Range of the t-J model parameters for CuO_2 planes: Experimental data constraints

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The t-J model effective hopping integral is determined from the three-band Hubbard model for the charge carriers in CuO_2 planes. For this purpose the values of the superexchange constant J and the charge-transfer gap E_{gap} are calculated in the framework of the three-band model. The energies of the local states for added electron and hole are obtained from the systematic analytical reduction of the three-band model and their kinetic energy is found from the accurate variational approach using the t-J model. Fitting values of J and E_{gap} to the experimental data allows us to narrow the uncertainty region of the three-band model parameters. As a result, the t/J ratio of the t-J model is fixed in the range 2.4–2.7 for holes and 2.5–3.0 for electrons. This approach explains the formation of the Frenkel exciton and correctly describes the main features of the charge-transfer spectrum.

I. INTRODUCTION

A large amount of work dedicated to high-T_c superconductors agrees that an appropriate electronic model which contains all essential orbitals is the three-band Hubbard model.[1–3] Some other works developing a multiband approach[4–7] also support this belief.

It is also widely accepted that the low-energy physics of insulating compounds can be described by the Heisenberg Hamiltonian. According to the earlier work by Zhang and Rice[8] and later studies[9–15] lightly doped systems are described by the simple t-J model. Different techniques provide different exactness of the three-band model to t-J model mapping and generate a wide range of generalizations.[11,14,15]

In our previous works[12,13,15] a consistent low-energy reduction of the three-band model to the generalized t-J model in a realistic range of parameters has been performed. It has been shown[15] that the second-order corrections to the local energy of the carrier and its hopping integral are small ($< 5\%$). The role of the next-nearest-neighbor terms has also been discussed.

It is commonly believed that the 90% or even higher accuracy of the t-J model as the low-energy electronic model for high-T_c superconductors justifies its wider study[16–18] and this model remains the main pretender in describing the superconductivity in cuprates.[19–21] Recent angle-resolved photoemission experiments[22] can be interpreted as direct support of some of the t-J model properties.[23] Both the anomalous behavior of these systems in normal state and the superconductivity itself seem to be described in the framework of the t-J model.[19–21,23]

The reasonable question from this point of view is what is the role of either the three-band or more complex first-principles models? There can be several answers: (i) calculating the t-J model parameters for real systems; (ii) giving insight into the experiments involving not only the simple t-J model degrees of freedom.

In this paper we mainly address the question of the t-J model parameters. The superexchange constant J for the t-J model is directly measurable.[24,25] Hence the parameter to be determined is the effective nearest-neighbor hopping integral. The problem of its calculation is not connected with the accuracy of low-energy mapping from the three-band model, which is always very high, but results from the uncertainty in the three-band model parameters. The three-band model in the conventional formulation contains as inner parameters two on-site and one intersite Coulomb repulsions, two hopping integrals, and splitting of the levels[1] which are not directly measurable. Some of them are rather badly determined. This makes the calculation of the hopping integral for real systems questionable and even controversial.

We develop an obvious idea of fixing the three-band model parameters by using experimental data. This idea has already been exploited in the cluster calculations for spectroscopic data[6] and for the superexchange constant J in our previous work.[15] A cluster-model analysis of the material dependence of the charge-transfer gap and superexchange interaction using the same model has been made in Ref. 26. This work[26] has shed some light on the charge-transfer nature of the high-temperature superconductors and some of their general properties. Now, on the basis of a better understanding of the low-energy model of the electronic system and the magnetic polaron nature of the t-J model carriers,[18,27,28] we can calculate quite accurately the charge-transfer gap. Self-consistent calculation provides a narrow range of possible values for t/J. Also, an excitonic feature of the charge-transfer spectrum is obtained in agreement with recent experiments.[29]

The paper is organized as follows. In Sec. II we dis-

cuss the low-energy limit of the three-band model and experimentally observable quantities. In Sec. III the calculation of the superexchange constant J and the charge-transfer gap $E_{\rm gap}$ is produced. In Sec. IV we discuss the dependence of J and $E_{\rm gap}$ on the parameters of the three-band model and determine the range of the hopping parameter t for electrons and holes. In Sec. V the properties of the excitonic state are considered. Section VI presents our results and discussions. The technical aspects of the work are given in Appendix.

II. THE LOW-ENERGY LIMIT OF THE THREE-BAND MODEL AND OBSERVABLE QUANTITIES

Previously, it was suggested that the three-band Hubbard model is an appropriate starting point for describing the electronic structure of CuO_2 planes.[1,2] The Cu $d_{x^2-y^2}$ orbital and $p\sigma(x,y)$ orbitals are strongly hybridized. These orbitals are explicitly treated in the three-band model with the justifiable assumption that other orbitals do not directly participate in the low-energy dynamics. The full Hamiltonian of the model is defined by[1]

$$
\begin{aligned}
&H = H_0 + H_t + \Delta H \ , \\
&H_0 = \epsilon_d \sum_{l,\alpha} n^d_{l\alpha} + \epsilon_p \sum_{m,\alpha} n^p_{m\alpha} + U_d \sum_l n^d_{l\uparrow} n^d_{l\downarrow} , \\
&H_t = t_{pd} \sum_{\langle lm\rangle,\alpha} (d^\dagger_{l\alpha} p_{m\alpha} + \text{H.c.}) \\
&\qquad - t_{pp} \sum_{\langle mm'\rangle,\alpha} (p^\dagger_{m\alpha} p_{m'\alpha} + \text{H.c.}) \ , \qquad (1) \\
&\Delta H = U_p \sum_m n^p_{m\uparrow} n^p_{m\downarrow} + V_{pd} \sum_{\langle lm\rangle,\alpha\beta} n^d_{l\alpha} n^p_{m\beta} , \qquad (2)
\end{aligned}
$$

in the standard notation of holes at O(p) and Cu(d) sites. The sign convention for oxygen orbitals in H_t is accepted.[15,30] Our approach to the description of low-energy properties of the above model[15] is based on taking into account the main Coulomb (U_d) interaction exactly and the others as perturbations.

