Magnetic frustration in the three-band Anderson lattice model for high-temperature superconductors

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(Received 8 March 1990)

The three-band Anderson lattice model for the CuO_2 planes in high-T_c superconductors is established. Treating this model by perturbation theory, the effective spin interactions are derived. The antiferromagnetic superexchange integrals are calculated as functions of the direct oxygen transfer and the hole concentration. It is found that frustration in the superexchange occurs, even in the undoped case, which increases with oxygen transfer and decreases with hole concentration.

The phase diagram of the high-T_c superconductors,[1] such as $La_{2-x}(Sr,Ba)_xCuO_4$, shows a variety of phases depending on temperature and dopant concentration;[1,2] e.g., the La-Sr compounds are antiferromagnetic below $T_N(x)$, where T_N drops quickly to zero at $x \simeq 0.02$, and becomes superconducting in the region $0.05 < x < 0.3$. This behavior suggests the existence of magnetic frustrations and a possible connection between antiferromagnetism and superconductivity. Concerning the description of frustration mechanisms, in the phenomenological models[2,3] a static O hole introduced by doping generates a ferromagnetic coupling between neighboring Cu spins. In the microscopic approaches, the t-J model (including three-site terms) was shown[4] to yield hole-induced frustration due to an effective second-neighbor antiferromagnetic coupling. In an extended Hubbard model, by numerical cluster calculations in an Anderson impurity approximation, magnetic frustration in undoped La_2CuO_4 was found.[5,6] The study of frustration in the realistic many-band Hubbard models may be extended by going beyond the impurity approximation and by calculating the concentration dependence.

In this paper we are concerned with those problems. Our goals are (i) to establish the three-band Anderson lattice model for the CuO_2 planes, (ii) to derive effective spin interactions by perturbation theory, and (iii) to calculate the frustration in the superexchange as function of the direct O transfer and the O-hole concentration.

We start from a simple version of the Emery model[7,8] for the Cu $3d_{x^2-y^2}$ and O $2p_{x,y}$ orbitals in the square CuO_2 lattice and include the direct O transfer (hole representation),

$$H=\varepsilon_d\sum_{i\sigma}d_{i\sigma}^{\dagger}d_{i\sigma}+U\sum_i d_{i\uparrow}^{\dagger}d_{i\uparrow}d_{i\downarrow}^{\dagger}d_{i\downarrow}+\varepsilon_p\sum_{l\sigma}p_{l\sigma}^{\dagger}p_{l\sigma}+t_p\sum_{\langle l,l'\rangle\sigma}(-1)^{N_{ll'}}p_{l\sigma}^{\dagger}p_{l'\sigma}+V\sum_{\langle i,l\rangle\sigma}(-1)^{M_{il}}(p_{l\sigma}^{\dagger}d_{i\sigma}+\text{H.c.})\,. \tag{1}$$

The energies $\varepsilon_d(\varepsilon_p)$ of the Cu(O) holes contain the chemical potential μ. In the transfer integrals, the phases of the orbitals are taken into account (as in the $t_p=0$ case[8]), where the integers $N_{ll'},M_{il}$ are given in Fig. 1. Concerning the parameters, we quote,[9] e.g., $U=10$ eV, $\Delta\varepsilon=\varepsilon_p-\varepsilon_d=4$ eV, $V=1.38$ eV, and $t_p=0.33$ eV.

After Fourier transforming the O-hole operators $p_{i\alpha\sigma}(\alpha=1,2$; see Fig. 1) to $p_{\mathbf{k}\alpha\sigma}(0\le k_x,k_y<2\pi)$, we construct the Wannier O-hole basis $(c_{\mathbf{k}\nu\sigma};\nu=1,2)$ diagonalizing the O part of (1) by the unitary transformation

$$c_{\mathbf{k}\sigma}=U_{\mathbf{k}}p_{\mathbf{k}\sigma}\,,\qquad U_{\mathbf{k}}=\frac{1}{\sqrt{2}}\begin{pmatrix}1 & e^{i[(k_x-k_y)/2]}\\ e^{-i[(k_x-k_y)/2]} & -1\end{pmatrix}. \tag{2}$$

We arrive at the model

$$H=\varepsilon_d\sum_{i\sigma}d_{i\sigma}^{\dagger}d_{i\sigma}+U\sum_i d_{i\uparrow}^{\dagger}d_{i\uparrow}d_{i\downarrow}^{\dagger}d_{i\downarrow}+\sum_{\mathbf{k}\nu\sigma}\varepsilon_{\mathbf{k}\nu}c_{\mathbf{k}\nu\sigma}^{\dagger}c_{\mathbf{k}\nu\sigma}+\frac{V}{\sqrt{\mathcal{N}}}\sum_{i\mathbf{k}\nu\sigma}(\gamma_{\mathbf{k}\nu}e^{-i\mathbf{k}\mathbf{R}_i}c_{\mathbf{k}\nu\sigma}^{\dagger}d_{i\sigma}+\text{H.c.})\,, \tag{3}$$

