Random-Phase Approximation Analysis of NMR and Neutron-Scattering Experiments on Layered Cuprates

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Using a random-phase-approximation expression to approximate the dynamic susceptibility $\chi(\mathbf{q},\omega)$ of a 2D Hubbard model, we have calculated the spin-relaxation rates and the neutron-scattering intensity. Here, using recent static magnetic measurements to set parameters which enter the hyperfine form factors, we show that this simple form for $\chi(\mathbf{q},\omega)$ exhibits the features of the spin fluctuations that are required to fit the NMR data, and explore its consequences for the temperature and energy dependence of the magnetic neutron scattering.

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Measurements of the Knight shifts, nuclear spin-lattice-relaxation times T_1, and neutron-scattering intensities[1-9] provide insight into the nature of the magnetic excitations in the cuprate superconductors. In the high-T_c superconductor $YBa_2Cu_3O_7$, the rate T_1^{-1} for the planar ^{17}O nuclei exhibits a linear ("Korringa-like") temperature dependence[4] above T_c, while T_1^{-1} for the planar ^{63}Cu nuclei is greatly enhanced relative to a simple Korringa behavior[1-3] and appears to saturate at higher temperatures. Nevertheless, below about 115 K, these two relaxation rates track each other,[6] both exhibiting a sharp decrease below T_c but maintaining a nearly fixed ratio $(T_1^{-1})_{Cu}/(T_1^{-1})_O \simeq 20$. Furthermore, below T_c, both the ^{17}O and the ^{63}Cu Knight shifts with the field in the *a-b* plane (i.e., the CuO_2 plane) are reduced, indicating pair formation on both sites.[5,8] Finally, recent inelastic-neutron-scattering measurements[9] on $La_{1.85}Sr_{0.15}CuO_4$ have been reported which exhibit a decrease in scattering intensity between 150 and 50 K for energy transfers less than 12 meV.

This rich variety of phenomena has led to many different interpretations ranging from two-component descriptions,[8,10-12] consisting of localized Cu^{+2} spin excitations and a planar hole Fermi liquid, to various one-component theories.[13-16] Here we present results obtained within a weak-coupling random-phase-approximation (RPA) treatment of a 2D Hubbard model. This approach provides a simple parametrization for a dynamic spin susceptibility $\chi(\mathbf{q},\omega)$ with strong antiferromagnetic spin fluctuations. The hyperfine form factors which describe the coupling of electronic and nuclear spins filter $\chi(\mathbf{q},\omega)$, leaving the strong antiferromagnetic fluctuations at the Cu sites and suppressing them at the O sites.[1,6] As we have previously shown,[15] this leads to results which are qualitatively similar to NMR observations on $YBa_2Cu_3O_7$. Here we use the results of recent experiments to make a quantitative analysis within this approach. We also examine the RPA predictions for the temperature dependence of the neutron-scattering intensities at various energy transfers. Within the context of this model the experimental data reflect the saturation below a characteristic temperature T^* of the spectral weight of the low-frequency antiferromagnetic fluctuations.

The nuclear relaxation rate is given by

$$T_1^{-1} = \frac{T}{N}\sum_{\mathbf{q}} |A(\mathbf{q})|^2 \frac{\mathrm{Im}\chi(\mathbf{q},\omega)}{\omega}. \quad (1)$$

Here $\chi(\mathbf{q},\omega)$ is the Fourier transform of the transverse spin susceptibility, and ω is the electronic Zeeman frequency. The form factor $|A(\mathbf{q})|^2$ depends upon the site. For the planar ^{63}Cu nuclei, Mila and Rice[17] have proposed a hyperfine Hamiltonian which includes an isotropic transfer coupling B between the nuclear spin and the electron spins of the nearest-neighbor Cu atoms along with an on-site anisotropic coupling $A_{\alpha\alpha}$ ($A_{xx}=A_{yy} \neq A_{zz}$):

$$H_{\mathrm{hf}} = \sum_{\alpha} A_{\alpha\alpha} I^{\alpha} S_0^{\alpha} + \sum_{i=1}^{4} B\mathbf{I}\cdot\mathbf{S}_i. \quad (2)$$

