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Theoretical study of effective magnetic interactions in high-T_c Cu oxides

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Starting from a two-band Hubbard model for cupric oxides and using perturbation theory, we study the effect of a small O-O hopping term t_p on the parameters of the single-band t-J model. It is found that as long as the wave function on the oxygen site is extended and Bloch-like, the antiferromagnetic coupling between the nearest-neighbor copper spins decreases rapidly with increasing hole doping. This behavior seems to be insensitive to the value of t_p and dispersion of the oxygen band and is consistent with the results of recent experiments. We also show that, between next-nearest-neighbor copper spins, there exists a non-negligible antiferromagnetic coupling even in the zero-doping limit.

One of the most important questions in the high-T_c superconductors is the effect of hole doping on the superexchange interaction J between copper spins. Recent experiments[1,2] have shown that the effective J decreases rapidly with doping. In particular, Johnston[1] has shown that for $La_{2-x}SrCuO_{4-y}$ the contribution to the magnetic susceptibility due to Cu spins in the presence of holes is the same as that without holes except for much reduced values of J and the magnetic moment. At 20% of hole concentration, J is already reduced almost 1 order of magnitude from its maximum value at half-filling. In this paper we shall present a possible explanation of this surprising result.

It is now generally accepted that even though the doped holes reside on the oxygen sites, the two-band model[3] is equivalent to the t-J model in a certain parameter region.[4-6] Zhang and Rice[4] calculated the coupling constant J by using the localized wave function of the unoccupied oxygen state. The direct oxygen-oxygen hopping t_p was completely neglected. However, recent band-structure calculations[7,8] have consistently obtained a value of $\bar{t}_p \leq 0.5$ eV for the effective O-O hopping. Hence the wave function of the hole should be quite extended. It will be shown below that the phase of the wave function and its oscillating behavior cause the rapid decrease of J by doping.

Our goal is to follow Zhang and Rice[4] and reproduce the t-J model from the two-band model which takes into account oxygen-oxygen hopping. In order to accomplish this in a simple perturbative approach, we must confine $8t_p \ll \Delta$, with $\Delta = \varepsilon_p - \varepsilon_d$ as the energy difference between $2p$(O) and $3d$(Cu) levels. Later it will be shown that our main conclusion is insensitive to the magnitude of t_p.

Let us start with the following two-band Hubbard model[3]

$$H = \varepsilon_d \sum_{i,\sigma} d_{i\sigma}^{\dagger} d_{i\sigma} + \varepsilon_p \sum_{l,\sigma} p_{l\sigma}^{\dagger} p_{l\sigma} - t_p \sum_{\langle l,l' \rangle,\sigma} (p_{l\sigma}^{\dagger} p_{l'\sigma} + \text{H.c.}) + U_d \sum_i d_{i\uparrow}^{\dagger} d_{i\uparrow} d_{i\downarrow}^{\dagger} d_{i\downarrow} + H_I , \quad (1)$$

$$H_I = -t_0 \sum_{i,\sigma} \sum_{l \in i} (d_{i\sigma}^{\dagger} p_{l\sigma} + \text{H.c.}) , \quad (2)$$

where $d_{i\sigma}^{\dagger}$ creates a Cu hole, and $p_{l\sigma}^{\dagger}$ creates an O hole. U_d is the Cu on-site Coulomb repulsion. ε_d and ε_p are, respectively, the atomic-energy levels for Cu and O holes. t_p is the direct (or bare) O-O hopping matrix and t_0 is the Cu-O hybridization. The O-hole Brillouin zone can be divided into two parts,[9]

$$p_{l\sigma} = \frac{1}{(2N)^{1/2}} \left(\sum_{k,\sigma} e^{i\mathbf{k}\cdot\mathbf{R}_l} p_{k\sigma} + \sum_{k,\sigma} e^{i(\mathbf{k}+\mathbf{K})\cdot\mathbf{R}_l} p_{k+K,\sigma} \right)$$

$$= \frac{1}{(2N)^{1/2}} \sum_{k,\sigma} e^{i\mathbf{k}\cdot\mathbf{R}_l} (p_{k\sigma} \pm p_{k+K,\sigma}) , \quad (3)$$

where $\mathbf{K} = 2\pi\hat{\mathbf{x}}$ and the summation over $\mathbf{k}$ in Eq. (3) is confined to the Cu-hole Brillouin zone. i represents the site of a Cu atom and l is the site of its neighboring O atom. The "+" sign corresponds to $l = i \pm \hat{\mathbf{y}}/2$ and the "−" sign to $l = i \pm \hat{\mathbf{x}}/2$, where $\hat{\mathbf{x}}$ and $\hat{\mathbf{y}}$ denote, respectively, the unit vectors along the x and y directions, and N is the Cu-site number. We can define the symmetric state $P_{i\sigma}$ from the four oxygen-hole states around a Cu ion by[4]

