t-J Model and Nuclear Magnetic Relaxation in High-T_c Materials

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We obtain the Fermi contact term of the electron-nuclear effective Hamiltonian describing the coupling between the planar Cu(2) nucleus and the O(2,3) nuclei, on the one hand, and the effective degrees of freedom of the one-band t-J (or Heisenberg-Hubbard) model, on the other. The details of the resulting couplings suggest that the copper and oxygen relaxation rates are sensitive to antiferromagnetic correlations in different degrees. In the insulating phase we predict that the effective dynamical critical exponent for O(2,3) is reduced by two from that of Cu(2).

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Recent experiments[1-4] on nuclear relaxation in the 1:2:3 and 2:1:4 compounds have yielded a wealth of detailed data, and indicate[1,2] that the planar Cu(2) nuclear relaxation differs substantially in its temperature dependence from that of O(2) relaxation, at least in the superconducting materials above T_c. Below T_c, however, the two relaxation rates are similar, suggesting that there is a single species of fluctuating electronic spins relaxing the two neighboring nuclei. It is therefore surprising that above T_c one is seeing drastically different relaxation rates.

In this work we take the point of view, proposed by Anderson,[5] that there is an effective one-band description of the copper-oxide plane—namely the t-J (Heisenberg-Hubbard) model. The variables in this model are projected ones, obtained by eliminating many variables which are expected to be irrelevant in the low-energy sector of the problem. The NMR probes different bare degrees of freedom in the problem, and hence a difference in behavior of the Cu(2) and O(2) rates indicates that we may be able to test in a nontrivial sense the proposed projection.

These considerations have motivated this work. Our aim is to take a well defined projection of a declared set of bare variables to a one-band model, to add on to this the electron-nuclear interactions specific to Cu(2) and O(2,3) nuclei, and to examine whether these could derive from a single set of variables and yet display different behaviors within a controlled calculation. We find that the reduction proposed by Zhang and Rice[6] can be interpreted as a controlled projection—provided one uses a particular ratio of Fourier components of a certain hybridization function as an additional small parameter. We then extend the analysis of Mila and Rice[7] to the case of O(2,3), wherein we propose that the oxygen $3s^2$-like states mix with the active degrees of freedom. The picture that results is that the two nuclei experience contact with the same set of spins of the t-J model via two different well defined form factors. The differences then are argued as arising from these form factors. In the stoichiometric case (La_2CuO_4 or $YBa_2Cu_3O_6$), the present calculation predicts that the effective dynamical exponent for the oxygen nucleus is reduced relative to that of copper by two. In the doped case (say, $YBa_2Cu_3O_7$), we do not have a good justification for using a single-length scaling hypothesis for the dynamics. However, heuristically, such an *Ansatz* leads to interesting suggestions regarding spin correlations in the doped case as well.

We begin by writing the three-band model (in hole notation) with bare parameters in momentum space with $H=H_1+H_2$,

$$H_1=\epsilon_d\sum d^\dagger_{r\sigma}d_{r\sigma}+\epsilon_p\sum(p^{x\dagger}_{k\sigma}p^x_{k\sigma}+p^{y\dagger}_{k\sigma}p^y_{k\sigma})+U_d\sum n^d_{r\uparrow}n^d_{r\downarrow} \quad (1)$$

and

$$H_2=-(2i)V_{p\text{-}d}\sum d^\dagger_{k\sigma}[\sin(k_x/2)p^x_{k\sigma}+\sin(k_y/2)p^y_{k\sigma}]+\text{H.c.} \quad (2)$$

