RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 47, NUMBER 2 1 JANUARY 1993-II

Spin and charge dynamics in a one-dimensional two-band Hubbard model

H.-Q. Ding and W. A. Goddard III
Materials Simulation Center, Beckman Institute, California Institute of Technology, Pasadena, California 91125
(Received 20 August 1992)

A CuO chain with one or two holes doped is studied via the two-band Hubbard model in the large-U limit on a finite cluster. The quasiparticle band is $E_k \sim 0.87J\cos(2ka)$ with the bottom at $ka=\pi/2$. The quasiparticles are kinks which have the character of three-spin polarons. The copper spin excitations are incommensurate spin waves. The two doped holes repel each other very slightly, in contrast to the slight attraction in the t-J model. Spin-charge separation also occurs.

As prototypes of strongly correlated systems, Hubbard models are interesting because of the difficulty in obtaining analytical solutions and also in many fundamental concepts. The discovery of high-T_c superconductivity has brought a surge of interest in these (both single-band and multiband) Hubbard models. In particular, the one-dimensional (1D) single-band model and its strong-coupling version, the t-J model,[1] have been widely studied[2] because of their simplicity in 1D. In special cases, exact solutions are obtained. The general features include spin-charge separation and incommensurate spin waves. However, the realistic models for the cuprate superconductors are the multiband models[3,4] which include the oxygens explicitly, e.g., a two-band model for the CuO chain in $YBa_2Cu_3O_7$. The exact properties of this multiband 1D model are far less well known, although it is generally expected that the general features in the one-band model may persist.

In this paper, we report exact calculations of the two-band model in the strong-coupling limit on finite clusters (16 Cu and 16 oxygen sites). With one or two doped holes at the oxygen sites, the Cu spin correlations becomes incommensurate. The quasiparticle band can be approximated by $E(k)\sim 0.87J\cos(2k)$; the bottom is at $k=\pi/2$ with an effective mass $m^*\simeq 1.2m_e$. The dressing excitations around the doped oxygen hole is short ranged (about 2a) and the average spin of the hole is $\langle S_O^z\rangle \simeq 0$. In both one- and two-hole cases, the quasiparticle has characteristics of the three-spin polaron[3] except that this polaron is constantly scattered between spin-up and spin-down states, i.e., polarons are incoherent kinks. The spin correlation between the two doped holes are weak and its sign oscillates as their separation goes through even and odd number of unit cells. In broad parameter range, the motions of two doped holes are very similar to that of noninteracting fermions, as seen from the pair (density-density) correlations, although the holes experience a very slight repulsion in contrast to the slight attraction occurring in the t-J model.

The model for the CuO chain in the present study is the standard 1D version of the three-band extended Hubbard model[3,4]

$$\begin{aligned} H=&\epsilon_d\sum_i n_i+\epsilon_p\sum_k n_k+U_d\sum_i n_{i\uparrow}n_{i\downarrow}+U_p\sum_k n_{k\uparrow}n_{k\downarrow}\\ &+U_{pd}\sum_{\langle ik\rangle} n_i n_k+t_{pd}\sum_{ik\sigma}(d_{i\sigma}^\dagger p_{k\sigma}+\text{H.c.})\\ &+K_{pd}\sum_{ik\sigma\sigma'} d_{i\sigma'}^\dagger p_{k\sigma}^\dagger d_{k\sigma}p_{i\sigma'}\ , \end{aligned} \tag{1}$$

where i,j label the Cu sites, k,l the O sites, σ the spin. $p_k^\dagger$ (p_k) are the creation (destruction) operators for oxygen $2p_y$ states and $d_i^\dagger, d_i$ for Cu $3d_{x^2-y^2}$ states, and the t_{pd} term is the hybridization between them. U_p, U_d are on-site Coulomb repulsions and ϵ_p, ϵ_d are site energies. U_{pd} is near-neighbor repulsion, and K_{pd} the direct spin exchange between Cu and O sites. $n_i=\sum_\sigma d_{i\sigma}^\dagger d_{i\sigma}$ and $n_k=\sum_\sigma p_{k\sigma}^\dagger p_{k\sigma}$. The 2D version of this model has been studied in various cases.[3-7]

