RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 48, NUMBER 6 1 AUGUST 1993-II

Validity of the t-J model

C. D. Batista and A. A. Aligia

Comisión Nacional de Energía Atómica, Centro Atómico Bariloche, 8400 Bariloche, Argentina

(Received 20 April 1993)

We have diagonalized exactly an effective Hamiltonian $H_{\rm eff}$ that describes the low-energy eigenstates of a Cu_4O_8 cluster with five holes. Projecting these eigenstates onto Zhang-Rice states we find that for total spin $S=1/2$, they have more than 90% of local singlet character for realistic parameters. This percentage depends on S and the symmetry of the states and is larger for the ground state. The fit of the energy levels using a model similar to the t-J model, is improved substantially if the model includes a term that combines a nearest-neighbor exchange with a next-nearest-neighbor hopping.

The validity of the widely studied t-J model[1] for the description of the superconducting CuO_2 planes is still a controversial issue.[2–8] The original derivation[1] starts from the three-band Hubbard model H (Refs. 9 and 10) [see Eq. (1)], assuming $t_{pp}=U_{pd}=0$ and that the hopping t_{pd} is small. In this limit H reduces to a simpler effective Hamiltonian $H_{\rm eff}$.[2,6] Then, the Hilbert space is further reduced retaining only states in which the O states enter the many-body wave function in singlet combinations of the form $(\alpha^\dagger_{i\uparrow}d^\dagger_{i\downarrow}-\alpha^\dagger_{i\downarrow}d^\dagger_{i\uparrow})$, where the $\alpha^\dagger_{i\sigma}$ creates a hole with spin σ in the Wannier function of the O states at the Cu site i with the same symmetry of the d orbital. The O Wannier functions orthogonal to the $\alpha_{i\sigma}$, which we call $\gamma_{i\sigma}$, and the local triplets are neglected. While $H_{\rm eff}$ is a good representation of H, even for large t_{pd} if the parameters are renormalized,[11] the t-J model $H_{t\text{-}J}$ is not an exact representation of $H_{\rm eff}$ in any limit.[6]

The Cu_4O_8 cluster with periodic boundary conditions is the smaller cluster for which all atoms have the same coordination number as the corresponding ones for the infinite system. A smaller coordination of the O atoms, like in a Cu_2O_7 cluster decreases the singlet-triplet admixture.[5] In a previous study[6] we tried to fit the lowest eigenenergy of $H_{\rm eff}$ for each irreducible representation of the complete symmetry group with the corresponding one of $H_{t\text{-}J}$. We found that for one added hole, the resulting mean square deviation σ is of the order of the optimum value of t. Thus, the fit was not very good, particularly for the levels of lowest energy. Similar results were obtained in a recent paper.[7] There are two possible explanations of this failure of $H_{t\text{-}J}$. (a) The low-energy eigenstates have an important admixture of local triplet states and/or the above-mentioned O $\gamma_{i\sigma}$ states. (b) The interactions between Zhang-Rice states (local singlets) included in $H_{t\text{-}J}$ are not enough to describe the energy spectrum.

Here we report on our study of these two possibilities. For realistic parameters and minimum total spin $S=1/2$, our results favor the second one. In addition to next-nearest-neighbor (NNN) hopping t', which arises naturally in the derivation of the t-J model because of the structure of the Wannier functions $\alpha_{i\sigma}$,[1] there is another important term absent in $H_{t\text{-}J}$. It takes into account processes of second order in the nearest-neighbor (NN) α-d hopping [which is in turn a fraction of t_{pd} (Refs. 1, 12, and 13)], in which a Cu hole at a singly occupied site j, NN to a site i occupied by a singlet, jumps to another NN site l building a singlet there and then, one of the O states $\alpha_{i\sigma}$ jumps to site j leaving it again singly occupied by a Cu hole. This term, the energy of which we denote by t'' [see Eq. (2)] arises naturally from a canonical transformation which eliminates the hopping term in effective one-band Hubbard-like models used to represent the CuO_2 planes.[4,12,13] We find that the resulting modified Hamiltonian, which we call $H_{t,t',t'',J}$, for optimized parameters provides a very good fit of the energy levels of $H_{\rm eff}$ for realistic undoped and single-hole doped systems.

