Application of the antiferromagnetic-Fermi-liquid theory to NMR experiments on $YBa_2Cu_3O_{6.63}$

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NMR experiments on the $YBa_2Cu_3O_{6.63}$ material provide an important test of the antiferromagnetic-Fermi-liquid model for the spin-spin correlation function proposed by Millis, Monien, and Pines. We show that their theory provides a quantitative fit, with parameters determined from experiment, to the NMR experiments of Takigawa *et al.* on the Cu(2),O(2,3) nuclei, and of Alloul *et al.* on the Y nuclei in the $YBa_2Cu_3O_{6.63}$ material. We find that the hyperfine couplings do not change in going from the $YBa_2Cu_3O_7$ to the $YBa_2Cu_3O_{6.63}$ material, whereas the antiferromagnetic correlation length is increased. We present results for the changes in relevant magnetic parameters brought about by reducing the oxygen content from $YBa_2Cu_3O_7$ to $YBa_2Cu_3O_{6.63}$ and discuss the implications of a small energy scale $\hbar\omega_{SF} < k_B T_c$ emerging from the analysis of the NMR experiments. Our results support the proposal by Pines that the excitations in the normal state of superconducting Y-Ba-Cu-O are best described as those of an antiferromagnetic Fermi liquid and suggest that this description is equally applicable to other cuprate oxide superconductors.

I. INTRODUCTION

The nature of the normal state of the cuprate oxide superconductors has remained a key issue since their discovery over three years ago. Models that have been proposed and discussed in varying detail, including the resonating-valence-bond model, normal Fermi liquids, spin liquids, and more recently "Luttinger" liquids, marginal Fermi liquids, and nearly antiferromagnetic Fermi liquids, as well as both one-component and two-component descriptions.[1] Experiments must play the key role in deciding this question, but the question remains as to what experiment or experiments will prove to be the Rosetta stone. During the past eighteen months, NMR measurements on $YBa_2Cu_3O_7$, which provide strong constraints on the description of the low-frequency spin excitations, have emerged as a promising candidate.[2–7] These show that antiferromagnetic spin correlations are important; the Cu spin-lattice relation rate is enhanced by antiferromagnetic spin correlations,[2,3] while the markedly different temperature dependence of the ^{17}O spin relaxation rate from that measured for ^{63}Cu nuclei, in a one-component description, can only be explained as resulting from strong antiferromagnetic (AF) correlations in combination with the form factors of the different nuclei.[7,8]

A quantitative account of the role played by antiferromagnetic correlations in the spin-lattice relaxation of Cu, O, and Y nuclei in $YBa_2Cu_3O_7$ has been developed by Millis, Monien, and Pines,[9] hereafter referred to as MMP. They proposed a phenomenological model in which the leading role played by antiferromagnetic correlations is explicitly recognized by describing the system as *nearly antiferromagnetic Fermi liquid* (AFL), in which the dominant contribution to the short-wavelength part of the spin-spin correlations comes from antiferromagnetic paramagnons, whose characteristic energies are small compared to those that characterize the longer-wavelength more nearly quasiparticle behavior. Around the same time Bulut *et al.*[10] were able to show using random-phase approximation (RPA) for the Hubbard model that very close to the antiferromagnetic instability the antiferromagnetic correlations due to nesting give qualitatively the right behavior for the Cu and O relaxation rates. These results left open the question whether a one-component description was necessary, or simply sufficient,[11] and of the doping dependence of the spin-spin correlations. In principal, doping might affect the matrix elements of the hyperfine coupling as well as the spin-spin correlations. It is important to find out which changes in the NMR experiments with doping reflect a change in the properties of the fundamental excitations and which reflect a change in the hyperfine Hamiltonian.

The recent NMR measurements on the oxygen-deficient Y-Ba-Cu-O materials by Warren *et al.*,[12] Alloul, Ohno, and Mendels,[13] Shimizu *et al.*,[14] Yasuoka, Imai, and Shimizu,[15] Walstedt *et al.*,[16] Takigawa,[17] and Takigawa *et al.*,[18] not only provide an answer to the above questions, but offer a significant test of the applicability of antiferromagnetic-Fermi-liquid theory to the cuprate superconductors. The experiments of Shimizu *et al.*[14] and Walstedt *et al.*[16] on the ^{63}Cu Knight shift show that the planar static susceptibility of the Cu^{2+} spins is strongly temperature dependent and that the temperature dependence of the ^{63}Cu relaxation rate is markedly different from that found for the $YBa_2Cu_3O_7$ material. As we shall see, both of these results are a natural consequence of the MMP antiferromagnetic-Fermi-liquid theory. Takigawa *et al.*[18] have measured not only the ^{63}Cu Knight shift and relaxation rate (with results which are quite similar to those of Walstedt *et al.*), but the corresponding quantities for the ^{17}O nuclei. They find that while the Knight shift of the Cu and O nuclei have the same temperature dependence, the spin-lattice relaxation rates of these nuclei display a quite different temperature dependence. Moreover, for a suitable choice of the chemical shift of the ^{89}Y nuclei (which agrees with that suggested by Walstedt and Warren), the temperature dependence of the ^{89}Y Knight shift may be shown to be identical to that found by Takigawa *et al.* for the Cu and O nuclei. We conclude from the similarity in the temperature dependence of the Knight shift of the Cu, O, and Y nuclei that the measurements of Takigawa *et al.*[18] and Alloul, Ohno, and Mendels[13] provide direct and decisive evidence for a one-component description of the planar spin excitations, and we use these results to demonstrate that the hyperfine couplings, within experimental accuracy, do not change in going from $YBa_2Cu_3O_7$ to $YBa_2Cu_3O_{6.63}$ and to obtain the planar spin susceptibility.

Since there is only one spin degree of freedom, one is forced to the conclusion that in the $YBa_2Cu_3O_{6.63}$ material, as in the $YBa_2Cu_3O_7$ material, the dynamics of the Cu, O, and Y nuclear spins differ because the form factors lead the spin to sample different parts of the *same* spin-spin correlation function. The question remains, can one understand *quantitatively* the different temperature dependence of the relaxation rates of the Cu, O, *and* Y nuclei. We show in this paper that *antiferromagnetic-Fermi-liquid theory* provides such a quantitative account, and that the relaxation rates of the Cu, O, and Y nuclei serve uniquely to determine the temperature-dependent phenomenological parameters of the theory. Our fit to experiment enables us to demonstrate that the antiferromagnetic correlations are maximum at the zone corner $(\pi/a,\pi/a)$, in $YBa_2Cu_3O_{6.63}$, as is the case for the $YBa_2Cu_3O_7$ material, and provides a number of instructive comparisons between the phenomenological parameters that characterize the low-frequency spin excitation spectra of the two materials.

The paper is organized as follows. In Sec. II we discuss the hyperfine Hamiltonian, which is essential for the understanding of the anisotropy and magnitude of the Knight shifts and relaxation rates, and introduce our phenomenological model for the spin-spin correlation function $\chi(\mathbf{q},\omega)$. In Sec. III we discuss the Knight-shift experiments on Cu and O nuclei and the determination of the hyperfine couplings from the Knight-shift experiments and the resonance frequency of the Cu in the antiferromagnetic state. In Sec. IV we present the results of the MMP model for the relaxation rates and show how it provides a quantitative determination of the role played by antiferromagnetic correlations in determining the relaxation rates. In the following section we calculate the ^{63}Cu and ^{17}O relaxation rates and compare our results with measurements by Takigawa *et al.*[18] We then show how the experimental results of Alloul, Ohno, and Mendels[13] on the ^{89}Y Knight shift and relaxation rate may be combined with those of Takigawa *et al.* to determine uniquely the temperature dependence of the parameters that enter antiferromagnetic-Fermi-liquid theory. In Sec. VI we discuss antiferromagnetic-Fermi-liquid theory, and the physical origin of the measured changes in the basic parameters of the theory with temperature and oxygen content. We discuss related work in Sec. VII and present our conclusions in Sec. VIII.

II. THE SPIN-SPIN CORRELATION FUNCTION AND THE HYPERFINE HAMILTONIAN

We assume that there is only one electron spin $\mathbf{S}$ per unit cell composed of the planar Cu and O spins. This spin may interact with the nucleus at the same site (direct hyperfine coupling) or with a nucleus at a different site (transferred hyperfine coupling). We will assume, following Mila and Rice[19] and MMP, that the Cu nuclear spin $\mathbf{I}$ has a direct as well as a transferred hyperfine coupling, while the other nuclei, oxygen and yttrium, couple to the electron spin only via a transferred hyperfine coupling. With these assumptions we can write down the hyperfine Hamiltonian for the planar Cu and O atoms and the interplanar yttrium:

$$H_{\rm hf}=\sum_{i,\alpha}{}^{63}I_{i,\alpha}A_{\alpha\alpha}S_{i,\alpha}+\sum_{\langle ij\rangle,\alpha}{}^{63}I_{i,\alpha}BS_{j,\alpha}+\sum_{\langle ij\rangle,\alpha}{}^{17}I_{i,\alpha}CS_{j,\alpha}+\sum_{\langle ij\rangle,\alpha}{}^{89}I_{i,\alpha}DS_{j,\alpha}\ , \tag{2.1}$$

where $A_{\alpha\alpha}$ is the Cu direct hyperfine tensor with the two components $A_\perp$ and $A_\parallel$ and B is the transferred hyperfine coupling of the Cu nucleus. C denotes the oxygen and D the yttrium hyperfine coupling. $I_{i,\alpha}$ is the αth component of the nuclear spin at the site i, and $\langle ij\rangle$ is the sum over the nearest neighbors j. We would like to emphasize that the hyperfine couplings are material constants and not parameters.

The electron spins $\mathbf{S}$ are assumed to be at the Cu sites, and to interact antiferromagnetically with a finite temperature-dependent correlation length $\xi(T)$ which increases with decreasing temperature. MMP model the spin correlation function by two separate parts. One describes the more normal Fermi-liquid-like or quasiparticle part $\chi_{\rm QP}$ and the other more important part, $\chi_{\rm AF}$, describes the short-wavelength antiferromagnetic correlations; thus

$$\chi(\mathbf{q},\omega)=\chi_{\rm QP}(\mathbf{q},\omega)+\chi_{\rm AF}(\mathbf{q},\omega)\ . \tag{2.2}$$

We denote the real part of the quasiparticle-like contribution to the susceptibility, $\chi_{\rm QP}$, as $\bar{\chi}_0=\chi_{\rm QP}(\mathbf{q}=0,\ \omega=0)$. We will assume that the imaginary part of $\chi_{\rm QP}$ is related to the real part for small energies, ω, by

$${\rm Im}(\chi_{\rm QP})\cong\frac{\pi\omega}{\Gamma}\bar{\chi}_0\ , \tag{2.3}$$

independent of the wave vector $\mathbf{q}$. Here Γ is the characteristic spin fluctuation energy for the quasiparticle part. The antiferromagnetic part of the spin-spin correlation function can be modeled around the antiferromagnetic wave vector $\mathbf{Q}=(\pi/a,\pi/a)$ by (see, e.g., Ref. 9):

$$\chi_{\rm AF}(\mathbf{q},\omega)=\frac{\chi_Q}{1+\xi^2(\mathbf{Q}-\mathbf{q})^2-i(\omega/\omega_{\rm SF})}\ ; \tag{2.4}$$

here χ_Q is the static spin susceptibility at the antiferromagnetic wave vector $\mathbf{Q}$, which is related to the static susceptibility at $\mathbf{q}=0$ by $\chi_Q=\bar{\chi}_0(\xi/\xi_0)^2$, ξ is the antiferromagnetic correlation length, and $1/\xi_0$ is the wave vector at which the antiferromagnetic part of the correlation function, χ_{AF}, starts to dominate the quasiparticle contribution. We can relate the size of the quasiparticle contribution to the measured static susceptibility χ_0 with the aid of Eq. (2.2):

$$\chi_0=\chi_{QP}(0,0)+\chi_{AF}(0,0)=\bar{\chi}_0\left[1+\frac{1}{2\pi^2}\left[\frac{a}{\xi_0}\right]^2\right] . \quad (2.5)$$

For a typical value of $(a/\xi_0)^2\approx 3$ we find that the quasiparticle bit contributes 87% of the static susceptibility, whereas the antiferromagnetic part contributes some 13% at the wave vector $\mathbf{q}=0$.