In order to justify this method we briefly describe here the magnitudes of the three-band model's parameters. Different experimental,[6,7,31] atomic,[32] and band calculations[4,33,34] show that U_d =5–7 eV,[32] 7–11 eV,[4,6] U_p =3–8 eV,[7] and V_{pd} =0–1.7 eV. U_p is always less than U_d. There is a general agreement for the Cu-O system that $\Delta = \epsilon_p - \epsilon_d$ is always > 0 and $< U_d$.[31] It reflects the facts that the first hole in the unit cell is predominantly at the Cu site and the added hole has oxygen character. t_{pd} =1–1.6 eV (and it is unlikely that it is less than 1 eV) and t_{pp} =0.5–0.7 eV.[4,33,34] This set of magnitudes will be called hereafter the *realistic region* of parameters.

The consistent low-energy reduction of the three-band model to the generalized t-J model has been performed in previous works.[12,13,15] Our method of low-energy reduction has been based on construction of a set of local states with different numbers of holes over the filled atomic orbitals. The most essential states are the following.

(1) The vacuum state or the vacancy which is simply

$$|v\rangle = |0\rangle \ . \qquad (3)$$

(2) The one-hole states which represent the ground state of the CuO_2 plane,

$$|f\alpha\rangle \equiv |\alpha\rangle = U|d\alpha\rangle - V|q\alpha\rangle \ , \qquad (4)$$

where $|d\alpha\rangle$ and $|q\alpha\rangle$ are the copper and symmetrical oxygen hole states with spin projection α, respectively.

(3) The two-hole states which are the Zhang-Rice singlets:

$$
\begin{aligned}
|c\rangle = U_1|d\uparrow d\downarrow\rangle + V_1|q\uparrow q\downarrow\rangle \\
+ W_1(|d\uparrow q\downarrow\rangle - |d\downarrow q\uparrow\rangle)/\sqrt{2}. \qquad (5)
\end{aligned}
$$

The coefficients U, V, U_1, V_1, W_1 are functions of the parameters of the three-band model.[15] At half filling Hamiltonian (1),(2) is reduced to the Heisenberg Hamiltonian with spins 1/2 which are antiferromagnetically ordered due to the second-order virtual transitions through the set of two-hole states. Note that the above named spins 1/2 are exactly states $|f\alpha\rangle$ [Eq. (4)].

It has been shown[13,15] that for the case of near to half filling the Hamiltonian of the three-band model is reduced to the Hamiltonian of the t-J like model of singlets, vacancies, and spins:

$$
\begin{aligned}
H_{t\text{-}J} = (E_v - \mu) \sum_l X^{vv}_l + (E_c + \mu) \sum_l X^{cc}_l \\
+ t_e \sum_{\langle ll'\rangle,\alpha} X^{v\alpha}_{l'} X^{\alpha v}_l + t_h \sum_{\langle ll'\rangle,\alpha} X^{c\alpha}_{l'} X^{\alpha c}_l \\
+ J \sum_{\langle ll'\rangle} \mathbf{S}_l \mathbf{S}_{l'} \ , \qquad (6)
\end{aligned}
$$

where $X^{ab}_l \equiv |al\rangle\langle lb|$ are the Hubbard operators at the site l, and $\mathbf{S}_l = \sigma_{\alpha\beta} X^{\alpha\beta}_l/2$. The constants E_v and E_c are the local energies of the vacancy and singlet; μ is the chemical potential; t_e and t_h are the hopping integrals for the vacancy and singlet (electron and hole), respectively; J is the exchange constant. All five parameters E_v, E_c, t_e, t_h, and J are functions of the three-band model parameters. J is defined in the next section, other parameters are defined in the Appendix. It has been shown that the relative magnitudes of the omitted terms in the Hamiltonian $H_{t\text{-}J}$ [Eq. (6)] are of the order of 10%.[13,15]

Since the Hamiltonian (6) describes many important properties of the cuprates the real values of its parameters are of great interest. As was noted above, the parameters of the primary model (1),(2) are known with low precision. In this situation calculation of the observable quantities is an urgent issue since it provides a way to fix the parameters of Hamiltonians (1), (2), and (6). The most reliable experimental values which we can describe quite accurately are the superexchange constant J and the charge-transfer gap $E_{\rm gap}$. The experimental values of J are 0.14 eV and 0.17 eV for the lanthruman and yttrium systems, respectively.[24,25] These values of

J follow from measurements of the velocity of sound for magnons. The value of the charge-transfer gap is known from a variety of optical measurements[29,31,34,35] and is close to 2.0 eV. Observation of photoconductivity at the same energies shows that the excitations result in separated electrons and holes.[29,35] We have taken most of the clear experimental features of the charge-transfer spectrum from Ref. 29 where photoconductivity as well as reflectivity data for La_2CuO_4 are presented.

III. CALCULATION OF THE OBSERVABLE QUANTITIES

The expression for the antiferromagnetic (AF) coupling constant J in the framework of our approach is[15]

$$J = -2h_1 V^4 U_p + \sum_n x_n \frac{|D_n|^2}{\Delta E_n}. \tag{7}$$

The first term in Eq. (7) represents the exchange energy between two holes (spins) due to the repulsion at an oxygen atom. This contribution has the ferromagnetic sign and arises as an exchange interaction between the hole states (4) due to their nonlocal nature. The constant V is defined in Eq. (4); h_1 is in the Appendix. The second term in Eq. (7) represents the correction to the energy due to the virtual transition of the hole from the state Eq. (4) into the two-hole states and back.[15] Here n enumerates the two-hole states; matrix elements of transitions D_n were calculated in Ref. 15; ΔE_n are differences in energies between the vacancy and two-hole states at neighbor sites and the ground state energy (see Fig. 1); the coefficients $x_n = 4$ for the singlet and $x_n = -2$ for triplet two-hole states. It has been shown in Ref. 15 that the parameter of our low-energy reduction scheme ($|D_n|/\Delta E_n$) is of the order of 0.1. Since the next term of $\mathbf{S}_l\mathbf{S}_{l'}$ type arises only in the fourth order, the *direct* correction to the superexchange constant J (7) is very small [$\sim J/(2E^0_{\rm gap}) < 2\%$]. The term $K_N \mathbf{S}_l \mathbf{S}_{l'} N_{l''}$ arises in the third order of perturbation theory. It is omitted in the effective Hamiltonian (6) for the following reasons. Naively, it could be supposed that its magnitude $K_N/J \sim 10\%$, but the main part of this correction is almost exactly canceled due to compensation of the direct and exchange processes. To be more specific, the amplitudes of the processes which differ by permutation of two fermions in the final state are almost equal. In this situation only transitions including highly excited states (triplet, etc.) contribute. Thus, the relative magnitude of the above term is small, $K_N/J \leq 3\%$, and Eq. (7) is correct.