The three-band Hubbard model[9,10] can be written in the form

$$\begin{aligned}H=&\Delta\sum_{j\sigma}p^\dagger_{j\sigma}p_{j\sigma}+U_d\sum_i d^\dagger_{i\uparrow}d_{i\uparrow}d^\dagger_{i\downarrow}d_{i\downarrow}\\&+U_p\sum_j p^\dagger_{j\uparrow}p_{j\uparrow}p^\dagger_{j\downarrow}p_{j\downarrow}+U_{pd}\sum_{i\delta\sigma\sigma'}d^\dagger_{i\sigma}d_{i\sigma}p^\dagger_{i+\delta\sigma'}p_{i+\delta\sigma'}\\&+t_{pd}\sum_{i\delta\sigma}(p^\dagger_{i+\delta\sigma}d_{i\sigma}+\text{H.c.})-t_{pp}\sum_{j\gamma\sigma}p^\dagger_{j+\gamma\sigma}p_{j\sigma}.\end{aligned}\quad(1)$$

$d^\dagger_{i\sigma}$ ($p^\dagger_{j\sigma}$) creates a hole with spin σ on the Cu $3d_{x^2-y^2}$ (O σ) orbital at site i (j). $i+\delta$ ($j+\gamma$) label the four O atoms nearest to the Cu(O) atom at site $i(j)$. The phases of half of the orbitals have been changed in such a way that t_{pd} and t_{pp} are both positive and independent of δ and γ, respectively. To describe the low-energy eigenstates of H we use an $H_{\rm eff}$ obtained eliminating the linear term in t_{pd} by means of a canonical transformation and neglecting terms of order t^3_{pd} or higher except the Cu-Cu exchange term J.[6] For realistic parameters, it is necessary to renormalize the parameters of $H_{\rm eff}$ to obtain a good description of H.[11] The procedure used in Ref. 11 seems to work very well for undoped and single-hole doped systems, but two cautionary points should be mentioned. (a) The Cu_4O_8 cluster with periodic boundary conditions, allows an exchange process between two Cu spins of order $t^4_{pd}/(\Delta+U_{pd})^3$ (without involving either U_d or U_p), which consists of four successive Cu-O hoppings along the line joining two NN Cu atoms. Thus, the exchange interaction J is increased considerably with

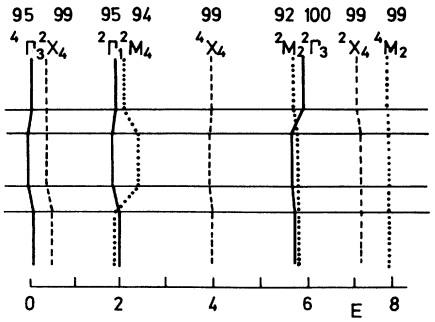

FIG. 1. Middle row: lowest energy levels of a Cu_4O_8 cluster with five holes. On top we denote the percentage of local character of the corresponding eigenstates. Bottom: best fit of these levels using the t-J model. Top row of levels: best fit using the extended t-J model $H_{t,t',t'',J}$ [Eq. (2)]. The notation for each irreducible representation is the same as in Ref. 6. The superscript denotes the spin multiplicity and Γ, X, and M denote wave vectors (0,0), $(\pi,0)$, and (π,π), respectively, with the convention of phases used in Eq. (1). Parameters are $t_{pp}=0, U_d, \Delta \to +\infty, U_p, U_{pd}$ arbitrary $t_{pd}^2/(U_d-\Delta-2U_{pd})=1 << t_{pd}$, but $J=0.1$.

respect to the thermodynamic limit. To represent better the latter we have calculated J as the difference between the energy of the triplet and the singlet of H for a Cu_2O_7 cluster with two holes. (b) In the covalent regime $\Delta \lesssim 2t_{pd}$, a four-spin cyclic exchange of leading order $t_{pd}^8/(\Delta+U_{pd})^7$ between Cu spins becomes important[14] and should be included in H_{eff}- and (t-J)-like models to accurately describe the spin dynamics.

The extended t-J model can be written as

$$H_{t,t',t'',J} = C + \epsilon \sum_{i\sigma} c_{i\sigma}^{\dagger} c_{i\sigma} + t \sum_{i\delta\sigma} c_{i+\delta\sigma}^{+} c_{i\sigma} - t' \sum_{i\gamma\sigma} c_{i+\gamma\sigma}^{\dagger} c_{i\sigma} + \frac{J}{2} \sum_{i\delta\sigma} \mathbf{S}_i \cdot \mathbf{S}_{i+\delta} - t'' \sum_{i\delta\delta'\sigma} c_{i+\delta'\sigma}^{\dagger} c_{i+\delta\sigma} \left(\frac{1}{2} - 2\mathbf{S}_i \cdot \mathbf{S}_{i+\delta} \right), \quad (2)$$

where $c_{i\sigma}^{\dagger} = \tilde{c}_{i\sigma}^{\dagger} (1 - \tilde{c}_{i\bar{\sigma}}^{\dagger} \tilde{c}_{i\bar{\sigma}})$ in terms of ordinary fermion operators, $\mathbf{S}_i$ is the spin operator at site i, and the sum

