Anomalous Long Time Tail Behavior of Spin-Spin Correlation in the Normal State of $Tl_2Ba_2CuO_y$

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(Received 4 February 1994)

We have found a frequency (ω) dependence of the spin-lattice relaxation time in the normal state of $Tl_2Ba_2CuO_y$ ($T_c = 15$ and 85 K). The value of $1/T_1T$ at the Tl and Cu sites is proportional to $\ln\omega^{-1}$ at low temperature. This type of ω dependence suggests a long time tail behavior of the spin-spin correlation. On the other hand, no frequency dependence is observed at the planar oxygen site; thus the observed long time tail behavior is enhanced around the wave vector $q = (\pi, \pi)$. The characteristic cutoff frequency for the long time tail decreases with decreasing T, indicating that the confinement of spin freedom occurs above T_c.

PACS numbers: 76.60.−k, 74.72.Fq, 75.40.Gb

For more than a decade, the special features of low-dimensional magnetic systems have received continuous interest. The high T_c cuprates are quite interesting systems from this point of view because of their strong two dimensionality. It has been revealed that low-dimensional magnetic systems have unique magnetic properties which are not observed in three-dimensional systems [1]. The long time tail (LTT) of the spin-spin correlation is an especially unique behavior, which has been theoretically predicted and experimentally observed in many one-dimensional organic systems [2]. In general, the spectral function of spin-spin correlation $g_q(\omega)$ is expressed as [1]

$$g_q(\omega) = \int_0^\infty \langle S_+^q(t) \cdot S_-^q(0)\rangle \cos\omega t\, dt . \qquad (1)$$

In the ideal low-dimensional system, $g_q(\omega)$ diverges at $\omega = 0$ due to the LTT [3]. The spin-lattice relaxation rate ($1/T_1$) is sensitive to the LTT behavior because it is proportional to $g_q(\omega)$ at $\omega \approx 0$. For example, Boucher *et al.* found this frequency (ω) dependence of T_1 in the one-dimensional antiferromagnetic insulator (TMMC) [4]; in addition a similar ω dependence of T_1 was also revealed in the one-dimensional organic conductor (TCNQ) [5].

Theoretically the relations of $1/T_1 \propto \omega^{-1/2}$ and $1/T_1 \propto \ln\omega^{-1}$ around $q \approx 0$ are predicted for the one- and two-dimensional cases, respectively [6]. In fact the relation of $1/T_1 \propto \omega^{-1/2}$ is observed in several one-dimensional systems [2, 4, 5]. On the contrary, experimental evidence for the relation of $1/T_1 \propto \ln\omega^{-1}$ in the two-dimensional system is confirmed only for the case of NH_3 molecular diffusion in an intercalation complex [7]. Although the high T_c cuprates have very strong two-dimensional magnetic nature, up to now no frequency dependence has been reported in $(La,Sr)_2CuO_4$ [8] and $YBa_2Cu_3O_y$ [9].

In the present study, we have studied the ω dependence of T_1 in the normal state of the $Tl_2Ba_2CuO_y$ system. This is the first clear observation of the LTT behavior in the two-dimensional magnetic system. This system is known to be an overdoped compound which has a high carrier concentration as compared with $YBa_2Cu_3O_y$ and $(La,Sr)_2CuO_4$ [10]. The unit cell contains a CuO_2 monolayer which is separated by double TlO layers [10]. The T dependence of the Knight shift and T_1 at each site in $Tl_2Ba_2CuO_y$ of $T_c = 85$ and 0 K has been reported previously [11, 12].

The characteristic spin-fluctuation energy at $q \approx Q = (\pi, \pi)$ is estimated to be large (90 meV) in this system [11]; thus the antiferromagnetic correlation in the normal state of this system is considered to be weaker than that of $YBa_2Cu_3O_y$ and $(La,Sr)_2CuO_4$ [8].

We have prepared the samples of $Tl_2Ba_2CuO_y$ by the standard solid reaction method in a closed system [11]. In the present report, we study two specimens ($T_c = 15$ K, sample A, and $T_c = 85$ K, sample B). Sample A is regarded as the overdoped one which has a higher carrier concentration than sample B. NMR measurements were performed using the standard pulse method. For measurements of T_1, we have used the free induction decay signal of ^{205}Tl NMR, and the spin echo signals of ^{17}O and ^{63}Cu NMR, respectively. Details of the experiments have been reported in Refs. [11, 12].

