Reduction of a three-band model for copper oxides to a single-band generalized t-J model

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A three-band model for copper oxides in the region of parameters where the second hole on the copper has energy close to the first hole on the oxygen is considered. The exact solution for one hole on a ferromagnetic background of the ordered copper spins is obtained. A general procedure for transformation of the primary Hamiltonian to the Hamiltonian of singlet and triplet excitations is proposed. Reduction of the singlet-triplet Hamiltonian to the single-band Hamiltonian of the generalized t-J model is performed. A comparison of the solution for the generalized t-J model on a ferromagnetic background with the exact solution shows very good agreement.

I. INTRODUCTION

Some time ago the extended Hubbard model or the Emery model was proposed for a description of holes in the CuO_2 plane.[1] The next essential step was made by Zhang and Rice.[2] They proposed that holes on the oxygen move over the crystal in the form of spin-singlets formed with the copper spins and can be described by the single-band t-J model. It should be noted that consistent reduction of the three-band model Hamiltonian to the single-band model t-J Hamiltonian was not presented in Ref. 2. Therefore a polemic concerning the validity of the t-J model has arisen.

In the work by Emery and Reiter,[3] the exact solution of the three-band model on a ferromagnetic background of Cu spins was obtained. They have shown that this exact solution can be interpreted in the region of small momenta as the motion of a spin triplet formed by the O hole and two adjacent Cu spins. Zhang and Rice[4] have shown that the exact solution on a ferromagnetic background can be interpreted as motion of the local spin singlet. In the work by Zhang[5] it has been shown that the spectra of the t-J model and of the three-band model are identical: if the eigenstate of the t-J model is known one can construct the eigenstate of the three-band model with the same energy with the help of the local spin-singlet. However, this does not mean the physical equivalence of the two models because the wave functions of the local spin singlets are not orthogonal, as was stressed by Emery and Reiter.[6]

The effective Hamiltonian in terms of singlet and triplet operators was obtained by Shen and Ting.[7] The contribution of the triplet state was estimated to be of the order of 10% on an antiferromagnetic background. This value determines the precision of the single-band approximation. Notice that it is sufficiently difficult to recognize the Hamiltonian of the t-J model in the final formula of Ref. 7. The work by Pang, Xiang, Su, and Yu[8] was devoted to the construction of the singlet and triplet states and to a comparison of the hopping parameters on a ferromagnetic background with the exact solution.[3] A sufficiently good agreement was obtained.

All above-named works were dealing with parameters of the Emery model[1] in the region $U_d - \epsilon, \epsilon \gg |t|$, where U_d is the Coulomb repulsion at the Cu site, ϵ is the difference in energy between the O($2p$) and Cu($3d$) holes, and t is the Cu-O hopping parameter. This condition means that the energy of the p_x, p_y oxygen levels lies between and sufficiently far from the energy of the $d_{x^2-y^2}$ copper levels split by the Coulomb repulsion U_d.

A more accurate estimation (see work by Lovtsov and Yushankhai[9] and this work below) shows that in fact the condition of applicability of the perturbation theory is more rigid: $U_d - \epsilon, \epsilon \gg 4\sqrt{2}|t|$. Different band calculations[10,11] give $t \approx -1.4$ eV and the perturbation theory over $|t|$ for computing the properties of charge carriers works at U_d >16 eV. Known estimations[10,11] give $U_d \lesssim 8$ eV. The situation is simpler if oxygen levels are close to the lower or the higher $d_{x^2-y^2}$ copper level. We use the hole classification of the energy levels. The case $\epsilon \ll U_d$ was considered in the work by Lovtsov and Yushankhai,[9] where local singlet and triplet states were constructed and the hopping of these states over the crystal was studied.

In this work we study the case $U_d - \epsilon \ll U_d$ when the oxygen level is close to the higher copper $d_{x^2-y^2}$ level. Such level position was proposed as a result of band calculations in the work by Flambaum and Sushkov[11] and does not contradict the photoemission data.[12] Actually, the difference in position between the lower $d_{x^2-y^2}$ and the p_x, p_y levels is approximately 4 eV.[12] For U_d =8 eV the p_x, p_y levels are in the middle between the split Cu $d_{x^2-y^2}$ levels, but for $U_d = 6$ eV, as proposed in Ref. 11, the O p_x, p_y levels are closer to the higher $d_{x^2-y^2}$ level. In this work the direct oxygen-oxygen hopping is not taken into account.

The work can be divided into three parts. In the first part we will get the exact solution of the three-band model on a ferromagnetic background and discuss its properties. This solution is an analog of the corre-

sponding solution of Emery and Reiter.[3] The consideration of the hole motion on a ferromagnetic background is of certain methodical interest. This solution is exact but it does not describe the ground state but the high-excitation state. Such a solution is used for testing the approximate Hamiltonian of the generalized t-J model obtained in the present work from the three-band Hamiltonian. This allows us to make a simple estimation of the magnitude of corrections to the t-J model that appear in the reduction of the three-band model. An estimation of such corrections for the hole motion on an antiferromagnetic background of the copper spins represents a separate problem.

In the second part of the work we transform the three-band Hamiltonian to the Hamiltonian describing hopping and transition between two singlet and one triplet state. These singlet and triplet states are formed by the spin of the hole and the spin of the copper. For performing the transformation the technique of representation of the Hubbard operators in terms of the hole and the spin-$\frac{1}{2}$ operators was used. This Hamiltonian, also containing three bands (two singlet and one triplet), with the help of the Schrieffer-Wolff transformation is reduced to the low-energy Hamiltonian for the lower singlet. It is the Hamiltonian of the generalized t-J model.

In the third part of the work a detailed comparison of the properties of the generalized t-J model on a ferromagnetic background with the exact solution of the three-band model on a ferromagnetic background is made. Corrections to the t-J model Hamiltonian providing an agreement with the exact result are estimated. An excellent agreement between the approximate and exact solutions is shown.

In the Appendix corrections to a single-band Hamiltonian of third- and fourth-order over nondiagonal hopping terms in the case $U_d = \epsilon$ are derived.

II. EXACT SOLUTION FOR THE THREE-BAND MODEL ON A FERROMAGNETIC BACKGROUND

A. Three-band Hamiltonian and the exact solution

We want to remind the reader that the Hamiltonian of the t-J model is usually represented in the form

$$\begin{aligned} &H_{t\text{-}J} = t\sum_{\langle ll'\rangle,\alpha} \tilde{c}^{\dagger}_{l\alpha}\tilde{c}_{l'\alpha} + J\sum_{ll'} \mathbf{S}_l\cdot\mathbf{S}_{l'},\\ &\tilde{c}_{l\alpha} = c_{l\alpha}(1-\hat{n}_{l,-\alpha}),\quad \tilde{c}^{\dagger}_{l\alpha} = (\tilde{c}_{l\alpha})^{\dagger},\\ &\mathbf{S}_l = (1/2)c^{\dagger}_l\sigma c_l,\quad \hat{n}_{l\alpha} = c^{\dagger}_{l\alpha}c_{l\alpha}. \end{aligned} \tag{2.1}$$

Here $c^{\dagger}_{l\alpha}, c_{l\alpha}$ are the electron creation and annihilation operators at the lattice site l, $\alpha = \uparrow, \downarrow$ or $\pm\frac{1}{2}$ is the spin projection, σ are Pauli matrices, the symbol $\langle ll'\rangle$ denotes summation over the nearest-neighbors, t is the hopping integral, and J is the superexchange energy. It will be more convenient for us to use another form of representation of the Hamiltonian (2.1) in terms of Hubbard operators:

$$H_{tJ} = E_0\sum_l X^{00}_l + t\sum_{\langle ll'\rangle} X^{\alpha 0}_l X^{0\alpha}_{l'} + J\sum_{\langle ll'\rangle}\mathbf{S}_l\mathbf{S}_{l'}. \tag{2.2}$$

Here X^{ab}_l are Hubbard operators at the site l: $X^{ab}_l = |al\rangle\langle lb|$ for the states $|a\rangle, |b\rangle = |0\rangle, |\alpha\rangle$. The connection between the Hamiltonians (2.1), (2.2) is given by the following relations:

$$\begin{aligned} &X^{\alpha 0}_l \Rightarrow \tilde{c}^{\dagger}_{l\alpha},\quad X^{0\alpha}_l \Rightarrow \tilde{c}_{l\alpha},\\ &\mathbf{S}_l = (1/2)\sigma_{\alpha\beta}X^{\alpha\beta}_l \Rightarrow (1/2)c^{\dagger}_l\sigma c_l. \end{aligned} \tag{2.3}$$

We have added in Eq. (2.2) the first term, which describes the energy of the quenched hole.