$\hbar\omega_{SF}$ is a typical energy scale for the antiferromagnetic paramagnons that describe the AF spin dynamics. It can be very small, since it is related to the energy scale of the spin dynamics of the noninteracting system, Γ, by

$$\omega_{SF}=\frac{\Gamma}{\pi}\left[\frac{\xi_0}{\xi}\right]^2 . \quad (2.6)$$

If we identify the expression (2.4) with the spin-spin correlation function of a Fermi gas we can identity Γ with $\Gamma\sim v_F Q\sim \varepsilon_F$. For details we refer the reader to MMP. As we shall see, over a considerable temperature range the temperature dependence of the antiferromagnetic correlation length is described by the following expression:

$$\left[\frac{\xi(T)}{a}\right]^2=\left[\frac{\xi}{a}\right]^2_{T=0}\frac{|T_x|}{T_x+T} . \quad (2.7)$$

with temperature scale T_x for the variations of the antiferromagnetic correlation length, which is small compared to T_c. Under these circumstances the energy scale of the spin-spin correlation function is proportional to the temperature

$$\omega_{SF}\cong\frac{\Gamma}{\pi}\left[\frac{\xi_0}{\xi(0)}\right]^2\frac{T}{|T_x|}\sim T , \quad (2.8)$$

which implies that in the low-energy regime the imaginary part of the average paramagnon contribution to the spin-spin correlation function goes like ω/T. Varma *et al.*[20] proposed such behavior for the imaginary part of the charge (density-density) correlation function for low energies but did not allow for the substantial differences in magnitude between the charge and spin-correlation function at low energies that emerges in antiferromagnetic-Fermi-liquid theory.

III. THE KNIGHT-SHIFT EXPERIMENTS

In a one-band picture the Knight shifts of *all* nuclei scale the same way. With the hyperfine Hamiltonian equation (2.1) the Knight shifts are given by

$$^{63}K_{\parallel}=\frac{A_{\parallel}+4B}{^{63}\gamma_n\gamma_e\hbar^2}\chi_0 , \quad (3.1a)$$

$$^{63}K_{\perp}=\frac{A_{\perp}+4B}{^{63}\gamma_n\gamma_e\hbar^2}\chi_0 , \quad (3.1b)$$

$$^{17}K_{iso}=\frac{2C}{^{17}\gamma_n\gamma_e\hbar^2}\chi_0 , \quad (3.1c)$$

$$^{89}K_{iso}=\frac{8D}{^{89}\gamma_n\gamma_e\hbar^2}\chi_0 , \quad (3.1d)$$

where the γ_n are the various nuclear magnetic moments and γ_e the electron magnetic moment. There is no way of checking the one-band picture for the $YBa_2Cu_3O_7$ material, since the Knight shifts, Cu as well as O, are all temperature independent, a result that is consistent with a one-component picture but does not prove it. However, in the $YBa_2Cu_3O_{6.63}$ material, the Knight shifts, measured by Takigawa *et al.*,[18] are all temperature dependent with the same temperature dependence as shown in Fig. 1. Takigawa *et al.*[18] find that for $T > 60$ K, the temperature dependence of the total frequency shifts can be expressed in terms of $^{17}K_c(T)$ as

$$^{63}K_{ab}(T)=1.522\,^{17}K_c(T)+0.32\% , \quad (3.2a)$$

$$^{17}K_{iso}(T)=1.057\,^{17}K_c(T)+0.039\% , \quad (3.2b)$$

$$^{17}K_{ax}(T)=0.189\,^{17}K_c(T)+0.015\% . \quad (3.2c)$$

After subtracting the temperature-independent chemical shift all Knight shifts are proportional to each other with a temperature-independent proportionality constant that is given by the ratio of the hyperfine couplings. Thus the Knight-shift experiments of Takigawa *et al.*[18] demonstrate unambiguously that there is only *one* spin degree of freedom determining the response for *all* nuclei in the long-wavelength limit ($\mathbf{q}\rightarrow 0$).

The Cu Knight shift in the c direction does not vary with temperature within experimental accuracy. Thus it can only consist of the chemical shift. This proves that the delicate cancellation of the direct and transferred hyperfine coupling strength of the planar Cu(2) nucleus, found for $YBa_2Cu_3O_7$,[9] persists in the $YBa_2Cu_3O_{6.63}$ material. Therefore we will assume that $A_{\parallel}\approx -4B$. Repeating a similar analysis to that in Ref. 9 we find, on comparing the Cu and O Knight shift, that the oxygen hyperfine coupling C in the $YBa_2Cu_3O_{6.63}$ material is

$$C=0.18(A_{\perp}+4B) , \quad (3.3)$$

which has to be compared to the result previously obtained by MMP for the $YBa_2Cu_3O_7$ material:

$$C=(0.18\pm 0.01)(A_{\perp}+4B) . \quad (3.4)$$

The value of the transferred hyperfine coupling B can be obtained from the Cu nuclear resonance frequency in the *antiferromagnetic* state (see, e.g., Ref. 21) if we assume that the transferred hyperfine coupling does not change substantially with doping. From the experimental data of Yasuoka *et al.*[22] we obtain

$$\mu_{eff}|A_{\perp}-4B|=80 \quad (3.5)$$

in kOe. If we accept the effective magnetic moment

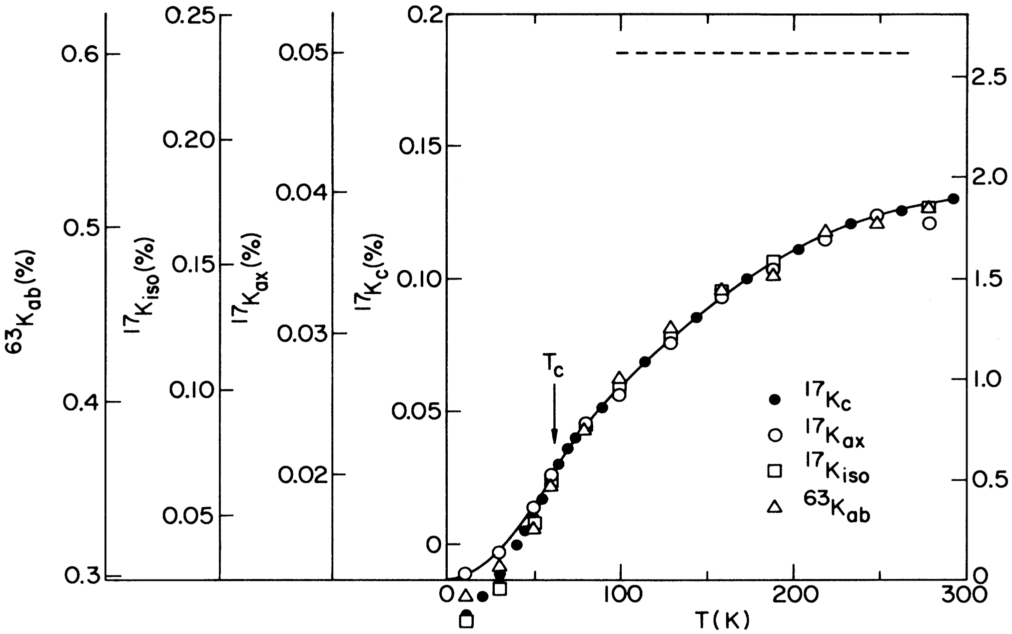

FIG. 1. The experimental Cu and O Knight shifts are plotted vs the temperature T. The values for the static planar susceptibility $\chi_0(T)/\mu_B^2$ are given on the right-hand scale. Note that the Knight shifts have exactly the same common temperature dependence.

$\mu_{\rm eff}=0.62\pm0.02\mu_B$ obtained by Manousakis[23] in his review of theoretical and experimental work on the antiferromagnetic insulator, $LaCu_2O_4$, we can determine the transferred hyperfine coupling B to the extent that we know the ratio of the transferred to the direct hyperfine coupling from the anisotropy of the Cu relaxation rate. If we take the ratio $A_\perp/4B=0.21$ determined for $YBa_2Cu_3O_7$,[9] we can determine the absolute value of $B=41$ kOe/μ_B. This value of the transferred hyperfine coupling B gives for the isotropic oxygen hyperfine coupling $C=69$ kOe/μ_B, which is in good agreement with the value proposed by Butaud *et al.*[24] and Imai.[25] In the Appendix, we present the corresponding results for slightly different choices of $A_\perp/4B$. We see that within experimental error the hyperfine couplings for the $YBa_2Cu_3O_{6.63}$ are identical to those for $YBa_2Cu_3O_7$ material, as was already suggested by Shimizu *et al.*[14] from plots of the Knight shift versus the total bulk susceptibility (see Table I).

We use the values derived from $A_\perp/4B=0.84$ to determine the temperature-dependent planar static long-wavelength susceptibility χ_0, with the results indicated by the scale on the right-hand side of Fig. 1. We find at $T=280$ K, $\chi_0(T)/\mu_B^2=1.87$ states/eV Cu(2), a value that is slightly reduced from the temperature-independent value $\chi_0/\mu_B^2=2.62$ states/eV Cu(2) of the $YBa_2Cu_3O_7$ material and is to be compared with the predicted band-structure value for $YBa_2Cu_3O_7$ of 1.8 states/eV Cu(2);

TABLE I. (a) Hyperfine couplings measured in kOe/μ_B, with $\mu_{\rm eff}[\mathrm{Cu(2)}]=0.62\mu_B$. (b) Values for the Cu(2) long-wavelength static susceptibility at 280 K.

(a)

$A_\perp/4B$	B	$A_\parallel$	$A_\perp$	C	D
0.2	40.3	−161	32.3	67.2	−3.0
0.21	40.8	−163	34.3	68.6	−3.0
0.225	41.6	−167	37.5	70.8	−3.0

(b)

$A_\perp/4B$	$YBa_2Cu_3O_7$ χ_0/μ_B^2	$YBa_2Cu_3O_{6.63}$ χ_0/μ_B^2
0.2	2.67 states/eV Cu(2)	1.91 states/eV Cu(2)
0.21	2.62 states/eV Cu(2)	1.87 states/eV Cu(2)
0.225	2.53 states/eV Cu(2)	1.81 states/eV Cu(2)

see, e.g., Ref. 26. Here we do *not* explain the temperature dependence of the static susceptibility. We will come back to this problem later on.

