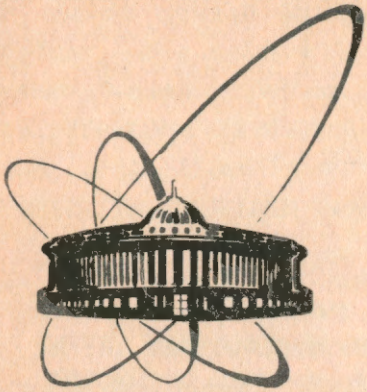
91-227



объединенный
институт
ядерных
исследований
дубна

E17-91-227

S.V.Lovtsov*, V.Yu.Yushankhai

EFFECTIVE SINGLET-TRIPLET MODEL FOR CuO_2 PLANE IN OXIDE SUPERCONDUCTORS: THE CHARGE FLUCTUATION REGIME

Submitted to "Physica C"

*Faculty of Physics, Irkutsk University,
Irkutsk 664003, USSR

1991

Ловцов С.В., Юшанхай В.Ю.

E17-91-227

Эффективная синглет-триплетная модель для CuO_2 плоскости в оксидных сверхпроводниках: режим зарядовых флуктуаций

На основе двухзонной модели Эмери дан вывод эффективного гамильтониана, описывающего низкоэнергетические свойства CuO_2 плоскости в оксидных сверхпроводниках. Подход основан на разложении по амплитуде межузельной p-d-гибридизации $2\,t^{dp}\lambda_{ij}$ ($\lambda_{ij} \leq 0{,}14$) между Cu-орбиталью и симметризованной O-орбиталью в представлении Ванье. Результирующий гамильтониан представлен в терминах синглетных и триплетных локальных состояний. Дальнейшее сведение к однозонной t-J модели представляется правдоподобным.

Работа выполнена в Лаборатории теоретической физики ОИЯИ.

Препринт Объединенного института ядерных исследований. Дубна 1991

Lovtsov S.V., Yushankhai V.Yu.

E17-91-227

Effective Singlet-Triplet Model for CuO_2 Plane in Oxide Superconductors: the Charge Fluctuation Regime

A reduction from the two-band Emery model to an effective Hamiltonian describing low-energy properties of the CuO_2 plane in oxide superconductors is presented. The approach is based on an expansion in the inter-site p-d hybridization amplitude $2\lambda_{ij}\,t^{dp}$ ($\lambda_{ij} \leq 0.14$) between Cu-orbital and symmetric O-orbital in the Wannier representation. The resulting Hamiltonian is presented in terms of Hubbard operators for singlet and triplet local states. A further reduction to the single-band t-J model seems to be plausible.

The investigation has been performed at the Laboratory of Theoretical Physics, JINR.

Preprint of the Joint Institute for Nuclear Research. Dubna 1991

1. Introduction

Since the discovery of high-T_c superconductivity a great deal of efforts have been undertaken to obtain an effective Hamiltonian that describes the low-energy properties of the CuO_2 plane in oxide superconductors. As has been pointed out by Rice [1], the aim is to find the simplest Hamiltonian eliminating all terms which are not relevant to the determination of the superconducting fixed point. Anderson was the first to propose [2] a Hamiltonian of that sort which is now well-known as the t-J model. This model can be derived from the single-band Hubbard model in the large U-limit [3,4]. However, it is believed now that a generalized (multi-band) Hubbard model proposed by Emery [5] should be taken as a starting point of this reduction. The first step in this way was made by Zhang and Rice [6] (ZR hereafter) who suggested a perturbation scheme on the basis of a simplified version of the Emery model. They have found that the Cu-O hybridization strongly binds a hole on each square of O-ions with the central Cu-ion to form a local singlet state with a binding energy (with respect to the triplet state and the nonbonding state) greater that the singlet bandwidth. After neglecting the triplet hole band ZR arrived at the t-J model. This has led to considerable discussion in the literature on the validity and limitations of the reduction from a multi-band description to a one-band model [7-13]. Particularly, it has been pointed out that the approach suggested by ZR is not controlled by a small parameter, i.e. the same physical quantities determine the binding energy of the singlet, its band width, and the parameter of the singlet-triplet mixing as well.