In the case of half filling, the Hamiltonian (2.2) reduces to the Heisenberg Hamiltonian, and for $J > 0$ the antiferromagnetic state is its ground state. However, the ferromagnetic state is an eigenstate of this Hamiltonian and we can easily get a simple exact eigenstate $|\mathbf{k}\rangle$ for H_{tJ} with one hole over the ferromagnetic background,

$$\begin{aligned} &|\mathbf{k}\rangle = \sum_l \exp(i\mathbf{k}\cdot\mathbf{r}_l)X^{0\downarrow}_l|f\rangle,\qquad |f\rangle = \prod_l c^{\dagger}_{l\downarrow}|0\rangle\\ &\epsilon_{\mathbf{k}} = E_0 + 4t\gamma_{\mathbf{k}},\qquad \gamma_{\mathbf{k}} = (1/2)[\cos(k_x a) + \cos(k_y a)], \end{aligned} \tag{2.4}$$

where $|f\rangle$ is the ferromagnetic state at half filling and all electron spins down, and $\epsilon_{\mathbf{k}}$ is the electron energy. We will construct the exact solution for the state with one hole over a ferromagnetic background for the three-band model in the region of parameters discussed above.

The Hamiltonian has the form

$$\begin{aligned} H = {}&\epsilon^0_d\sum_{l,\alpha} n^d_{l\alpha} + \epsilon^0_p\sum_{m,\alpha} n^p_{m\alpha} + U_d\sum_l n^d_{l\uparrow}n^d_{l\downarrow}\\ &+V\sum_{\langle lm\rangle\alpha\beta} n^d_{l\alpha}n^p_{m\beta} + t\sum_{\langle lm\rangle\alpha}(d^{\dagger}_{l\alpha}p_{m\alpha} + p^{\dagger}_{m\alpha}d_{l\alpha}), \end{aligned} \tag{2.5}$$

where $d^{\dagger}_{l\alpha}(d_{l\alpha})$ creates (annihilates) the $d_{x^2-y^2}$ hole of spin projection α at the Cu site l and $p^{\dagger}_{m\beta}(p_{m\beta})$ creates (annihilates) the p_β hole of spin projection β at the O site m, $n^d_{l\alpha} = d^{\dagger}_{l\alpha}d_{l\alpha}, n^p_{m\beta} = p^{\dagger}_{m\beta}p_{m\beta}$. The sign convention in the last term of Eqs. (2.5) corresponds to the change of the signs of wave functions in all the odd cells, which corresponds to the quasimomentum redefinition $(k_x, k_y) \to (k_x + \pi/a, k_y + \pi/a)$.

In the case of one hole over unit filling of the $d_{x^2-y^2}$ copper states at each site, one can get the reduced Hamiltonian. Using the representation of $d^{\dagger}_{l\alpha}, d_{l\alpha}$ in terms of the Hubbard operators $X^{\alpha 0}_l, X^{0\alpha}_l, X^{\alpha 2}_l, X^{2\alpha}_l$

$$d^{\dagger}_{l\alpha} = X^{\alpha 0}_l + 2\alpha X^{-\alpha 2}_l,\quad d_{l\alpha} = X^{0\alpha}_l + 2\alpha X^{2-\alpha}_l \tag{2.6}$$

and omitting the contribution of the $X^{\alpha 0}_l, X^{0\alpha}_l$ operators, one can get

$$\begin{aligned} H_{pd} = {}&\epsilon_p\sum_{m\alpha} n^p_{m\alpha} + \epsilon_d\sum_l X^{22}_l\\ &+t\sum_{\langle lm\rangle\alpha} 2\alpha(p^{\dagger}_{m\alpha}X^{-\alpha 2}_l + X^{2-\alpha}_l p_{m\alpha}), \end{aligned} \tag{2.7}$$

where $\epsilon_d = \epsilon_d^0 + U_d$ and $\epsilon_p = \epsilon_p^0 + 2V$ are the renormalized energies of the d and p states, $X_l^{22} = |2l\rangle\langle l2|$, $X_l^{\alpha 2} = |1\alpha l\rangle\langle l2|$, $X_l^{2\alpha} = |2l\rangle\langle l\alpha|$ are the Hubbard operators for the $d_{x^2-y^2}$ Cu states at site l. The Hubbard operators $X_l^{22}, X_l^{\alpha 2}, X_l^{2\alpha}$ can be expressed in terms of the d operators of the copper

$$X_l^{22} = n_{l\uparrow}^d n_{l\downarrow}^d, \quad X_l^{\alpha 2} = d_{l\alpha}^\dagger d_{l\downarrow} d_{l\uparrow}, \quad X_l^{2\alpha} = d_{l\uparrow}^\dagger d_{l\downarrow}^\dagger d_{l\alpha}. \tag{2.8}$$

The Hamiltonian in the form similar to (2.5) was used in many works where the slave boson (fermion) method was applied to the three-band model.[13–16] We will show that eigenstates of the Hamiltonian (2.5) on a ferromagnetic background can be represented in the form

$$\begin{aligned} &|\mathbf{k}\rangle = \sum_l \exp(i\mathbf{k}\cdot\mathbf{r}_l)\hat{Y}_l(\mathbf{k})|f\rangle, \quad |f\rangle = \prod_l d_{l\downarrow}^\dagger|0\rangle, \\ &\hat{Y}_l(\mathbf{k}) = (\alpha(\mathbf{k})d_{l\uparrow}^\dagger d_{l\downarrow}^\dagger + \beta(\mathbf{k})\pi_l)d_{l\downarrow}, \quad |pl\rangle = \pi_l d_{l\downarrow}|f\rangle, \\ &\pi_l = (1/\sqrt{2})(P_{l\uparrow}^\dagger d_{l\downarrow}^\dagger - P_{l\downarrow}^\dagger d_{l\uparrow}^\dagger), \quad P_{l\alpha} = (1/2)\sum_{m\in\langle l\rangle} p_{m\alpha}, \end{aligned} \tag{2.9}$$

where $\langle l\rangle$ are the nearest-neighbor sites to site l, $|0\rangle$ is the vacuum state of the CuO_2 plane that corresponds to the completely filled $3d^{10}$ shell of Cu and the $2p^6$ shell of O. The energy of this state $|\mathbf{k}\rangle$ is equal to

$$\begin{aligned} &\epsilon(\mathbf{k}) = \bar{\epsilon} - R(\mathbf{k}), \quad R(\mathbf{k}) = \sqrt{\Delta^2 + 8t^2\eta^2(\mathbf{k})}, \\ &\bar{\epsilon} = (\epsilon_d + \epsilon_p)/2, \quad \Delta = (\epsilon_d - \epsilon_p)/2, \\ &\eta(\mathbf{k})^2 = 1 + (1/2)\gamma_{\mathbf{k}}, \quad \gamma_{\mathbf{k}} = (1/2)[\cos(k_x a) + \cos(k_y a)], \end{aligned} \tag{2.10}$$

where a is the distance between the Cu sites. Below we will count the energy from $\bar{\epsilon}$. The coefficients $\alpha(\mathbf{k}), \beta(\mathbf{k})$ have the form

$$\begin{aligned} &\alpha(\mathbf{k}) = -\sqrt{(R(\mathbf{k}) - \Delta)/2R(\mathbf{k})}, \\ &\beta(\mathbf{k}) = \eta(\mathbf{k})^{-1}\sqrt{(R(\mathbf{k}) + \Delta)/2R(\mathbf{k})} \end{aligned} \tag{2.11}$$

and satisfy the normalization condition

$$|\alpha(\mathbf{k})|^2 + \eta(\mathbf{k})^2|\beta(\mathbf{k})|^2 = 1. \tag{2.12}$$

