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Anisotropy and magnetic field dependence of the planar copper NMR spin-lattice relaxation rate in $YBa_2Cu_4O_8$

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We have measured the temperature and magnetic field dependence of the ^{63}Cu nuclear spin-lattice relaxation rate W and its anisotropy r at plain Cu(2) sites in normal and superconducting $YBa_2Cu_4O_8$. Below T_c we observed that an applied magnetic field $B \parallel c$ enhances $W = W_c$, whereas $B \perp c$ suppresses $W = W_{ab}$. Such a behavior seems to rule out the spin diffusion to the fluxoid cores and the fluxoid motion as being responsible for the effect. It indicates more an unexpected field-related breaking of the spin-rotation invariance in the superconducting state. The anisotropy r defined as the ratio W_{ab}/W_c is almost field and temperature independent in the normal state but develops a nonmonotonic temperature dependence below T_c with a flat minimum at 45 K in B = 5.17 T and a much more pronounced minimum at 55 K in B = 0.58 T. A qualitatively similar behavior of r has been reported previously for $YBa_2Cu_3O_7$. Comparing r in both compounds, we note one essential difference at low B. Namely, the slope dr/dT just below T_c is large for $YBa_2Cu_3O_7$ but almost zero for $YBa_2Cu_4O_8$.

Nuclear magnetic resonance and neutron-scattering experiments have shown that spin fluctuations in high-temperature superconductors have strong antiferromagnetic (AFM) correlations, which persist into the superconducting state. The study of the temperature and field dependence of the NMR spin-lattice relaxation rate W_α, where $\alpha = a$, b, or c specifies the orientation of the static applied field B, and its anisotropy $r = W_{ab}/W_c$ at the Cu plane site [(Cu(2)] may help to understand how these AFM correlations are affected by superconductivity.

Recently, it was reported[1,2] that in $YBa_2Cu_3O_7$ (abbreviated 1-2-3) measurements of W_α and r in the superconducting state appear to require a modification of theories such as that by Millis, Monien and Pines[3] but agree qualitatively with the orbital-*d*-wave fits by Bulut and Scalapino[4,5] and Lu.[6] In addition, Martindale *et al.*[2] found that in the superconducting state W_c, and to a lesser degree W_{ab}, become enhanced in a magnetic field, with the enhancement growing at lower temperature. A temperature-dependent anisotropy below T_c in 1-2-3 has also been studied in low magnetic field by Takigawa, Smith, and Hulst.[7]

We have reported previously[8] similar investigations of W_α anisotropy and its field dependence for Cu(2) in the stoichiometric double-chain compound $YBa_2Cu_4O_8$ (1-2-4), which has the same Cu-O plane structure as 1-2-3 but lower charge-carrier concentration. In particular, the Cu(2) rate W_c, field independent in the normal state, shows a field-dependent enhancement in the superconducting state that already begins 13 K above T_c in a 5.17 T field. Consequently the anisotropy r that is temperature and field independent above $T_c + 13$ K ($r = 3.3$) starts to diminish below this temperature. Down to 80 K the reduction of r is hardly noticeable. However, below that temperature r drops very rapidly to a value of 2.2 and after passing a flat minimum at 45 K it increases again at lower temperature.

Since in the superconducting state the change of r due to a magnetic field could be a secondary effect caused, for example, by fluxoid cores or by T_c suppression in a magnetic field, we decided to extend our previous high-field experiments to low fields, where the field-induced anisotropy effects may be neglected. In this Rapid Communication we will show that the anisotropy r behaves similarly as in 1-2-3, but with some pronounced differences. In addition, new results on the low-temperature behavior of W_{ab} and W_c will be presented.

We briefly discuss the procedure to determine the rate anisotropy. For a strong magnetic field and pure magnetic relaxation the nuclear spin-lattice relaxation rate W involves fluctuating hyperfine fields H_α perpendicular to the applied external field. In case of $B \parallel c$ the rate may be expressed as $W_c = \frac{3}{2}(H_a^2+H_b^2)\gamma_n^2\tau$, where γ_n is the nuclear gyromagnetic ratio and τ is the (isotropic) correlation time.[9] For $B \perp c$, the rate is $W_{ab} = \frac{3}{2}(H_a^2+H_c^2)\gamma_n^2\tau$, since a and b are not distinguishable for the Cu(2) site.