IV. SPIN-LATTICE RELAXATION

In a one-component model for the spin-spin correlation function the nuclear-spin-lattice relaxation rates depend on a product of the form factors, which differ from one nucleus to the other, and the dynamical structure factor $S(\mathbf{q},\omega)$. The latter is related to the imaginary part of the spin-spin correlation function by

$$S(\mathbf{q},\omega)=\frac{1}{1-e^{-\omega/k_B T}}\chi''(\mathbf{q},\omega) . \quad (4.1)$$

In the small frequency regime $\omega \ll T$ in which the NMR experiments are done Eq. (4.1) simplifies to

$$S(\mathbf{q},\omega)\approx \left[\frac{T}{\omega}\right]\chi''(\mathbf{q},\omega) . \quad (4.2)$$

The relaxation rates are determined by a $\mathbf{q}$ average over the Brillouin zone of the structure factor multiplied with the appropriate form factors. MMP have obtained the following explicit expressions for the relaxation rates:

$$^{63}W_{\perp}=\frac{3}{4}\frac{1}{\mu_B^2\hbar}\lim_{\omega\to 0}\sum_q \{A_{\perp}-2B[\cos(q_x a)+\cos(q_y a)]\}^2 S(\mathbf{q},\omega) , \quad (4.3a)$$

$$^{63}W_{\parallel}=\frac{3}{8}\frac{1}{\mu_B^2\hbar}\lim_{\omega\to 0}\sum_q (\{A_{\parallel}-2B[\cos(q_x a)+\cos(q_y a)]\}^2+\{A_{\perp}-2B[\cos(q_x a)+\cos(q_y a)]\}^2) S(\mathbf{q},\omega) , \quad (4.3b)$$

$$^{17}W=\frac{3}{4}\frac{1}{\mu_B^2\hbar}\lim_{\omega\to 0}\sum_q \{2C^2[1-\cos(q_x a)]\}S(\mathbf{q},\omega) , \quad (4.3c)$$

$$^{89}W=\frac{3}{4}\frac{1}{\mu_B^2\hbar}\lim_{\omega\to 0}\sum_q \{16D^2[1-\cos(q_x a)][1-\cos(q_y a)]\}S(\mathbf{q},\omega) . \quad (4.3d)$$

Here $\mathbf{q}$ is measured from the antiferromagnetic wave vector $\mathbf{Q}=(\pi/a,\pi/a)$. Whereas the Cu relaxation rate picks up all the antiferromagnetic correlations, the O relaxation rate is not strongly enhanced by the AF correlations since the hyperfine field of the Cu spins cancels at the oxygen site. It is useful to introduce the following moments of the structure factor $S(\mathbf{q},\omega)$; see, e.g., Ref. 9:

$$S_0=\left[\frac{a}{2\pi}\right]^2\int d^2q S(\mathbf{q},\omega) , \quad (4.4a)$$

$$S_1=\left[\frac{a}{2\pi}\right]^2\int d^2q\{1-\tfrac{1}{2}[\cos(q_x a)+\cos(q_y a)]\}S(\mathbf{q},\omega) , \quad (4.4b)$$

$$S_2=\left[\frac{a}{2\pi}\right]^2\int d^2q\tfrac{4}{5}\{1-\tfrac{1}{2}[\cos(q_x a)+\cos(q_y a)]\}^2 S(\mathbf{q},\omega) , \quad (4.4c)$$

$$S_3=\left[\frac{a}{2\pi}\right]^2\int d^2q[1-\cos(q_x a)][1-\cos(q_y a)]S(\mathbf{q},\omega) . \quad (4.4d)$$

For the MMP model the integrals can be evaluated quite easily, see, e.g., Ref. 8, and one finds

$$S_0=\frac{\pi\chi_0 k_B T}{\hbar\Gamma}\left\{1+\frac{\beta}{\pi^2}\left[\frac{\pi}{4}\left[\frac{\xi}{a}\right]^2-\left[\frac{1}{8\pi}+\frac{1}{4\pi^2}\right]\right]\right\} , \quad (4.5a)$$

$$S_1=\frac{\pi\chi_0 k_B T}{\hbar\Gamma}\left\{1+\frac{\beta}{\pi^2}\left[\frac{\pi}{8}\ln\left[\frac{\xi}{a}\right]+0.1703\right]\right\} , \quad (4.5b)$$

$$S_2=\frac{\pi\chi_0 k_B T}{\hbar\Gamma}\left[1+\frac{\beta}{\pi^2}0.2522\right] , \quad (4.5c)$$

$$S_3=\frac{\pi\chi_0 k_B T}{\hbar\Gamma}\left[1+\frac{\beta}{\pi^2}0.1986\right] , \quad (4.5d)$$

where β is defined as $\beta=(a/\xi_0)^4$. Since these moments depend on the detailed form of the spin-spin correlation function, the numerical coefficients might change a little bit if one does not have perfect commensurability.

One word of caution has to be said to avoid confusion. For a Cu^{2+}, $\frac{3}{2}$ nuclear spin the relaxation rates are $W(0,\frac{2}{3},2,4)$ where W is the fundamental rate. In a nuclear magnetic-resonance experiment in a strong field the energy levels of the nuclear spin are dominated by the Zeeman energy; therefore the rate is $(1|T_1)_{NMR}=\frac{2}{3}W$. In a nuclear quadrupolar resonance experiment the $\pm\frac{1}{2}$ and the $\pm\frac{3}{2}$ levels are degenerate, so that the rate is given by $(1|T_1)_{NQR}=2W$. Since the actual relaxation times T_1 depend on the experimental condition, namely, on the applied field, we prefer to work with the basic rate W.

We see on inspection of Eqs. (4.5) that the zeroth moment S_0 is dominated by the correlation length ξ/a for large correlation lengths. The first moment S_1 depends only logarithmically on the correlation length, while the moments S_2 and S_3 are independent of the correlation length. Now we can express the different spin-lattice relaxation times in terms of the explicit moments:

$$^{63}W_{\parallel}=\frac{3}{4\mu_B^2\hbar}[(A_\perp-4B)^2S_0+8B(A_\perp-4B)S_1+20B^2S_2] , \tag{4.6a}$$

$$^{63}W_{\perp}=\frac{3}{8\mu_B^2\hbar}\{[(A_\perp-4B)^2+(A_\parallel-4B)^2]S_0+8B(A_\perp+A_\parallel-8B)S_1+40B^2S_2\} , \tag{4.6b}$$

$$^{17}W=\frac{3}{2\mu_B^2\hbar}C^2S_1 , \tag{4.6c}$$

$$^{89}W=\frac{6}{\mu_B^2\hbar}D^2S_3 . \tag{4.6d}$$

The copper relaxation is dominated by the moment S_0 and is therefore proportional to ξ^2, whereas the oxygen relaxation rate depends on the antiferromagnetic correlations only through $\ln(\xi)$. In the MMP model the basic difference between the two relaxation rates arises from the temperature dependence of the antiferromagnetic correlation length $\xi(T)$.

We turn next to a quantitative determination of the role played by antiferromagnetic correlations in determining relaxation rates. Since, as we have seen, the hyperfine couplings, $A_\perp$ and B, change by less than 5% on going from $YBa_2Cu_3O_6$ to $YBa_2Cu_3O_7$, we may use the result[9] for the $YBa_2Cu_3O_7$ material, $A_\perp=0.84B$, in determining $^{63}W_\parallel$ and $^{63}W_\perp$. On combining Eqs. (4.3) and (4.5) we then obtain

$$^{63}W_\perp(T)=\frac{12\pi}{\mu_B^2\hbar^2}B^2k_BT\left[\frac{\chi_0(T)}{\Gamma(T)}\right]\left\{0.294+\frac{\beta}{\pi^2}\left[0.49\left[\frac{\xi(T)}{a}\right]^2-0.62\ln\left[\frac{\xi(T)}{a}\right]+0.0175\right]\right\} , \tag{4.7a}$$

$$^{63}W_\parallel(T)=\frac{12\pi}{\mu_B^2\hbar^2}B^2k_BT\left[\frac{\chi_0(T)}{\Gamma(T)}\right]\left\{0.772+\frac{\beta}{\pi^2}\left[1.83\left[\frac{\xi(T)}{a}\right]^2-1.10\ln\left[\frac{\xi(T)}{a}\right]-0.297\right]\right\} , \tag{4.7b}$$

$$^{17}W(T)=\frac{3\pi}{2\mu_B^2\hbar^2}C^2k_BT\left[\frac{\chi_0(T)}{\Gamma(T)}\right]\left\{1+\frac{\beta}{\pi^2}\left[0.39\ln\left[\frac{\xi(T)}{a}\right]+0.17\right]\right\} , \tag{4.7c}$$

$$^{89}W(T)=\frac{6\pi}{\mu_B^2\hbar^2}D^2k_BT\left[\frac{\chi_0(T)}{\Gamma(T)}\right]\left[1+\frac{\beta}{\pi^2}0.2\right] . \tag{4.7d}$$

In this form we can easily separate out the contribution made by antiferromagnetic paramagnons to the spin-lattice relaxation rate; the "quasiparticle" result is obtained if one takes $\beta=0$ in Eqs. (4.7), while the importance of antiferromagnetic paramagnons is measured by defining the quantity

$$^{63}(R_{AF})_\parallel=\frac{^{63}W_\parallel(\beta)}{^{63}W_\parallel(0)} , \tag{4.8}$$

with corresponding expressions for $(^{63}R_{AF})_\perp$, $^{17}R_{AF}$ and $^{89}R_{AF}$. On making use of the result, $\beta\approx\pi^2$,[9] we thereby obtain

$$(^{63}R_{AF})_\parallel(T)=0.914+1.44\left[\frac{\xi}{a}\right]^2-1.82\ln\left[\frac{\xi}{a}\right] , \tag{4.9a}$$

$$^{17}R_{AF}(T)=1.01+0.336\ln\left[\frac{\xi}{a}\right] , \tag{4.9b}$$

$$^{89}R_{AF}(T)=1.04 . \tag{4.9c}$$

The relative importance of antiferromagnetic correlations, as one goes from ^{63}W to ^{89}W, is seen clearly in Eqs. (4.9). As MMP have emphasized, for $\xi^2/a^2\gg1$ antiferromagnetic correlations play a dominant role in determining $^{63}W(T)$, with the leading term being proportional to ξ^2, and a logarithmic contribution of antiferromagnetic origin (which is of opposite sign from the Fermi-liquid contribution), playing a more significant role than the latter for $(\xi/a)\geq1.5$. For ^{17}W antiferromagnetic paramagnons continue to enhance the relaxation rate; for example with $(\xi/a)\sim4$, the antiferromagnetic enhancement ratio is $^{17}R_{AF}=1.5$, while for $(\xi/a)\sim2.5$ it is 1.31. For ^{89}W, on the other hand, antiferromagnetic paramagnons contribute only 4% of the total relaxation rate, independent of the temperature.

V. DETERMINATION OF THE TEMPERATURE-DEPENDENT AF CORRELATION LENGTH AND FIT TO THE EXPERIMENTAL DATA

We consider first the copper relaxation rate. According to MMP, if the energy Γ, which defines the spin dynamics of the noninteracting spin system (and is approximately ε_F for a normal Fermi liquid), is independent of temperature, the temperature dependence of $^{63}W(T)/T$ reflects the temperature dependence of *both* the static susceptibility χ_0, *and* the antiferromagnetic correlation length ξ, as may be seen in Eq. (4.7a). Moreover, in the limit $(\xi/a)\gg1$, in which the antiferromagnetic correlations are quite strong, to the extent that $\xi(T)$ displays the mean-field behavior, Eq. (2.5), the product, $^{63}T_1T\chi_0(T)$ should display a linear temperature dependence. To see whether this is the case, we combine the results of Sec.

III for $\chi_0(T)$ with the experimental results of Takigawa *et al.*[18] to determine this product, with the result shown in Fig. 2. We see that $^{63}T_1T\chi_0(T)$ is indeed linear in temperature above 100 K. That the product $^{63}T_1T\chi_0(T)$ displays such simple behavior, when no such simplicity is evident in ^{63}W or $(^{63}W/T)$, provides strong support for the application of the MMP phenomenological theory to materials in which χ_0 varies with temperature and for their assumption that Γ is nearly independent of T.