Here we have chosen to ignore several additional terms that may be added, such as U_p or $U_{p\text{-}d}$ which have been suggested in the literature,[8,9] in order to focus on the spin degrees of freedom, and will in fact set $U_d=\infty$ in order to bring out the qualitative feature of our argument. In common with Zhang and Rice, we will assume that the direct oxygen-oxygen overlap is small; specifically, the following analysis requires a two-sided inequality,

$$1\gg\frac{V_{p\text{-}d}}{\epsilon_p-\epsilon_d}\gg\frac{V_{\text{O-O}}}{V_{p\text{-}d}}, \quad (3)$$

to be fulfilled. The operators $p^x_{k\sigma}$, $p^y_{k\sigma}$, and $d_{k\sigma}$ are Fourier components of $p^x_\sigma(r+\hat{x}/2)$, $p^y_\sigma(r+\hat{y}/2)$, and $d_\sigma(r)$, which are defined at the two midpoints of the square lattice links, and the corners, respectively; the factors of $\sin(k_x/2)$, etc., are a consequence of the phase factors assumed in Ref. 6 and reflect the local "quantum chemistry." We shall write all hybridizations in k space for brevity. In order to proceed, we work with "canonical" fermions obtained by inspection from Eq. (2) as $\alpha_k=i(s_xp^x+s_yp^y)/\mu_k$ and $\beta_k=(-i)(s_yp^x-s_xp^y)/\mu_k$, where $s_x\equiv\sin(k_x/2)$, $s_y\equiv\sin(k_y/2)$, and $\mu_k\equiv\{s_x^2+s_y^2\}^{1/2}$ (we shall use these abbreviations elsewhere in this paper). The species of fermions α_k and β_k then are independent, and the β's do not mix with the d's, thereby

remaining at an energy ϵ_p and hence eliminated from the problem. Thus $H_2 = -2V_{p\text{-}d}\sum_k \mu_k (d^\dagger_{k\sigma} a_{k\sigma} + \text{H.c.})$. We can now use degenerate perturbation theory to mix the states of H_1 by various orders of H_2 and find $H_{\text{eff}} = H_3 + H_{\text{ex}}$, where

$$H_{\text{ex}} = J_{\text{ex}} \sum_{\langle ij \rangle} (\mathbf{S}_i \cdot \mathbf{S}_j + \tfrac{1}{4}) ,$$

with $J_{\text{ex}} = 32 V^4_{p\text{-}d}/(\epsilon_p - \epsilon_d)^3$, and

$$H_3 = [4V^2_{p\text{-}d}/(\epsilon_p - \epsilon_d)] \sum_{ijk} d^\dagger_{r_i\sigma_1} d_{r_i\sigma_2} \alpha^\dagger_{r_j\sigma_2} \alpha_{r_i\sigma_1} \hat{\mu}(r_i - r_j)\hat{\mu}(r_i - r_l) . \tag{4}$$

Here $\hat{\mu}(\mathbf{r})$ is the Fourier transform of μ_k, and as usual, one argues that H_{ex} is operative at the insulating limit and that H_3 comes into play when more holes are added. Now H_3 contains several terms all at the energy scale $V^2_{p\text{-}d}/(\epsilon_p - \epsilon_d)$ and it is not obvious as to whether a further reduction is possible. A somewhat similar, but formally clearer problem was considered by LaCroix,[10] who considered a Kondo lattice with coupling J_K between local moments and an otherwise free set of tight-binding electrons with hopping t_k in the limit of large J_K/t_k. Here one has a controlled expansion in which local singlets are formed by the Kondo coupling and the hopping term mixes this manifold within first order, leading to an effective Hamiltonian of the $U = \infty$ Hubbard model. The triplet manifold is at a higher energy $\sim J_K$ and ignorable provided $t_k/J_K \ll 1$, and the tightly bound singlets are describable as $U = \infty$, Hubbard-model holes. In the present problem we do not have an *a priori* small parameter until we examine the details of the function $\hat{\mu}(\mathbf{r})$. We first note that its values at distances (0,0), (1,0), (1,1), and (2,0) are, respectively, ~ 0.96, -0.15, -0.02, and -0.01. Hence we could use the ratio $\hat{\mu}(1,0)/\hat{\mu}(0,0) \sim 1/7$ to organize the term H_3 [Eq. (4)] into a Kondo-lattice-type form. Within this spirit we find $H_{\text{eff}} = H_{\text{kin}} + H_{\text{ex}}$, where