By a zero-field nuclear quadrupole resonance (NQR) experiment, only W_c can be obtained for the Cu(2) nuclei because the largest component V_{zz} of the axially symmetric electric-field gradient at the Cu(2) sites, defining the quantization direction, is parallel to the c axis. To determine W_{ab}, a nonvanishing magnetic field perpendicular to V_{zz} has to be applied. To keep the anisotropy effect of the applied magnetic field on T_c and relaxation possibly small, we studied the temperature dependence of r at rather low field of 0.58 T. A choice of appreciably lower fields is limited by the rapid deterioration of the signal to noise ratio S/N with decreasing field. S/N of the Zeeman splitted $+1/2 \longleftrightarrow -1/2$ resonance used in the experiment is proportional to the square of the applied field. Our measurements were performed on a c-axis-oriented powdered sample imbedded in epoxy, with a random orientation of the a and b axis in the plane perpendicular to the c axis. The 1-2-4 powder used exhibits $T_c = (81.0 \pm 0.5)K$.[10]

Since the quadrupole splitting of the Cu(2) nuclear spin levels is much larger than the Zeeman splitting for a small field, a special procedure is required to obtain the anisotropy of W.[2,7] For a weak magnetic field applied perpendicular to the c axis there is no obvious quantization axis, and therefore the relaxation of the Zeeman splitted $+1/2 \longleftrightarrow -1/2$ resonance is caused by the in-plane and the out-of-plane components of the fluctuating hyperfine fields. For a spin-$\frac{3}{2}$ nucleus such as Cu(2), the magnetization recovery following an inversion pulse is described by

$$M(t) = M(\infty) \left\{ 1 - 2 \sum_{k=1}^{3} \beta_k \exp\left(-\lambda_k W_c t\right) \right\} , \quad (1)$$

where $\sum \beta_k = 1$. The β_k and λ_k are functions of the anisotropy r and of the ratio between the Zeeman ν_L and the quadrupole frequency ν_Q. In Fig. 1, β_k and λ_k are plotted as a function of r calculated for $\nu_L = 6.44$ MHz ($B = 0.58$ T) and $\nu_Q = 29.75$ MHz. Using the values of W_c as measured by NQR,[11] we fitted the magnetization recovery data by Eq. (1) to obtain λ and, hence, r.

Figure 2 shows the temperature dependence of the anisotropy ratio r measured in a low magnetic field $B = 0.58$ T (solid circles) and high magnetic field $B = 5.17$ T (open circles). Within experimental errors of ±10%, the weak field ratio $r = 3.2$ in the normal conducting phase agrees with our previously reported high-field result $r = 3.3$.

Below T_c, both the high- and low-field r values decrease. While the high-field values level off around $r = 2.2$,[8] the low-field r passes a pronounced minimum at 55 K, increases again at lower temperatures and reaches at 30 K a value of 5.7. We did not continue our low-field measurements below 30 K because of an inhomogenious distribution of W_c seen in zero-field NQR arising most probably from extrinsic effects as disorder and impurities. Figure 3 compares our low field r values for 1-2-4 with those of Takigawa, Smith, and Hulst[7] for 1-2-3. The arrows indicate the value of r above T_c for 1-2-4 and 1-2-3. The two sets of data are quite similar. However, the minimum of r in 1-2-4 seems to be deeper and is positioned about $0.1T/T_c$ lower than in 1-2-3. The upturn of r with decreasing temperature is much more pronounced as compared to 1-2-3.

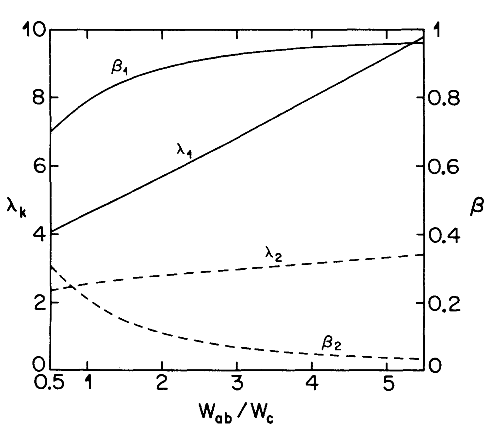

FIG. 1. Calculated r dependence of β_k and λ_k for $\nu_L = 6.44$ MHz ($B = 0.58$ T) and $\nu_Q = 29.75$ MHz. β_3 is zero and not plotted.