In the undoped case such as La_2CuO_4, each Cu has $3d^9$ configuration with no holes in oxygen sites. This half-filled system is a charge-transfer insulator with antiferromagnetic spin coupling J_{dd} between the localized Cu spins, i.e., a Heisenberg model. With small doping, the additional holes go largely to oxygen sites and we can similarly simplify the two-band model into an effective large-U Hamiltonian through the second-order perturbation theory[3,8,9]

$$\begin{aligned} H_{\text{eff}}=&J_{dd}\sum_{\langle ij\rangle}' S_iS_j+J_{pd}\sum_{\langle ik\rangle} S_iS_k\\ &+\sum_{\langle kil\rangle\sigma\sigma'}(t_a p_{k\sigma}^\dagger n_{i,\sigma'}p_{l\sigma}\\ &\qquad -t_c p_{k,\sigma}^\dagger d_{i,\sigma'}^\dagger d_{i,\sigma}p_{l,\sigma'}+\text{H.c.})\ . \end{aligned} \tag{2}$$

Here the J_{pd} term is a Kondo-like spin coupling between the oxygen hole and the two nearest Cu spins, arising from the strong d-p hybridization t_{pd}. The hybridization also induces effective spin-preserving hopping (t_a) and spin-exchanging hopping (t_c) terms, implicitly taking into account the charge-transfer process.

The parameters of H_{eff} are obtained by exactly diagonalizing an O-Cu-O cluster using the original three-band Hamiltonian and extracting the effective hopping

parameters t_a, t_c as the energy splitting between the symmetric and antisymmetric states. The Kondo coupling J_{pd} together with a modified Heisenberg term J'_{dd} (the Cu-Cu spin coupling when a doped hole resides at the middle oxygen site) is obtained as the energy splitting between the three lowest states of a Cu-O-Cu cluster. Using the Hubbard parameters of McMahan, Annett, and Martin[10] (parameters of Hybertsen *et al.*[11] and of Langlois and co-workers[12] lead to slightly different effective parameters, but all physical properties remain very much the same), we obtained the set (A) of parameters (all in eV): $J_{dd} \simeq 0.132(1)$, $J'_{dd} \simeq 0.054(0.4)$, and $J_{pd} \simeq 0.92(7)$, $t_a \simeq 0.43(3.3)$, $t_c \simeq 0.82(6.2)$. (In detailed calculation, we express all energies in units of J_{dd} as indicated in the parenthesis.) This J_{dd} is quite close to the values[13] determined in neutron scattering (0.125 eV) and Raman scattering (0.128 eV) and from the spin-wave velocity.

To see how a larger Cu correlation would change the final results, we double U_d in the Hubbard parameters and thus obtain another set (B) of effective parameters: $J_{dd} \simeq 0.095(1)$, $J'_{dd} \simeq 0.010(0.1)$, $J_{pd} \simeq 0.46(4.8)$, $t_a \simeq 0.16(1.7)$, $t_c \simeq 0.63(6.6)$. To see how a smaller charge-transfer barrier would change the results, we reduce by half the value of $\epsilon = \epsilon_p - \epsilon_d$ and thus obtain the third set (C) of effective parameters: $J_{dd} \simeq 0.290(1)$, $J'_{dd} \simeq 0.036(0.1)$, $J_{pd} \simeq 0.086(3)$, $t_a \simeq 0.16(1.1)$, $t_c \simeq 0.93(3.2)$. These three sets of effective parameters cover a reasonably broad range of phase space. A surprising result of our study is that these three sets of parameters lead to very much the same (quantitatively) results. For comparison to the t-J model, we used $t_{t\text{-}J} \simeq 0.46$ eV $= 3.5 J_{dd}$.[11] All calculations use a cluster of 16 unit cells with periodic boundary conditions. In the two-hole case, we restrict the two holes not to occupy the same O site. The Hilbert space has a dimension of 5 834 400, which is reduced to about 0.4×10^6 after using translation symmetry with a given total crystal momenta q. Using standard Lanczos tridiagonalization method, the first few lowest states in each q space can be easily obtained.

