# NMR and neutron-scattering experiments on the cuprate superconductors: A critical re-examination

Y. Zha
*Department of Physics and Science and Technology Center for Superconductivity, University of Illinois at Urbana-Champaign, 1110 West Green Street, Urbana, Illinois 61801*

V. Barzykin
*Department of Physics, University of British Columbia, 6224 Agricultural Rd., Vancouver, British Columbia, Canada V6T 1Z1*

D. Pines
*Department of Physics and Science and Technology Center for Superconductivity, University of Illinois at Urbana-Champaign, 1110 West Green Street, Urbana, Illinois 61801*

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We show that it is possible to reconcile NMR and neutron-scattering experiments on both $La_{2-x}Sr_xCuO_4$ and $YBa_2Cu_3O_{6+x}$, by making use of the Millis-Monien-Pines mean-field phenomenological expression for the dynamic spin-spin response function, and re-examining the standard Shastry-Mila-Rice hyperfine Hamiltonian for NMR experiments. The recent neutron-scattering results of Aeppli *et al.* on $La_{1.86}Sr_{0.14}CuO_4$ are shown to agree quantitatively with the NMR measurements of $^{63}T_1$ and the magnetic scaling behavior proposed by Barzykin and Pines. The reconciliation of the $^{17}O$ relaxation rates with the degree of incommensuration in the spin-fluctuation spectrum seen in neutron experiments is achieved by introducing a transferred hyperfine coupling $C'$ between $^{17}O$ nuclei and their next-nearest-neighbor $Cu^{2+}$ spins; this leads to a near-perfect cancellation of the influence of the incommensurate spin-fluctuation peaks on the $^{17}O$ relaxation rates of $La_{2-x}Sr_xCuO_4$. The inclusion of the $C'$ term also leads to a natural explanation, within the one-component model, the different temperature dependence of the anisotropic $^{17}O$ relaxation rates for different field orientations, recently observed by Martindale *et al.* The measured significant decrease with doping of the anisotropy ratio, $^{63}R={}^{63}T_{1ab}/{}^{63}T_{1c}$ in the $La_{2-x}Sr_xCuO_4$ system, from $^{63}R=3.9$ for $La_2CuO_4$ to $^{63}R\simeq 3.0$ for $La_{1.85}Sr_{0.15}CuO_4$ is made compatible with the doping dependence of the shift in the incommensurate spin-fluctuation peaks measured in neutron experiments, by suitable choices of the direct and transferred hyperfine coupling constants $A_\beta$ and $B$. [S0163-1829(96)04830-8]

## I. INTRODUCTION

The magnetic behavior of the planar excitations in the cuprate superconductors continues to be of central concern to the high-temperature superconductivity community. Not only does it provide significant constraints on candidate theoretical descriptions of their anomalous normal-state behavior, but it may also hold the key to the physical origin of high-temperature superconductivity. Recently two of us have used the results of NMR experiments to determine the magnetic phase diagram for the $La_{2-x}Sr_xCuO_4$ and $YBa_2Cu_3O_{6+x}$ systems.[2] We found that for both systems bulk properties, such as the spin susceptibility, and probes in the vicinity of the commensurate antiferromagnetic wave vector $(\pi,\pi)$, such as $^{63}T_1$, the $^{63}Cu$ spin relaxation time, and $^{63}T_{2G}$, the spin-echo decay time, display $z=1$ scaling and spin-pseudogap behavior over a wide regime of temperatures. On the other hand, the neutron-scattering experimental results of Aeppli *et al.*[1] on $La_{1.86}Sr_{0.14}CuO_4$ which probe directly $\chi''(q,\omega)$, the imaginary part of the spin-spin response function, while supporting this proposed scaling behavior, at first sight appear incapable of explaining NMR experiments on this system.

This apparent contradiction between the results of NMR and neutron-scattering experiments, both of which probe $\chi(q,\omega)$ in $La_{1.86}Sr_{0.14}CuO_4$, is but one of a series of such apparent contradictions. For example, in the $YBa_2Cu_3O_{6+x}$ system, NMR experiments on $^{63}Cu$ and $^{17}O$ nuclei in both $YBa_2Cu_3O_7$ (Ref. 4) and $YBa_2Cu_3O_{6.63}$ (Ref. 5) require the presence of strong antiferromagnetic correlations between the planar $Cu^{2+}$ spins, and a simple mean-field description of the spin-spin response function with a temperature-dependent magnetic correlation length $\xi\gtrsim 2$, was shown to provide a quantitative description of the measured results for $^{63}T$ and $^{17}T_1$ in $YBa_2Cu_3O_7$,[6] and $YBa_2Cu_3O_{6.63}$.[7] Yet neutron-scattering experiments on $YBa_2Cu_3O_7$ (Refs. 8–10) and $YBa_2Cu_3O_{6.63}$,[11] find only comparatively broad, temperature-independent, peaks in $\chi''(q,\omega)$, corresponding to a quite short ($\xi\lesssim 1$) temperature-independent magnetic correlation length. The apparent contradiction is especially severe for the $La_{2-x}Sr_xCuO_4$ system, where neutron-scattering experiments show at low temperatures four incommensurate peaks in the spin-fluctuation spectrum, whose position depends on the level of Sr doping,[12] while the quantitative explanation (using the same one-component phenomenological description which worked for the $YBa_2Cu_3O_{6+x}$ system) of the measurements of $^{63}T_1$ and $^{17}T_1$ in this system requires that the spin fluctuations be peaked at $(\pi,\pi)$, or nearly so.[13,14] Viewed from the NMR perspective, there are two major problems with four incommensurate spin-fluctuation peaks. First, the Shastry-Mila-Rice (SMR) form factor,[15,16] which, provided the peaks are nearly at $(\pi,\pi)$, effectively screens neighboring $^{17}$O nuclei from the presence of the strong peaks in the nearly localized $Cu^{2+}$ spin spectrum required to explain the anomalous temperature-dependence behavior of $^{63}T_1$, fails to do so for the considerable degree of incommensuration in the peaks at $(\pi,[\pi\pm\delta])$ and $([\pi\pm\delta],\pi)$ seen in $La_{1.86}Sr_{0.14}CuO_4$.[14,17,18] As a result $^{17}T_1$ picks up a substantial anomalous temperature dependence which is not seen experimentally. Second, with the doping-independent values of the hyperfine couplings which appear in the SMR form factors for a commensurate spectrum, the calculated anisotropy of $^{63}T_1$ for the incommensurate peaks seen by neutrons is in sharp variance with what is seen in the NMR experiments.[14]

Two ways out of these apparent contradictions have been proposed. One view is that the spin-fluctuation peaks seen in the neutron-scattering experiments reflect the appearance of discommensuration, not incommensuration; on this view, the $La_{2-x}Sr_xCuO_4$ system contains domains in which the spin-fluctuation peaks are commensurate (so that there are no problems with $^{17}T_1$), but what neutrons, a global probe, see is the periodic array of the domain walls.[19] A second view is that a one-component description of $\chi(\mathbf{q},\omega)$ is not feasible; rather, the transferred hyperfine coupling between the nearly localized $Cu^{2+}$ spins and the $^{17}$O nuclei is presumed to be very weak, and the $^{17}$O nuclei are assumed to be relaxed by a different mechanism, whence the nearly Korringa-like behavior of $^{17}T_1$.[18] A further challenge to a one-component description has come from the very recent work of Martindale *et al.*[3] who find that their results for the temperature dependence of the planar anisotropy of $^{17}T_{1\alpha}$ for different field orientations appear incompatible with a one-component description.

In the present paper we present a third view: that the one-component phenomenological description is valid, but what requires modification are the hyperfine couplings which appear in the SMR Hamiltonian which describes planar nuclei coupled to nearly localized $Cu^{2+}$ spins. We find that by introducing a transferred hyperfine coupling $C'$, between the next-nearest-neighbor $Cu^{2+}$ spins and a $^{17}$O nucleus, the nearly antiferromagnetic part of the strong signals emanating from the $Cu^{2+}$ spins can be far more effectively screened than is possible with only a nearest-neighbor transferred hyperfine coupling, so that the existence of four incommensurate peaks in the $La_{2-x}Sr_xCuO_4$ system can be made compatible with the $^{17}T_1$ results. We also find that by permitting the transferred hyperfine coupling, $B$, between a $Cu^{2+}$ spin and its nearest-neighbor $^{63}$Cu nucleus to vary with doping, we can explain the trend with doping of the anisotropy of $^{63}T_1$ in this system. We then use these revised hyperfine couplings to re-examine the extent to which the recent results of Aeppli *et al.*[1] on $La_{1.86}Sr_{0.14}CuO_4$ can be explained quantitatively by combining the Millis-Monien-Pines (hereafter MMP) response function[6] with the scaling arguments put forth by Barzykin and Pines.[2] We find that they can, and are thus able to reconcile the neutron-scattering and NMR experiments on this member of the $La_{2-x}Sr_xCuO_4$ system.

We present as well the results of a reexamination of the NMR and neutron results for the $YBa_2Cu_3O_{6+x}$ system. Here we begin by making the ansatz that it is the presence of incompletely resolved incommensurate peaks which is responsible for the broad lines seen in neutron experiments. We follow Dai *et al.*[9] who suggest the increased linewidth for $YBa_2Cu_3O_7$ seen along the zone diagonal directions reflects the presence of four incommensurate peaks, located at $\mathbf{Q}_i=(\pi\pm\delta,\pi\pm\delta)$, a proposal which is consistent with the earlier measurements of Tranquada *et al.* for $YBa_2Cu_3O_{6.6}$.[11] We then find that incommensuration can be made compatible with NMR experimental results provided the transferred hyperfine coupling constant $B$ is somewhat doping dependent in this system as well. Moreover, on considering $^{17}T_1$ for $YBa_2Cu_3O_7$, we find that incommensuration combined with the presence of the next-nearest-neighbor coupling, $C'$, leads to results which are consistent with the experimental results of Martindale *et al.*,[3] who find an anomalous temperature dependence of the planar anisotropy of $^{17}T_1$. This agreement with experiment preserves the one-component description of the planar spin excitation spectrum and provides an independent check on the presence of $C'$-like terms in the hyperfine Hamiltonian.

The outline of our paper is as follows: In Sec. II we review the SMR description of coupled $Cu^{2+}$ spins and nuclei as well as the mean-field description of $\chi(\mathbf{q},\omega)$, and examine the modifications brought about by incommensuration and next-nearest-neighbor coupling between $Cu^{2+}$ spins and a $^{17}$O nucleus. In Sec. III we review the experimental constraints on the hyperfine coupling parameters, and present our results for their variation with doping in both the $La_{2-x}Sr_xCuO_4$ and $YBa_2Cu_3O_{6+x}$ systems. We show in Sec. IV how the $^{63}$Cu NMR results can be reconciled with neutron-scattering results on $La_{1.86}Sr_{0.14}CuO_4$, while in Sec. V we present a quantitative fit to the $^{17}T_{1c}$ results for the $La_{1.85}Sr_{0.15}CuO_4$ based on the four incommensurate peaks in the spin fluctuation spectrum expected from neutron scattering. We show in Sec. VI how the anomalous results of Martindale *et al.*[3] for the $YBa_2Cu_3O_{6+x}$ system can be explained using our modified one-component model, and in Sec. VII, we give a quantitative comparison of the predictions of $\chi''(\mathbf{Q},\omega)$ in $YBa_2Cu_3O_7$ based on our analysis of the NMR experiments, with the neutron-scattering results of Fong *et al.*[23] Finally, in Sec. VIII, we present our conclusions.