The most general expression for the charge-transfer energy is

$$E_{\rm gap} = E^{N-1}_{\rm min} - E^N_g + E^{N+1}_{\rm min} - E^N_g, \tag{8}$$

where N refers to the total number of electrons, E^N_g is the ground state energy, and $E^{N\pm1}_{\rm min}$ is the minimal energy of a system with one removed or added electron. For our system Eq. (8) can be specified as

$$E_{\rm gap} = E^0_{\rm gap} + \Delta E_e + \Delta E_h, \tag{9}$$

where $E^0_{\rm gap}$ is the difference in energies between a singlet and vacancy at local states separated by a large distance and the ground state (see Fig. 2); ΔE_e and ΔE_h are the depths of the bands for electron and hole (vacancy and singlet), i.e., the kinetic energy gain. $E^0_{\rm gap}$ can be calculated in the framework of the three-band model, whereas for the calculation of ΔE_e and ΔE_h we will use the t-J model.

The expression for $E^0_{\rm gap}$ in terms of Eq. (6) is very simple:

$$E^0_{\rm gap} = E_c + E_v. \tag{10}$$

The values of ΔE_e and ΔE_h can be determined from numerous analytical and numerical calculations[16–19,23,27,28,36,37] of the dispersion relation $\epsilon(\mathbf{k})$ for one hole in the t-J model on an antiferromagnetic background. There is a general agreement that the hole (or vacancy) on the antiferromagnetic background creates a magnetic polaron of a small radius,[17,27] or, in

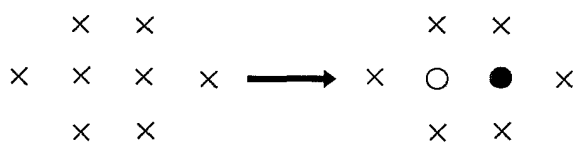


FIG. 1. The nearest-neighbor two-hole state and vacancy. Black circle denotes two-hole state; empty circle denotes vacancy. Crosses are one-hole states (spins).

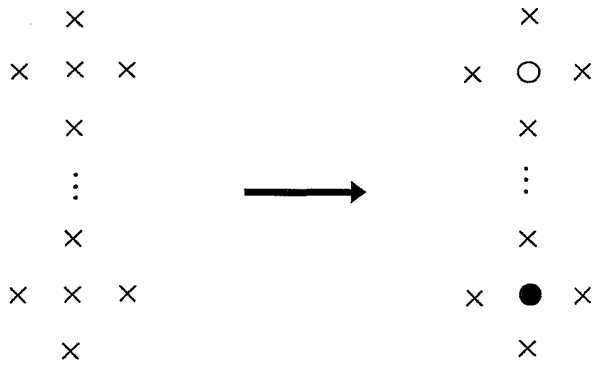


FIG. 2. The separated Zhang-Rice (ZR) singlet (hole) and vacancy (electron). Black circle denotes singlet; empty circle denotes vacancy. Crosses are one-hole states (spins).

other words, the carriers are strongly dressed by the spin waves.[23] The influence of antiferromagnetism and strong correlations is manifested in a special form of dispersion relation $\epsilon(\mathbf{k})$. For our calculations we use the results from the earlier work by Sushkov[18] where the hole wave function and $\epsilon(\mathbf{k})$ were obtained variationally:

$$\epsilon(\mathbf{k}) = 1.32J + \tfrac{1}{2}\left\{\tilde{\Delta}J - \sqrt{\tilde{\Delta}^2J^2 + 16t^2[(1+y) - (x+y)\gamma_{\mathbf{k}}^2]}\right\}$$

and

$$\Delta E = 1.32J + \tfrac{1}{2}\left\{\tilde{\Delta}J - \sqrt{\tilde{\Delta}^2J^2 + 16t^2(1+y)}\right\}, \quad (11)$$

where for Néel background $\tilde{\Delta} = 1.33, x = 0.56, y = 0.14$. Loss of energy due to the broken AF bonds (four per carrier) is included. The result for the bottom of the band at $t/J = 2.5$ from Eq. (11), $\Delta E = -1.2t$, coincides almost exactly with the recent results of a Green function Monte Carlo calculation by Dagotto, Nazarenko, and Boninsegni,[23] $\Delta E = -1.255t$. Formula (11) is quite good up to $t/J \approx 5$.[18] As was noted in the number of works (see for example Ref. 23), direct next-nearest-neighbor hopping which is omitted in the t-J model (6) almost does not change the depth of the band (11). It only shifts the quasiparticle minima from points $(\pm\pi/2, \pm\pi/2)$ to $(0, \pm\pi)$ and $(\pm\pi, 0)$.

Let us discuss now the characteristic values of all essential parameters that determine the observable quantities J and E_{gap} [(7) and (9)]. In the *realistic region* of parameters of the three-band model one can easily obtain results for J which are close to the experimental values $J = 0.14$ eV and $J = 0.17$ eV for lanthanum and yttrium systems, respectively. The value for E_{gap}^0 (10) was obtained in Ref. 15 and is equal to 3.2 eV in the same region of parameters. The characteristic values of ΔE_e and ΔE_h (11) depend on the t/J ratios for electrons and holes. These ratios weakly differ and for a typical value of $t/J = 2.5$ we have $\Delta E_e \approx \Delta E_h \approx 0.42$ eV. Thus the overall gain in energy due to magnetic polaron formation is of the order of 1 eV, which is comparable with the experimentally observed $E_{\mathrm{gap}} \approx 2.0 \pm 0.1$ eV. Therefore, the magnetopolaron effect gives an essential contribution to the value of the charge-transfer gap.

