Transverse nuclear magnetic relaxation rate of the cuprate superconductors

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We simplify the Pennington-Slichter expression for the transverse NMR relaxation rate (T_{2G}^{-1}) in the cuprate superconductors, and use our result to calculate spin-fluctuation parameters in the normal state and T_{2G} for the superconducting state of $YBa_2Cu_3O_7$.

I. INTRODUCTION

In comparing theories[1,2] of the low-frequency magnetic behavior of the cuprate superconductors with experiment, the longitudinal nuclear magnetic relaxation rate at various nuclear sites (Cu, O, and Y) provides a number of significant constraints on the imaginary part of the spin-spin response function. Pennington and Slichter[3] have shown that the transverse relaxation rate provides a constraint on the real part of that response function. They concluded that the experimentally measured large transverse relaxation rate was due to an effective copper nuclear-nuclear spin coupling induced through an indirect interaction with electron spins. Itoh *et al.*[4] simplified the Pennington Slichter expression for T_{2G} for a spin susceptibility using the phenomenological expression for the spin susceptibility introduced by Millis, Monien, and Pines[1] (hereby referred to as MMP), an asymptotic expansion valid for large correlation lengths, and a continuum approximation. More recently, Imai and Slichter[5] have reported on measurements made at several temperatures in the normal state of $YBa_2Cu_3O_7$, and have used their results to discuss the consequences of that constraint on the parameters which enter into the MMP theory. In the present paper, we show how the Pennington-Slichter expression for the transverse relaxation rate may be simplified and use this result to calculate revised values for the parameters which characterize the normal-state spin-fluctuation excitation spectrum in the vicinity of the commensurate peak at $(\pi/a,\pi/a)$, and T_{2G} for the superconducting state of $YBa_2Cu_3O_7$.

II. EFFECTIVE NUCLEAR-NUCLEAR HAMILTONIAN AND TRANSVERSE NUCLEAR RELAXATION RATE OF $YBa_2Cu_3O_7$

As Pennington and Slichter have emphasized, the transverse nuclear relaxation rate for ^{63}Cu nuclei spins at site i, I_i, is described by the coupling to the planar electron spins S_i. We follow Mila and Rice[7] and write that coupling in the following form:

$$H_i = {}^{63}\gamma_n \gamma_e \hbar^2 I_z(\mathbf{r}_i)\cdot[A_{\parallel}(\mathbf{r}_i) + B\Sigma_\delta S_z(\mathbf{r}_{i+\delta})] , \quad (1)$$

where δ is the sum over nearest neighbors. The Fourier transform of this interaction then gives us the Mila-Rice form factor

$$F(\mathbf{q}) = A_{\parallel} + 2B[\cos(q_x a) + \cos(q_y a)] , \quad (2)$$

where a is the planar copper to copper lattice constant. The coupling of the nuclear spins is mediated by the electron spins so that the Hamiltonian describing the coupling of nuclear spins at positions $\mathbf{r}_1$ and $\mathbf{r}_2$, $I_z(\mathbf{r}_1)$ and $I_z(\mathbf{r}_2)$ respectively, is given by Imai and Slichter[5] to be

$$H_{12} = -({}^{63}\gamma_n \hbar)^2 \sum_{\mathbf{r}',\mathbf{r}} I_z(\mathbf{r}_2) F(\mathbf{r}_2,\mathbf{r}')\chi'(\mathbf{r}',\mathbf{r}) \times F(\mathbf{r},\mathbf{r}_1) I_z(\mathbf{r}_1) , \quad (3)$$

where $F(\mathbf{r},\mathbf{r}')$ is the Mila-Rice form factor in real space and $\chi(\mathbf{r}',\mathbf{r})$ is the Fourier transform of the real part of the electron spin susceptibility $\chi'(\mathbf{q})$

$$\chi(\mathbf{r}',\mathbf{r}) = \sum_{\mathbf{q}} e^{i\mathbf{q}\cdot(\mathbf{r}'-\mathbf{r})}\chi'(\mathbf{q})/N . \quad (4)$$