In the one-hole case, we plotted the elementary excitations at all nonequivalent **k** in Fig. 1(a), for all three parameter sets. They are surprisingly close to one another. All states are doublet states: $S = \frac{1}{2}$. The bottom of the bands are at $q = \pi/2$. The bands are asymmetric: they are higher on the $q = \pi$ side. These bands can be well approximated by $E(q)/J_{dd} = 0.865 - 0.126\cos(qa) + 0.865\cos(2qa) - 0.042\cos(3q)$, leading to an effective mass $m^* \simeq 0.29\hbar^2/J_{dd}a^2 = 1.2m_e$ (using $J_{dd} = 0.13$ eV and $a = 3.8$ Å). In Fig. 1(b), we also plotted the band from the t-J model and a mean-field band[8] $E(q)/J_{dd} = [t_a^2 + t_b^2 + 2t_a t_b \cos(2qa)]^{1/2}$ where $t_b = t_c - t_a$. Compared to the exact band, the t-J band is similar but the mean-field band has too large a bandwidth. This invalidates the mean-field assumptions that the Cu spins remain in a Néel state and that the effect of the spin-flip hopping (t_c term) is small. In the two-hole case, the ground state is singlet $S = 0$ at $k = 0$. The $k = \pi$ state is a triplet $S = 1$ state with an energy $1.6J_{dd}$ higher.

As doped oxygen holes hop from site to site, the Cu spin background of the Heisenberg ground state is severely distorted. In Fig. 2, we show the average Cu-Cu spin correlations in the two-hole case in comparison to that in the pure Heisenberg case. Clearly, the spin correlations become weaker in the doped case. More importantly, the oscillation of the correlation in the doped case becomes out of phase relative to that of the Heisenberg case. The phase change occurs at $r = 4$. In the structure factor (Fourier transform), a peak is clearly developed at $k = 7\pi/8$. This indicates that the spin correlation acquire a phase $\cos[(7\pi/8)r]$ [rather than the usual antiferromagnetic phase $\cos(\pi r) = (-1)^r$]. This phase is incom-

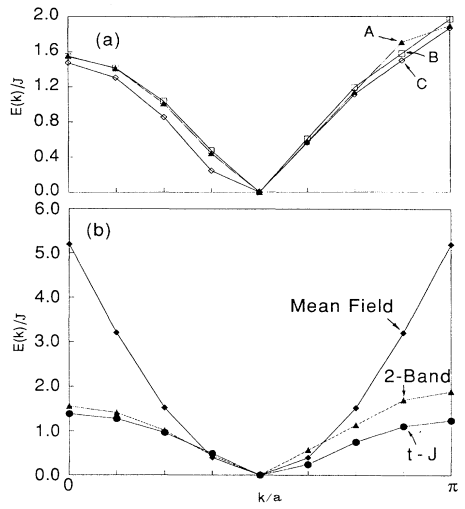

FIG. 1. (a) Quasiparticle band for the two-band model for the parameter sets A, B, C. (b) A comparison with the bands obtained in the t-J model or through the mean-field theory.

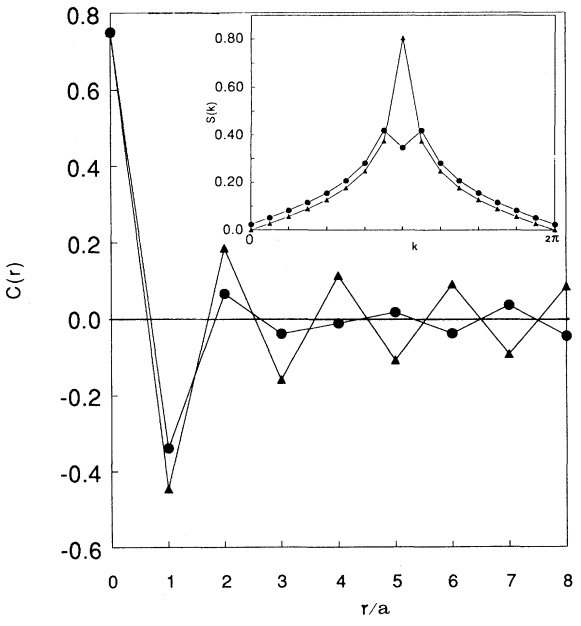

FIG. 2. Cu-Cu spin correlations in the two-hole case (●) compared to that in the Heisenberg case (▲), in real space and in momentum space (inset).