We next consider the oxygen relaxation rate. According to Eqs. (4.5b) and (4.5c) the product $^{17}T_1T\chi_0(T)$, being only logarithmically dependent on $\xi(T)$, should vary only weakly with temperature. We combine our result for $\chi_0(T)$ with the experimental results of Takigawa *et al.*[18] for ^{17}W to determine this product and find, as may be seen in Fig. 3, that this is likewise the case. However, the weak temperature dependence displayed there does not reflect the predicted logarithmic dependence on $\xi(T)$, which would reduce $^{17}T_1T\chi_0(T)$ at low temperatures in contrast to the observation. What is the physical origin of this behavior? One possibility is a departure of $\mathbf{Q}$ from $(\pi/a, \pi/a)$. Since, however, any departure from commensurability introduces a term $\sim(\xi/a)^2$ in ^{17}W, which with even very small incommensurability would easily dominate the logarithmic term, this would make $^{17}T_1T\chi_0(T)$ decrease still more rapidly with increasing temperature and so exacerbate the problem. A more promising explanation is a weakly temperature-dependent $\Gamma(T)$. To explore this alternative, we first assume the trial value, $\beta=\pi^2$, and determine $\xi(T)/a$ from the ratio, $^{63}W_{\parallel}(T)/^{17}W(T)$, which is independent of $\Gamma(T)$; we then use this result, shown in Fig. 4, to calculate $\Gamma(T)$ from the product $^{17}T_1T\chi_0(T)[1.17+0.39\ln(\xi/a)]$. Our result for $\Gamma(T)$ is shown in Fig. 5. We see that a 20% increase in $\Gamma(T)$ between 280 and 60 K suffices to explain the measured increase of $^{17}T_1T\chi_0(T)$.

To check whether this variation is real, we analyze the NMR experiments on ^{89}Y in $YBa_2Cu_3O_{6+x}$ by Alloul, Ohno, and Mendels.[13] We note first that our finding that ^{17}W is proportional to the static spin susceptibility (and hence Γ is nearly independent of temperature) is in contradiction with the conclusion reached by Alloul, Ohno, and Mendels.[13] They found, on taking the ^{89}Y chemical shift, $^{89}K^L$, equal to 300 ppm, that $1/T_1T$ at the ^{89}Y site is proportional to the square of the ^{89}Y Knight shift, $^{89}K^s(T)$, and hence to $\chi_0^2(T)$, whereas according to the MMP theory it should be nearly proportional to $\chi_0(T)$. (The hyperfine form factor at the Y sites is zero at $\mathbf{q}=\mathbf{Q}$, so that, as is the case for the O nuclei, the AF correlations should contribute only minimally to the ^{89}Y relaxation rate.)

This apparent contradiction is resolved if, in fact, the Y chemical shift, which is difficult to measure directly, is *not* 300 ppm but is close to 200 ppm, as a number of authors (Walstedt *et al.*,[16] Butaud *et al.*,[24] and Imai[25]) have recently proposed. With this latter value, one finds on plotting the measurements by Alloul, Ohno, and Men-

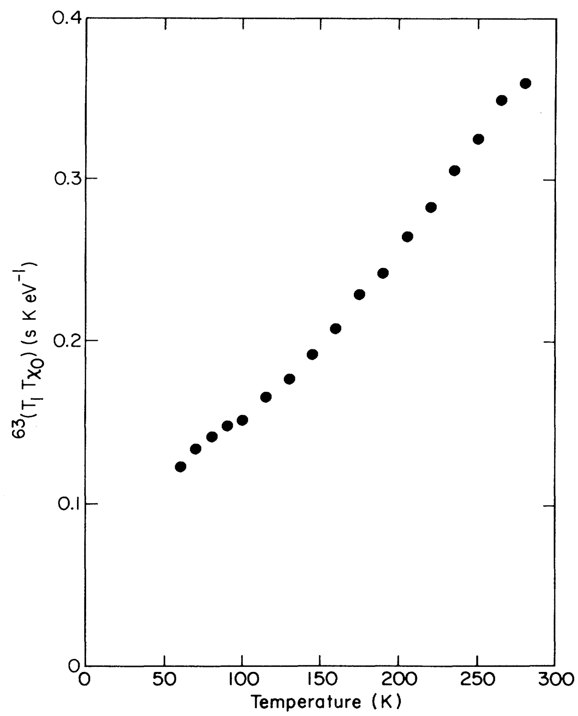

FIG. 2. The experimentally determined product $^{63}T_1T\chi_0(T)$ is plotted vs the temperature. The value of $\chi_0(T)$ is determined from the Knight-shift experiments [see Fig. 1, especially $^{17}K_c^s(T)$].

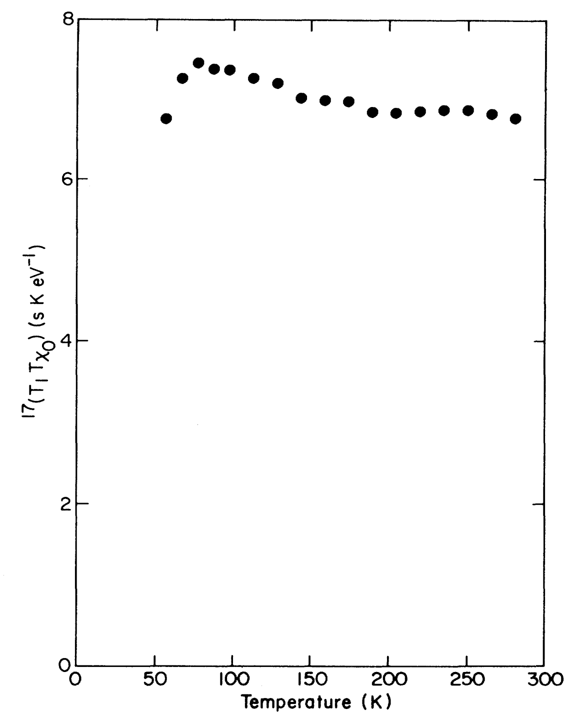

FIG. 3. The experimentally determined product $^{17}T_1T\chi_0(T)$ is plotted vs the temperature.

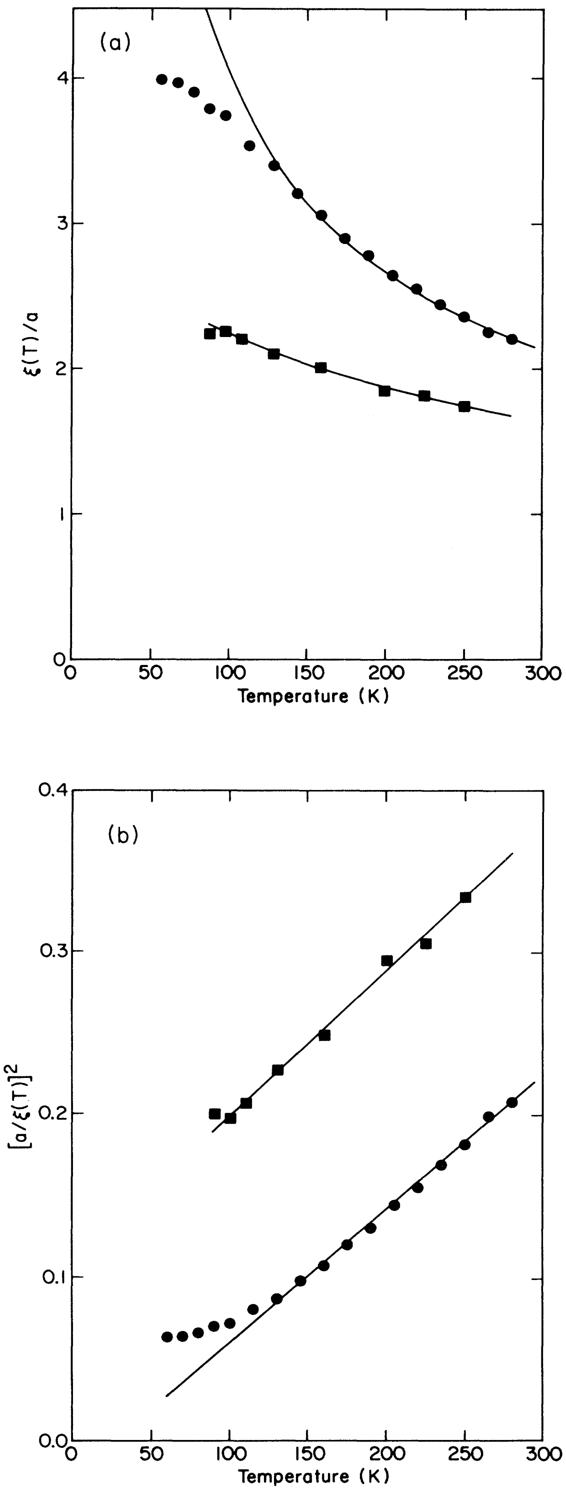

FIG. 4. (a) The correlation length $\xi(T)/a$ as determined from the ratio of the relaxation rates ${}^{63}W_{\perp}/{}^{17}W$ plotted vs the temperature for $YBa_2Cu_3O_{6.63}$ (circles) and $YBa_2Cu_3O_7$ (squares). (b) The inverse correlation length squared $[a/\xi(T)]^2$ plotted as a function of temperature for $YBa_2Cu_3O_7$ (squares) and $YBa_2Cu_3O_{6.63}$ (circles). The temperature dependence is close to a straight line, which is the behavior for a three-dimensional mean-field theory.

dels of ${}^{89}(T_1T)$, against their measured Knight shift, that within experimental accuracy the two are proportional. Since, moreover, the overall temperature dependence of the Y Knight shift measured by Alloul, Ohno, and Mendels for their samples ranging from $YBa_2Cu_3O_{6.85}$ to $YBa_2Cu_3O_{6.63}$ is quite similar to that found here for $\chi_0(T)$, it would seem plausible that the values of $\Gamma(T)$ would likewise be similar. If we make this assumption, and take $\Gamma_{\mathrm{Alloul}}(T)\cong\Gamma_{6.63}(T)$ (the value given in Fig. 5), we can use our expression, Eq. (4.7d), for ${}^{89}W(T)$, to deduce, from the Alloul data for a given hole concentration x, the hyperfine coupling D (which we expect to be independent of x), the Y chemical shift, and the planar susceptibility for the Alloul samples with $x\approx 0.63$, which we write as

$$[\chi_{\mathrm{Alloul}}(T)]_{6+x}=\delta\chi_0(T)\ , \tag{5.1}$$

where $\chi_0(T)$ is the spin susceptibility we have determined in Sec. III using the experimental data of Takigawa *et al.* In Fig. 6 we plot the total Knight shift ${}^{89}K(T)$ measured by Alloul, Ohno, and Mendels for their $YBa_2Cu_3O_{6.63}$ sample against $\chi_0(T)$. From the slope we find

$${}^{89}K^{L}=-198\ \mathrm{ppm}\ , \tag{5.2a}$$

$$D\delta=-0.011\ \mathrm{B}\ . \tag{5.2b}$$

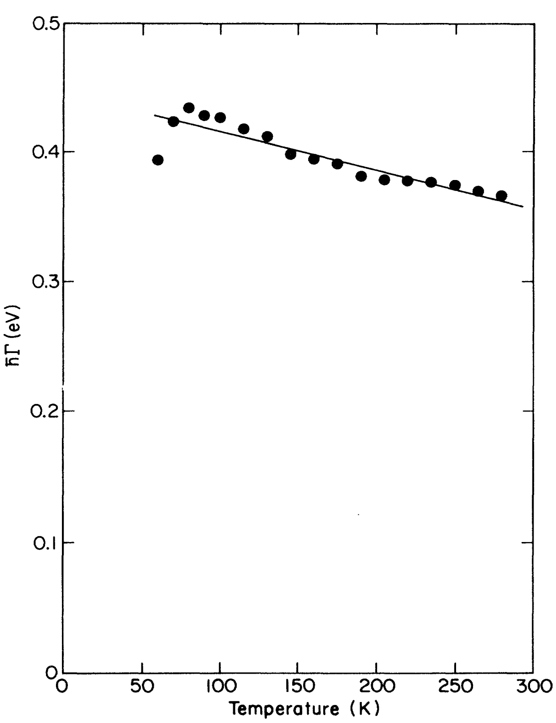

FIG. 5. The spin-fluctuation energy of the noninteracting system $\hbar\Gamma$ as determined from the oxygen relaxation rate, ${}^{17}(T_1T)\chi_0(T)$, corrected for the logarithmic dependence of the relaxation rate on the correlation length, $1.17+0.39\ln[\xi(T)/a]$, plotted vs the temperature in units of eV. The dashed line is a linear interpolation for $\Gamma(T)$, which was used to calculate the relaxation rates.

