | \( \frac{M_\infty - M(t)}{M_\infty} = \) |
|-----|---------------------|
| \( -\frac{5}{2} \leftrightarrow -\frac{3}{2} \) | \( \frac{1}{5} e^{\frac{T}{T_1}} + \frac{2}{5} e^{\frac{T}{T_1}} + \frac{2}{5} e^{\frac{3T}{T_1}} \) |
| \( -\frac{3}{2} \leftrightarrow -\frac{3}{2} \) | \( \frac{1}{5} e^{\frac{T}{T_1}} + \frac{2}{5} e^{\frac{T}{T_1}} + \frac{2}{5} e^{\frac{3T}{T_1}} + \frac{3}{5} e^{\frac{10T}{T_1}} \) |

TABLE 1. Recovery profiles for \( (\pm \frac{5}{2} \leftrightarrow \pm \frac{3}{2}) \) transitions.

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 \( \chi(q, \omega) \), which is the response function entering most theoretical descriptions and is also the quantity experi-
Cu-O coupling and probes $\chi'(q, 0)$ in the intermediate region of the Brillouin zone between $(\pi, \pi)$ 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 $q = 0$, $\chi'(0, 0)$ which we have reported earlier$^{12}$ for sample $A$ 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:

- $^{63}T_1^{-1} \propto \chi''/\omega$ for $q$ near $(\pi, \pi)$,
- $^{63-17}T_{2G}^{-1}_{\text{ind}} \propto \chi'$ for $q$ between $(0, 0)$ and $(\pi, \pi)$,
- $^{17}T_1^{-1} \propto \chi''/\omega$ for $q$ near $(0, 0)$.

Using these tools, we investigate the response of $\chi(q, 0)$ to a magnetic field near $T_c$ to determine which processes affect $\chi$.

IV. $^{63}$T$_1$ RESULTS

In Fig. 2 and 3 we show $^{63}T_1$ for samples $A$ and $B$ respectively. For both samples, we observe no discernible field dependence in the normal state within experimental accuracy of $\pm 2\%$. This result is consistent with that reported by Gorny et al. Above $\sim 100$ 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$^{14}$. The peak in $^{63}(T_1T)^{-1}$ is observed at $T^* \sim 100$ K. Reduction of

\[
\frac{1}{T_1} \propto \lim_{\omega \to 0} \sum_{\mathbf{q}, \alpha' \neq \alpha} \left[ |F_{\alpha'\alpha}(q)|^2 \frac{\chi''_{\alpha\alpha}(\mathbf{q}, \omega_n)}{\omega_n} \right] \quad (1)
\]

where $i$ identifies the nuclear species; $\alpha$ is the direction of $\mathbf{H}_0$ (taken to be parallel to one of the principal axes of the $F_{\alpha'\alpha}$ and $\chi''_{\alpha\alpha}$ tensors); $F_{\alpha'\alpha}(q)$, referred to as a form factor, is the Fourier transform of the hyperfine coupling between nuclei and electrons; and $\chi''_{\alpha'\alpha}(q, \omega)$ is the imaginary part of the dynamic spin susceptibility for the wave vector $q$ and nuclear Larmor frequency, $\omega_n$, with the direction $\alpha'$ perpendicular to $\alpha$.

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. 4 in Appendix A. We see from Eq. (1) that these form factors identify 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 $q = (\pi, \pi)$, the AF wave vector. Since the imaginary part of the susceptibility is peaked at this wavevector, 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 vanishing small form factor at $q = (\pi, \pi)$.

In most solids, the spin-spin relaxation rate arises from the nuclear dipole-dipole interaction. This gives rise to a temperature independent rate, $T_2^{-1}$, and decay of the form $\exp(-t^2/2T_2^2)$ where $(T_2)^{-2}$ is equal to the second-moment of the homogeneous line-shape (excluding the broadening due to the finite lifetime of a spin in an eigenstate). However, nuclei can also interact indirectly via conduction electrons depending on the real part of their magnetic susceptibility. This coupling is an energy conserving process so that it contributes to $T_2$. Thus by measuring $T_2$ one can probe the real part of the electronic susceptibility. The importance of $T_{2G}$ of Cu in obtaining information about the AF exchange between the electronic spins was first pointed out by Pennington et al$^{14}$. Whereas these indirect processes dominate the Cu spin-spin relaxation, they are strongly reduced for oxygen by its vanishing form factor at $q = (\pi, \pi)$. The dominant contribution to oxygen $T_2$ is from direct nuclear dipole-dipole coupling between copper and 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,