We neglect the hyperfine coupling to the chain Cu spins. This hyperfine interaction leads to a Cu form factor

$$|A^{\mathrm{Cu}}(\mathbf{q})|^2/A_{zz}^2 = \begin{cases} [\frac{1}{2}(1-a_{xx})-4b\gamma_{\mathbf{q}}]^2+\frac{1}{4}(1+a_{xx})^3, & \mathbf{H}\parallel \mathbf{a},\mathbf{b}, \\ (a_{xx}+4b\gamma_{\mathbf{q}})^2, & \mathbf{H}\parallel \mathbf{c}, \end{cases} \quad (3)$$

where $a_{xx}=A_{xx}/|A_{zz}|$, $b=B/|A_{zz}|$, and $\gamma_{\mathbf{q}}=\frac{1}{2}(\cos q_x+\cos q_y)$. The values of the hyperfine factors are deduced from the Knight-shift measurements:[5,8,11,17] $A_{zz}/\gamma_n\hbar$ is reported to be about −400 kOe and $b=B/|A_{zz}|\approx 0.25$. There is more uncertainty in the value of $a=A_{xx}/|A_{zz}|$, and we have examined values ranging from 0.4 to small negative num-

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bers. For the planar ^{17}O nuclear spin, we will assume a simple isotropic near-neighbor Cu hyperfine coupling, $A_O \mathbf{I}_O \cdot (\mathbf{S}_1 + \mathbf{S}_2)$, so that the oxygen form factor is

$$|A_O(\mathbf{q})|^2/A_O^2 = \begin{cases} 4\cos^2(q_x/2), \\ 4\cos^2(q_y/2). \end{cases} \tag{4}$$

This vanishes for $(q_x,q_y)=(\pi,\pi)$, and the contributions of antiferromagnetic fluctuations to the nuclear spin relaxation of the ^{17}O nucleus are suppressed. The estimated value[18] of $A_O/\gamma_n \hbar$ is of order 120 kOe.

In order to model the dynamic spin susceptibility in a simple manner, we use an RPA expression for $\chi(\mathbf{q},\omega)$ obtained for a 2D Hubbard model:

$$\chi(\mathbf{q},\omega) = \frac{\chi_0(\mathbf{q},\omega)}{1 - U\chi_0(\mathbf{q},\omega)} \,. \tag{5}$$

Here U is the on-site Coulomb interaction and

$$\chi_0(\mathbf{q},\omega) = \frac{1}{N} \sum_{\mathbf{p}} \frac{f(\epsilon_{\mathbf{p}+\mathbf{q}}) - f(\epsilon_{\mathbf{p}})}{\omega - (\epsilon_{\mathbf{p}+\mathbf{q}} - \epsilon_{\mathbf{p}}) + i0^+} \,, \tag{6}$$

with $\epsilon_{\mathbf{p}} = -2t(\cos p_x + \cos p_y)$, where t is the hopping matrix element, and $f(\epsilon_{\mathbf{p}}) = (e^{\beta(\epsilon_{\mathbf{p}} - \mu)} + 1)^{-1}$ is the usual Fermi factor. The RPA gives an unphysical phase transition when $U\chi_0(\mathbf{q}^*,0)=1$, and here we avoid this by taking a band filling such that $U\chi_0(\mathbf{q},0)$ remains less than 1. Specifically, we take $U/t=2$ and consider band fillings $\langle n \rangle$ less than $n_c=0.865$. Our point of view is that U and $\langle n \rangle$ are simply parameters[19] which provide a means of tuning the RPA form for $\chi(\mathbf{q},\omega)$. Thus U is clearly a suitably renormalized effective repulsion rather than the bare Hubbard value, and the shift in the chemical potential μ to move $\langle n \rangle$ from 1 to below n_c sets the characteristic temperature T^*.

The Knight shifts depend upon the $\mathbf{q} \to 0$ susceptibility, which has the usual Stoner enhancement at low temperatures

$$\chi(\mathbf{q} \to 0, 0) = \frac{N(\mu)}{1 - UN(\mu)} \,. \tag{7}$$

Here $N(\mu)$ is the density of states at the chemical potential, $K[1-(\mu/4t)^2]^{1/2}/2\pi^2 t$, with K the complete elliptic integral. In the parameter region we are considering, the Stoner enhancement is between 1.5 and 2.

