NMR, Neutron Scattering, and the One-Band Model of $La_{2-x}Sr_xCuO_4$

R. E. Walstedt, B. S. Shastry, and S-W. Cheong
AT&T Bell Laboratories, 600 Mountain Avenue, Murray Hill, New Jersey 07974
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NMR data on ^{17}O and ^{63}Cu in $La_{1.85}Sr_{0.15}CuO_4$ are presented and agree well with published work. Relaxation curves for ^{63}Cu and for both apical and planar ^{17}O sites are compared with calculations based on parametrization of recent absolute neutron susceptibilities $\chi''(\mathbf{q},\omega)$. A reasonable accord is found with the ^{63}Cu data. However, a significant discrepancy with the observed planar ^{17}O relaxation behavior poses an apparent contradiction with the widely accepted one-band dynamical model for this system.

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Several years ago a one-band interpretation of NMR shift and relaxation time T_1 data on high-T_c superconducting compounds was proposed [1], wherein the sharp contrast between relaxation behaviors at various sites in the lattice could be accounted for via **q**-dependent hyperfine terms. In particular, one hoped to be able to explain how Korringa-like relaxation behavior (T_1T =const) at the planar oxygen sites [2] could coexist with sharply non-Korringa behavior at the (planar) copper sites [3,4] in the system $YBa_2Cu_3O_7$ ($YBCO_7$) without invoking multiple carrier bands. Subsequent work [5–9] has developed the one-band approach to this problem extensively, using mean-field model dynamic susceptibilities and the spin Hamiltonian formulation [1,10] for the magnetic properties of the Cu^{2+} ions in these systems. NMR data on oxygen-deficient material $YBCO_{6.6-7}$ ($T_c \sim 60$ K) [11,12] are thought to give additional support to a one-band picture in that the NMR shifts at planar Cu and O sites, strongly temperature dependent in this case because of a spin gap effect [13,14], show nearly coincident temperature profiles as though derived from a common source. However, whether the one-band picture can simultaneously give a satisfactory account of the ^{63}Cu, ^{89}Y, and ^{17}O relaxation behavior for $YBCO_7$ [6] depends on there being a sufficiently narrow, commensurate antiferromagnetic (AFM) peak in the dynamic susceptibility $\chi''(\mathbf{q},\omega)$. The success of this scheme has been questioned extensively on theoretical grounds [7]. Support for it from neutron scattering data is also marginal [15,16], leaving the one-band approach in a state of some uncertainty.

On the other hand, recent neutron data on superconducting $La_{2-x}Sr_xCuO_4$ (LSCO) [17,18] have revealed distinctly incommensurate fluctuation peaks in $\chi''(\mathbf{q},\omega)$ for that system. Such an incommensurability prevents the planar O-site hyperfine form factor from canceling out the fluctuation peak contribution to the T_1 process at that site. Whether the latter contribution is appreciable, and, if so, is observed, provides a more stringent test of the one-band picture for these systems than the YBCO cases cited above. In this paper we present NMR shift and relaxation data for ^{17}O and ^{63}Cu in $La_{1.85}Sr_{0.15}CuO_4$ [19–21]. The ^{17}O data are the first complete set to cover the range from T_c up to 250 K. Using a numerical representation of the neutron data for $\chi''(\mathbf{q},\omega)$, including absolute amplitude, and hyperfine constants estimated here as well as from the literature, we have evaluated the incommensurate fluctuation peak contribution to the ^{63}Cu and ^{17}O T_1 processes. The ^{63}Cu data are in approximate agreement with values calculated in this way. In contrast, the planar ^{17}O T_1 data are in striking disagreement with the sizable AFM fluctuation contribution which the above procedure yields. Instead, the planar ^{17}O site T_1 process appears to be completely dominated by a background ($\sim$**q** independent) susceptibility term, much as in the YBCO phases.

