Phenomenological model of nuclear relaxation in the normal state of $YBa_2Cu_3O_7$

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A phenomenological model of a system of antiferromagnetically correlated spins is shown to give a good quantitative description of NMR, nuclear-quadrupole-resonance, and Knight-shift measurements on yttrium, planar copper, and planar oxygen sites in $YBa_2Cu_3O_7$. The antiferromagnetic correlation length is estimated to be ~ 2.5 lattice constants at $T = 100$ K. The temperature dependence of the correlation length ceases at $T_x \simeq 100$ K. The enhancement of the observed relaxation rates over what is expected for weakly interacting electrons is calculated and shown to be large. Extension of the calculation to other cuprate superconductors is discussed.

I. INTRODUCTION

NMR, nuclear-quadrupole resonance (NQR), and Knight-shift measurements on high-T_c CuO_2 superconducting materials have revealed several puzzling phenomena; the most striking of these is the observation, in $YBa_2Cu_3O_7$, of non-Korringa temperature dependence of the copper spin-lattice relaxation rate ^{63}W and Korringa temperature dependence of the oxygen and yttrium spin-lattice relaxation rates ^{17}W and ^{89}W. In this article we demonstrate that the various measurements in the nonsuperconducting state are consistent with a one-component system (i.e., one spin degree of freedom per CuO_2 unit) of disordered but antiferromagnetically correlated spins. The spin system could be interpreted as the spin excitations of an antiferromagnetically correlated Fermi liquid, or, more exotically, as the excitations of a hypothetical quantum disordered phase of a doped local-moment antiferromagnet.

The ideas behind this work are not new. Walstedt *et al.* proposed early on[1] that antiferromagnetic correlations were important for NMR in cuprate materials. The basic explanation of the difference between the T dependence of the Cu and O relaxation rates has been given by Hammel *et al.*[2] and developed by Shastry.[3] However, in this paper we give a quantitative and complete phenomenological description of the relevant measurements, using a reasonably realistic model and obtaining estimates of the antiferromagnetic correlation lengths and of other parameters.

A number of related theoretical papers have appeared. Mila and Rice[4] have stressed the importance of including a transferred hyperfine coupling in the model and have argued[5] that yttrium relaxation measurements support a one-component model. Monien, Pines, and Slichter,[6] Monien and Pines,[7] and Tree and Cox[8] have discussed some properties of two-component models. Independently of our work, Bulut *et al.*[9] have used the random-phase approximation (RPA) to calculate various relaxation rates in one-band and three-band Hubbard models. Their work is similar in spirit to ours and they conclude, as we do, that a one-component spin model with strong antiferromagnetic correlations suffices to explain the data. Varma *et al.*[10] have considered magnetic relaxation in the context of an attempt to make a unified phenomenology for high-T_c superconductors. The relation of all of this work to ours will be discussed further in Sec. V.

We believe our phenomenology is useful for several reasons. One is that we show quantitatively that the magnetic relaxation phenomena may be due to interaction of the nuclei with a one-component spin system. The question of whether low-T behavior of the cuprate superconductors may be described in terms of one quantum fluid or two (e.g., spins on Cu sites and holes on O sites) has been controversial. Another result of our analysis is an estimate of the antiferromagnetic correlation length. This is measurable via neutron scattering, and its observation or absence is an important test of the one-component model. Third, we have accurately determined how much the observed relaxation rates are enhanced over what would be expected were the electrons in cuprate superconductors well described by band theory. A fourth reason for making a quantitative phenomenological model is that analysis of the differences between the model and the data permits characterization of and, one hopes, insight into novel features of the spin dynamics in the cuprate superconductors.

Before proceeding with the details of our analysis, we outline the motivation for our work. In a conventional Fermi liquid, the spin-lattice relaxation rate, W, has the Korringa temperature dependence $W \sim T$. This temperature dependence may be understood as follows: for a Fermi liquid the density of states for spin excitations is constant at low energies. The rate is given by the number of thermally excited spin excitations, which in this case scales as T. In the high-T_c cuprate superconductors, the Cu relaxation rate ^{63}W varies more slowly than the Korringa rate for $T > T^*$. The crossover temperature T^*

varies from material to material, but is of order 100–150 K. Therefore, for $T > T^*$, the density of states for spin excitations must increase as T decreases. This increase in the density of states may occur roughly uniformly throughout the Brillouin zone, or it may be concentrated near a particular q value, as would be the case for a system near a magnetic instability. There are two arguments in favor of the second point of view. One has to do with explaining the magnitude and T dependence of the anisotropy of the Cu nuclear relaxation rate. It will be discussed in more detail. The other has to do with the T dependence of the oxygen relaxation rate ^{17}W. This argument will be given here; it is a restatement of ideas present or implicit in the work of Hammel *et al.*,[2] Shastry,[3] and Mila and Rice.[5]

In the $T_c = 90$ K $YBa_2Cu_3O_7$ material the oxygen relaxation rate ^{17}W is observed to be accurately Korringa (i.e., $^{17}W \simeq T$) for all experimentally accessible $T > T_c$.[2] Now the Cu and O sites are less than 2 Å apart, and the hybridization between them is strong ($t_{\text{Cu-O}} \gtrsim 1.3$ eV). It is thus hard to imagine that at temperatures lower than 300 K two distinct kinds of elementary excitation could exist, one coupling only to the Cu nuclei and one only to the O nuclei. It is sometimes argued that the holes added when the insulating "parent" CuO_2 compounds are doped go into oxygen π orbitals. It is further argued that the O π orbitals are sufficiently weakly coupled to the Cu $d_{x^2-y^2}$ states that the O holes are uncorrelated with the Cu spins at the temperatures of experimental interest, so that a "two-fluid" description of normal-state spin dynamics is appropriate. There is substantial experimental and theoretical evidence against this hypothesis.[11,12] However, even if it were correct, the hybridization between the Cu d and O $2s$ and $3s$ orbitals would still lead to a large transferred hyperfine coupling between the Cu spin and the O nucleus, so that the Cu spins would relax the O nuclei. Therefore, we conclude that some symmetry must cause the matrix element coupling the enhanced part of the Cu spin density of states to the O nucleus to vanish. Such a symmetry cannot be relevant if the Cu spin density of states is enhanced uniformly over the zone. Therefore, we conclude that the Cu spin density of states must be enhanced most strongly at some particular point of the Brillouin zone. If one makes the plausible assumption that a given O nucleus is coupled predominantly to the spins on its two nearest-neighbor copper sites, the spin density of states must be enhanced most strongly near the zone corner, $(\pi/a, \pi/a)$, i.e., the enhanced spin density of states must be associated with the development of nearly commensurate antiferromagnetic correlations. In an antiferromagnetically correlated spin fluctuation, each O would be between two essentially oppositely directed Cu moments, so the transferred hyperfine field from these moments would cancel at the O site.

The preceding argument suggests that the non-Korringa temperature dependence of the Cu spin-lattice relaxation rate in $YBa_2Cu_3O_7$ is due to the buildup of commensurate antiferromagnetic spin correlations in electronic magnetic moments centered on Cu sites. In the remainder of this paper we show that a phenomenological model involving only these moments can explain all of the presently available NMR data on the $YBa_2Cu_3O_7$, $T_c = 90$ K material. Less extensive data are available for the other high-T_c materials; we comment on the implications of our model for these materials later.

In Sec. II, we describe our model for the spin fluctuations and present explicit expressions for the Knight-shift and relaxation rates of the planar copper [^{63}CU(2)], planar oxygen [^{17}O(2)], and yttrium (^{89}Y) nuclei in terms of a correlation length, an energy scale, the long-wavelength planar spin susceptibility, and various hyperfine coupling constants. We show in Sec. III how the experimental results of Walstedt *et al.*[1], Pennington *et al.*,[13] Hammel *et al.*,[2] Imai *et al.*,[4] Alloul *et al.*,[15] Barrett *et al.*,[16] Oldfield *et al.*,[17] and Horvatić *et al.*[18] can be used to determine the basic quantities that enter our expressions, we demonstrate that our model provides a fit to existing experimental results for $YBa_2Cu_3O_7$, and we outline the uncertainties involved. In Sec. IV we discuss our results and consider the application of our approach to other cuprate oxides. In Sec. V we consider related work. Section VI is the Conclusion.