We show in the Appendix that the expression for the resulting Gaussian component of the transverse relaxation time takes the simple form

$$\left[\frac{1}{T_{2G}}\right]^2 = \frac{0.69({}^{63}\gamma_n \hbar)^4}{8\hbar^2}\left[\frac{1}{N}\sum_{\mathbf{q}} F(\mathbf{q})^4 \chi'(\mathbf{q})^2 - \left[\frac{1}{N}\sum_{\mathbf{q}} F(\mathbf{q})^2 \chi'(\mathbf{q})\right]^2\right] , \quad (5)$$

a result which has been independently obtained by Takigawa.[8] This expression simplifies considerably if one uses for spin-spin correlation function the phenomenological expression of Millis, Monien, and Pines[1] (hereby referred to as MMP).

$$\chi(\mathbf{q},\omega) = \frac{\chi_{\mathbf{Q}}}{1+(\mathbf{q}-\mathbf{Q})^2\xi^2 - i(\omega/\omega_{sf})} \equiv \frac{\chi_0\sqrt{\beta}(\xi/a)^2}{1+(\mathbf{q}-\mathbf{Q})^2\xi^2 - i(\omega/\omega_{sf})} , \quad (6)$$

where χ_0 is the static long-wavelength susceptibility in units of states/eV, $\chi_{\mathbf{Q}}$ is the corresponding static susceptibility at $\mathbf{Q}$, β reflects the scale of the antiferromagnetic enhancement, ξ is the correlation length, $\mathbf{Q}$ is the antiferromagnetic wave vector $(\pi/a,\pi/a)$, and

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$$\omega_{sf}=\Gamma_{AF}/[\sqrt{\beta}(\xi/a)^2\pi] \quad (7)$$

is the characteristic energy for spin fluctuations in the vicinity of **Q**. For $YBa_2Cu_3O_7$ in the expressions (6) and (7) only ξ varies with temperature. As shown in the Appendix, one finds in the limit of $\xi \gg a$, that

$$\left[\frac{1}{T_{2G}}\right]^2 \simeq \left[\frac{0.69({}^{63}\gamma_n\hbar)^4 F(\mathbf{Q})^4}{32\pi\hbar^2}\right] \times \left[\frac{\chi_Q^2(T)}{(\xi(T)/a)^2}=\beta\chi_0^2\left[\frac{\xi(T)}{a}\right]^2\right] . \quad (8)$$

It is instructive to compare this result with that for the longitudinal relaxation time at the copper sites, ${}^{63}T_1$, which involves many of these same quantities. In carrying out the comparison, it is useful to define the effective longitudinal rate with a static field applied perpendicular to the c direction by

$$\frac{1}{{}^{63}T_{1_\perp}^{\rm eff}}=\frac{2}{{}^{63}T_{1_\perp}}-\frac{1}{{}^{63}T_{1_\parallel}}=(2R_a-1)\frac{1}{{}^{63}T_{1_\parallel}} , \quad (9)$$

where $1/{}^{63}T_{1_\perp}$ and $1/{}^{63}T_{1_\parallel}$ are the copper longitudinal relaxation rates for static field applied perpendicular and parallel to the c direction and

$$R_a=\frac{(1/{}^{63}T_{1_\perp})}{(1/{}^{63}T_{1_\parallel})} .$$

The relaxation rate is then given by

$$\frac{1}{{}^{63}T_{1_\perp}^{\rm eff}T}=\frac{2k_b({}^{63}\gamma_n\hbar)^2}{\hbar}\sum_{\mathbf{q}} F(\mathbf{q})^2\frac{\chi''(\mathbf{q},\omega)}{\hbar\omega} ; \quad (10)$$

for MMP theory when $\xi \gg a$, one has

$$\frac{1}{{}^{63}T_{1_\perp}^{\rm eff}T}\simeq\frac{k_b({}^{63}\gamma_n\hbar)^2}{2\hbar}F(\mathbf{Q})^2 \times \left[\frac{\chi_Q}{\pi\hbar\omega_{sf}\xi^2}=\frac{\beta\chi_0(\xi/a)^2}{\hbar\Gamma_{AF}}\right] . \quad (11)$$

Because $1/T_{2G}^2$ and $1/{}^{63}T_{1_\perp}^{\rm eff}T$ are both proportional to the parameters β and ξ^2, one then has

$$\left[\frac{1}{T_{2G}}\right]^2 \simeq \left[\frac{0.69({}^{63}\gamma_n\hbar)^2F(\mathbf{Q})^2}{16\pi\hbar k_b}\right]\frac{2R_a-1}{{}^{63}T_{1_\parallel}T} \times(\chi_0\hbar\Gamma_{AF}=\chi_Q\hbar\omega_{sf}\pi) \quad (12)$$

so that in the limit of long correlation lengths, one can determine directly from experiment the product

$$F(\mathbf{Q})^2\chi_0\hbar\Gamma_{AF}=(8B)^2\chi_0\hbar\Gamma_{AF}$$

on making use of the Knight shift result, $A_\parallel=-4B$.

