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Measurement of the q-dependent static spin susceptibility $\chi'(\mathbf{q})$ in $YBa_2Cu_3O_{6.9}$

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We report the temperature dependence of the **q**-dependent static electron spin susceptibility $\chi'(\mathbf{q})$ in a high-T_c superconductor $YBa_2Cu_3O_{6.9}$ obtained by measuring the Gaussian component of ^{63}Cu nuclear spin-spin relaxation rate $1/T_{2G}$. We employ the low-field NMR technique, and explain why this difficult technique is necessary for obtaining reliable results. It was found that $\chi'(\mathbf{q})$ satisfies a Curie-Weiss law at the corner of the Brillouin zone above T_c. Comparison with the spin-lattice relaxation rate $1/T_1$ showed little evidence for pseudogap behavior.

It is widely recognized that the normal-state properties of high-T_c superconductors are quite unusual. The magnetic properties probed by various experimental techniques including NMR and nuclear quadrupole resonance (NQR) indeed exhibit very anomalous results.[1] Most NMR and NQR measurements have been of the Knight shift or the spin-lattice relaxation rate $1/T_1$. For a metal, Moriya[2] showed that $1/T_1T$ probes the **q**-averaged imaginary part of the dynamical electron spin susceptibility $\chi''(\mathbf{q},\omega)$,

$$\frac{1}{T_1T}=\frac{2\gamma_n^2 k_B}{\gamma_e^2\hbar^2}\sum_{\mathbf{q}} f(\mathbf{q})\frac{\chi''(\mathbf{q},\omega_n)}{\omega_n}, \quad (1)$$

where ω_n and $\gamma_n(\gamma_e)$ are the resonance frequency and nuclear (electronic) gyromagnetic ratios, respectively, and $f(\mathbf{q})$ is a weighting factor called the form factor. Because of the site and wave-vector dependence of the form factor, ^{63}Cu probes $\chi''(\mathbf{q},\omega_n)$ primarily around $\mathbf{q}=\mathbf{Q}\equiv(\pi/a,\pi/a)$ (i.e., antiferromagnetic component, a is the lattice constant), while ^{17}O and ^{89}Y probes $\chi''(\mathbf{q},\omega_n)$ around $\mathbf{q}=0$ (i.e., long-wavelength component).[1,3] It has been established by the measurements of $^{63,17,89}1/T_1$ that $\chi''(\mathbf{q},\omega_n)$ exhibits different temperature dependences around $\mathbf{q}=\mathbf{Q}$ and $\mathbf{q}=0$.[1] A number of theoretical models have been proposed to account for the complex behavior of dynamical susceptibility $\chi(\mathbf{q},\omega)$ in the anomalous normal state of high-T_c superconductors.[4] It is important, therefore, to obtain independent measurements of $\chi(\mathbf{q},\omega)$ and provide a test for the various theories. The purpose of the present paper is to report the quantitative study of the temperature dependence of $\chi'(\mathbf{q})$ based on the measurements of the Gaussian component of the ^{63}Cu NMR spin-spin relaxation rate $1/T_{2G}$ for the planar Cu sites.

Earlier Pennington *et al.* reported measurements of $1/T_{2G}$ at 100 K using oriented single crystals.[5] Subsequently Pennington and Slichter theoretically showed that $1/T_{2G}$ gives a strong constraint on the **q** dependence of the real part of $\chi(\mathbf{q},\omega)$ at zero frequency limit, i.e., static electron spin susceptibility $\chi'(\mathbf{q})$.[6] The origin of the relation between $\chi'(\mathbf{q})$ and $1/T_{2G}$ is as follows.[6] First, $1/T_{2G}$ is related with the z component of the nuclear spin-spin coupling constant $a_{ij}^z=(a_{ij}^z)_{\rm ind}+(a_{ij}^z)_{\rm dip}$ by $1/T_{2G}=\{\sum_j[(a_{ij}^z)^2/8]\}^{1/2}$, where the suffices ind and dip stand for the contributions of the indirect and dipole coupling, respectively, and the summation over j is taken for the lattice.[7] The Ruderman-Kittel-Kasuya-Yosida-like indirect nuclear spin-spin coupling[8] $(a_{ij}^z)_{\rm ind}$ between nuclear spins is defined as