The normalization condition (2.12) has a nontrivial form due to the fact that the states $|pl\rangle$ are not orthogonal

$$\langle pl'|pl\rangle = \delta_{ll'} + (1/8)\delta_{\langle ll'\rangle}, \tag{2.13}$$

where $\delta_{\langle ll'\rangle} = 1$ if l and l' are the nearest-neighbors and vanish otherwise. One can explicitly prove that the state (2.9) is an eigenstate of the Hamiltonian (2.7). Acting by the Hamiltonian (2.7) on the state $|\mathbf{k}\rangle$ given by (2.9) one can get the expression (2.11) for the coefficients $\alpha(\mathbf{k}), \beta(\mathbf{k})$.

B. Interpretation of the exact solution

This exact solution can be interpreted as the Bloch wave formed by the linear combination of two local singlets. One local singlet represents two holes on Cu. The other singlet consists of one hole on Cu and another hole on O (or more accurately of coherent sum of the hole states on the oxygen nearest the copper). The structure of the CuO_2 plane is shown in Fig.1.

Notice that at $\mathbf{k} \to 0$ the solution (2.10) can be represented in the form of Emery and Reiter triplets[3]

$$|\mathbf{k}\rangle = \sum_{m,j=\pm 1} [\gamma d_{mj\uparrow}^\dagger + \delta(p_{m\uparrow}^\dagger - p_{m\downarrow}^\dagger d_{mj\uparrow}^\dagger d_{mj\downarrow})]|f\rangle$$

$$\text{at } \mathbf{k} \to 0, \tag{2.14}$$

where summation over m is produced over all oxygen atoms in the CuO_2 plane and over j on two adjacent to the oxygen copper atoms for $j = \pm 1$. However, the solution (2.9) cannot be represented in the form (2.14) for all $\mathbf{k}$.

The solution represents the sum of the overlapping singlets. Due to this overlapping a spin density matrix ρ_O of an oxygen hole has a nontrivial form

$$\rho_O = (1/2)\{1 + [\gamma_{\mathbf{k}}/(2 + \gamma_{\mathbf{k}})]\sigma_z\}, \tag{2.15}$$

where $\gamma_{\mathbf{k}}$ is determined by Eq. (2.10) and σ_z is the Pauly matrix. An average of oxygen hole spin obtained with the help of the density matrix ρ_O is equal to $1/6$ at $\mathbf{k} \to 0$. This result corresponds to the calculations of Refs. 3 and 11.

In the region of small $\mathbf{k}$ we have the following expression for the energy of a singlet polaron:

$$\epsilon_{\mathbf{k}} = -R + (t^2/2R)k^2a^2 + \cdots, \quad R = \sqrt{\Delta^2 + 12t^2}. \tag{2.16}$$

The gain in energy in (2.16) is sufficiently high: 12 is a large number. According to the estimations of Refs. 10–12, $\Delta \simeq 1$–2 eV, $t \simeq -1.4$ eV, and the perturbation theory over t/Δ does not work: $\Delta^2 \le 4(\text{eV})^2$, $12t^2 \simeq 24(\text{eV})^2$, and $12t^2 \gg \Delta^2$, and we have at the small $\mathbf{k}$

$$\begin{aligned} &\epsilon_{\mathbf{k}} = -2\sqrt{3}t + t_{\text{eff}}k^2a^2, \\ &t_{\text{eff}} = t/4\sqrt{3} = 0.1443t \simeq 0.2 \text{ eV}. \end{aligned} \tag{2.17}$$

The band width w is equal to

$$w = R - \sqrt{\Delta^2 + 4t^2} \simeq 2(\sqrt{3} - 1)t = 1.469t, \tag{2.18}$$

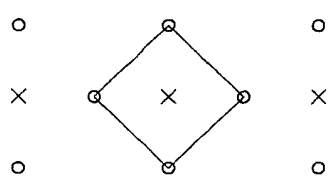

FIG. 1. The structure of the CuO_2 plane. The crosses denote copper, circles denote oxygen. The local cluster is separated by solid lines. The hole on the oxygen on the solid lines constitutes the coherent state, which forms the local singlet.

which is 1.25 times larger than the naive band width $2xt_{\rm eff} = 2t/\sqrt{3}$. These conclusions agree with results of Flambaum and Sushkov obtained by variational method.[11]

C. Reformulation of the exact solution

In this section we reformulate the exact solution in other notations that will be of further use. For this we produce a map of three Cu states at every site $|1\downarrow\rangle, |1\uparrow\rangle, |2\rangle = |\uparrow\downarrow\rangle$ into eight spin-hole states: $|0,\alpha\rangle, |1\beta,\alpha\rangle, |2,\alpha\rangle$ where $\alpha,\beta = \pm 1/2$ or $\downarrow,\uparrow$ are the spin-$\frac{1}{2}$ projections and the first index is the number of holes. One can introduce the Fermi operators for holes $h^\dagger_\alpha, h_\alpha$ and the spin-$\frac{1}{2}$ operators $\mathbf{s}$; then

$$s_z|0\alpha\rangle = \alpha|0\alpha\rangle, \quad |1\beta,\alpha\rangle = h^\dagger_\beta|0\alpha\rangle, \quad |2\alpha\rangle = h^\dagger_\uparrow h^\dagger_\downarrow|0\alpha\rangle. \tag{2.19}$$

The map has the following form:

$$\begin{aligned} &|1\alpha\rangle \Rightarrow |0\alpha\rangle, \\ &|2\rangle \Rightarrow |s\rangle \equiv (1/\sqrt{2})[|1\uparrow,\downarrow\rangle - |1\downarrow,\uparrow\rangle], \end{aligned} \tag{2.20}$$

where $|s\rangle$ is the hole-spin singlet state. This map generates the following representation for Hubbard operators on copper (2.8) (Ref. 17) in terms of $h^\dagger_\alpha, h_\alpha, \mathbf{s}$:

$$\begin{aligned} &X^{2\alpha} = \sqrt{2}2\alpha(h^\dagger\hat{s})_{-\alpha}(1-\hat{n}^h), \quad \hat{s} = (1-2\mathbf{s}\sigma)/4, \\ &X^{\alpha 2} = \sqrt{2}2\alpha(1-\hat{n}^h)(\hat{s}h)_{-\alpha}, \quad \hat{n}^h = (h^\dagger h), \quad \hat{d}^h = h^\dagger_\uparrow h_\uparrow h^\dagger_\downarrow h_\downarrow, \\ &X^{22} = h^\dagger(1-\hat{n}^h)\hat{s}h, \quad \hat{N} \equiv X^{\uparrow\uparrow} + X^{\downarrow\downarrow} = 1 - \hat{n}^h + \hat{d}^h. \end{aligned} \tag{2.21}$$

This representation can be used for a description of one-hole states if we omit the multiplier $(1-\hat{n}^h)$ in Eq. (2.21) for $X^{2\alpha}$, $X^{\alpha 2}$, and X^{22} that is essential for describing the two-hole states $|2\alpha\rangle$ (2.19). Substituting the representation (2.21) for $X^{2\alpha}$, $X^{\alpha 2}$, and X^{22} in the Hamiltonian (2.7), we can obtain a Hamiltonian in more usual terms

$$\begin{aligned} H_{pd} = {} & \epsilon_p \sum_{m,\alpha} n^p_{m\alpha} + \epsilon_d \sum_l h^\dagger_l \hat{s}_l h_l \\ & + \sqrt{2}t \sum_l (h^\dagger_l \hat{s}_l P_l + P^\dagger_l \hat{s}_l h_l). \end{aligned} \tag{2.22}$$