In the $YBa_2Cu_3O_7$ material, χ_0 is independent of temperature and Γ_{AF} is then found to be temperature independent from fits to the experimentally measured values of T_1. Experimentally, Imai and Slichter find the temperature dependence of $(1/T_{2G})^2$ to be nearly identical to that of $1/({}^{63}T_{1_\perp}^{\rm eff}T)$. Using $B=40.8$ KOe/μ_b and the results of Barrett *et al.*[9] for a temperature of 100 K,

$$1/({}^{63}T_{1_\parallel}T)=7.13(sK)^{-1}$$

and $R_a=3.74$, we find $\chi_0\hbar\Gamma_{AF}\simeq3.5$. Hence for $\chi_0=2.6$ states/eV, we find $\hbar\Gamma_{AF}=1.3$ eV, similar to the value found by Imai and Slichter[5] in their previous analysis.

This value of Γ_{AF} is considerably larger than that assumed by MMP, $\Gamma_{AF}\simeq0.4$ eV. As may be seen in Eq. (11), the various copper spin lattice relaxation rates depend on the product, $[\beta(\xi/a)^2/\Gamma_{AF}]$. If we substitute the result $\Gamma_{AF}=1.3$ eV into, for example, the expression for $({}^{63}T_\parallel)^{-1}$, we find

$$[\beta(\xi/a)^2]_{T=100\ \rm K}\simeq153 . \quad (13)$$

As discussed by Millis and Monien,[6] $(\xi/a)_{T=100\ \rm K}$ must be $\gtrsim 2$ for a spectrum of the MMP form to explain the large ratio ($\gtrsim 20$) of the copper to oxygen NMR spin lattice relaxation rates measured for a magnetic field directed along the c axis (otherwise the oxygen form factor would not produce sufficient cancellation of the antiferromagnetic spin fluctuation contribution). We adopt for β the value 32, since for this choice the resulting correlation length is essentially the same as that deduced in MMP.[1] A larger value of β in Eq. (13) would require a correlation length which is too short to explain the large ratio of the copper to oxygen NMR spin lattice relaxation rates, whereas a smaller value would lead to longer correlation lengths. The resulting values of $\xi(T)/a$, $\omega_{sf}(T)$, and $\chi_Q(T)$, obtained by neglecting logarithmic corrections and the quasiparticle contributions to $1/T_1$, are shown in Figs. 1–3.

III. THE TRANSVERSE RELAXATION TIME IN THE SUPERCONDUCTING STATE OF $YBa_2Cu_3O_7$

To calculate the Gaussian component of the transverse relaxation time in the superconducting state of $YBa_2Cu_3O_7$, we use the real part of the susceptibility calculated by Thelen, Pines, and Lu (Ref. 10) (hereafter re-

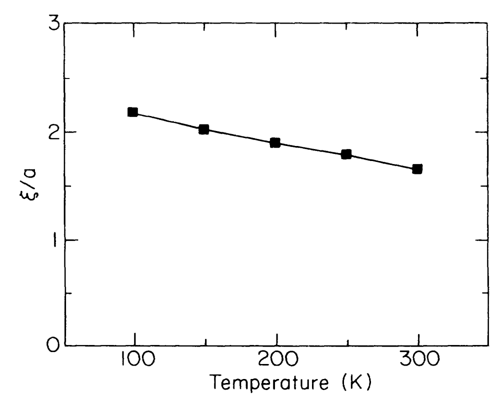

FIG. 1. The correlation length as a function of temperature for the normal state of $YBa_2Cu_3O_7$ as derived from ${}^{63}T_1$ and ${}^{63}T_{2G}$ relaxation rates measured by Imai and Slichter.[5]

