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NUCLEAR RELAXATION RATES AT COPPER AND OXYGEN SITES IN $YBa_2Cu_3O_7$

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We report NMR measurements of the nuclear relaxation rate at all copper and oxygen sites in magnetically aligned powder samples of $YBa_2Cu_3O_7$. There is no peak in the oxygen relaxation rate below T_c at any oxygen site supporting the possibility that d-wave pairs are formed in the superconducting state. Comparison of the oxygen and copper rates in the planes reveals a characteristic temperature greater than T_c.

We have measured the relaxation rate at all oxygen and copper sites in aligned powders of $YBa_2Cu_3O_7$. Relaxation measurements provide microscopic information about spin dynamics at these sites; comparison of data from the CuO_2 planes gives insight into the relationship between the doped holes of primarily O-2p and copper d-orbital character. Contrary to earlier reports[1] our measurements clearly show the absence of any increase of relaxation just below T_c at any of the four oxygen sites.

An earlier publication[2] describes the sample and shows an NMR spectrum. T_c is 93 K and the shielding is close to 100%. The ^{17}O (^{63}Cu) relaxation measurements were made in a 7.0 (7.4) Tesla field which reduces T_c to 86 K when the field direction is parallel to the crystal c-axis (H//c). The ^{17}O H//c measurements were made on the quadrupole satellites to ensure that signals from a single site only were detected. The time dependence of the recovery of the oxygen magnetization following a single 90° saturating pulse is well described by the expected[3] five component exponential recovery.

The O(2,3) (planar oxygen) relaxation rate, $^{17}T_1{}^{-1}$, contrasts dramatically with the Cu(2) rate, $^{63}T_1{}^{-1}$ (Fig. 1). Above T_c $^{17}T_1{}^{-1}$ is linear in temperature while $^{63}T_1{}^{-1}$ has a much weaker temperature dependence and is roughly 20 times larger. The Korringa[4] relation $(T_1TK^2)^{-1} = \pi\hbar k_B\gamma^2/\mu_B{}^2$ (K is the Knight shift) which describes relaxation of nuclei coupled to conduction electrons in a metal describes $^{17}T_1{}^{-1}$ well: its temperature dependence is linear and the magnitude of $(T_1TK^2)^{-1}$ is only 1.4 times the ideal value $\pi\hbar k_B\gamma^2/\mu_B{}^2$. To understand the magnitude of $^{63}T_1{}^{-1}$one is led to examine the consequences of antiferromagnetic interactions between d-hole spins. The relaxation rate is proportional to $k_BT\langle\chi''(q)\rangle_q$, (where $\langle\chi''(q)\rangle_q$ is the zero frequency limit of the average of the dynamical susceptibility, $\chi''(q,\omega)/\omega$, over all q spanning the Fermi surface). Antiferromagnetic interactions increase $\chi''_d(q,\omega)$ (d refers to the copper d-spin) at the antiferromagnetic wave vector, Q_{AF}, and thus increase $^{63}T_1{}^{-1}$. As a result $(T_1TK^2)^{-1}$ for copper will be enhanced with respect to the value $\pi\hbar k_B\gamma^2/\mu_B{}^2$: the

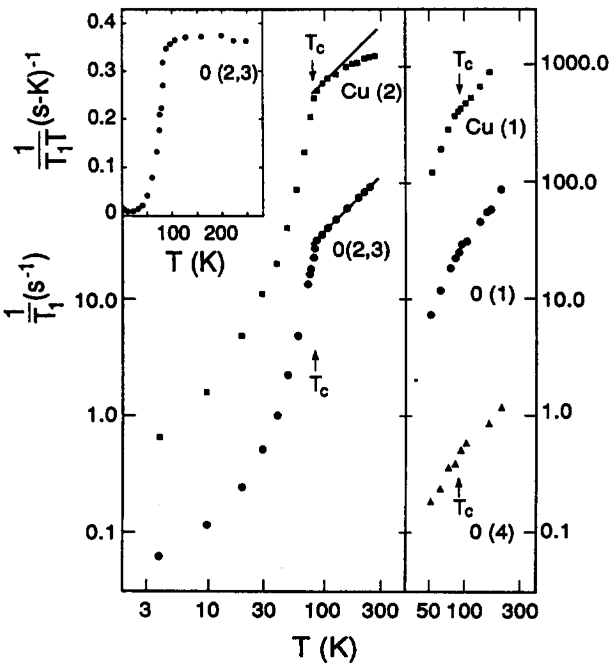

Figure 1. We show the H//c relaxation rates. The solid lines indicate a linear temperature dependence; the inset shows the linear behavior of the O(2,3) rate above T_c.

data[9] show this enhancement to be roughly 11 at 100 K. $^{17}T_1^{-1}$ is not enhanced by the large value of $\chi''_d(Q_{AF})$ because the transferred hyperfine coupling of the O(2,3) nuclear spin to the Cu-3d spin vanishes at Q_{AF} (the hyperfine fields from antiparallel spins on the neighboring coppers cancel at the oxygen site).