Nuclear spin echo measurements have been carried out on a single-phase, oriented powder sample of $La_{1.85}Sr_{0.15}CuO_4$. The source material for this specimen was synthesized using standard methods. Magnetic measurements gave $T_c \sim 35$ K for this material. ^{17}O NMR spectra at a series of temperatures are shown in Fig. 1 for this sample, where we see (a) a central ($\frac{1}{2} \leftrightarrow -\frac{1}{2}$) transition consisting of two separate maxima, (b) a first-order qua-

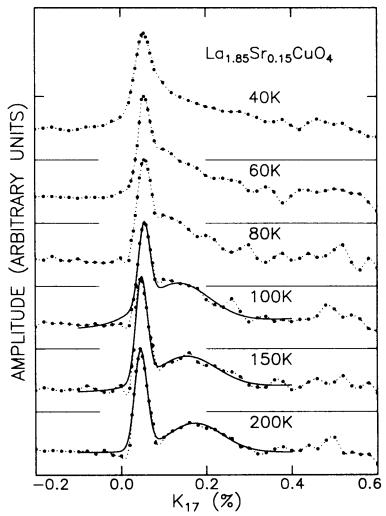

FIG. 1. ^{17}O NMR spectra on $La_{1.85}Sr_{0.15}CuO_4$, taken at 45.5 MHz, are shown at a series of temperatures. The ^{17}O shift is defined with respect to $\gamma_{17}=0.57719$ kHz/G. The dotted lines are spline fits to each data set. The solid lines shown for the three highest temperatures are fitted curves based on two Gaussians and an adjusted base line.

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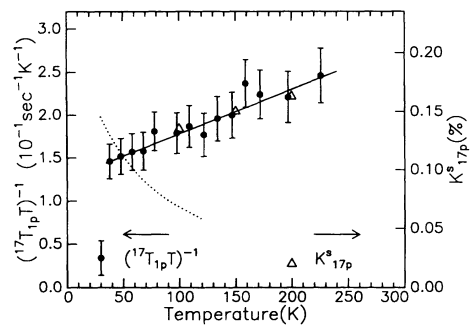

FIG. 2. Relaxation data for the planar ^{17}O spins are plotted as $(^{17}T_{1p}T)^{-1}$ vs T (dots). The open triangles are spin paramagnetic shift data scaled to illustrate their similar temperature dependence. The solid line is a guide to the eye. The dotted line shows the calculated contribution $(T_{1p}T)^{-1}_{neut}$ from the AFM fluctuation peaks (neutron data) based on a one-band model as described in the text.

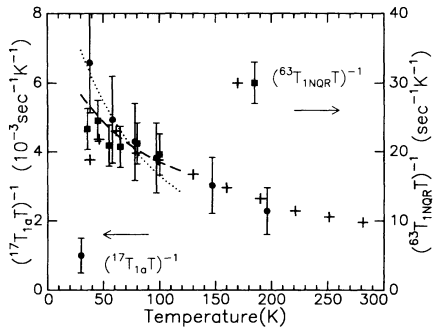

FIG. 3. Apical ^{17}O relaxation data are plotted vs T as in Fig. 2 (dots). Scaled to coincide at $T>80$ K are ^{63}Cu NQR data from this work (squares) and from Imai [21] (+'s). The dotted line is the calculated AFM fluctuation peak contribution $(T_1T)^{-1}_{neut}$ for the ^{63}Cu based on absolute neutron susceptibility results [30,31]. The dashed line is a curve fitted to the ^{63}Cu data as described in the text.

drupolar satellite at $K_{17}\sim 0.5\%$ similar to that found in published spectra [19], and (c) a substantial background smear of satellite intensity corresponding to unoriented sample material. The breadths and structure seen in the $(\frac{1}{2}\leftrightarrow -\frac{1}{2})$ spectra, entirely magnetic in origin, are also similar to examples from the literature [20]. The two peaks observed are identified [20] as the apical (narrow line) and planar (broad line) oxygen sites. The planar site line moves gradually to lower shift values at lower temperatures, reflecting the temperature dependence of the spin susceptibility. On account of its small shift, the apical line is very nearly stationary.

T_1 measurements have been carried out for ^{63}Cu using nuclear quadrupole resonance (NQR) and for both oxygen sites using NMR. The planar ^{17}O relaxation data are plotted in Fig. 2 as $(^{17}T_{1p}T)^{-1}$ vs T. The solid line is a guide for the eye. These data agree within estimated errors with measurements reported earlier [20,22], the overall temperature variation being, however, quite different from that indicated in [20]. The positive temperature slope of $(T_1T)^{-1}$ seen in Fig. 2 indicates a deviation from Korringa behavior characteristic of the spin gap effect [12–14]. We underscore this point by also plotting in Fig. 2 shift values (triangles) from the Gaussian fits to the spectra in Fig. 1 at $T=100$, 150, and 200 K. These values are scaled to show the approximate relationship $K_s(T)\propto(T_1T)^{-1}$, which is found in a number of spin gap cases [12,23]. In Fig. 3 we plot, in the same fashion as Fig. 2, relaxation data for the apical ^{17}O site as well as NQR data for the ^{63}Cu, scaled to show an approximate correspondence at the high-temperature end. As with the YBCO phases, both these sites display a sharp contrast in temperature variation with the planar ^{17}O site. This contrast has traditionally been attributed to contrasting hyperfine form factors [1,2,5,6,9,12]. Quite unexpectedly, there is also an apparent disparity between the apical ^{17}O and the ^{63}Cu at temperatures below $T\sim 80$ K, in spite of similar form factors for these sites.