mensurate with the underlining discrete lattice. Its sign becomes negative relative to that of $\cos(\pi r)$ at $r=4$, which clearly manifested itself in the real space (Fig. 2). In the one-hole case, the spin correlations acquire a phase $\cos[(7\pi/16)r]$. A detailed discussion on incommensurate spin waves and their close relation to the Fermi surface is given elsewhere.[14] These incommensurate spin waves are clearly observed in experiments.[15]

It has been suggested that due to the large Kondo coupling J_{pd} between the oxygen hole spin and Cu spins, the quasiparticle is the three-spin polaron

$$\chi_\uparrow=(1/\sqrt{6})(2|\mathrm{Cu}_\uparrow\mathrm{O}_\downarrow\mathrm{Cu}_\uparrow\rangle-|\mathrm{Cu}_\uparrow\mathrm{O}_\uparrow\mathrm{Cu}_\downarrow\rangle$$

$$-|\mathrm{Cu}_\downarrow\mathrm{O}_\uparrow\mathrm{Cu}_\uparrow\rangle)\ .$$

A simple verification of the polaron is to calculate the projection probability, $|\langle\chi_\uparrow|g\rangle|^2=0.529$ and $|\langle\chi_\downarrow|g\rangle|^2=0.453$, where $|g\rangle$ is the ground state. Such a 98% projection is rather good evidence for the polaron. Furthermore, correlation functions characteristic of the polaron can be measured. For an isolated polaron, the correlation between the two Cu spins is $\langle\chi_\uparrow|S_{\mathrm{Cu}(1)}S_{\mathrm{Cu}(2)}|\chi_\uparrow\rangle=\frac{1}{4}$ and the correlation between the O hole spin and the Cu spin is $\langle\chi_\uparrow|S_{\mathrm{O}}S_{\mathrm{Cu}(1)}|\chi_\uparrow\rangle=-\frac{1}{2}$. In the ground state of the many-body system, $\langle S_{\mathrm{Cu}(1)}S_{\mathrm{Cu}(1)}\rangle=0.24$ and $\langle S_{\mathrm{O}}S_{\mathrm{Cu}(1)}\rangle=-0.493$ as shown in Fig. 3. These values are 96% and 99% of the saturation, respectively. The characteristics can also be seen in the two-hole cases as shown in Fig. 3.

A difficulty in interpreting the polaron as a coherent propagation of a quasiparticle is the fact that the spin polaron has been scattered constantly between up and down states as it propagates [Figs. 3(d) and 3(e)]. This is reflected in the fact that $|\langle\chi_\uparrow|g\rangle^2$ is close to $|\langle\chi_\downarrow|g\rangle|^2$ and the fact that $\langle S^z_{\mathrm{O}}\rangle\sim 0$. Thus it seems that a coherent (i.e., the spin state remains fixed) quasiparticle is a combination of the spin-up and spin-down states $\chi=c_1\chi_\uparrow+c_2\chi_\downarrow$, $|c_1|^2\simeq 0.529$, $|c_2|^2\simeq 0.453$. However, within the second-order theory, we cannot determine the relative phase between the two components because $\langle g|\chi_\uparrow\rangle\langle\chi_\downarrow|g\rangle=0$ due to spin conservation. Given the phase incoherence between $\chi_\uparrow$ and $\chi_\downarrow$, polarons are essentially kinks which separate individual pieces of unfrustrated Heisenberg Cu spin systems [see Figs. 3(d) and 3(e)]. The process of kink hop is essentially the dominant spin-exchange t_c hopping (note $t_c>2t_a$), which do not cause any frustration in the spin configurations.