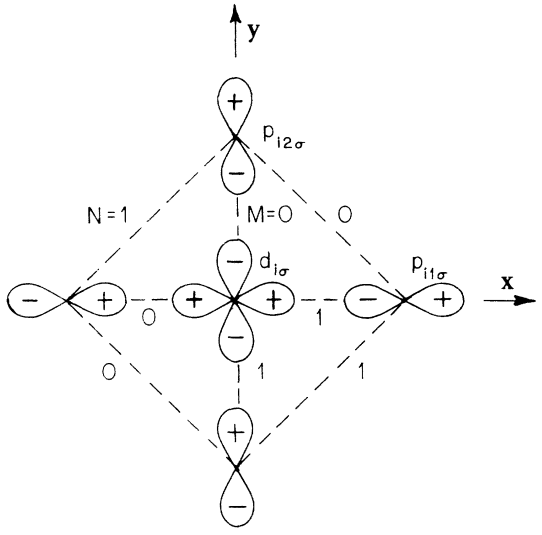

FIG. 1. Bonding Cu and O orbitals and the $N_{ll'},M_{il}$ in Eq. (1) defined according to the phases of the orbitals.

where

$$\varepsilon_{\mathbf{k}\nu}=\varepsilon_p-(-1)^{\nu}4|t_p|\sin\frac{k_x}{2}\sin\frac{k_y}{2},$$

$$\gamma_{\mathbf{k}\nu}=\sqrt{2}ie^{i(k_\nu/2)}\left(\sin\frac{k_x}{2}+(-1)^{\nu}\sin\frac{k_y}{2}\right)$$

($k_1=k_x$, $k_2=k_y$, and $\mathcal{N}$ is the number of Cu sites). This model is the three-band Anderson lattice model with the explicit O band and hybridization dispersions $\varepsilon_{\mathbf{k}\nu}$ and $\gamma_{\mathbf{k}\nu}$, respectively. In contrast to heavy-fermion systems,[10] the O bandwidth $4|t_p|$ has about the magnitude of V, and the O bands are slightly filled for small doping. The inclusion of the nonbonding O orbitals (perpendicular to those shown in Fig. 1) yields additional O bands with $\varepsilon_{\mathbf{k}3}=\varepsilon_{\mathbf{k}1}$, $\varepsilon_{\mathbf{k}4}=\varepsilon_{\mathbf{k}2}$, which are decoupled from (3).

In the limit $t_p=0$,[7] it is convenient to rotate the Wannier O basis by the unitary transformation

$$c_{\mathbf{k}\sigma}=W_{\mathbf{k}}\Phi_{\mathbf{k}\sigma},\quad W_{\mathbf{k}}=\frac{1}{\gamma_{\mathbf{k}}}\begin{pmatrix}\gamma_{\mathbf{k}1} & \gamma_{\mathbf{k}2}^{*}\\ \gamma_{\mathbf{k}2} & -\gamma_{\mathbf{k}1}^{*}\end{pmatrix},\tag{4}$$

where

$$\gamma_{\mathbf{k}}^2=4\left(\sin^2\frac{k_x}{2}+\sin^2\frac{k_y}{2}\right)=\sum_{\nu}|\gamma_{\mathbf{k}\nu}|^2$$

and $\gamma_{\mathbf{k}}\geq 0$. Since $\sum_\nu \gamma_{\mathbf{k}\nu}c_{\mathbf{k}\nu\sigma}^{\dagger}=\gamma_{\mathbf{k}}\Phi_{\mathbf{k}1\sigma}^{\dagger}$, the orbital $\Phi_{\mathbf{k}2\sigma}^{\dagger}$ at energy ε_p is decoupled, and (3) reduces to a two-band model.[8,11]

For the derivation of effective spin interactions from (3), we take the limit $U\rightarrow\infty$ by means of Hubbard operators ($X_i^{00}+\sum_\sigma X_i^{\sigma\sigma}=1$) and the replacements $d_{i\sigma}^{\dagger}\rightarrow X_i^{\sigma 0}$, $d_{i\sigma}\rightarrow X_i^{0\sigma}$. In our perturbation theory, we consider the Kondo region ($V\ll\Delta\varepsilon$) of $H=H_0+H_V$, where H_V is the hybridization term in (3), and construct an effective Hamiltonian by the unitary transformation $\tilde{H}=e^{S}He^{-S}$ with S determined from $[S,H_0]=-H_V$.[12] The exact solution is

$$S=\frac{V}{\sqrt{N}}\sum_{i\mathbf{k}\sigma}\left(\frac{\gamma_{\mathbf{k}\nu}}{\Delta\varepsilon_{\mathbf{k}\nu}}e^{-i\mathbf{k}\mathbf{R}_i}c_{k\nu\sigma}^{\dagger}X_i^{0\sigma}-\text{H.c.}\right),\tag{5}$$