## II. A GENERALIZED SHASTRY-MILA-RICE HAMILTONIAN

On introducing a hyperfine coupling $C'_{\alpha,\beta}$ between the the $^{17}$O nuclei and their next-nearest-neighbor $Cu^{2+}$ spins, we can rewrite the SMR hyperfine Hamiltonian for the $^{63}$Cu and $^{17}$O nuclei as

$$H_{\rm hf}={}^{63}I_\alpha(\mathbf{r}_i)\left[\sum_\beta A_{\alpha,\beta}S_\beta(\mathbf{r}_i)+B\sum_j^{\rm NN} S_\alpha(\mathbf{r}_j)\right]+{}^{17}I_\alpha(\mathbf{r}_i)\times\left[C_{\alpha,\beta}\sum_{j,\beta}^{\rm NN} S_\beta(\mathbf{r}_j)+C'_{\alpha,\beta}\sum_{j,\beta}^{\rm NNN} S_\beta(\mathbf{r}_j)\right], \qquad (1)$$

where $A_{\alpha,\beta}$ is the tensor for the direct, on-site coupling of the $^{63}$Cu nuclei to the $Cu^{2+}$ spins, $B$ is the strength of the transferred hyperfine coupling of the $^{63}$Cu nuclear spin to the four nearest-neighbor $Cu^{2+}$ spins, $C_{\alpha,\beta}$ is the transferred hyperfine coupling of the $^{17}O$ nuclear spin to its nearest-neighbor $Cu^{2+}$ spins, and $C'_{\alpha,\beta}$ its coupling to the next-nearest-neighbor $Cu^{2+}$ spins. The indices ''NN'' represent nearest-neighbor electron spins to the specific nuclei, and ''NNN'' the next-nearest-neighbor $Cu^{2+}$ spins. As we shall see below, inclusion of the $C'_{\alpha,\beta}$ term enhances the cancellation of the anomalous antiferromagnetic spin fluctuations seen by the $^{17}O$ nucleus, and therefore reduces the leakage from incommensurate spin-fluctuation peaks to the $^{17}O$ relaxation rates. It thus enables us to reconcile the measured $^{17}O$ relaxation rates with the neutron-scattering experiments for both $La_{2-x}Sr_xCuO_4$ and $YBa_2Cu_3O_{6+x}$.

The spin contributions to the NMR Knight shift for the various nuclei are[6]

$$^{63}K_c=\frac{(A_c+4B)\chi_0}{^{63}\gamma_n\gamma_e\hbar^2},$$

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

$$^{17}K_\beta=\frac{2(C_\beta+2C'_\beta)\chi_0}{^{17}\gamma_n\gamma_e\hbar^2}. \tag{2}$$

Here, we have incorporated the $C'_\beta$ term into the $^{17}O$ Knight-shift expression for $^{17}K_\beta$, while the others remain their standard form as in Ref. 6; $\gamma_n$ are various nuclei gyromagnetic ratios, $\gamma_e$ is the electron gyromagnetic ratio, and $\chi_0$ is the static spin susceptibility. The indices $c$ and $ab$ refer to the direction of the applied static magnetic field along the $c$ axis and the $ab$ plane. The spin-lattice relaxation rate, $(^\alpha T_1)^{-1}_\beta$ for nuclei $\alpha$ responding to a magnetic field in the $\beta$ direction, is

$$^\alpha T^{-1}_{1\beta}=\frac{k_BT}{2\mu_B^2\hbar^2\omega}\sum_{\mathbf{q}}\ ^\alpha F_\beta(\mathbf{q})\chi''(\mathbf{q},\omega\to 0), \tag{3}$$

where the modified SMR form factors, $^\alpha F_\beta(\mathbf{q})$, are now given by

$$^{63}F_c=[A_{ab}+2B(\cos q_xa+\cos q_ya)]^2,$$

$$^{63}F_{ab}=\frac{1}{2}[^{63}F_c+{}^{63}F^{\text{eff}}_{ab}],$$

$$^{63}F^{\text{eff}}_{ab}=[A_c+2B(\cos q_xa+\cos q_ya)]^2,$$

$$^{17}F_\alpha=2\sum_{\alpha_i=\alpha',\alpha''}\cos^2\frac{q_xa}{2}(C_{\alpha_i}+2C'_{\alpha_i}\cos q_ya)^2, \tag{4}$$

Here, $\alpha'$ and $\alpha''$ are the directions perpendicular to $\alpha$. The form factor $^{63}F^{\text{eff}}_{ab}$ is the filter for the $^{63}Cu$ spin-echo decay time $^{63}T_{2G}$:[20]

$$^{63}T^{-2}_{2G}=\frac{0.69}{128\hbar^2\mu_B^4}\left\{\frac{1}{N}\sum_{\mathbf{q}}\ F^{\text{eff}}_{ab}(\mathbf{q})^2[\chi'(\mathbf{q},0)]^2 -\left[\frac{1}{N}\sum_{\mathbf{q}}\ F^{\text{eff}}_{ab}(\mathbf{q})\chi'(\mathbf{q},0)\right]^2\right\}. \tag{5}$$

The values of the hyperfine constant $C_\alpha$ and $C'_\alpha$ can be determined from the various $^{17}O$ Knight-shift data. In fact, we may obtain these new values from the ''old'' values of the hyperfine coupling constant, $C^{\text{old}}_\alpha$ which have been well established by fitting the Knight-shift data.[2] Note we use $C_\alpha$ to represent the new nearest-neighbor hyperfine coupling constant, while the old hyperfine coupling constant is written explicitly as $C^{\text{old}}_\alpha$ throughout the paper. In order not to change the Knight-shift result of the previous analysis,[2] the new hyperfine coupling constants should satisfy the following requirement:

$$C_\alpha+2C'_\alpha=C^{\text{old}}_\alpha=\zeta_\alpha C^{\text{old}}_c, \tag{6}$$

where $\zeta_\alpha=C^{\text{old}}_\alpha/C^{\text{old}}_c$ and $c$ denotes the case of a magnetic field along the $c$ axis. For $YBa_2Cu_3O_{6+x}$, from the previous analysis of Yoshinari[21] and Martindale *et al.*,[3] we have for a field parallel to the Cu-O bond, $\zeta_\parallel=1.42$, and $\zeta_\perp=0.91$ for a field perpendicular to the Cu-O bond direction, while $\zeta_c=1$. On introducing $r_\alpha\equiv C'_\alpha/C_c$ we obtain

$$C_\alpha=C^{\text{old}}_c\left(\zeta_\alpha-\frac{2r_\alpha}{2r_c+1}\right),$$

$$C'_\alpha=C^{\text{old}}_c\frac{r_\alpha}{2r_c+1}. \tag{7}$$

Substituting these values of $C_\alpha$ and $C'_\alpha$ into Eq. (4), we obtain the new $^{17}O$ form factor in terms of $C^{\text{old}}_\alpha$:

$$^{17}F_\alpha=\frac{2(C^{\text{old}}_c)^2}{(1+2r_c)^2}\sum_{\alpha_i=\alpha',\alpha''}\cos^2\frac{q_xa}{2}[\zeta_{\alpha_i}(1+2r_c)-2r_{\alpha_i}+2r_{\alpha_i}\cos q_ya]^2. \tag{8}$$

Although $C'_\alpha$ may well be anisotropic (as $C_\alpha$ is), in the absence of detailed quantum chemistry calculations, (which lie beyond the purview of the present paper) we assume $C'_\alpha$ to be isotropic for illustrative purposes, in which case $r_\perp=r_\parallel=r_c\equiv r\equiv C'/C_c$. In Fig. 1, we compare our modified form factor $^{17}F_c$, Eq. (8), with the standard SMR form. It is seen that with a comparatively small amount of next-nearest-neighbor coupling, corresponding to $r\equiv C'/C_c=0.25$, the new form factor is reduced significantly near $(\pi/a,\pi/a)$, and is some 30% narrower near $\mathbf{q}=0$. This indicates that the oxygen $(^{17}T_1T)^{-1}$ is less likely to pick up the anomalous antiferromagnetic contribution near $(\pi/a,\pi/a)$, even when the anomalous spin fluctuation is slightly spread away from $(\pi/a,\pi/a)$.

We adopt the phenomenological MMP expression for the spin-spin correlation function, modified to take into account the presence of four incommensurate peaks at $\mathbf{Q}_i$ near $(\pi/a,\pi/a)$,[2]

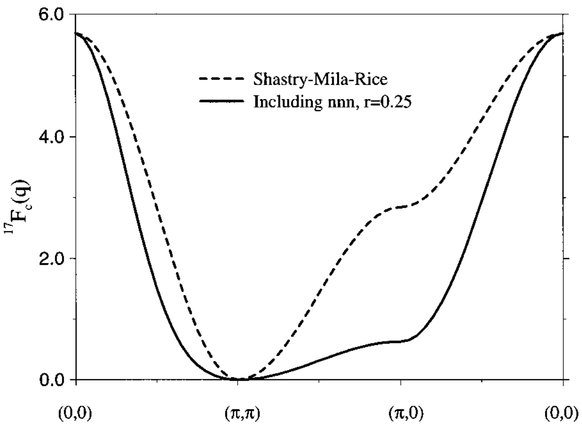


FIG. 1. Comparison of the modified form factor of $^{17}F_c$ in Eq. (8) with $r=0.25$ (solid line) with the standard Shastry-Mila-Rice form (dashed line). $^{17}F_c$ is plotted in units of $(C_c^{\rm old})^2$.

$$\chi(\mathbf{q},\omega)=\frac{1}{4}\sum_i \frac{\alpha\xi^2\mu_B^2}{1+(\mathbf{q}-\mathbf{Q}_i)^2\xi^2-i\omega/\omega_{\rm SF}}+\frac{\chi_0(T)}{1-i\pi\omega/\Gamma}. \tag{9}$$

Here the first term, often called $\chi_{\rm AF}$, represents the anomalous contribution to the spin spectrum, brought about by the close approach to antiferromagnetism of the Fermi liquid in the vicinity of the peaks at $\mathbf{q}=\mathbf{Q}_i$ determined by neutron-scattering experiments.[1,24] For $La_{1.86}Sr_{0.14}CuO_4$, $\mathbf{Q}_i=(\pi/a,[\pi\pm\delta]/a),([\pi\pm\delta]/a,\pi/a)$, with $\delta=0.245\pi$. In Eq. (9), $\omega_{\rm SF}$ is the characteristic frequency of the spin fluctuations, $\xi$ is the correlation length, and $\alpha$ is the scale factor (in units of states/eV, where $\mu_B$ is the Bohr magneton), which relates $\chi_{\mathbf{Q}_i}$ to $\xi^2$; thus the height of each of the four peaks is

$$\chi_{\mathbf{Q}_i}=\frac{\alpha}{4}\xi^2\mu_B^2. \tag{10}$$

The second term on the right-hand side of Eq. (9), usually called $\chi_{\rm FL}$, is a parameterized form of the normal Fermi-liquid contribution, which, apart from the usual Fermi-liquid pile up as $\mathbf{q}\to 0$, is found to be remarkably wave-vector independent over most of the Brillouin zone, according to the tight-binding model calculations which are expected to describe the cuprates; $\Gamma$ is of order the Fermi energy. The static bulk susceptibility $\chi_0$, which is generally temperature dependent, has been determined for $La_{2-x}Sr_xCuO_4$ and $YBa_2Cu_3O_{6+x}$ from copper and oxygen Knight-shift experiments.[2] For a system with any appreciable antiferromagnetic correlations ($\xi\gtrsim a$), the normal Fermi-liquid contribution is small compared to $\chi_{\rm AF}$ for wave vectors in the vicinity of $\mathbf{Q}_i$, and plays a negligible role in determining $(^{63}T_1T_c)^{-1}$; however, because of the filtering action of $^{17}F_\alpha$, it makes a significant contribution to $(^{17}T_1T)^{-1}$. Note that because the MMP expression for $\chi_{\rm AF}$ is a good approximation only for wave vectors in the vicinity of the antiferromagnetic wave vector $\mathbf{Q}_i$, it should not be used in calculating long-wavelength properties, such as the Knight shift of $^{17}$O, and should be cut off in calculations of $^{17}T_1$.