We next use our result, Eq. (4.7d) for $^{89}W(T)$, to obtain from the experimental results of Alloul, Ohno, and Mendels for $^{89}W_{6.63}(T)$, the product

$$D^2\delta = 1.4\times10^{-4}\ \mathrm{B}^2\ . \tag{5.3}$$

On combining (5.2b) and (5.3), we find $\delta \cong 0.86$, so that

$$D = -3.0\ \mathrm{kOe}/\mu_B\ , \tag{5.4a}$$

$$[\chi_{\mathrm{Alloul}}(T)]_{6.63} = 0.86\chi_0(T)\ . \tag{5.4b}$$

A measure of the accuracy of these results is obtained by comparing our calculated value of $\Gamma(T)$ determined from the product $^{63}T_1T\delta\chi_0(T)$ with the result for $\Gamma(T)$, Fig. 5, obtained from the ^{17}O experiments; as may be seen in Fig. 7(a), the agreement is quite good. Our deduced values of $\chi_0(T)$ for the Los Alamos $YBa_2Cu_3O_{6.63}$ sample are compared with those for the Orsay $YBa_2Cu_3O_{6.63}$ sample, as well as those for the nearby oxygen concentrations, in Fig. 7(b). We believe the difference between the "$YBa_2Cu_3O_{6.63}$" samples reflects a difference in oxygen content for the two samples. Indeed the similarity between the results we have obtained for $\chi_0(T)$ for the

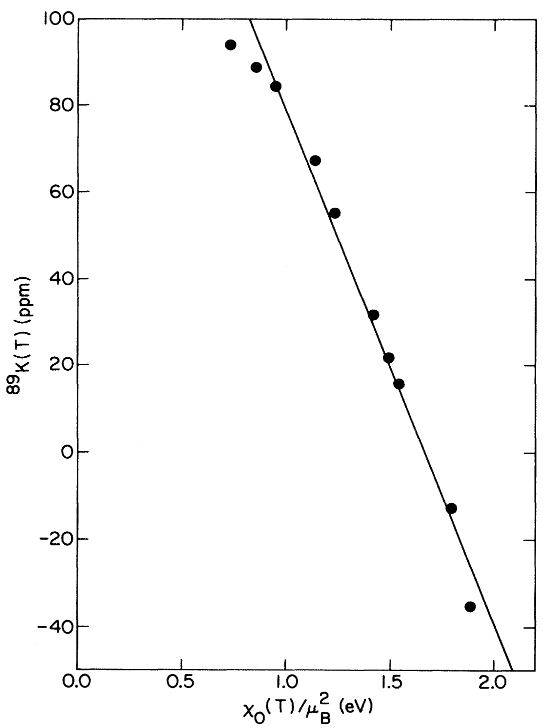

FIG. 6. The yttrium Knight shift, $^{89}K^s(T)$, as measured by Alloul, Ohno, and Mendels (Ref. 13) in the $YBa_2Cu_3O_{6.63}$ material plotted vs the temperature-dependent static planar susceptibility as measured by Takigawa *et al.* (Ref. 18) in the $YBa_2Cu_3O_{6.63}$ material. If $[\chi_0(T)]_{\mathrm{Alloul}} \cong \delta\chi_0(T)$, the temperature dependence of the Knight shift should be proportional $\chi_0(T)$. This is indeed the case. From the intercept we determine the yttrium chemical shift to be $\sim$200 ppm and the yttrium hyperfine coupling $D = -(2.6\ \mathrm{kOe}/\mu_B)/\delta$. The solid line is a guide to the eye.

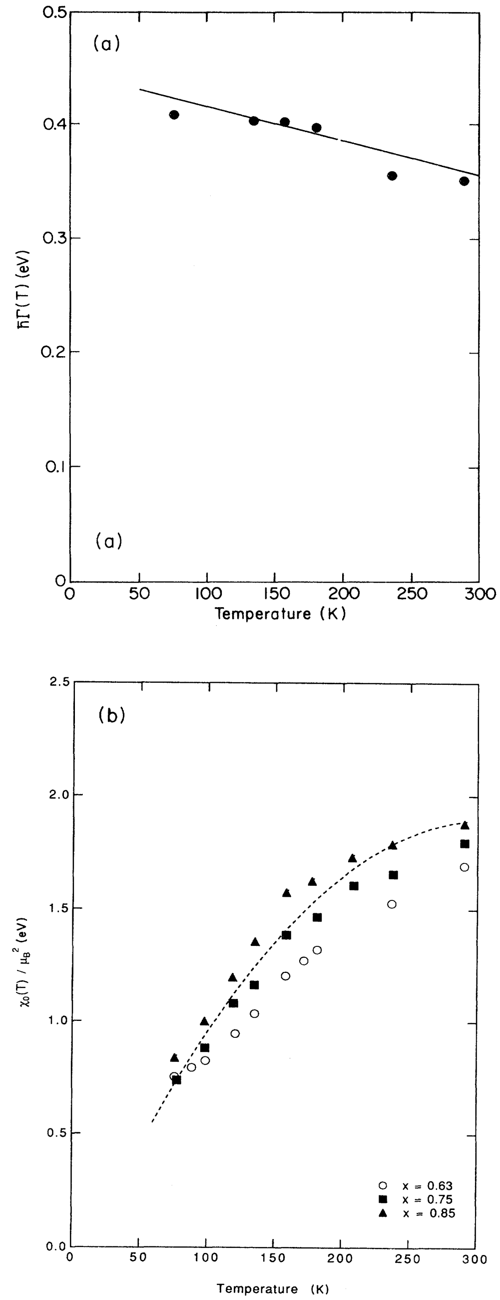

FIG. 7. (a) The energy $\hbar\Gamma$ for the $YBa_2Cu_3O_{6.63}$ sample of Alloul, Ohno, and Mendels as determined from the product $^{89}\{T_1T[\chi_0(T)]_{\mathrm{Alloul}}\}$ is plotted vs the temperature. We have used the measurements of the spin-lattice relaxation rate and the Knight shift of Alloul, Ohno, and Mendels (Ref. 13), as described in the text. The yttrium hyperfine coupling was taken to be $D = -3.0\ \mathrm{kOe}/\mu_B$. The dashed line is a linear interpolation for $\Gamma(T)$, which was used to calculate the relaxation rates (compare Fig. 5). (b) The values of the spin susceptibility, $[\chi_0(T)]_{\mathrm{Alloul}}$, we have deduced from the experiments of Alloul, Ohno, and Mendels (Ref. 13), using a chemical shift of 200 ppm and $D = -3.0\ \mathrm{kOe}/\mu_B$ are compared with $\chi_0(T)$. The oxygen concentration given represents the sample designation by Alloul, Ohno, and Mendels. The dashed line is the $\chi_0(T)$ determined from the experimental data of Takigawa *et al.* (Ref. 18).

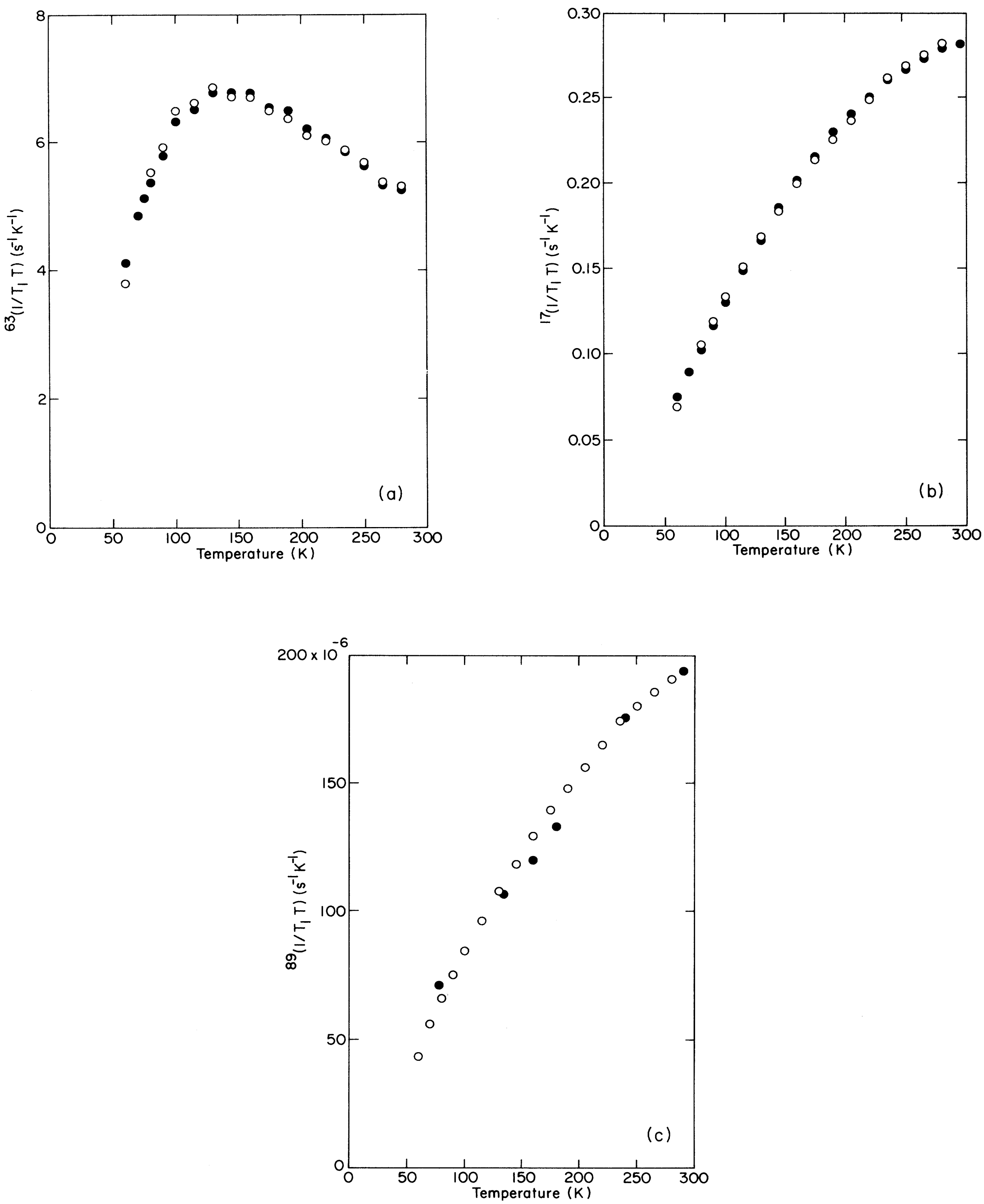

FIG. 8. (a) The theoretical result of the MMP theory for the copper spin-lattice relaxation rate (open circles) are compared to the measured $^{63}(T_1T)_{\parallel}$ (solid circles) vs the temperature. (b) The theoretical result of the MMP theory for the oxygen spin-lattice relaxation (open circles) rate are compared to the measured $^{17}(T_1T)_{\parallel}$ (solid circles) vs the temperature. (c) The theoretical result of the MMP theory for the yttrium spin-lattice relaxation rate (open circles) are compared to the measured $^{89}(T_1T)_{\parallel}$ (solid circles) vs the temperature.