$$H_{12}=\mathbf{I}_z(\mathbf{r}_1)(a_{12}^z)_{\rm ind}\mathbf{I}_z(\mathbf{r}_2) =-\gamma_n^2\hbar^2\sum_{\mathbf{r},\mathbf{r}'}\mathbf{I}_z(\mathbf{r}_1)G(\mathbf{r}_1,\mathbf{r}')\chi'(\mathbf{r}',\mathbf{r}) \times G(\mathbf{r},\mathbf{r}_2)\mathbf{I}_z(\mathbf{r}_2). \quad (2)$$

In Eq. (2), $\mathbf{I}_z(\mathbf{r}_i)$ is a z component of the nuclear spin operator at position $\mathbf{r}_i$, $G(\mathbf{r},\mathbf{r}_1)=A_z\delta\mathbf{r},\mathbf{r}_1+\sum_{\mathbf{r}''}B\delta\mathbf{r},\mathbf{r}''$ is the Green function for the Mila-Rice hyperfine Hamiltonian[9] and $\mathbf{r}''$ runs over the four nearest-neighbor planar Cu sites of a Cu site at $\mathbf{r}_1$, A_z $(=-164\ \mathrm{KOe}/\mu_B)^{4(c)}$ and B $(=42\ \mathrm{KOe}/\mu_B)^{4(c)}$ are the z component of the on-site hyperfine coupling constant and the isotropic transferred hyperfine coupling constant, respectively. Writing the nonlocal electron spin susceptibility $\chi'(\mathbf{r}',\mathbf{r})$ as the Fourier transform of $\chi'(\mathbf{q})$, $\chi'(\mathbf{r}',\mathbf{r}) =\sum_{\mathbf{q}}e^{i\mathbf{q}(\mathbf{r}'-\mathbf{r})}\chi'(\mathbf{q})/N$, where N is the number of Cu atoms per unit volume, it is clear that one obtains information about the **q** dependence of $\chi'(\mathbf{q})$ from the measurement of $1/T_{2G}$.

To measure $1/T_{2G}$, it is essential that one flips not only the nuclei observed but all neighbors as well. Our earlier experiments on single crystals at 100 K had narrow enough lines to be reliable,[5] but the sample volume is so small that the signal-to-noise ratio is inadequate for the studies of the temperature dependence of $\chi'(\mathbf{q})$ above 100 K. To increase signal to noise, one goes to powders. If one employs nuclear quadrupole resonance (NQR),[10] there is no guarantee that one can flip all nuclear spins by the RF excitation pulse H_1 because of the great breadth of the resonance line profile of $YBa_2Cu_3O_x$ (typically more than 300 KHz). The dramatic change of the experimental results observed when H_1 is reduced[10(f),10(g)] as well as a number of contradictory results reported by various groups[10] should be attributed to the artifact caused by the broad linewidth of NQR.[10] Moreover the typical values of $1/T_{2G}$ measured by NQR (Ref. 10) are comparable or less than those determined by NMR,[5] nevertheless theoretically the former should be $\sqrt{2}$ times faster as pointed out by Song and Halperin.[10(h)] These facts clearly indicate that one cannot rely on NQR results. On the other hand, high-field ($\sim$80 KOe) NMR measurements for oriented ceramic powders suffer from a background signal arising from adjacent transitions and the line broadening at low temperatures. Therefore again there is no guarantee that high-field NMR measurements provide accurate values of $1/T_{2G}$, unless one confirms that the results agree with the single-crystal data.[11] We therefore carried out our NMR measurements of $1/T_{2G}$ by a method which avoids these difficulties. We observe the $I_z=\frac{1}{2}$ to $-\frac{1}{2}$ transition using oriented ceramic powders *at a low field* 6.5 KOe (7.3 MHz), so that the resonance linewidth is narrow enough ($\approx$20 KHz) for us to flip all nuclei. This method has another advantage that the background signal is reduced.