This Hamiltonian contains the operators of holes $p^\dagger_{m\alpha}, p_{m\alpha}$ at the O sites, the operators of holes $h^\dagger_\alpha, h_\alpha$ at the Cu sites, and the operators of spin-$\frac{1}{2}$ $\mathbf{s}$ at the Cu sites. These operators act on the ground state where there is a spin-$\frac{1}{2}$ at every Cu site. The operator $\hat{s}_l$ represents a projector on the singlet state in the one-particle sector. The eigenstate of the Hamiltonian (2.7),(2.22) can be represented as the sum of two singlets

$$\begin{aligned} &|\mathbf{k}\rangle = \sum_l \exp(i\mathbf{k}\cdot\mathbf{r}_l)\hat{Z}_l(\mathbf{k})|f\rangle, \\ &\hat{Z}_l(\mathbf{k}) = \alpha(\mathbf{k})\hat{S}^d_l + \beta(\mathbf{k})\hat{S}^p_l, \\ &\hat{S}^d_l = (1/\sqrt{2})(h^\dagger_{l\uparrow} - h^\dagger_{l\downarrow}s^\dagger_l), \\ &\hat{S}^p_l = (1/\sqrt{2})(P^\dagger_{l\uparrow} - P^\dagger_{l\downarrow}s^\dagger_l), \end{aligned} \tag{2.23}$$

where $|f\rangle$ is the ground state without holes with all Cu spins $\mathbf{s}$ having the down projection. The state (2.9) in the form (2.23) is explicitly a one-particle state. One can easily prove that the state (2.23) is the eigenstate of the Hamiltonian (2.7) in the form (2.22). For this we note that the operator $\hat{Z}_l(\mathbf{k})$ can be represented in the form

$$\hat{Z}_l(\mathbf{k}) = \sqrt{2}[\alpha(\mathbf{k})h^\dagger_l + \beta(\mathbf{k})P^\dagger_l]\hat{s}_{l\downarrow}, \quad H|f\rangle = 0. \tag{2.24}$$

Then commuting the Hamiltonian (2.22) with the operator $\hat{Z}_l(\mathbf{k})$ (2.24) and using the identity

$$\hat{s}_l\hat{s}_{l'}|f\rangle = (1/2)\hat{s}_l|f\rangle \quad \text{for } l \neq l', \tag{2.25}$$

we can obtain the expression (2.11) for the coefficients $\alpha(\mathbf{k}), \beta(\mathbf{k})$.

III. REDUCTION TO THE GENERALIZED t-J MODEL

A. Transformation of the Hamiltonian to the Hubbard form

For deduction of the low-energy Hamiltonian it will be convenient to transform the primary Hamiltonian (2.7) to the form containing exclusively the Hubbard operators. Such transformation is based on a solution of the local or cluster problem for one electron or hole, when cluster energy levels and cluster eigenfunctions are found. After this we can express all operators contained in the primary Hamiltonian (2.7), such as $X^{22}_l, X^{\alpha 2}_l, X^{2\alpha}_l, P^\dagger_{l\alpha}, P_{l\alpha}$, through the Hubbard operators $X^{a'b'}_l = |a'\rangle\langle b'|$ characterizing cluster system, where $|a'\rangle, |b'\rangle$ are eigenfunctions of the cluster problem. Since only low-energy levels of the cluster problem are essential for description of low-energy excitations, such transformation creates the basis for such a description.

For realization of the program described above, let us introduce the Wannier representation for the oxygen hole operators $P^\dagger_{l\alpha}, P_{l\alpha}$ (Ref. 2)

$$\begin{aligned} &P_l = \sum_{l'} \lambda(l,l')q_{l'}, \quad P^\dagger_l = \sum_{l'} \lambda(l,l')q^\dagger_{l'}, \\ &\lambda(l,l') = \sum_{\mathbf{k}} \sqrt{(1+\gamma_{\mathbf{k}})}\exp[i\mathbf{k}(\mathbf{r}_l - \mathbf{r}_{l'})]. \end{aligned} \tag{3.1}$$

Since the Wannier-oxygen operators $q_l, q_l^\dagger$ are independent at different sites, the primary Hamiltonian (2.7) can be expressed through them. After this, the local or cluster problem can be solved. But we will use another method of deduction of the extended Hubbard Hamiltonian. This method is based on the use of the representation (2.21) for Hubbard operators. Hence we substitute the representation (3.1) for $P_{l\alpha}^\dagger, P_{l\alpha}$ into the Hamiltonian (2.22) and get

$$H_{pd} = \epsilon_p \sum_l (q_l^\dagger q_l) + \epsilon_d \sum_l h_l^\dagger \hat{s}_l h_l + 2\sqrt{2}t \sum_{ll'} \lambda(l,l')(h_l^\dagger \hat{s}_l q_{l'} + q_{l'}^\dagger \hat{s}_l h_l). \quad (3.2)$$

For solving the one-site problem we divide the operators $q_{l\alpha}^\dagger, q_{l\alpha}$ into the singlet and triplet parts

$$q_l = q_l^s + q_l^t, \quad q_l^s = \hat{s}_l q_l, \quad q_l^t = \hat{t}_l q_l, \quad (3.3)$$
$$\hat{s}_l = (1/4)(1 - 2\mathbf{s}_l\sigma), \quad \hat{t}_l = (1/4)(3 + 2\mathbf{s}_l\sigma), \quad \hat{t}_l + \hat{s}_l = 1.$$

Then the one-site Hamiltonian has a simple quadratic form

$$H_{pd}^0 = \sum_l [\ \epsilon_p q_l^\dagger \hat{s}_l q_l + \epsilon_d h_l^\dagger \hat{s}_l h_l + \epsilon_p q_l^\dagger \hat{t}_l q_l + 2\sqrt{2}t\lambda_0 (h_l^\dagger \hat{s}_l q_l + q_l^\dagger \hat{s}_l h_l)] \quad (3.4)$$

and can be easily diagonalized

$$H_{pd}^0 = \sum_l (E_- c_l^\dagger \hat{s}_l c_l + E_+ b_l^\dagger \hat{s}_l b_l + \epsilon_p q_l^\dagger \hat{t}_l q_l), \quad (3.5)$$

where $E_\pm = \pm r$ with $r = \sqrt{\Delta^2 + 8\lambda_0^2 t^2}$ and $\lambda_0 \equiv \lambda_{00}$. New Fermi operators $c_l^\dagger, c_l, b_l^\dagger, b_l$ have the form

$$c_l = bq_l - ah_l, \quad b_l = aq_l + bh_l, \quad (3.6)$$

where $a = 2\sqrt{2}t\lambda_0/\sqrt{2r(r+\Delta)}$, $b = \sqrt{(r+\Delta)/2r}$. The additional part of the Hamiltonian $H_{pd}^{\rm int}$ can be represented in the form

$$H_{pd}^{\rm int} = H_{pd}^1 + H_{pd}^2,$$
$$H_{pd}^1 = -4\sqrt{2}abt \sum_{ll'} \lambda_{ll'} (c_l^\dagger \hat{s}_l \hat{s}_{l'} c_{l'} - b_l^\dagger \hat{s}_l \hat{s}_{l'} b_{l'}), \quad (3.7)$$
$$H_{pd}^2 = 2\sqrt{2}t \sum_{ll'} \lambda_{ll'} [\ (b^2 - a^2) c_l^\dagger \hat{s}_l \hat{s}_{l'} b_{l'} - a c_l^\dagger \hat{s}_l \hat{t}_{l'} q_{l'} + b b_l^\dagger \hat{s}_l \hat{t}_{l'} q_{l'} + \text{H.c.}].$$

