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Quasiparticles in the copper-oxygen planes of high-T_c superconductors: An exact solution for a ferromagnetic background

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A model for a mobile hole in the copper oxide planes of high-temperature superconductors is solved exactly. The hole moves on the oxygen atoms through a lattice of spins localized on the copper atoms. In order to obtain a solvable problem, it is assumed that the copper atoms provide a ferromagnetic background. The resulting quasiparticles have both charge and spin in contrast to the Cu-O singlets occurring in proposed effective single-band Hubbard models derived from the Cu-O network. Thus these two models of high-temperature superconductors may have different low-energy physics.

A central problem in the theory of high-temperature superconductors[1] such as $La_{2-x}Sr_xCuO_4$ and $YBa_2Cu_3O_{6+x}$ is to determine the nature of the quasiparticle states in the copper oxide planes. This is a nontrivial problem because we have to deal with a strongly coupled system and the charge carriers are not free electrons or holes.

In a single-band Hubbard model[2] it is assumed that there is one available state per unit cell. For a strong on-site interaction U and one hole per unit cell, it is a good approximation to assume that each state is singly occupied and that the spins are the only remaining degrees of freedom.[2] Such a system is an antiferromagnetic insulator, consistent with many experiments on high-temperature superconductors.[3] When further holes are added by doping or by increasing the oxygen content, some states will contain two holes of opposite spin,[4] and their mobility is strongly dependent on the background configuration. The problem is then to construct the corresponding quasiparticle states and to understand how the antiferromagnetism is destroyed giving way to a superconducting state. An alternative picture to be assumed in the present paper is to use an extended model,[5] allowing single states on both copper and oxygen sites. There is by now much evidence to support the view that the antiferromagnetic spins are mainly on copper sites and the mobile holes on oxygen sites.[5] It is then a more complicated problem to understand the motion of the charge carriers, since we must first determine what replaces the simple doubly occupied sites of the single-band model and find out how many degrees of freedom there are. In order to address this first step in the process of constructing quasiparticle states, we give in this paper the exact solution of a model of an oxygen hole moving in a *ferromagnetic* Cu background. This is a trivial problem in the single-band model—the doubly occupied site is a freely moving spin-0 entity. For the entended copper-oxygen model, we find that the oxygen hole is equally mobile but it carries a spin. Thus, the two models are not equivalent. In a separate paper[6] (to be referred to as I) we calculate the interactions between the mobile holes, mediated by the background of Cu holes, and show how they may lead to high-temperature superconductivity.

Zhang and Rice[7] suggested that the two models are in fact equivalent in their low-energy physics because an oxygen hole forms a singlet with a copper hole in the same cell, and this behaves in essentially the same way as a doubly occupied site of the single-band model. Our calculation shows explicitly that this is not so. The admixture of singlets formed between holes in different cells may not be neglected and gives rise to the additional spin quantum number.

We emphasize that the ferromagnetic background of Cu spins is not the same as the physical state of the CuO_2 planes of the high-T_c superconductors. The reason for using such a background as a first step is that it enables us to obtain an exact solution of the model and hence to construct quasiparticle states and to consider the relationship to the single-band model without having to worry about the merits of particular approximations. As we shall show, the essential point is that the superexchange between holes on Cu sites and oxygen Wannier states is nonlocal. This will lead to differences from the single-band model whatever the state of the background Cu spins.

The Hamiltonian to be studied is

$$H=(t_1+t_2)\sum_{\substack{\mathbf{m},\Delta,\Delta\\ \sigma,\sigma'}} a^{\dagger}_{\mathbf{m}\sigma'}a_{\mathbf{m}\sigma}a^{\dagger}_{\mathbf{m}+\Delta,\sigma}a_{\mathbf{m}+\Delta',\sigma'} - t_2\sum_{\substack{\Delta,\Delta'\\ \mathbf{m},\sigma}} a^{\dagger}_{\mathbf{m}+\Delta,\sigma}a_{\mathbf{m}+\Delta',\sigma} - 4t_1\sum_{\mathbf{m},\sigma} a^{\dagger}_{\mathbf{m}\sigma}a_{\mathbf{m}\sigma}, \qquad (1)$$

