Superconducting Fluctuation Effects on the Spin-Lattice Relaxation Rate in $YBa_2Cu_3O_{6.95}$

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We report ^{63}Cu(2) spin-lattice relaxation rate measurements of $YBa_2Cu_3O_{6.95}$ in magnetic fields from 2.1 to 27.3 T obtained from ^{17}O(2, 3) nuclear magnetic resonance spin-spin relaxation. For $T < 120$ K, the spin-lattice rate increases with increasing magnetic field. We identify this magnetic field dependence with the change in the low-energy spectral weight originating from d-wave pairing fluctuation corrections to the density of states. [S0031-9007(99)08771-2]

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Nuclear magnetic resonance has played an important role in elucidating the nature of high-T_c superconductivity [1]. In the normal state of many high-T_c superconductors an increase in the nuclear spin-lattice relaxation rate divided by the temperature, $1/T_1T$, of planar Cu with decreasing temperature has been attributed to antiferromagnetic (AFM) spin fluctuations [2]. At lower temperatures, in the superconducting state, the rate of planar Cu decreases strongly with decreasing temperature as the gap in the quasiparticle spectrum develops. The crossover from normal to superconducting behavior occurs around 100 K, substantially above the transition temperature of optimally doped $YBa_2Cu_3O_{7-\delta}$(YBCO). We have investigated this crossover experimentally and theoretically. We show that the crossover can be understood quantitatively in terms of pairing fluctuation corrections to the spin-lattice relaxation rate in optimally doped YBCO.

Because of their large anisotropy and small coherence lengths, the onset of superconductivity in high-T_c materials is preceded by the effects of strong superconducting fluctuations on the normal-state properties, including the specific heat [3], diamagnetism [4], nuclear spin-lattice relaxation rate [5–8], and Pauli susceptibility [6,8,9]. Here we report on the field dependence of $1/T_1T$ of planar copper, ^{63}Cu(2), in optimally doped YBCO. We find that below 120 K the relaxation rate increases with increasing field with a typical field scale of 10 T. We quantitatively account for this behavior in terms of pairing fluctuations with d-wave symmetry [8].

Our aligned powder sample of 30%–40% ^{17}O-enriched $YBa_2Cu_3O_{6.95}$ has been investigated previously [9–13]. Our measurements cover the temperature range 70 to 160 K over a wide range of magnetic fields, from 2.1 to 27.3 T. 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. In order to study planar copper nuclear spin-lattice relaxation, we take advantage of its direct effect on the ^{17}O(2, 3) NMR spin-spin relaxation which we can accurately measure using a Hahn echo sequence: $\pi/2$-τ-π acquire. Our typical $\pi/2$ pulse lengths were 1.5 μs, giving us a bandwidth >100 kHz. After the $\pi/2$ pulse, the precessing nuclear spins dephase because of variations in the z component of the magnetic field in the sample. The dephasing from static processes is recovered after the π pulse, leaving the echo intensity to be determined predominantly by copper spin-lattice relaxation, as has been recently demonstrated [14]. The ^{17}O (2, 3) ($1/2 \leftrightarrow -1/2$) resonance has a low frequency tail owing to oxygen deficiency in a small portion of the sample [11]. Its effect on our measurements can be eliminated by performing a nonlinear least squares fit in the frequency domain for each echo, a method similar to that of Keren *et al.* [15]. The oxygen resonance is much narrower than that of copper (by a factor of 6 at 8.4 T) and thus ^{17}O NMR is more favorable for our experiments. This is particularly true for the high field experiments, $H_0 > 15$ T, performed in a Bitter magnet at the National High Magnetic Field Laboratory in Tallahassee, Florida. The measurements for $H_0 \leq 14.8$ T were obtained with superconducting magnets. The reliability of this technique for measuring T_1 of ^{63}Cu(2) was tested by comparison with direct measurements of T_1 performed on the same sample.