Here and below we will separate λ_0 from $\lambda_{ll'}$ for $l \neq l'$ and suppose that all summations over l, l' are performed for $l \neq l'$. The Hamiltonians H_{pd}^0 and H_{pd}^1 sufficiently correctly describe the lower c-singlet band position and c-singlet hopping to the nearest-neighbor sites. If we exclude the double occupancy of the c-singlet sites, we can reduce this part of the Hamiltonian H_{pd}^0 and H_{pd}^1 to the Hamiltonian of the t-J model (2.2) with the nearest-neighbor hopping. For this we estimate the energy of the third additional hole on the elementary Cu-O plaque. Choosing the hole wave function in the form

$$|3l\alpha\rangle = \xi_3 P_{l\alpha}^\dagger d_{l\uparrow}^\dagger d_{l\downarrow}^\dagger |0\rangle + \eta_3 d_{l\alpha}^\dagger P_{l\uparrow}^\dagger P_{l\downarrow}^\dagger |0\rangle \quad (3.8)$$

and solving a simple variational problem, we can get for the energy of the three-hole state

$$E_3 = V/2 + U_p/8 - \sqrt{(\Delta + V/2 - U_p/8)^2 + 4t^2\lambda_0^2}. \quad (3.9)$$

At $V = U_p = 0$ the energy E_3 almost coincides with the top of the singlet band on a ferromagnetic background, and the constants V and U_p give an additional gap.

Due to this estimation we can neglect the contribution of the three-hole state in low-energy physics and rewrite the Hamiltonian H_{pd} in terms of the Hubbard operators. For this we introduce Hubbard operators connected with the triplet states:

$$X^{\pm 1\alpha} = (1/4)q^\dagger [(1 \pm 2s^z)(1 \pm \sigma^z) + s^\pm \sigma^\pm]_\alpha (1 - \hat{n}^q),$$
$$X^{0\alpha} = (1/2\sqrt{2})q^\dagger [1 - 4s^z\sigma^- + 2s\sigma]\sigma_\alpha^x (1 - \hat{n}^q), \quad (3.10)$$
$$X^{\pm 1\pm 1} = (1/4)q^\dagger (1 - \hat{n}^q)(1 \pm 2s^z)(1 \pm \sigma^z)q,$$
$$X^{00} = (1/4)q^\dagger (1 - \hat{n}^q)[1 - 4s^z\sigma^- + 2s\sigma]q.$$

Here the operators $X^{\mu\alpha}$ for $\alpha = \pm 1/2$ and $\mu = 0, \pm 1$ transform the spin-$\frac{1}{2}$ state with projection α into the triplet spin-hole state with projection μ and $X^{\alpha\mu} = (X^{\mu\alpha})^+$. The operators $X^{\mu\mu}$ act inside the triplet states.

Substituting expressions of the operators $c_l^\dagger \hat{s}_l, \hat{s}_l c_l, b_l^\dagger \hat{s}_l, \hat{s}_l b_l, q_l^\dagger \hat{t}_l, \hat{t}_l q_l$ in terms of the Hubbard operators, after some calculations we get the following expression for the Hamiltonian H_{pd}:

$$H_{pd}^0 = \sum_l (E_- X_l^{cc} + E_+ X_l^{bb}) + \epsilon_p \sum_{l\mu} X_l^{\mu\mu},$$
$$H_{pd}^1 = -2\sqrt{2}abt \sum_{ll'} \lambda_{ll'} (X_l^{c\alpha} X_{l'}^{\alpha c} - X_l^{b\alpha} X_{l'}^{\alpha b}), \quad (3.11)$$
$$H_{pd}^2 = \sqrt{2}t \sum_{ll'} \lambda_{ll'} [\ (b^2 - a^2) X_l^{c\alpha} X_{l'}^{\alpha b} - \sqrt{3}a(\alpha\beta\mu) X_l^{c\beta} X_{l'}^{\alpha\mu} + \sqrt{3}b(\alpha\beta\mu) X_l^{b\beta} X_{l'}^{\alpha\mu} + \text{H.c.}].$$

Here the Hubbard operators $X_l^{cc}, X_l^{bb}, X_l^{c\alpha}, X_l^{\alpha c}, X_l^{b\alpha}, X_l^{\alpha b}$ are determined by Eq. (2.21) if we replace h operators by c and b operators, respectively. The operators $X_l^{\mu\mu}, X_l^{\mu\alpha}, X_l^{\alpha\mu}$ are determined in Eq. (3.10). $(\alpha\beta\mu) \equiv \langle 1/2\alpha | 1/2\beta, 1\mu \rangle$ are Clebsh-Gordon coefficients for angular momentum summation.

B. Low-energy reduction of the extended Hubbard Hamiltonian

The Hamiltonian (3.11) is equivalent to the primary Hamiltonian (2.7), but in this form it is substantially more convenient for description of low-energy excitations. If we retain in Eq. (3.11) only the first two terms con-

taining operators $X_l^{cc}, X_l^{c\alpha}, X_l^{\alpha c}$ and if we add the J term from (2.2), we get the t-J the model with hopping to all sites. Indeed, the constants $\lambda_{ll'}$ are different from zero for all sites and decrease rapidly with increasing distance between the sites l and l':

$$\lambda_0 \equiv \lambda_{00} = 0.9581, \quad \lambda_1 \equiv \lambda_{01} = 0.1401,$$
$$\lambda_2 \equiv \lambda_{11} = -0.02351, \quad \lambda_3 \equiv \lambda_{02} = -0.01373,$$
$$\lambda_4 \equiv \lambda_{12} = 0.006851, \quad \lambda_5 \equiv \lambda_{03} = 0.003520,$$

$$\lambda_{n,m} \Longrightarrow (-1)^{m+n+1}/2\pi(n^2+m^2+nm)^{3/2} \quad \text{for } n+m \gg 1, \quad (3.12)$$

where $\lambda_{nm} \equiv \lambda_{0,l}$ for $l = ne_x + me_y$. Since the constants λ_{nm} decrease sufficiently rapidly with increasing $m+n$, we can construct the perturbation theory over the Hamiltonian H_{pd}^2 with the help of the Schrieffer-Wolff transformation

$$H_{pd} \Rightarrow \tilde{H}_{pd} = \exp(-S) H_{pd} \exp(S), \quad S^\dagger = -S. \quad (3.13)$$

In the first order of perturbation theory over H_{pd}^2 the generator of transformation and the second-order correction to the Hamiltonian are

$$[H_{pd}^0, S] = -H_{pd}^2, \quad \delta H_{pd} = (1/2)[H_{pd}^2, S]. \quad (3.14)$$

For the Hamiltonian H_{pd} in the form (3.11) the generator S can be easily found using the properties of the Hubbard algebra

$$S = \sqrt{2}t \sum_{ll'} \lambda_{ll'} \left(\frac{b^2-a^2}{E_+ - E_-} X_l^{c\alpha} X_{l'}^{\alpha b} - \frac{\sqrt{3}a}{\epsilon_p - E_-} (\alpha\beta\mu) X_l^{c\beta} X_{l'}^{\alpha\mu} - \text{H.c.} \right). \quad (3.15)$$

We retain here the contribution to the generator S essential for the correction to the lower c-singlet Hamiltonian. On the basis of this formula for the generator S, using an explicit form of the parameters $E\pm, a, b$ and the summation formulas for the Clebsh-Gordon coefficients, one can get the expression for the correction δH_{pd} to the Hamiltonians H_{pd}^0 and H_{pd}^1,

$$\delta H_{pd} = (t^2/r) \sum_{lnl'} \lambda_{ln}\lambda_{nl'} [(1-\Delta^2/r^2) X_l^{c\alpha} X_n^{\alpha\beta} X_{l'}^{\beta c} - 2\hat{N}_n X_l^{c\alpha} X_{l'}^{\alpha c}],$$
$$\hat{N}_n \equiv X_n^{\uparrow\uparrow} + X_n^{\downarrow\downarrow}. \quad (3.16)$$

The presence of the operators $X_n^{\alpha\beta}$ in Eq. (3.16) reflects the Fermi statistics of holes. Hopping from site l to site l' through site n depends on the filling and the spin state of a hole at site n.