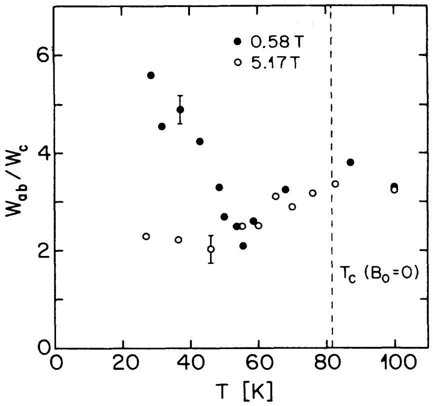

FIG. 2. Temperature dependence of Cu(2) spin-lattice relaxation rate anisotropy r for $YBa_2Cu_4O_8$ in two different magnetic fields $B = 0.58$ T (•) and $B = 5.17$ T (○).

New explanations of the temperature dependence of r in the superconducting state have been presented recently by Bulut and Scalapino[4,5] and by Lu[6] using a BCS pairing theory. Both groups use spin-singlet pairing and assume temperature-dependent energy-level broadening and include pair-creation and -annihilation terms in the calculation of the susceptibility. They also include an

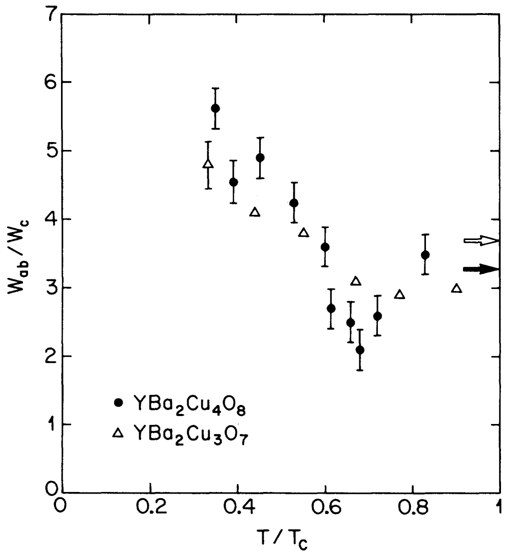

FIG. 3. Cu(2) spin-lattice relaxation rate anisotropy r vs the reduced temperature T/T_c in a weak magnetic field: $YBa_2Cu_4O_8$ in $B = 0.58$ T (•) and $YBa_2Cu_3O_7$ in $B = 0.44$ T (△) (Ref. 7).

anisotropic on-site and an isotropic transferred hyperfine coupling of the Cu(2) nuclei to Cu(2) electron spins. Figure 2 of Ref. 2 compares the theoretical results with experimental data for 1-2-3.

The sharp decrease of r just below T_c and its upturn at lower temperature is in qualitative agreement with these theories, which predict such a behavior as a result of nodes in the gap (d-wave pairing). In addition, the change of r close to the normal-to-superconducting transition reveals the importance of coherence factors.

However, our data for 1-2-4 exhibit just below T_c a much softer decrease of r; the derivative dr/dT is almost zero at T_c. It remains to be shown whether such a behavior can be reproduced by the above-mentioned theories. Thus, at present it cannot be decided whether our anisotropy data for 1-2-4 favor d-wave pairing or not. Theoretical calculations of r for 1-2-4 by Eremin and Markendorf[12] are in progress.

We now discuss the field dependence of the relaxation rate. We have measured W_{ab} and W_c in strong, weak and zero fields in an oriented powder sample. A summary of the temperature dependence of the absolute values is given in Fig. 4. The results normalized to the respective rate at T_c are plotted as a function of the reduced temperature $T/T_c(B)$ in Figs. 5 and 6. The T_c values for a fixed field were derived from H_{c2} measurements done on a 1-2-4 single crystal by Bucher *et al.*[13]

We first note that W_c depends more strongly on the field than W_{ab} does. The field dependence increases with decreasing temperature. At $T = 0.4T_c$ the high-field rate becomes about twice as large as the zero-field rate. A similar enhancement has been found in 1-2-3.[2] On the other hand, W_{ab} exhibits quite a different behavior. Down to about T = $0.7T_c$, an applied field slightly enhances the relaxation rate as previously observed in 1-2-3.[2,14] However, below $0.7T_c$ the enhancement gives way to a suppression that becomes more evident at lower temperatures. At $0.35T_c$ the high 5.17-T field reduces W_{ab} to about 40% of the value measured in the low 0.58-T field. Such an unique field dependence is in contrast to the behavior of W_{ab} in 1-2-3.