With the RPA form, Eq. (5), for $\chi(\mathbf{q},\omega)$, Eq. (1) then gives the nuclear relaxation rate as

$$T_1^{-1} = T \left\langle \frac{|A(\mathbf{p}-\mathbf{p}')|^2}{[1 - U\chi_0(\mathbf{p}-\mathbf{p}',0)]^2} \right\rangle , \tag{8}$$

where $\mathbf{p}$ and $\mathbf{p}' = \mathbf{p} - \mathbf{q}$ are averaged over the Fermi surface. Thus an enhancement of $(T_1 T K^2)^{-1}$, with K the Knight shift, for ^{63}Cu reflects the strong antiferromagnetic fluctuations,[1,6] which enter the expression Eq. (8) for T_1^{-1} but not the Knight shift, which is proportional to $\chi(\mathbf{q} \to 0, 0)$, Eq. (7).

Using the RPA expression for $\chi(\mathbf{q},\omega)$ and the ap-

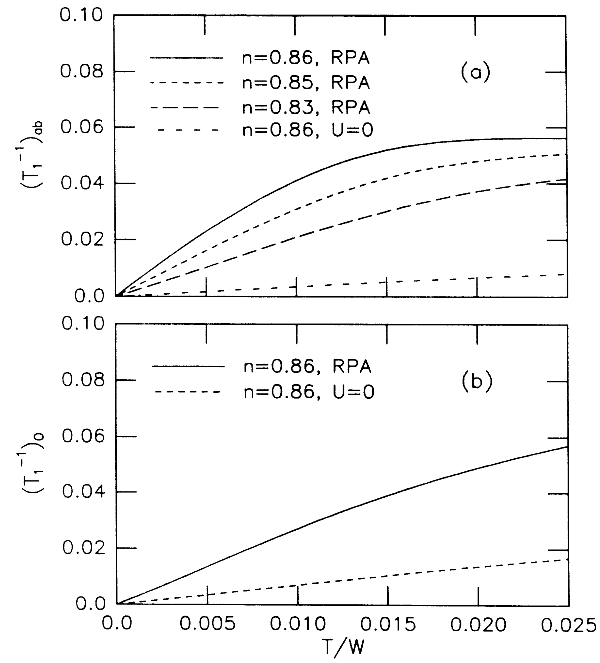

FIG. 1. T_1^{-1} vs T for $U/t=2$ and various values of $\langle n \rangle$. (a) Results for Cu in units of $\pi A_{zz}^2/\hbar t$ with **H** in the a-b plane and (b) for O in units of $\pi A_O^2/\hbar t$.

propriate form factors, we have evaluated T_1^{-1} for Cu and O. In Fig. 1 we show results obtained using $a=0.4$ and $b=0.25$ for $U/t=2$ and several values of $\langle n \rangle$. The temperature is measured in units of the bandwidth[20] $W=8t$. If the enhancement factor (within angular brackets) in Eq. (8) were temperature independent, then that equation would predict Korringa ($T_1^{-1} \propto T$) behavior. However, as $\langle n \rangle$ approaches n_c, $U\chi_0(\mathbf{q},0)$ becomes close to unity for some $\mathbf{q}$. Then even small changes in $\chi_0(\mathbf{q},0)$ with T have large impact on the enhancement of T_1^{-1}, as long as $|A(\mathbf{q})|^2$ is substantial near the same value of $\mathbf{q}$. The non-Korringa behavior seen for ^{63}Cu relaxation is due to the fact that the spectral weight of antiferromagnetic fluctuations saturates at T^* as T is lowered. Within the RPA,[21] the peak in $[1 - U\chi_0(\mathbf{q},0)]^{-2}$ is displaced[22] from (π,π) to (π,q^*) and (q^*,π). This both weakens the effect of these fluctuations at the ^{63}Cu nuclei, since the corresponding form factor $|A(\mathbf{q})|^2$ peaks at $\mathbf{q}=(\pi,\pi)$, and allows some leakage of these fluctuations onto the ^{17}O site, since $\cos^2(q_x/2)$ is finite for $q_x \neq \pi$. The latter gives some deviation of T_1^{-1} from linear behavior in O [see Fig. 1(b)]. But the effect for the same value of $\langle n \rangle = 0.86$ is much more dramatic for Cu, where T_1^{-1} is seen to saturate at high T.