We extract T_1 of ^{63}Cu(2) from ^{17}O(2, 3) spin-spin relaxation data following the proposal of Walstedt and Cheong [16] that the dominant mechanism for spin-echo decay of ^{17}O is the copper spin-lattice coupling. 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.* [14] derived an expression for the ^{17}O spin echo height, $M(\tau)$, as a function of pulse spacing τ,

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$$M = M_0 \exp\left\{ -{}^{17}\gamma^2 k^2 \sum_{i=1}^{\nu} \left[\frac{{}^{63,65}\gamma \hbar}{r_i^3} (1 - 3\cos^2\theta_i) \right]^2 \frac{I(I+1)}{3} (T_1^{(i)})^2 \times [2\tau/T_1^{(i)} + 4e^{-\tau/T_1^{(i)}} - e^{-2\tau/T_1^{(i)}} - 3] - 2\tau/T_{2R} \right\}. \quad (1)$$

We performed a nonlinear least squares fit of the data to Eq. (1) in the range $50 < \tau < 350$ μs, with T_1 of ^{63}Cu(2) as a fitting parameter. The sum was performed over all Cu neighbors in a radius of 12 Å; r_i is the Cu-O distance; θ_i is the angle between the applied field and the Cu-O axis; $T_1^{(i)}$ is T_1 of the ith copper nucleus; $I = 3/2$ is the copper nuclear spin; k is an enhancement factor due to the Cu-O indirect coupling which we determine to be 1.57; and T_{2R} is the Redfield contribution to the rate. An example of the fit is presented in the inset of Fig. 1, at 19 T and 95 K, and is compared with the measured relaxation profile. The fit to Eq. (1) is sufficiently accurate that we can rely on its systematic behavior. We have also compared our data with direct measurements of $1/T_1T$ of ^{63}Cu(2) taken from earlier work [7,13,17] for several magnetic fields, as shown in Fig. 3 (below). The measurement at 7.4 T was performed on our sample [13].

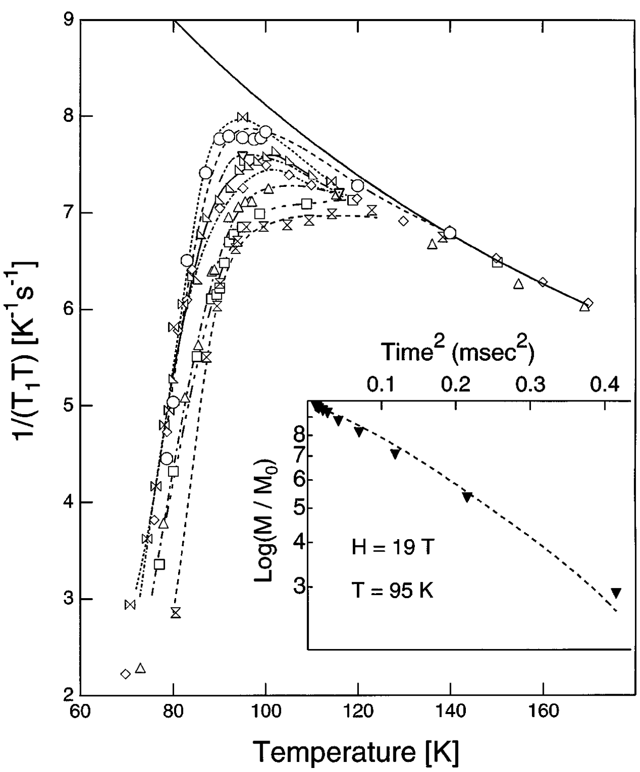

FIG. 1. Spin-lattice relaxation rate of ^{63}Cu(2) in YBCO as a function of temperature for the following fields: 27.3 T (⋈), 22.8 T (○), 19 T (▽), 14.8 T (⊾), 8.4 T (◇), 5.9 T (△), 3.2 T (□), 2.1 T (⧖). Dashed lines are guides to the eye. The solid line is a fit to $(T_1T)_n^{-1} \propto T_x/(T + T_x)$, $T_x = 103$ K [2]. Inset: Spin-spin relaxation of ^{17}O NMR at 19 T, $T = 95$ K, and a fit to Eq. (1).

Our results for $1/T_1T$ are presented in Fig. 1. Above 120 K, there is no discernible field dependence within experimental accuracy of $\pm 2\%$. However, near the peak in $1/T_1T$, we find that the rate increases with increasing magnetic field. At 95 K, the rates at 2.1 and 27.3 T differ by 17%. The peak in $1/T_1T$ versus T shifts toward lower temperature as the field increases and the rate drops sharply in the superconducting state, consistent with reduction of T_c by the field [9]. We show below that pairing fluctuations are in quantitative agreement with this behavior, and that a purely magnetic mechanism with a spin pseudogap is difficult to reconcile with the field scale.