In this paper, starting with the same Hamiltonian as in Ref.[6] we present a different perturbative approach to the reduction which allows us to overcome the objection mentioned

above and to obtain the effective singlet-triplet Hamiltonian for the CuO_2 plane in oxide superconductors. At the same time, based on the Wannier representation for oxygen hole operators our consideration is similar to that by ZR. Thus, the Hamiltonian obtained, though somewhat more complicated, in main features resembles the t-J model. We believe that the latter is contained in our resulting Hamiltonian as a limiting case for a certain range of relevant model parameters and low temperatures.

2. Model Hamiltonian

In the Emery model [5] the largest energy parameter is the on-site Coulomb interaction between copper d-electrons, U_d, and the largest hopping integral, t^{dp}, corresponds to the σ-bond between copper $d(x^2-y^2)$- and oxygen p(x,y)- orbitals. The simplified version of the Emery model, we start with, is gained by keeping these parameters and ignoring the on-site Coulomb interaction on the O-ions, the intersite Cu-O Coulomb interaction and direct O-O hopping terms. Two different regimes should be distinguished for a hole dynamics. The first one, the so-called spin fluctuation regime, corresponds to a weak Cu-O hybridization, $t^{dp}/\Delta = t_1' << 1$ and $t^{dp}/(U_d - \Delta) = t_2' << 1$, where $\Delta = \varepsilon_p - \varepsilon_d > 0$ is the energy distance between p- and d-atomic levels and $(U_d - \Delta)$ is the energy of the higher Hubbard sublevel at copper sites with respect to the p-level. Small t_1' and t_2' provide a commonly used perturbation expansion used by ZR [6,10] and other authors [9,12,13] in deriving an effective one-band description. However, more representative values of the parameters in copper oxides do not give not so weak hybridization. Actually, according to [7,11,16] one has $t_1' \cong 1/2$ and $t_2' \cong 1/4$ and hence only the latter parameter may provide a good perturbation expansion. That is the change fluctuation regime because the charge degrees of freedom at copper sites should be taken into account. Just this regime is treated below.

After projecting out the upper Hubbard sublevel the two-band Emery model can be written to the second order in t_2' as

Объединенный институт ядерных исследований БИБЛИОТЕКА

(in hole notation):

$$H = -\Delta \sum_{i\sigma} X_i^{\sigma\sigma} + \sum_{im\sigma} t_{im}^{dp} \left[X_i^{\sigma 0} p_{m\sigma} + \text{h.c.} \right] + \qquad (1)$$

$$+ \sum_{imn\sigma} \frac{t_{im}^{dp} t_{in}^{dp}}{(U_d - \Delta)} \left[X_i^{\sigma\bar{\sigma}} p_{m\bar{\sigma}}^{+} p_{n\sigma} - X_i^{\bar{\sigma}\bar{\sigma}} p_{m\sigma}^{+} p_{n\sigma} \right].$$

Here Hubbard operators for a hole at Cu $\vec{i}$-sites are introduced:

$$X_i^{\sigma 0} \equiv d_{i\sigma}^{+}(1-n_{i-\sigma}), \quad X_i^{\sigma\sigma} \equiv n_{i\sigma}(1-n_{i-\sigma}), \quad X_i^{\sigma\bar{\sigma}} \equiv d_{i\sigma}^{+} d_{i\bar{\sigma}}$$

where $\bar{\sigma} = -\sigma$, and $t_{im}^{dp} = t^{dp} S_{im}$ is the hybridization amplitude which is nonzero for nearest neighbours. The sign convension $S_{im}=\pm 1$ is chosen in agreement with ZR [6]. The atomic energy of the p-level is chosen to be zero, $\varepsilon_p=0$. According to ZR let us now define a symmetric combination of four O-hole states around a Cu site

$$p_{i\sigma}^{(s)} = \frac{1}{2} \sum_{m} S_{im} \, p_{m\sigma} \qquad (2)$$

which are not orthogonal. The Wannier representation provides the orthogonal symmetric O-states with corresponding operators $c_{i\sigma}$ instead of $p_{i\sigma}^{(s)}$. The relation between them is familiar,

$$p_{i\sigma}^{(s)} = \sum_{j} \lambda(\vec{i}-\vec{j}) \, c_{j\sigma} \qquad (3)$$

where (N being the number of Cu sites)

$$\lambda(\vec{i}-\vec{j}) = \frac{1}{N} \sum_{k} \left[1 - \frac{1}{2} (\cos k_x + \cos k_y) \right]^{1/2} e^{i\vec{k}(\vec{i}-\vec{j})}. \qquad (4)$$

One can check that $\lambda(\vec{i}-\vec{j})$ rapidly decreases with the distance $(\vec{i}-\vec{j})$. Particularly, several largest values of $\lambda(\vec{i}-\vec{j})$ are presented in Table 1.