To interpret these relaxation data, we first note that in a multiband situation with n_B species of fluctuating spins, the hyperfine coupling Hamiltonian is

$$^{a}H_{hf}=\sum_{\mathbf{q};\mu=1}^{n_B}\sum_{\rho}{}^{a}\mathcal{H}_{\rho}(\mathbf{q}|\mu)I_{a\rho}(\mathbf{q})S_{\mu\rho}(-\mathbf{q}),$$

with μ representing a band index. The system is described by susceptibilities $\chi^{\rho}_{\mu\nu}(\mathbf{q},\omega)$ $(=\langle\langle S^{\rho}_{\mu}(\mathbf{q});S^{\rho}_{\nu}(-\mathbf{q})\rangle\rangle_{\omega})$ $(\rho=a,b,c)$, and the nuclear gyromagnetic ratio γ_a, so that the relaxation rate (say with H along the c axis) is given by

$$(^{a}T_{1c}T)^{-1}=\frac{\gamma_a^2k_B}{\mu_B^2}\sum_{\mathbf{q};\mu,\nu=1}^{n_B}\sum_{\rho=a,b}{}^{a}\mathcal{H}_{\rho}(\mathbf{q}|\mu)\,{}^{a}\mathcal{H}_{\rho}(\mathbf{q}|\nu)[\chi''^{\rho}_{\mu\nu}(\mathbf{q},\omega)/(g_{\rho}^2\omega)]_{\omega\to 0}. \quad (1)$$

The Knight shift of nuclear spins a with the field in the ρ direction is given by $^{a}K_{\rho}=\sum_{\mu,\nu=1}^{n_B}{}^{a}\mathcal{H}_{\rho}(0|\mu)\chi^{\rho}_{\mu\nu}(0,0)/2\mu_B\times N_{\text{Avogadro}}$. In these formulas we have allowed for anisotropic, diagonal hyperfine coupling and g-shift tensors. If we insist that there is only one species of fluctuating spins, then the sums over μ,ν are omitted and we set $n_B=1$. In this case the form factors are given for the planar ^{17}O nucleus as [1] $^{17}\mathcal{H}_{\rho}(\mathbf{q})=2C^{\rho}_{p}\cos(q_xa/2)$ $(\rho=a,b,c)$, for the apical ^{17}O nucleus as $^{17}\mathcal{H}_{a}(\mathbf{q})=C^{a}$, and for the ^{63}Cu we take $^{63}\mathcal{H}(\mathbf{q})=A_{ab}+2B(\cos q_xa+\cos q_ya)$ [1,24]. The ^{63}Cu hyperfine constants are estimated to be [25,26] $A_{ab}=18$ and $B=92$ in units of kG/spin. The C^{ρ}_{p} tensor for LSCO has been extracted from shift and susceptibility data [27–29] with the results given in Table I.

Calculations of ^{17}O and ^{63}Cu relaxation rates have been carried out using Eq. (1) and dynamic susceptibility data derived from neutron scattering results [17]. The latter measurements yield a susceptibility in a region of the Brillouin zone where the intensity is sufficiently large (accounting for $\sim 13.5\%$ of the area). We write for the neutron susceptibility [17,30]

TABLE I. NMR shift and T_1 data, spin susceptibilities, and g factors for LSCO (A) and the 60 K (B) and 90 K (C) phases of YBCO are tabulated ($T=300$ K) along with values for the bandwidth parameter Γ_B [Eq. (3)]. Shift values are ± 20 and are corrected for orbital shifts $\chi^{orb}_{(a,b,c)}=(46,8,-27)$ [12]. Units are as follows: K^s_ρ, 10^{-5}; C^s_ρ, kG/spin; χ^s_ρ, 10^{-6} emu/mole; $T_{1c}T$, sec K; Γ_B, meV.