II. THE MODEL

We assume that the low-frequency spin dynamics in the CuO_2 planes are described by a one-component model. In other words, we assume that in the CuO_2 planes there is at most one $S = \frac{1}{2}$ electronic spin degree of freedom per CuO_2 unit cell. In a band-theory picture the spin degree of freedom would be due to an electron residing in a Wannier orbital obtained by Fourier transforming k states in the conduction band. The spin would have some Cu and some O character. A band-theory picture is not necessary. Shastry[3] has shown that in a doped charge-transfer insulator with spins on Cu sites and holes on O sites, a one-band model describes the low-frequency spin dynamics. This spin degree of freedom is assumed to be responsible for spin-lattice relaxation and Knight shifts for the planar copper (^{63}Cu) and oxygen (^{17}O) nuclei and for the yttrium (^{89}Y) nuclei. We assume that the spins are centered on the Cu sites and are antiferromagnetically correlated, with correlations that increase with decreasing temperature. The spin dynamics are described by $\chi''(q,\omega)$, the imaginary part of the spin susceptibility. We assume that for $\omega \to 0$

$$\chi''(q,\omega \to 0) = \frac{\pi \chi_0 \omega}{\Gamma} \left[1 + \beta \frac{(\xi/a)^4}{(1+\xi^2 q^2)^2} \right] . \tag{2.1}$$

Here χ_0 is the uniform susceptibility. If χ_0 is also a reasonable estimate for the static susceptibility at a typical q not near $(\pi/a, \pi/a)$, then one may interpret spin fluctuation frequency. We shall make this interpretation in what follows. ξ is the spin correlation length and a is the lattice constant. β is a parameter, which will be discussed later. Momentum q is measured from the zone corner $(\pi/a, \pi/a)$. Our form for χ'' is plotted in Fig. 1.

The two key assumptions underlying Eq. (2.1) are that $\chi''(q,\omega \to 0) \sim \omega$ at all q, and that the antiferromagnetic correlations have relaxational dynamics. Both assumptions would hold for a Fermi liquid with antiferromagnet-

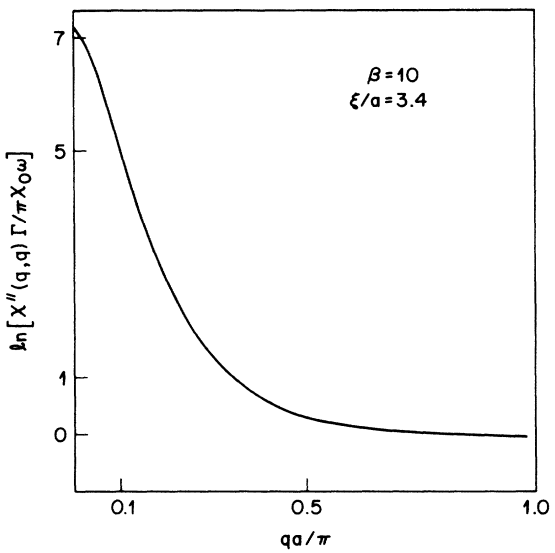

FIG. 1. Plot of assumed form of $\chi''(\mathbf{q},\omega)$ [Eq. (2.1)] along the zone diagonal, $\mathbf{q}=\alpha(\pi/a,\pi/a)$. We have chosen the origin of coordinates to be at the zone boundary so that $\alpha=1$ corresponds to zone center.

ic correlations. It is not clear whether they would hold for localized spin models. In many localized spin models a gap opens up at low T in the spin spectrum at all but a discrete set of points in q space, so that at typical q vectors,

$$\lim_{\omega\to 0}\frac{\chi''(q,\omega)}{\omega}\sim e^{-\Delta_q/T},$$

where Δ_q is a wave-vector-dependent spin gap typically of order the magnetic exchange energy J. As we show in Sec. III, the possibility of such a gap in the $YBa_2Cu_3O_7$ material is ruled out by the oxygen and yttrium relaxation data. In writing Eq. (2.1) we have also assumed the conventional (van Hove) value 2 for the dynamic exponent z, and the mean-field value 2 for the susceptibility exponent $2-\eta$. These assumptions are plausible in view of the relatively short coherence lengths we find but are not essential.

One motivation for our phenomenological form for χ'' may be obtained by considering a mean-field expression for the χ of an interacting spin system in terms of the susceptibility $\bar{\chi}$ of a noninteracting spin system. One obtains

$$\chi(q,\omega)=\frac{\bar{\chi}(q,\omega)}{1-J_q\bar{\chi}(q,\omega)}\ . \tag{2.2}$$

If we assume that

$$\lim_{\omega\to 0}\bar{\chi}''(q,\omega)=\pi\frac{\omega}{\Gamma_q}\bar{\chi}'(q,\omega=0)\equiv\pi\frac{\omega}{\Gamma_q}\bar{\chi}_q\ , \tag{2.3}$$

where Γ_q is a characteristic energy of spin fluctuations at wave vector q, then

$$\lim_{\omega\to 0}\chi''(q,\omega)=\pi\frac{\omega}{\Gamma_q}\bar{\chi}_q\frac{1}{(1-F_q)^2+[(\pi\omega/\Gamma_q)F_q]^2}\ , \tag{2.4}$$

where $J_q\bar{\chi}_q=F_q$. We expand F_q about its value F_Q at the zone corner

$$F_q=F_Q-q^2\xi_0^2\ , \tag{2.5}$$

define ξ via

$$1-F_Q=(\xi_0/\xi)^2\ , \tag{2.6}$$

and set $F_q=1$ in the second term of the denominator of Eq. (2.4). We then obtain

$$\chi''(q,\omega)=\frac{\pi\omega\bar{\chi}_q}{\Gamma_q}\frac{(\xi/\xi_0)^4}{(1+q^2\xi^2)^2+(\pi^2\omega^2/\Gamma_q^2)(\xi^4/\xi_0^4)}\ . \tag{2.7}$$

For large ξ we expect the q dependence of $\bar{\chi}_q$ and Γ_q to be a small correction to the q dependence in the denominator of Eq. (2.7); we therefore ignore this q dependence, setting $\bar{\chi}_q=\bar{\chi}$ and $\Gamma_q=\bar{\Gamma}$.

The $\omega\to 0$ limit of this expression yields one term of Eq. (2.1). Equation (2.7) is, however, only valid for small q. For larger q, say $qa>1$, one expects χ'' to be comparable to the uncorrelated value $\pi\bar{\chi}\omega/\bar{\Gamma}$, while Eq. (2.7) would imply a much smaller value. Therefore, we add a q independent term to Eq. (2.7), obtaining Eq. (2.1); we identify

$$\beta=(a/\xi_0)^4(\bar{\chi}/\chi)(\Gamma/\bar{\Gamma})\ . \tag{2.7a}$$

From Eq. (2.7) we see that the characteristic energy for a zone corner spin fluctuation in this mean-field theory is

$$\hbar\omega_{\rm SF}=\hbar\Gamma\xi_0^2/\xi^2\ . \tag{2.8}$$

This is much smaller than Γ if ξ^2 is large, i.e., if the system is near an antiferromagnetic instability. Because $\omega_{\rm SF}\sim\xi^{-2}$, the dynamic exponent $z=2$. Similarly, from (2.2) and (2.6) we see $\chi(0)\sim\xi^{-2}$; the susceptibility exponent $2-\eta$ therefore takes the mean-field value of two.

Equation (2.8) is to be regarded as a rough estimate. In particular, for sufficiently large ξ, mean-field theory is not applicable. The correct theory would then presumably have, e.g., a dynamic exponent $z=1$ appropriate to $d=2$ antiferromagnets.