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

where $^{17}F_{\alpha'}(q)$ and $^{63}F_{\alpha'}(q)$ are form factors of O and Cu respectively for $\alpha' = c$ for the case $\hat{c} || \hat{z}$. Unlike the case of $^{63}(T_2)^{-1}_{\text{ind}}$ which arises from Cu-Cu indirect coupling and probes $\chi'(q, 0)$ near $(\pi, \pi)$, $^{63-17}(T_{2G})_{\text{ind}}$ arises from

\[ \text{FIG. 2. Spin-lattice relaxation rate of } ^{63}\text{Cu(2) in YBCO sample A as a function of temperature in the magnetic fields of 1.1, 2.4, 8, and 14.7 T.} \]
$^{63}(T_1T)^{-1}$ below $T^*$ has been associated with the loss of low-energy spectral weight which is caused by the opening of a pseudogap. It is interesting to note (for Sample A) that in spite of the fact that $T_c$ decreases with field, $^{63}(T_1T)^{-1}$ falls off independently of the magnetic field, indicating that down to $\sim 63$ limit of as a spin-pseudogap.

The $^{63}(T_1T)^{-1}$ opening of a pseudogap. It is interesting to note (for Sample A) that in spite of the fact that $T_c$ decreases with field, $^{63}(T_1T)^{-1}$ falls off independently of the magnetic field, indicating that down to $\sim 80$ K the low frequency limit of $\chi''(q, \omega)/\omega$ for $q = (\pi, \pi)$ is not sensitive to superconductivity and is dominated by a process with a high energy scale that becomes gapped and which we refer to as a spin-pseudogap.

For sample B the maximum value of $^{63}(T_1T)^{-1}$ is shifted to slightly lower values and higher temperatures compared to sample A. In the superconducting state, $^{63}(T_1T)^{-1}$ decreases with decreasing temperature with slight magnetic field dependence, not as much as expected from $T_c$ reduction by the field.

In a previous report[1] we inferred $^{63}T_1$ from $^{17}T_2$ invoking the theory of Recchia et al.[12] where we found significant magnetic field dependence of $^{63}T_1$. We now believe that this interpretation of our $^{17}T_2$ experiment is incorrect as we will discuss in the next section.

V. $^{63}T_1$ FROM $^{17}T_2$ AND CU-O INDIRECT COUPLING

Walstedt and Cheong[13] proposed that the main source of spin-echo decay of $^{17}O$ is the copper spin-lattice relaxation. The z-component fluctuating fields from copper nuclear spin flips are transferred to the oxygen nuclei by Cu-O nuclear dipolar interactions. To account for this process Recchia et al.[12] derived an expression for $^{89}Y$ and $^{17}O$ spin echo height, $M(\tau)$, as a function of pulse spacing $\tau$. In order to fully account for the $^{17}O$ spin-echo decay two additional mechanisms have to be invoked. First is a Redfield contribution[14] to the spin-echo decay, caused by the finite lifetime of a spin in an eigenstate as a result of the $^{17}O$ spin-lattice relaxation. This contribution is $\sim 15\%$ at $T_c$ and can be evaluated using the measured $^{17}T_1$. The Redfield contribution is taken to be magnetic field independent and, even if we introduce a weak magnetic field dependence to this contribution, our fit results do not significantly change. The second contribution is an indirect Cu-O nuclear coupling, $k$, mediated by the conduction electrons[15]. This effect was assumed by Recchia et al.[12] and ourselves[13] to be a temperature and field independent enhancement of the effective Cu-O dipolar coupling strength. The following expression from Recchia et al.[12] gives the $^{17}O$ spin echo height, $M(\tau)$ as a function of pulse spacing $\tau$,

\[
M = M_0 e^{\left[\frac{15}{2}k^2 \sum_{i=1}^{12} \left(\frac{63.65}{\chi_i^2} (1-3\cos^2 \theta_i)\right)^2 \frac{1}{r_i} \left(T_1^{(1)}\right)^2 \left(2\tau/T_1^{(1)} + 4e^{-\tau/T_1^{(1)}} + e^{-2\tau/T_1^{(1)}} - 3 - 2\tau/T_2R\right)e^{-\tau/T_2R} \right]}
\]