$$P_{i\sigma} = \tfrac{1}{2} \sum_{l \in i} p_{l\sigma} = \frac{1}{N^{1/2}} \sum_{k,\sigma} \beta_k^{-1} P_{k\sigma} e^{i\mathbf{k}\cdot\mathbf{R}_i} , \quad (4)$$

with

$$\beta_k = \left(\cos^2 \frac{k_x}{2} + \cos^2 \frac{k_y}{2} \right)^{-1/2} . \quad (5)$$

We notice that the local states $P_{i\sigma}$ are not orthogonal to each other for the neighboring sites, but the $P_{k\sigma}$ are orthogonal operators in the k space due to additional factor β_k^{-1} in Eq. (4). Here the symmetric states (or bonding orbitals) $P_{i\sigma}$ coupling directly to Cu ions are made from half of the total states in the O-hole Brillouin zone. The other half states in the brillouin zone that do not couple to Cu ions are referred to as the nonbonding states. To compare Eq. (3) with Eq. (4), we introduce respectively, the bonding and the nonbonding operators in k space:

$$P_{k\sigma}=\frac{1}{\sqrt{2}}\beta_k\left[\left(\cos\frac{k_x}{2}+\cos\frac{k_y}{2}\right)p_{k\sigma}-\left(\cos\frac{k_x}{2}-\cos\frac{k_y}{2}\right)p_{k+K,\sigma}\right], \tag{6}$$

$$Q_{k\sigma}=\frac{1}{\sqrt{2}}\beta_k\left[\left(\cos\frac{k_x}{2}-\cos\frac{k_y}{2}\right)p_{k\sigma}+\left(\cos\frac{k_x}{2}+\cos\frac{k_y}{2}\right)p_{k+K,\sigma}\right]. \tag{7}$$

In terms of these operators, the Hamiltonian in Eqs. (1) and (2) can be rewritten as

$$\begin{aligned}H=&\varepsilon_d\sum_{i,\sigma}d_{i\sigma}^{\dagger}d_{i\sigma}+\varepsilon_p\sum_{k,\sigma}(P_{k\sigma}^{\dagger}P_{k\sigma}+Q_{k\sigma}^{\dagger}Q_{k\sigma})\\&-4t_p\sum_{k,\sigma}\cos\frac{k_x}{2}\cos\frac{k_y}{2}\left[2\cos\frac{k_x}{2}\cos\frac{k_y}{2}(P_{k\sigma}^{\dagger}P_{k\sigma}-Q_{k\sigma}^{\dagger}Q_{k\sigma})+\left(\cos^2\frac{k_x}{2}-\cos^2\frac{k_y}{2}\right)(P_{k\sigma}^{\dagger}Q_{k\sigma}+\text{H.c.})\right]\\&+U_d\sum_i d_{i\uparrow}^{\dagger}d_{i\uparrow}d_{i\downarrow}^{\dagger}d_{i\downarrow}+H_I,\end{aligned} \tag{8}$$

$$H_I=-\frac{2t_0}{\sqrt{N}}\sum_{i,k,\sigma}(\beta_k^{-1}e^{-i\mathbf{k}\cdot\mathbf{R}_i}P_{k\sigma}^{\dagger}d_{i\sigma}+\text{H.c.}). \tag{9}$$

It should be noted that the hybridization term H_I given in Eq. (9) connects the copper sites only with the bonding states of O holes. Although the nonbonding states described by the operators $Q_{k\sigma}$ in Eq. (8) are coupled to the bonding states $P_{k\sigma}$ through a small t_p term, they have no direct coupling with Cu ions. In the case of finite doping, the holes will first occupy the bonding states because of their lower energies as compared with those of the nonbonding states [see Eq. (8)]. It is easy to see from Eq. (8) that the low-energy bonding states with $k_x \ll 1$ and $k_y \ll 1$ have very small overlap with the nonbonding states. The contribution from the nonbonding states will thus be neglected below.