For the frequently encountered case of long correlation lengths ($\xi\gtrsim 2a$), $\Sigma_i(\pi/\xi^2)$ in calculating the various $^{63}$Cu relaxation rates one can approximate $\chi''(\mathbf{q},\omega)$ by $\chi''(\mathbf{Q}_i,\omega)\delta(\mathbf{q}-\mathbf{Q}_i)$. One can then replace Eqs. (3) and (5) by the following analytic expressions:

$$\frac{1}{^{63}T_{1\beta}T}\simeq\frac{k_B}{8\pi\hbar}{}^{63}F_\beta(\mathbf{Q}_i)\frac{\alpha}{\hbar\omega_{\rm SF}}, \tag{11}$$

$$(1/^{63}T_{2G})^2\simeq\frac{0.69}{512}\frac{^{63}F_{ab}^{\rm eff}(\mathbf{Q}_i)^2\alpha^2\xi^2}{\pi\hbar^2}. \tag{12}$$

Another important quantity, the anisotropy ratio of the $^{63}$Cu spin-lattice relaxation rates, which has been measured for $La_{2-x}Sr_xCuO_4$ and $YBa_2Cu_3O_{6+x}$ at various doping concentrations, provides a direct constraint on the hyperfine coupling constants, $A_\alpha$ and $B$. For $\xi\gtrsim 2$, this anisotropy ratio, $^{63}R$ can be written as

$$^{63}R\equiv\frac{T_{1c}}{T_{1ab}}\simeq\frac{^{63}F_{ab}(\mathbf{Q}_i)}{^{63}F_c(\mathbf{Q}_i)}. \tag{13}$$

For the case of $La_{2-x}Sr_xCuO_4$, where the peaks are located at $\mathbf{Q}_i=(\pi/a,[\pi\pm\delta]/a),([\pi\pm\delta]/a,\pi/a)$, we then have

$$^{63}R\simeq\frac{1}{2}\left[1+\frac{[A_c-2B(1+\cos\delta)]^2}{[A_{ab}-2B(1+\cos\delta)]^2}\right]. \tag{14}$$

For $YBa_2Cu_3O_{6+x}$, as indicated in the Introduction, on assuming the broad $(\pi/a,\pi/a)$ peak seen in neutron-scattering experiments[8–11] reflects the presence of four unresolved overlapping incommensurate peaks located along the zone diagonal directions,[9,11] we may write

$$\mathbf{Q}_i=([\pi\pm\delta]/a,[\pi\pm\delta]/a), \tag{15}$$

and the anisotropy ratio becomes

$$^{63}R\simeq\frac{1}{2}\left[1+\frac{(A_c-4B\cos\delta)^2}{(A_{ab}-4B\cos\delta)^2}\right]. \tag{16}$$

Numerical calculations of the $^{17}$O relaxation rates show that these rates can deviate significantly from those obtained by approximating the $\chi''_{\rm AF}$ by a $\delta(\mathbf{q}-\mathbf{Q}_i)$ function. We therefore calculate the $^{17}$O relaxation rates numerically, using Eqs. (3) and (4), and introducing a cutoff at $|\mathbf{Q}_i-\mathbf{q}|\sim\xi^{-1}$, since the MMP form is not expected to be valid for $(\mathbf{Q}_i-\mathbf{q})^2\xi^2\gtrsim 1$.

## III. THE DIRECT AND TRANSFERRED HYPERFINE CONSTANTS

Seven years of NMR experiments on aligned powders and single crystals of the cuprates have produced a significant number of constraints which must be taken into account in selecting the hyperfine constants which enter the SMR Hamiltonian. Thus experiments which determine the $^{63}$Cu nuclear resonance frequency in the AF insulators, $YBa_2Cu_3O_6$ (Ref. 25) and $La_2CuO_4$,[26] yield similar results for the product of $(4B-A_{ab})$ and $\mu_{\rm eff}$, the effective moment of the localized $Cu^{2+}$ spins,[27]

$$\mu_{\rm eff}(4B-A_{ab})=79.65\pm 0.05\ \text{kOe}\quad (YBa_2Cu_3O_6), \tag{17}$$

TABLE I. Spin-lattice anisotropy and incommensuration in the $La_{2-x}Sr_xCuO_4$ system.

| System | $\delta$ | $(^{63}R)_{\rm expt}$ | Ref. | $(^{63}R)_{\rm Eq.(22)}$ | $^{63}R_{\rm Eq.(14)}$ |
|---|---|---|---|---|---|
| $La_2CuO_4$ | 0 | 3.9±0.3 | 31 | 3.7 | 3.9 |
| $La_{1.9}Sr_{0.1}CuO_4$ | 0.175 | 3.5±? | 34 | 4.11 | 3.2 |
| $La_{1.85}Sr_{0.15}CuO_4$ | 0.263 | 3.0±0.20 | 32 | 4.78 | 3.2 |

$$\mu_{\rm eff}(4B-A_{ab})=78.78\ \ {\rm kOe}\quad ({\rm La_2CuO_4}). \tag{18}$$

On using the value, $\mu_{\rm eff}=0.62\mu_B$, determined by Manousakis[28] for the two-dimensional spin-1/2 Heisenberg antiferromagnet, we then find

$$4B-A_{ab}=128.5\ \ {\rm kOe}/\mu_B\quad ({\rm YBa_2Cu_3O_6}), \tag{19}$$

$$4B-A_{ab}=127\ \ {\rm kOe}/\mu_B\quad ({\rm La_2CuO_4}). \tag{20}$$

A second set of constraints comes from $^{63}$Cu Knight-shift experiments. To a high degree of accuracy, in the $YBa_2Cu_3O_{6+x}$ system the $^{63}$Cu Knight shift in a magnetic field along the $c$ axis is temperature independent in both the normal and superconducting state, and hence reflects only the chemical shift. The absence of a spin contribution means that for this system,

$$A_c+4B\simeq 0, \tag{21}$$

independent of doping level. A third set of constraints is obtained from measurements of the anisotropy of the $^{63}$Cu spin-lattice relaxation rates; for $YBa_2Cu_3O_7$ one finds $^{63}R=3.7\pm0.1$.[29] To the extent that $A_{ab}$, $A_c$, and $B$ are independent of doping level in $YBa_2Cu_3O_{6+x}$, and the spin-fluctuation peaks are commensurate (or nearly so) for this system, one then finds from Eqs. (16), (19), and (21), that

$$B=40.8\ \ {\rm kOe}/\mu_B,$$

$$A_c=-163\ \ {\rm kOe}/\mu_B,$$

$$A_{ab}=34\ \ {\rm kOe}/\mu_B, \tag{22}$$

in agreement with the analysis of Monien, Pines, and Takigawa.[7] These values are consistent with the constraint on $(4B+A_{ab})$ obtained by Ishida *et al.* for $La_{1.85}Sr_{0.15}CuO_4$; from the slope of a plot of their direct measurement of $\chi_0(T)$ against their measured value of $^{63}K_{ab}(T)$, they found[30]

$$4B+A_{ab}=189\ \ {\rm kOe}/\mu_B. \tag{23}$$

It seemed natural therefore to conclude that not only were $A_{ab}$, $A_c$, and $B$ independent of doping for the $YBa_2Cu_3O_{6+x}$ system, but that the corresponding values for the $La_{2-x}Sr_xCuO_4$ system were likewise doping independent and were virtually identical with those deduced for $YBa_2Cu_3O_{6+x}$.

If, however, the spin-fluctuation peaks in the $La_{2-x}Sr_xCuO_4$ system are incommensurate, the assumption that the hyperfine constraints for this system are doping independent is no longer tenable for this system, as may be seen by comparing the measured values of $^{63}R$ for the $La_{2-x}Sr_xCuO_4$ system shown in Table I with the values calculated using Eqs. (22), and using the doping dependence of the degree of incommensuration determined in neutron-scattering experiments,[12] $\delta\sim1.75x$, where $x$ is the Sr doping level. As may be seen in Table I, the calculated trend with doping is opposite to that seen experimentally. Since the quantum chemical environment responsible for the direct hyperfine interaction $A_\alpha$ is not expected to vary substantially with doping, the most likely culprit in Eqs. (22) is the assumption that the transferred hyperfine coupling constant does not vary appreciably with doping; indeed, if $B$ increases sufficiently rapidly with doping, with $A_{ab}$ and $A_c$ fixed, one can find a doping dependence of $^{63}R$ which is more nearly in accord with experiment. This means abandoning for the $La_{2-x}Sr_xCuO_4$ system the constraint, $A_c\simeq-4B$, which works so well for the $YBa_2Cu_3O_{6+x}$ system.

Suppose then one starts anew with the insulator, $La_2CuO_4$. On making use of Eqs. (14) and (20) and taking $^{63}R=3.9$, in accord with the result of Imai *et al.*[31] at 475 K, one finds readily that

$$A_{ab}-A_c=203\ \ {\rm kOe}/\mu_B. \tag{24}$$

On turning next to $La_{1.85}Sr_{0.15}CuO_4$, taking $^{63}R=3.0$, in accord with the recent measurement of Milling and Slichter,[32] using the result of Ishida *et al.*,[30] Eq. (23), and assuming that $A_{\alpha\beta}$ is independent of doping, one then finds $B=48$ kOe/$\mu_B$ and $A_{ab}=-3\ \ kOe/\mu_B$. This result is, however, unrealistic. A straightforward calculation using the expressions adapted by Monien *et al.*[27] from the work of Bleaney *et al.*,[33]

$$A_c=395\left[-\hat\kappa-\frac{4}{7}-\frac{62}{7}\gamma\right]\ \ {\rm kOe}/\mu_B,$$

$$A_{ab}=395\left[-\hat\kappa+\frac{2}{7}-\frac{11}{7}\gamma\right]\ \ {\rm kOe}/\mu_B. \tag{25}$$

In Eqs. (25), $\gamma\equiv\lambda/E_{xy}$ is the dimensionless ratio of the spin-orbit coupling for a $Cu^{2+}$ ion, $\lambda\sim-710\ {\rm cm}^{-1}$, to the excitation energy from the ground state of the $^{63}$Cu $d_{x^2-y^2}$ orbital of the various $^{63}$Cu $d$ states, $E_{xy}\sim E_{xz}\sim E_{yz}\sim2$ eV; with these typical values, $\gamma=-0.044\pm0.009$; $\langle 1/r^3\rangle$ which enters as a multiplicative factor in Eq. (25) is taken to be $6.3a_0^{-3}$. With the value of $\gamma=-0.0471$ obtained using Eq. (24),

$$A_{ab}=(-395\hat\kappa+142)\ \ {\rm kOe}/\mu_B. \tag{26}$$

On taking the core polarization $\hat\kappa=0.26\pm0.06$,[27] we then get, for $\hat\kappa$ in the vicinity of its plausible upper limit, 0.32,

$$A_{ab}\geqslant 16\ \ {\rm kOe}/\mu_B. \tag{27}$$

In order to satisfy the above constraints, we next assume that the anisotropy, $^{63}R$, for $La_{1.85}Sr_{0.15}CuO_4$ is at the upper end of the range quoted by Milling and Slichter, and take $^{63}R=3.2$; we next take $A_{ab}=18$ kOe/$\mu_B$ (corresponding to $\hat\kappa=0.316$), a value close, but not at, the estimated minimum value for $A_{ab}$. We then have, from Eq. (24), $A_c=-185$ kOe/$\mu_B$ and, from Eq. (14) for $^{63}R$, $B_{0.15}=51$ kOe/$\mu_B$, while for the insulator, we find from Eq. (20), $B_0=36.1$ kOe/$\mu_B$. With these hyperfine constants we find for $La_{1.85}Sr_{0.15}CuO_4$ that $4B+A_{ab}=222$ kOe/$\mu_B$, some 17% above the value obtained by Ishida *et al.*,[30] while for this