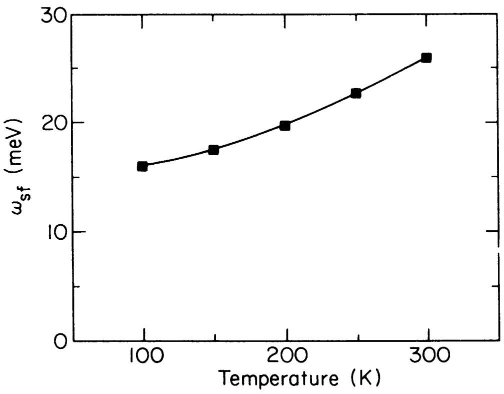

FIG. 2. The characteristic energy for spin fluctuations for the normal state of $YBa_2Cu_3O_7$ as a function of temperature derived from measurements of $^{63}T_1$ by Imai and Slichter.[5]

ferred to as TPL). TPL calculate the noninteracting susceptibility using a BCS d-wave expression with a band structure which includes nearest- and next-nearest-neighbor hopping; the interacting susceptibility is then calculated within the random-phase approximation (RPA) through the use of a momentum-dependent effective spin-spin interaction.

The real part of the TPL susceptibility for very low frequency is shown in Fig. 4 as a function of momentum at $T_c(=93$ K) and $T=40$ K. The real part of the susceptibility at low momentum vanishes at low temperatures due to the opening of the gap and agrees well with the susceptibility inferred from Knight shift experiments. At wave vectors that connect nodes of the d-wave gap function on the Fermi surface such as $(0.742\pi/a, 0.742\pi/a)$, no change in the real part of the susceptibility is found. At the antiferromagnetic wave vector, the real part of the susceptibility increases slightly as one enters the superconducting state. The physical origin of this increase is a combination of the reduction of the large quasiparticle scattering rate and the opening of the superconducting gap as one enters the superconducting state. Note that

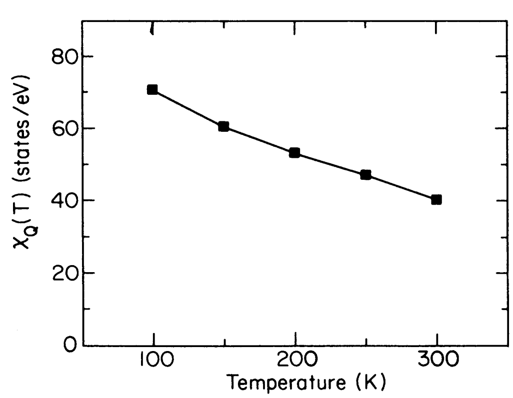

FIG. 3. The real part of the susceptibility at the antiferromagnetic wave vector as a function of temperature for the normal state of $YBa_2Cu_3O_7$ as derived from measurements of $^{63}T_{2G}$ by Imai and Slichter.[5]

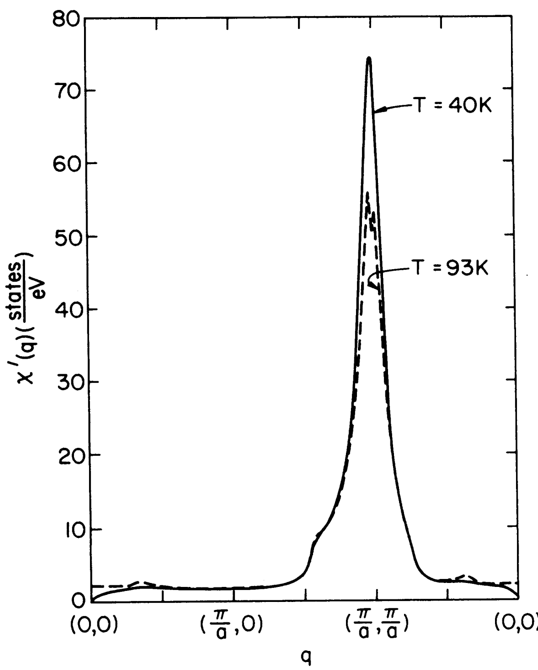

FIG. 4. The calculated real part of the susceptibility as a function of momentum for the superconducting state of $YBa_2Cu_3O_7$ using TPL theory[10] for a temperature of $T_c=93$ K (dashed line) and 40 K (solid line).

above T_c, the large quasiparticle scattering rate acts to suppress the susceptibility, as shown, for example, by the microscopic strong-coupling calculations of Monthoux and Pines.[11] If we take the scattering rate determined by microwave experiments of Bonn *et al.*,[12] we find the results for $1/T_{2G}$ shown in Fig. 5. There, we see that the predicted transverse relaxation rate $1/T_{2G}$, which is dominated by the susceptibility at the antiferromagnetic wave vector, increase by some 18% from its value at T_c. Also shown in Fig. 5 is the result we obtain if we use the TPL values for $\chi'(\mathbf{q})$ calculated assuming that the scattering rate is very small ($\ll k_b T_c$).