(3)

In our earlier work we performed a nonlinear least squares fit of our data to Eq. (3) with $^{63}T_1$ as a fitting parameter choosing $k = 1.57$ to match the high temperature results. The sum was performed over all Cu neighbors in a radius of 12 A; $r_i$ is the Cu-O distance; $\theta_i$ is the angle between the applied field and the Cu-O axis; $T_1^{(i)}$ is $T_1$ of the $i$th copper nucleus; $I = 3/2$ is the copper nuclear spin; and $T_{2R}$ is the Redfield contribution to the rate.

We now examine in more detail the relaxation described by the parameter $k$. We can extract that 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 using Eq. (3) with $k$ set to 1. We take into account the relaxation from unmediated Cu spin-flips using our direct measurements of $^{63}T_1^{-1}$. We then approximately fit the residual decay with a gaussian function of time and show the resulting relaxation times in Fig. 4 versus temperature for magnetic fields from 2.1 to 22.8 T. There is a well-defined field dependence for $T < 120$ K. The qualitative behavior of the rate resembles the behavior observed in the $^{17}O$ Knight shift[16] a quantity proportional to $\chi'(0,0)$, although we do not intend special significance by this comparison. Above $T_c$, $T > 90$ K, there is a small but
clear dependence on field as reported earlier by Mitrović et al. using a different interpretative framework. The relatively low field scale for this dependence, in contrast to \(63(T_1 T)^{-1}\) in Fig. 2 and 3 suggests a connection to superconductivity, most likely from pairing fluctuations.

Below \(T_c\), the spin-spin relaxation rates shift 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 \(T = 0.9 T_c(H)\) the rate drops by \(\sim 20\%\) from its value at \(T_c(H)\). For higher applied fields the decrease is smaller. Superconductivity can affect \(T_2\) data in two ways: through vortex vibrations, whose precise contribution to the rate is not known; and, through the suppression of \(\chi'\) due to pair-formation. We have shown in previous work that vortex vibrations give rise to a sharp increase in spin-spin relaxation with a lorentzian spectral density that onset at the vortex melting transition (at least in low fields, \(H < 10 T_c\)). So it seems unlikely that vortex vibrations are primarily responsible for the field dependence we report in Fig. 3 at relatively higher temperatures. To study the effect of pairing on suppression of the indirect interaction we have calculated both the temperature dependence of \(\chi'\) and \((63^{-1} T_{2G}^{-1})_{ind}\) in the superconducting state following the RPA-like approach of Balut and Scalapino. We take an RPA form for \(\chi\):

\[
\chi(q, \omega) = \frac{\chi_0(q, \omega)}{1 - J_0 \chi_0(q, \omega)},
\]

where \(J_0 = -J_0 (\cos q_x + \cos q_y)/2\) is an enhancement factor. The value of \(J_0\) was constrained by matching \((63^{-1} T_{2G}^{-1})_{ind}\) experiments from Cu NQR, measurements that are unaffected by vortices since they are at zero field. Below \(T_c\), the measured Cu-Cu indirect coupling contribution to relaxation \((63^{-1} T_{2G}^{-1})_{ind}\), decreases by \(\sim 10 - 20\%\) of its value at \(T_c\). We have reproduced this result with our calculation as shown in Fig. 4. We find that the reduction of \((63^{-1} T_{2G}^{-1})_{ind}\) using the same parameters for \(\chi(q)\) and we find that in the superconducting state it decreases by \(\sim 5 - 10\%\) of its value at \(T_c\), as shown in Fig. 4. It is conceivable that pairing fluctuations could modify this rate giving raise to field dependence above \(T_c\). However, this effect is probably too small to account for the observed field dependence shown in Fig. 4.

**FIG. 4.** Spin-spin relaxation rate of \(^{17}\text{O}(2,3)\) (sample A) after dividing out the part of the relaxation coming from the direct Cu-O dipolar interaction as described in the text. The solid line is the calculated spin-spin relaxation from Cu-O indirect coupling using the same susceptibility parameters as we used to calculate \((T_1 T)^{-1}\) and \((63(T_1 T)^{-1}\) discussed later in Sec. VI.