In Ref. 29 it was proposed that the usual phonon polaron effect contributes to the observable values of the charge-transfer spectrum. The corresponding gain in energy was estimated as 0.5 eV. In our opinion the usual polaron effect does not contribute in the optical transition due to the Frank-Condon principle. The magnetopolaron effect has no such restriction since it involves electronic degrees of freedom only. We will return to this question in the Discussion.

IV. PARAMETER SENSITIVITY

Thus, we have found the superexchange constant J (7) and charge-transfer gap E_{gap} (9) (Refs. 7, 25, 38) as functions of the three-band model parameters:

$$J = J(t_{pd}, t_{pp}, \Delta, U_d, U_p, V_{pd}),$$
$$E_{\mathrm{gap}} = E_{\mathrm{gap}}(t_{pd}, t_{pp}, \Delta, U_d, U_p, V_{pd}). \quad (12)$$

Both observable quantities strongly depend on hopping integrals and $\Delta = \epsilon_p - \epsilon_d$, which provides the way of fixing these latter by the experimental values of the first.

As was discussed earlier, the abundance of the parameters makes questionable the calculation of the effective hopping integral for the t-J model from the three-band model for real CuO_2 planes. While Coulomb repulsions are known with fair precision (30%–50%), the situation is complicated due to the very low precision of the direct determination of t_{pd}, t_{pp}, and Δ, which affect all effective parameters more than others. Previously, the above parameters have been determined from the analysis of spectroscopic data.[6,7] In our recent work we fitted Δ to the experimental value of J.[15]

Now, on the basis of a better understanding of the

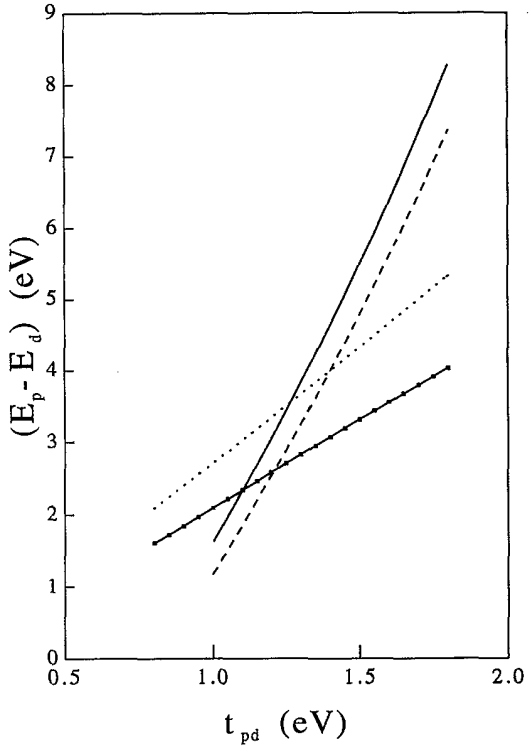


FIG. 3. Δ vs t_{pd} at constant J or E_{gap}. $U_d = 7$ eV, $U_p = 3$ eV, $V_{pd} = 1$ eV, $t_{pp}/t_{pd} = 0.5$. Full curve, $J = 140$ meV, dashed curve, $J = 170$ meV, full curve with markers, $E_{\mathrm{gap}} = 2.0$ eV; dotted curve, $E_{\mathrm{gap}} = 2.05$ eV.

charge-transfer process and more accurate calculations, fixing the worse known parameters using experimental values of J and $E_{\rm gap}$ suggests itself. We will show that this procedure keeps the effective t_h inside a narrow enough region.

First, for further discussion we define O-O hopping as $t_{pp} = \gamma t_{pd}$. In order to characterize the above mentioned strong dependence of the parameters (12) one can calculate $\Delta(t_{pd})$ at fixed J or at fixed $E_{\rm gap}$, with other parameters (Coulomb repulsions and γ) as constants in the *realistic region*. We evaluate Δ vs t_{pd} at $J = 140$ meV and 170 meV; $E_{\rm gap} = 2.0$ eV and 2.5 eV (see Fig. 3). Note that the profiles of the curves resemble those in the diagram of $U(t)$ for the simple one-band Hubbard model where $E_{\rm gap} = U + 2W$, $W = -\alpha t$, $J = 4t^2/U$, and the crossing point uniquely determines U and t.

To be more specific, we first determined Δ for constant J at an arbitrary t_{pd} and further move up or down along the curve $J = \text{const}$ to fix the value of $E_{\rm gap}$. We used the data for La_2CuO_4: $J = 140$ meV Ref. 24 and $E_{\rm gap} = 2.1$ eV (photoconductivity).[29] Figures 4–6 show the parameter of our prime interest: the effective integral for the hole in the t-J model. The parameter $\gamma = t_{pp}/t_{pd}$ is 0.5, 0.7, and 0.3 for Figs. 4, 5, and 6, respectively. In all figures the simple dotted curve corresponds to $V_{pd} = U_p = 0$; dotted curves with crosses, $V_{pd} = 0$, $U_p = 3$ eV, 6 eV; dotted curves with triangles, $V_{pd} = 0.5$ eV, 1 eV, U_p=0; and full curves correspond

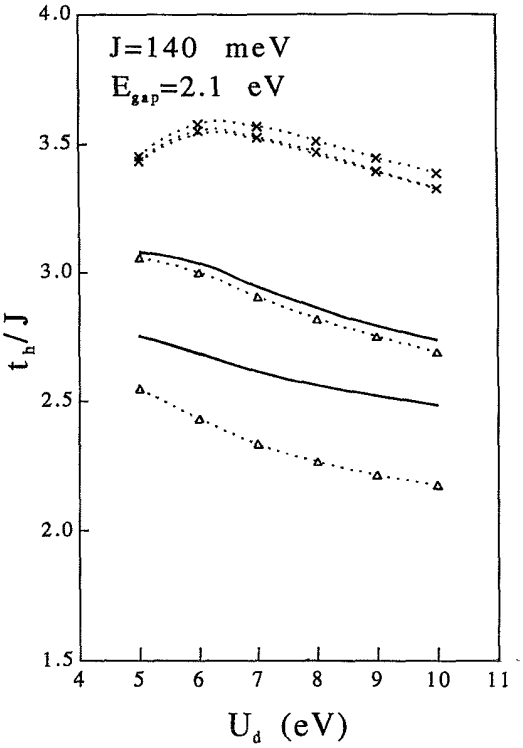


FIG. 5. All notations as for Fig. 4; $\gamma = 0.7$.