The transverse relaxation time in the superconducting state has also been calculated previously by Bulut and Scalapino.[13] Their model susceptibility was calculated with a band structure with only nearest-neighbor hopping and a momentum-independent interaction for both a d and s wave superconducting gap. In their theory, they choose the filling of their band to be near half-filling. This makes the antiferromagnetic wave vector connect nodes on the Fermi surface for a $d_{x^2-y^2}$ superconductor. Therefore, the real part of their susceptibility at the antiferromagnetic wave vector does not see the d-wave gap, and remains constant as one enters the superconducting state. The resultant $1/T_{2G}$ they predict for d-wave then decreases slightly as one enters the superconducting state with nearly the same temperature dependence as the TPL based calculation with a small scattering rate mentioned above. For s-wave pairing, Bulut and Scalapino find a decrease in the real part of the susceptibility for all momenta, including Q. Their corresponding calculated $1/T_{2G}$

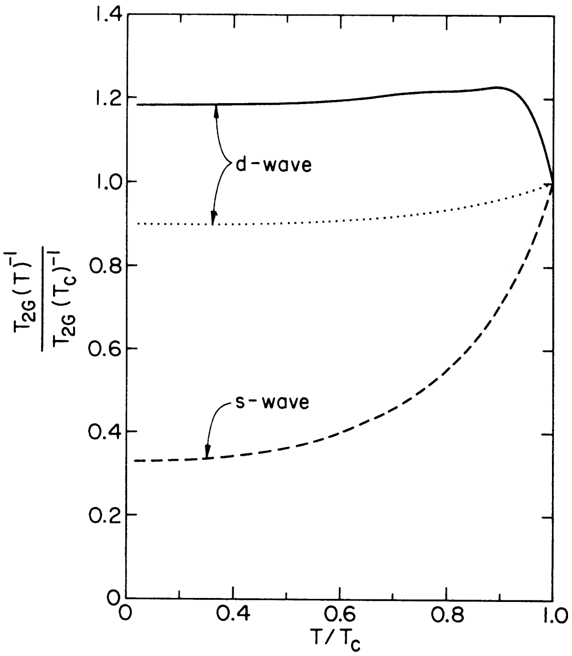

FIG. 5. The transverse nuclear magnetic relaxation rates in the superconducting state of $YBa_2Cu_3O_7$ as a function of temperature calculated using the *d*-wave theory of TPL (Ref. 10) for a temperature-dependent scattering rate (solid line) and a negligible scattering rate (dotted line). The dotted line is very similar to the *d*-wave calculation of Bulut *et al.* (Ref. 13). The calculated $1/T_{2G}$ for *s*-wave theory from the calculations of Bulut *et al.* (Ref. 13) is also shown (dashed line).

for an *s*-wave pairing gap then decreases substantially as one enters the superconducting state as shown in Fig. 5.

Experimentally, it has not yet proved possible to obtain an accurate measurement of $1/T_{2G}$ in the superconducting state. This difficulty is due to the stricter requirements needed to flip spins in the NMR experiment for T_{2G} where the interaction is nuclear spin-spin coupling than for the T_1 experiment where the interaction is between a nuclear spin and electron spin. Theoretically, the large decrease in the *s*-wave $1/T_{2G}$ as one enters the superconducting state would clearly distinguish between *s* and *d*-wave states. This temperature dependence of T_{2G} can more clearly determine the pairing state than a T_1 experiment because, in RPA, the temperature dependence of T_{2G} only depends on the real part of the noninteracting electron spin susceptibility, whereas a T_1 experiment is influenced by the real and imaginary parts of the noninteracting electron spin susceptibility. As a result, T_{2G} depends less on the details of the theory but depends principally on whether the pairing state is *s* or *d* wave.

IV. CONCLUSION

We have shown that an accurate measurement of the temperature dependence of T_{2G} in the superconducting state of $YBa_2Cu_3O_7$ will provide valuable information on the superconducting pairing state. We have used the constraint provided by the T_{2G} measurements of Imai and Slichter[5] to obtain revised values of the parameters that characterize the spin-fluctuation excitation spectrum in the vicinity of $(\pi/a,\pi/a)$. In a forthcoming paper, Monthoux and Pines[14] use this new spectrum to calculate T_c, the normal-state resistivity, and optical properties of $YBa_2Cu_3O_7$; they find that it leads to an improved agreement with experiment for the later two quantities.

