Magnetic Field Independence of the Spin Gap in $YBa_2Cu_3O_{7-\delta}$

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We report, for magnetic fields of 0, 8.8, and 14.8 T, measurements of the temperature dependent ^{63}Cu NMR spin lattice relaxation rate for near optimally doped $YBa_2Cu_3O_{7-\delta}$, near and above T_c. In sharp contrast with previous work we find no magnetic field dependence. We discuss experimental issues arising in measurements of this required precision and implications of the experiment regarding issues including the spin gap or pseudogap. [S0031-9007(98)08138-1]

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A dominant feature of optimally and underdoped cuprates is the appearance of a pseudogap in the normal state excitation spectrum. The microscopic mechanism which is responsible remains a mystery. A number of scenarios for explaining the pseudogap have been proposed (see Ref. [1] for a recent review). However, no calculations of the consequences of a large applied field for the pseudogap have been published. The high magnetic field behavior of the pseudogap provides additional experimental characterization of the pseudogap which is crucial for differentiating between various pictures.

We report very high accuracy measurements of the magnetic field dependence of the ^{63}Cu spin lattice relaxation rate in near optimally doped $YBa_2Cu_3O_{7-\delta}$. Our measurements demonstrate, in sharp contrast with previous work [2–6], that there is *no* magnetic field dependence to $^{63}(T_1T)^{-1}$ in $YBa_2Cu_3O_{7-\delta}$. This result has three important ramifications. Although the magnetic fields we apply shift T_c down by as much as 8 K, the onset of pseudogap effects does not shift down in temperature. Hence the pseudogap is unrelated to superconducting fluctuations, even in near optimally doped $YBa_2Cu_3O_{7-\delta}$ where the gap behavior appears just above T_c. The onset of the pseudogap is very rapid, clearly demonstrating that its magnitude is temperature dependent, opening very rapidly near 110 K. Finally, the absence of any field effect indicates a relatively large energy scale for the gap mechanism. If dynamical pairing correlations or preformed pairs are involved, the length scales must be very short.

The ^{63}Cu NMR spin lattice relaxation rate reveals the spin part of pseudogap behavior, the "spin gap." In underdoped $YBa_2Cu_3O_{6.6}$, $^{63}(T_1T)^{-1}$ famously exhibits a broad maximum in the vicinity of room temperature and then decreases as T approaches T_c from above. In optimally doped $YBa_2Cu_3O_{7-\delta}$ (data shown in Fig. 1) the maximum occurs at $\approx$110 K, and commences a quite steep descent as T is lowered towards $T_c = 93$ K, although the magnitude and onset temperature of the effect seem to have a significant dependence on the doping level even for samples with the same T_c [7]. The steepness of the downturn of $^{63}(T_1T)^{-1}$ in $YBa_2Cu_3O_{7-\delta}$ enables a sensitive measurement.

The core experimental finding of this work is presented in Fig. 1, which shows, for magnetic fields of 0, 8.8, and 14.8 T, the ^{63}Cu spin lattice relaxation rate $^{63}(T_1T)^{-1}$ vs temperature. All data shown in Fig. 1 are normal state measurements, with temperatures greater than $T_c(H)$.

The results of Fig. 1 contrast sharply with previous measurements. For example, Carretta *et al.* [3,4] have found that increasing the magnetic field from 0 to 6 T results in a decrease in $^{63}(T_1T)^{-1}$ by some 20%, for temperatures just above T_c. They ascribe the decrease to the field suppression of phase sensitive Maki-Thompson effects and conclude that their findings support s-wave pairing. Borsa *et al.* [2] observe similar behavior but suspect a field effect upon antiferromagnetic fluctuations