TABLE II. Parameters for $La_{2-x}Sr_xCuO_4$.

| | $La_2CuO_4$ | $La_{1.90}Sr_{0.10}CuO_4$ | $La_{1.86}Sr_{0.14}CuO_4$ | $La_{1.85}Sr_{0.15}CuO_4$ |
|---|---|---|---|---|
| $A_c$ (kOe/$\mu_B$) | -185 | -185 | -185 | -185 |
| $A_{ab}$ (kOe/$\mu_B$) | 18 | 18 | 18 | 18 |
| B (kOe/$\mu_B$) | 36.1 | 46 | 50 | 51 |
| $C_c$ (kOe/$\mu_B$) | 33 | 33 | 33 | 33 |
| $^{63}R_{\rm exp}$ | $3.9\pm0.3$ | $3.5\pm?$ | | $3.0\pm0.2$ |
| $^{63}R_{\rm cal}$ | 3.9 | 3.2 | 3.2 | 3.2 |
| 4B-$A_{ab}$ (kOe/$\mu_B$) | 127 | 166 | 182 | 186 |
| 4B+$A_{ab}$ (kOe/$\mu_B$) | 162.4 | 202 | 218 | 222 |
| $^{63}K_c/^{63}K_{ab}$ | -25% | -0.5% | 7% | 8.6% |
| $^{63}T_1T$ | 138 (s K/eV$^2$)$\omega_{\rm SF}/\alpha$ | 95.2 (s K/eV$^2$)$\omega_{SF}/\alpha$ | 93.5 (s K/eV$^2$)$\omega_{\rm SF}/\alpha$ | 94.2 (s K/eV$^2$)$\omega_{\rm SF}/\alpha$ |
| $1/T_{2G}$ | 298 (eV/s)$\alpha\xi$ | 347 (eV/s)$\alpha\xi$ | 350 (eV/s)$\alpha\xi$ | 348 (eV/s)$\alpha\xi$ |
| $^{63}T_1T/T_{2G}$ | $4.12\times10^4$ (K/eV)$\omega_{\rm SF}\xi$ | $3.30\times10^4$ (K/eV)$\omega_{\rm SF}\xi$ | $3.27\times10^4$ (K/eV)$\omega_{\rm SF}\xi$ | $3.28\times10^4$ (K/eV)$\omega_{\rm SF}\xi$ |
| $^{63}T_1T/T_{2G}^2$ | $1.23\times10^7$ (K/s)$\alpha\omega_{\rm SF}\xi^2$ | $1.15\times10^7$ (K/s)$\alpha\omega_{\rm SF}\xi^2$ | $1.14\times10^7$ (K/s)$\alpha\omega_{\rm SF}\xi^2$ | $1.14\times10^7$ (K/s)$\alpha\omega_{\rm SF}\xi^2$ |
| $\Gamma$ (meV) | | | | 565 |
| $r$ | | | | 0.25 |
| $\delta$ | | 0.175 | 0.245 | 0.263 |

system, the ratio of the spin contributions to the Knight shift for fields parallel and perpendicular to the $c$ axis is

$$\frac{^{63}K_c}{^{63}K_{ab}}=\frac{4B+A_c}{4B+A_{ab}}=8.6\%. \qquad (28)$$

The slight temperature variation of $^{63}K_c$ which follows from this choice of parameters would not be detectable, consistent with the measurements of Ohsugi *et al.*[34]

For intermediate levels of Sr doping, if we assume that the change in $B$ induced by doping scales with the doping level, we obtain the results for $La_{1.9}Sr_{0.1}CuO_4$ and $La_{1.86}Sr_{0.14}CuO_4$ given in Table II. Also given there are the corresponding results for $^{63}T_1$ and $^{63}T_{2G}$ and related quantities of interest in analyzing NMR experiments. We note that to obtain $^{63}R=3.5$ for $La_{1.9}Sr_{0.1}CuO_4$, one needs a transferred hyperfine coupling, $B=37.8$ kOe/$\mu_B$, which is considerably lower than that obtained by direct interpolation.

We turn next to the $YBa_2Cu_3O_{6+x}$ system. For $YBa_2Cu_3O_6$, the only constraint on the hyperfine constants is the AF resonance result, Eq. (19). However, as noted above, for $YBa_2Cu_3O_7$ one has two further constraints: $4B=A_c$, and $^{63}R=3.7\pm0.1$.[29] Moreover, as is the case for $YBa_2Cu_3O_{6.63}$, neutron-scattering experiments on $YBa_2Cu_3O_7$ suggest that one has four incommensurate and largely unresolved peaks along the zone diagonal direction whose positions, $\mathbf{Q}_i$, are given by Eq. (15). On taking $\delta=0.1$, a value consistent with the experimental results of Dai *et al.*[9] we then find, on making use of Eq. (16), that

$$A_{ab}=0.721B. \qquad (29)$$

If now we assume that the spin orbit coupling of a $Cu^{2+}$ ion in $YBa_2Cu_3O_7$ is little changed from that found for $La_2CuO_4$, $\gamma=0.471$, we have a third relation between the coupling constants,

$$A_{ab}-A_c=4.721B=203 \text{ kOe}/\mu_B \qquad (30)$$

from which we find

$$B=43 \text{ kOe}/\mu_B,$$

$$A_{ab}=31 \text{ kOe}/\mu_B,$$

$$A_c=-172 \text{ kOe}/\mu_B, \qquad (31)$$

while from the AF resonance constraint, Eq. (19), we find for the insulator $YBa_2Cu_3O_6$, that $B=39.8$ kOe/$\mu_B$.

Confirmation of this choice of parameters comes by determining the slope from the linear temperature dependence found in a plot of $^{63}K_{ab}$ versus $\chi_0(T)$ for $O_{6.63}$. We find $4B+A_{ab}\sim200$ kOe/$\mu_B$, in agreement with Eq. (30). Moreover, Shimizu *et al.*[35] find, from a similar plot for $YBa_2Cu_3O_{6.48}$, that for this system, $4B+A_{ab}\simeq200$ kOe/$\mu_B$.

We adopt these values in our subsequent calculations. We note that the value of $B$ we obtain for $YBa_2Cu_3O_6$ is some 10% larger than that found for $La_2CuO_4$, while the doping dependence of $B$ is considerably smaller in the $YBa_2Cu_3O_{6+x}$ system than in the $La_{2-x}Sr_xCuO_4$ system. Both effects may plausibly be attributed to the presence of chains in the $YBa_2Cu_3O_{6+x}$ system. The core polarization parameter, $\hat{\kappa}=0.281$ we find for the $YBa_2Cu_3O_{6+x}$ system is some 10% smaller than that inferred for the $La_{2-x}Sr_xCuO_4$ system. We tabulate in Table III our results for the $YBa_2Cu_3O_{6+x}$ system at three doping levels; we estimate $B=40.6$ kOe/$\mu_B$ for $YBa_2Cu_3O_{6.63}$ by interpolating between an assumed value, $B=39.8$ for $YBa_2Cu_3O_{6.5}$, and that we found above for $YBa_2Cu_3O_7$.

## IV. RECONCILING NEUTRON SCATTERING AND $^{63}$Cu NMR MEASUREMENTS IN $La_{2-x}Sr_xCuO_4$

We now explore whether, with the revised hyperfine constants proposed above, we can reconcile the recent neutron-scattering results of Aeppli *et al.*[1] for $La_{1.86}Sr_{0.14}CuO_4$ with the NMR measurements of Ohsugi *et al.*[34] on the two adjacent systems, $La_{1.87}Sr_{0.13}CuO_4$, and $La_{1.85}Sr_{0.15}CuO_4$. We as-

TABLE III. Parameters for $YBa_2Cu_3O_{6+x}$.

| | $YBa_2Cu_3O_6$ | $YBa_2Cu_3O_{6.63}$ | $YBa_2Cu_3O_7$ |
|---|---|---|---|
| $A_c$ (kOe/$\mu_B$) | -172 | -172 | -172 |
| $A_{ab}$ (kOe/$\mu_B$) | 31 | 31 | 312 |
| $B$ (kOe/$\mu_B$) | 39.8 | 40.6 | 43 |
| $C_c$ (kOe/$\mu_B$) | 33 | 33 | 33 |
| $^{63}R_{\rm exp}$ | | | 3.7±0.1 |
| $^{63}R_{\rm cal}$ | 3.8 | 4.0 | 3.7 |
| $4B-A_{ab}$ (kOe/$\mu_B$) | 128.5 | 131.4 | 141 |
| $4B+A_{ab}$ (kOe/$\mu_B$) | 190 | 193 | 203 |
| $^{63}K_c/^{63}K_{ab}$ | −7% | −5% | 0 |
| $^{63}T_1T$ | 135 (s K/eV$^2$)$\omega_{\rm SF}/\alpha$ | 145 (s K/eV$^2$)$\omega_{\rm SF}/\alpha$ | 126 (s K/eV$^2$)$\omega_{\rm SF}/\alpha$ |
| $1/T_{2G}$ | 301 (eV/s)$\alpha\ \xi$ | 293 (eV/s)$\alpha\ \xi$ | 310 (eV/s)$\alpha\xi$ |
| $^{63}T_1T/T_{2G}$ | $4.06\times10^4$ (K/eV)$\omega_{\rm SF}\ \xi$ | $4.25\times10^4$ (K/eV)$\omega_{\rm SF}\xi$ | $3.9\times10^4$ (K/eV)$\omega_{\rm SF}\xi$ |
| $^{63}T_1T/T_{2G}^2$ | $1.22\times10^7$ (K/s)$\alpha\omega_{\rm SF}\xi^2$ | $1.25\times10^7$ (K/s)$\alpha\omega_{\rm SF}\xi^2$ | $1.21\times10^7$ (K/s)$\alpha\omega_{\rm SF}\xi^2$ |
| $\alpha$ | | 8.34 | 14.8 |
| $\Gamma$ (meV) | | 226 | 308 |
| $r$ | | 0.25 | 0.25 |
| $\delta$ | | 0.1 | 0.1 |

sume that $\chi(q,\omega)$ takes the MMP form, Eq. (9), in which case

$$\chi''(q,\omega)=\sum_i \frac{\chi_{Q_i}(\omega/\omega_{\rm SF})}{[1+(\mathbf{Q}_i-\mathbf{q})^2\xi^2]^2+(\omega/\omega_{\rm SF})^2}, \qquad (32)$$

where $\chi_{Q_i}$ is given by Eq. (10). There are three undetermined parameters; $\alpha$, $\xi$, and $\omega_{\rm SF}$. We begin by deducing $\chi_{Q_i}$ and $\omega_{\rm SF}$ from the results of Aeppli *et al.* for $\chi''(Q_i,\omega)$ at 35 K; as may be seen in Fig. 2, a good fit to their results is found with $\chi_{Q_i}(35\ {\rm K})=350$ states/eV and $\omega_{\rm SF}=8.75$ meV. To determine $\alpha$, and hence $\xi(35\ {\rm K})$, we turn to the NMR results of Ohsugi *et al.*;[34] on interpolating between their results for the adjacent systems, as shown in Fig. 3, we find $^{63}T_1T=34(10^{-3}$ s K), while according to Table II, one has

$$^{63}T_1T=93.5(\omega_{\rm SF}/\alpha)\ \ {\rm s\,K/(eV)^2}. \qquad (33)$$

Equating these results, we obtain $\alpha=23.9$ states/eV and $\xi=7.6$.

A first check then on our use of Eq. (32) to fit both NMR and neutron-scattering results is to compare this value of $\xi$ with the measurements of the intrinsic linewidth of each peak by Aeppli *et al.*[1] We find on converting units, that at 35 K the linewidth parameter of Aeppli *et al.* corresponds to a correlation length, $\xi=7.7$ in the low- ($\omega=0$ meV) frequency limit. The agreement is quite good.