We must also postulate a temperature dependence of ξ. We choose the dependence

$$\left[\frac{\xi(T)}{a}\right]^2=\left[\frac{\xi(T=0)}{a}\right]^2\frac{T_x}{T+T_x}\quad(T>T_c)\ . \tag{2.9}$$

We consider only temperatures T larger than the superconducting transition temperature T_c. Equation (2.9) is the low-T temperature dependence of ξ expected from a mean-field-theory treatment of a model in which the coupling is not quite strong enough to produce a magnetic phase transition. In a mean-field treatment of a model with coupling strong enough to produce a magnetic phase transition at a temperature $T_N>0$, one would have $\xi^{-2}\sim(T-T_N)$.

T_x is to be identified with the temperature scale at which the Cu relaxation rate crosses over from its high-temperature to its low-temperature behavior. Indeed at $T\gg T_x$, $\xi^2\sim 1/T$. The increase of ξ as T decreases leads to an increase in the density of low-frequency spin excita-

tions and hence a relaxation rate that falls more slowly than the Korringa rate. However, for $T<T_x$, $\xi^2\sim$constant, leading to an approximately Korringa behavior.

Having specified the spin dynamics, we now turn to the coupling with the nuclei. Our Hamiltonian is

$$H^{e\text{-}n}={}^{63}\mathbf{I}_n\mathbf{A}\mathbf{S}_n+B\sum_{\delta}{}^{63}\mathbf{I}_n\cdot\mathbf{S}_{n+\delta}+C\sum_{\delta'}{}^{17}\mathbf{I}_n\cdot\mathbf{S}_{n+\delta'}+D\sum_{\delta''}{}^{89}\mathbf{I}_n\cdot\mathbf{S}_{n+\delta''}\ . \quad (2.10)$$

Here δ labels the four unit cells nearest to Cu site n, δ' labels the two unit cells sharing O site n, and δ'' labels the eight unit cells (four in one plane, four in another) nearest Y site n. **A**, B, C, and D are hyperfine coupling constants. They have the dimension of energy. **A** is the direct hyperfine tensor. It has two independent components: $A_\parallel$ pertains to spins oriented along the c axis of the $YBa_2Cu_3O_7$ crystal, and $A_\perp$ pertains to spins oriented perpendicular to this axis. B is the transferred hyperfine coupling first proposed for this problem by Mila and Rice.[4] It is assumed to be isotropic, as are the other transferred couplings C and D. The geometry and the various couplings are illustrated in Fig. 2.

In terms of these parameters the components of the spin Knight shift are

$${}^{63}\mathbf{K}_\parallel=\frac{(A_\parallel+4B)\chi_0}{\gamma_e{}^{63}\gamma_n\hbar^2}\ , \quad (2.11a)$$

$${}^{63}\mathbf{K}_\perp=\frac{(A_\perp+4B)\chi_0}{\gamma_e{}^{63}\gamma_n\hbar^2}\ , \quad (2.11b)$$

$${}^{17}\mathbf{K}_{iso}=\frac{2C\chi_0}{\gamma_e{}^{17}\gamma_n\hbar^2}\ , \quad (2.11c)$$

$${}^{89}\mathbf{K}_{iso}=\frac{8D\chi_0}{\gamma_e{}^{89}\gamma_n\hbar^2}\ . \quad (2.11d)$$

The various nuclear moments are denoted by γ_n, and the electron moment $\gamma_e=2\mu_B/\hbar$, where μ_B is the Bohr magneton.

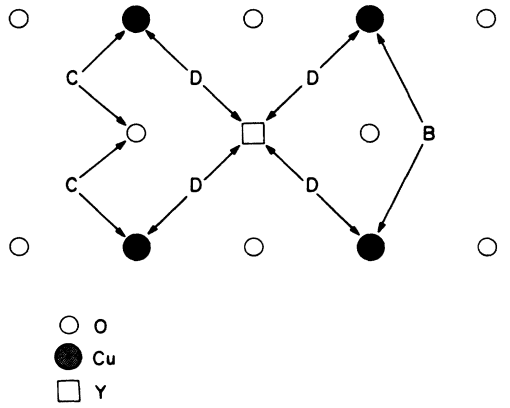

FIG. 2. Sketch of atoms near one CuO_2 plane with hyperfine couplings [Eq. (2.10)] indicated. Note that the Y atom couples to two Cu-O planes, although only the coupling to one is shown.

The ^{63}Cu(2) spin-lattice relaxation rate W, for a field applied parallel to the c axis, may be written in the form

$${}^{63}W_\parallel=\frac{3}{4}\frac{1}{\mu_B^2\hbar^2}\lim_{\omega\to0}\times\sum_q[A_\perp-2B(\cos q_xa+\cos q_ya)]^2S(q,\omega)\ , \quad (2.12)$$

where $S(q,\omega)$, the spin dynamic structure factor, is in general related to the imaginary part of the spin-spin correlation function, $\chi''(q,\omega)$, by

$$S(q,\omega)=\frac{e^{\beta\omega}}{e^{\beta\omega}-1}\chi''(q,\omega)\ . \quad (2.13a)$$

For the frequencies of interest for NMR experiments one has

$$S(q,\omega)\cong(kT/\hbar\omega)\chi''(q,\omega)\ . \quad (2.13b)$$

The other relaxation rates of interest are given by

$${}^{63}W_\perp=\frac{3}{8}\frac{1}{\mu_B^2\hbar}\sum_q[A_\parallel-2B(\cos q_xa+\cos q_ya)]^2+[A_\perp-2B(\cos q_xa+\cos qa)^2]^2S(q,\omega) \quad (2.14a)$$

for a magnetic field applied in the a-b plane,

$${}^{17}W(2)=\frac{3}{4}\frac{1}{\mu_B^2\hbar}\sum_q[2C^2(1-\cos q_xa)]S(q,\omega) \quad (2.14b)$$

for an ^{17}O(2) nucleus, and

$${}^{89}W=\frac{3}{4}\frac{1}{\mu_B^2\hbar}\sum_q\left[16D^2\left[\cos^2\frac{q_za}{2}\right]\times(1-\cos qa)(1-\cos q_ya)\right]S(q,\omega) \quad (2.14c)$$

for an ^{89}Y nucleus.

As an intermediate step in the calculation of the W it is convenient to define four moments of $S(q,\omega)$:

$$S_0=\left[\frac{a}{2\pi}\right]^2\int d^2qS(q,\omega)\ , \quad (2.15a)$$

$$S_1=\left[\frac{a}{2\pi}\right]^2\int d^2q[1-\tfrac{1}{2}(\cos q_xa+\cos q_ya)]S(q,\omega)\ , \quad (2.15b)$$

$$S_2=\left[\frac{a}{2\pi}\right]^2\tfrac{4}{5}\int d^2q[1-\tfrac{1}{2}(\cos q_xa+\cos q_ya)]^2S(q,\omega)\ , \quad (2.15c)$$

$$S_3=\left[\frac{a}{2\pi}\right]^2\int d^2q(1-\cos q_xa)(1-\cos q_ya)S(q,\omega)\ . \quad (2.15d)$$