Some typical examples of the spin-echo decay $M_G(2\tau)$ observed at 7.3 MHz are presented in Fig. 1 as a function of the square of the separation time τ between the $\pi/2$ and π pulses, where $M_G(2\tau)$ is defined as $M(2\tau)=M_G(2\tau)\exp(-2\tau/T_{2R})$ and $M(2\tau)$ is the experimentally observed decay. The Lorentzian contribution of T_1 processes to the spin-echo decay, T_{2R}, was estimated based on the Redfield theory,[8] $1/T_{2R}=3(1/T_1)_c+(1/T_1)_{ab}=6.7(1/T_1)_c$,[5] where the suffices c and ab stand for the quantization axis of the nuclear spins and we inserted $(1/T_1)_{ab}/(1/T_1)_c=3.7$.[12] We confirmed that the results do not change even if we reduce H_1 by a factor of 2, in contrast with the dramatic change observed for NQR.[10(f),10(g)] Evidently the observed $M_G(2\tau)$ can be fitted to a Gaussian as expected, $M_G(2\tau)=\exp[-(2\tau)^2/(2T_{2G}^2)]$. We stress that the fitted value of $1/T_{2G}$ is rather insensitive to the choice of the value of $1/T_{2R}$, and varies no more than $\pm3\%$ even if $1/T_{2R}$ is changed by $\pm10\%$. It is also worth noting that the high-field NMR measurements for oriented powders by Imai *et al.*[11] turned out to yield nearly identical $M(2\tau)$ as those in Fig. 1. The short recovery time of the signal detection circuits after high-voltage pulses ($\approx$8 μsec) for the high-field measurements allowed them to fit $M(2\tau)$ by choosing both T_{2R} and T_{2G} as independent parameters, and the fitted value of T_{2R} agreed very well

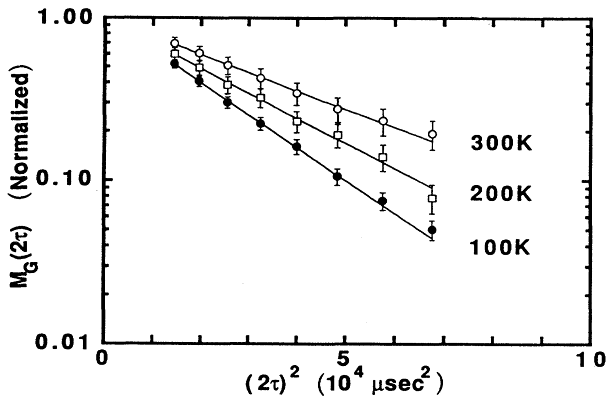

FIG. 1. Gaussian component of the spin-echo decay $M_G(2\tau)$.

with the theoretical values above 200 K where the linewidth is sufficiently narrow ($\approx$35 KHz).[11] Therefore our procedure inserting the theoretical value of T_{2R} in the fit of low-field NMR data should not cause any significant systematic errors to T_{2G}.

The temperature dependence of $1/T_{2G}$ determined by low-field NMR is presented in Fig. 2(a). The result at 100 K (9.6 m sec^{-1}) is in reasonable agreement with our previous single-crystal measurement (8.4 msec^{-1}).[5] We also found that random-phase-approximation (RPA) calculations by Bulut and Scalapino (not shown)[13] reproduce our results fairly well.