$$H_{\text{kin}} = -t_{\text{eff}} \sum_{\langle ij \rangle} P_d (c^\dagger_{i\sigma} c_{j\sigma} + c^\dagger_{j\sigma} c_{i\sigma}) P_d \tag{5}$$

and $t_{\text{eff}} = [2V^2_{p\text{-}d}/(\epsilon_p - \epsilon_d)][\hat{\mu}(0,0)\hat{\mu}(1,0)]$. Here the c's are the Fermi operators of the $U = \infty$ Hubbard model (P_d is the Gutzwiller projection operator). It should be mentioned that the purpose of this somewhat elaborate derivation of the t-J model is to stress the fact (implicit in Ref. 6) that the energy cost of breaking one of the singlets (i.e., unsublimating the oxygen hole spin) is $\sim 8 \times \hat{\mu}(0.0)/\hat{\mu}(1,0)$ times the effective hopping, which in turn cannot be logically allowed to be smaller than J_{ex} in this scheme. The scheme generates second-neighbor hops as well with $t' = [\mu(2,0)/\mu(1,0)]t_{\text{eff}}$, etc.

We now turn to the calculation of the Fermi contact term in the model. The basic hypothesis is that the electron-nuclear interaction term is $[\alpha_0 \equiv (8\pi/3) \mid \psi_{4s}(0) \mid^2 \times \gamma_n g \mu_B \hbar$ and $\beta_0 = (8\pi/3) \mid \psi_{3s}(0) \mid^2 \gamma_n g \mu_B \hbar]$

$$H_{e\text{-}n}(\text{Cu}(2)) = \tfrac{1}{2} \alpha_0 \sum_i I^\alpha_{r_i} a^\dagger_{r_i\sigma_1} \tau^\alpha_{\sigma_1\sigma_2} a_{r_i\sigma_2} , \tag{6}$$

$$H_{e\text{-}n}(\text{O}(2)) = \tfrac{1}{2} \beta_0 \sum_i J^\alpha_{r_i + \hat{x}/2} b^\dagger_{r_i + \hat{x}/2\sigma_1} \tau^\alpha_{\sigma_1\sigma_2} b_{r_i + \hat{x}/2\sigma_2} . \tag{7}$$

Here $\psi_{4s}(0)$ and $\psi_{3s}(0)$ refer to the Cu $4s$ wave function and the O $3s$ wave function at the respective nuclei and a,b are the corresponding field operators. The case of Cu(2) was considered previously by Mila and Rice[7] and the case of O(2) is new. In the electron picture, these ($4s$ and $3s$, respectively) s orbitals are the ones closest to the Fermi level with the correct symmetry to mix with the active orbitals, and the lower ones (i.e., Cu $3s$, $2s$, $1s$, and O $2s$, $1s$) are unable to contribute to the Fermi term due to the Pauli principle (they do so to the core polarization terms which are expected to be smaller in magnitude).

These orbitals a and b are, in the hole picture, doubly occupied and placed at a deeper energy, $\epsilon_s(\text{Cu})$ and $\epsilon_s(\text{O})$, respectively. These have nontrivial dynamics only because of the hybridization with the active orbitals, which we now write down directly in k space:

$$H^{(1)}_a \equiv -2V(4s \mid 2p) \sum_k a^\dagger_{k\sigma} [(s^2_x - s^2_y)\alpha_{k\sigma} + (2s_x s_y)\beta_{k\sigma}]/\mu_k + \text{H.c.} , \tag{8}$$

$$H^{(2)}_a \equiv -2V(4s \mid 3d) \sum_k (a^\dagger_{k\sigma} d_{k\sigma} + \text{H.c.})(\cos k_y - \cos k_x) , \tag{9}$$

and

$$H_b = -(2i)V(3s \mid 3d) \sum_k s_x (b^\dagger_{k\sigma} d_{k\sigma} - \text{H.c.}) . \tag{10}$$