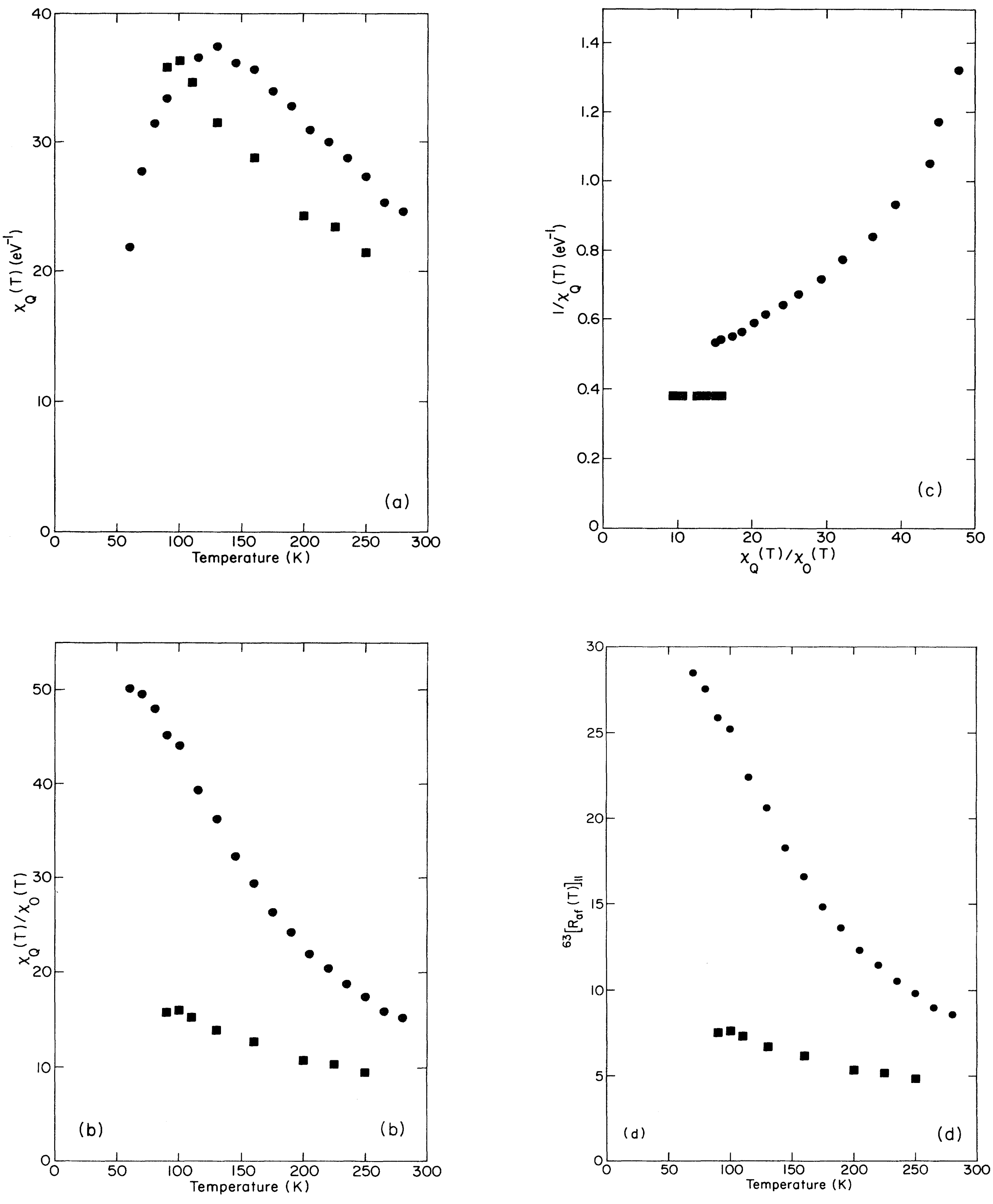

FIG. 9. (a) The calculated values of $\chi_Q(T)$ are plotted as a function of temperature for $YBa_2Cu_3O_{6.63}$ (circles) and $YBa_2Cu_3O_7$ (squares). (b) The calculated values of $\chi_Q(T)/\chi_0(T)$ are plotted as a function of temperature for $YBa_2Cu_3O_{6.63}$ (circles) and $YBa_2Cu_3O_7$ (squares). (c) The calculated values of $\chi_0(T)$ are plotted as a function of $\chi_Q(T)/\chi_0(T)$ for $YBa_2Cu_3O_{6.63}$ (circles) and $YBa_2Cu_3O_7$ (squares). (d) The calculated values of $^{63}[R_{AF}(T)]_{\parallel}$ are plotted as a function of temperature for $YBa_2Cu_3O_{6.63}$ (circles) and $YBa_2Cu_3O_7$ (squares).

$YBa_2Cu_3O_{6.85}$ sample of Alloul, Ohno, and Mendels, and the susceptibility obtained for the $YBa_2Cu_3O_{6.63}$ sample of Takigawa *et al.* suggests that the two samples are quite similar.

We next present our calculated results for a number of quantities of physical interest; our theoretical results are obtained using the values of $\chi_0(T)$, $\xi(T)/a$, and $\Gamma(T)$ depicted in Figs. 1, 4(a), and 5, and using $B=41$ kOe/μ_B, $C=69$ kOe/μ_B, $D=-3.0$ kOe/μ_B, and $\beta=\pi^2$. Our results for the relaxation rates, $^{63}(1/T_1T)$, $^{17}(1/T_1T)$, and $^{89}(1/T_1T)$, are compared with experiment in Figs. 8(a), 8(b), and 8(c). In Figs. 9(a), 9(b), 9(c), and 9(d) we present our calculated results for $\chi_Q(T)$, $\chi_Q(T)/\chi_0(T)$, and $^{63}(R_{AF})_{\|}(T)$, respectively, and compare these with the corresponding quantities calculated by MMP for $YBa_2Cu_3O_7$. The corresponding results for the spin fluctuation energy, $\hbar\omega_{SF}(T)$, are given in Fig. 10, while in Fig. 11, we show our predicted value of the anisotropy ratio $[^{63}W_{\perp}(T)/^{63}W_{\|}(T)]$.

The fit between theory and experiment for the relaxation rates is obviously quite good. Does it, however, constitute a real test of the antiferromagnetic Fermi-liquid theory of Millis, Monien, and Pines? We believe that it does, for the following reason. Once $\chi_0(T)$ and the hyperfine coupling parameters have been fixed by Knight-shift experiments on the $YBa_2Cu_3O_{6.63}$ and $YBa_2Cu_3O_7$ material, and by the choice of $B=41$ kOe/μ_B from the fit to the antiferromagnetic resonance results for the $YBa_2Cu_3O_6$ material, one has three parameters to determine: $\xi(T)/a$, $\Gamma(T)$, and $(\xi_0/a)\equiv\beta^{-1/4}$. At a specific temperature T, one has, in principle, three experimental results—$^{63}W(T)$, $^{17}W(T)$, and $^{89}W(T)$—available on the same compound to fix three parameters, although at present, as we have seen, $^{89}W(T)$ is known only for a closely related compound. Once, however, these three parameters are determined at that temperature, since ξ_0 is taken to be independent of temperature, at all remaining temperatures there are only two free parameters: $\Gamma(T)$ and $\xi(T)/a$; hence *at all other temperatures,* one is fitting three independent experimental results with two free parameters. Put another way, given arbitrarily *accurate* results for $^{89}W(T)$, one could use these to fix the temperature dependence of $\Gamma(T)$, since ^{89}W is independent of ξ/a. One could then fix β, the magnitude of Γ and ξ/a, uniquely at a specific temperature T_0, from a knowledge of ^{17}W, ^{89}W, and ^{63}W at that temperature, and then determine the behavior of ξ/a at other temperatures by using, say, $^{63}W_{\|}$. Using these results for $\xi(T)/a$, the agreement between theory and experiment for $^{17}W(T)$ (at a temperature other than T_0), then constitutes a test of the accuracy of the MMP antiferromagnetic-Fermi-liquid theory, a test that it would seem to meet with ease.

A related question is how accurately we know the four parameters, $\chi_0(T)$, $\Gamma(T)$, $\xi(T)$, and ξ_0 that enter the

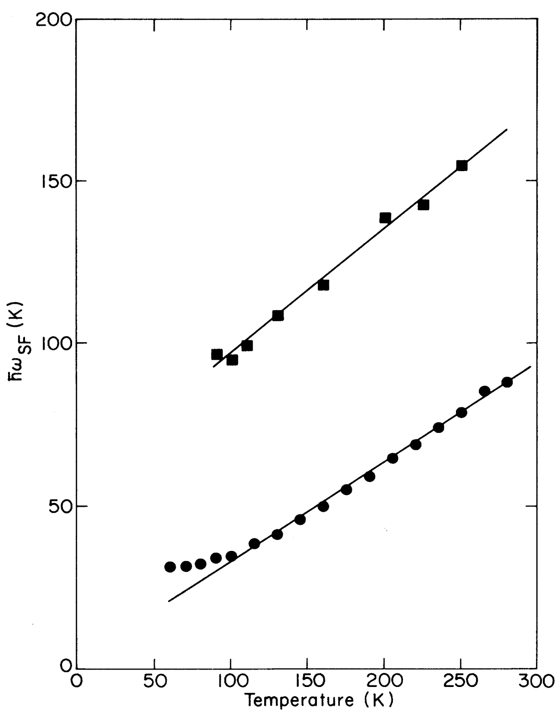

FIG. 10. The spin fluctuation temperature ω_{SF} plotted as function of temperature for the $YBa_2Cu_3O_{6.63}$ (circles) and the $YBa_2Cu_3O_7$ (squares) material. The lines are guides to the eye.

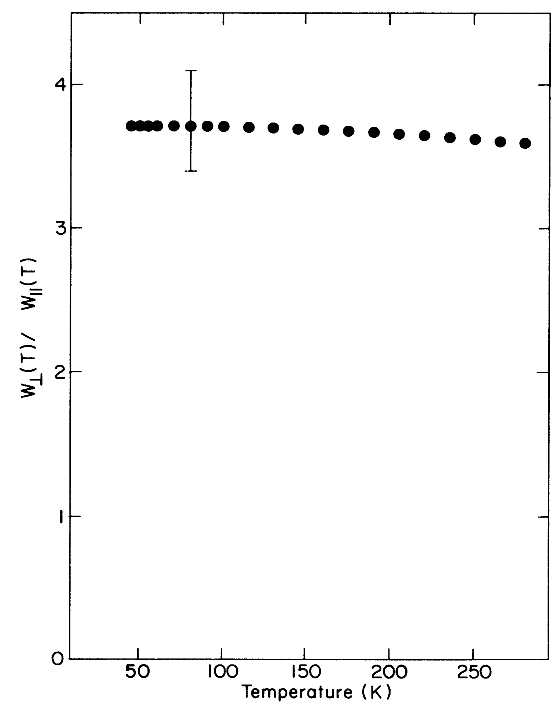

FIG. 11. The predicted value for the anisotropy of the copper spin-lattice relaxation rate $^{63}W_{\perp}(T)/^{63}W_{\|}(T)$ is shown as a function of temperature for $YBa_2Cu_3O_{6.63}$. A typical range of measured anisotropies is indicated by the error bar. The measured anisotropy does not change very much with temperature.

theory. Given present experimental accuracies of $\sim 10\%$ for the Knight shift and the relaxation rate experiments, we believe that the long-wavelength susceptibility $\chi_0(T)$, is known to that accuracy, as is the product $\beta(\xi/a)^2 \equiv (\xi/a)^2(a/\xi_0)^4$, since for long AF correlation lengths the latter quantity may be determined quite accurately from $^{63}W_{\parallel}(T)$. On the basis of our examination of choices of β that lie within 20% of our best fit value $\beta \sim \pi^2$, we conclude that our deduced values of β and ξ/a, and of Γ, which depends on our choice of β, are reliable to within 20%.