where $a^{\dagger}_{\mathbf{m},\sigma}$ creates a hole of spin σ in a $Cu(3d_{x^2-y^2})$ state at $\mathbf{m}\equiv(m,n)$ and $a^{\dagger}_{\mathbf{m}+\Delta,\sigma}$ creates a hole of spin σ in an $O(2p_x)$ or $O(2p_y)$ state at $\mathbf{m}+\Delta$. Here Δ is $(\pm\frac{1}{2},0)$ or $(0,\pm\frac{1}{2})$, and a factor $(-1)^{m+n}$ has been included in the definition of $a^{\dagger}_{m+\Delta,\sigma}$ to remove signs associated with the symmetry of the states. The simplest derivation of H from an extended Hubbard model[6] uses second-order degenerate perturbation theory in the Cu-O hopping parameter t and gives $t_1=t^2/\varepsilon$ and $t_2=t^2/(U_d-\varepsilon)$, where ε is the difference in the energy of $O(2p)$ and $Cu(3d)$ holes

and U_d is the Coulomb repulsion at a Cu site. The Cu-Cu superexchange has been neglected because it is $O(t^4)$ and is much smaller than t_1 or t_2. Also, in order to simplify the presentation, the Cu-O Coulomb repulsion V and oxygen on-site interaction U_p have been omitted: Their effects are discussed in detail in I. With these modifications, Eq. (1) is the same as Eq. (2.4) of I. Furthermore H is the Hamiltonian implicitly used by Zhang and Rice[7] to discuss the relationship to the single-band Hubbard model. We believe that H contains the essential physics of the quasiparticles, even when second-order degenerate perturbation theory cannot be used.[8]

It is also possible to solve the model when the Cu-Cu superexchange is included, as we shall show in a future publication.

Since the number operator for Cu holes commutes with H, it is possible to work with an effective Hamiltonian H' acting in a subspace with one hole per copper site, and to introduce copper spin operators $\vec{S}_{\mathbf{m}}$. Using the Fourier transformation

$$\vec{S}_{\mathbf{k}}=\sum_{\mathbf{m}}\vec{S}_{\mathbf{m}}e^{i\mathbf{k}\cdot\mathbf{m}} \tag{2}$$

and the canonical transformation[9]

$$b_{\mathbf{k}\sigma}=(a_{\mathbf{k}}N^{1/2})^{-1}\sum_{\mathbf{m},\Delta}a_{\mathbf{m}+\Delta,\sigma}e^{-i\mathbf{k}\cdot\mathbf{m}}, \tag{3}$$

where

$$a_k^2=2(2+\cos k_x+\cos k_y). \tag{4}$$

H' may be written

$$H'=[2(t_1+t_2)]/N\sum_{\substack{\mathbf{k},\mathbf{k}'\\ \sigma,\sigma'}}a_{\mathbf{k}}a_{\mathbf{k}'}\vec{S}_{\mathbf{k}-\mathbf{k}'}\cdot\vec{s}_{\sigma\sigma'}b^{\dagger}_{\mathbf{k}'\sigma}b_{\mathbf{k}\sigma'}+\tfrac{1}{2}(t_1-t_2)\times\sum_{\mathbf{k},\sigma}a_{\mathbf{k}}^2b^{\dagger}_{\mathbf{k}\sigma}b_{\mathbf{k}\sigma}-4Nt_1. \tag{5}$$

Here $\vec{s}_{\sigma\sigma'}$ are the matrix elements of the spin operator. Now imagine adding a hole with an up spin to a ferromagnetic (FM) background $|\mathrm{FM}\downarrow\rangle$ in which all Cu spins are down. The hole will hop between oxygen sites (since all Cu states are singly occupied) and will exchange its spin with the Cu holes. The exact eigenstates are of the form

$$|\psi_{\mathbf{k}}\rangle=\left(b^{\dagger}_{\mathbf{k}\uparrow}+1/N\sum_{\mathbf{q}}W(\mathbf{k},\mathbf{q})b^{\dagger}_{\mathbf{q}\downarrow}S^{\dagger}_{-\mathbf{q}}\right)|\mathrm{FM}\downarrow\rangle, \tag{6}$$