To analyze the result of $1/T_{2G}$ quantitatively, we employ the RPA form of the dynamical susceptibility following Millis, Monien, and Pines,[4(c)] $\chi_{AF}(\mathbf{q},\omega)=\chi_0(\mathbf{q},\omega)/[1-J_q\chi_0(\mathbf{q},\omega)]$, where the suffices AF and 0 stand for antiferromagnetic and noninteracting systems, respectively, and J_q is the Weiss molecular field interaction. Assuming an exponential decay with a $\mathbf{q}$-independent rate $1/\Gamma_0$ for electron spin relaxation processes of the noninteracting system, i.e., $\chi_0(\mathbf{q},\omega)=\chi_0(\mathbf{q},0)/[1-i\omega/\Gamma_0]$, $\chi_{AF}(\mathbf{q},\omega)$ may be written in the low-frequency limit as

$$\chi_{AF}(\mathbf{q},\omega)=\chi'_{AF}(\mathbf{q})+i\omega\frac{\chi'_{AF}(\mathbf{q})}{\Gamma(\mathbf{q})}\ , \tag{3}$$

where

$$\chi'_{AF}(\mathbf{q})=\frac{\chi'_0(\mathbf{q})}{[1-J_q\chi'_0(\mathbf{q})]}\ , \tag{4a}$$

$$\Gamma(\mathbf{q})=\Gamma_0[1-J_q\chi'_0(\mathbf{q})]\ . \tag{4b}$$

Then we expand $1-J_q\chi'_0(\mathbf{q})$ so that $\chi_{AF}(\mathbf{q},\omega)$ is peaked at $\mathbf{q}=\mathbf{Q}$.[1] In what follows, we utilize a Gaussian form, $[1-J_q\chi'_0(\mathbf{q})]=(\xi_0/\xi)^2\exp(|\mathbf{q}-\mathbf{Q}|^2\xi^2)$, where ξ is the antiferromagnetic correlation length and ξ_0 is another parameter to be determined by experiment. All that is assumed is a $\mathbf{q}$-dependent susceptibility which is peaked at $\mathbf{q}=\mathbf{Q}$. Even if one assumes Lorentzian form[4(c),6] instead of Gaussian, the final results are semiquantitatively the same. For $\chi'_0(\mathbf{Q})$, we substitute the calculated result of the Lindhard function by Xu *et al.*, 2.9 states/eV–both spins.[14] Since the value of $\chi'_{AF}(\mathbf{q})$ is vanishingly small at $\mathbf{q}=0$, we add a long-wavelength susceptibility $\chi'_{LW}(\mathbf{q})$ to satisfy the experimental constraint on the static susceptibility obtained by NMR shift measurements. We thus obtain the total susceptibility,

$$\chi'(\mathbf{q})=\chi'_{AF}(\mathbf{q})+\chi'_{LW}(\mathbf{q}) . \qquad (5)$$

We take $\chi'_{LW}(\mathbf{q})=2.53$ states/eV-both spins as its $\mathbf{q}=0$ value,[4(c)] neglecting its $\mathbf{q}$ dependence for simplicity. Since the $\mathbf{q}$ dependence of the calculated Lindhard function is only about 40% in the first Brillouin zone[14] and the value at $\mathbf{q}=\mathbf{Q}$ is an order of magnitude smaller than that of $\chi'_{AF}(\mathbf{q})$ as shown below, neglecting the $\mathbf{q}$ dependence of $\chi'_{LW}(\mathbf{q})$ will not affect our conclusions.