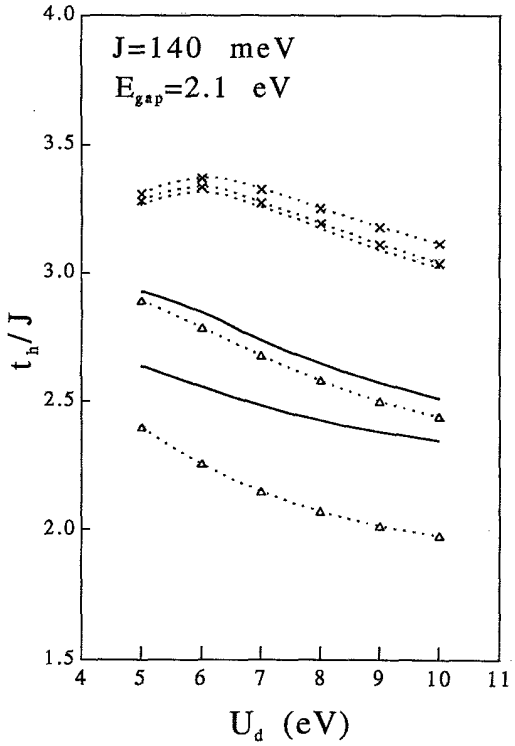


FIG. 4. Effective hopping integral for t-J model hole vs U_d. Dotted line, $V_{pd} = U_p = 0$; dotted curve with crosses, $V_{pd} = 0$, $U_p = 3$, 6 eV; dotted curve with triangles, $U_p = 0, V_{pd} = 0.5$, 1 eV; full curves, $V_{pd} = 0.5$ eV, $U_p = 3$ eV (upper), $V_{pd} = 1$ eV, $U_p = 6$ eV (lower), $\gamma = 0.5$.

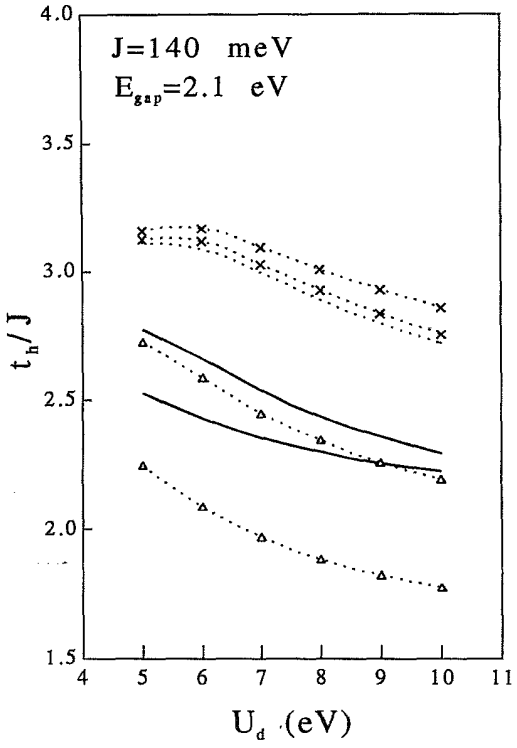


FIG. 6. All notations as for Fig. 4; $\gamma = 0.3$.

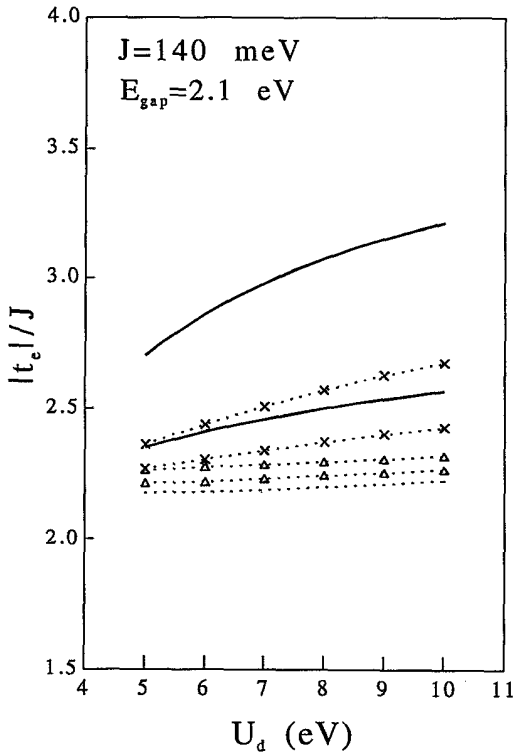


FIG. 7. Effective hopping integral for electron in the t-J model vs U_d; curve markers as for Fig. 4; $\gamma = 0.5$.

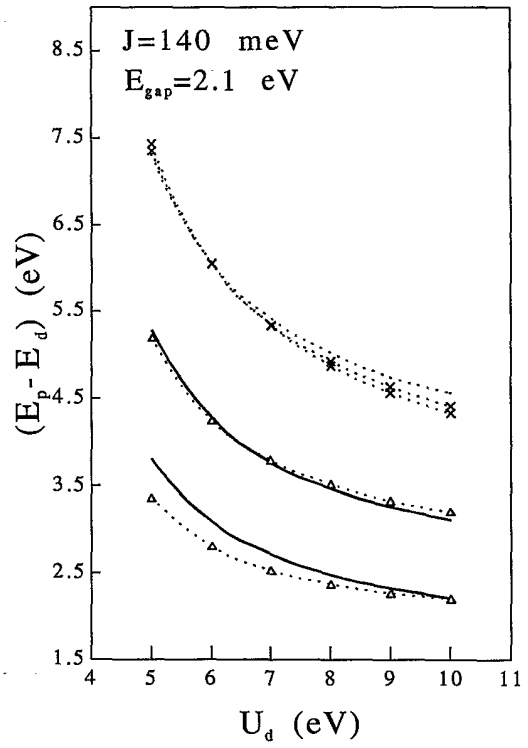


FIG. 9. Δ vs U_d; $\gamma = 0.5$.