**FIG. 5.** Calculated \((63^{-1} T_{2G}^{-1})_{ind}\), arising from Cu-Cu indirect coupling, (open circles) and \((63^{-1} T_{2G}^{-1})_{ind}\), arising from Cu-O indirect coupling, (open squares) as a function of the reduced temperature, \(T/T_c\), for a \(d\)-wave superconductor and \(J_0 = 10\).

Well above \(T_c\) we see in Fig. 4 that \((63^{-1} T_{2G}^{-1})_{ind}\) decreases as a function of decreasing temperature. This appears to be in contrast with our calculation of the rate using a phenomenological form for the susceptibility as discussed in the next section. Nonetheless the magnitude of the observed effect is similar to the calculation. One might also argue that 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 which 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. They showed in YBa\(_2\)Cu\(_4\)O\(_8\) 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. In addition they found evidence that these fluctuations are associated with superconductivity. It might be that these fluctuations also have spin character and are sensitive to the magnetic field providing a possible channel for spin-spin relaxation.

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.4

VI. $^{17}T_1$ RESULTS

As previously pointed out $^{17}(T_1)^{-1}$ probes the imaginary part of electronic spin susceptibility, $\chi''(q,0)$, close to $q = 0$. In Fig. 6 we show the $^{17}O$ spin-lattice relaxation rate, of the $A$ sample, as a function of temperature in different magnetic fields. We find that the rate increases with increasing magnetic field, on the scale of $10^3$ in different magnetic fields. We find that the rate drops sharply in the superconducting state, consistent with previous assumptions.4

Our Knight shift data6 indicate a $T_c$ shift of $\sim 2$ K from 3.2 to 8 T. However, $^{17}(T_1)^{-1}$ has a value of 0.367 (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 analysis12,24. As the magnetic field increases it suppresses the negative DOS pairing fluctuation contribution to the rate causing the overall rate to increase with increasing field, as observed in Fig. 6. Once the fluctuations are completely suppressed by the field, we expect the rate to drop sharply at $T_c(H)$. However, we observe that the field dependence of the rate saturates around 10 T and that at “high” field, $H \geq 10$ T, it has well-defined curvature near $T_c$ indicating that DOS pairing fluctuations cannot be the only process affecting $^{17}(T_1)^{-1}$. In the following we try to model the influence of a spin-pseudogap on $^{63}(T_1)^{-1}$ and, with the same parameters, estimate the effect of $^{17}(T_1)^{-1}$.

We take the MMP phenomenological expression for the dynamical susceptibility, altered so as to include the incommensurations in the susceptibility peaks at $Q_i = (\pi \pm \delta, \pi \pm \delta)$ AF wave vector,24

$$\chi(q, \omega) = \chi_{AF} + \chi_{FL} = \frac{1}{4} \sum_i \frac{\alpha \xi^2 q^2}{1 + \xi^2 (q - Q_i)^2 - i \omega / \omega_{SF}} + \frac{\chi_0}{1 - i \pi \omega / T}. \quad (5)$$

where $\xi$ is the spin fluctuation correlation length in units of the lattice constant $a$, $\alpha$ is a scaling factor, $\omega_{SF}$ the frequency of spin fluctuations, and $\xi_0$ and $\Gamma$ are terms added to describe the Fermi-liquid background for AF fluctuations.

The imaginary part of $\chi(q, \omega)$ divided by frequency in the limit of $\omega \rightarrow 0$ is given by,

$$\lim_{\omega \rightarrow 0} \chi''(q, \omega) / \omega = \frac{1}{4} \sum_i \frac{\alpha \xi (T)^2 q^2 / \omega_{SF}}{[1 + \xi (T)^2 (q - Q_i)^2]^2} + \frac{\chi_0 \pi}{T}. \quad (6)$$

The rate divided by the temperatures, for $H_0||\hat{c}$ is then evaluated by summing the product of the form factor and $\chi''$ over all $q$,

$$\frac{1}{T_1 T} = \lim_{\omega \rightarrow 0} \frac{k_B}{2 \mu_B^2 h^2 \sum_q F_c(q) \frac{\chi''(q, \omega)}{\omega}}. \quad (7)$$