Having determined $\alpha$, we can then use our interpolated NMR results to obtain $\omega_{\rm SF}(T)$ for 35 K$\leqslant T\leqslant$300 K from Eq. (33). That leaves only one parameter, $\chi_{Q_i}$ (or $\xi$) to be determined over this temperature range. As a first step toward its determination, we use the results of Aeppli *et al.* for $\chi''(Q_i,\omega)$ at 80 K. As shown in Fig. 2, a good fit to the experimental data is obtained with $\chi_{Q_i}(80\ {\rm K})=175$ states/eV. From Eq. (10), we then get $\xi(80\ {\rm K})=5.41$.

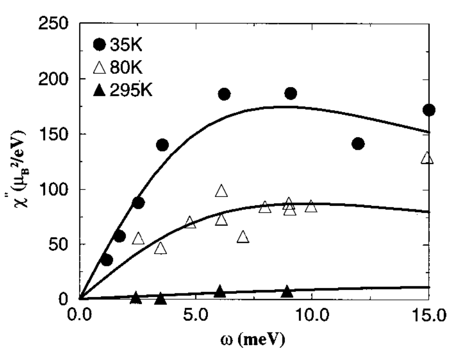


FIG. 2. The frequency dependence of $\chi''(\mathbf{Q}_i,\omega)$ at three temperatures. The experimental points are the results obtained by Aeppli *et al.* (Ref. 1); the solid curves are the fits obtained using a mean-field description of $\chi''(\mathbf{q},\omega)$ shown in Eq. (32) and parameters compatible with NMR results.

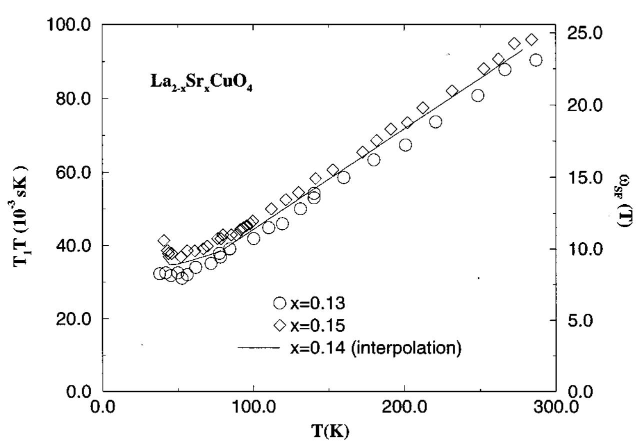


FIG. 3. The interpolated $^{63}T_{1c}T$ for $La_{1.86}Sr_{0.14}CuO_4$ is shown together with the measured values of $^{63}T_{1c}T$ for $La_{1.87}Sr_{0.13}CuO_4$ and $La_{1.85}Sr_{0.15}CuO_4$ of Ohsugi *et al.* (Ref. 34). Shown on the right-hand side is the scale for $\omega_{\rm SF}(T)\propto{}^{63}T_{1c}T$ for $La_{1.86}Sr_{0.14}CuO_4$ inferred from the fit to the neutron-scattering experiments.

We next make use of the Barzykin-Pines magnetic phase diagram. From their analysis of NMR, transport and static susceptibility experiments, they conclude that the $La_{2-x}Sr_xCuO_4$ system will, like its $YBa_2Cu_3O_{6+x}$ counterpart, exhibit nonuniversal scaling behavior, perhaps best described as pseudoscaling, between two crossover temperatures, $T^*$ and $T_{cr}$. In this regime, the system exhibits apparent $z=1$ dynamic scaling behavior, with $\omega_{SF}$ varying linearly with temperature and

$$\omega_{SF}=c'/\xi \tag{34}$$

where $c'$ depends on the doping level. They propose that the upper crossover temperature, $T_{cr}$, which marks the onset of pseudoscaling behavior, can be identified as the maximum in the measured value of $\chi_0(T)$, and corresponds to a magnetic correlation length, $\xi\sim 2$. The lower temperature $T^*$ is determined from $^{63}T_1$ measurements as the lower limit of the linear variation of $\omega_{SF}$ (or $^{63}T_1T$) with temperature. Inspection of Fig. 3 shows that for $La_{1.86}Sr_{0.14}CuO_4$, one has a comparatively weak crossover at $T^*\sim 80$ K. Since $\omega_{SF}(T)$ has already been determined, a knowledge of $c'$, obtained at one temperature between $T^*$ and $T_{cr}$, enables one to fix $\xi(T)$ over the entire temperature range. From our fit to the neutron data at 80 K, we find $c'=52.9$ meV, and use this result to conclude that $T_{cr}\sim 325$ K, and that

$$\frac{1}{\xi}=0.0828+0.128\left(\frac{T}{100}\right) \quad 80\ \mathrm{K}<T<325\ \mathrm{K}. \tag{35}$$

We can interpolate between this result for $\xi(T)$ and our result at 35 K to obtain $\xi(T)$ over the region, 35 K$\leqslant T \leqslant$300 K. The result of that interpolation, which is very nearly a continuation of the linear behavior found above 80 K, is given in the inset of Fig. 4.

A first check on the correctness of this procedure is to compare our ''NMR'' derived results at 295 K, shown in Table IV, with the neutron-scattering results at this temperature. As may be seen in Fig. 2, the slope, obtained from the NMR results, $[\chi''(Q,\omega)/\omega]=\chi_{Q_i}/\omega_{SF}=1.16\mu_B^2/(\mathrm{eV\ meV})$ is in good agreement with experiment. A second check is to compare our results for $\xi(T)$ with the values deduced from the half width of the incommensurate peaks in $\mathrm{Im}\chi(\mathbf{q},\omega)$ observed in neutron scattering over the entire temperature domain (35$\leqslant T \leqslant$300 K); that comparison is given in the main portion of Fig. 4. Finally, we can compare the predictions of Eqs. (34) and (35) (the parameters being specified in Table IV) with the combined frequency and temperature dependence of the half width found by Aeppli *et al.* in Fig. 5. In obtaining this figure, we calculated the theoretical inverse correlation length $\kappa$ from the $\xi(T)$ shown in Fig. 4, by matching the full width at half maximum of the incommensurate peaks of Eq. (32) to those of the experiments of Ref. 1. Our comparison of the calculated $\kappa(\omega,T)$ to the experimental values is shown in Fig. 5. The extent of the agreement between our calculations and experiment suggests that we have succeeded in reconciling the $^{63}$Cu NMR results with the neutron-scattering results, and it suggests as well that the neutron scattering results are consistent with $z=1$ pseudoscaling behavior for temperatures less than 300 K. The latter conclusion was also reached by Aeppli *et al.* from their analysis of their neutron-scattering experiments. Moreover, the agreement at 300 K provides independent support for the Barzykin-Pines proposal that $\xi(T_{cr})=\xi(325\ \mathrm{K})\simeq 2$.

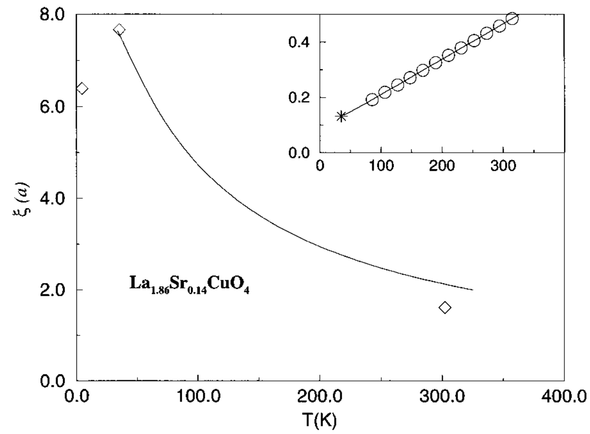


FIG. 4. A comparison of the NMR-deduced values of $\xi(T)$ (solid line) for $La_{1.86}Sr_{0.14}CuO_4$ with those obtained (diamonds) from the neutron-scattering experiments of Ref. 1. The experimental points (diamonds) are derived by first fitting the half-width of the neutron scattering peak at $\mathbf{Q}_i$ for low-energy transfer ($\omega$=2.5 meV), and then extrapolating to $\omega$=0, following the formula $\kappa^2(\omega,T)=\kappa^2(T)+a^{-2}\omega^2/E_\omega^2$ given in Ref. 1. The inset shows the interpolation procedure used to obtain $1/\xi(T)$ between 35 and 80 K: the point at 35 K (star) is obtained from the MMP fit to the neutron scattering data at 35 K, while the points above 80 K are deduced from the scaling analysis of Eq. (35) (circles); the solid line shows the extrapolation between 35 and 80 K.

## V. $^{17}$O RELAXATION RATES FOR $La_{1.85}Sr_{0.15}CuO_4$

We now demonstrate that by choosing a reasonable next-nearest-neighbor hyperfine coupling contribution $C'$, we can reconcile the incommensurate peaks in $\chi''(\mathbf{q},\omega)$ with the measured NMR relaxation rates $(^{17}T_1T)^{-1}$ for $La_{1.85}Sr_{0.15}CuO_4$.

In calculating $(^{17}T_{1c}T)^{-1}$ for $La_{1.85}Sr_{0.15}CuO_4$, we simply use the previously determined parameters as inputs to Eqs. (4) and (9), where the next-nearest-neighbor Cu-oxygen hyperfine coupling $C'$ is included in the form factor $^{17}F_c$. For $La_{2-x}Sr_xCuO_4$ materials, there is still not enough experimental data to determine the exact values of $C_\alpha^{\mathrm{old}}$ for different field orientations; we therefore assume that these values are the same as those of the $YBa_2Cu_3O_{6+x}$ family. Following Monien *et al.*[7] and Yoshinari *et al.*,[21] we take $C_c^{\mathrm{old}}=33$ kOe

TABLE IV. Fits to neutron scattering experiments of $La_{1.86}Sr_{0.14}CuO_4$.

| | $T$=35 K | $T$=80 K | $T$=295 K |
|---|---|---|---|
| $^{63}T_1T$ ($\times 10^{-3}$ s K) | 34 | 38 | 96 |
| $\omega_{SF}$ (meV) | 8.75 | 9.78 | 24.3 |
| $\alpha$ (states/eV) | 23.9 | 23.9 | 23.9 |
| $\chi_{\mathbf{Q}}$($\mu_{sB}^2$/eV) | 350 | 175 | 28.2 |
| $\xi$ (a) | 7.6 | 5.41 | 2.17 |
| $c'$ | | 52.9 | |
| $1/T_{2G}$ (m s$^{-1}$) | 63 | 45 | 18 |

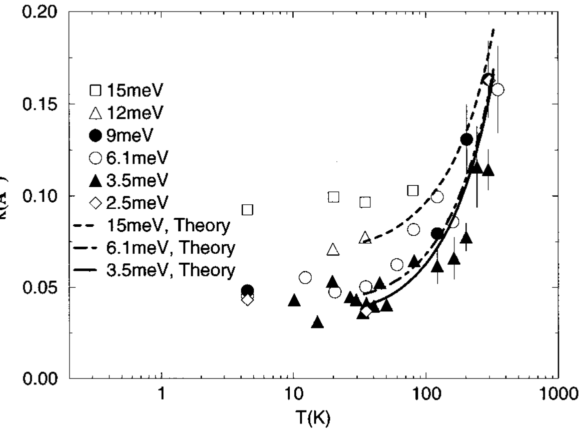


FIG. 5. A comparison of the NMR-deduced values of the frequency dependence inverse correlations length, $\kappa$, at $\omega$ =3.5, 6.1, and 15 meV (lines), with the experimental results of Aeppli *et al.* (Ref. 1) (symbols). It is seen that the consistency is quite good at low frequencies, while at high frequencies, the NMR-deduced **q** width is smaller than those seen in neutron-scattering experiments.