It is interesting to note that although the large J_{pd} motivates the formation of the three-spin polaron, the dominant factor is in fact the large spin-exchange hopping (t_c). Setting $J_{pd}=0$ still leads to a clear polaron: $\sum_\sigma|\langle\chi_\sigma|g\rangle|^2=96.8\%$. This spin-flip process is even stronger for the symmetrized singlet states, which have a hopping amplitude $(2t_c-t_a)$ instead of t_c for the nonsymmetrized up and down spin states. This brings us to an important point regarding the quasiparticle: the singlet pairing the O hole with one of the neighboring Cu spins. The ground-state projection to this singlet pair is quite high, 74%. This singlet pairing well explains the above-

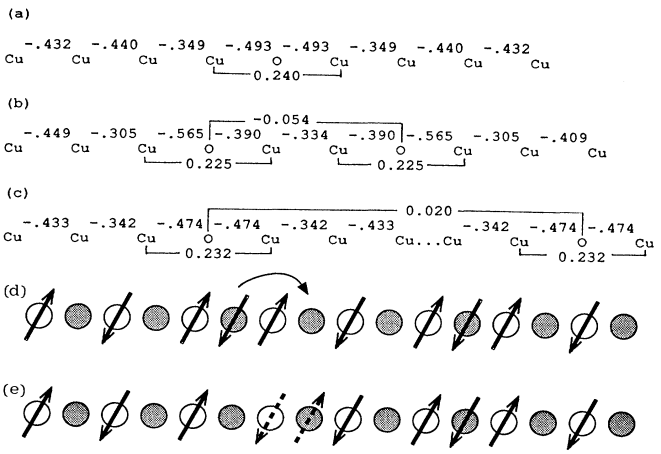

FIG. 3. Near-neighbor Cu-Cu or Cu-O spin correlations in the one-hole case (a), two-hole case when the separation is 2 (b) or 8 (c). Also shown are the hole-hole spin correlations. All correlations are normalized such that the hole configuration occurs with a probability of 1. An interpretation of (a), (b), and (c) is sketched in (d) where open (filled) circles denote Cu (O) sites. In (e) the effect of the hopping is indicated by the dashed arrow.

mentioned characteristic correlations for the polaron which, in fact, can be regarded as a symmetrized combination of the two pairs in Cu-O-Cu.

The question of binding of holes to form Cooper pairs is sometimes studied by the binding energy $E_b=E_2-2E_1+E_0$, where E_2,E_1,E_0 are the minimum energies in the two-, one-, and zero-hole cases. For the two-band model, E_b could be either positive or negative, as listed in Table I for all three parameter sets. A more direct indicator about binding is the hole-hole pair (or density-density) correlation $g(r)=\langle n_0 n_r\rangle$. This pair correlation is listed in Table I, along with those in the t-J model and those for two free spin-$\frac{1}{2}$ fermions, which are $g(r)=(2/L)\sin^2(\pi r/La)$.[16] The pair correlations of both the t-J model and the two-band model are very close to the free fermion case, although a small but qualitative difference exists. $g(r)$ in the two-band model is smaller than the free case at short distance $r\leq 4$ while greater at larger distances. This indicates a small repulsion between the two doped holes. The correlation in the t-J model behaves just opposite—the two holes in the t-J model experience a slight attraction, even though E_b indicates binding for $t/J=3.5$ but not for $t/J=7$. This small but qualitatively different behavior between the two-band model and the t-J model may be understood as the following: in the t-J model, the number of broken bonds favor the binding, i.e., the two holes tend to stay next to each other (so as to break three bonds) instead of being separated (thus breaking four bonds). This mechanism does not exist in the two-band model: the kink (polaron) formation favors no particular separation distance [see Figs. 3(d) and 3(e)]. In addition, the kinks have no frustration effect on the Cu spins. These two facts suggest that the kinks move almost freely in the 1D model, in-

TABLE I. Hole-hole density correlation on the 16-site lattice for the two-band model with parameter sets A,B,C and the t-J model with $t/J=3.5$ and 7. The binding energy is also listed. For comparison, the case of two free fermions with no double occupancy is also given.

r	1	2	3	4	5	6	7	8	E_b
Two-band	0.0043	0.0173	0.0375	0.0619	0.0865	0.1076	0.1217	0.1266	0.0093
	0.0045	0.0178	0.0381	0.0622	0.0865	0.1071	0.1209	0.1257	0.0119
	0.0045	0.0178	0.0380	0.0622	0.0865	0.1071	0.1210	0.1258	−0.1529
t-J	0.0055	0.0192	0.0395	0.0628	0.0863	0.1058	0.1191	0.1236	−0.1261
	0.0051	0.0188	0.0390	0.0627	0.0863	0.1063	0.1196	0.1243	0.0648
Free	0.0048	0.0183	0.0386	0.0625	0.0864	0.1067	0.1202	0.1250	

dependent of their spin state (up or down). In this sense, the spin degrees of freedom are separated from those of the charges. This is quite consistent with the charge (density) correlations which are very close to the free-particle case.