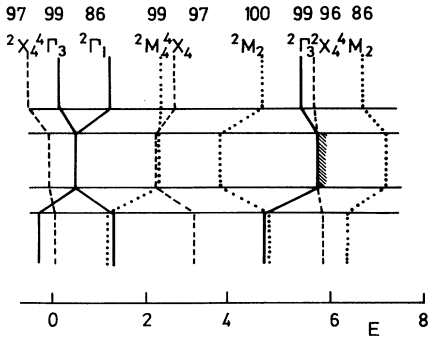

FIG. 2. Same as in Fig. 1 except for the parameters $U_p=0$, and Δ such that $t_{pd}^2/\Delta = 1 << t_{pd}$. The hashed region denotes a series of levels originated by nonbonding O orbitals, without correspondence in $H_{t\text{-}J}$.

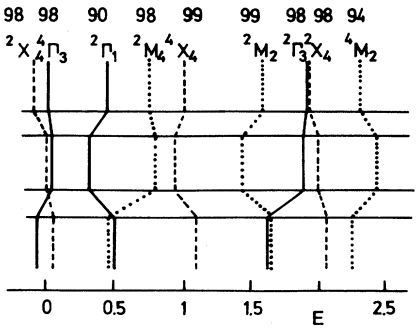

FIG. 3. Same as in Fig. 1 for $t_{pp}=U_{pd}=0$, $U_d=6, U_p=3$, and $\Delta=4$.

over δ (γ) runs over the four vectors which connect a site with each of its NN (NNN). In the square 2×2 cluster there are only two (one) NN (NNN) to each site and the sum over δ or δ' (γ) introduces a factor 2 (4).

For the undoped system, H_{eff}, $H_{t\text{-}J}$, and $H_{t,t',t'',J}$ all have the same form. Thus, C and J should be the same for all models. We have determined the remaining parameters of $H_{t\text{-}J}$ and $H_{t,t',t'',J}$ minimizing the mean square deviation σ between all the levels for one added hole and total spin projection $S_z = 1/2$ of these models and the corresponding ones E_i' of H_{eff}:

$$\sigma = \left[\frac{\sum_i (E_i - E_i')^2}{\sum_i 1} \right]^{1/2} . \quad (3)$$

The correspondence between the eigenstates of the (extended) t-J model and some of the eigenstates of H_{eff} was done in the following way: for each irreducible representation of the complete symmetry group one takes the n states of $H_{t\text{-}J}$ which transform according to it. These states are transformed into states of H_{eff} replacing the hole by a singlet composed of a Cu hole at the same site and an O hole in the symmetric orbital around it.[1] Then, the eigenstates of H_{eff} belonging to the same irreducible representation are projected onto the subspace defined by these Zhang-Rice states. Finally the n eigenstates of largest projection are assumed to correspond to the n states of the (t-J)-like model with the same order of energies.

In Figs. 1–6 we show the results of the fitting of the levels of H_{eff} using $H_{t\text{-}J}$ and $H_{t,t',t'',J}$, and the amount of Zhang-Rice states (local nonorthogonal singlets) in

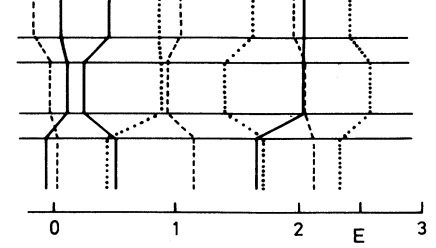

FIG. 4. Same as in Fig. 3 changing $\Delta=2$, $U_{pd}=1$.

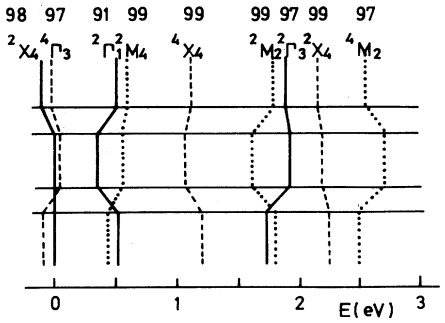

FIG. 5. Same as in Fig. 1 for $U_d = 10$ eV, $U_p = 5$ eV, $\Delta = 3$ eV, $U_{pd} = 1$ eV, $t_{pd} = 1.6$ eV, and $t_{pp} = 0.2t_{pd}$.

the eigenstates of $H_{\rm eff}$. This amount is slightly less if the symmetric linear combinations of the four O orbitals around any Cu site i (used to build the local singlets) are replaced by the Wannier functions $\alpha_{i\sigma}$. Before discussing each figure, we point out some general features of the eigenstates of $H_{\rm eff}$.