Figure 1 shows the ^{205}Tl fast Fourier transform (FFT) spectra in sample A. No impurity phases were observed in the two samples measured. The time decay curve for the spin-lattice relaxation is well fitted by a unique T_1 for every measurement (see inset of Fig. 1). Because the nuclear spin of Tl is $\frac{1}{2}$, this site is insensitive to the quadrupole interaction, and thus the time decay curve of the spin-lattice relaxation obeys the single exponential type. Even in a low magnetic field at high temperature, we could therefore determine T_1 exactly at the Tl site, although it is more difficult at the Cu site. Furthermore, the T dependence of T_1 at the Tl site is almost scaled to that of the Cu site because the transferred hyperfine field from the Cu site dominates the spin-lattice relaxation process at the Tl site [12], so we could probe the spin

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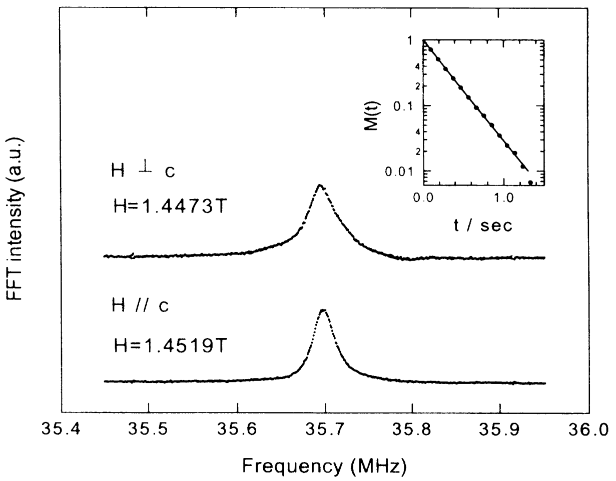

FIG. 1. The FFT spectra of ^{205}Tl NMR at 50 K in the $Tl_2Ba_2CuO_y$ of $T_c = 15$ K (sample A). The nuclear magnetization decay curve for the ^{205}Tl nuclear spin-lattice relaxation at 50 K ($H \perp c$) is shown in the inset.

fluctuation in the CuO_2 layer by T_1 at the Tl site. In addition, we have confirmed that T_1 of ^{203}Tl NMR is proportional to that of ^{205}Tl NMR by the factor of the squared gyromagnetic ratio at each frequency; thus there is no mixing in the excitation of ^{205}Tl and ^{203}Tl nuclei by the applied rf pulse. In the present study, we have intensively measured T_1 at the Tl site in order to clarify the LTT behavior in the CuO_2 layer.

Figure 2 shows the ω dependence of $1/T_1T$ at the Tl site for $H \parallel c$ and $H \perp c$ in sample A. In general, $1/T_1$ could be expressed as

$$1/T_1 = \sum_q A(q)^2 g_q(\omega), \tag{2}$$

where $A(q)$ is the hyperfine coupling constant. The value of T_1 for $H \perp c$ is proportional to that for $H \parallel c$ by the factor of the squared transferred hyperfine coupling constant $[A(q)^2_{H\parallel c}/A(q)^2_{H\perp c}]$ at each frequency; thus the $g_q(\omega)$ is concluded to be isotropic in the measured frequency range at 20 K. This behavior is different from that in the one-dimensional case where the ω dependence of $g_q(\omega)$ is quite anisotropic [4, 5].

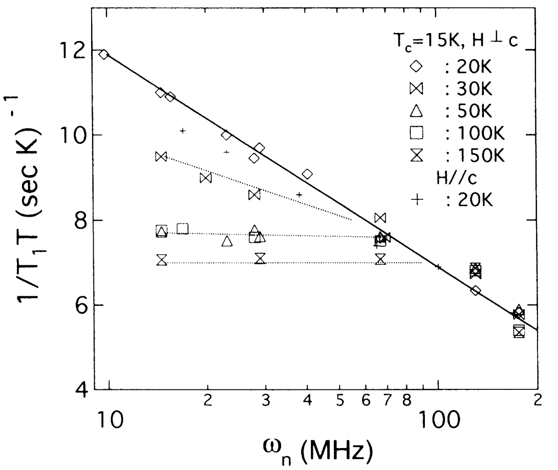

FIG. 2. The frequency dependence of $1/T_1T$ at the Tl site in sample A. The ω_n is the resonance frequency in the NMR measurement. The solid line is obtained by a least squares fitting procedure for the data at 20 K ($H \perp c$). The dashed lines are guides to the eye.

It should be noted here that no ω dependence of the Knight shift is observed at the Tl site. Since the spin part of the Knight shift at the Tl site is proportional to that at the Cu site in this system [12], the present result suggests that the static spin susceptibility in the CuO_2 layer is independent of ω, and hence the external field. This confirms that the observed ω dependence is not caused by changes in the density of states around the Fermi energy due to the external field.