Furthermore, we can make quantitative comparisons with the experimental results. Within our model it is possible to fit the experimental data in more than one way using different sets of parameters. For example, if we choose the Hubbard model parameters $U/t=2$, $\langle n \rangle = 0.86$, and the bandwidth $W \sim 1.2$ eV, one possible fit is provided by the hyperfine couplings $A_{zz}/\gamma_n \hbar$

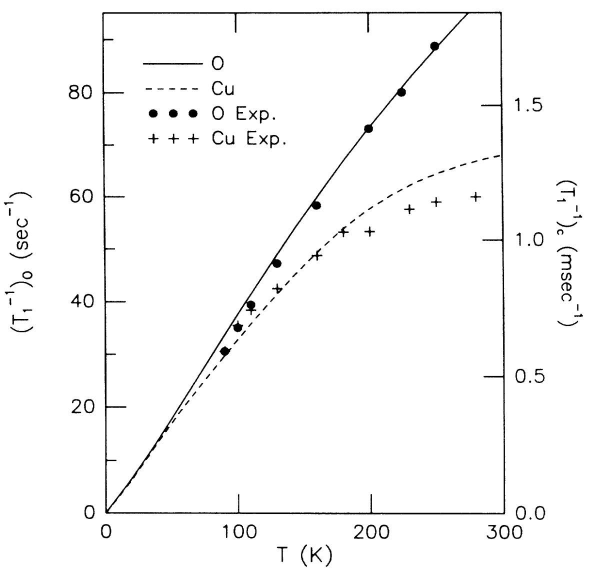

FIG. 2. A possible fit to the experimental data. The planar ^{63}Cu (dashed line, right axis) results have been obtained using the hyperfine couplings $A_{zz}/\gamma_n\hbar = -380$ kOe, $A_{xx}/\gamma_n\hbar = 80$ kOe, and $B/\gamma_n\hbar = 85$ kOe. For ^{17}O (solid line, left axis) we have used $A_O/\gamma_n\hbar = 110$ kOe. The Hubbard model parameters are $U/t = 2.0$, $\langle n\rangle = 0.86$, and $W = 1.2$ eV. The orienting magnetic field is taken in the c direction. Experimental points shown for $YBa_2Cu_3O_7$ are from Refs. 6 and 24.

$= -380$ kOe, $a = 0.2$, $b = 0.22$, and $A_O/\gamma_n\hbar = 110$ kOe as seen in Fig. 2.

Other important NMR features are a large anisotropy ratio $(T_1^{-1})_{ab}/(T_1^{-1})_c$ and a large enhancement of the Korringa ratio $(T_1TK^2)^{-1}$. For $a = 0.4$, $b = 0.25$, and $\langle n\rangle = 0.86$, we find that the ratio $(T_1^{-1})_{ab}/(T_1^{-1})_c$ varies linearly from 4.0 to 3.4 as the temperature is increased from $0.01W$ to $0.02W$. Smaller values for a yield a smaller anisotropy ratio. For example, using $\langle n\rangle = 0.86$, $a = 0.0$, and $b = 0.25$ we obtain $(T_1^{-1})_{ab}/(T_1^{-1})_c \sim 2.6$ with little temperature dependence. The RPA enhancement of the Korringa ratio $(T_1TK^2)^{-1}$ vs T for Cu and O is shown in Fig. 3 for various values of $\langle n\rangle$ with $a = 0.4$ and $b = 0.25$. For these parameters the enhancement of $(T_1TK^2)^{-1}$ is 4 at $T = 0.01W$. Using $a = 0.0$ and $b = 0.25$ one obtains an enhancement of about 5.5 at $T = 0.01W$. Experimentally, Hammel *et al.*[6] find an enhancement of $\frac{11}{2}$ for Cu at 100 K and a temperature-independent enhancement of order 1.4 for O. The large enhancement of the Korringa ratio for Cu implies significant antiferromagnetic fluctuations, which are modeled in the RPA by taking $\langle n\rangle$ near n_c. A measure of the strength of these fluctuations is the correlation length ξ. Defining ξ^{-1} to be the half width at half maximum of the peak in $\chi(\mathbf{q},0)$, we find the correlation length to be about four lattice spacings at $T = 0.01W$. However, at lower temperatures such a definition is not appropriate since the susceptibility is no longer symmetric around $\mathbf{q} = \mathbf{q}^*$. In fact,[23] as $T \to 0$ the noninteracting susceptibility $\chi_0(\mathbf{q},0)$ develops a square-root cusp at $\mathbf{q} = \mathbf{q}^*$, with an infinite slope on the high-$|\mathbf{q}|$ side.