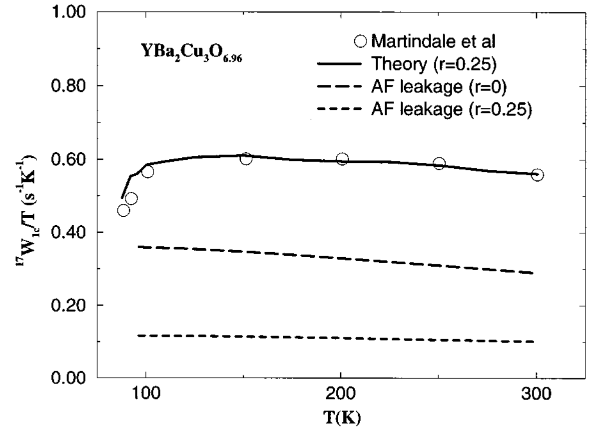


FIG. 7. The $^{17}$O spin-lattice relaxation rate $^{17}W_{1c}/T$ calculated by assuming $r=0.25$ (solid line), plotted against the experimental data of Martindale *et al.* (Ref. 3) (circles) for $YBa_2Cu_3O_{6.6}$. Also shown is the contribution from AF leakage to the relaxation rates $^{17}W_{1c}/T$ calculated using our oxygen form factor with $r=0.25$ and the standard Shastry-Mila-Rice form factor $(r=0)$.

and $\zeta_{\parallel}=1.42$, $\zeta_c=1$, and $\zeta_{\perp}=0.91$. We further assume an isotropic $C'$, with $r=C'/C_c=0.25$, and obtain $\chi_0(T)$ by modifying the results of Ref. 2 to reflect the new values of $A_{ab}$ and $B$ presented in Sec. III. We use the $\alpha$ for $La_{1.86}Sr_{0.14}CuO_4$ obtained from the neutron-scattering fits from the last section, and obtain $\omega_{SF}$, and $\xi(T)$ from NMR data of Ohsugi *et al.*[34] These numbers are almost the same as those of $La_{1.86}Sr_{0.14}CuO_4$. The remaining parameter in Eq. (9) which is sensitive to our choice of a cutoff in the applicability of the MMP expression for $\chi_{AF}$, $\Gamma$ is chosen to get the best fit to the experimental results for $(^{17}T_{1c}T)^{-1}$. It is important to point out that our choice of $\Gamma$ does not affect $(^{63}T_{1c}T)^{-1}$ and $1/^{63}T_{2G}$, because the Fermi-liquid contribution to these quantities is negligible compared to that of the anomalous antiferromagnetic spin fluctuations.

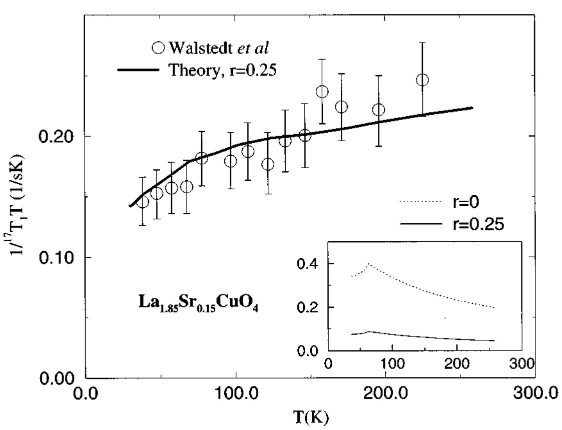


FIG. 6. Our calculated spin-lattice relaxation rates $(^{17}T_{1c}T)^{-1}$ for $^{17}$O (solid line) compared to the experimental data of Walstedt *et al.* (Ref. 18) (circles) for $La_{1.85}Sr_{0.15}CuO_4$; we use $r=0.25$ for the calculation. The inset shows the contribution of the AF spin fluctuations [first term only in Eq. (9) with a cutoff of $|\mathbf{q}-\mathbf{Q}_i|=1/\xi$] to $(^{17}T_{1c}T)^{-1}$, calculated with the present form factor ($r$=0.25) and with the standard Shastry-Mila-Rice form factor ($r$=0).

In Fig. 6 we compare our calculated $^{17}$O NMR relaxation rate $(^{17}T_{1c}T)^{-1}$, using $\Gamma=565$ meV, with the experimental data of Walstedt *et al.*[18] The agreement is quite good. Note, however, the choice of $r$ and $\Gamma$ is not unique in our calculations; fits of the same quality can be obtained by choosing other values for $r$ and $\Gamma$. The inset of Fig. 6 shows the substantial leakage of the anomalous spin fluctuations [the first term only in Eq. (9)] to $(^{17}T_{1c}T)^{-1}$, calculated with the standard SMR form factor $(r=0)$. The $(^{17}T_{1c}T)^{-1}$ thus calculated has a temperature dependence similar to that of the $^{63}$Cu relaxation rates, much faster than seen experimentally. Also shown in the inset is the substantially smaller AF leakage calculated from the present hyperfine coupling $^{17}F_c$ with $r=0.25$.

## VI. NEUTRON-SCATTERING LINE WIDTHS AND $^{17}$O SPIN-LATTICE RELAXATION RATES IN $YBa_2Cu_3O_{6+x}$

We turn now to the neutron-scattering and NMR experiments for the $YBa_2Cu_3O_{6+x}$ system. As noted in the introduction, one apparent problem here has been that the large **q** width of the antiferromagnetic peak, as observed in the neutron-scattering experiments,[8–11] appeared to be in contradiction with size of the correlation length ($\xi \gtrsim 2$) required to explain the $^{17}$O NMR experiments. As Thelen and Pines demonstrated,[20] the half width at half maximum for the antiferromagnetic peak in $\chi''(\mathbf{q},\omega)$ should have been $q_{1/2} \lesssim 0.4/a$ in order to be consistent with the Mila-Rice-Shastry model and the oxygen relaxation data for $YBa_2Cu_3O_7$. They found that in order to be consistent with experiment the leakage from the antiferromagnetic peak should account for no more than 1/3 of the total measured oxygen rate. This upper bound from NMR is much smaller than the actual **q** width of the antiferromagnetic peak, $q_{1/2} \simeq 0.7/a$, observed in the neutron-scattering experiments.[9] Assuming the measured width is produced by incommensuration, we plot the antiferromagnetic ‘‘leakage’’ contribution [i.e., that from the antiferromagnetic part of Eq. (9)] to the

$^{17}$O relaxation rate in Fig. 7, using the incommensuration $\delta = 0.1\pi$, which provides a fit to the neutron-scattering experiments.[9] Obviously, as in the $La_{2-x}Sr_xCuO_4$ material, the temperature dependence of the measured NMR relaxation rate is remarkably different, and the amplitude of the "leakage" term is too large. This problem can be avoided by introducing $C'$, as we have done on the $La_{2-x}Sr_xCuO_4$ system. In fact, the much smaller degree of presumed incommensurability in the $YBa_2Cu_3O_{6+x}$ system than that measured directly for the $La_{2-x}Sr_xCuO_4$ system makes it almost evident that any problem produced by AF leakage can be reconciled by the same method as used above. We show, in Fig. 7, that the AF leakage contribution for $r = C'/C_c = 0.25$ indeed becomes negligible. If we assume the same ratio of the AF part to the total rate as Thelen and Pines[20] did, we obtain a constraint on $C'$. We note that the oxygen form factors Eq. (8) are quadratic in $\delta\mathbf{q} = (\mathbf{q} - \mathbf{Q})$ in the vicinity of the antiferromagnetic wave vector $\mathbf{Q}$:

$$^{17}F_{\alpha} = (\delta q_x)^2 \frac{(C_c^{\mathrm{old}})^2}{2(1+2r_c)^2} \sum_{\alpha_i = \alpha', \alpha''} [(2r_c+1)\zeta_{\alpha_i} - 4r_{\alpha_i}]^2$$
$$= \eta(\delta q_x)^2. \tag{36}$$

As a result, the antiferromagnetic contribution to the oxygen relaxation rate (which we keep as a constant when we change the form factor) is

$$\left(\frac{1}{^{17}T_1 T}\right)_{\mathrm{AF}} \propto \eta(\mathbf{Q_i} - \mathbf{Q})^2, \tag{37}$$

and a change of the oxygen form factor, which alters $\eta$, produces a constraint on the acceptable width (or incommensurability) of the neutron-scattering peak. Since Thelen and Pines[20] used the isotropic form of the Mila-Rice-Shastry Hamiltonian, with $C_{\mathrm{iso}} = C_c^{\mathrm{old}}$ we easily obtain from Eq. (36):

$$q_{1/2\alpha} \lesssim 0.4/a \frac{1+2r_c}{\sqrt{\frac{1}{2}\sum_{\alpha_i=\alpha',\alpha''}[(2r_c+1)\zeta_{\alpha_i} - 4r_{\alpha_i}]^2}}, \tag{38}$$

where we have neglected possible slow (logarithmic) dependence. In particular, with $r_{\alpha_i} = 0.25$, Eq. (38) gives the upper limit: $q_{1/2} \lesssim 0.7/a$. This crude estimate shows that indeed, our hyperfine Hamiltonian is consistent with both NMR and neutron-scattering experiments. However, the antiferromagnetic leakage contribution to the oxygen relaxation rate in $YBa_2Cu_3O_{6+x}$ can become important, and should therefore be calculated numerically, since the spin-spin correlation length is very short.

For our numerical calculation of the antiferromagnetic peak contribution to the $^{17}$O relaxation rates we assume, as indicated in the Introduction, that the neutron-scattering data of Tranquada *et al.*[11] and Dai *et al.*[9] can be interpreted as indicating that the magnetic response function $\chi(\mathbf{q},\omega)$ possesses four incommensurate peaks located at $\mathbf{Q}_i = (\pi \pm \delta, \pi \pm \delta)$, and take $\delta \simeq 0.1\pi$, an incommensuration consistent with the measured experimental widths. We also assume that the temperature-dependent spectral weight for these incommensurate peaks, as in case of $La_{2-x}Sr_xCuO_4$, comes from the temperature dependence of the correlation length, and adopt the MMP form Eq. (9) for each of the four peaks. It should be emphasized, however, that accord between the inelastic neutron scattering and the oxygen NMR can be reached for any bell-shaped curve for $\chi''(\mathbf{q},\omega)$ which has the characteristic width measured in the neutron-scattering experiments, and a sufficiently abrupt falloff at large $(\mathbf{q} - \mathbf{Q})$. In Fig. 7, we show our calculated antiferromagnetic leakage to the oxygen relaxation $^{17}W_{1c}/T \equiv 1.5(^{17}T_{1c}T)^{-1}$, for the case of both $r=0$ and $r=0.25$; again, we see that the form factor with $r=0.25$ greatly reduces the AF leakage. Also shown in Fig. 7 is our calculated $^{17}W_{1c}/T$ plotted against the experimental data of Martindale *et al.*[3] In obtaining our theoretical result, we have used as an input to Eq. (9), $\chi_0(T)$ deduced from the Knight-shift $K_c(T)$ data on the same sample, provided by Martindale *et al.*[22] and used the $\xi$ and $\alpha$ from Ref. 2. Again, we take $C_c^{\mathrm{old}} = 33$ kOe/$\mu_B$ for $YBa_2Cu_3O_{6+x}$ system. By assuming $r_{\alpha_i} = 0.25$, we obtain a good fit to the experimental data with $\Gamma \sim 308$ meV. These parameters are listed in Table III.