The linear temperature dependence of $^{17}T_1^{-1}$ shows that $<\chi''_{O\text{-}p}(q)>_q$ (O-p refers to holes primarily resident in O-2p orbitals) is independent of temperature. The temperature dependence of R (Fig. 2) above 1.35 T_c shows that $<\chi''_d(q)>_q$ is increasing with decreasing temperature. From the copper Knight shift[5] we know that $\chi(q=0)$ is temperature independent above T_c showing that it is the large q (near Q_{AF}) component of $\chi''_d(q,\omega)$ which is increasing. Through the Kramers-Kronig relation, the temperature dependence of $\chi''_d(Q_{AF},\omega)/\omega$ indicates that $\chi_d(Q_{AF})$, is increasing with decreasing temperature. Fig. 2 shows that the increase of $\chi_d(Q_{AF})$ with cooling ceases at a characteristic temperature greater than T_c. Below 1.35 T_c $\chi''_{O\text{-}p}(q,\omega)$ and $\chi''_d(q,\omega)$ become strongly coupled and $\chi_d(Q_{AF})$ is temperature independent. This could imply that the coupling between the doped holes and the d-holes themselves becomes much stronger at this temperature. This situation would bear some similarity to the heavy fermion systems. That R never decreases with decreasing temperature means the enhancement of copper relaxation relative to that of oxygen does not decrease even in the superconducting state. Thus the rapid decrease of copper relaxation which occurs in the vicinity of T_c is not the result of the loss of the enhancement.

The complete absence of any peak in the relaxation rate immediately below T_c is significant. Seen[6] in s-wave superconductors, this peak will be absent in d-wave superconductivity because the existence of quasiparticle states in the gap reduces the accumulation of states at the gap edge and the coherence factor will be zero for the simplest d-wave states one might consider for a square Fermi surface.

In the chains (Fig. 1) both O(1) and O(4) show the same temperature dependence as Cu(1). From this we conclude that the doped holes are strongly bound to the Cu(1) holes to form a single band. As in the planes, neither the O(1) nor O(4) sites show a peak in T_1^{-1} below T_c again supporting the possibility of d-wave pairing.

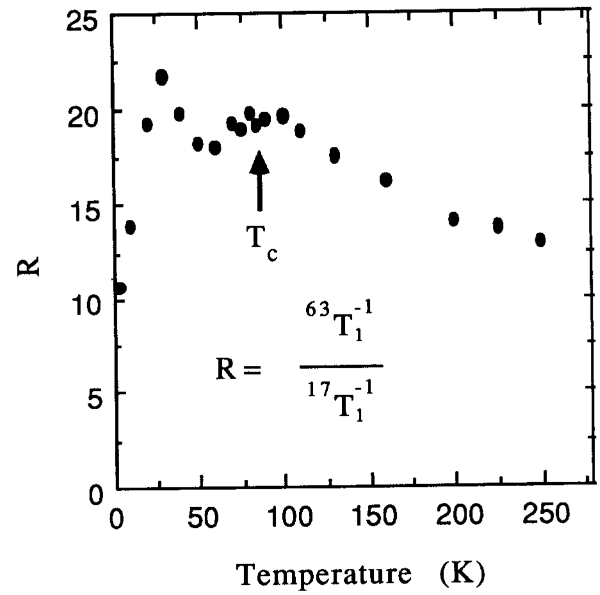

Figure 2. Comparison of copper and oxygen relaxation in the planes reveals a characteristic temperature greater than T_c, 1.35 T_c, at which the antiferromagnetic spin fluctuations stop growing with decreasing temperature.

The clear conclusion from the comparison of oxygen and copper relaxation is the existence of a characteristic temperature other than T_c. At 1.35 T_c spin degrees of freedom which have some independence at higher temperatures become coupled: $\chi_d(Q_{AF})$ ceases to grow with decreasing temperature and becomes temperature independent. The rapid decrease of copper relaxation in the vicinity of T_c is not due to the loss of enhancement from antiferromagnetic spin fluctuations. Finally, the clear absence of any increase in $^{17}T_1^{-1}$ below T_c supports the possibility of d-wave superconductive pairing.

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