Substituting our ansatz for χ'' into Eqs. (2.15), evaluating the integrals, and dropping terms of relative order (a/ξ) and higher yields

$$S_0=\frac{\pi\chi_0 k_B T}{\hbar\Gamma}[0.080\beta(\xi/a)^2-0.007\beta+1] ,$$

$$S_1=\frac{\pi\chi_0 k_B T}{\hbar\Gamma}[0.040\beta\ln(\xi/a)+0.017\beta+1] ,$$

$$S_2=\frac{\pi\chi_0 k_B T}{\hbar\Gamma}(0.026\beta+1) ,$$

$$S_3=\frac{\pi\chi_0 k_B T}{\hbar\Gamma}(0.020\beta+1) .$$

III. DETERMINATION OF PARAMETERS

We begin with the Knight shifts, which are by now reasonably well established experimentally. Barrett *et al.*[16] find the ^{63}Cu(2) Knight shift for a field applied in the c direction is

$$^{63}K_{\parallel}(2)=(-0.01\pm0.01)\% . \tag{3.1}$$

We can therefore, within the present experimental accuracy, take

$$A_{\parallel}=-4B , \tag{3.2}$$

and use Eqs. (2.11) and the nuclear moments $^{63}\gamma_n\hbar=7.5\times10^{-24}$ erg/G, $^{17}\gamma_n\hbar=3.8\times10^{-24}$ erg/G, $^{89}\gamma_n\hbar=1.3\times10^{-24}$ erg/G, and the experimental results $^{63}K_{\perp}=(0.30\pm0.02)\%$,[16] $^{17}K_{\rm iso}=(0.21\pm0.02)\%$,[12,17,18] $^{89}K_{\rm iso}=(0.03\pm0.01)\%$,[15] to determine the hyperfine couplings in terms of $B+A_{\perp}/4=B(1+\alpha)$. We find

$$K\equiv(1+\alpha)B\chi_0/\mu_B^2=1.21\times10^{-6} , \tag{3.3a}$$

$$C=(0.72\pm0.05)B(1+\alpha) , \tag{3.3b}$$

$$D=(1\pm0.3)\times10^{-2}B(1+\alpha) . \tag{3.3c}$$

Direct measurements of the spin susceptibility χ_0 are complicated by the need to determine the core and van Vleck contributions, subtract them from the measured bulk susceptibility to obtain a Pauli susceptibility, and then separate the plane from the chain contributions to $\chi_{\rm Pauli}$. A provisional estimate[4] is

$$\frac{\chi_0}{\mu_B^2}=3 \tag{3.4}$$

in states/eV Cu(2), implying

$$B(1+\alpha)=4\times10^{-7}\text{ eV}=42.9\text{ kOe}/\mu_B . \tag{3.5}$$

This estimate is only slightly larger than the value $B(1+\alpha)=3.6\times10^{-7}$ obtained by Yasuoka *et al.*[9] for $YBa_2Cu_3O_{6.48}$.

Antiferromagnetic resonance experiments in $YBa_2Cu_3O_{6.0}$ imply $B(1-\alpha)=2.8\times10^{-7}$ eV.[6] Our subsequent analysis of the anisotropy of the copper relaxation rate suggests $\alpha\approx0.20$–0.25, in which case the results of Ref. 6 imply $B=3.5$–3.7×10^{-7} eV, while Eq. (3.5) would imply the slightly smaller values $B=3.2$–3.3×10^{-7} eV. The consistency (within 15%) of the various estimates suggests that the hyperfine couplings do not change much upon doping and suggests that the Mila-Rice estimate of χ_0 (Ref. 4) is reasonable. Our derived values of B are somewhat smaller than the value $B=3.8\times10^{-7}$ eV estimated by Mila and Rice[4] from a quantum chemical analysis. Our values for α and $A_{\parallel}$ differ somewhat from those of Mila and Rice[4] because in their analysis they used a preliminary and, we now believe, incorrect experimental value for $^{63}K_{s\parallel}$.

We now consider the relaxation rates. It is convenient to define another dimensionless parameter Δ,

$$\Delta=\chi_0\hbar\Gamma/\mu_B^2 . \tag{3.6}$$

Using Eqs. (2.14), (2.16), (3.1), and (3.5) we find

$$^{63}W_{\parallel}=\frac{k_B T}{\hbar}\frac{2K^2}{(1+\alpha)^2\Delta}[1.51(1-\alpha)^2\beta(\xi/a)^2-1.51(1-\alpha)\beta\ln(\xi/a)-(0.15-0.90\alpha+0.13\alpha^2)\beta+4.71+18.9\alpha^2] , \tag{3.7a}$$

$$^{63}W_{\perp}=\tfrac{1}{2}{}^{63}W_{\parallel}+\frac{k_B T}{\hbar}\frac{2K^2}{(1+\alpha)^2\Delta}[3.02\beta(\xi/a)^2-1.51\beta\ln(\xi/a)-0.60\beta+11.78] , \tag{3.7b}$$

$$^{17}W=\frac{k_B T}{\hbar}\frac{2K^2}{\Delta}\frac{C^2}{(1+\alpha)^2B^2}[0.094\beta\ln(\xi/a)+0.040\beta+2.36] , \tag{3.7c}$$

$$^{89}W=\frac{k_B T}{\hbar}\frac{2K^2}{\Delta}\frac{D^2}{(1+\alpha)^2B^2}(0.19\beta+9.43) . \tag{3.7d}$$

We obtain values for β, Δ, ξ/a, and α by fitting Eqs. (3.7a)–(3.7c) to experiment, and then we test the consistency of the model by comparing Eq. (3.7d) with the observed yttrium relaxation rate. Before discussing the details of our analysis, we find it useful to estimate the order of magnitude of the enhancements involved by comparing the data with what one would expect for a noninteracting Fermi gas.

In a noninteracting Fermi gas the dynamic susceptibility at low ω is proportional to $(\omega/v_F q)$. It is easy to verify that

$$\sum_q S(q,\omega\to 0)=\frac{k_B T}{\hbar\omega}\sum_q \chi''(q,\omega\to 0)=\frac{\pi k_B T}{2\mu_B^2}\chi_0^2 \tag{3.8}$$

where, as before, χ_0 is the uniform susceptibility. If this Fermi gas were coupled via an isotropic contact interaction E to a nuclear moment, the dimensionless spin Knight shift K would be given by

$$K=E\chi_0/\mu_B^2 \tag{3.9}$$

and the rate W by

$$W=\frac{3E^2}{4\mu_B^2}\sum_q S(q,\omega\to 0)=\frac{3\pi}{8}K^2 k_B T\ . \tag{3.10}$$

The Korringa relation in this notation is

$$\Lambda=\frac{8W}{3\pi K^2 k_B T}=1\ . \tag{3.11}$$

The expression differs from that usually found in the literature because in our dimensionless Knight-shift parameter $K=4(\gamma_n/\gamma_e)K^{\text{expt}}$ we have absorbed the nuclear moment factor γ_n and normalized susceptibilities to μ_B^2, not $(g\mu_B)^2$.

In the phenomenology developed in Sec. II we would represent the susceptibility of a noninteracting Fermi gas by the form given in Eq. (2.1) but with the parameter $\beta=0$. Writing $\chi''(q,\omega)=\pi\chi_0\omega/\Gamma$, performing the sum over q, and comparing with Eq. (3.8) shows that the Fermi-gas value Δ^F of the parameter Δ is

$$\Delta^F=\chi_0\hbar\Gamma_F/\mu_B^2=2\ . \tag{3.12}$$

Thus to obtain Fermi-gas estimates for relaxation rates in $YBa_2Cu_3O_7$ we set $\beta=0$ and $\Delta=2$ in Eqs. (3.7). We note that the numerical factors will differ from those in Eq. (3.10) because each nucleus is coupled to electrons in several different cells, whereas in the derivation of Eq. (3.10) the coupling was assumed to be local. Using our ansatz, and Eqs. (3.7) we may compute the relaxation rates. From Eqs. (2.11) we may find the Knight shifts and form the Korringa product defined in Eq. (3.11). We find [using K defined in (3.3a), $\alpha=0.2$, $\beta=0$, and $\Delta=2$]

$$^{63}\Lambda\equiv\frac{8\,{}^{63}W_{\parallel}\hbar}{(3\pi)16K^2 k_B T}=0.20\ , \tag{3.13a}$$

$$^{17}\Lambda\equiv\frac{8\,{}^{17}W(1+\alpha)^2B^2\hbar}{(3\pi)4K^2C^2 k_B T}=0.50\ , \tag{3.13b}$$

$$^{89}\Lambda\equiv\frac{8\,{}^{89}W(1+\alpha)^2B^2\hbar}{(3\pi)64K^2D^2 k_B T}=0.125\ . \tag{3.13c}$$

In all cases the noninteracting value for the Korringa product is substantially smaller than unity. The observed values are larger; at $T=100$ K one finds

$$^{63}\Lambda_{\text{expt}}=2.9\pm0.2\ , \tag{3.14a}$$

$$^{17}\Lambda_{\text{expt}}=1.2\pm0.2\ , \tag{3.14b}$$

$$^{89}\Lambda_{\text{expt}}=0.15\pm0.1\ . \tag{3.14c}$$

Thus the oxygen Korringa product is enhanced by a factor of ~ 3 and the copper by a factor of ~ 15 over the naive estimate. Within our model the Cu enhancements will come principally from antiferromagnetic correlations (i.e., a nonzero β) and the oxygen and yttrium enhancements in part from a nonzero β and in part from a value of Δ smaller than the Fermi liquid value $\Delta=2$. We also note that as discussed in Sec. IV the uniform susceptibility is enhanced over the band-structure value by a factor of ~ 2.5, so that the relaxation rates are further enhanced over the noninteracting values.