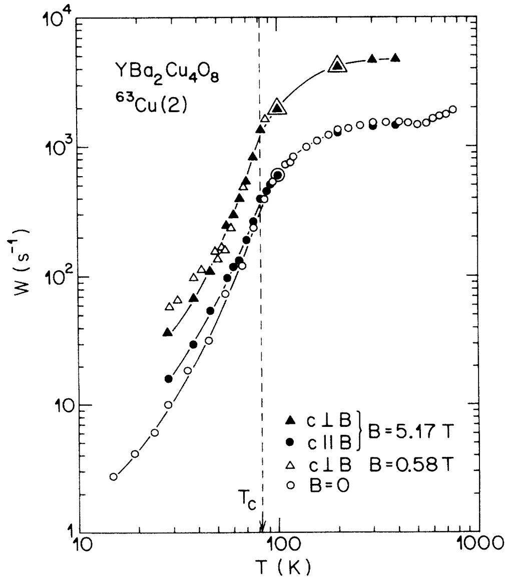

FIG. 4. Spin lattice relaxation rates vs temperature for Cu(2) for different magnetic fields and orientations. The triangles are for $B \perp c$ and the circles for $B \parallel c$.

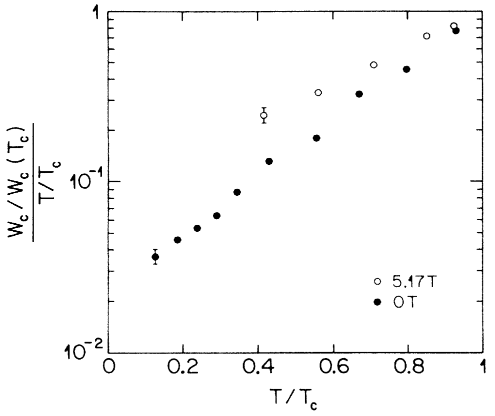

FIG. 5. Temperature dependence of the normalized Cu(2) spin-lattice relaxation rate W_c divided by T/T_c in $YBa_2Cu_4O_8$ for $B = 0$ T (●) and $B = 5.17$ T (○).

The opposite response of W_{ab} and W_c to the application of a magnetic field seems to rule out the possibility that fluxoid cores or thermally activated fluxoid motions cause the field dependence as it has been discussed for 1-2-3.[2] To account for the opposite response of W_{ab} and W_c an unexpected field-related breaking of the spin-rotation invariance in the superconducting state has to be considered. Finally, we want to stress the fact that in 1-2-4 the high field W_c is larger than the NQR rate from the lowest temperatures used in our experiment, up to T_c + 13 K (see Fig. 4). This is in contrast with a recent obser-

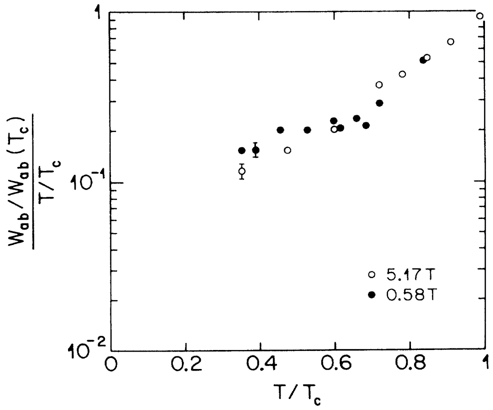

FIG. 6. Temperature dependence of the normalized Cu(2) spin-lattice relaxation rate W_{ab} divided by T/T_c in $YBa_2Cu_4O_8$ for $B = 0.58$ T (●) and $B = 5.17$ T (○).

vation by Borsa *et al.*,[14] who found in the temperature region just above T_c an opposite behavior for 1-2-3 and $La_{1.85}Sr_{0.15}CuO_4$.

In conclusion, our new data for 1-2-4 and the comparison with results from 1-2-3 clearly show that at the moment there is no consistency with regard to the influence of an applied magnetic field on the planar copper spin-lattice relaxation rates in the superconducting state and the normal state just above T_c.

ACKNOWLEDGMENTS

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