where $S^{\dagger}_{\mathbf{k}}\equiv S^{x}_{\mathbf{k}}+iS^{y}_{\mathbf{k}}$ is a Cu spin-raising operator. Substituting this form into the Schrödinger equation leads to the following equations for $W(\mathbf{k},\mathbf{q})$ and the eigenvalue $\lambda_{\mathbf{k}}$:

$$[(t_1+t_2)/N]a_{\mathbf{k}}\sum_{q}W(\mathbf{k},\mathbf{q})a_{\mathbf{q}}=\lambda_{\mathbf{k}}+t_2a_{\mathbf{k}}^2, \tag{7}$$

$$(\lambda_{\mathbf{k}}-t_1a_{\mathbf{q}}^2)W(\mathbf{k},\mathbf{q})+[(t_1+t_2)/N]a_{\mathbf{q}}\times\sum_{\mathbf{q}'}W(\mathbf{k},\mathbf{q}')a_{\mathbf{q}'}=(t_1+t_2)a_{\mathbf{k}}a_{\mathbf{q}}. \tag{8}$$

It is straightforward to solve for $W(\mathbf{k},\mathbf{q})$ to find

$$W(\mathbf{k},\mathbf{q})=-a_{\mathbf{q}}/a_{\mathbf{k}}[(\lambda_{\mathbf{k}}-t_1a_{\mathbf{k}}^2)/(\lambda_{\mathbf{k}}-t_1a_{\mathbf{q}}^2)], \tag{9}$$

where $\lambda_{\mathbf{k}}$ is given by

$$a_{\mathbf{k}}^2/(\lambda_{\mathbf{k}}-t_1a_{\mathbf{k}}^2)+1/N\sum_{\mathbf{q}}\frac{a_{\mathbf{q}}^2}{\lambda_{\mathbf{k}}-t_1a_{\mathbf{q}}^2}=-1/(t_1+t_2). \tag{10}$$

When $t_1=0$, these equations may be solved explicitly to obtain

$$W^0(\mathbf{k},\mathbf{q})=-a_{\mathbf{q}}/a_{\mathbf{k}}, \tag{11}$$

$$\lambda_{\mathbf{k}}^0=-t_2(4+a_{\mathbf{k}}^2)=-8t_2-2t_2(\cos k_x+\cos k_y), \tag{12}$$

$$|\psi_{\mathbf{k}}^0\rangle=\left(b^{\dagger}_{\mathbf{k}\uparrow}-a_{\mathbf{k}}^{-1}\sum_{q}a_{\mathbf{q}}b^{\dagger}_{\mathbf{q}\downarrow}S^{\dagger}_{\mathbf{k}-\mathbf{q}}\right)|\mathrm{FM}\downarrow\rangle. \tag{13}$$

When $\mathbf{k}=0$, there is another way of writing the wave function. Substituting Eq. (3) into Eq (13),

$$|\psi_0^0\rangle=1/(a_0N^{1/2})\sum_{m,j=1,2}[2a^{\dagger}_{\mathbf{m}+\Delta_j\uparrow}-a^{\dagger}_{\mathbf{m}+\Delta_j\downarrow}\times(S^{\dagger}_{\mathbf{m}}+S^{\dagger}_{\mathbf{m}+2\Delta_j})]|\mathrm{FM}\downarrow\rangle, \tag{14}$$

where $\Delta_1=(\frac{1}{2},0)$, $\Delta_2=(0,\frac{1}{2})$. This is a plane wave made up of the Cu-O-Cu spin configuration $[2\downarrow\uparrow\downarrow-\uparrow\downarrow\downarrow-\downarrow\downarrow\uparrow]$ on a horizontal bond ($j=1$) or a vertical bond ($j=2$). This is the ground state of the Hamiltonian that describes the superexchange between an oxygen at site i and its neighbors:

$$H=Js_{i+\Delta_j}\cdot(S_i+S_{i+2\Delta_j}), \tag{15}$$

where $J=2(t_1+t_2)$ for the present model. It is useful to think of the states (6) this way: We can regard the oxygen hole as hopping between sites whose local copper spin configuration is given by the ground state of (15). This is rigorously true when $k=0$ and $t_1=0$, but remains a good approximation for small k and any t_1.