In underdoped materials, the temperature dependence of the Knight shift, $K(T)$, and the peak in $1/T_1T$ has been associated with the opening of a spin pseudogap [18] in the spin excitation spectrum below a temperature $T^* > 100$ K. The temperature scale T^* was suggested to be a rough measure of the pseudogap, with a magnetic field scale of $H^* = k_BT^*/\mu_B$, ≥ 140 T. This exceeds by far the field scale of $\sim$10 T that we observe in $1/T_1T$ in our optimally doped sample. The large field scale, $H^* \gg 10$ T, for a spin pseudogap is consistent with recent neutron scattering measurements that show that the resonance peak of optimally doped YBCO remains almost unaffected in a field of 11.5 T [19].

In high-T_c materials, superconducting fluctuations are expected to have a significant effect on $1/T_1$ near T_c. Diamagnetic fluctuations do not play a role in our measurements of T_1 since they alter the magnetic field mainly along the axis parallel to the applied field; only transverse fields contribute to relaxation of the z component of the nuclear spin. The pairing fluctuation contributions to the rate result from fluctuation corrections to the density of states (DOS) and from the Maki-Thompson (MT) corrections to the local dynamical susceptibility. The corresponding Feynman diagrams for these corrections are shown in Fig. 2. The propagators and vertices are defined below and in Ref. [8]. The pairing fluctuation correction is sensitive to the symmetry of the order parameter fluctuations because of the difference in sign of the MT (positive) and DOS (negative) corrections, and because of the sensitivity of the non-s-wave pairing fluctuations to disorder. In the case of s-wave pairing fluctuations, the dominant contributions to the rate come from the positive MT processes [5], which are insensitive to nonmagnetic disorder. A magnetic field suppresses the MT and DOS contributions, and leads to a suppression of the rate for s wave. In the case of d-wave pairing, the field dependence of $1/T_1$ is reversed compared to that for s-wave pairing. Scattering by nonmagnetic disorder leads to strong

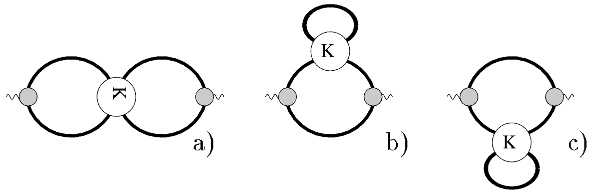

FIG. 2. Pairing fluctuation corrections, to leading order in T_c/E_F, where E_F is the Fermi energy, for the nuclear spin-lattice relaxation rate. (a) is the Maki-Thompson process; (b) and (c) are the density of states corrections to the rate. K is the impurity-renormalized pair fluctuation propagator.

suppression of the MT corrections for d-wave fluctuations. The DOS corrections survive nonmagnetic scattering, but are suppressed by a magnetic field leading to an increase in $1/T_1$ with increasing field, even for modest levels of disorder. As we show below, our results provide a consistent and quantitative account of the field dependence of the nuclear spin-lattice relaxation rate above T_c.

In order to make a quantitative comparison between the leading order pairing fluctuation corrections and the experimental field dependence of the rate, we isolate the fluctuation corrections to the experimental rate by writing $(T_1T)^{-1}_{\text{tot}} = (T_1T)^{-1}_n + \delta(T_1T)^{-1}$, where the normal-state rate is fit to the AFM Fermi-liquid model [2], $(T_1T)^{-1}_n \propto T_x/(T + T_x)$. We obtain $T_x = 103$ K from a fit to high temperature data at 8.4 T. The fluctuation contributions are indicated by $\delta(T_1T)^{-1}$. These values, normalized by $(T_1T)^{-1}_n$, are plotted in Fig. 3 as a function of magnetic field at 95 K along with our theoretical calculations of the pairing fluctuation corrections.

The calculations of $1/T_1$ assume a quasi-2D cylindrical Fermi surface, with an isotropic in-plane Fermi velocity $\vec{v}_f$. We expect the pairing fluctuations to be predominantly 2D in a magnetic field because of Landau-level quantization. A summary of the calculation is provided here; more details can be found in Ref. [8]. The pairing interaction is $V(\vec{p}, \vec{p}\,') = \eta(\vec{p}) g \eta(\vec{p}\,')$, where $\eta(\vec{p})$ is the normalized pairing amplitude; for s-wave pairing $\eta(\vec{p}) = 1$ while for d-wave pairing $\eta(\vec{p}) = \sqrt{2}\cos 2\psi$, where ψ is the angle between the crystallographic $\hat{a}$ axis and $\vec{p}$.