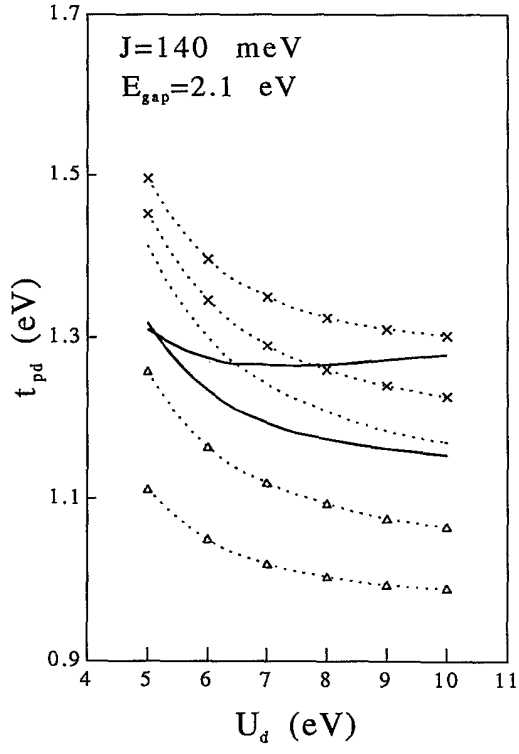


FIG. 8. Cu-O hopping integral vs U_d, curve markers as for Fig. 4; $\gamma = 0.5$.

to including both Coulomb interactions $V_{pd} = 0.5$ eV, $U_p = 3$ eV (upper), $V_{pd} = 1$ eV, $U_p = 6$ eV (lower). The maximum in the first three curves is due to transition from $\Delta > U_d$ (unrealistic range) to $\Delta < U_d$. All variations of $t_h(U_d, V_{pd}, U_p, \gamma)$ actually show only weak dependence, and in the most preferential region, when all Coulomb interactions are included, t_h lies between 2.4 J and 2.7 J. We believe that our consideration is quite accurate and well justified. Hence one can hope that the interval for t/J obtained above provides the basis for *quantitatively* correct calculations in the framework of the t-J model; for example, for the recently proposed mechanism of superconductivity in the t-J model which provides a very (exponentially) t/J-sensitive gap value.[20]

Some other features can also be explained. Figures 7–9 represent effective hopping $|t_e|$ for vacancy, t_{pd}, and Δ, respectively, vs U_d. Here always $\gamma = 0.5$. Strong support of our fitting procedure is the fact that self-consistently determined t_{pd} and Δ (Figs. 8 and 9) lie in the most appropriate region. From our calculation t_{pd} =1.2–1.4 eV and Δ =2.5–4.5 eV, which are really close to cluster calculations of Eskes and Sawatzky[7] and to the results of other groups.[4,33,34]

V. EXCITONIC STATE

The problem of consistently taking into account the Coulomb interaction of the carriers in the framework of the t-J-like model remains open. Several recent works are devoted to this problem.[39]

As was suggested earlier,[1] the short-range part of the Coulomb interaction may be kept by inclusion of the nearest-neighbor Cu-O repulsion. Since V_{pd} has been included in our effective model, one can expect that at least some of the effects will be caught.

In recent work[29] a kink in the optical reflectivity somewhere below the charge-transfer peak (at 1.75 eV) was observed. Since it had no associated photoconductivity, it was related to the creation of an exciton. An essential role of the short-range Coulomb interaction was also discussed in Ref. 29. In our way of reasoning the exciton state, if it does exist, is the Frenkel exciton state, because of the short-range nature of the interactions.

Recently it has become evident that an effective attraction can result from a pure magnetopolaron effect in the t-J model.[18,37] As was shown in Refs. 36 and 37, "contact" interaction of two holes (without charge) dressed by spin fluctuations is attractive for a special symmetry of the wave function. However, the associated energy is very small ($\leq J/3 \approx 0.04$ eV). Contact means that the interaction is due to exchange by spin excitations with momentum $q \sim \pi$, i.e., when polarons are overlapped (see Fig. 10). It has been shown[37] that this magnetopolaron interaction is quantitatively the same for the Néel and Ising backgrounds. (Obviously that additional Coulomb interaction is independent of the magnetic order.) The following consideration was performed on the Ising background since some functions have simpler form for it.

It is possible to combine the ideas of the short-range Coulomb and magnetopolaron effects. The difference between $E^0_{\rm gap}$ (10) when the hole and electron are separated and ΔE_n [Eq. (7), where n denotes the lowest singlet] when they are close is the effective Coulomb attraction of the "bare" hole and electron (singlet and vacancy). We can write an addition to Eq. (6) as

$$\Delta H_c = -V_c \sum_{\langle ll' \rangle, \alpha\beta} n^e_{l\alpha} n^h_{l'\beta}, \tag{13}$$

where n^e is the electron number operator and n^h is the hole number operator. We have found that V_c is really almost independent of U_d and U_p and $V_c \approx 0.4 V_{pd}$. Bare electron-hole attraction in itself does not necessarily mean Frenkel's exciton effect. One has to show that the dressed electron and hole placed closely possess lower energy than the mobile ones. In order to specify the magnetopolaron language we reproduce here the wave function of the magnetic polaron from Ref. 18. For the Ising background it has the very simple form