We turn now to determining the parameters. In Fig. 3 we have plotted the T dependence of the Cu-relaxation rate $^{63}W_{\parallel}$ for several choices of parameters. Observe that the form of the curve is qualitatively very similar to the data for the high-T_c cuprates, with $^{63}W_{\parallel}\sim a+bT$ for large T and $^{63}W_{\parallel}\sim cT$ for small T. In our model the departure from the high-temperature behavior occurs at a temperature T^* somewhat larger than the reference temperature T_x, and appears to be relatively abrupt even though our correlation length has a rather smooth T dependence. In $YBa_2Cu_3O_7$ the departure form the high-temperature from becomes noticeable at $T\sim 100$ K, so we have chosen $T_x=100$ K although somewhat lower or higher values would also fit.

Figure 4(a) shows the anisotropy $^{63}W_{\perp}/{}^{63}W_{\parallel}$ for $\alpha=0.2$ and two of the $(\beta,\xi/a)$ pairs considered in Fig. 3. The anisotropy is seen to be large and weakly T dependent. The observed anisotropies[20,21] are also indicated on the graph. In Fig. 4(b) the anisotropy at $T=T_x$ is plotted as a function of α, for two (β,ξ) pairs. We find that

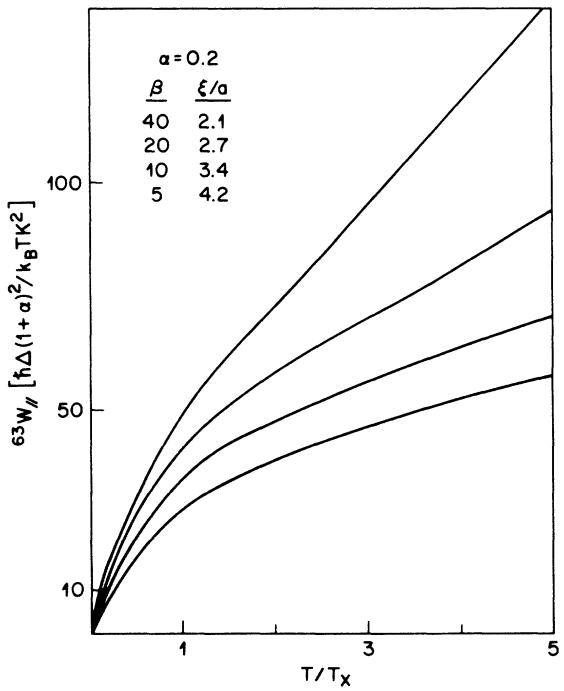

FIG. 3. Temperature dependence of normalized Cu relaxation rate computed from Eqs. (3.7a) and (2.9) for $\alpha=0.2$ and $(\beta,\xi/a)$ pairs indicated in the figure. $\beta=40$, $\xi/a=2.1$ corresponds to top curve, etc.

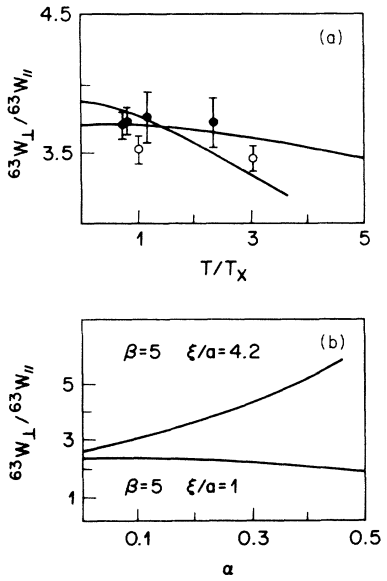

FIG. 4. Anisotropy of the Cu relaxation rate. (a) solid lines: T dependence of anisotropy for computed from Eqs. (2.9), (3.7a), and (3.7b) for $\alpha=0.2$ and $\beta=5$, $\xi/a=4.2$ (curve with less T dependence), and $\beta=20$, $\xi/a=2.7$ (curve with more T dependence). Open circles: experimental results (Ref. 20). Solid circles: experimental results (Ref. 21). (b) Dependence of anisotropy at $T=T_x$ on the ratio of transferred to direct hyperfine coupling $A_\perp/4B\equiv\alpha$.

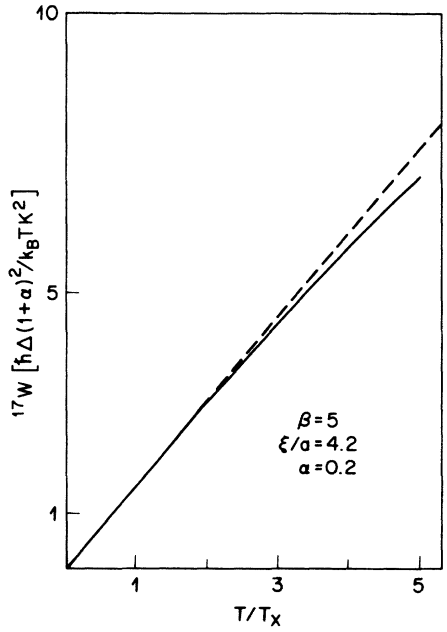

FIG. 5. T dependence of the normalized oxygen relaxation rate computed from Eqs. (2.9) and (3.7c) for the parameters indicated in the figure. The dashed line is $^{17}W\sim T$.

the magnitude, α, and T dependence of the anisotropy are only weakly dependent on β and ξ/a, *provided* that the product $\beta(\xi/a)^2$ is large enough. However, as shown in Fig. 4(b), if $\beta(\xi/a)^2$ is too small, the theory yields too small an anisotropy for any α. Thus we believe that the observed anisotropy of the Cu relaxation rates implies that the antiferromagnetic fluctuations provide an important contribution to the Cu relaxation rates. Monien, Pines, and Slichter[6] have shown that the maximum anisotropy obtainable in a one-band model with no antiferromagnetic correlations is 3, regardless of how α is varied. This result may be derived simply in our formalism by setting $\beta=0$ in Eqs. (3.7a) and (3.7b). They showed that the observed larger anisotropy could be explained by a two-band model; we see it is consistent with a one-band model with sufficiently large antiferromagnetic correlations.

We note that although the α dependence and order of magnitude of $^{63}W_\perp/^{63}W_\parallel$ are determined by the leading terms of Eqs. (3.7), the T dependence of this ratio is fixed by the interplay between leading and nonleading terms. Changes in the nonleading terms can change the sign of the T dependence. Thus it is possible that the agreement between our result for the T dependence of $^{63}W_\perp/^{63}W_\parallel$ and the experimental result is fortuitous.

Note the strong α dependence of the anisotropy in Fig. 4(b). The results of Refs. 20 and 21 constrain α to be $\alpha\sim0.20$, consistent with single-ion estimates.