At this final step we can add the Hamiltonian δH_{pd} to the Hamiltonians H_{pd}^0, H_{pd}^1 and obtain the Hamiltonian correctly describing the energy of the lower c singlet and its hopping to the nearest neighbors. Using the identity

$$X_n^{\alpha\beta} = (\hat{N}_n/2)\delta_{\alpha\beta} + \mathbf{s}_n \sigma_{\beta\alpha}, \quad (3.17)$$

where $\mathbf{s}_n$ is the Cu spin-$\frac{1}{2}$ operator, one can get

$$H_{pd}^{01} = E_0 \sum_l X_l^{cc} + t_1 \sum_{\langle ll' \rangle} X_l^{\alpha c} X_{l'}^{c\alpha} + E_{0N} \sum_{ln} \lambda_{ln}^2 \hat{N}_n X_l^{cc} + \sum_{\langle ll' \rangle n} \lambda_{ln}\lambda_{nl'} [t_{1N} \hat{N}_n X_l^{\alpha c} X_{l'}^{c\alpha} + t_{1S} \mathbf{s}_n X_l^{\alpha c} \sigma_{\alpha\beta} X_{l'}^{c\beta}], \quad (3.18)$$

where $E_0 = -r$, $t_1 = 4\lambda_0\lambda_1 t^2/r$, $E_{0N} = -(3+\Delta^2/r^2)(t^2/r)$, $t_{1N} = (t^2/2r)(3+\Delta^2/r^2)$, $t_{1S} = -(t^2/r)(1-\Delta^2/r^2)$. Using the representation (2.3) of the Hubbard operators in terms of the primary electron operators $c_{l\alpha}^\dagger, c_{l\alpha}$, we can rewrite the expression for the Hamiltonian (3.18) in a more usual form

$$H_{pd}^{01} = E_0 \sum_l (1 - \hat{n}_l^c + \hat{d}_l^c) + t_1 \sum_{\langle ll' \rangle} (\tilde{c}_l^\dagger \tilde{c}_{l'}) + E_{0N} \sum_{ln} \lambda_{ln}^2 (\hat{n}_n^c - 2\hat{d}_n^c)(1 - \hat{n}_l^c + \hat{d}_l^c)$$
$$+ \sum_{\langle ll' \rangle n} \lambda_{ln}\lambda_{nl'} [t_{1N} (\hat{n}_n^c - 2\hat{d}_n^c)(\tilde{c}_l^\dagger \tilde{c}_{l'}) + t_{1S} (\tilde{c}_n^\dagger \sigma \tilde{c}_n)(\tilde{c}_l^\dagger \sigma \tilde{c}_{l'})], \quad \hat{d}_l^c \equiv c_\uparrow^\dagger c_\uparrow c_\downarrow^\dagger c_\downarrow. \quad (3.19)$$

The first two terms of these Hamiltonians (3.18) and (3.19) coincide with the first two terms of the t-J Hamiltonian (2.2). The second two terms represent the second-order corrections which depend on the filling and the spin state of the neighbor sites. The relative magnitude of these additional terms is approximately 10% of the first two terms. In this case summation over index n can be limited by the nearest neighbors, next-nearest neighbors, and next-to-next-nearest neighbors of sites l and l'. A more detailed comparison of the relative contribution of different terms in the Hamiltonian (3.18) for the case of a ferromagnetic background is presented below. If we add to the Hamiltonian (3.18) the J term with the nearest-neighbor exchange,

$$J = 4t^4 (U_d - 2\Delta)^{-2} [1/U_d + 1/(U_d - 2\Delta)] \simeq 0.13 \text{ eV}$$

(see Refs. 1 and 2), we get the effective one-band Hamiltonian for our case.

C. The structure of hopping to the next neighbors

Some works[18–22] on the t-J model consider a different generalization of the usual t-J Hamiltonian. The

reason for such consideration is a dependence of the one-particle energy for the antiferromagnetic spin ordering on the details of the Hamiltonian. In some works the exchange Hamiltonian for the next neighbors (frustration) was considered.[19] Such terms were deduced from the three-band Hamiltonian in Ref. 23. The last two terms in the Hamiltonian (3.18) represent the corrections to the energy-level position and to the hopping to the nearest neighbors. But the three-band Hamiltonian also generates hopping to the next-nearest neighbors. The structure of these additional terms in the total Hamiltonian is the following:

$$H_{pd}^{23}=\sum_{i=2,3}\left(t_i\sum_{\langle ll'i\rangle}X_l^{\alpha c}X_{l'}^{c\alpha}+\sum_{\langle ll'i\rangle n}\lambda_{ln}\lambda_{nl'}(t_{iN}N_nX_l^{\alpha c}X_{l'}^{c\alpha}+t_{iS}\mathbf{s}_n X_l^{\alpha c}\sigma_{\alpha\beta}X_{l'}^{c\beta})\right), \tag{3.20}$$

where $t_i=4t^2\lambda_i\lambda_0/r$, $t_{iN}=(t^2/r)(3+\Delta^2/r^2)$, $t_{iS}=-2(t^2/r)(1-\Delta^2/r^2)$ for $i=2,3$ and $\langle ll'i\rangle$ denotes summation over the second or third neighbors. The last term formally coincides with the last term of the Hamiltonian H_{01} but summation over l and l' is performed over the second and third neighbors. The summation over n is performed over the sites nearest l and l'. The physical interpretation of the Hamiltonian is similar to that of H_{01}. Corrections of the third and the fourth order to the Hamiltonian $H_{01}+H_{23}$ are considered in the Appendix.

IV. COMPARISON WITH THE EXACT SOLUTION ON A FERROMAGNETIC BACKGROUND

A. Comparison of the second-order Hamiltonian

For the one-hole problem on a ferromagnetic background, the Hamiltonian (3.18) is substantially simplified and can be represented in the form

$$H_{01}=E_0^f\sum_l X_l^{cc}+t_1^f\sum_{\langle ll'\rangle}X_l^{\downarrow c}X_{l'}^{c\downarrow}, \tag{4.1}$$

where the parameters E_0^f, t_1^f have the form

$$\begin{aligned}E_0^f&=-r-(4t^2/r)(3+\Delta^2/r^2)(\lambda_1^2+\lambda_2^2+\lambda_3^2),\\ t_1^f&=(2\lambda_1t^2/r)[2\lambda_0+(1+\Delta^2/r^2)(2\lambda_2+\lambda_3)].\end{aligned} \tag{4.2}$$

We want to discuss two questions: (1) the relative magnitude of the second-order corrections and (2) a comparison with the exact result. We can compare these parameters E_0^f, t_1^f of the approximate Hamiltonian (3.18) with the exact parameters $E_0^{\rm ex}, t_1^{\rm ex}$ of the exact solution on the ferromagnetic background

$$t_i^{\rm ex}=-\sum_{\mathbf{k}}\epsilon_{\mathbf{k}}\cos(\mathbf{k}\cdot\mathbf{r}_i),\quad E_0^{\rm ex}=-t_0^{\rm ex}, \tag{4.3}$$

where the energy of a hole on a ferromagnetic background $\epsilon_{\mathbf{k}}$ is represented by a very simple equation (2.10), and $\mathbf{r}_i$ is the vector from the origin to the i neighbor. At the first step let us compare Eq. (2.10) and Eq. (4.1) in two limiting cases $\Delta\gg t$ and $\Delta\ll t$. In the first case $\Delta\gg t$ we have the exact parameters $E_0^{\rm ex}, t_1^{\rm ex}$,

$$E_0^{\rm ex}=-\Delta-4t^2/\Delta,\quad t_1^{\rm ex}=t^2/2\Delta \tag{4.4}$$

and for the approximate parameters E_0^f, t_1^f

$$\begin{aligned}E_0^f&=-\Delta-(4t^2/\Delta)[\lambda_0^2+4(\lambda_1^2+\lambda_2^2+\lambda_3^2)]\\ &\simeq-\Delta-3.998t^2/\Delta,\\ t_1^f&=(4\lambda_1t^2/2\Delta)(\lambda_0+2\lambda_2+\lambda_3)\simeq0.5031t^2/\Delta.\end{aligned} \tag{4.5}$$