(a) The states containing the Wannier functions $\gamma_{i\sigma}$ which do not hybridize with the $d_{i\sigma}$ at the same Cu site are practically decoupled from the rest.[15] They all lie inside a narrow region of energy which except for particular parameters (Fig. 2) lies above the highest energy of the t-J model.

(b) The states with total spin $S = 5/2$ are also outside the low-energy region.

(c) All states which have a correspondence with states of the t-J model have more than 85% of local singlet character.

(d) For S=1/2, the ground state has momentum $(\pi,0)$ and 97% or more of local singlet character, while the state of largest triplet character has momentum (0,0), in agreement with Ref. 8.

(e) The fitting with $H_{t,t',t'',J}$ is better than the one with $H_{t,J}$. The variance σ of the former is lower by a factor ranging from 1/4 to 3/4. The improvement is due to the term in t'', since t' is smaller (see Table I) and the number of hopping processes associated with t' (4) is much smaller than the corresponding number for t'' (24 for a Néel background).

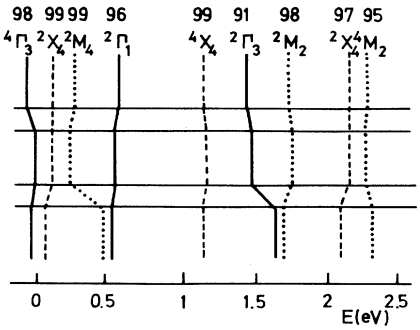

FIG. 6. Same as in Fig. 5 changing $t_{pd} = 1.3$ eV and $t_{pp} = 0.4t_{pd}$.

(f) The fitting improves with increasing t_{pp} and increasing importance of the Cu^{3+}-excited configuration with respect to the Cu^{+} configuration. Both effects increase the difference between the energy of local triplets and that of local singlets.[11]

(g) Probably because of its mixing with states of local triplet character, the $^2\Gamma_1$ and $^4X_4$ levels of $H_{\rm eff}$ lie systematically at lower energies than the corresponding levels of $H_{t,t',t'',J}$.

In Figs. 1 and 2 we represent two limiting opposite situations in which $t_{pp} = 0$ and t_{pd} is small enough to be treated perturbatively.[2,6] In both cases the Cu^{2+} configuration is stable. In the first case only fluctuations via Cu^{3+} are allowed. The fitting with $H_{t,J}$ is already good, but the separation between the levels $^2\Gamma_1$ and $^2M_4$ is always J, smaller and of opposite sign as the corresponding one for $H_{\rm eff}$. Including t'', the fitting improves but a crossing of levels at higher energies occurs. The situation represented in Fig. 2, where only fluctuations via Cu^{+} are allowed, is the worst for the (t-J)-like models. In agreement with Ref. 3, we find that the ground state of $H_{\rm eff}$ has low spin, while the ground state of $H_{t\text{-}J}$ is a Nagaoka state. Also, the state of minimum energy for S=1/2 and wave vector (0,0) has an important admixture of local triplet states, as found earlier.[8] The addition of t'' corrects most of the the crossings between the energy levels of $H_{\rm eff}$ and the corresponding ones of $H_{t\text{-}J}$, but the fitting does not improve too much: σ is reduced from 0.66 to 0.43. However, if the states of largest triplet character $^2\Gamma_1$, $^4X_4$, and $^4M_2$ are eliminated from the fit, σ is reduced to 0.19.

In Figs. 3 and 4, the comparison is made for more realistic parameters, used in Monte Carlo calculations,[16] for which the photoemission spectra calculated by H and $H_{\rm eff}$ are very similar.[11] In both cases the fitting of the levels including the t'' term in $H_{t\text{-}J}$ improves considerably. In particular the ordering of the energy levels is the right one for $H_{t,t',t'',J}$. For the parameters of Fig. 3 but with U_p=0, a canonical transformation from an effective one-band Hubbard model[13] gives $t'' = -t_{AB}^2/U = 0.020$ in reasonable agreement with the results of the fitting, taking into account the particular form of the Wannier functions in this small cluster.