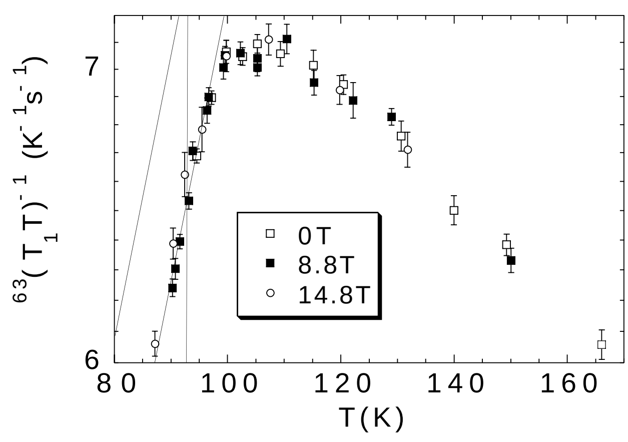

FIG. 1. $^{63}(T_1T)^{-1}$ vs T for magnetic fields (applied along the crystal c axis) of 0, 8.8, and 14.8 T. Only the data points such that T greater than $T_c(H)$ are plotted. A straight line is given which coincides approximately with the falloff in $^{63}(T_1T)^{-1}$, along with a parallel line shifted to a lower temperature by an amount equal to 7.8 K, the difference in T_c for fields of 0 and 14.8 T. The vertical line indicates $T_c(H = 0)$.

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as the mechanism. In contrast, Mitrovic *et al.* [5,6] have probed $^{63}(T_1T)^{-1}$ indirectly through effects upon the T_2 of ^{17}O [8], and find that as the field is increased from 2 to 24 T the rate $^{63}(T_1T)^{-1}$ *increases* by some 18%. Note that the field dependencies of 10%–20% observed in all of these previous measurements are approximately the same as the entire vertical scale of Fig. 1.

Now, we shall describe experimental procedures, focusing on possibly significant differences with previous work. Then we shall discuss inferences which can be drawn from the results.

Our measurements were carried out on powder samples aligned in epoxy with the crystallite c axes parallel. These samples were extensively characterized in earlier studies [9]. For all T_1 measurements the magnetic field is applied along the sample c axis. The sample was prepared using the procedure described in Ref. [10]. Figure 2 shows the high frequency ^{65}Cu (3/2,1/2) satellite transition line shape, with a full width at half maximum of ≈400 kHz. One important feature of Fig. 2 is that the line shape is not symmetric—there is essentially no intensity at frequencies much greater than the line position of 136.7 MHz, but at lower frequencies there is a significant "background" intensity. This kind of behavior for Cu NMR in aligned powder samples is well understood: in a perfectly aligned powder sample there would be background intensity neither above nor below the satellite transition. The source of this background intensity, then, is the crystallites which for whatever reason are not perfectly aligned. Considering both the plane and chain ^{63,65}Cu and the full interaction between the nucleus and the local electric field gradient tensor, a misaligned crystallite in an 8.8 T field can contribute NMR intensity anywhere in the range from 69.1 to 136.7 MHz. However, there can be *no* intensity outside this range. Furthermore, intensity appearing at the upper (lower) frequency limit derives exclusively from the ^{65}Cu (^{63}Cu) upper (lower) satellite transition with the field parallel to the c axis.

Conventionally Cu spin lattice relaxation has been measured on the central $(1/2, -1/2)$ transition, where one expects that background intensity from misaligned crystallites will be present. Martindale *et al.* [11], however, have documented that in these circumstances the background intensity can significantly contaminate the signal and reduce the accuracy of the relaxation measurement. For this reason, the T_1 measurements reported here have been performed on satellite transitions having the highest (or lowest) frequency which can be present. For the 14.8 T measurements we used the low frequency ^{63}Cu satellite. For the 8.8 T measurements we used the high frequency ^{65}Cu satellite of Fig. 2, and we plotted in Fig. 1 the measured rate multiplied by $(^{63}\gamma/^{65}\gamma)^2 = 0.8713$. Finally, for zero field we used the ^{63}Cu nuclear quadrupole resonance (NQR) transition at 31.5 MHz.