$\Delta\varepsilon_{\mathbf{k}\nu}=\varepsilon_{\mathbf{k}\nu}-\varepsilon_d$. Note that, unlike the perturbation approach by Zaanen and Oleś,[13] in our theory the effects of the O-band terms in H_0 on the effective interactions in $\tilde{H}$ can be taken into account. We project $\tilde{H}$ onto the subspace of single Cu-hole occupancy ($\sum_\sigma X_i^{\sigma\sigma}=1$) and obtain H_{eff} containing only terms of even order in V. Up to fourth order, we get $H_{\text{eff}}=H_0+H_2+H_4$, where

$$H_2=\frac{V^2}{4}\sum_{\mathbf{k}\nu\nu'\sigma}\gamma_{\mathbf{k}\nu}\gamma_{\mathbf{k}\nu'}^{*}\left(\frac{1}{\Delta\varepsilon_{\mathbf{k}\nu}}+\frac{1}{\Delta\varepsilon_{\mathbf{k}\nu'}}\right)c_{k\nu\sigma}^{\dagger}c_{\mathbf{k}\nu'\sigma}-V^2\sum_{\mathbf{k}\nu}\frac{|\gamma_{\mathbf{k}\nu}|^2}{\Delta\varepsilon_{\mathbf{k}\nu}}+\frac{V^2}{\mathcal{N}}\sum_{i\mathbf{k}\nu\mathbf{k}'\nu'}\gamma_{\mathbf{k}\nu}\gamma_{\mathbf{k}'\nu'}^{*}\left(\frac{1}{\Delta\varepsilon_{\mathbf{k}\nu}}+\frac{1}{\Delta\varepsilon_{\mathbf{k}'\nu'}}\right)e^{-i(\mathbf{k}-\mathbf{k}')\mathbf{R}_i}\mathbf{S}_i\cdot\mathbf{R}_{\mathbf{k}\nu,\mathbf{k}'\nu'};$$

$$\mathbf{R}_{\mathbf{k}\nu,\mathbf{k}'\nu'}=\tfrac{1}{2}\sum_{\sigma\sigma'}c_{k\nu\sigma}^{\dagger}\boldsymbol{\sigma}_{\sigma\sigma'}c_{\mathbf{k}'\nu'\sigma'}\tag{6}$$

is a generalized O spin. In second order there occur indirect O-band and interband terms and an antiferromagnetic exchange coupling between O and Cu spins (Kondo coupling).

The fourth-order contribution contains spin-spin, spin-spin-hole, hole-hole, and hole-hole-spin interactions and irrelevant renormalizations of H_0, H_2. Considering small O-hole concentrations, we neglect the four-hole-operator terms and treat the spin-spin-hole term in the mean-field approximation (replacing the hole operators by the average with respect to H_0). Accordingly, we obtain the Heisenberg term

$$H_4^J=\sum_{i\neq j}J_{ij}\mathbf{S}_i\cdot\mathbf{S}_j,\tag{7}$$

where

$$J_{ij}=\frac{V^4}{2\mathcal{N}^2}\sum_{\mathbf{k}\nu\mathbf{q}\mu}A_{\mathbf{k}\nu\mathbf{q}\mu}(1-\langle c_{\mathbf{k}\nu\sigma}^{\dagger}c_{\mathbf{k}\nu\sigma}\rangle_0)e^{i(\mathbf{k}-\mathbf{q})(\mathbf{R}_i-\mathbf{R}_j)},$$

$$A_{\mathbf{k}\nu\mathbf{q}\nu}=\frac{|\gamma_{\mathbf{k}\nu}|^2|\gamma_{\mathbf{q}\mu}|^2}{\Delta\varepsilon_{\mathbf{k}\nu}\Delta\varepsilon_{\mathbf{q}\mu}}\left(\frac{3}{\Delta\varepsilon_{\mathbf{k}\nu}}+\frac{1}{\Delta\varepsilon_{\mathbf{q}\mu}}\right).$$