$$\psi^\dagger_{\uparrow \mathbf{k}} = \frac{1}{\sqrt{N/2}} \sum_n d^\dagger_{n\uparrow} \exp(i\mathbf{k}\mathbf{r}_n),$$
$$d^\dagger_{n\uparrow} = \nu h^\dagger_{n\uparrow} + \mu S^+_n \sum_{n' \in \langle n \rangle} h^\dagger_{n'\downarrow}, \tag{14}$$

where $n \in$ sublattice with the spin $s = -1/2$, $h^\dagger_{n\alpha}$ are primarily hole operators; at $t/J > 1$, $\nu^2 \approx 1/2$, $\mu^2 \approx 1/8$, and n' is the nearest neighbor site to n. Thus, the ansatz consists of a mixture of bare holes and holes with one overturned spin. Contact interaction of these polarons with opposite spins was considered in Ref. 37 (see Fig. 10). The gain in energy for "attracting polarons" arises from the pure t-J model effects[37] and effective Coulomb attraction (13). The loss in energy results from the restriction of mobility. Competition of these evident effects (without the Coulomb interaction) provides bound states up to t/J =2–3.[36,37] Simply acting in the spirit of magnetic polaron interaction[37] we obtain $\Delta E_{\rm exc} \approx 0.35 V_c$. Thus at $V_{pd} = 1$ eV, $\Delta E_{\rm exc} = 0.14$ eV.

More accurate variational construction of the exciton-magnetopolaron wave function yields $\Delta E_{\rm exc} \approx 0.5 V_c$ (at t/J =2.5–3). This wave function consists of the mixture of "bare" hole and electron at neighbor sites and hole and electron with overturned spins. It is schematically shown in Fig. 11(a). It differs from the simple product of the two polaron wave functions (electron and hole) at the nearest neighbors by the different weights of its components. Thus, at $V_{pd} = 1$ eV, $\Delta E_{\rm exc} \simeq 0.2$ eV, which is slightly less than the observed $\Delta E^{\rm exp}_{\rm exc}$ =0.25–0.35 eV.[29] In our calculations the interaction between next-nearest-neighbor magnetic polarons was neglected, which produced a small effect for the pure t-J model[37] but may be essential for the problem with attraction. [These configurations are shown in Figs. 11(b) and 11(c).]

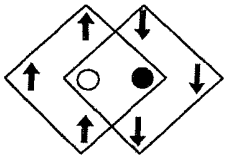
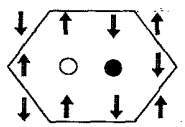
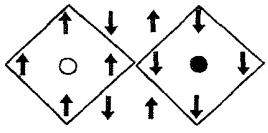


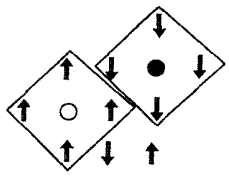
FIG. 10. Configuration of interacting magnetic polarons from Ref. 36.



a



b



c

FIG. 11. (a) Exciton magnetopolaron; (b) and (c) next-nearest-neighbor magnetic polarons.

The answer may also partly lie in the rest of the long-range Coulomb interaction.

VI. DISCUSSION

The detailed quantitative consideration of some of the effective parameters of the low-energy models related to description of the high-T_c superconductors presented in this work relies heavily on our earlier works. In these works consistent mapping of the three-band Hubbard model onto the effective t-J model[12,13,15] has been produced. Taking into account all essential interactions enables us to correctly calculate local energies of various sets of states with different numbers of particles and matrix elements of interesting transitions. The combination of properties of the local bare hole and electron (ZR singlet and vacancy) and their magnetic polaron nature as the carriers allows us to approach the calculation of some observable quantities adequately.

We have calculated the superexchange constant J and charge-transfer gap $E_{\rm gap}$. Their experimental values strongly constrict a possible variation interval for the quantity of great interest: the t/J ratio in the t-J model. Self-consistent calculation of this ratio for a wide range of parameters places it into the region t/J =2.4–2.7. Narrowed ranges for the three-band model parameters have also been determined: t_{pd} =1.2–1.4 eV and Δ =2.5–4.5 eV. They coincide quite well with earlier cluster calculations, which supports our self-consistent procedure. An excitonic state of the Frenkel type induced by the short-range Coulomb interaction with energy lower than the charge-transfer transition by approximately 0.2 eV is found.

We have also compared the width of the peak in $\epsilon_2(\omega)$ at 2.3 eV from Ref. 29 that is of the order of 0.5 eV with the total width of the charge-transfer spectrum. This total width is equal to the combined widths of the vacancy and singlet bands. According to Eq. (11), the width of the hole band is $W_h = 2.0J$ at $t_h = 2.55J$ and the width of the electron band is $W_e = 2.2J$ at $t_e = 2.75J$. The resulting total width of the charge-transfer spectrum is about 0.6 eV. Thus the narrowness of the $\epsilon_2(\omega)$ spectrum can also be easily reproduced by the magnetic polaron approach.

One of the essential questions for the CuO_2-plane systems, which we only briefly touched, is the phonon polaron effect. Its importance for real carriers in the CuO_2 plane is intensively investigated.[40,41] If it does not have a projection on optics, or, as we believe, the Frank-Condon principle is applicable, our calculated t_h/J ratio is the upper limit of the real parameter. This is due to mass renormalization for real carriers. There is another view on the polaron effect (see Refs. 29 and 42). It is stated that electron-phonon interaction lies in an intermediate range and thus the Frank-Condon principle is not obeyed. If such is the situation, we underestimate the depth of the bands (or overestimate $E_{\rm gap}$), and the effective t_h should be increased. Our estimation shows that this increasing of t_h is no more than 30%. Naturally, this problem requires additional investigation.