It is necessary to assume $\Delta-8t_p \gg 2\sqrt{2}t_0$ to ensure that the above Hamiltonian has an insulating ground state in the absence of O holes. Using this assumption and the perturbation theory, we obtain the second- and the fourth-order effective Hamiltonian from Eqs. (8) and (9),

$$\begin{aligned}H_{\text{eff}}^{(2)}=&-(2t_0)^2\sum_{k\sigma}\frac{1}{\Delta_k}(1-\beta_k^{-2})P_{k\sigma}^{\dagger}P_{k\sigma}+\frac{(2t_0)^2}{N}\sum_{i,\sigma}\sum_{k,k'}e^{i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{R}_i}\beta_k^{-1}\beta_{k'}^{-1}\left(\frac{1}{\Delta_{k'}}+\frac{1}{U_d-\Delta_k}\right)\\&\times(d_{i\sigma}^{\dagger}d_{i-\sigma}P_{k'-\sigma}^{\dagger}P_{k\sigma}-d_{i\sigma}^{\dagger}d_{i\sigma}P_{k'-\sigma}^{\dagger}P_{k-\sigma}),\end{aligned} \tag{10}$$

$$H_{\text{eff}}^{(4)}=\sum_{i,j}J(\mathbf{R}_{ij})S_i\cdot S_j, \tag{11}$$

with

$$\begin{aligned}J(\mathbf{R}_{ij})=&\frac{(2t_0)^4}{N}\sum_{k,k'}\sum_{\sigma,\sigma'}e^{i(\mathbf{k}-\mathbf{k}')\cdot(\mathbf{R}_i-\mathbf{R}_j)}\beta_k^{-2}\beta_{k'}^{-2}\left[\frac{1}{\Delta_k\Delta_{k'}}\left(\frac{1}{U_d}+\frac{1}{\Delta_k}\right)(1-f_{k\sigma})(1-f_{k'\sigma'})\right.\\&\left.-\frac{1}{U_d\Delta_{k'}(U_d-\Delta_k)}f_{k\sigma}(1-f_{k'\sigma'})\right],\end{aligned} \tag{12}$$

where $\Delta_k=\Delta+\varepsilon_k$ with

$$\varepsilon_k=-8t_p\cos^2\frac{k_x}{2}\cos^2\frac{k_y}{2}.$$

$f_{k\sigma}$ is the hole distribution function.

Employing the procedure of Ref. 4 and recognizing that the second term of $H_{\text{eff}}^{(2)}$ in Eq. (10) produces a very large splitting between singlet and triplet states as discussed in detail by Shen and Ting,[6] one obtains the effective single-band Hamiltonian describing Cu-Cu hopping between the singlet states:

$$H^{(2)}=\sum_{i\neq j}t_{ij}d_{i\sigma}^{\dagger}d_{j\sigma}, \tag{13}$$

where t_{ij} is the effective hopping integral between the singlet states, which can be calculated from the following equation similar to that in Ref. 4 at the $U_d\rightarrow\infty$ limit

$$\begin{aligned}t_{ij}=&-\frac{(2t_0)^2}{N}\sum_k e^{-i\mathbf{k}\cdot(\mathbf{R}_i-\mathbf{R}_j)}\left(\lambda\beta_k^{-1}\frac{1}{\Delta_k}+\beta_k^{-1}\frac{1}{N}\sum_{k'}\beta_{k'}^{-1}\frac{1}{\Delta_{k'}}-\frac{1}{2\Delta_k}(\beta_k^{-2}-1)+\frac{1}{2\Delta}\right)\\&-\frac{4t_p}{N}\sum_k e^{-i\mathbf{k}\cdot(\mathbf{R}_i-\mathbf{R}_j)}\cos^2\frac{k_x}{2}\cos^2\frac{k_y}{2}.\end{aligned} \tag{14}$$

Here $\lambda=0.96$. If $t_p=0$ is put in the above equation, one obtains $t_{ij}=t=-0.57$ and $t_{ij}=t'=0.18$ in units of t_0^2/Δ for the nearest neighbor $\mathbf{R}_{ij}=\mathbf{R}_i-\mathbf{R}_j=(1,0)$ and for the next-nearest neighbor $\mathbf{R}_{ij}=(1,1)$, respectively. These results are consistent with those obtained by Zhang and Rice[4] at the large-U_d limit. In fact, the effective O-O hopping term $\bar{t}_p=t_p+t_R\leq 0.5$ eV (Refs. 7 and 8) includes two parts: one part (t_p) comes from the bare (or direct) O-O hopping term, and another part (t_R) from the indirect hopping which corresponds to the first term on the right-hand side of Eq. (10). If one takes the bare hopping term $t_p=0.2$ eV and $\Delta=4$ eV, one gets $t=-0.67$ and $t'=0.13$. It is clear that the dominant hopping process is between the nearest-neighbor sites, but the next-nearest-neighbor hopping term is obviously not negligible. It should be noted that t_{ij} has different signs for the nearest-neighbor and the next-nearest-neighbor hoppings. As expected, the above results indicate that the kinetic energy of the doped hole is increased by the direct O-O hopping term.