Figure 5 shows the T dependence of the oxygen relaxation ^{17}W for one choice of parameters $\beta,\xi/a$. The behavior for other choices is very similar. Hammel *et al.*[2] find that the ratio of the copper to oxygen relaxation rates, $^{63}W_\parallel/^{17}W$, is 19 for temperatures less than 100 K in the $YBa_2Cu_3O_7$ system. This ratio determines β if ξ/a is fixed, or ξ/a if β is fixed. Figure 6 shows the T dependence of this ratio. The $(\beta,\xi/a)$ pairs used to generate the curves in Figs. 2 and 3(a) give $^{63}W_\parallel/^{17}W=19$ at $T=100$ K $=T_x$ if $\alpha=0.2$.

Thus far we have shown that the phenomenological model yields a qualitatively correct T dependence for the

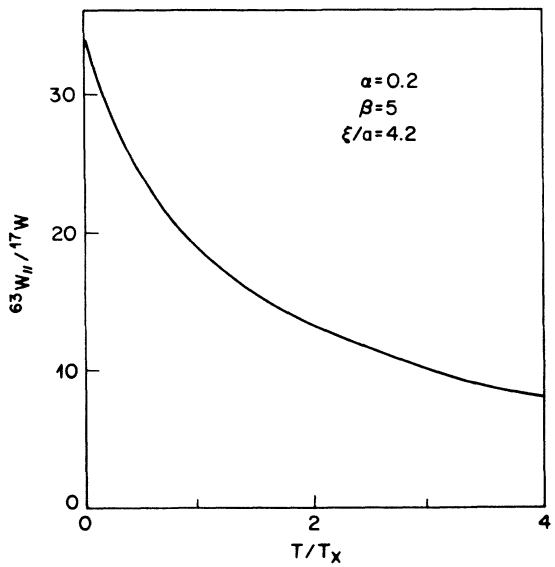

FIG. 6. T dependence of ratio of relaxation rates $^{63}W_\parallel/^{17}W$.

normal-state Cu and O relaxation rates, and we have used the anisotropy and the $^{63}W_{\parallel}/^{17}W$ ratio to constrain some of the parameters. We wish, however, to point out that our form Eq. (2.9) for the T dependence of ξ can only apply for temperatures greater than the superconducting transition temperature T_c. For all $T<T_c$ the ratio $^{63}W_{\parallel}/^{17}W$ is claimed to equal 19 ± 2,[2] even though both rates change by several orders of magnitude. If our Eq. (2.9) applied for $T<T_c$, the ratio would show substantial temperature dependence. The T dependence of the ratio for one particular choice of parameters is shown in Fig. 6. We return to this issue in Sec. IV.

To determine the remaining parameters we consider more quantitatively the T dependence of $^{63}W_{\parallel}$ given in Fig. 2. At high $T(T\gtrsim 2T_x)$ we find that the relaxation rate is approximately of the form $^{63}W_{\parallel}=a+bT$. By comparing the values of a and b determined from Fig. 3 with the experimental values $a=1000$ sec^{-1}, $b=2.9$ sec^{-1}/K, and using $T_x=100$ K we find that the curves with $\beta=5$ and 10 yield roughly the experimental value for b/a, while the curves with larger or smaller β do not. The curve with $\beta=10$ implies $\Delta\sim1.4$, rather smaller than the noninteracting Fermi liquid value $\Delta=2$, while the curve with $\beta=5$ implies $\Delta=1.0$.

Thus far we have obtained estimates for our parameters by considering relatively general features of the data, which have been observed by several groups. It is also of interest to fit as precisely as possible the experimental data of one group, although the significance of the detailed values of the parameters is unclear in view of the remaining differences (e.g., in the Cu anisotropy) between the results of various experimental groups. Our best fit to the Cu-relaxation data of Ref. 16 and 20, subject to the constraint that $^{63}W_{\parallel}/^{17}\mathrm{O}=19$ at $T=100$ K,[2] is shown in Fig. 7. The Cu anisotropy with these parameters is found to be 3.9 at $T=100$ K and 3.8 at $T=300$ K. As can be seen from Fig. 7, the agreement between our calculation and the data of Ref. 16 is very good. We note, however, that reasonably good fits could probably be obtained with other parameter values, provided $0.2\lesssim\alpha\lesssim0.25$ and $5\lesssim\beta\lesssim15$.

To test our parameter values, we calculate the yttrium relaxation rate, finding at $T=100$ K

$$^{89}W=(4\pm2)\times10^{-2}\ \mathrm{sec}^{-1},$$

consistent with the experimental result $^{89}W=3\times10^{-2}$ sec^{-1}.[15] The large error bars come principally from the 30% uncertainty in the hyperfine coupling D, which in turn comes from uncertainty in the magnitude of the spin part of the yttrium Knight shift; the uncertainty in Δ is a smaller effect.

In this section we have shown that Knight shifts and relaxation measurements in the $YBa_2Cu_3O_7$, $T_c=90$ compound can be quantitatively accounted for by a one-component model of antiferromagnetically correlated spins. In conclusion, we wish to draw attention to some uncertainties in our analysis. First, a range of β (and hence ξ/a) yield consistent accounts of experiment. Second, the mean-field form for χ'' we used may not be appropriate, and a different form for χ'' may yield a somewhat different value for the characteristic temperature T_x and a dramatically different correlation length.[3] Third, a χ'' peaked at an incommensurate Q only slightly different from $(\pi/a,\pi/a)$ would still lead to an approximately Korringa behavior for ^{17}W; however, the parameters we obtain will change somewhat. Fourth, the interpretation of our parameter β is not clear. Within a RPA treatment of the spin dynamics, β is defined by Eq. (2.7a). In this context a value of β greater than unity implies the existence of a microscopic length scale slightly shorter than a lattice constant. Whether this is reasonable is not known. Finally, the T dependence of $^{63}W_{\parallel}$ for $T>T_x$, which fixed the value of β, is determined by the interplay between leading and nonleading terms in Eq. (3.7a) and could change if a different form for χ'' were used.

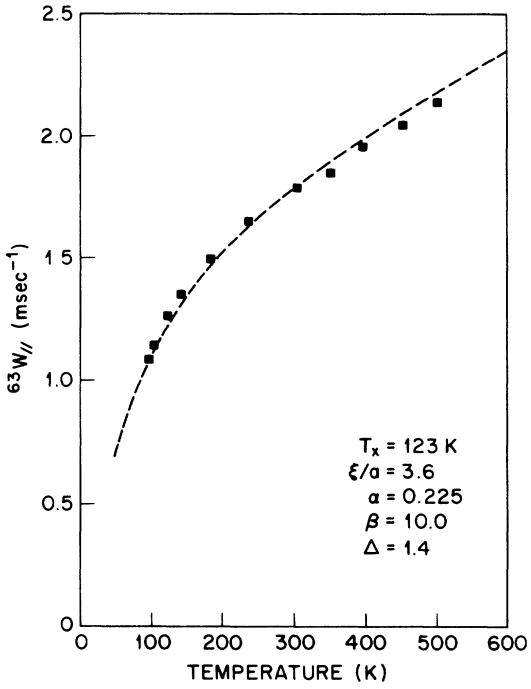

FIG. 7. Comparison of calculated and observed (Ref. 16) Cu relaxation rates for "best-fit" parameters indicated in the figure.

IV. DISCUSSION

In this section we will first discuss the implications of our analysis for the $YBa_2Cu_3O_7$, $T_c=90$ K material and then consider the extension of our phenomenology to other cuprate superconductors.