At 20 K, the value of $1/T_1T$ is proportional to $\ln\omega^{-1}$ for the measured frequency range in sample A. However, at high temperatures, the ω dependence is observed only above a certain cutoff frequency, which seems to decrease with decreasing T. This implies that the saturation of $g_q(\omega)$ occurs below this cutoff frequency at high temperature. In a real system, a three-dimensional coupling introduces a truncation of the LTT in the spin-spin correlation. Thus the divergent behavior of the $g_q(\omega)$ vanishes below a certain characteristic cutoff frequency. The present results indicate that the magnetic coupling between the CuO_2 layers decreases with decreasing T in the normal state of $Tl_2Ba_2CuO_y$. Recently, Uchida *et al.* reported that a confinement of charge freedom occurs in the normal state of several high T_c oxides [13]. The observed T dependence of the magnetic coupling suggests that a confinement of spin freedom in the CuO_2 layer is enhanced in low temperature.

Figure 3 shows the ω dependence of $1/T_1T$ at the Cu and planar O sites for $H \parallel c$ at 20 K in sample A. The NMR spectra at these sites have been reported in Refs. [11, 12]. At the Cu site, the value of $1/T_1T$ is proportional to $\ln\omega^{-1}$, in accordance with the result of the Tl site.

It is remarkable than no ω dependence is observed at the O site. In contrast to the Cu and Tl sites, the spin fluctuation around $q \approx Q$ does not contribute the spin-lattice relaxation at the O site [9]; thus this fact indicates that *in the present case, the long time tail behavior is enhanced around* $q = Q$. So far the $\ln\omega^{-1}$ type ω dependence has been theoretically predicted for the $q \approx 0$ mode of the spin-spin correlation when this mode dominates the spin-lattice relaxation process. In the present case, however, the dynamical spin susceptibility is enhanced around $q \approx Q$ in the wide temperature range because of the large two-dimensional intralayer exchange interaction [11]. Although a theory of the LLT behavior for $q \approx Q$ in such a situation is still an open question, the observed new phenomenon is supposed to be caused by a

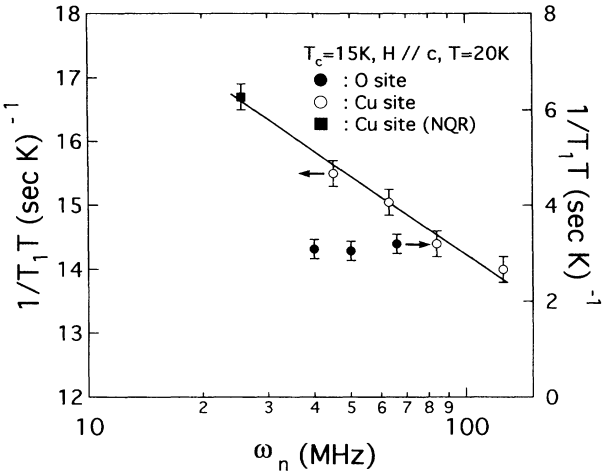

FIG. 3. The frequency dependence of $1/T_1T$ at the Cu and planar O sites at 20 K in sample A. The square point is obtained by the nuclear quadrupole resonance measurement in zero field. The solid line is obtained by a least squares fitting procedure for the data for the Cu site.

strong quantum spin effect which may be remarkable and general in the low-dimensional quantum spin system.

Figure 4 shows the ω dependence of $1/T_1T$ at the Tl site for $H \perp c$ in sample B. The cutoff frequency in sample B seems to be smaller than that of sample A at each temperature; thus the magnetic coupling between the $\mathrm{CuO_2}$ layers is considered to be weak in the higher T_c sample. This tendency is also confirmed by measurements of T_1 at the apical O site in the same samples [12]. On the other hand, a comparison of the cutoff frequency at 100 and 150 K indicates that the confinement of spin freedom also occurs in sample B.

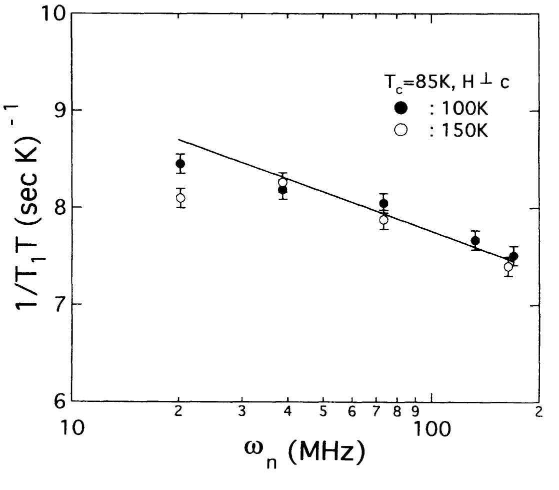

FIG. 4. The frequency dependence of $1/T_1T$ at the Tl site in the $\mathrm{Tl_2Ba_2CuO_y}$ of $T_c = 85$ K (sample B). The solid line is obtained by a least squares fitting procedure for the data with $\omega_n > 75$ MHz.