Let us now discuss the interaction J in Eq. (12). The second term inside the large brackets in Eq. (12) corresponds to part of the so-called Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction obtained in the Fermi-liquid treatment of Si, Lu, and Levin[10] if one sets the phase factors $\beta_k=\beta_{k'}=1$. In the local picture, where we take $t_p=0$, this term represents the ferromagnetic interaction[6] between the nearest-neighbor Cu spins while a hole is occupying the oxygen site in the middle. It is usually thought that this term is responsible for the strong reduction of J by doping.[6] But because $U_d\gg\Delta_k$, this term is, in fact, negligible compared to the first term in Eq. (12) which in the local picture is exactly the superexchange interaction J.

It should be noticed that there is another RKKY interaction to be obtained from the square of the second term in $H_{\rm eff}^{(2)}$. This term with $\beta_k=\beta_{k'}=1$ as shown by Si *et al.*[10] produces a strong ferromagnetic interaction; but by taking into account the large energy difference between triplet and singlet states as discussed above, we will no longer have this kind of RKKY interaction. Far away Cu spins only see the spinless singlet, while nearest-neighbor ferromagnetic correlation is adequately taken into account by the formation of the singlet Wannier state. Because t_p is much smaller than the energy difference between the triplet and singlet states, the Kondo behavior will not occur here. At this stage we wish to emphasize that the primary difference between $J(\mathbf{R}_{ij})$ in Eq. (12) and that of Ref. 10 is the existence of the phase factors β_k and $\beta_{k'}$ in Eq. (12). It is exactly because of these phase factors, $J(\mathbf{R}_{ij})$ in the local picture could reduce back to the result of Ref. 6.

The first term in Eq. (12) represents the antiferromagnetic interaction when $t_p=0$. Assuming f_k to be the Fermi-Dirac distribution function and evaluating $J(\mathbf{R}_{ij})$ for nearest neighbors in the limit $U_d\rightarrow\infty$ and $t_p/\Delta=0.05$, J in unit of $(4t_0^4/\Delta^3)$ is plotted (the solid curve) as a function of the hole concentration δ in Fig. 1. In our calculations, the parabolic dispersion for the oxygen band has been employed to avoid large-scale computational problems. If we take the local limit $t_p=0$, and replace $1-f_{k\sigma}$ by $1-\delta$ to represent the effect of finite-hole density, this result is represented by the dot-dashed curve in Fig. 1. To understand this result further, we have calculated J by taking $t_p=0$ or $\Delta_k=\Delta$ which still assumes the existence of a Fermi surface due to the indirect oxygen-oxygen hopping via the neighboring Cu sites. The strong reduction of J by doping is again obtained as shown by the dotted curve in Fig. 1. For comparison we have used values of t_0^4/Δ^3 for the dotted and dot-dashed curves that are different from the solid curve so that at $\delta=0$ all cases have same value of J.

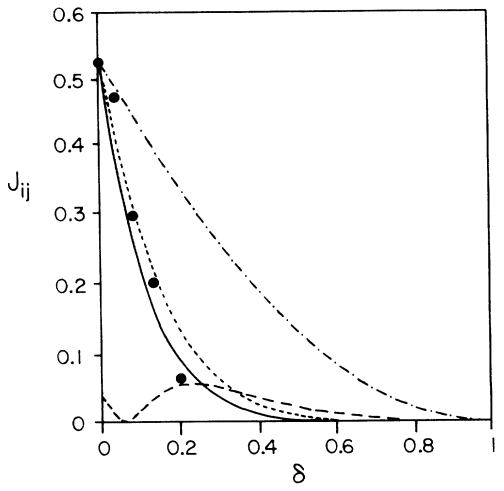

FIG. 1. The doping dependences of the superexchange coupling between the Cu spins. Here the solid line is the nearest-neighbor coupling J, and the dashed line corresponding to the next-nearest-neighbor coupling J'. The dotted line is the result of J when $t_p=0$. The dot-dashed line represents the nearest-neighbor coupling $J_{\rm loc}$ at local limit and the solid circles indicate the measurement for the nearest-neighbor coupling from Ref. 1.

For comparison we have also shown the experimental results of Johnston[1] by solid circles in Fig. 1. The agreement between the present result and the experiment is quite astonishing considering the fact that Eq. (12) is derived in the lowest-order perturbation theory. It should be noted that this agreement by itself is not to be taken as the support of a Fermi-liquid solution for the hole since dynamical effects not included in our calculation may play an important role.