We comment briefly on the influence of oxygen content on the antiferromagnetic-Fermi-liquid parameters. The most dramatic changes come in $\xi(T)$, or $^{63}R_{\mathrm{AF}}(T)$, and $\chi_0(T)$. We see that in the reduced oxygen material the antiferromagnetic correlations at any given temperature are much stronger than in $YBa_2Cu_3O_7$ and that $\chi_0(T)$ is not only reduced somewhat at 280 K, but develops a dramatic temperature dependence, while $\Gamma(T)$ possesses a much weaker temperature dependence.

The alert reader will note the strong similarity between the plots of $^{63}[R_{\mathrm{AF}}(T)]_{\parallel}$ and $\chi_Q(T)/\chi_0(T)$. The similarity is no accident; inspection of Eq. (4.7a) shows that for all temperatures of interest $^{63}W_{\parallel}(T)$ is dominated by the leading term in $(\xi/a)^2$,

$$^{63}W_{\parallel}(\beta,T) \sim \frac{\bar{\chi}_0(T)}{\Gamma(T)}\beta\left[\frac{\xi(T)}{a}\right]^2 \sim \frac{\chi_Q(T)}{\Gamma(T)}\sqrt{\beta}\ , \quad (5.5a)$$

while

$$^{63}W_{\parallel}(0,T) \equiv (^{63}W_{\parallel})_{\mathrm{QP}} \sim \frac{\bar{\chi}_0(T)}{\Gamma(T)}\ , \quad (5.5b)$$

so that for a fixed value of β,

$$[R_{\mathrm{AF}}(T)]_{\parallel} \sim \chi_Q(T)/\chi_0(T)\ .$$

An interesting feature of our results for $\chi_Q(T)$, [Fig. 9(a)], is the presence of a maximum at ~ 100 K. The physical origin of this feature is the leveling off of $\xi(T)$ for $T < 120$ K, depicted in Fig. 4. Below 100 K, $\chi_0(T)$ falls off far more rapidly than $\xi(T)$ increases; as a result $\chi_Q(T)$, and hence, according to Eq. (5.1a), $^{63}W_{\parallel}(T)$ both display a maximum followed by a comparatively rapid decrease.

We call attention to the fact that the tendency toward antiferromagnetism in the $YBa_2Cu_2O_{6.63}$ material is so strong that above $T \sim 130$ K, say, $\xi(T)$ follows approximately the temperature dependence

$$\left[\frac{\xi(T)}{a}\right]^2 \cong \frac{1060\ \mathrm{K}}{T-30\ \mathrm{K}}\ . \quad (5.6)$$

In other words, as the temperature is lowered from 300 K, the system appears to be on its way toward becoming an antiferromagnet with a Néel temperature of 30 K. What stops it? Here we can only speculate, but a natural speculation is that as the temperature is lowered below ~ 130 K the system begins to be aware of the fact that it is going to become a superconductor at 60 K rather than an antiferromagnet at 30 K. We know that for $T \le T_c$, in the superconducting state, the antiferromagnetic correlation length is a constant, since $^{63}W(T)/^{17}W(T) \approx$ const. It follows that the behavior, Eq. (5.6), must stop somewhere. It would seem that for the $YBa_2Cu_3O_{6.63}$ material, the transition from Eq. (5.6) to ξ=const, which may be thought of as a precursor effect, begins at a temperature $\sim 2T_c$. A corollary of this remark is that there likely is an intrinsic connection between failed antiferromagnetic behavior and the appearance of superconductivity; we refer the interested reader to Pines[27] for a discussion of this possibility.

Finally, we note that as a consequence of the much stronger antiferromagnetic correlations present in $YBa_2Cu_3O_{6.63}$, $\hbar\omega_{\mathrm{SF}}$ is very much reduced compared to its value for the $YBa_2Cu_3O_7$ material. We discuss the physical consequences of this quite low energy scale in the following section.

VI. ANTIFERROMAGNETIC-FERMI-LIQUID THEORY

We have seen that *antiferromagnetic-Fermi-liquid theory* provides a *detailed* quantitative description of NMR experiments, not simply a useful qualitative description. It contains four basic parameters, each of which can be determined experimentally. Two relate to the short-wavelength behavior of the spin-spin correlation function: the temperature-dependent antiferromagnetic correlation length ξ, and the quasiparticle correlation length ξ_0, which marks the transition from antiferromagnetic paramagnon to quasiparticle behavior. Two relate to the long-wavelength ($q\xi_0 < 1$) quasiparticle behavior: the (usually) temperature-dependent static spin susceptibility χ_0 and the (weakly) temperature-dependent quasiparticle spin fluctuation energy Γ. One significant measure of the applicability of the theory is when one takes out the **q**-independent scaling factor, $\chi_0(T)$, the spin-lattice relaxation rates behave in the same way in the $YBa_2Cu_3O_{6.63}$ and $YBa_2Cu_3O_7$ material. This tells that in addition to the temperature-dependent coherence length ξ the spin-spin correlation function has an overall temperature dependence that is the same at each wave vector.

As one of us has proposed,[28] the properties of a nearly antiferromagnetic Fermi liquid are *genuinely novel* and may be expected to be quite different from those of a normal Fermi liquid. For the latter, Landau theory provides an exact description of long-wavelength, low-frequency properties in terms of a minimal number of phenomenological macroscopic parameters; the short-wavelength high-frequency properties must be calculated microscopically. On the other hand, for an antiferromagnetic Fermi liquid, MMP theory provides an exact description of the *short*-wavelength, low-frequency part of the spin-spin correlation function in terms of a minimal number of phenomenological macroscopic parameters; it is the longer-wavelength quasiparticle properties that must be calculated microscopically. Moreover, in general, the latter *cannot* be described in terms of the usual Landau Fermi liquid, for which *both* χ_0 and Γ would be temperature independent.

From this perspective, it is the antiferromagnetic

Fermi-liquid part, $\chi_{AF}(\mathbf{q},\omega)$, of the spin-spin response function, $\chi(\mathbf{q},\omega)$, Eq. (2.4), that we can determine quite accurately from experiment, while in the absence of an accurate microscopic theory we must content ourselves with a phenomenological expression for the quasiparticle part, $\chi_{QP}(\mathbf{q},\omega)$ [as given for instance in Eq. (2.3)], rather than being able to use normal Fermi-liquid theory. In fact, there are not *two* independent parts of $\chi(\mathbf{q},\omega)$; there is a single function $\chi(\mathbf{q},\omega)$, which for strong antiferromagnetic correlations and wave vectors $|\mathbf{Q}-\mathbf{q}|\xi_0<1$ will be given by $\chi_{AF}(\mathbf{q},\omega)$, while for $|\mathbf{Q}-\mathbf{q}|\xi_0>1$ it will go over to a function that is close to $\chi_{QP}(\mathbf{q},\omega)$. In our calculation we have permitted both functions to run over all wave vectors, and have minimized the double counting problem by introducing the ξ_0- (or β-) dependent parameter $\bar{\chi}_0$. Our final results may always be expressed in terms of χ_0, by making use of the relation Eq. (2.5), $\chi=\bar{\chi}_0(1+\sqrt{\beta}/2\pi^2)$.

It is instructive to try to link the short-wavelength and long-wavelength behavior of $\chi(\mathbf{q},\omega)$. To the extent that the long-wavelength portion is Fermi liquid like, albeit with a temperature-dependent $\chi_0(T)$, we can identity Γ with an inverse density of states, $\Gamma\approx[N(0)]^{-1}$. To see this we note that for a Fermi liquid, in the long-wavelength limit, $\Gamma\sim qv_F$; however, we are interested in a wave-vector-independent value of Γ that will yield the usual noninteracting quasiparticle relaxation rate. That value is $\Gamma\approx\varepsilon_F$, the Fermi energy, from which the above result follows. Continuing the normal Fermi-liquid analogy, we would write, for the long-wavelength static susceptibility,

$$\chi_0(T)=\frac{\mu_B^2 N(0)}{1+F_0^a(T)}\cong\frac{\mu_B^2/\Gamma}{1+F_0^a(T)} . \tag{6.1}$$

In this expression, we would argue that the temperature variation of $\chi_0(T)$ would primarily arise from the change of F_0^a, since $\Gamma(T)$ varies by some 20% in the temperature range (from 100 to 300 K), over which the experimental $\chi_0(T)$ varies by a factor of 3. A further indication that the density of states does not change appreciably with temperature is given by the interpretation of optical experiments.[29] Moreover, if the primary effect is a density of states that is temperature dependent, the spin-lattice relaxation rate would again be proportional to the Knight-shift squared, and not to the Knight shift as seen in experiment.

What is the physical origin of the temperature variation of $\chi_0(T)$ [or $F_0^a(T)$]? It does not appear to be caused by a pseudogap,[30] since a pseudogap would mainly diminish the density of states at the Fermi surface, in disagreement with the optical experiments. A second possibility is that it is a natural consequence of the much stronger short-wavelength antiferromagnetic correlations found in $YBa_2Cu_3O_{6.63}$, since a system of spins that are strongly correlated at short distances respond less easily to a uniform magnetic field [hence the positive sign of $F_0^a(T)$].[27,28] In this view, the stronger the antiferromagnetic correlations [as measured by $\chi_Q(T)/\chi_0(T)$], the more $\chi_0(T)$ will be reduced. To examine the extent to which this idea might be correct, we plot in Fig. 9(c), $\chi_0^{-1}(T)$ versus $\chi_Q(T)/\chi_0(T)$ and note that if there is a threshold, $\chi_Q/\chi_0\approx 12$, for the influence of the AF correlations, one can understand in this way both the temperature-dependent $\chi_0(T)$ found in the $YBa_2Cu_3O_{6.63}$ material and the temperature-independent χ_0 found in the $YBa_2Cu_3O_7$ material. On this interpretation of "temperature-independent" offset between $(1/\chi_0)$ for $T>280$ K for the $YBa_2Cu_3O_{6.63}$ material and $1/\chi_0$ (300 K) for the $YBa_2Cu_3O_7$ material would not be associated with antiferromagnetic correlations but would reflect the direct influence of doping. It is natural to ascribe the comparatively small temperature dependence measured for $\Gamma(T)$ to a similar physical origin: a feedback of the strong and temperature-dependent antiferromagnetic correlations on the "long-wavelength" quasiparticle spin fluctuation energy $\Gamma(T)$.

The physical origin of the failure of Landau theory for antiferromagnetic Fermi liquids is the role played by the antiferromagnetic paramagnons, whose characteristic energy scale, $\hbar\omega_{SF}$, we find to be less than $k_B T_c$ and, hence, less than $k_B T$.[28] In an antiferromagnetic Fermi liquid the strong scattering of quasiparticles by these paramagnons, reduces the energy region near the Fermi surface in which the lifetime of the quasiparticles τ goes like $\sim T^2$, as in a normal Fermi liquid, to a very small energy, while above this energy the lifetime behaves as $\tau\sim T$ in a very large energy range around the Fermi surface. Support for this point of view comes from the present analysis, in which we find that over a substantial temperature region, $\omega_{SF}\sim T$, which indicates that q-averaged imaginary part of the spin-spin correlation function is proportional to ω/T for small energies ω. This behavior is also observed in Raman-scattering experiments[31] that measure the imaginary part of the density-density correlations. The latter result led Varma *et al.*[20] to propose a phenomenological theory based on the assumption that the imaginary part of the self-energy is proportional to the energy for low energies and constant at energies larger than the temperature, so that the only energy scale present in this problem is the temperature T. In several theoretical model calculations[32,33] a similar behavior has been obtained on a more microscopic basis.