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APPENDIX

The effective planar copper nuclear-nuclear spin Hamiltonian is[5]

$$H_{12} = -({}^{63}\gamma_n \hbar)^2 \sum_{\mathbf{r}',\mathbf{r}} I_z(\mathbf{r}_2) F(\mathbf{r}_2,\mathbf{r}') \chi'(\mathbf{r}',\mathbf{r}) F(\mathbf{r},\mathbf{r}_1) I_z(\mathbf{r}_1) \ , \tag{A1}$$

where $\chi(\mathbf{r}',\mathbf{r})$ is the Fourier transform of $\chi'(\mathbf{q})$

$$\chi(\mathbf{r}',\mathbf{r}) = \sum_{\mathbf{q}} e^{i\mathbf{q}\cdot(\mathbf{r}'-\mathbf{r})} \chi'(\mathbf{q})/N \ . \tag{A2}$$

Defining a_{12}^z by the equation

$$H_{12} = a_{12}^z I_z(\mathbf{r}_1) I_z(\mathbf{r}_2) \tag{A3}$$

gives

$$a_{12}^z = -\frac{({}^{63}\gamma_n \hbar)^2}{N} \sum_{\mathbf{r}',\mathbf{r},\mathbf{q}} F(\mathbf{r}_2,\mathbf{r}') e^{i\mathbf{q}\cdot(\mathbf{r}'-\mathbf{r})} \times \chi'(\mathbf{q}) F(\mathbf{r},\mathbf{r}_1) \ . \tag{A4}$$

The Gaussian component of the transverse relaxation rate squared is then given by Pennington and Slichter[3] to be

$$\left[\frac{1}{T_{2G}}\right]^2 = \frac{0.69}{8\hbar^2} \sum_{\mathbf{r}_2 \neq \mathbf{r}_1} (a_{12}^z)^2 \tag{A5}$$

for the $(\frac{1}{2},-\frac{1}{2})$ transition. The factor of 0.69 takes into account the natural abundance of the ${}^{63}Cu$ isotope. Here, we will simplify the expressions for a_{12}^z and $1/T_{2G}$. Equation (A4) can be rewritten as

$$a_{12}^z = -\frac{({}^{63}\gamma_n \hbar)^2}{N} \sum_{\mathbf{q}} e^{i\mathbf{q}\cdot(\mathbf{r}_2-\mathbf{r}_1)} \chi'(\mathbf{q}) \sum_{\mathbf{r}'} f(\mathbf{r}_2,\mathbf{r}') e^{i\mathbf{q}\cdot(\mathbf{r}'-\mathbf{r}_2)} \sum_{\mathbf{r}} F(\mathbf{r},\mathbf{r}_1) e^{i\mathbf{q}\cdot(\mathbf{r}_1-\mathbf{r})} \ . \tag{A6}$$

It then follows that

$$a_{12}^{z}=-\frac{({}^{63}\gamma_n\hbar)^2}{N}\sum_{\mathbf{q}}e^{i\mathbf{q}\cdot(\mathbf{r}_2-\mathbf{r}_1)}F(\mathbf{q})^2\chi'(\mathbf{q}) . \tag{A7}$$

To simplify the expression for $(1/T_{2G})^2$, one can rewrite Eq. (A5) to eliminate the restricted sum over $\mathbf{r}_2$ so that

$$\left[\frac{1}{T_{2G}}\right]^2=\frac{0.69}{8\hbar^2}\left[\sum_{\mathbf{r}_2}(a_{12}^{z})^2-(a_{11}^{z})^2\right] . \tag{A8}$$

The expression for $\sum_{\mathbf{r}_2}(a_{12}^{z})^2$ can be simplified by substituting for a_{12}^{z} from Eq. (A7):

$$\sum_{\mathbf{r}_2}(a_{12}^{z})^2=\sum_{\mathbf{r}_2}\left[-\frac{({}^{63}\gamma_n\hbar)^2}{N}\sum_{\mathbf{q}}e^{i\mathbf{q}\cdot(\mathbf{r}_2-\mathbf{r}_1)}F(\mathbf{q})^2\chi'(\mathbf{q})\right]\left[-\frac{({}^{63}\gamma_n\hbar)^2}{N}\sum_{\mathbf{q}'}e^{-i\mathbf{q}'\cdot(\mathbf{r}_2-\mathbf{r}_1)}F(\mathbf{q}')^2\chi'(\mathbf{q}')\right] , \tag{A9}$$