Here $V(4s \mid 2p)$ and $V(4s \mid 3d)$ are the Cu-$4s$-O-$2p$ and Cu-$4s$-next-neighbor-Cu-$3d$ overlaps and $V(3s \mid 3d)$ is the O-$3s$-Cu-$3d$ overlap. In addition to these, we add $\epsilon_s(\text{O}) \sum b^\dagger_{k\sigma} b_{k\sigma} + \epsilon_s(\text{Cu}) \sum a^\dagger_{k\sigma} a_{k\sigma}$ to the full Hamiltonian. These three hybridization terms arise by writing the possible overlaps in the sense of Refs. 6 and 7, and rewriting in k space using the canonical fermions α and β. We now use the small ratios $V(4s \mid 2p)/[\epsilon_p - \epsilon_s(\text{Cu})]$, etc., as expansion parameters to leading order. First we ignore the coupling of a's to β's in (8) since the β's are the passive orbitals. The calculation is illustrated in the case of $H^{(1)}_a$. We first hybridize a's and α's at a fixed k value from Eq. (8) and the site energies of a's and α's [i.e., $\epsilon_s(\text{Cu})$ and ϵ_p] to find two new canonical fermions called $\hat{a}$ and $\hat{\alpha}$ (which are a- and α-like for small mixing). We may invert the relation and to lowest order find

$$a_{k\sigma} = \hat{a}_{k\sigma} - \frac{2V(4s \mid 2p)}{\epsilon_p - \epsilon_s(\text{Cu})} (s^2_x - s^2_y) \frac{1}{\mu_k} \hat{\alpha}_{k\sigma} . \tag{11}$$

The levels $\hat{a}$ are now assumed to be inert (i.e., doubly oc-

cupied), and hence dropped, and the distinction between $\hat{\alpha}$ and α ignored in the mixing Hamiltonian H_2 to lowest order. This leads to the effective Hamiltonian that we write down finally (truncating various terms to their leading nonvanishing orders),

$$H_{e\text{-}n}(\mathrm{Cu}(2)) = \alpha_1 \sum_{r,\delta} \tfrac{1}{2}\, \sigma_r \cdot \mathbf{I}_{r+\delta} + \alpha_2 \sum_{r,\delta} \mathbf{S}_r \cdot \mathbf{I}_{r+\delta}, \quad (12)$$

$$H_{e\text{-}n}(\mathrm{O}(2)) = \alpha_3 \sum_{r} \mathbf{J}_{r+\hat{x}/2} \cdot (\mathbf{S}_r + \mathbf{S}_{r+\hat{x}}). \quad (13)$$

Here δ is the nearest-neighbor vector, and the constants are

$$\alpha_1 = \alpha_0 \{V(4s \mid 2p)/[\epsilon_p - \epsilon_s(\mathrm{Cu})]\}^2 \Delta,$$

$$\alpha_2 = 4\alpha_0 \{V(4s \mid 3d)/[\epsilon_d - \epsilon_s(\mathrm{Cu})]\}^2,$$

and

$$\alpha_3 = \beta_0 \{V(3s \mid 3d)/[\epsilon_d - \epsilon_s(\mathrm{O})]\}^2,$$

with

$$\Delta^{1/2} \equiv \langle \cos k_x (\cos k_x - \cos k_y)/\mu_k \rangle,$$

the bracket denoting average over the Brillouin zone. To make contact with Ref. 7, we note that the second term in Eq. (12) is of the same nature as the one found by the authors; however, they do not have the analog of the first term (i.e., the coefficient of α_1). All of these terms represent "transferred hyperfine couplings," in the sense that every nucleus senses the spins on neighboring sites.