It is not *a priori* clear how antiferromagnetic-Fermi-liquid theory, which provides an exact description of the spin-spin correlation function, $\chi(\mathbf{q},\omega)$, for small derivations from the antiferromagnetic wave vector, $\mathbf{Q}=(\pi/a,\pi/a)$ and not too large energies, say $\hbar\omega<\hbar\omega_{SF}$ or $\hbar\omega<k_B T$, should be extended to larger energy transfers. There are experimental indications mainly from the Raman-scattering experiments[31] as well as theoretical indications (e.g., Refs. 32 and 33) that the imaginary part of the spin-spin correlation function $\mathrm{Im}[\chi(\mathbf{q},\omega)]$ saturates at energies somewhat larger than the spin-fluctuation energy ω_{SF} and does not decay at large ω like

$$\mathrm{Im}[\chi(\mathbf{q},\omega)]\sim(\omega_{SF}/\omega)^2$$

as predicted by the simple expression Eq. (2.3). Apart from ω_{SF} and T, the only other energy scale that might give a cutoff at very large energies is the bandwidth. Neutron-scattering experiments that measure the dynamical structure factor $S(\mathbf{q},\omega)$ at energies much larger than the NMR experiments do not necessarily see the same $\mathbf{q}$ dependence as the NMR experiments, since the spin fluctuation energy ω_{SF} is quite small. Even for $\omega \sim T$, the structure in $\mathbf{q}$ space of the MMP model spin-spin correlation function $\chi(\mathbf{q},\omega)$ will tend to be obscured for those materials for which $\omega_{SF} \ll T$, since the damping term $i(\omega/\omega_{SF})$ would be large. Still one should be able to obtain from neutron-scattering experiments performed in the $YBa_2Cu_3O_{6.63}$ or $YBa_2Cu_3O_7$ samples at sufficiently low energies (e.g., 3–6 meV) information on the departure (for energies large compared to $\hbar\omega_{SF}$) of the spin-spin correlation function from the AFL form, Eq. (2.3).

Another interesting experimental quantity, which is, in principle, measurable in a neutron-scattering experiment, is the static susceptibility, χ_Q, for wave vectors in the neighborhood of $\mathbf{Q}$. As we have seen, for $T \sim 100$ K, χ_Q reaches its maximum value of some 38 eV^{-1}, representing an antiferromagnetic enhancement of a factor of ~ 22 over the room-temperature long-wavelength susceptibility ~ 1.7 eV^{-1}. A measurement of χ_Q would then provide a direct measurement of (ξ/ξ_0), and hence of ξ_0, since ξ is determined by the ratio, ${}^{63}W_{\parallel}/{}^{17}W$. Moreover, should the angular resolution of the experiment be sufficiently good, one could, in principle, not only obtain an independent measure of ξ (from the half width of the peak at $\mathbf{Q}$) but, in principle, of ξ_0 [from the wave vector at which $\chi(\mathbf{Q}-\mathbf{q}) \cong \chi_0$]. The latter measurement is, however, likely ruled out by background problems.

VII. RELATED WORK

The importance of antiferromagnetic correlations for the Cu nuclear magnetic resonance experiments was pointed out rather early by Walstedt *et al.*[2] and Imai *et al.*[3] Hammel *et al.*[7] were the first to propose that the difference between the Cu and O relaxation rates could be understood in terms of an interplay between $\mathbf{q}$-dependent form factors and strong antiferromagnetic correlations.

Mila and Rice[19] analyzed the experiments of Takigawa *et al.*[5] and pointed out that in a *one-component* picture a transferred hyperfine interaction was necessary to understand the difference between the Cu Knight-shift and spin-lattice relaxation time anisotropies. Shastry[8] extended their analysis to include the oxygen sites. He pointed out that if the oxygen nuclear spin interacts with the electron spins only via a hyperfine coupling, the hyperfine fields of the antiferromagnetically coupled electron spins residing on the Cu sites nearly cancel, in accord with the picture of Hammel *et al.*[7]

Earlier attempts to explain the difference between the Cu and the O spin-lattice relaxation rates using a two-component model include the work of Monien, Pines, and Slichter[21] and Cox and Tree.[34] In the view of the Knight-shift experiments in the $YBa_2Cu_3O_{6.63}$ material, we now believe that these attempts fail because they lead to results that are inconsistent with the fact that the Knight shifts have the same temperature dependence.

A somewhat different version of the two-component model has recently been put forward by Anderson and Ren,[11] who wish to avoid strong antiferromagnetic correlations by invoking an orbital relaxation mechanism, not associated with a measurable Knight shift, to explain the differences between the ${}^{17}W$ and ${}^{63}W$ relaxation rates. While such an approach could, in principle, explain these differences for, say, the $YBa_2Cu_3O_7$ material, it cannot provide either a qualitative or a quantitative explanation for the basic similarities one finds between relaxation rates for the $YBa_2Cu_3O_7$ and $YBa_2Cu_3O_{6.63}$ materials, once one takes into account the measured temperature dependence of $\chi_0(T)$ for the latter material.

Closest in spirit to the antiferromagnetic theory of Millis, Monien, and Pines is the work by Bulut *et al.*[10] who used the random-phase approximation to calculate the spin-spin correlation function for a three- and a two-band Hamiltonian. In their theory, the antiferromagnetic correlations arise from the nesting in the tight-binding Fermi surface. The doping they introduce is a parameter describing how strong the antiferromagnetic correlations are. It is in some sense related to our parameter β, which measures the strength of the antiferromagnetic correlations in the MMP theory. (Their "doping" parameter does not necessarily reflect the hole concentration.) It turns out that many of the physical features they obtained in their model calculation can be related to MMP theory if one is only looking at the $YBa_2Cu_3O_7$ compound, for which both theories describe the quantitative behavior of the copper and the oxygen relaxation rate successfully. Unfortunately their model cannot be extended to describe the $YBa_2Cu_3O_{6.63}$ material, since this approach does not contain the interaction effects that lead to a temperature dependence of F_0^a. On the other hand, their model gives some insight into how the MMP theory could be extended for energies large compared to the spin-fluctuation temperature.

The phenomenological approach by Varma *et al.*[20] is mainly motivated by the Raman scattering experiments,[31] and is therefore an attempt to describe the density-density correlations. In their approach the correlation functions are assumed to be nearly $\mathbf{q}$ independent. In the region of the NMR experiments, ω very small, their theory proposes that the correlation function goes like ω/T as a function of frequency ω and temperature T, which is in agreement with our findings from the NMR experiments for the spin-spin correlation function. Virosztek and Ruvalds[32] substantiated the proposal of Varma *et al.* by a more microscopic self-consistent theory, which, however, neglects the $\mathbf{q}$ dependence of the correlation functions.

We have recently received unpublished work by Imai[25] in which he used a much simplified version of the MMP model to analyze NMR experiments in different CuO high-temperature superconductors. In agreement with our conclusions, he finds that antiferromagnetic correlation plays a crucial role in understanding the NMR experiments.

We should also like to mention a very recent preprint of Moriya *et al.*,[35] who arrive at qualitative conclusions quite similar to our own on the basis of an interesting microscopic model calculation. As is the case with the

RPA calculation of Bulut *et al.*, it is not clear how their theory will yield the temperature-dependent $\chi_0(T)$ required for all cuprate superconductors other than $YBa_2Cu_3O_7$. We note that if one makes use of their expression for the contribution to the resistivity, $R(T)$ from quasiparticle-paramagnon scattering, one finds that $R(T) \sim T$ is a quite general consequence of the linear spectrum $\omega_{SF} \sim T$ that we have deduced here from our analysis of the NMR experiments.

VIII. CONCLUSIONS

There are a number of important conclusions that we can draw from our analysis. The first is that the one-component hyperfine Hamiltonian, which describes the $YBa_2Cu_3O_7$ material quite well, does not change in going from the $YBa_2Cu_3O_7$ to the $YBa_2Cu_3O_{6.63}$ material. It seems as if the Wannier orbitals of the Cu ions are not markedly disturbed by the presence of the oxygen holes, and the ionic picture for the Cu orbitals applies all the way from the $YBa_2Cu_3O_7$ to the $YBa_2Cu_3O_6$. It is, moreover, possible to describe the nuclear-magnetic-resonance experiments in the $LaCu_2O_4$ materials with the same hyperfine Hamiltonian.[36]

Second, as we have emphasized, the success of antiferromagnetic-Fermi-liquid theory in providing a quantitative explanation of the quite different measured low-frequency magnetic properties of $YBa_2Cu_3O_{6.63}$ and $YBa_2Cu_3O_7$ leaves little doubt that it will provide the correct explanation for the magnetic properties of the entire range of superconducting Y-Ba-Cu-O samples. Two questions naturally arise. Will the theory work equally well for the magnetic properties of the other cuprate superconductors? Can one use it to explain as well the charge or density response of the cuprate oxides?

The answer to the first question is almost certainly "yes." Monien, Monthoux, and Pines[36] have used antiferromagnetic-Fermi-liquid theory to analyze the Knight-shift and relaxation rate experiments on $La_{1.85}Sr_{0.15}CuO_4$, and they find that the values they calculate for $\chi_0(T)$, $^{63}W_{\parallel}(T)$, and $\xi(T)$ fit naturally into the Y-Ba-Cu-O family. A similar conclusion has been reached by Hammel *et al.*[37] and Reyes from their analysis of corresponding experiments on $Y_xPr_{1-x}Ba_2Cu_3O_7$ and $La_2CuO_{4.032}$ samples.

The answer to the second question is "quite possibly." As we have noted, there is a good reason to expect that because of quasiparticle antiferromagnon scattering the transport properties, density-density response function, and quasiparticle energy near the Fermi surface for an antiferromagnetic Fermi liquid will differ from those of a normal Fermi liquid. It remains to be seen which of the nonmagnetic properties of the normal state of the cuprate oxide superconductors can be attributed to their being antiferromagnetic Fermi liquids and which have their physical origin elsewhere. From an antiferromagnetic-Fermi-liquid perspective, the results that Bulut and coworkers[10,33] have obtained using their RPA model for $YBa_2Cu_3O_7$ are encouraging, as are the results of naive calculations that yield a linear resistivity result close, but further work is clearly required.

Quite generally we expect that the phenomenological parameters that determine the low-frequency magnetic behavior, and which we have extracted from our analysis of NMR experiments, may be viewed both as a proper goal for, and useful constraint on, microscopic calculations. In this connection we cannot emphasize too strongly how useful it is to have experimental results available for all three nuclei, Cu, O, and Y, in the Y-Ba-Cu-O series, as well the importance of having these results available for the same sample, or having an independent check on whether the samples at two different laboratories are sufficiently close in oxygen content to make possible meaningful comparison of experimental data. The virtue of antiferromagnetic-Fermi-liquid theory is that it constitutes an accurate description of nearly antiferromagnetic behavior, regardless of the physical origin of that behavior. That is also its defect, in a sense, since in and of itself it does not enable one to decide whether the physical origin of the near antiferromagnetism is Fermi surface nesting (as would be expected for a weakly localized spin system) or a temperature-dependent interaction between quasiparticles (as would be the case for an almost localized set of spins). It is clear that a comparison of the results for the $YBa_2Cu_3O_7$ and $YBa_2Cu_3O_{6.63}$ materials, obtained with these rather different approaches, will help provide an answer to this question.

In conclusion, we note that even though both the charge and spin elementary excitations that characterize the normal state of the cuprate oxides may turn out to be those of an antiferromangetic Fermi liquid, this result, in itself, does not settle the question of the physical origin of their superconductivity. Antiferromagnetic paramagnons are certainly an attractive candidate for that superconductivity,[27,35] but further work will be required to demonstrate that these provide a unique explanation.

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