Figure 3 gives measured spin lattice relaxation recovery curves at $T = 100$ K, demonstrating the importance, when making precise T_1 measurements, of probing at frequencies not subject to background effects. Experimental data are given for 0 T (^{63}Cu NQR), and for both the high frequency ^{65}Cu satellite $(3/2 \leftrightarrow 1/2)$ and the central $(1/2 \leftrightarrow -1/2)$ ^{63}Cu transition at 8.8 T.

Relaxation measurements were made by first inverting the nuclear spin magnetization M at time $t = 0$.

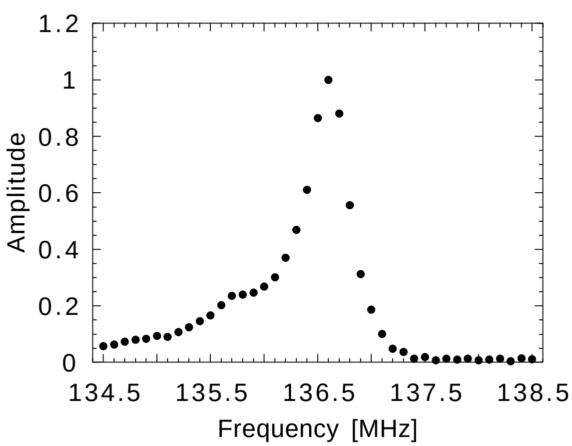

FIG. 2. ^{65}Cu NMR satellite $(3/2 \leftrightarrow 1/2$ transition) line shape with 8.8 T magnetic field applied along the c axis of the aligned powder sample. No intensity is observed at frequencies significantly higher than the peak frequency of 136.6 MHz, because that frequency is the highest frequency which can occur for any Cu NMR transition and for any orientation in $YBa_2Cu_3O_7$. Thus, T_1 measurements performed on this transition are not corrupted by background.

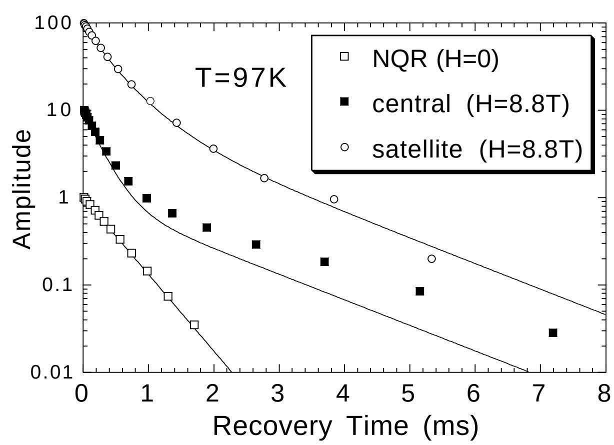

FIG. 3. ^{63}Cu spin lattice relaxation experimental and theoretical recovery curve for zero field (NQR), and for $H = 8.8$ T, both central transition and satellite transition. For the case of the satellite transition data reported are for ^{65}Cu, and the times are multiplied by $(^{63}\gamma/^{65}\gamma)^2 = 0.8713$. Theoretical curves are as given in Table I, using $T_1 = 1.484$ ms for all three sets of data. The disagreement between the theoretical curve and experiment for the case of the central transition demonstrates the error resulting from background intensity, which we avoid in the data of Fig. 1 by observing the highest or lowest frequency satellite transitions.

The recovery of $M(t)$ to its thermal equilibrium value M_∞ was then monitored, using the CYCLOPS phase cycling sequence to remove the effects of coil ringdown, gain imbalance, and stimulated echoes [12]. ^{63,65}Cu is a four-level, spin $3/2$ system, and thus the relaxation curve $M(t) - M_\infty$ is expected to be multiexponential with known coefficients [13] but with only a single adjustable time constant T_1 which is to be measured. The expected functional forms for the relaxation of the magnetization in the three situations are given in Table I. In Fig. 3 we use for all three relaxation curves a time constant T_1 equal to 1.484 ms, chosen to best fit the NQR data. We see clearly from the figure that while the satellite and NQR measurements follow the expected functional form beautifully, the central transition has a significant deviation. We interpret this effect as arising from background intensity, having a different T_1, at the frequency of the central transition. Such an effect may be the source of the apparent field dependence observed in previous measurements, with the exception of those of Refs. [5,6], which would not be susceptible to this problem. We note that the excellent, single exponential recoveries observed in the NQR experiment rule out any effects due to spectral diffusion.