The results discussed above seem to suggest that the existence of a sharp Fermi surface for the hole is the main reason for the rapid drop of J when the hole density is increased slightly. To verify this idea, we have also calculated J by replacing the Fermi function $f_{k\sigma}=\theta(\varepsilon_F-\varepsilon_k)$ in Eq. (12) by a more smooth varying function $f_{k\sigma}=\{\exp[(\varepsilon_k-\varepsilon_F)/\alpha]+1\}^{-1}$. The parameters α and ε_F are related by the condition that the hole density is δ. In Fig. 2, J is plotted as a function of δ for several different values of α, and f_k as a function of ε_k for $\delta=0.2$ is also displayed in the inset. It is quite clear that smoothing out the Fermi surface would indeed reduce the effect of doping upon J. But a reduction of J beyond the value obtained from the local approach is still expected.

In a much more sophisticated treatment we would in-

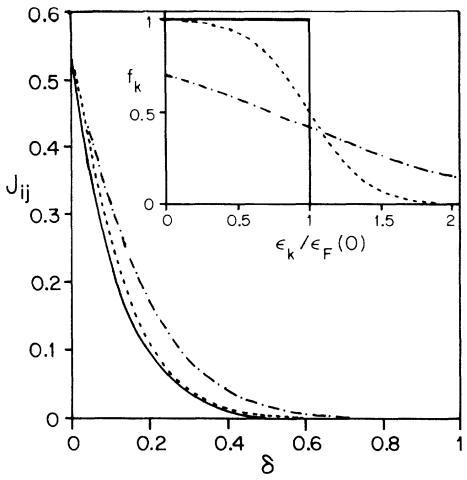

FIG. 2. The doping dependences of the nearest-neighbor coupling J between Cu spins which is calculated from distribution function $f_k = \{\exp[(\varepsilon_k - \varepsilon_F)/\alpha + 1]\}^{-1}$. In the inset the distribution function f_k is plotted as a function ε_k with $\delta = 0.2$.

clude that effect of singlet formation on those unoccupied oxygen states. This presumably would cause self-energy renormalization of Δ_k and phase shifts. But if a Fermi surface is still present, our result should be unchanged.

We have also calculated the next-nearest-neighbor superexchange interaction J' between Cu spins, it is shown as the dashed line in Fig. 1. This interaction is also antiferromagnetic. We wish to point out here (not shown in Fig. 1) that if taking $t_p = 0$, then $J' = 0$ at $\delta = 0$ and J' increases with increasing δ in the small doping limit. The presence of J' could help to destroy the long-range antiferromagnetic order as discussed by Doniach *et al.*[11] More interestingly, it could help to stabilize the parity and time reversal symmetry breaking state—the chiral spin state as discussed by Wen, Wilczek, and Zee.[12] Therefore, from the above results we construct the t-t'-J-J' Hamiltonian

$$H_{\text{eff}} = -\sum_{\langle n,n\rangle\sigma} t_{ij} d^{\dagger}_{i\sigma} d_{j\sigma} + \sum_{\langle n,n,n\rangle\sigma} t'_{ik} d^{\dagger}_{i\sigma} d_{k\sigma} + \sum_{\langle n,n\rangle} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j + \sum_{\langle n,n,n\rangle} J'_{ij} \mathbf{S}_i \cdot \mathbf{S}_k \,. \quad (15)$$

Here $t_{ij} > 0$ and $t'_{ik} > 0$.

In summary, we studied the effect of a small O-O direct hopping term on the parameters of a single-band t-J model. Since our numerical analysis is carried out in the limit $U_d \rightarrow \infty$, the Cu-O triplet state does not contribute to or modify the t-J model[6] and thus has been neglected. The dominant hopping of the singlet state is between the nearest-neighbor Cu sites, the next-nearest-neighbor hopping, although small, but is non-negligible. The direct O-O hopping increases the hopping probability of both nearest-neighbor and next-nearest-neighbor Cu sites which are represented by t and t', respectively. We find that the next-nearest-neighbor coupling J' between copper spins is antiferromagnetic and its magnitude is non-negligible. It will be interesting to study the t-t'-J-J' model by both numerical and analytic methods, because it may provide far richer physics than the t-J model itself.

We also show that the doping dependence of the superexchange interaction J changes drastically depending upon whether a localized wave function or an extended wave function is used for the holes. This result indicates that J should be self-consistently calculated when a more accurate solution of the model is found.

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