The superexchange integrals J_{ij} are functions of $t_p/\Delta\varepsilon$ and the O-hole concentration δ (number of holes per elementary cell). For $t_p=0$ we get $J_{ij}=J_1\delta_{\langle i,j\rangle,0}$; $J_1=[2V^4/(\Delta\varepsilon)^3][1-(\delta/\delta_{\max})]$; adding the nonbonding orbitals to (3), we have $\delta_{\max}=8$. Since only the nearest-neighbor coupling J_1 occurs, for $t_p=0$ there is no frustration for all δ. This result is contrary to that by Entel *et al.*,[14] who have found frustration for $t_p=0$ and $\delta\neq 0$. Moreover, we have an antiferromagnetic superexchange ($J_1>0$) for all δ, which contradicts the result by Muramatsu, Zeyher, and Schmeltzer[15] showing a change in sign of J_1 at δ_c [in hole representation and for $U\rightarrow\infty$, J_1 of Ref. 15 reads $J_1=2V^4/(\Delta\varepsilon)^3(1-\frac{3}{2}\delta)$, so that $\delta_c=\frac{2}{3}$].

For $t_p\neq 0$ and $\delta=0$, the dependences of J_1 and the second-neighbor (diagonal bond) superexchange J_2 on $|t_p|/\Delta\varepsilon$ are shown in Fig. 2 (all further neighbor couplings are neglected). J_1 increases with direct O-band dispersion, which is due to the decrease of the gap for virtual d-p transitions. The qualitative effect of the direct O transfer is the appearance of the antiferromagnetic coupling J_2 which results in frustration measured by J_2/J_1 increasing with $|t_p|/\Delta\varepsilon$. Taking the parameters quoted above[9] ($|t_p|/\Delta\varepsilon=0.083$), we get $J_1=0.36$ eV and $J_2/J_1=0.05$, where J_1 has the correct order of magnitude [$J_1^{\text{expt}}\simeq 0.1$ eV (Ref. 9)] and the frustration agrees with the cluster results by Annett *et al.*[5] ($J_2/J_1=5$–8%).

In the calculation of the δ dependences of J_1 and J_2 we have determined the chemical potential and the O-hole average in (7) at zero temperature; the results are shown in Fig. 3. With increasing δ, the exchange integral J_2 decreases faster than J_1, so that J_2/J_1 decreases too. From this result we conclude that frustration in the superexchange cannot explain the drop in T_N with doping, and additional frustration mechanisms in H_{eff} must be in-

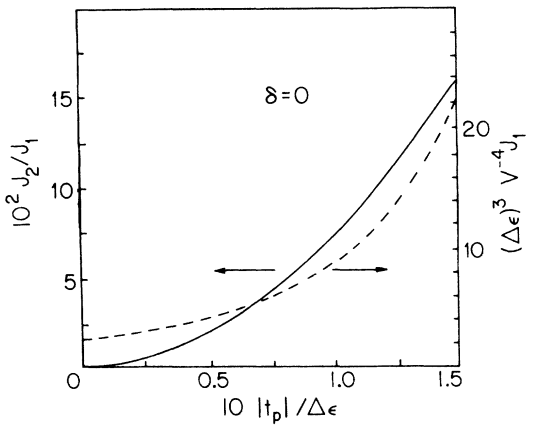

FIG. 2. Nearest-neighbor superexchange integral J_1 (dashed curve) and frustration J_2/J_1 (solid curve) as functions of $|t_p|/\Delta\epsilon$ for $\delta=0$.

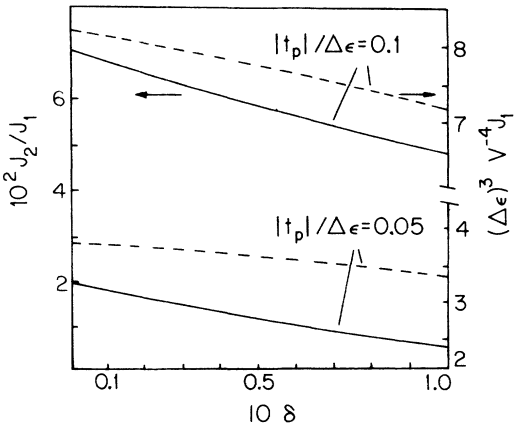

FIG. 3. Superexchange integral J_1 (dashed curve) and frustration J_2/J_1 (solid curve) as functions of δ.

voked. Analogous to the situation in the phenomenological frustration models,[2,3] where the spin-hole coupling creates a ferromagnetic exchange between neighboring Cu spins, in our microscopic model the second-order spin-hole term in (6) may create a fourth-order Ruderman-Kittel-Kasuya-Yosida exchange superimposing the antiferromagnetic superexchange. The net effect may be an increase of frustration upon doping. This problem remains to be studied.

We would like to thank A. Muramatsu, W. Weller, and R. Zeyher for useful discussions.

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