In conclusion, we have characterized the band, the magnetic correlations, the quasiparticle nature, and the binding of quasiparticles in the 1D two-band model. The free motion of the kinks independent of their spin state suggests that the charge-spin separation also happens in the two-band model.

This research was supported by the NSF-DMR-MRG and NSF-CHE. The facilities of the MSC are also supported by grants from DOE-AICD, BP America, Allied-Signal, Asahi Chemical, Asahi Glass, Zerox, and Chevron.

[1]P. W. Anderson and Y. Ren, in *High Temperature Superconductivity Proceedings: The Los Alamos Symposium 1989*, edited by K. Bedell *et al.* (Addison-Wesley, Reading, MA, 1990); P. A. Bares and G. Blatter, Phys. Rev. Lett. **64**, 2576 (1990); H. J. Schulz, *ibid.* **64**, 2831 (1990); N. Kawakami and S.-K. Yang, *ibid.* **65**, 2390 (1990); M. Ogata, M. U. Luchini, S. Sorella, and F. F. Assaad, *ibid.* **66**, 2388 (1991).

[2]F. C. Zhang and T. M. Rice, Phys. Rev. B **37**, 3759 (1988).

[3]V. J. Emery, Phys. Rev. Lett. **58**, 2794 (1987); V. J. Emery and G. Reiter, Phys. Rev. B **38**, 4547 (1988).

[4]P. B. Littlewood, C. M. Varma, and E. Abrahams, Phys. Rev. Lett. **60**, 379 (1987); C. M. Varma, S. Schmitt-Rink, and E. Abrahams, Solid State Commun. **62**, 681 (1887).

[5]J. E. Hirsch, Phys. Rev. Lett. **59**, 228 (1987); J. E. Hirsch, S. Tang, E. Loh, and D. J. Scalapino, Phys. Rev. B **69**, 1668 (1988).

[6]R. T. Scalettar, D. J. Scalapino, R. L. Sugar, and S. R. White, Phys. Rev. B **44**, 770 (1991); J. Wagner, W. Hanke, and D. J. Scalapino, *ibid.* **43**, 10 517 (1991).

[7]G. Kotliar, P. A. Lee, and N. Read, Physica C **153-155**, 538 (1988); J. H. Kim, K. Levin, and A. Auerbach, Phys. Rev. B **39**, 11 633 (1989); M. Grilli *et al. ibid.* **42**, 329 (1990); C. A. Balseiro *et al.* Phys. Rev. Lett. **62**, 2624 (1989).

[8]J. Zaanen and A. M. Oles, Phys. Rev. B **37**, 9423 (1988).

[9]M. Ogata and H. Shiba, J. Phys. Soc. Jpn. **57**, 3074 (1988); D. M. Frenkel, R. J. Gooding, B. I. Shraiman, and E. D. Siggia, Phys. Rev. B **41**, 350 (1990).

[10]A. K. McMahan, J. F. Annett, and R. M. Martin, Phys. Rev. B **42**, 6268 (1990).

[11]M. S. Hybertsen, E. B. Stechel, M. Schluter, and D. R. Jennison, Phys. Rev. B **41**, 11 068 (1990).

[12]Y. Guo, J-M. Langlois, and W. A. Goddard, Science **239**, 896 (1988); J-M. Langlois and W. A. Goddard (unpublished).

[13]H.-Q. Ding and M. S. Makivic, Phys. Rev. Lett. **64**, 1449 (1990); R. R. P. Singh *et al.*, *ibid.* **62**, 2736 (1989); G. Aeppli *et al.*, *ibid.* **62**, 2052 (1989).

[14]H. Q. Ding, Physica C (to be published).

[15]R. J. Birgeneau *et al.*, Phys. Rev. B **39**, 2868 (1989); G. Shirane *et al.*, Phys. Rev. Lett. **63**, 330 (1989); S.-W. Cheong *et al.*, *ibid.* **67**, 1791 (1991).

[16]The t-J model with two holes and $J=0$ have the same pair correlations.