We take Shastry-Mila-Rice22 form factors given in Eq. 11. In addition, $\omega_{SF}$ is assumed to be proportional to $\xi(T)^{-2}$ and that $\xi(T) = \xi_0 |T_c/(T_c + T)|^{1/2}$. Temperature dependence of $\xi(T)$, $\omega_{SF}$, and other parameters were determined so that both calculated $^{17}(T_1)^{-1}$ and $^{17}(T_1)^{-1}$ coincide with our data. Assuming that $Q_{AF} = (\pi \pm 0.1, \pi \pm 0.1)$ we find the following values for the parameters used to calculate $(T_1)^{-1}$: $\xi(T) = 3.07 [114 K/(114 K + T)]^{1/2}$, $\omega_{SF} = 6.09 \ast \xi(T)^{-2}$ meV, $\alpha = 14.8$ (eV)$^{-1}$, and for the Fermi liquid part, $\chi_0 \pi / \mu_B^2 h_\gamma = 8.885$ eV$^{-2}$.

We obtain values of $(T_1 T)^{-1}$, for both $^{17}O$ and $^{63}Cu$, shown as the solid curve (extending to dashed below 120

![FIG. 6. Spin-lattice relaxation rate of $^{17}O(2,3)$ in YBCO (sample $A$) as a function of temperature in the magnetic fields of 3.15, 8, 13.7, and 22.92 T.](image-url)
We notice that $^{17}(T_1 T)^{-1}$ increases slightly with decreasing temperature similar to $^{63}(T_1 T)^{-1}$ 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.

We then model the opening of the pseudogap by assuming that it only affects $\omega_{SF}$. We take a phenomenological form for $\omega_{SF}^{-1} \propto (\tanh((T - T_p)/c_1))([\xi(T)^2])$, 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 $^{63}(T_1 T)^{-1}$, giving the solid curve in Fig. 7b below $T \approx 100$ K. Using exactly the same pseudogap parameterization, we calculate $^{17}(T_1 T)^{-1}$ giving the corresponding solid curve in Fig. 7a. We clearly see that the suppression of $^{63}(T_1 T)^{-1}$ due to the opening of the spin pseudogap, as modeled here, will also cause a small suppression of $^{17}(T_1 T)^{-1}$ that reproduces the observed curvature of the high field $^{17}(T_1 T)^{-1}$ data near $T_c$. From our simplistic model we have shown phenomenologically that $^{17}(T_1 T)^{-1}$ 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 have also calculated the rates using the oxygen form factor suggested by Zha et al. Eq. II. Results similar to the ones shown in Fig. 7 were obtained using the following parameters: $\xi_0 = 2.98$, $T_x = 105$ K, $\omega_{SF} = 5.6 \cdot \xi(T)^2$ meV, $\chi_0 \pi/\mu_B^2 h\Gamma = 13.835$ eV$^{-2}$ $c_1 = 17$ K, $T_p = 69.5$ K.

VII. DISCUSSION AND SUMMARY

We summarize our relaxation experiments by showing the relative effect of magnetic field on $\chi(q, 0)$ for sample A. This can be conveniently represented by $R(H)$ defined as $R(H) = ((T_{1,2})_{n=1}^1 - (T_{1,2})_{n=0})/\langle(T_{1,2})_{n=1}^1$, where the normal-state rate, $(T_{1,2})_{n=1}^1$, is a fit to the field independent high temperature behavior ($T > 120$ K) of the appropriate rate. The results at $T = 95$ K are given in Fig. 8. The two upper graphs indicate that both real and imaginary parts of the spin susceptibility away from $q = (\pi, \pi)$ have magnetic field dependence above $T_c$ on the scale of 10 T. This field dependence is likely caused by superconducting pair fluctuations that can be attributed to the field induced suppression of the negative contribution to the rate from the density of states.
The latter can be expected since the superconducting fluctuations as a precursory effect of thermodynamic critical field, \( \mathcal{H}_c \). This is consistent with the fact that Zeeman contributions to the excitation spectrum from spin are necessarily much less than \( \mathcal{H}_c \). For \( \mathcal{H}_c \) less than the inverse of the superconducting coherence length, \( \xi(\mathcal{H}_c) \), the susceptibility is influenced by superconducting fluctuations. This is consistent with a Fermi-liquid like behavior in which the susceptibility is suppressed by superconducting fluctuations for \( \mathcal{H}_c \) less than the inverse of the superconducting coherence length. The magnetic field behavior of \( \chi(q,0) \) indicates the coexistence of two pseudogaps of different origins. One pseudogap dominating \( \chi(q,0) \) near \( q = (\pi, \pi) \) is insensitive to magnetic fields in our experimental range \( \mathcal{H}_c \). This insensitivity indicates that this pseudogap is not intimately tied to superconductivity and that its possible origin is on a high energy scale which we call a spin-pseudogap. This nomenclature is motivated by the fact that Zeeman contributions to the excitation spectrum from spin are necessarily much less than \( k_B T_c \) in the range of experiments we discuss here. The second pseudogap, evident in \( \chi(q,0) \) away from \( q \sim (\pi, \pi) \) 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 by the thermodynamic critical field, \( H_c \).