After introducing the notation $V_{ij}=2t^{dp}\lambda(\vec{i}-\vec{j})$, the Hamiltonian (1) can be represented in the following form

$$H = H_0 + H_{int}, \quad H_0 = \sum_i H_0^{(i)}, \quad H_{int} = \sum_{i \neq j} H_{int}^{(i,j)} \qquad (5)$$

where

$$H_0^{(i)} = -\Delta \sum_{0} X_i^{\sigma\sigma} + V_0 \left[X_i^{\sigma 0} c_{i\sigma} + \text{h.c.} \right] + J \sum_{\sigma} \left[X_i^{\sigma\bar{\sigma}} c_{i\bar{\sigma}}^{+} \, c_{i\sigma} - X_i^{\bar{\sigma}\bar{\sigma}} c_{i\sigma}^{+} c_{i\sigma} \right], \qquad (6)$$

$$H_{int}^{(i,j)} = V_{ij} \sum_{0} \left[X_i^{\sigma 0} c_{j\sigma} + \gamma \left[X_i^{\sigma\bar{\sigma}} c_{i\bar{\sigma}}^{+} \, c_{j\sigma} - X_i^{\bar{\sigma}\bar{\sigma}} c_{i\sigma}^{+} c_{j\sigma} \right] + \text{h.c.} \right] \qquad (7)$$

and $\gamma = V_0/(U_d-\Delta)$, $J=\gamma V_0$. Here $H_0^{(i)}$ gives the on-site interaction with the Cu-O hybridization term $\sim V_0 = 2t^{dp}\lambda_0$ (where $\lambda_0=0.96$), the hopping term $H_{int}^{(i,j)}$ with any distance between $\vec{i}$ and $\vec{j}$ sites is governed by rather small parameters $V_{ij}=2t^{pd}\lambda(\vec{i}-\vec{j})$. Hence, the on-site Hamiltonian will be diagonalized exactly and the hopping terms will be treated as a perturbation.

3. Diagonalization of H_0 and perturbation expansion in $2t^{dp}\lambda_{ij}/\Delta$

The diagonalization of H_0 is performed at each site independently. There are two different fermionic degrees of freedom at a site and we choose the auxiliary representation for them following Long [14] in main features. Namely, we define two kinds of creation (annihilation) operators: $g_{i\sigma}^{+}(g_{i\sigma})$ for an oxygen and $f_{i\sigma}^{+}(f_{i\sigma})$ for a copper hole, respectively. Then the complete set of basic vectors for one-site states can be subdivided into two sectors. The first one is given by (a site index is dropped):

$$f_\sigma^{+}|0\rangle \equiv |f_\sigma\rangle, \quad f_0^{+} g_\uparrow^{+} g_\downarrow^{+} |0\rangle \equiv |\xi_\sigma\rangle, \quad \frac{1}{\sqrt{2}} \sum_{\sigma} 2\sigma \, f_0^{+} g_{\bar{\sigma}}^{+} |0\rangle \equiv |\psi\rangle, \qquad (8)$$

$$\left(f_\uparrow^{+} g_\uparrow^{+}, \; f_\downarrow^{+} g_\downarrow^{+}, \; \frac{1}{\sqrt{2}} \sum_{\sigma} f_\sigma^{+} g_{\bar{\sigma}}^{+} \right) |0\rangle \equiv \left(|\tau_1\rangle, |\tau_{-1}\rangle, |\tau_0\rangle \right)$$

with one copper hole at the site; $\sigma = \pm 1/2$. The vectors $|\psi\rangle$

and $|\tau_\alpha\rangle$ in (8) give the singlet and triplet states, respectively. The vector $|\xi_0\rangle$ represents the only three-particle state at the site. The second sector is given by the following basic vectors

$$|0\rangle, \quad g_0^+|0\rangle \equiv |g_\sigma\rangle, \quad g_\uparrow^+ g_\downarrow^+ |0\rangle \equiv |\varphi\rangle \qquad (9)$$

with no copper hole operator.