In order to proceed further, we note that within the t-J model, the oxygen hole spins are exhausted by singlet formation (i.e., σ's are not available as free variables) and hence the effective coupling is exactly as in Eqs. (12) and (13) except that the term multiplying α_1 should be ignored.

We next note that the relaxation rates obtained from these terms follow from standard arguments and lead to

$$\left.\frac{1}{T_1}\right|_{\mathrm{Cu}(2)} \propto k_B T \sum_q \frac{\chi_q''(\omega_0)}{\omega_0} (\cos q_x + \cos q_y)^2 \quad (14)$$

and

$$\left.\frac{1}{T_1}\right|_{\mathrm{O}(2)} \propto k_B T \sum_q \frac{\chi_q''(\omega_0)}{\omega_0} (1 + \cos q_x). \quad (15)$$

Here $\chi_q''(\omega_0)$ is the imaginary part of the t-J-model spin susceptibility at nuclear frequency ω_0 (assumed to be very small). The two rates thus differ in the two form factors multiplying the susceptibility.

We first consider the insulating case, where it is clear from considerable neutron scattering data that the paramagnetic state ($T > T_N$) possesses considerable 2D correlations described by a long correlation length driving the dynamics. The σ-model analysis of Refs. 11 and 12 gives a detailed description of the paramagnetic phase that is assumed to be governed by a zero-temperature critical point. The dynamical scaling hypothesis[13] says that in the scaling regime, the relevant susceptibility given by $\chi_q''(\omega_0)/\pi\omega_0 = \chi_q/\Gamma_q$, where Γ_q is some characteristic frequency, has a singular term $\chi_q \sim \xi^{2-\eta} f(\hat{q}\xi)$ and $\Gamma_q^{-1} = \xi^z h(\hat{q}\zeta)$, where $\hat{q}$ is the wave vector relative to the dominant value of $q = (\pi,\pi)$, f and h are appropriate scaling functions, η is the anomalous dimension, and z is the dynamical critical exponent ($z = 1$ in the present case, Refs. 12 and 13). It is clear from the form factors in (14) and (15) that the behavior is very different since in the proximity of (π,π) the function $\cos q_x + \cos q_y \sim -2$ whereas $\cos q_x + 1 \sim -\hat{q}_x^2/2$. The second-degree node of the oxygen form factor leads, via standard arguments (as in Ref. 13), to the following prediction:

$$z_{\mathrm{O}}^{\mathrm{eff}} = z_{\mathrm{Cu}}^{\mathrm{eff}} - 2. \quad (16)$$

With $z_{\mathrm{Cu}}^{\mathrm{eff}} = 1$, this implies that the momentum-space integral involved[14] in evaluating $1/T_1$/oxygen is convergent, and hence we should expect a striking difference between the behavior of the relaxation rates in Cu(2) and O(2) in this case. As an oversimplified but explicit functional form, one may take a classical spherical model to describe the susceptibility as

$$\chi_q = (k_B T)^{-1} C_q = [\chi_s^{-1} + 2J\gamma_q + 4J]^{-1},$$

where $\gamma_q \equiv \cos q_x + \cos q_y$. The staggered susceptibility χ_s is obtained by using the sum rule $\langle C_q \rangle_q = \frac{1}{3} S(S+1) = \frac{1}{4}$ and goes at low temperature as $\chi_s \sim (1/32J) \times \exp(\pi J\beta)$. The characteristic frequency may be crudely estimated[15] as $\Gamma_q \sim (4\pi \langle \omega^2 \rangle_q)^{1/2}$, where the second moment is

$$\langle \omega^2 \rangle_q = J^2 (2 - \gamma_q)(2 + \kappa^2/2 - 2t)(2 + \kappa^2/2 + \gamma_q),$$

with $\kappa^2 \equiv \chi_s^{-1} J^{-1}$ and $t \equiv k_B T/J$. This leads to a $1/T_1$ relaxation rate for Cu(2) that is proportional to $T\xi$, with $\xi \sim \chi_s^{1/2} \sim e^{\pi J\beta/2}$, and hence diverging at low temperature, but a rate for O(2) that is essentially proportional to $k_B T$. A quantitative measure of the spin fluctuations is provided by an interesting dimensionless ratio defined as