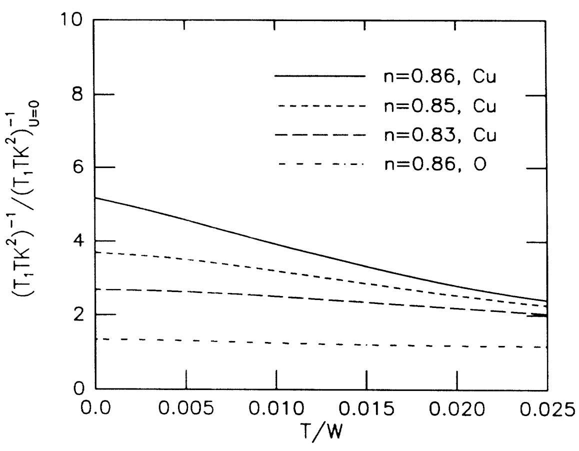

FIG. 3. Enhancement of the Korringa ratios for ^{63}Cu and ^{17}O with $\mathbf{H}$ in the a-b plane for various fillings $\langle n\rangle$ using $U/t = 2.0$, $a = 0.4$, and $b = 0.25$.

Using the same RPA form for $\chi(\mathbf{q},\omega)$, one can ask what the neutron scattering would give. The inelastic-neutron-scattering cross section is proportional to $[n(\omega)+1]\mathrm{Im}\chi(\mathbf{q},\omega)$, where $n(\omega)$ is the Bose occupation number. Integrating this over $q = q_x = q_y$ from $\pi/2$ to $3\pi/2$ for various values of ω/W gives results for the integrated intensity versus temperature shown in Fig. 4. Since $n(\omega)+1 \to k_BT/\omega$ for $\omega \ll k_BT$, the integrated intensity versus T at a Zeeman energy transfer should be similar to T_1^{-1} for Cu [Fig. 1(a)]. At larger values of ω, when k_BT decreases below $\hbar\omega$, $n(\omega)+1 \to 1$ and the intensity levels off. In this way, when $\hbar\omega$ exceeds the temperature at which T_1^{-1} saturates, one finds a relatively constant behavior versus T. The RPA results appear

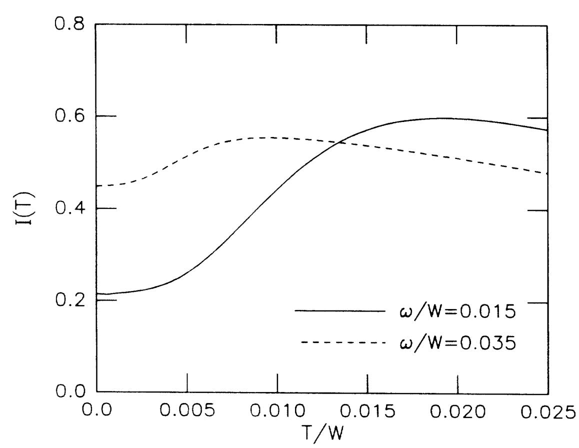

FIG. 4. Integrated inelastic scattering intensity vs T for $U/t = 2.0$, $\langle n\rangle = 0.86$, and different energy transfers ω.

similar to the features observed[9] in neutron-scattering experiments on $La_{2-x}Sr_xCuO_4$. Our interpretation of the data is that in these materials $\mathrm{Im}\chi(\mathbf{q},\omega)$ initially increases as T is lowered and then saturates at and below a characteristic temperature T^*. Thus we believe it is possible to interpret the neutron-scattering-intensity data without introducing a gap in the magnetic excitation spectrum.

From these results we conclude that it is possible to understand the observed differences in the NMR data on the Cu and O sites and the occurrence of structure in the integrated inelastic-scattering intensity within a one-component Fermi-liquid picture. However, it is clear that in order to see this we have had to push the RPA form very close to the spin-density-wave instability. In addition, $(T_1^{-1})_{\rm O}$ shows some deviation from a simple Korringa behavior. This latter feature may be associated with the failure of the RPA to generate correlations which peak closer to (π,π). Nevertheless, this simple approach suggests that a Fermi liquid with strong antiferromagnetic fluctuations may well provide an appropriate description of the normal state of the layered superconducting cuprates.

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