Equation (14) involves correlation only between neighboring sites because, when $t_1=0$, a down-spin oxygen hole cannot propagate in a down-spin copper background. When $t_1\neq0$, the state spreads somewhat, although it never becomes very large. We show in Fig. 1 the energy surface obtained by solving Eq. (10) numerically for $t_1=t_2=0.5$. The surface is very close to that which would be generated by simple hopping on a square lattice, $\lambda_{\mathbf{k}}=\text{const}-2\bar{t}(\cos k_x+\cos k_y)$, with a value of $\bar{t}$ of 0.63.

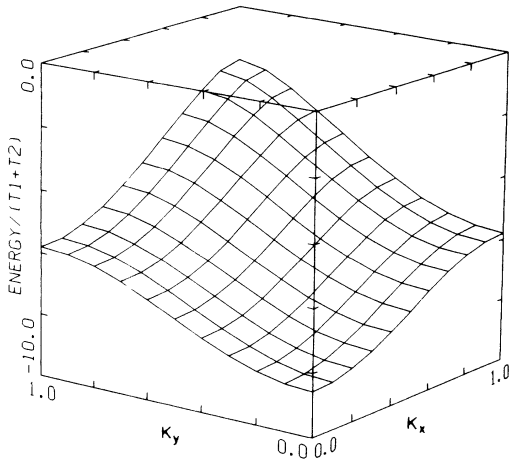

FIG. 1. $\lambda_{\mathbf{k}}$ as a function of (k_x,k_y) for $t_1=t_2=0.5$.

Note that the oxygen states may be written in the site representation [Eq. (14)] or the Wannier representation [Eq. (13)]. Despite the fact that only one of the two Wannier states is involved, Eqs. (13) and (14) are completely equivalent and there is no overcounting in using the site representation. The interaction between holes is, however, easier to see in the site representation, and is discussed in detail in Ref. 6.

Although the quasiparticle is a spin-$\frac{1}{2}$ object, only part, $\frac{1}{2}\sigma_{\mathbf{k}}^0$, of the spin resides on the oxygen site. Indeed, when $t_1 \neq 0$ and $k \neq 0$, the value of $\sigma_{\mathbf{k}}^0$ provides a convenient measure of the difference between the true quasiparticle state and the approximation described below Eq. (14). Using Eq. (6),

$$\sigma_{\mathbf{k}}^0 = \left(1 - 1/N \sum_{\mathbf{q}} |W(\mathbf{k},\mathbf{q})|^2\right) \times \left(1 + 1/N \sum_{\mathbf{q}} |W(\mathbf{k},\mathbf{q})|^2\right)^{-1}. \quad (16)$$

Once again, this may be evaluated explicitly for $t_1 = 0$, using Eqs. (4) and (11) to obtain

$$\sigma_{\mathbf{k}}^0 = (\cos k_x + \cos k_y)/(4 + \cos k_x + \cos k_y), \quad (17)$$

when $\mathbf{k} = 0$, $\sigma_{\mathbf{k}}^0 = \frac{1}{3}$ as for the Cu-O-Cu spin configuration appearing in Eq. (15). As k increases, $\sigma_{\mathbf{k}}^0$ decreases, passing through zero along the lines $\pm k_x \pm k_y = \pi$ and going to -1 when $k = (\pi,\pi)$. A graph of σ_k^0 for another case, $t_1 = t_2$, is shown in Fig. 2. It can be seen that σ_k^0 shows qualitatively the same behavior as for $t_1 = 0$.