	K^s_a	K^s_b	K^s_c	C^p_a	C^p_b	C^p_c	g_{ab}	g_c	χ^s_{ab}	χ^s_c	$T_{1c}T$	Γ_B
A	274	162	214	108	64	77	2.06	2.27	146	177	3.57	82
B	214	133	157	181	113	118	2.08	2.34	69	87	3.16	106
C	264	162	207	159	97	110	2.08	2.34	96	123	2.70	97

$$[\chi''_n(\mathbf{q},\omega)/\omega]_{\omega\to 0}=\hbar\frac{k_0a^2}{\Gamma_n^2(q)}\theta(\Gamma_c-\Gamma_n(q))\,. \tag{2}$$

This term represents the incommensurate peaks in $S(\mathbf{q},\omega)$ near $\mathbf{Q}_{AFM}=\pi(0.755,1)$ and the symmetry related vectors. The absolute scale parameter k_0 has been estimated [31]. We have computed relaxation rates $(T_1T)^{-1}_{neut}$, using the hyperfine constants given with Eq. (1) and assuming that only the neutron determined term [Eq. (2)] is operative for various nuclei. The dotted line in Fig. 3 shows the result for the ^{63}Cu nucleus. The magnitude of $(T_1T)^{-1}_{neut}$ is approximately in accord with the ^{63}Cu data of Fig. 3. The temperature variation is also in approximate accord with the apical ^{17}O data, but is much steeper at the low temperature end than the ^{63}Cu data. Clarification of this point awaits a more extensive body of neutron data.

For the planar ^{17}O, the picture is substantially different. We plot the result calculated from Eqs. (1) and (2) using the hyperfine tensor components for C^p_ρ from Table I in Fig. 2 as a dashed line. Despite the large cancellation produced by the oxygen form factor [$\langle\cos(q)\rangle\sim -0.87$], there is a substantial contribution to the rate with a temperature dependence completely contrary to the experimental data. It is our opinion that the tensor components C^p_ρ cannot be revised downward significantly [27] and downward revision of the fluctuation peak susceptibility will only worsen the agreement with the ^{63}Cu.

The clear implication of the comparison in Fig. 2 is that the transferred hyperfine coupling between the planar oxygen and the copper susceptibility is substantially smaller than the total couplings listed in Table I. The only way we see for this to be incorrect is if the discommensurations δ reported [17,18] are not applicable in Eq. (1). We discuss two scenarios whereby this might be the case: First, one might conjecture that δ is frequency dependent in such a way that at NMR frequencies it is much smaller than the value $\delta=0.245$ used here [30]. One cannot, of course, rule this out, but there are reasons to suppose that it is improbable. For example, in the data reported [17,18], there is no observed change in δ over energies ranging from 30 mV down to 1 mV to suggest that δ declines at energies below 1 mV. On the other hand, the ^{63}Cu results show that at least the magnitude of $\chi''(\mathbf{q},\omega)$ extrapolates successfully to NMR frequencies, giving confidence that the form of Eq. (2) remains valid. Finally, we note that 1 mV is well below k_BT here, which should be well into the asymptotic low frequency regime. There is no known energy scale smaller than this, below which a transition $\delta\to 0$ might be expected to take place. Further neutron data at lower energies would, of course, be very desirable. Second, one may suggest the possibility of magnetic domains of short-range order separated by disclinations, wherein commensurate peaks may consequently be shifted and broadened in k space. If this were the case, then there would also be harmonic peaks at points further from (π,π). Such peaks do not appear to be present [32]. With minor reservations, then, the data in Fig. 2 lead us to conclude that ^{17}O hyperfine coupling with the susceptibility of Eq. (2) is very small. It then follows from the derivation of Eq. (1) (see also [6]) that the shift and relaxation of the planar oxygen nuclei *are not driven by Cu-site susceptibilities at all.* The one-band model therefore appears to be contradicted by these results. Because of the wide susceptibility peaks reported for YBCO [16,33], we believe that a similar conclusion holds for that system as well, evidence to the contrary notwithstanding [12]. A detailed discussion is, however, beyond the scope of this paper.