$$\sum_{\mathbf{r}_2}(a_{12}^{z})^2=\frac{({}^{63}\gamma_n\hbar)^4}{N^2}\sum_{\mathbf{q},\mathbf{q}'}F(\mathbf{q})^2\chi'(\mathbf{q})F(\mathbf{q}')^2\chi'(\mathbf{q}')e^{-i(\mathbf{q}-\mathbf{q}')\cdot\mathbf{r}_1}\sum_{\mathbf{r}_2}e^{i(\mathbf{q}-\mathbf{q}')\cdot\mathbf{r}_2} . \tag{A10}$$

Now since $\sum_{\mathbf{r}_2}e^{i(\mathbf{q}-\mathbf{q}')\cdot\mathbf{r}_2}=N\delta_{\mathbf{q}\mathbf{q}'}$,

$$\sum_{\mathbf{r}_2}(a_{12}^{z})^2=\frac{({}^{63}\gamma_n\hbar)^4}{N}\sum_{\mathbf{q}}F(\mathbf{q})^4\chi'(\mathbf{q})^2 . \tag{A11}$$

Substituting into Eq. (A8) one finds

$$\left[\frac{1}{T_{2G}}\right]^2=\frac{0.69({}^{63}\gamma_n\hbar)^4}{8\hbar^2}\left[\frac{1}{N}\sum_{\mathbf{q}}F(\mathbf{q})^4\chi'(\mathbf{q})^2-\left[\frac{1}{N}\sum_{\mathbf{q}}F(\mathbf{q})^2\chi'(\mathbf{q})\right]^2\right] , \tag{A12}$$

a result which has been obtained independently by Takigawa.[8] One can then substitute in the real part of the susceptibility $\chi'(\mathbf{q})$ at low frequency into Eq. (A12) for any normal or superconducting state theory. Here, we will substitute the real part of the normal-state susceptibility as given by the phenomenological theory of Millis, Monien, and Pines.[1] This theory has a spin susceptibility near the antiferromagnetic wave vector $\mathbf{Q}=(\pi/a,\pi/a)$ of

$$\chi'(\mathbf{q}\approx\mathbf{Q})=\frac{\chi_{\mathbf{Q}}}{1+\xi^2(\mathbf{q}-\mathbf{Q})^2}=\frac{\sqrt{\beta}(\xi/a)^2\chi_0}{1+\xi^2(\mathbf{q}-\mathbf{Q})^2} . \tag{A13}$$

With this susceptibility, the calculation of $1/T_{2G}$ from Eq. (A12) is dominated by the first integral in parentheses which involves the susceptibility squared. $1/T_{2G}$ is then approximately

$$\left[\frac{1}{T_{2g}}\right]^2\simeq\frac{0.69({}^{63}\gamma_n\hbar)^4}{8\hbar^2}\frac{1}{N}\times\sum_{\mathbf{q}}F(\mathbf{q})^4\frac{\chi_{\mathbf{Q}}^2}{[1+\xi^2(\mathbf{q}-\mathbf{Q})^2]^2} . \tag{A14}$$

This integral is dominated by contributions coming from the vicinity of the antiferromagnetic wave vector; when $\xi\gg a$, one finds

$$\left[\frac{1}{T_{2G}}\right]^2\simeq\frac{0.69({}^{63}\gamma_n\hbar)^4F(\mathbf{Q})^4}{32\pi\hbar^2}\times\left[\frac{\chi_{\mathbf{Q}}^2}{(\xi/a)^2}=\beta\chi_0^2\left[\frac{\xi}{a}\right]^2\right] . \tag{A15}$$

An expression of the form of Eq. (A15) has been obtained previously by Itoh *et al.*[4]; however, their prefactor differs from that found here, perhaps because they replaced the latter sums by a continuum approximation.

We thus see that in the long correlation length limit, $(T_{2G})^{-1}$ provides a direct measurement of $(\chi_{\mathbf{Q}}/\xi)$; while $(1/{}^{63}T_1T)$ measures $\chi_{\mathbf{Q}}/\omega_{sf}\xi^2$. Hence, from their ratio, one may obtain $\omega_{sf}\xi$, while the ratio $({}^{63}T_1T/T_{2G}^2)$ yields the product $\omega_{sf}(T)\chi_{\mathbf{Q}}(T)$.

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