Zhang and Rice[7] considered the same problem, implicitly working with the effective Hamiltonian H' of Eq. (5). They proposed that the quasiparticles be regarded as singlets formed by two holes occupying a copper state and an oxygen Wannier state *in the same cell.* The motion of the singlet could then be described by the same Hamiltonian as for a doubly occupied site in a single-band Hubbard model and the low-energy physics should be the same. This approximation is equivalent to retaining only one term ($n = 0$) of the Fourier transform $a(n)$ of a_k. Indeed,[10] the amplitude $a(0) \simeq 1.92$ for an in-cell singlet is larger than that for neighboring cells $a(2\Delta) \approx 0.26$. Furthermore, the overlap between the in-cell singlet state $|\psi_k^s\rangle$, for which $W(\mathbf{k},\mathbf{q}) = -1$, and the exact state, is quite large:

$$\frac{\langle \psi_{\mathbf{k}}^s | \psi_{\mathbf{k}} \rangle}{(\langle \psi_{\mathbf{k}} | \psi_{\mathbf{k}} \rangle \langle \psi_{\mathbf{k}}^s | \psi_{\mathbf{k}}^s \rangle)^{1/2}} = \frac{1 - 1/N \sum_{\mathbf{q}} W(\mathbf{k},\mathbf{q})}{\left[2\left(1 + 1/N \sum_{\mathbf{q}} |W(\mathbf{k},\mathbf{q})|^2\right)\right]^{1/2}}, \quad (18)$$

which is equal to $[a_k + a(0)]/[(8 + 2a_k^2)^{1/2}]$ when $t_1 = 0$. For $\mathbf{k} = 0$, the overlap is 0.97 and for $\pm k_x \pm k_y = \pi$ it is 0.98. Nevertheless, the spin on the oxygen site, which is a symptom of the spin of the true quasiparticle is substantial whereas it is zero for the singlet. To get a feeling for how this can be so, define the overlap in Eq. (18) to be $\cos\theta$ and make the approximation that $[1/N \sum_q W(\mathbf{k},\mathbf{q})]^2$ is the same as $1/N \sum_q |W(\mathbf{k},\mathbf{q})|^2$ (which is in error by 4% when $t_1 = 0$ and $k = 0$). Then it is straightforward to show from Eq. (16) that $\sigma_{\mathbf{k}}^0 = \sin 2\theta$. Thus when $\sigma_{\mathbf{k}}^0 = \frac{1}{3}$, $\theta \approx \frac{1}{6}$, and $\cos\theta \approx 71/72 = 0.99$. In other words the overlap is not a sensitive indicator of the value of the spin on the oxygen site which in turn is a manifestation of the difference between the in-cell Wannier singlet and the

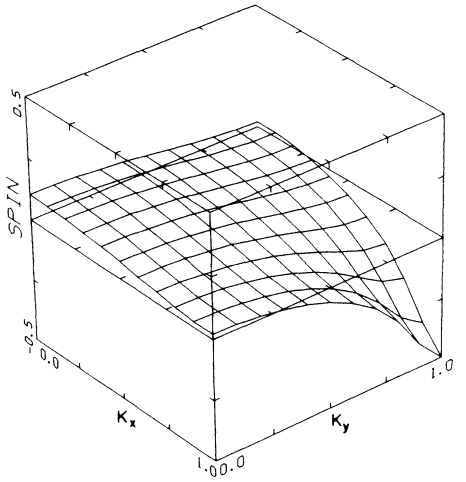

FIG. 2. The average value of the spin on the oxygen hole at any particular site, in the eigenstates of the Hamiltonian of wave vector (k_x,k_y). $t_1 = t_2 = 0.5$.