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APPENDIX

In this Appendix we present some details of the technical treatment of the problems discussed in the paper. According to our previous work,[15] we use the following transformation from the primary oxygen p_{lx}, p_{ly} operators to the operators $q_l, \tilde{q}_l$ of the symmetrical and antisymmetrical oxygen states:

$$(q_l, \tilde{q}_l) = \sum_{\mathbf{k}} [p_{\mathbf{k}a} \cos(k_x/2) \pm p_{\mathbf{k}b} \cos(k_y/2)] \times (1+\gamma_{\mathbf{k}})^{-1/2} \exp(ikl), \tag{A1}$$

where $p_{\mathbf{k}a}$ is the Fourier image of p_{lx} for q_l and p_{ly} for $\tilde{q}_l$; p_{kb} is the Fourier image of p_{ly} for q_l and p_{lx} for $\tilde{q}_l$; $\gamma_{\mathbf{k}}$=[cos(k$_x$a)+cos(k$_y$a)]/2. The summation in Eq. (A1) is produced over the Brillouin zone, and the lattice constant $a = 1$.

Since the ground states of both undoped and doped systems do not consist of the antisymmetrical oxygen state,[6,7,15] the reformulated Hamiltonian (1), (2) where only essential degrees of freedom are kept is conveniently expressed through the local and hopping parts.[15] The local part is

$$\begin{aligned} H_{\rm loc} &= \epsilon_d \sum_{l,\alpha} n^d_{l\alpha} + (\epsilon_p - \mu_0 t_{pp}) \sum_{l,\alpha} n^q_{l\alpha} \\ &\quad + U_d \sum_l n^d_{l\uparrow} n^d_{l\downarrow} + V_{pd} f_0 \sum_{l,\alpha\beta} n^d_{l\alpha} n^q_{l\beta} \\ &= + U_p h_0 \sum_l n^q_{l\uparrow} n^q_{l\downarrow} + 2 t_{pd} \lambda_0 \sum_{l,\alpha} (d^\dagger_{l\alpha} q_{l\alpha} + \text{H.c.}) , \end{aligned}$$

$$\begin{aligned} \Delta H_{\rm int} &= V_{pd} f_1 \sum_{\langle ll' \rangle, \alpha\beta} n^d_{l\alpha} n^q_{l'\beta} \\ &= -2 U_p h_1 \sum_{\langle ll' \rangle} (\mathbf{S}^q_l \mathbf{S}^q_{l'} - \tfrac{1}{4} n^q_l n^q_{l'}) , \end{aligned} \tag{A2}$$

with

$$\mathbf{S}^q_l = \tfrac{1}{2} q^\dagger_{l\alpha} \boldsymbol{\sigma}_{\alpha\beta} q_{l\beta}, \quad n^q = n^q_{l\uparrow} + n^q_{l\downarrow}. \tag{A3}$$

The hopping part is

$$H_{\rm hop} = 2t_{pd}\lambda_1 \sum_{\langle ll'\rangle,\alpha} (d^\dagger_{l\alpha} q_{l'\alpha} + \text{H.c.})$$
$$= -2t_{pp}\mu_1 \sum_{\langle ll'\rangle,\alpha} q^\dagger_{l\alpha} q_{l'\alpha},$$

$$\Delta H_{\rm hop} = V_{pd} f' \sum_{\langle ll'\rangle,\alpha\beta} n^d_{l\alpha}[q^\dagger_{l\beta} q_{l'\beta} + \text{H.c.}]$$
$$+U_p h' \sum_{\langle ll'\rangle,\alpha} n^q_{l\alpha}[q^+_{l\bar{\alpha}} q_{l'\bar{\alpha}} + \text{H.c.}]. \qquad \text{(A4)}$$

All constants λ, μ, f, h in Eqs. (A2) and (A4) are of the Wannier nature. Their Fourier images and magnitudes are given in Ref. 15. In order to group them together we reproduce

λ_0=0.9581, λ_1=0.1401, μ_0=1.4567, μ_1=0.2678 ,
f_0=0.9180, f_1=0.2430, h_0=0.211, h_1=0.059 ,
f'=0.1342, h'=0.030 .

We have treated the Hamiltonian $H_{\rm loc} + \Delta H_{\rm int}$ (A2) in the self-consistent mean-field approximation[15] which enables us to solve the problem of the local states at sites with different numbers of holes. The matrix elements of the Hamiltonian (A4) between states with a singlet or vacancy at different sites in initial and final states lead to the following expressions for the hopping constants (6):

$$t_h = 2t\lambda_1(W_1V' - \sqrt{2}U_1U')(W_1U' - \sqrt{2}V_1V')$$
$$+ t_p\mu_1(W_1U' - \sqrt{2}V_1V')^2/2$$
$$- (V_{pd}f_1W_1U' - \sqrt{2}U_ph_1V_1V')(W_1U' - \sqrt{2}V_1V'),$$
$$t_e = -4t\lambda_1 U''V'' - t_p\mu_1(V'')^2, \qquad \text{(A5)}$$

where U', V' are the coefficients of the $|f\rangle$ state nearest to the singlet, and U'', V'' of those nearest to the vacancy. The coefficients U', V' and U'', V'' are slightly different from the ones in Eq. (4) due to a distortion of $|f\rangle$ states by the nearest vacancy or singlet. This distortion has its origin in the short-range Coulomb repulsions due to U_p and V_{pd} terms in the Hamiltonian (2). Note that it changes the energy of the $|f\rangle$ states nearest to a vacancy or singlet. We take this effect into consideration when we calculate the quantity $E^0_{\rm gap}$. Thus, we have for the total energies E_c and E_v for a singlet and a vacancy [Eqs. (6) and (10)]

$$E_c = E_s + 4E_{fs} - 5E_f,$$
$$E_v = E_0 + 4E_{f0} - 5E_f, \qquad \text{(A6)}$$

where E_s and E_0 are the local energies of singlet and vacancy; E_{fs} and E_{f0} are the energies of the $|f\rangle$ states nearest to a singlet or vacancy and E_f is the energy of the $|f\rangle$ state in an undoped sample. These states are schematically shown in Fig. 2.

When the vacancy and the two-hole state are created at the neighbour sites (see Fig. 1) the difference in energy between this state and the ground state is determined by the relation

$$\Delta E_n = E_n + E_0 + 3E_{fs} + 3E_{f0} - 8E_f. \qquad \text{(A7)}$$

These energies ΔE_n are involved in calculation of the superexchange constant J [Eq. (7)] and in the energy of the Coulomb attraction of a singlet and vacancy in Sec. V.

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