$$S(T) = \lim_{\omega \to 0} \frac{1}{\pi} \frac{\langle f_q \chi_q''(\omega)/\omega \rangle_q}{[\chi_0(0)]^2}, \quad (17)$$

where f_q is the form factor normalized to unity at $q = 0$. S is akin to the Korringa ratio and equals unity (for $T \ll T_F$) in ideal metals. Although $S(T)$ is not usually discussed in insulating systems, it is instructive to compute it within the simple scheme indicated above. We find S_{Cu} varying by several orders of magnitude $\{S_{\mathrm{Cu}} \sim (2/\pi^3)^{1/2} [\exp(\pi/2t)]/(1-t)^{1/2}$ for $t \leq 0.5\}$, whereas S_{O} is slowly varying around unity ($S_{\mathrm{O}} \sim 1.11 + 0.67t$ for $t \leq 0.5$).

The more elaborate scaling theory in Refs. 11 and 12 would lead to qualitatively similar results but with different values for exponents, etc. We will not attempt to present these details at this stage. It would be interesting to look for such differences in the insulating materials above the 3D ordering temperatures.

Turning to the doped cases, such as $YBa_2Cu_3O_7$, the present scheme should, in principle, be applicable provided the dynamical susceptibility χ_q'' is calculated within the t-J model, i.e., provided the spin fluctuations are relaxed by exchange terms as well as by hole hopping terms. In the absence of a reliable detailed calculation of χ'', it seems worthwhile to invert the question, and to ask whether the relaxation data could be used to infer some property of the spin relaxation, at least phenomenologically. In order to do so, we are tempted to make an extra hypothesis, that spin fluctuations continue to be dominated by a single "long" correlation length, and that the susceptibility is maximum at some value of the wave vector $\mathbf{Q}_0$. We emphasize that such an *Ansatz* is not *a priori* justifiable since there is no compelling experimental or theoretical reason to believe in a magnetic critical point in any physical temperature in the doped cases. However, the resulting picture is suggestive and the analysis may be regarded as heuristic asymptotic behavior.

Assuming that a long correlation length dominates the spin response, we now attempt to reconcile the data[1,2] with our rate formulas. First, the marked departure from Korringa linearity (with respect to T) of the Cu(2) relaxation and the linear relaxation of O(2) can only be understood in this framework provided the wave vector $\mathbf{Q}_0$ is not too far from (π,π); specifically, if we assume $|\mathbf{Q}_0-(\pi,\pi)|\xi$ is of order unity then the suppression of the O(2) relaxation continues to hold. Second, if the Cu(2) relaxation rate is fitted by $1/T_1 \propto T\xi^p$ (with an expectation $p=1$) then we find that the temperature dependence of the correlation length required is $\xi^p \sim \xi_0^p[T_A/(T+300\text{ K})]^{1.5}$ in order to fit the data of Refs. 1 and 2 on $YBa_2Cu_3O_7$ with ξ_0 and T_A undetermined by the fit. This form suggests that the Cu(2) rate may be regarded as crossing over from a linear to a sublinear behavior roughly around $\sim$150 K. The appearance of the characteristic temperature (300 K) cutting off the zero-temperature divergence has a possible interpretation[16,17] in terms of the hole kinetic energy removing the most singular antiferromagnetic fluctuations. The internal consistency of the argument would require the correlation length to be of the order of at least 10 Å.

Third, we should note that the normalized relaxation rates of Cu(2) and O(2), as in Eq. (17), should be numerically quite different, with Cu(2) considerably larger [since the integrand is large for $q\sim(\pi,\pi)$]. The identical temperature dependence of Cu and O rates in the superconducting state would then imply that the susceptibility does not have a residual peak near (π,π) in the superconducting state.

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