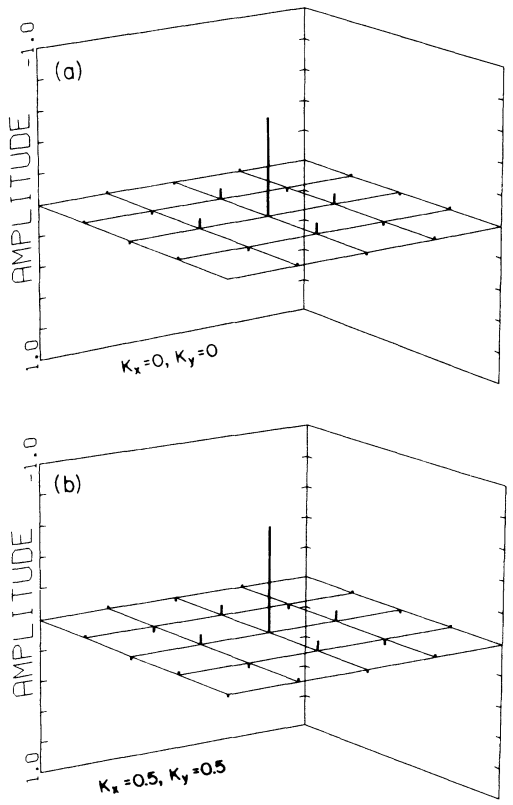

FIG. 3. The amplitude for a spin to be reversed on a given copper site, when there is a hole on the oxygen Wannier state at the center of the region shown for two values of $\mathbf{k}$. Note that the wave function changes very little, although the average spin varies considerably.

Cu-O-Cu correlated spin system. It may be seen in Eq. (13) that the true state is a superposition of singlets involving an oxygen Wannier state and holes in all Cu states. The individual components are not orthogonal and hence the net spin is not zero.

We show in Fig. 3 the amplitude for the reversed copper spin to be on a particular site given that the oxygen Wannier state is located at the origin, for $k=0$ and $k=(\pi/2,\pi/2)(t_1=t_2)$. The wave function does not change dramatically even though the spin is nearly zero for the second case.

It is of course significant that there is a local spin as well as a charge associated with the oxygen hole—in contrast to the single-band model. In a spin-fluid background, either quantity may be measured to determine the location of the quasiparticle. Furthermore, since the quasiparticle has a spin, it can bind in either a triplet or a singlet state, as for a free electron, possibilities that are not available to a spinless hole.

In the present model, the value of J in Eq. (16) is fixed by the hopping parameters. More realistically, J could be smaller than $2(t_1+t_2)$ due to direct ferromagnetic exchange or the effect of additional Coulomb repulsion parameters. We point out here that the wave function of the form (14) may also remain a good representation of the states at the bottom of the band, even when the total exchange is slightly ferromagnetic. This is a consequence of the energy gained from the high mobility of a Cu-O singlet. This more complex case will be treated in detail in a subsequent paper.

The existence of a local spin associated with an oxygen hole has been demonstrated for a ferromagnetic background of Cu spins but it is clearly more general since it is a consequence of the nonlocality of the first term of H' in Eq. (5)—the one describing copper-oxygen exchange. Of course, it is still necessary to solve for the motion of an oxygen hole in an arbitrary background in order to construct the general quasiparticle state, and this is a complicated problem. It is conceivable that, for some backgrounds, the spin of the quasiparticle turns out to be compensated. But that would have to be proven, and it is clear that the starting point for such a proof is not the same as the single-band model.

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[4]We find it convenient to use the name "holes" for missing electrons from Cu^+ (filled $3d$ shell) and O^{2-} (filled $2p$ shell) as described later. In the literature of the single-band Hubbard-model "holes" are vacant sites.

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[8]N. Andrei and P. Coleman have studied a related model but have omitted the interaction between a copper spin and a hole in an oxygen Wannier state on a different cell. These interactions make the spin of the quasiparticle nonzero and are the terms which make the behavior of the two models different. S. Maekawa, T. Matsuma, Y. Isawa, and H. Ebisawa have given a derivation of Eq. (5) that incorrectly leaves out some of the terms in the Hamiltonian.

[9]The Wannier states $\phi_{i\sigma}$ used by Zhang and Rice (Ref. 7) [their Eq. (7)] are the Fourier transforms of our $b_{\mathbf{k}\sigma}$ defined in Eq. (3). To show the equivalence, note that, as described below Eq. (1), we have included a factor $(-1)^{m+n}$ in the definition of $a_{\mathbf{m}+\Delta,\sigma}$ which removes factors of (-1) in the hopping Hamiltonian and displaces all wave vectors by (π,π). Thus, $P^{s}_{\mathbf{m},\sigma}$ of Zhang and Rice (Ref. 7) is equal to our $\frac{1}{2}\sum_{\Delta} a_{\mathbf{m}+\Delta,\sigma}$ and their quantity $\beta_{\mathbf{k}}$ is equal to $2/\alpha_{\mathbf{k}}$ in our notation.

[10]These quantities were evaluated by Zhang and Rice (Ref. 7).