We begin with the uniform susceptibility, $\chi_0/\mu_B^2=3$ states/eV Cu(2) [Eq. (3.4)]. We are interested in the contribution to χ_0 from spin excitations in the CuO_2 planes; as discussed earlier, this quantity is difficult to determine unambiguously from experiment. It is of interest to compare the experimental value with the value predicted by band theory. The susceptibility for CuO_2 planes may be obtained from calculation for $La_{2-x}Sr_xCuO_4$.[22] Provided that the doping in the planes in $YBa_2Cu_3O_7$ is not too close to $x=0.15$ (where the band calculation predicts a

van Hove singularity that enhances χ_{band} by a factor of almost 2) we find that χ_0 is enhanced over the band value by a factor of ~ 2.5. We note that the band-structure density of states for $YBa_2Cu_3O_7$ is 5.6 states/eV cell.[22] Naively dividing by 3 would yield $N_0 \sim 1.9$ states /eV Cu $\lesssim \chi_0/2\mu_B^2$. We believe, however, that the calculation for La_2CuO_4 yields a better estimate of the band-theory prediction for the CuO_2 planes.

From the oxygen relaxation rate (and, with less certainty, from the yttrium) we find that the product $\Delta = \chi_0 \hbar\Gamma/\mu_B^2$ is about $\frac{2}{3}$ of the noninteracting Fermi-liquid value of 2. This indicates that the physics leading to the enhancement of χ_0 over the band-structure value also leads to a comparable or greater decrease in the typical spin fluctuation frequency Γ. Using the estimate for χ_0 given in Eq. (3.4) we find $\hbar\Gamma \sim 0.4$ eV. As will be shown, this value for Γ is only weakly doping dependent. Such behavior would be expected within "t-J models"[4] and is apparently also found in the RPA calculations of Bulut *et al.*[9] Further calculations of χ_0 and Δ for specific microscopic models would be useful.

Because the Cu Knight shift K is proportional to the susceptibility χ_0 and the relaxation rates are proportional to K^2/Δ, it is clear that all of the relaxation rates and Knight shifts would be substantially enhanced (by a factor of 5 or more) over their band-structure values even if ξ/a or β were to vanish.

It is also worth pointing out that the magnitude of the Cu relaxation rate provides further evidence for antiferromagnetic correlations, if the one-component hypothesis is accepted. In a one-component model the Cu and O relaxation rates can differ only via coupling constants, which are fixed by the Knight shifts. After these are divided out, an enhancement of the magnitude of Cu over O remains at any T. This can be due only to antiferromagnetic correlations.

We now consider the T dependence of the spin-correlation length. We found in Sec. III that the increase of ξ as T decreases is cut off on the surprisingly low energy scale of $T_x \sim 100$ K. Of course the small energy scale is directly implied by the non-Korringa behavior of the Cu relaxation, which persists down to ~ 100 K in the $YBa_2Cu_3O_7$ material. However, it is puzzling that such a small energy scale should appear in a material where the basic electronic energy scales (set, e.g., by $\chi_0 \sim 3$ states/eV Cu [Eq. (3.4)] or by the optically determined quasiparticle Fermi energy $E_F^* \sim 0.4$ eV) (Ref. 23) are of the order of several tenths of a volt and where the correlation lengths are not too long.

As mentioned in Sec. III, the observed[2] T independence of the ratio ${}^{63}W_{\parallel}/{}^{17}W$ for $T < 100$ K suggests that ξ becomes T independent below this temperature. It is not clear to us why, in the absence of a magnetic transition, the correlation length should so abruptly saturate. Further, it is remarkable that the antiferromagnetic correlations whose characteristic energy scale is $T_x \sim 100$ K are apparently unaffected by the onset of superconductivity at $T_c \sim 90$ K. This issue is discussed further by one of us.[24]

We have asserted, following Hammel *et al.*[2] and Shastry[3] that in order to explain the different T dependence of the Cu and O relaxation rates one must postulate a χ'' that is strongly peaked about the zone corner. Such a peak should be observable via neutron scattering; if it is not, the whole theory must be questioned. The energy scale associated with the peak may be estimated from Eq. (2.8). It is T dependent; using the results $[\xi(T=100\ K)/a] \sim 2.5$, $\hbar\Gamma \sim 0.4$ eV, we estimate

$$\hbar\omega_{\text{SF}} \sim 20 \text{ meV } (T = 100 \text{ K}) .$$

Neutron-scattering experiments on superconducting cuprates are difficult to perform because of the lack of sufficiently large single crystals and are difficult to interpret because of doubts concerning the homogeneity of available samples. Neutron studies of $\chi''(q,\omega)$ for q along the zone diagonal for $La_{2-x}Sr_xCuO_{4-\delta}$ samples have detected antiferromagnetic correlations peaked near, but not at, $(\pi/a, \pi/a)$ with a correlation length of approximately 3–5 lattice constants.[25] The correlation length is claimed to have only a weak T dependence for $T < 300$ K. Subsequent studies of the ω dependence of $\sum_q \chi''(q,\omega)$ (Ref. 26) have detected interesting behavior consistent[27] with the development of antiferromagnetic correlations as we have discussed here. The energy scale associated with this behavior is ~ 6 meV, somewhat smaller than our value for $\hbar\omega_{\text{SF}}$. Studies of $YBa_2Cu_3O_{6+x}$ samples with $x \sim 0.4$–0.5, which are semiconducting but are close to the metal insulator transition, have revealed similar behavior.[28] However, an early study of an $YBa_2Cu_3O_{7-\delta}$ ($T_c = 90$) sample apparently detected no magnetic scattering in the frequency range $\omega < 25$ meV.[29] This result is inconsistent with our analysis.

In sum, neutron-scattering experiments on the $La_{2-x}Sr_xCuO_4$ system and on low-x $YBa_2Cu_3O_{6+x}$ samples have shown the existence of dynamic antiferromagnetic correlations. The T dependence of the neutron-measured correlation length may be different than the T dependence we require to explain the magnetic relaxation measurements. Whether this behavior persists in the $YBa_2Cu_3O_7$ material has in our opinion not yet been determined. If no antiferromagnetic correlations are found in the $YBa_2Cu_3O_7$ material, then new physics (including a two-band model for the magnetic susceptibility) will be required to explain the magnetic resonance data in this superconductor.

We turn now to NMR experiments on lower T_c superconducting oxides. There is less information available than in the $T_c = 90$ K case and, perhaps because the materials problems are more severe in reduced T_c samples, the agreement between experimental groups is less good. For these reasons we have deferred detailed fits of data from reduced T_c samples to a future paper.[30] We wish, however, to comment on two issues.

The first is the temperature dependence of the Knight shifts and relaxation rates in reduced T_c materials. For $T < 300$ K the spin Knight shifts on all sites decrease as T is lowered.[15,31] The only reasonable interpretation of this result is that the static susceptibility χ_0 is strongly T dependent, decreasing substantially as T is lowered from room temperature to T_c. The microscopic origin of this

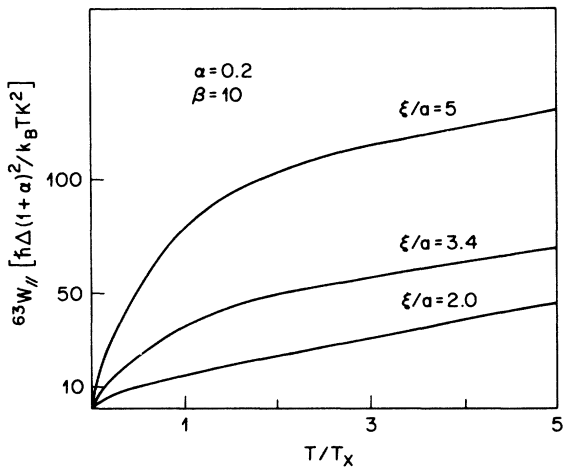

FIG. 8. Effect of varying magnetic correlation length upon T dependence of the Cu relaxation rate. Parameters are indicated in the figure.