The observed magnitude of ω dependence (the slope of the $1/T_1T$ vs $\ln\omega_n$ plot) in sample B is smaller than that in sample A. In the present case, the $g_q(\omega)$ around $q \approx Q$ in low temperature may be expressed as

$$g_q(\omega) = k_B T(\alpha \ln \omega^{-1} + \beta), \tag{3}$$

where α and β are certain constants. From Eqs. (2) and (3), the value of $1/T_1T$ at the Tl site can be expressed as

$$1/T_1T = k_B \sum_{q \approx Q} \alpha A(q)^2 \ln \omega^{-1} + \beta A(q)^2. \tag{4}$$

Thus the difference in the magnitude of the ω dependence is attributable to the difference of the $A(q)$ and/or the α term. The magnitude of the ω dependence in sample A is 3.7 times larger than that in sample B (obtained from the solid line in Figs. 2 and 4). Then, if we assume that $A(q)$ is 1.5 times larger in sample A than in sample B [14], the α term of sample A is estimated to be $1.6(= 3.7/1.5^2)$ times larger than that of sample B.

One possible explanation for this difference of the α term is as follows: In the one-dimensional Hubbard model of the nearly half-filled and large $U(\gg t)$ case, $g_q(\omega)$ in the high temperature limit is expressed as

$$g_q(\omega) = \frac{3}{16t}\left(\frac{\hbar U}{2}\right)^{1/2} \gamma(c, U/t)\omega^{-1/2}, \tag{5}$$

where c, t, and U are the carrier concentration, the transfer, and the correlation energies, respectively [15]. We discuss the present results based on this one-dimensional model, because so far no quantitative theoretical treatment has been obtained in the two-dimensional case. In this model, the γ term increases strongly with increasing c around the half-filled state [16]. The values of U and t are considered to be nearly the same in samples A and B; however, the γ term is larger in sample A than in sample B because the value of c is larger in sample A than in sample B. Consequently, the prefactor of the $\omega^{-1/2}$ term increases with increasing c around the half-filled state of this system. If we could expect an analogous behavior in the two-dimensional case, the α term is considered to be larger in sample A than in sample B, in accordance with the present results. It should be noted, however, that Eq. (5) should be applied for the exchange narrowing case, which is different from the present one.

The characteristic decay time τ of the LTT due to the interlayer coupling can be estimated as follows [17]:

$$\tau^{-1} \approx [S(S+1)]^{1/2} \frac{J_c^2}{J_{ab}\hbar}, \tag{6}$$

where J_c and J_{ab} are the interlayer and intralayer exchange energies, respectively. If we adopt the values of J_c and J_{ab} in $\mathrm{La_2CuO_4}$ [18], the τ is estimated to be $\approx 10^{-6}$ sec, which is considerably larger than the typical NMR measurement time, $\omega_n^{-1} \approx 10^{-8}$ sec. Thus, in principle, the existence of the LTT behavior in high T_c oxides seems to be detectable by T_1, although no ω dependence has been observed in $YBa_2Cu_3O_y$ and $(La,Sr)_2CuO_4$ so far. Here we propose possible reasons for the absence of ω dependence in these oxides. In the case of $YBa_2Cu_3O_y$, the existence of the chain Cu site and CuO_2 bilayer may introduce three-dimensional coupling. Indeed, the antiferromagnetic coupling between the CuO_2 layers is confirmed by neutron scattering measurements [19]. On the other hand, the γ term is considered to be small in the light doped system of $(La,Sr)_2CuO_4$; thus the α term may be too small to observe the LTT behavior. We could observe the ω dependence of T_1 in $Tl_2Ba_2CuO_y$ because this compound has the well separated CuO_2 monolayer and the suitable carrier concentration.

In conclusion, we have reported the $\ln\omega^{-1}$ type ω dependence of T_1 in the two-dimensional magnetic system for the first time in a high T_c superconductor. A remarkable fact is that the long time tail behavior is found around $q \approx Q$ but not around $q \approx 0$, which is quite different from the usual cases. Recently, the confinement of charge freedom is suggested theoretically and experimentally in the high T_c oxides [13,20]. In this context, we have presented that the magnetic coupling between CuO_2 layers decreases with decreasing T in the normal state of the $Tl_2Ba_2CuO_y$ system, indicating that the confinement of spin freedom is enhanced in low temperature.

The authors thank Y. Itoh for stimulating discussions. This work has been supported by Grant-in-Aid for Scientific Research by the Ministry of Education, Science and Culture.

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