Calculations of $1/T_{2G}$ based on Eqs. (2), (4), and (5) are straightforward.[6] It was found from the fit of the data that two unknown parameters ξ and ξ_0 satisfy a simple relation at each temperature, $a\xi/\xi_0^2=c(T)$, where $c(T)$ is a dimensionless temperature-dependent parameter. The temperature dependences of $c(T)^{-2}$ and $c(T)^{-1}$ are shown in Fig. 2(b). Both quantities fit to a Curie-Weiss law well with a negative Weiss temperature as predicted by several theoretical models.[4(d),4(e)] Since $\chi'_{AF}(\mathbf{Q})$ is proportional to $c(T)^2$ or $c(T)^1$ when $\xi(T)$ or $\xi_0(T)$ dominates the temperature dependence of $1/T_{2G}$, we arrived at one of our most important results here: The staggered component of the static susceptibility $\chi'_{AF}(\mathbf{Q})$ satisfies a Curie-Weiss law, and the conclusion does not depend on the assumption of whether or not ξ is temperature dependent. The Stoner enhancement $\chi'_{AF}(\mathbf{Q})/\chi'_0(\mathbf{Q})$ for $\mathbf{q}=\mathbf{Q}$ is estimated to be $c(T)\xi/a\sim10$ (~20) for $\xi/a\sim1$ ($\xi/a\sim2$) indicating strong antiferromagnetic correlation. The negative Weiss temperature observed implies that the electronic system is not approaching toward antiferromagnetic ordering below 300 K. It is also worthwhile noting that a possible temperature dependence of ξ determined from $1/T_{2G}$ for a particular case of temperature-independent ξ_0 of $(a/\xi_0)^2\equiv7$, $[a/\xi(T)]^2=8.9\times10^{-4}(T+125)$, is in excellent agreement with that determined by the analysis of $1/T_1$ by Monien, Pines, and Takigawa.[4(c)]

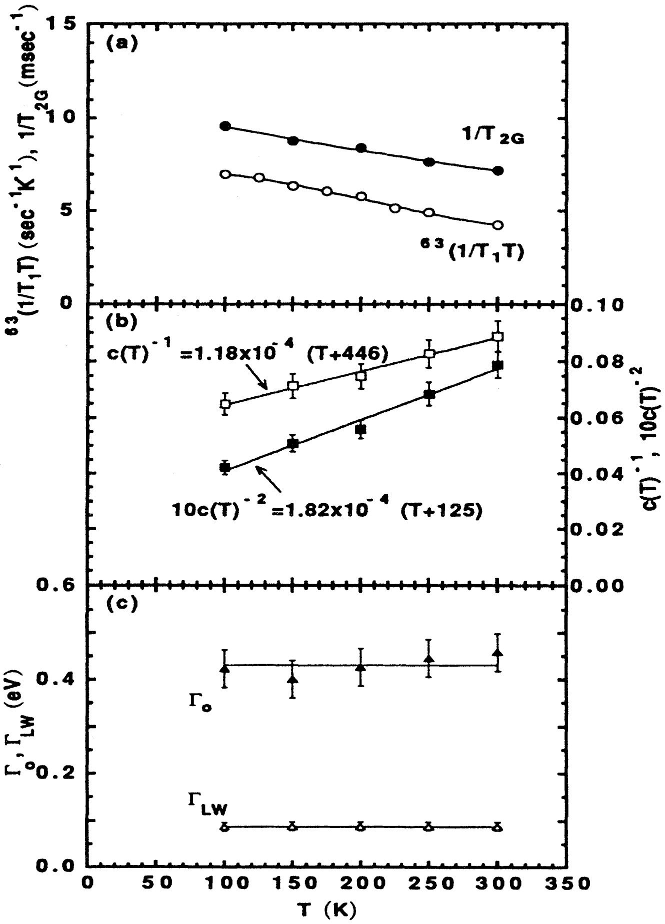

FIG. 2. (a) Temperature dependence of $^{63}1/T_{2G}$ and $^{63}1/T_1T$ at the planar Cu(2) sites. Experimental errors are about the size of the symbols. Solid curves are guides for eyes. (b) Temperature dependences of $c(T)^{-1}$ [$\approx\xi_0(T)^2/\xi$] and $c(T)^{-2}$ [$\approx\xi_0^4/\xi(T)^2$]. Solid lines are the fits to Curie-Weiss law. (c) Typical results of Γ_0 and Γ_{LW}, which are obtained for the case of temperature-dependent ξ, $[a/\xi(T)]^2=8.9\times10^{-4}(T+125)$. Solid lines are guides for the eye.