We can see the agreement up to the third digit. The relative magnitudes of the corrections to E_0^f and t_1^f are 0.089 and 0.061, respectively. In the opposite case $t\gg\Delta$ we can compute the integrals (4.3) for $E_0^{\rm ex}, t_1^{\rm ex}$,

$$E_-^{\rm ex}=-2.8053t,\quad t_1^{\rm ex}=0.1801t, \tag{4.6}$$

and have for the approximate case

$$\begin{aligned}E_0^f&=-2\sqrt{2}\lambda_0t(1+3(\lambda_1^2+\lambda_2^2+\lambda_3^2)/\lambda_0^2)\\ &=-2.8001t,\\ t_1^f&=2\sqrt{2}\lambda_1(1+(2\lambda_2+\lambda_3)/\lambda_0)\\ &=0.19118t.\end{aligned} \tag{4.7}$$

The agreement between $E_0^{\rm ex}$ and E_0^f is also up to the third digit, but agreement between $t_1^{\rm ex}$ and t_1^f is of about 5%. The relative magnitudes of the corrections in this case to E_0^f and t_1^f are 0.066 and 0.031, respectively. In Table I we give the values of the parameters $E_0^{\rm ex}$, E_0^f, $t_1^{\rm ex}$, and t_1^f for different values of the Δ^2/t^2 ratio.

B. More detailed comparison of the fourth-order Hamiltonian

We will make a more detailed comparison of the parameters of the effective Hamiltonian on a ferromagnetic background with the exact solution in the practically important limit $\Delta\ll t$. The corrections of the third order to the one-band Hamiltonian, obtained in the Appendix, lead to the following corrections to the parameters E_0^f, t_1^f of the Hamiltonian (4.1):

TABLE I. Parameters of the effective t-J model on a ferromagnetic background for the exact solution $E_0^{\rm ex}, t_1^{\rm ex}$ and for the reduced Hamiltonian (4.1) E_0^f, t_1^f as a function of Δ^2/t^2.

Δ^2/t^2	0.01	0.1	1.0	10.0	100.0
$\lvert E_0^{\rm ex}\rvert$	2.8077	2.8233	2.9808	4.2360	10.3919
$\lvert E_0^f\rvert$	2.8758	2.8917	3.0460	4.2835	10.4117
$t_1^{\rm ex}$	0.1800	0.1789	0.1691	0.1182	0.04807
t_1^f	0.1865	0.1854	0.1751	0.1211	0.04858

$$\delta E_0^f = (\lambda_2 t/\sqrt{2}\lambda_0^2)(3\lambda_1^2 + 2\lambda_2\lambda_3) - 54t\lambda_1^4/16\sqrt{2}\lambda_0^3 \simeq -0.0021t, \quad (4.8)$$

$$\delta t_1^f = -(\lambda_1 t/4\sqrt{2}\lambda_0^2)(17\lambda_1^2 + 20\lambda_2^2 + 18\lambda_3^2) \simeq -0.00939t.$$

Summing up these expressions for δE_0^f and δt_1^f with E_0^f and t_1^f from Eq. (4.7), we get

$$E_0^f = -2.8023t, \quad t_1^f = 0.1824t. \quad (4.9)$$

Comparing with the exact values E_0^{ex}, t_1^{ex} of Eq. (4.6) we see an excellent quantitative agreement.

We also will compare the hopping Hamiltonian for the second and the third neighbors on a ferromagnetic background,

$$H_{23} = \sum_{i=2,3} t_i^f \sum_{\langle ll'i\rangle} X_l^{\downarrow c} X_{l'}^{c\downarrow} \quad (4.10)$$

with the exact solution (2.9). The exact hopping parameters t_2^{ex}, t_3^{ex} on a ferromagnetic background are equal to

$$t_2^{ex} = -0.011\,77t, \quad t_3^{ex} = -0.006\,03t. \quad (4.11)$$

We represent the expression for the approximate values of the parameters t_2^f, t_3^f in the form

$$t_i^f = t_i^{(1)} + t_i^{(2)} + t_i^{(3)} + t_i^{(4)}, \quad (4.12)$$

where $t_i^{(j)}$ for j=1,2,3,4 correspond to the contribution of the jth order of the perturbation theory. The explicit expressions for the parameters $t_2^{(j)}/t$, as shown in the Appendix, are the following:

$$t_2^{(1)}/t = \sqrt{2}\lambda_2 = -0.0332,$$
$$t_2^{(2)}/t = (1/\sqrt{2}\lambda_0)(\lambda_1^2 + 2\lambda_2\lambda_3 + 2\lambda_1\lambda_4) = 0.0164,$$
$$t_2^{(3)}/t = -(1/4\sqrt{2}\lambda_0^2)(26\lambda_1^2\lambda_2 + 8\lambda_1^2\lambda_3 + 24\lambda_2\lambda_3^2 + 17\lambda_2^3) = 0.0028,$$
$$t_2^{(4)}/t = 13\lambda_1^4/4\sqrt{2}\lambda_0^3 = 0.0010. \quad (4.13)$$

and in a more compact form for $t_3^{(j)}/t$

$$t_3^{(j)}/t = (-0.0194, 0.0095, 0.0021, 0.0006). \quad (4.14)$$

As a result we have

$$t_2^f = -0.013t, \quad t_3^f = -0.0072t. \quad (4.15)$$

In this case we can see that the agreement between the exact and approximate values is of the order of 10%. We have a substantial compensation of the direct hopping constants $t_2^{(1)}, t_3^{(1)}$ up to the final values t_2^f, t_3^f on a ferromagnetic background due to the higher-order correction. This means that the corrections to hopping on the next neighbors have a very complicated nature and include hopping processes depending on the Cu spin states at the neighbor sites.

V. CONCLUSION

It has been shown that in the case of the three-band model for $\epsilon \gg U_d - \epsilon$ the Bloch waves constructed from the local Cu-O singlets are the ground states for one-hole excitations. The Zhang-Rice Cu-O singlets form the basis for reduction of the three-band model to the single-band generalized t-J model. The method of the reduction developed in this work is rather specific and is based on the representations of the Hubbard operators in terms of the Fermi and spin-$\frac{1}{2}$ operators. This reflects the history of work on the paper.

In fact, the method of obtaining a single-band Hamiltonian is sufficiently general: at the first step the cluster problem is solved and local electron (hole) energy levels and wave functions are found with correlations being taken into account. At the second step the initial Hamiltonian can be expressed in terms of the Hubbard operators that transfer these states in each other, including the ground state. This Hamiltonian includes hopping terms that describe a hole transition from one lattice site to another, including mixing of the local energy positions. If such mixing is small, at the third step with the help of the Schrieffer-Wolff transformation one can get the single-band Hamiltonian for description of the low-energy excitations.

In the framework of such an approach one can consider the general case of the three-band model parameters when $\epsilon, U_d - \epsilon$ and $4\sqrt{2}t$ are of the same order of magnitude. One can also take the direct oxygen-oxygen hopping into consideration.