Now, what can be said about the field dependence of spin gap behavior from Fig. 1? For illustration we have included in Fig. 1 a straight line which coincides approximately with the falloff in $^{63}(T_1T)^{-1}$ between $\approx$96 K and $T_c(H)$. Then for comparison we include the same line, but shifted to lower temperature by an amount equal to 7.8 K, which is the expected difference in T_c for fields of 0 and 14.8 T, assuming $dH_{c2}/dT = -1.9$ T/K [14]. One suspects that if the spin gap phenomenon were a superconducting fluctuation effect, then the 14.8 T data would be shifted relative to the 0 T data by a comparable amount, but that plainly is not the case. In fact, within experimental error the 14.8 T data display the same onset temperature as the 0 T data, and from a close analysis of the data and error bars we find that any decline in the temperature for the onset of spin gap effects over this range of fields must be less than 2 K. Thus, we find that application of a field (14.8 T) representing a Zeeman energy $g\mu_BH = 20$ K, which is $\approx$20% of T_c, results in no decrease in onset temperature within an uncertainty of 2%.

TABLE I. Functional forms of T_1 recovery curves expected for the zero field NQR, and the high field central $(1/2, -1/2)$ and satellite (3/2,1/2) transitions, assuming a magnetic relaxation mechanism [13].

Transition	Recovery curve: $[M(t) - M_\infty]/[M_0 - M_\infty]$
NQR	$\exp(-\frac{3t}{T_1})$
Central	$0.1\exp(-\frac{t}{T_1}) + 0.9\exp(-\frac{6t}{T_1})$
Satellite	$0.1\exp(-\frac{t}{T_1}) + 0.5\exp(-\frac{3t}{T_1}) + 0.4\exp(-\frac{6t}{T_1})$

Of course, a comparison of this result with quantitative predictions of various theoretical models is necessary, but nevertheless we suspect that this null result will pose a serious challenge to some theories. Possible exceptions include models [15] based upon the t-J model, which call for local singlet pairing with an energy scale governed by the exchange coupling J (of the order of 1000 K). We expect there would be a coupling of the applied magnetic field to the orbital motion of pairs [16] formed above T_c, in this case the absence of a field effect will constrain the length scale of the pair. Gaps associated with the formation of ladderlike structures [17] also involve large energy scales and so would not be expected to be sensitive to the fields applied here. Finally, the antiferromagnetic Fermi liquid based approaches [18] have treated the pseudogap effect, but the extent of any magnetic field dependence which would be predicted is not clear.

The fact that the onset temperature for spin gap effects is not shifted down in temperature along with the known suppression of T_c demonstrates that the gap even in optimally doped $YBa_2Cu_3O_{7-\delta}$ is not closely tied to the onset of superconductivity and thus has nothing to do with superconducting fluctuations. The abrupt decrease in $^{63}(T_1T)^{-1}$ then requires a strongly temperature dependent gap. Clearly the gap is not present at high temperatures, rather there must be an abrupt transition near 100 K that causes the gap to open. This abrupt opening could reflect the onset of charge ordering into fluctuating structures which would enable the development of a gap [16,17]. Finally, the mechanism for the gap must have a large energy scale compared to the electron spin Zeeman energy scale $g\mu_BH$ ($\approx$20 K for a 14 T field), even at optimal doping where the gap appears only slightly above 100 K.

To conclude, we find that the spin gap effect in the cuprates is insensitive to magnetic field, with the onset temperature remaining unchanged, within an uncertainty of 2 K, by a 14.8 T field which suppresses T_c by 8 K. These results would appear to call for relatively large energy and short length scales in a scenario involving dynamical pairing correlations or preformed pairs. However, in order to make a more definite statement, quantitative predictions from alternative theories will be necessary.

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