Finally, in Figs. 5 and 6 we use a set of realistic param-

TABLE I. Parameters which describe the best fit of the low-energy levels of the Cu_4O_8 cluster represented in Figs. 1–6 using $H_{t,t',t'',J}$ [Eq. (1)]. J is obtained fitting the levels of the undoped system and the remaining parameters are determined adding one hole to the system. The changes that occur if $t' = t'' = 0$ is assumed are indicated between brackets. For Figs. 1–4 the units are arbitrary, while for Figs. 5 and 6 the units are eV.

Fig.	t	t'	t''	J	σ
1	0.98	0	0.020	0.10	0.12(0.16)
2	0.75(0.76)	0.092	0.116	0.10	0.43(0.66)
3	0.29	0.018	0.031	0.039	0.09(0.18)
4	0.29(0.30)	0.026	0.043	0.063	0.12(0.24)
5	0.43	0.026	0.028	0.10	0.12(0.18)
6	0.31	0.004	0.017	0.065	0.025(0.097)

eters justified elsewhere[11] which include t_{pp}. The general features of the fitting are the same as those of Figs. 3 and 4, but for important values of t_{pp} the fitting obtained with $H_{t,t',t'',J}$ is excellent. The addition of the direct spin exchange between O and Cu sites K_{pd} (Refs. 4 and 7) in Eq. (1) does not modify essentially the fitting.

In summary, the validity of the t-J model depends crucially on the parameters and the precision required. Direct O-O hopping, higher energy of states containing Cu^+, and inclusion of the term in t'' of Eq. (2) in the t-J model improve considerably the comparison of the energy levels. An analysis of the processes in t'' lost when holes are brought together suggests that this term might work against pairing, but more effectively against phase separation. However, when more than one hole is added to undoped CuO_2 planes, probably it is necessary to add more terms to $H_{t,t',t'',J}$ to represent the interaction between them. In particular when U_{pd} is large enough, there is an effective attraction between holes which is present in H (Ref. 10) and in $H_{\rm eff}$ (Ref. 17) but not in $H_{t,t',t'',J}$. It is of interest to find out if a simple modification of the latter can account for this effective attraction. Work in this direction is in progress.

We thank M. E. Simón for useful discussions. One of us (C.D.B.) was supported by the Consejo Nacional de Investigaciones Científicas y Técnicas (CONICET), Argentina. A.A.A. was partially supported by CONICET.

[1] F.C. Zhang and T.M. Rice, Phys. Rev. B **37**, 3759 (1988).
[2] V.J. Emery and G. Reiter, Phys. Rev. B **38**, 4547 (1988).
[3] R.J. Gooding and V. Elser, Phys. Rev. B **41**, 2557 (1990).
[4] M.S. Hybertsen *et al.*, Phys. Rev. B **41**, 11068 (1990).
[5] H. Eskes and G.A. Sawatzky, Phys. Rev. B **44**, 9656 (1991).
[6] C. Batista and A.A. Aligia, Solid State Commun. **83**, 419 (1992), and references therein. There was a mistake in two energy levels ($^2\Gamma_1$ of $H_{\rm eff}$ and $^2X_4$ of $H_{t\text{-}J}$), but the main conclusions are not affected.
[7] H.-Q. Ding, G.H. Lang, and W.A. Goddard III, Phys. Rev. B **46**, 14317 (1992).
[8] V.V. Kabanov and A. Vagov, Phys. Rev. B **47**, 12134 (1993), and references therein.
[9] V.J. Emery, Phys. Rev. Lett. **58**, 2794 (1987).
[10] P.B. Littlewood, C.M. Varma, and E. Abrahams, Phys. Rev. Lett. **63**, 2602 (1989), and references therein.
[11] C. Batista and A.A. Aligia, Phys. Rev. B **47**, 8929 (1993).
[12] H.B. Schüttler and A.J. Fedro, Phys. Rev. B **45**, 7588 (1992).
[13] M.E. Simón, M. Baliña, and A.A. Aligia, Physica C **206**, 297 (1993).
[14] H.J. Schmidt and Y. Kuramoto, Physica C **167**, 263 (1990).
[15] The decoupling of the states containing antisymmetric $\gamma_{i\sigma}$ Wannier functions is perfect in the Cu_4O_8 cluster if the parameters of $H_{\rm eff}$ satisfy $J_K = 2(t_1 + t_2)$, which is always approximately true (Refs. 6 and 11).
[16] G. Dopf, A. Muramatsu, and W. Hanke, Phys. Rev. Lett. **68**, 353 (1992).
[17] A.A. Aligia, Int. J. Mod. Phys. **5**, 2109 (1991).