Another problem with the one-component Shastry-Mila-Rice picture has been pointed out recently by Martindale *et al.*,[3] who measured planar $^{17}$O relaxation rates for different magnetic-field directions. They have found that the *temperature dependences* of the relaxation rates for magnetic fields parallel and perpendicular to the Cu-O bond axis directions were different, in contradiction with the predictions based on the SMR hyperfine Hamiltonian for which the oxygen form factor is given by Eq. (4), without $C'$:

$$^{17}F_{\alpha} = 2 \sum_{\alpha_i = \alpha', \alpha''} \cos^2 \frac{q_x a}{2} C_{\alpha_i}^2. \tag{39}$$

From Eq. (39) it follows that the ratios of the oxygen relaxation rates for different magnetic-field orientations should be temperature independent, and determined only by the hyperfine $C$ couplings

$$\frac{^{17}(1/T_{1,\alpha_i})}{^{17}(1/T_{1,\alpha_j})} = \frac{C_{\alpha_i'}^2 + C_{\alpha_i''}^2}{C_{\alpha_j'}^2 + C_{\alpha_j''}^2}. \tag{40}$$

Experimentally, as shown by Martindale *et al.*[3] these ratios turn out to be mildly temperature dependent, although numerically close to the values of Eq. (40).

This apparent contradiction can, in fact, be turned into a proof of the validity of the modified one-component model Eq. (1). It can easily be seen that for our oxygen form factors, Eq. (8), the $^{17}$O relaxation rates for different field directions do not have the same $\mathbf{q}$ dependence for the whole Brillouin zone. As a result, ratios such as Eq. (40) should indeed become temperature dependent. Since we do not know the precise values of the couplings once we go beyond the nearest-neighbor Mila-Rice-Shastry approximation, we use here the expressions for the oxygen form-factors in the most general form. To derive the form of the temperature dependence, we separate the antiferromagnetic and the Fermi-liquid or short-wavelength ($\chi_0$) contributions to $(1/^{17}T_{1\alpha})$, according to Eq. (9):

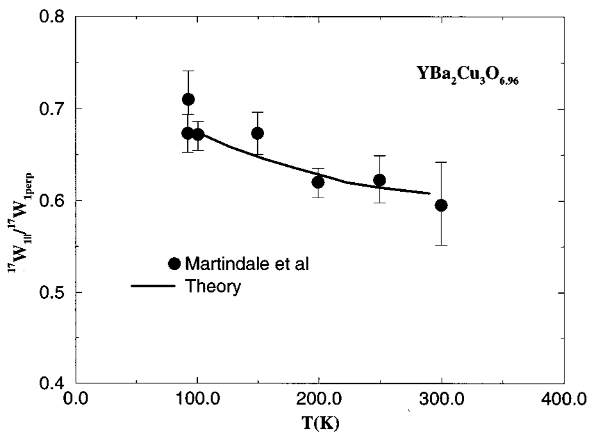


FIG. 8. The temperature dependence of the oxygen relaxation rate ratios in $YBa_2Cu_3O_{6.6}$ measured by Martindale *et al.* (Ref. 3), and fits using our theoretical expression Eq. (43).

$$\frac{1}{^{17}T_{1\alpha}T}=\left(\frac{1}{^{17}T_{1\alpha}T}\right)_{\chi_0}+\left(\frac{1}{^{17}T_{1\alpha}T}\right)_{\mathrm{AF}}. \tag{41}$$

Here the short-wavelength part $(1/^{17}T_1T)_{\chi_0}$ is proportional to the bulk magnetic susceptibility $\chi_0(T)$, while the antiferromagnetic part follows the copper relaxation rate:

$$\left(\frac{1}{^{17}T_{1\alpha}T}\right)_{\chi_0}=S_\alpha\chi_0(T),$$

$$\left(\frac{1}{^{17}T_{1\alpha}T}\right)_{\mathrm{AF}}=\frac{^{17}F_\alpha(\mathbf{Q_i})}{^{63}F_c(\mathbf{Q_i})}\left(\frac{1}{^{63}T_{1c}T}\right). \tag{42}$$

As we have demonstrated above, the temperature dependence of the antiferromagnetic leakage term is very different from what is observed in experiment. Since the empirical modified Korringa law $[1/^{17}T_1T\chi_0(T)]=$const is rather well satisfied for these materials, the short-wavelength part should be dominant. Therefore, we can write for the different $^{17}O$ relaxation rate ratios:

$$\frac{(1/^{17}T_1)_{\alpha_i}}{(1/^{17}T_1)_{\alpha_j}}=\frac{S_i\chi_0(T)+P_i/^{63}T_{1c}T}{S_j\chi_0(T)+P_j/^{63}T_{1c}T}$$
$$\simeq\frac{S_i}{S_j}\left[1+\left(\frac{P_i}{S_i}-\frac{P_j}{S_j}\right)\left(\frac{1}{^{63}T_{1c}T\chi_0(T)}\right)\right], \tag{43}$$

where $P_j={}^{17}F_{\alpha_j}(\mathbf{Q_i})/^{63}F_c(\mathbf{Q_i})$, while the $S_j$ are coefficients determined by integrating the product of the short-range part of the magnetic susceptibility with the oxygen form factor. If the short-range part of $\chi''(\mathbf{q},\omega)$ is only mildly $\mathbf{q}$ dependent, $S_j$ is determined primarily by the momentum average of $^{17}F_{\alpha j}$,

$$S_j=\frac{\pi}{\Gamma}\int {}^{17}F_{\alpha_j}(\mathbf{q})d^2q. \tag{44}$$

In this case the temperature-independent part of the ratio of the oxygen relaxation rates is determined again only by the ratio of the form factors:

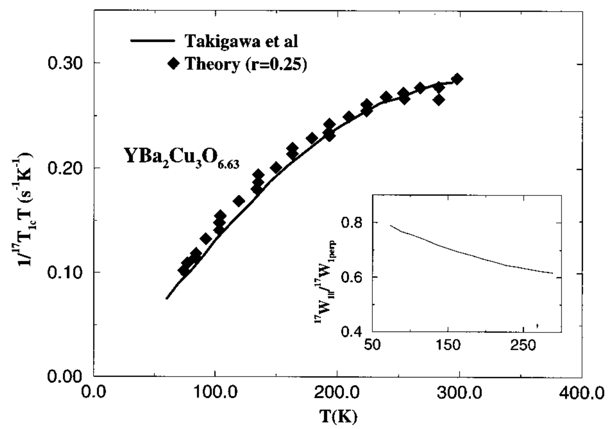


FIG. 9. The calculated spin-lattice relaxation rates $(^{17}T_{1c}T)^{-1}$ for $YBa_2Cu_3O_{6.63}$, compared with the data of Takigawa *et al.* (Ref. 5). The inset shows the predicted anisotropy ratio as described in the text. The predicted anisotropy lies slightly above the calculated curve for $YBa_2Cu_3O_{6.6}$ in Fig. 8, yet both are within the experimental error bars of the $YBa_2Cu_3O_{6.6}$ data of Martindale *et al.* (Ref. 3).

$$\frac{S_i}{S_j}=\frac{^{17}F_{\alpha_i}(\mathbf{r}=0)}{^{17}F_{\alpha_j}(\mathbf{r}=0)}. \tag{45}$$

If a realistic band-structure $\mathbf{q}$ dependence of $\chi''(\mathbf{q},\omega)$ is taken into account, this ratio will have a somewhat different value. We demonstrate, in Fig. 8, that expression Eq. (43) indeed provides a consistent explanation of the temperature-dependent term for the oxygen relaxation rates in $YBa_2Cu_3O_7$; on using Eq. (43) to fit the observed anisotropy ratio of $^{17}W_\parallel/^{17}W_\perp$, we find

$$\frac{S_\parallel}{S_\perp}=0.5,$$

$$\frac{S_\parallel}{S_\perp}\left(\frac{P_\parallel}{S_\parallel}-\frac{P_\perp}{S_\perp}\right)=0.06\ \ (\mathrm{s\,K})\mu_B^2/\mathrm{eV}. \tag{46}$$

These values of $S_{i,j}$ and $P_{ij}$ impose certain constraints on the parametric space of the hyperfine couplings. There are not enough of these constraints to enable us to deduce unambiguously the values of the hyperfine couplings, so that specific quantum chemical calculations are needed to determine the hyperfine coupling constants for these materials. However, as we have shown, the temperature dependence of the rates can be accounted for by assuming a finite incommensurability for the antiferromagnetic peak.

Using our formalism, and the constants $S_j$ and $P_j$ for $YBa_2Cu_3O_7$, we can predict the oxygen relaxation rates for $YBa_2Cu_3O_{6.63}$. It is easy to see that if the hyperfine $C$ couplings do not depend significantly on doping, the product, $S_j\Gamma$, for $YBa_2Cu_3O_{6.63}$ is the same as for $YBa_2Cu_3O_7$. $P_j$, however, can be somewhat different, corresponding to a different amount of incommensuration for $YBa_2Cu_3O_{6.63}$. Since the oxygen form factor is quadratic in the vicinity of $(\pi/a,\pi/a)$, we can write

$$P_{j\,\mathrm{YBa_2Cu_3O_{6.63}}}=P_{j\mathrm{YBa_2Cu_3O_7}}\frac{\delta^2_{\mathrm{YBa_2Cu_3O_{6.63}}}}{\delta^2_{\mathrm{YBa_2Cu_3O_7}}}. \tag{47}$$

However, the degree of incommensurability in $YBa_2Cu_3O_{6.63}$ (Ref. 11) is roughly the same as in $YBa_2Cu_3O_7$, both have $\delta\sim 0.1\pi$, so that $P_j$ will remain unchanged from the $YBa_2Cu_3O_7$ values. This makes it possible to predict the behavior of the oxygen relaxation rates ratios in $YBa_2Cu_3O_{6.63}$, once the parameter $\Gamma$ in Eq. (9) is determined from experiment. In Fig. 9, we fit the $^{17}$O relaxation rates $(^{17}T_{1c}T)^{-1}$ of Takigawa *et al.* to determine $S_c$. Again, we use the $\chi_0$, $\alpha$, and $\xi$ given in Ref. 2. It is seen in the main portion of Fig. 9 that the fit is very satisfactory; from this fit we obtain $\Gamma=226$ meV for $YBa_2Cu_3O_{6.63}$, if we assume $r_{\alpha_i}=0.25$. These parameters are also listed in Table III. From Eq. (44), we have $S_i\Gamma$ being the same in $YBa_2Cu_3O_7$ and $YBa_2Cu_3O_{6.63}$, because their form factors do not change. Therefore, we get for $YBa_2Cu_3O_{6.63}$,

$$\frac{S_\parallel}{S_\perp}=0.5,$$

$$\frac{S_\parallel}{S_\perp}\left(\frac{P_\parallel}{S_\parallel}-\frac{P_\perp}{S_\perp}\right)_{\mathrm{YBa_2Cu_3O_{6.63}}}=\frac{S_\parallel}{S_\perp}\left(\frac{P_\parallel}{S_\parallel}-\frac{P_\perp}{S_\perp}\right)_{\mathrm{YBa_2Cu_3O_7}}\times\frac{S_c(\mathrm{YBa_2Cu_3O_7})}{S_c(\mathrm{YBa_2Cu_3O_{6.63}})}$$

$$=0.06\times\frac{\Gamma_{\mathrm{YBa_2Cu_3O_{6.63}}}}{\Gamma_{\mathrm{YBa_2Cu_3O_7}}}$$

$$=0.044\ \ (\mathrm{s\,K})\mu_B^2/\mathrm{eV}. \tag{48}$$

We show our calculated relaxation rate ratio for $YBa_2Cu_3O_{6.63}$ in the inset of Fig. 9.