Regarding the planar ^{17}O T_1 data, we note that the rate follows the temperature dependence of the uniform susceptibility in the LSCO system (Fig. 2), as is also found in the YBCO phases [12]. This suggests that we write, phenomenologically, an additional $\mathbf{q}$-independent susceptibility term in the form $[\chi''_\rho(\mathbf{q},\omega)/\omega]_{\omega\to 0}=\chi^s_\rho(T)/\Gamma_B$, where $\chi^s_\rho(T)$ is the uniform spin susceptibility in direction ρ and Γ_B is a bandwidth parameter. This leads to an expression for the relaxation rate in terms of shift and hyperfine tensor components [27],

$$(T_{1\rho}T)^{-1}=\frac{\hbar\gamma_{17}^2k_B}{\mu_B\Gamma_B}\left[\frac{C^p_aK^s_a}{g_a}+\frac{C^p_bK^s_b}{g_b}\right]. \tag{3}$$

It is instructive to fit Eq. (3) to the planar ^{17}O relaxation data in Fig. 2 so as to determine Γ_B, and to compare this result with similarly derived values for the YBCO phases. The relevant parameter values for the latter systems have been derived from data in the literature [2,12,25,34,35]. They and the resulting Γ_B values are listed in Table I. The Γ_B's are seen to be roughly 0.1 eV, which is of the order of the exchange coupling J in the antiferromagnetic phases of these compounds. It is interesting that the values of Γ_B for these systems correspond rather closely

in spite of strikingly different temperature dependences for the shifts and relaxation rates [9]. This correspondence supports our earlier conjecture that a similar background susceptibility dominates the planar ^{17}O relaxation in all three systems. Moreover, it implies that Γ_B is essentially a structural property of the hole-doped CuO_2 planar conduction band.

In conclusion, we find that calculated nuclear relaxation rates based on recent neutron data for dynamic susceptibilities in LSCO do not agree with planar ^{17}O T_1 data, in apparent contradiction with the traditional one-band model of dynamics for this system.

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[26] We adopt the values given in Ref. [25], because they incorporate three features not found elsewhere in the literature: (a) Only static shift and susceptibility data are used, (b) chain and plane-site spin susceptibilities are partitioned using these data, and (c) the important effects of anisotropic g factors are included.
[27] The hyperfine tensor components [see Eq. (1)] are derived from the equation [25] ${}^{\alpha}\mathcal{H}_{\rho}(0)=2C_{\rho}^{\beta}=g_{\rho}K_{\rho}^{s}/(0.179\times\chi_{\rho}^{s})$, where χ_{ρ}^{s} is in emu/mole and C_{ρ}^{β} is in kG/spin. All parameters are given in Table I. The shift values are extracted from fits to room temperature spectra, and are corrected for chemical shifts (see table caption). The g factors are estimated from those for YBCO [25], noting that the ^{63}Cu orbital shift in LSCO is $\sim 20\%$ less than for YBCO ([29] and present work). We attribute this to an increased crystal field splitting, giving rise to correspondingly smaller g shifts and orbital (Van Vleck) susceptibilities. Room temperature susceptibility data for the LSCO material give $\chi_{\rm expt}(T)=98\times10^{-6}$ emu/mole, within $\pm 5\%$ of values reported in Ref. [19] and by D. C. Johnston, J. Magn. Magn. Mater. **100**, 218 (1991). To obtain $\chi_{\rho}^{s}(300\ \mathrm{K})$ one must subtract off the Van Vleck (χ_{ρ}^{VV}) and core diamagnetic ($\chi^{\rm core}$) terms. References [19] and [27] give $\chi_c=-99\times10^{-6}$. The χ_{ρ}^{VV} are taken to be 80% of the YBCO values [25]. The resulting susceptibilities (Table I) and other parameters are inserted into the above equation to yield the tabulated C_{ρ}^{β}'s.
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[30] The neutron energy scale is conveniently parametrized in Ref. [17] as $\Gamma_n(q)=a^2[\kappa^2+R(q)]$, with
$$R(\pi+q_1,\pi+q_2)=(8\pi^2a_L^2\delta^2)^{-1}\{[(q_1-q_2)^2-(\pi\delta)^2]^2+[(q_1+q_2)^2-(\pi\delta)^2]^2\},$$
where $a_L=3.8$ Å is the lattice constant, $\delta=0.245$, $a^2=3.858$ eV(Å)^2, and the correlation wave vector $\kappa=\sqrt{(0.043)^2+0.5k_BT}\,(\text{Å})^{-1}$, with k_BT expressed in eV. The cutoff in energy Γ_c is the maximum magnon energy, i.e., ~ 0.3 eV.
[31] The overall scale factor k_0 has been roughly (i.e., within say a factor of 2) estimated [17] to be $k_0a^2\sim 2.5\mu_B^2$ per Cu. Converting to units convenient for NMR, this gives us an absolute scale $\chi_n''(q,\omega_0)/\omega_0=0.83/\Gamma_n^2(q)\times10^{-4}$ emu per eV mole, with Γ_n in eV. We thank G. Aeppli and P. Littlewood for providing us with this estimate.
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