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

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APPENDIX A: FORM FACTORS

The form factors, relevant for this work, are Shastry-Mila-Rice\(^a\) form factors given by,

\[
\begin{align*}
(63)^{F_a} & = [A_{ab} + 2B(\cos q_x + \cos q_y)]^2 \\
(63)^{F_{eff}} & = [A_c + 2B(\cos q_x + \cos q_y)]^2 \\
17F_b & = 2C^2[\cos(q_x/2) + \cos(q_y/2)]^2 \\
17 - (63)^{F_c} & = 63 F_{eff} F_b
\end{align*}
\]

where \( A_{ab} = 0.84B \), \( A_c = -4B \), \( C = 0.91B \), and \( B = 3.82 \times 10^{-7} \) eV. Their \( q \)-dependence is shown in Fig. 10 along with the imaginary part of the susceptibility which is dominated by AF-spin fluctuations.

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 \( q = (\pi, \pi) \) antiferromagnetic spin fluctuations, insensitive to superconducting fluctuations, dominate the spin susceptibility. In the region away from \( q = (\pi, \pi) \) the susceptibility is influenced by superconducting fluctuations. This is consistent with a Fermi-liquid like behavior in which the susceptibility is suppressed by superconducting fluctuations for \( q \) less than the inverse of the superconducting coherence length. The magnetic field behavior of \( \chi(q,0) \) indicates the coexistence of two pseudogaps of different origins. One pseudogap dominating \( \chi(q,0) \) near \( q = (\pi, \pi) \) is insensitive to magnetic fields in our experimental range \( \mathcal{H}_c \). This insensitivity indicates that this pseudogap is not intimately tied to superconductivity and that its possible origin is on a high energy scale which we call a spin-pseudogap. This nomenclature is motivated by the fact that Zeeman contributions to the excitation spectrum from spin are necessarily much less than \( k_B T_c \) in the range of experiments we discuss here. The second pseudogap, evident in \( \chi(q,0) \) away from \( q \sim (\pi, \pi) \) 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 by the thermodynamic critical field, \( H_c \).

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\[\begin{align*}
\text{FIG. 9. Field dependence of the relaxation } R(H) =
\end{align*}\]
The exact form of the oxygen form factor is not generally accepted. In order to assure a near perfect cancellation of the influence of the incommensurate spin-fluctuation peaks (observed by neutron scattering) on the $^{17}$O relaxation rates, Zha et al. suggested that the oxygen form factor should be altered to include coupling of $^{17}$O nuclei to both nearest-neighbor and next-nearest-neighbor Cu$^{2+}$ spins. In this case the oxygen form factor, $F_r$, includes extra terms which they take to be
eq 0.25, \zeta_\perp = 0.91, \text{ and } \zeta_\parallel = 1.42.\)

\begin{align}
\frac{2C^2}{(1 + 2r_c)} \{ & \cos(q_\parallel a/2)^2 \zeta_\parallel (1 + 2r) - 2r + 2 \cos(q_\perp a) \}^2 \\
& + \cos(q_\parallel a/2)^2 \zeta_\parallel (1 + 2r) - 2r + 2 \cos(q_\perp a) \}^2, \quad (A2)
\end{align}

where $r \equiv 0.25$, $\zeta_\perp = 0.91$, and $\zeta_\parallel = 1.42$. 

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