Finally, we note that it is straightforward to extend to the superconducting state the analysis which led to Eq. (43); one obtains in this way a modified version of Eq. (43) in which the parameters $P_\alpha$ and $S_\alpha$ will depend on the coherence factors in the superconducting state. We note that once the energy gap has opened up ($T\lesssim 0.7T_c$, say) the system behavior is determined by quasiparticles in the vicinity of the nodes of the $d_{x^2-y^2}$ gap function. Under these circumstances, the form factors, $^{17}F_\alpha$ and $^{63}F_c$ will be those appropriate to the momentum transfers which characterize the transitions between the nodal quasiparticles, while, $\chi_0(T)$ falls off linearly with decreasing $T$, and $(^{63}T_{1c}\chi_0)^{-1}$ falls off as $T^2$. As a result, the anisotropy at low temperatures may be expected to fall off linearly with temperature for $T\lesssim 0.7T_c$, approaching to constant $S_i/S_j$ at very low temperatures. Detailed calculations of $^{17}W_\alpha(T)$ using the modified SMR Hamiltonian will be required before any quantitative comparison can be made with the superconducting state results of Martindale *et al.*[3]

## VII. ABSOLUTE INTENSITY OF THE DYNAMICAL SPIN SUSCEPTIBILITY FOR $YBa_2Cu_3O_7$

In this section, we consider the dynamical spin susceptibility $\chi''(\mathbf{q},\omega)$ derived from various NMR experiments and compare it with the energy scales and absolute intensities determined in neutron-scattering experiments.

As shown by Barzykin and Pines,[2] $T_{\mathrm{cr}}\sim 125$ K for $YBa_2Cu_3O_7$, which indicates $\xi\sim 2$ at 125 K. From the $T_{2G}^{-1}$ data of Imai *et al.*[36] at 125 K, we get $\alpha=14.8$ states/eV on using the relevant expression shown in Table III. We then find $\xi=2.08$ for T=100 K from the $T_{2G}^{-1}$ data, and $\omega_{\mathrm{SF}}=16.6$ meV at $T=100$ K from the $T_1T$ data of Imai *et al.*[36]

We next examine the dynamical spin susceptibility at $\mathbf{Q}=(\pi/a,\pi/a)$ obtained from the above NMR fit. As indicated in Sec. II, on assuming the spin-fluctuation spectrum consists of four incommensurate peaks located at $\mathbf{Q}_i=([1\pm 0.1]\pi/a,[1\pm 0.1]\pi/a)$, the anomalous antiferromagnetic contribution to $\chi''(\mathbf{q},\omega)$ can be written as

$$\chi''(\mathbf{q},\omega)=\frac{1}{4}\sum_i\frac{\alpha\xi^2(\omega/\omega_{\mathrm{SF}})}{[1+(\mathbf{q}-\mathbf{Q}_i)^2\xi^2]^2+(\omega/\omega_{\mathrm{SF}})^2}. \tag{49}$$

Therefore at $T=100$ K, we can write

$$\chi''(\pi,\pi,\omega)=\frac{34\omega/\tilde{\omega}_{\mathrm{SF}}}{1+\omega^2/\tilde{\omega}_{\mathrm{SF}}^2}\ \text{states/eV}, \tag{50}$$

where

$$\tilde{\omega}_{\mathrm{SF}}\equiv\omega_{\mathrm{SF}}\times[1+(\mathbf{Q}-\mathbf{Q}_i)^2\xi^2]=1.85\omega_{\mathrm{SF}}=31\ \text{meV}. \tag{51}$$

Thus from our assumption of incommensuration and our fit to the NMR experiments, we predict a weak spin-fluctuation peak, at $(\pi,\pi)$, with energy 31 meV, and strength 17 states/eV.

Experimentally, it is still controversial whether there exists a measurable spin-fluctuation peak in the normal state. Dai *et al.*[9] report the observation of a weak peak near $\omega\sim 35-40$ meV, while Fong *et al.*[23] do not see any peak structure, but provide an upper limit, 30 states/eV for the strength of the spin-susceptibility spectrum between 10 meV $\leqslant\omega\leqslant 40$ meV. Thus, our prediction of a characteristic spin-fluctuation energy scale of $\tilde{\omega}_{\mathrm{SF}}\sim 31$ meV and a maximum intensity of 17 states/eV is compatible with both neutron-scattering experiments.

We next consider the integrated intensity, $\int d\omega\chi''(\pi,\pi,\omega)$, in the normal and superconducting states of $YBa_2Cu_3O_7$. From Eq. (49), we have

$$\int_0^{\omega_c}d\omega\chi''(\pi,\pi,\omega)=\frac{\alpha\omega_{\mathrm{SF}}\xi^2}{2}\ln\left[1+\left(\frac{\omega_c}{\tilde{\omega}_{SF}}\right)^2\right]$$

$$=0.53\ln\left[1+\left(\frac{\omega_c}{\tilde{\omega}_{SF}}\right)^2\right]. \tag{52}$$

For the normal state, at $T=100$ K, we may expect $\omega_c$, the cutoff energy, will lie between $\omega_{\mathrm{SF}}\xi^2$ and $\tilde{\omega}_{\mathrm{SF}}\xi^2$. On taking $\omega_c\sim 100$ meV, we find the integrated total weight at $(\pi,\pi)$ is $\int d\omega\chi''(\pi,\pi,\omega)=1.3$. We may also estimate the integrated intensity of the 41 meV peak in the superconducting state to compare with the experimental value of Fong *et al.*[23] For this purpose, we assume that at low temperatures, the entire normal-state spectrum below the superconducting gap

($2\Delta\sim 40$ meV) is turned into the 41 meV resonant peak, observed in the superconducting state;[8–10] we then have

$$\int_{\omega=41\ \mathrm{meV}} d\omega\chi''(\pi,\pi,\omega)|_{\mathrm{sup}}=\int_0^{40\ \mathrm{meV}} d\omega\chi''(\pi,\pi,\omega)|_{\mathrm{normal}}$$
$$=0.52, \tag{53}$$

in excellent agreement with the experimental value $0.49\pm 0.1$ of Fong *et al.*[23]

## VIII. DISCUSSION AND CONCLUSIONS

We have seen that by modifying the SMR hyperfine Hamiltonian we can use the MMP one-component spin-spin response function to reconcile the results of a number of neutron-scattering and NMR experiments on the cuprate superconductors. With the aid of the scaling arguments of Barzykin and Pines,[2] we are able to obtain a quantitative fit to both the NMR and the neutron-scattering data for $La_{1.86}Sr_{0.14}CuO_4$. We find that for the $YBa_2Cu_3O_{6+x}$ system, we can reconcile the **q** width of the antiferromagnetic peak seen in neutron scattering experiments with the substantial temperature-dependent AF correlation required to explain the NMR experiments on $YBa_2Cu_3O_7$ and $YBa_2Cu_3O_{6.63}$. Moreover, in the recent results of Martindale *et al.*[3] on the anomalous temperature dependence of the anisotropy of the $^{17}$O relaxation rates, the small amount of the AF leakage is shown not only to be explicable using our modified one-component description; but to provide an independent consistency check for our one-component picture. Our ability to reconcile so many different experiments leads us to conclude that a transferred hyperfine coupling between next-nearest-neighbor $Cu^{2+}$ spins and $^{17}$O nuclei spin plays a significant role, and that the transferred hyperfine coupling $B$, changes as one goes from the $La_{2-x}Sr_xCuO_4$ to the $YBa_2Cu_3O_{6+x}$ system, and is moreover comparatively sensitive to hole doping in the former system. It will be interesting to see whether the presence of these terms can be justified microscopically through detailed quantum chemical calculations in these systems. We doubt that our modified SMR hyperfine Hamiltonian is unique. Thus it may eventually prove desirable to devise other qualitatively new hyperfine couplings. What we have attempted to do is to devise the minimal modification of the SMR model required to obtain agreement with the present generation of experiments. We have no ready explanation for the doping dependence of $B$ which is required to explain the experimental results on the anisotropy of the $^{63}$Cu spin-lattice relaxation rates in the $La_{2-x}Sr_xCuO_4$ system or why it is much less dependent on doping in the $YBa_2Cu_3O_{6+x}$ system. A check on whether $C$ and $C'$ are doping dependent in either system will come from measurements of the doping dependence of the anisotropy of the planar $^{17}$O spin-lattice relaxation rates. If $C$ and $C'$ are not markedly doping dependent, then this anisotropy will exhibit the dependence on $^{63}T_{1c}T\chi_0(T)$ which is given in Eq. (43).

For the $La_{2-x}Sr_xCuO_4$ system, the values of $\xi$, $\omega_{\mathrm{SF}}$, and $\alpha$ we have deduced in the present paper are sensitive to our choice of the hyperfine constants, $A_{ab}$ and $B$; these in turn depend sensitively on the measurements of $^{63}R$, which is not known to better than 10% accuracy. Moreover, in calculating $^{17}T_{1c}$, for $La_{1.86}Sr_{0.14}CuO_4$ we have used hyperfine constants and values for $\chi_0(T)$ which do not reflect any possible anisotropy of the $g$ factor such as that found by Walstedt *et al.*[40] for the $YBa_2Cu_3O_{6+x}$ system, while our choice of $\Gamma$ is obviously sensitive to our choice of a cut-off wave vector for the applicability of the MMP expression for $\chi_{\mathrm{AF}}(\mathbf{q},\omega)$. For the $YBa_2Cu_3O_{6+x}$ system, the spin fluctuation parameters and hyperfine constants depend on the extent to which the spin fluctuation spectrum is incommensurate. We have assumed a degree of incommensuration which is compatible with current INS experiment, but until direct measurements of that incommensuration can be carried out, there remains a considerable degree of uncertainty. Given these unavoidable uncertainties, an overall assignment of an accuracy of some 20% in the results we have presented here would seem a consistent chioce.

Our results have a number of interesting implications for (nearly antiferromagnetic Fermi-liquid theory (NAFL) calculations of other properties of the superconducting cuprates. For example, Pines and Monthoux[37] have shown that incommensuration acts to lower the superconducting transition temperature, $T_c$; it is tempting therefore to attribute much of the substantial difference in $T_c$ found for the $La_{2-x}Sr_xCuO_4$ and $YBa_2Cu_3O_{6+x}$ systems to the much greater degree of incommensuration found in the former materials. In their calculation of planar resistivities, Stojkovic and Pines[38] find that $\rho_{ab}$ depends sensitively on the size and distribution of ''hot spots''( regions of the Fermi surface connected by $\mathbf{Q}_i$), and thus is markedly changed by incommensuration. To cite a third example, in NAFL theory, the location in momentum space of the peak in the spin-fluctuation spectrum depends on the interplay of the peaks in the irreducible particle-hole susceptibility, $\tilde{\chi}(\mathbf{q},0)$, produced by band structure and the momentum dependence of the restoring force, $J_{\mathbf{q}}$, which acts to shift those peaks according to Ref. 39,

$$\chi(\mathbf{q},0)=\frac{\tilde{\chi}(\mathbf{q},0)}{1-J_{\mathbf{q}}\tilde{\chi}(\mathbf{q},0)}. \tag{54}$$

Since the peaks in $\tilde{\chi}(\mathbf{q},0)$ move away from $(\pi/a,\pi/a)$ as one moves away from half-filling, less peaking in $J_{\mathbf{q}}$ is required to produce four incommensurate peaks than was needed by Monthoux and Pines[39] to keep the peak at $(\pi/a,\pi/a)$ in the presence of substantial hole doping.

Further NMR and neutron experiments on the $YBa_2Cu_3O_{6+x}$ and $La_{2-x}Sr_xCuO_4$ systems can also help verify the correctness of our proposed hyperfine Hamiltonian and our assignment of incommensurate peaks in the $YBa_2Cu_3O_{6+x}$ system. For example, our results, Eqs. (36) and (43), lead us to predict substantial temperature dependence in the anisotropy of $1/^{17}T_{1\alpha}$ in the $La_{2-x}Sr_xCuO_4$ system for magnetic fields parallel and perpendicular to the Cu-O bond axis, and it will be instructive to see whether this can be measured. It is, moreover, to be hoped that improvements both in neutron-scattering facilities and the availability of large single crystals will make possible a direct experimental check on our assignment of incommensuration in the $YBa_2Cu_3O_{6+x}$ system. Resolution of those peaks, together with a direct measurement of their intensities would also enable one to carry out a detailed comparison of NMR and neutron-scattering experiments on $YBa_2Cu_3O_{6.63}$ analogous to that presented here for the $La_{1.86}Sr_{0.14}CO_4$ system.

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