Besides information on $\chi'(\mathbf{q})$ itself, the NMR measurement of $1/T_{2G}$ is a clue for the energy scale of antiferromagnetic spin fluctuations $\hbar\Gamma(\mathbf{Q})$ when combined with the result of $1/T_1$, and provides a test for *the pseudogap*[15,16] as pointed out by Imai.[10(d)] Corresponding to Eq. (5), we take

$$\frac{\chi''(\mathbf{q},\omega)}{\omega}=\frac{\chi'_{AF}(\mathbf{q})}{\Gamma(\mathbf{q})}+\frac{\chi'_{LW}(\mathbf{q})}{\Gamma_{LW}} , \qquad (6)$$

where $\Gamma(\mathbf{q})=\Gamma_0(\xi_0/\xi)^2\exp(|\mathbf{q}-\mathbf{Q}|^2\xi^2)$.[17] Since we already know ξ and ξ_0 from $1/T_{2G}$, Γ_0 and Γ_{LW} are the two parameters to be determined by fitting $^{63,17,89}(1/T_1)$. A typical result is presented in Fig. 2(c), which indicates that Γ_0 is temperature independent within $\sim15\%$, and therefore $\hbar\Gamma(\mathbf{Q})=\hbar\Gamma_0(\xi_0/\xi)^2=\hbar\Gamma_0(a/\xi_0)^2c(T)^{-2}\approx20$ meV gradually decreases with temperature proportionally to $(\xi_0/\xi)^2$. Physically this decrease corresponds to the slowing down of spin fluctuations by a growing antiferromagnetic correlation, and is consistent with the total momentum sum rule $\sum_{\mathbf{q},\omega}\chi''(\mathbf{q},\omega)/[1-\exp(-\hbar\omega/k_BT)]=\text{const}$. Our finding that both $1/T_1$ and $1/T_{2G}$ can be fitted simultaneously with a simple RPA formalism with the essentially temperature-independent Γ_0 is in quite remarkable contrast with the case of $YBa_2Cu_4O_8$. Recently Itoh *et al.* reported that $1/T_{2G}$ for $YBa_2Cu_4O_8$ increases monotonically even below 200 K where $^{63}1/T_1T$ decreases.[10(g)] Their NQR results suggest that the RPA formalism does not work for the fit of $^{63}1/T_1T$ of $YBa_2Cu_4O_8$ unless Γ_0 is allowed to increase with lowering temperature. In other words, the growing antiferromagnetic correlation in $YBa_2Cu_4O_8$ does not reduce the characteristic energy $\hbar\Gamma(\mathbf{Q})$ as anticipated from the RPA theory. This is consistent with the observation of a pseudogap by inelastic-neutron-scattering experiments in reduced doping materials by

Rossat-Mignod *et al.*[15] However, since our result showed little indication of the opening of a pseudogap in $YBa_2Cu_3O_{6.9}$, the pseudogap phenomenon which has been detected so far for reduced doping material[15,16,10(g)] and has been attracting much theoretical attention may not have a fundamental connection with the mechanism of the high-temperature superconductivity.

In conclusion, we have demonstrated that low-field NMR measurements of the Gaussian component of ^{63}Cu nuclear spin-spin relaxation rate $1/T_{2G}$ shed much light on the dynamical spin susceptibility $\chi(\mathbf{q},\omega)$ of planar Cu sites in high-T_c oxides. Whether a particular theoretical model can reproduce the present results of $1/T_{2G}$ [i.e., $\chi'(\mathbf{q})$] quantitatively[13] as well as $1/T_1T$ [i.e., $\chi''(\mathbf{q},\omega)$] will be a crucial test for the model.

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