The pair fluctuation propagator is defined in terms of the sum over ladder diagrams in the particle-particle interaction channel; the propagator factorizes into $\eta(\vec{p})L(Q)\eta(\vec{p}\,')$, where $L(Q)^{-1} = g^{-1} - T\sum_{\epsilon_n} B_2(\epsilon_n, Q)$, $B_2(\epsilon_n, Q) = \sum_{\vec{p}} \eta(\vec{p})\tilde{\eta}(P, Q)G(P)G(Q - P)$, and $G(P)$ is the quasiparticle Green's function. We use a shorthand notation: $P \equiv (\epsilon_n, \vec{p})$, $P' \equiv (\epsilon_{n'}, \vec{p}\,')$ for fermion quasiparticles, and $Q \equiv (\omega_l, \vec{q})$ for bosonic Matsubara energy and pair momentum of the fluctuation modes; the pair momentum, $\vec{q}$, is quantized because of orbital quantization in a magnetic field. We include disorder via the standard averaging procedure for dilute impurity concentrations [20]. Impurity scattering introduces an elastic scattering time in the quasiparticle

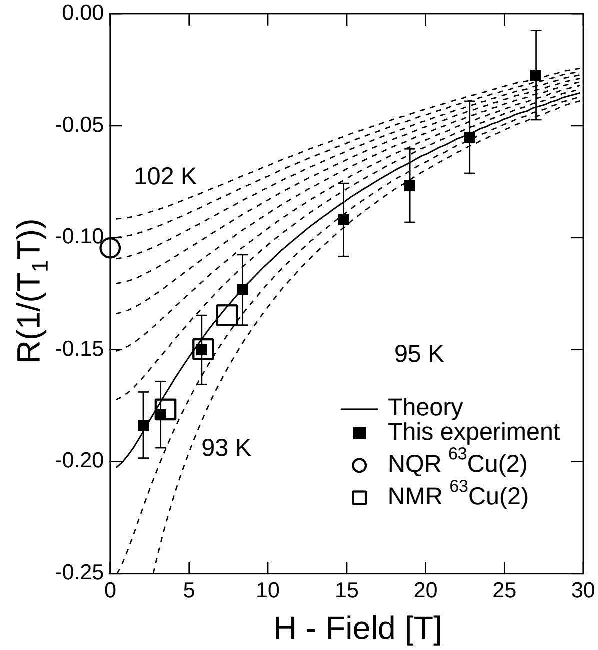

FIG. 3. Fluctuation contribution $R(1/(T_1T)) = \delta(T_1T)^{-1}/(T_1T)^{-1}_n$ of ^{63}Cu(2) spin-lattice relaxation rate as a function of magnetic field at 95 K. The dashed curves are d-wave calculations for temperatures ranging from 93 to 102 K in increments of 1 K. The solid curve is calculated for 95 K. The open circle is $R(1/(T_1T))_{NQR}$ at 95 K [17]. The open squares are from direct measurements of the ^{63}Cu(2) T_1 at 3.5 T by Y.-Q. Song [17], 5.9 T by Carretta *et al.* [7], and 7.4 T by Hammel *et al.* [13].

Green's function, $G(P) = [i\epsilon_n - \Sigma(\epsilon_n) - \xi(\vec{p})]^{-1}$, where $\xi(\vec{p}) = \epsilon(\vec{p}) - \mu$ is the quasiparticle excitation energy, $\Sigma(\epsilon_n) = -(i/2\tau + i/2\tau_\phi)\,\text{sign}(\epsilon_n)$ is the self-energy, and τ is the elastic scattering lifetime. We include inelastic scattering through the lifetime τ_ϕ. Impurity scattering modifies the fluctuation propagator directly through a vertex correction in the particle-particle channel, $\tilde{\eta}(P, Q) = \eta(\vec{p}) + \sum_{\vec{p}} \eta(\vec{p})G(P)G(Q - P)C(\epsilon_n, Q)$, where $C(\epsilon_n, Q)^{-1} = \tilde{\alpha}^{-1} - \sum_{\vec{p}} G(P)G(Q - P)$ is an impurity Cooperon-like propagator, $\tilde{\alpha} = 1/2\pi\tau N_F$ is the impurity scattering vertex, and N_F is the density of states at the Fermi level. The full impurity-renormalized pair propagator, $K(P, P', Q)$, which enters the dynamical susceptibility diagrams shown in Fig. 2, is given by $\tilde{\eta}(P, Q)L(Q)\tilde{\eta}(P', Q)$. The leading order fluctuation correction to $1/T_1$ then follows from the Feynman rules for evaluating the diagrams [20] and is given by