T dependence is not understood at present. For $T<300$ K the ratios $^{17}W/T$ and $^{89}W/T$ also decrease as T is lowered,[15,31] and are claimed to have T dependences identical to those of $\chi_0(T)$ (Ref. 21) or $[\chi_0(T)]^2$ (Ref. 15). The Cu rate $^{63}W_{\parallel}/T$ has a nonmonotonic T dependence:[32] for 120 K $\lesssim T \lesssim$ 300 K $^{63}W_{\parallel}/T$ increases as T decreases; for $T<120$ K it decreases as T decreases. Our model χ'' must be extended to account for these data. One possible modification is to assume the parameter χ_0 in Eq. (2.1) has the T dependence of the spin Knight shift. Whether this modification suffices to explain the data or whether additional T dependence is required is presently under investigation.[30]

The second is the trend in the Cu relaxation rate $^{63}W_{\parallel}$ as T_c is reduced. It is clear from the data of Imai *et al.*[14] that as oxygen is removed from the $YBa_2Cu_3O_{6+x}$ system (i) the magnitude of $^{63}W_{\parallel}$ increases, (ii) the crossover between the high- and low-T regimes apparently moves to higher temperature, and (iii) the slope in the high-T regime does not change much. Figure 8 shows that these three features are in at least qualitative agreement with the behavior of our model if the correlation length is increased and all other parameters are held fixed. Of course, the calculations in Fig. 8 assume T-independent Knight shifts.

V. RELATED WORK

The Hamiltonian with which we work was derived by Mila and Rice;[4] the derivation was improved and O sites were incorporated by Shastry.[3] A qualitative discussion of normal-state properties in this context was given by Shastry[3] for Cu and O sites and later by Mila and Rice[5] for Y sites. Our work is a more thorough and quantitative exploration of the model proposed by these authors, and our conclusions are in agreement with theirs.

Monien, Pines, and Slichter,[6] Monien and Pines,[7] and Cox and Tree[8] have considered two component models. It is our present opinion that the reasons given in Sec. I and the quantitative success of our one-component phenomenology render the two component models unlikely. Monien and Pines[7] have shown quantitatively that a phenomenological two-component model can account also for the Cu and O relaxation. Cox and Tree[8] have considered a more microscopic model in which antiferromagnetically correlated local moments on Cu sites are modeled via a Schwinger boson mean-field theory and are weakly coupled to a Fermi liquid of holes that are supposed to reside on the O p-π orbitals. They have obtained qualitative agreement with experiments. Cox and Tree conclude, as do we, that antiferromagnetic correlations of the Cu moments are required to explain the data. Cox and Tree also draw attention to the super-Korringa behavior of the Cu relaxation rate at low T in reduced T_c Y-Ba materials. They agree with Warren *et al.*[32] that this behavior is due to the opening of a gap; however, in contrast to Warren *et al.* they do not interpret the gap as a precursor to a superconducting gap, but instead suggest that it could be understood in a purely magnetic model. They obtain a qualitatively plausible T dependence from a calculation based on an anisotropic Heisenberg model. However, if the observed anisotropy of insulating $YBa_2Cu_3O_6$ is used, an unreasonably large correlation length $\xi/a \sim 100$ is apparently required.

A rather different phenomenology has been proposed by Varma *et al.*[10] In this phenomenology, susceptibilities such as $\chi''(q,\omega)$ are given by the sum of two terms; a quasiparticle contribution and another contribution coming from a polarization part, which is assumed to be roughly q independent and assumed to scale as ω/T for $\omega<T$ and to be ω independent for $\omega>T$. With this form they can account for the T dependence of $^{63}W_{\parallel}(T)$ for $T>T_x$. The oxygen and yttrium rates are not considered. We believe it will be difficult to account for the oxygen and yttrium data in this way unless either a two-component model is assumed (so that different degrees of freedom relax the Cu than the O or Y) or the polarization part is peaked near the zone corner. We note that our χ'' is approximately of the form proposed by Varma *et al.*, since $\sum_q \chi''$ may be written as the sum of a term proportional to $\omega/T+T_x$ and a constant term.

Finally, we note that Bulut *et al.*[9] have determined Cu, O, and Y relaxation rates using the hyperfine Hamiltonian used here and a RPA approximation to the Hubbard model to calculate $\chi''(q,\omega)$. They obtain results qualitatively similar to ours, and conclude, as we do, that a theory involving an antiferromagnetically correlated one-component spin system can explain the various data. They find it necessary to assume that the bandwidth of the conduction electrons is much smaller than the band-theory value. This is presumably related to the small value we find for our parameter Γ. They require, however, a larger antiferromagnetic correlation length ($\xi/a \sim 7$) than we do, presumably because in their theory the parameter β is not adjustable and is smaller than our fitted estimate of $\beta=10$. The extent to which varying our parameter β is theoretically justified is not understood.

VI. CONCLUSION

It is widely believed that magnetic relaxation experiments on high-T_c superconducting cuprates imply the existence of antiferromagnetic correlations in these materials. In this paper we have made this point of view more quantitative by analyzing a phenomenological model of a one-component system of antiferromagnetically correlated spins. We have found that all of the available normal-state NMR and NQR measurements in the $YBa_2Cu_3O_7$ system can be accounted for by a model susceptibility strongly peaked at the zone corner. Using a mean-field-theory ansatz we find an antiferromagnetic correlation length of 2.5 lattice constants and an energy scale for antiferromagnetic spin fluctuations of ~ 20 meV at $T=100$ K. The energy scale for a typical spin fluctuation at q away from the zone corner was estimated to be ~ 0.4 eV, much smaller than predicted by band theory but similar to that found in the insulator.

Our results thus demonstrate that a one-component model can explain the data, and show that if the one-component hypothesis is accepted, then antiferromagnetic correlations among the spins are necessary. The antiferromagnetic correlations ought to be observable via neutron scattering (which has not been done for $YBa_2Cu_3O_7$). If the magnetic correlations we have discussed are not observed, a radical rethinking of our approach to high-T_c superconductivity will be required.

Note added in proof. The analysis[30] of experiments[15,31] on $YBa_2Cu_3O_{6.63}$ using the model χ presented here has been completed. In $YBa_2Cu_3O_{6.63}$ the static susceptibility is strongly temperature dependent. The authors of Ref. 30 include this T dependence in the parameter χ_0 and use the same hyperfine couplings and β as were used to fit $YBa_2Cu_3O_7$. They obtain a quantitative fit to the data[15,31] and find a correlation length that is larger than in $YBa_2Cu_3O_7$ ($\xi/a=3$ at $T=100$ K) and saturates at a lower temperature. A similar analysis is in progress[33] for $La_{2-x}Sr_xCuO_4$. Refined estimates for the parameters appropriate to $YBa_2Cu_3O_7$ have also become available:[30] $B=3.8\times10^{-7}$ eV$=41$ kOe/μ_B, $C=6.4\times10^{-7}$ eV, $D=-3\times10^{-8}$ eV, $\alpha=0.2$, $\beta=9.9$, $[\xi(T=0)/a]=3.1$, and $T_x=115$ K. The results of this paper and Refs. 30 and 33 support the notion that antiferromagnetic correlations are present in all cuprate superconductors and raise again the question of whether the physics of the cuprates may be understood in terms of a strongly antiferromagnetically correlated Fermi liquid.

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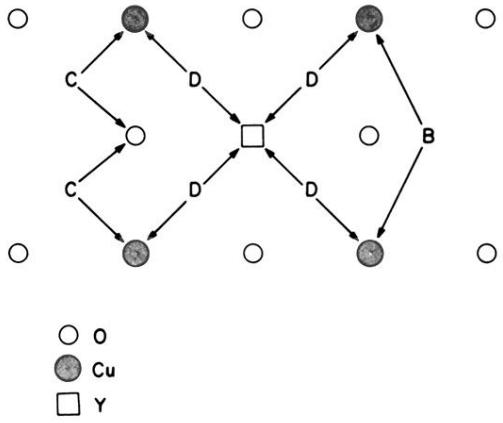

FIG. 2. Sketch of atoms near one CuO_2 plane with hyperfine couplings [Eq. (2.10)] indicated. Note that the Y atom couples to two Cu-O planes, although only the coupling to one is shown.