We want to stress that the singlet structure of hole excitations based on the Wannier functions provides a very low energy of hole excitations. In the case considered by Lovtzov and Yushankhai[9] and in the case discussed in this work, the situation is similar: the position of the bottom of the hole singlet band, measured from the middle of the spacing between the oxygen and copper local levels, is equal to (3.18)

$$E_b = -r - \xi t_1 \simeq -2\sqrt{2}(1 + 0.067\xi)t \simeq -3.1t, \quad (5.1)$$

where ξ is the parameter of the order of a unity which describes the dependence of the band-bottom position on the type of magnetic ordering of the copper spins. For the antiferromagnetic ordering $\xi = 3.13 - 2.83(J/t)^{0.73}$ (Ref. 24) and for our case, J=0.13 eV, $t_1 = 0.27$ eV $\rightarrow$ $\xi = 1.47$, and $E_b = -3.1t$. It is necessary to stress that this value of the bottom of the band position is very low. Attempts to develop the physics of the three-band model in terms of the slave-boson approach[13–16] give $E_b = -2\sqrt{2}r_0 t$ with $r_0 < 0.6$, which yields $E_b = -1.7t$ for the bottom of band for some variants of the spin-liquid state. This state is positioned sufficiently high. It is unlikely that the gain in the exchange energy of the spin-liquid state due to the presence of holes makes the energy of such a type of state lower than the energy of the singlet band.

Actually the gain in the exchange energy has a scale J, while the band position has a scale t, but $t/J \simeq 10$. Of

course we cannot prove a theorem that the hole singlet band has the lowest possible energy for the three-band model in the actual region of parameters. However, in our case such a theorem has been proved for a ferromagnetic background, and we believe that the consideration of the general magnetic state does not change the situation.

In our case ($U_d - \epsilon \ll \epsilon$) the fundamental parameter of the t-J model $J/t_1 \simeq J/0.19t \simeq 0.45$. This estimation of the ratio J/t_1 correlates fairly well with another estimation of this ratio.[11,25,26]

Hopping to the next neighbors has a complicated nature, depending on spin states of the neighbor copper ions and cannot be expressed by a simple t' term with hopping to the next-nearest neighbors. The order of magnitude of these terms in the Hamiltonian is (0.02–0.03)t, which is 10% of t_1.

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APPENDIX

We derive here the third- and fourth-order corrections to the effective Hamiltonian (3.11), and the forms of the corresponding energy additions. We restrict ourselves to the case $\Delta = 0$ and a ferromagnetic background. The full Hamiltonian in terms of the Hubbard operators (3.11) can be expressed in more convenient terms. Let us introduce

$$\begin{aligned} D &= \sum_{ll'} \lambda_{ll'} (X_l^{c\alpha} X_{l'}^{\alpha c} - X_l^{b\alpha} X_{l'}^{\alpha b}), \\ F &= \sum_{ll'} \lambda_{ll'} (\alpha\beta\mu)(X_l^{c\beta} X_{l'}^{\alpha\mu} - X_l^{\mu\alpha} X_{l'}^{\beta b}), \\ G &= \sum_{ll'} \lambda_{ll'} (\alpha\beta\mu)(X_l^{\mu\alpha} X_{l'}^{\beta c} - X_l^{b\beta} X_{l'}^{\alpha\mu}). \end{aligned} \tag{A1}$$

One can check that

$$[H_0, D] = 0, \quad [H_0, F] = -\eta t F, \quad [H_0, G] = \eta t G, \tag{A2}$$

where $\eta = 2\sqrt{2}\lambda_0$. Hence, G can be called "raising" and F "lowering" operators, because G transfers the low-singlet state to the triplet one, the triplet to the high-singlet, while F acts in the opposite way.

In these (A1) terms the Hamiltonian (3.11) has the form

$$H_1 = -\sqrt{2}tD, \quad H_2 = -\sqrt{3}t(F+G). \tag{A3}$$

The first-order generator of the Schrieffer-Wolff transformation and the second-order term of the effective Hamiltonian are given by

$$\begin{aligned} S_1 &= -(\sqrt{3}/\eta)(F-G), \\ \delta H^{(2)} &= -(3t/\eta)(FG - GF). \end{aligned} \tag{A4}$$

By projecting out highly excited states, the second-order term in $\delta H^{(2)}$ can be obtained. Equations for the second- and third-order generators of the Schrieffer-Wolff transformation and for the third- and fourth-order corrections to the interaction are[27]

$$\begin{aligned} &[H_0, S_2] = -[H_1, S_1], \\ &[H_0, S_3] = -[H_1, S_2] - (1/3)[[H_2, S_1], S_1], \end{aligned} \tag{A5}$$

$$\begin{aligned} \delta H^{(3)} &= (1/2)[H_2, S_2], \\ \delta H^{(4)} &= (1/2)[H_2, S_3] - (1/24)[[[H_2, S_1], S_1], S_1]. \end{aligned}$$

In terms of D, F, and G we get

$$S_2 = -(\sqrt{6}/\eta^2)[D(F+G) - (F+G)D], \tag{A6}$$

$$\begin{aligned} S_3 = -(2\sqrt{3}/\eta^3)[\ &(G-F)D^2 - 2D(G-F)D \\ &+D^2(G-F) - 2GFG + 2FGF \\ &-GFF + FGG + GGF - FFG], \end{aligned}$$

so that

$$\begin{aligned} \delta H^{(3)} = (3t/\sqrt{2}\eta^2)[\ &D(F+G)^2 + (F+G)^2 D \\ &-2(F+G)D(F+G)]. \end{aligned} \tag{A7}$$

Since we are interested in the low-energy states, all terms with F to the right and G to the left can be omitted. Also the third term in (A7) can be removed because the triplet state does not hop. Thus we get the effective

$$\delta H^{(3)} = (3t/\sqrt{2}\eta^2)[DFG + FGD]. \tag{A8}$$

Corresponding corrections to the effective hopping and the energy on a ferromagnetic background have the form

$$\begin{aligned} \delta t^{(3)f} = (\sqrt{2}t/\eta^2)\Bigg(&\sum_{l,l'} \lambda_{il}\ \lambda_{ll'}\ \lambda_{lj} \\ &+2\lambda_{ij} \sum_{l} (\lambda_{il}^2 + \lambda_{lj}^2) + 3\lambda_{ij}^3 \Bigg) \end{aligned} \tag{A9}$$

$$\delta E_0^{(3)f} = (\sqrt{2}t/\eta^2) \sum_{l,l'} \lambda_{il}\ \lambda_{ll'}\ \lambda_{l'i}.$$

By substitution of (A6) in (A5) and keeping low-energy terms, we have

$$\begin{aligned} \delta H^{(4)} = (3t/2\eta^3)[\ &6FGFG - 3F^2G^2 \\ &-FGD^2 - D^2FG]. \end{aligned} \tag{A10}$$

The fourth-order corrections to the hopping parameter and energy will be

$$\delta t^{(4)f} = -(t/2\eta^3)\Bigg(3\sum_{l,n,m} \lambda_{il}\,\lambda_{ln}\,\lambda_{nm}\,\lambda_{mj} + 5\sum_{l,n} \lambda_{il}\,\lambda_{lj}(\lambda_{in}^2+\lambda_{jn}^2) + 3\sum_{l,n} \lambda_{il}\,\lambda_{lj}\,\lambda_{ln}^2 + 5\sum_{l}(\lambda_{il}^3\,\lambda_{lj}+\lambda_{il}\,\lambda_{jl}^3)$$
$$+3\lambda_{ij}\sum_{l,n}(\lambda_{il}\,\lambda_{ln}\,\lambda_{ni}+\lambda_{jl}\,\lambda_{ln}\,\lambda_{jn}) + 13\lambda_{ij}^2\sum_{l}\lambda_{il}\,\lambda_{lj}\Bigg),$$

$$\delta E_0^{(4)f} = -(3t/2\eta^3)\Bigg(\sum_{l,n,m} \lambda_{il}\,\lambda_{ln}\,\lambda_{nm}\,\lambda_{mi} + \sum_{l,n}(\lambda_{il}^2\,\lambda_{ln}^2+\lambda_{il}^2\,\lambda_{in}^2) + \sum_{l}\lambda_{il}^4\Bigg). \tag{A11}$$

For hopping to the first, second, and third neighbors corrections may be easily calculated.

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