$$\delta\chi_M(\omega_m) = -2|\vec{A}|^2 \sum_{n,Q} B_1(\epsilon_n, Q)B_1(\epsilon_n - \omega_m, Q)L(Q),$$

$$\delta\chi_D(\omega_m) = 4|\vec{A}|^2 \sum_{n,Q} G_1(\epsilon_n - \omega_m)\frac{\delta B_2(\epsilon_n, Q)}{\delta\Sigma(\epsilon_n)} L(Q), \tag{2}$$

$$\delta(T_1T)^{-1} = \lim_{\omega\to 0} 2\,\text{Im}\,\frac{\delta\chi_M(\omega) + \delta\chi_D(\omega)}{\omega}, \tag{3}$$

with $B_1(\epsilon_n, Q) = \sum_{\vec{p}} \tilde{\eta}(P,Q)G(P)G(Q-P)$, $G_1(\epsilon_n) = \sum_{\vec{p}} G(P)$, and $|\vec{A}|^2$ are momentum-averaged hyperfine form factors [8]. We analytically continue Eqs. (2) to real energies using Eliashberg's technique [21] to obtain $\delta\chi_M(\omega)$ and $\delta\chi_D(\omega)$. The zero frequency limit in Eq. (3) is performed analytically and the resulting equations are evaluated numerically. The sum over Q includes a summation over all Landau levels and over all dynamical fluctuation modes.

The experimental zero-field transition temperature of 92.5 K determines the temperature scale for the theoretical calculations. The mean-field transition temperature, $T_c(8.4\ \mathrm{T}) = 80.9 \pm 0.3$ K, which is determined by the divergence of the pair fluctuations, is obtained from our fit to spin susceptibility [9]. We assumed $\hbar/2\pi\tau_\phi = 0.02k_BT_c$ and $\hbar/2\pi\tau = 0.2k_BT_c$, and there is one fitting parameter for the overall scale of the fluctuation contributions to $1/T_1$. Our theoretical calculation for the field dependence of the fluctuation correction is shown in Fig. 3 for d-wave pairing. The rate increases because of the suppression of the (negative) DOS contribution to the rate by the magnetic field. The results agree quantitatively with the experimental data at $T = 95$ K and provide strong evidence for d-wave pairing fluctuations. For s-wave pairing, the calculated rate (not shown) *decreases* with increasing magnetic field because of the suppression of the (positive) MT term.

Carretta *et al.* [7] reported experimental evidence for a positive contribution to the rate that was attributed to the MT process. These authors compared nuclear quadrupolar resonance (NQR) relaxation measurements and NMR relaxation at 5.9 T and found an NQR rate that is higher than the NMR rate in a range of $\sim$10 K above T_c, a result which is similar to our NQR measurement shown in Fig. 3. They interpret the decrease from the higher NQR to the lower NMR rate at 5.9 T in terms of s-wave pairing fluctuations, which implies a dominant MT term. However, our data in Fig. 3 shows that there is no significant MT contribution to the NMR rate at fields above 2.1 T. Our analysis of the field dependence of the data is in excellent agreement with the theory of d-wave pairing fluctuations, and disagrees with the theory based on s-wave fluctuations. Possible explanations for the apparent discrepancy between the NQR rate and the low-field NMR rate include an admixture of s-wave and d-wave fluctuations induced by orthorhombic anisotropy [8], and the 2D to 3D crossover regime at low fields.

In summary, we have determined the ^{63}Cu(2) spin-lattice relaxation rate as a function of magnetic field from 2.1 to 27.3 T. We found that $1/T_1T$ increases with increasing field in the temperature range $T < 120$ K, which we can account for quantitatively with the theory of d-wave pairing fluctuations in 2D. Our results are consistent with d-wave pairing in YBCO, and inconsistent with dominant s-wave pairing. We found that the characteristic field scale for the suppression of the fluctuation corrections, $\delta(T_1T)^{-1}$, is $\sim$10 T, which is an order of magnitude smaller than the expected field scale for a purely magnetic scenario for the pseudogap.

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