Now we represent how the operators $X_i^{\sigma 0}$ and $c_{i\sigma}^+$ act in the united state space defined above. That representation is given by the following formulas

$$X^{\sigma 0}=|f_\sigma\rangle\langle 0| + \frac{2\sigma}{\sqrt{2}}|\psi\rangle\langle g_{\bar\sigma}| + \frac{1}{\sqrt{2}}|\tau_0\rangle\langle g_{\bar\sigma}| + |\tau_{2\sigma}\rangle\langle g_\sigma| + |\xi_\sigma\rangle\langle\varphi|, \qquad (10)$$

$$c_\sigma^+=|g_\sigma\rangle\langle 0| + 2\sigma|\varphi\rangle\langle g_{\bar\sigma}| + \frac{2\sigma}{\sqrt{2}}|\psi\rangle\langle f_{\bar\sigma}| - \frac{1}{\sqrt{2}}|\tau_0\rangle\langle f_{\bar\sigma}| - \qquad (11)$$

$$- |\tau_{2\sigma}\rangle\langle f_\sigma| - \frac{1}{\sqrt{2}}|\xi_0\rangle\langle\psi| - \frac{2\sigma}{\sqrt{2}}|\xi_0\rangle\langle\tau_0| - 2\sigma|\xi_{\bar\sigma}\rangle\langle\tau_{2\bar\sigma}|.$$

Analogously, the on-site Hamiltonian $H_0^{(i)}$ is represented as

$$H_0^{(i)}=\sum_\sigma [\Delta |g_{i\sigma}\rangle\langle g_{i\sigma}| + V_0(|g_{i\sigma}\rangle\langle f_{i\sigma}| + h.c.)] +$$

$$+[\Delta |\varphi_i\rangle\langle\varphi_i| + \sqrt{2}\,V_0(|\varphi_i\rangle\langle\psi_i| + h.c.) - 2J|\psi_i\rangle\langle\psi_i|] +$$

$$+[\Delta |0_i\rangle\langle 0_i| - J\sum_\sigma |\xi_{i\sigma}\rangle\langle\xi_{i\sigma}| - \Delta]. \qquad (12)$$

We diagonalize $H_0^{(i)}$ by applying the canonical transformation

$$\tilde H_0^{(i)}=\exp(S_i)\, H_0^{(i)} \exp(-S_i)$$

with the generators S_i

$$S_i= \theta_1 \left[|g_{\sigma i}\rangle\langle f_{\sigma i}| - h.c.\right] + \theta_2 \left[|\varphi_i\rangle\langle\psi_i| - h.c.\right],$$

$$tg(2\theta_1) = (2V_0/\Delta), \quad tg(2\theta_2) = \sqrt{2}\,2\,V_0/(\Delta+2J). \qquad (13)$$

Finally, we obtain

$$\tilde H_0^{(i)}= \sum_{w\sigma} E(w)|w_{i\sigma}\rangle\langle w_{i\sigma}| + \sum_v E(v)|v_i\rangle\langle v_i| \qquad (14)$$

where $w = \xi, g, f$; $v = 0, \varphi, \psi$; and the energy of these localized states are defined as follows

$$E(\xi)=-J\,, \quad E(g\,,f\,) = \frac{1}{2}\left[\Delta \pm \sqrt{\Delta^2+(2V_0)^2}\,\right], \qquad (15)$$

$$E(0) = \Delta\,, \quad E(\varphi\,,\psi\,) = \frac{1}{2}\left[(\Delta-2J) \pm \sqrt{(\Delta+2J)^2+2(2V_0)^2}\,\right].$$

We note that three triplet states from (8) possess the same energy which is zero in our notation, i.e. $E(\tau)=0$. The on-site energy spectrum given by (15) is presented in Fig.1 for various values of the ratio t^{dp}/Δ, (in numerical calculations throughout in this paper we choose U_d by putting $t^{dp}/(U_d-\Delta) \simeq 1/4$)

One can clearly see that at any value of the on-site hybridization parameter $V_0(=2t^{dp}\lambda_0)$ the states from the sector (9) are separated from the f-sector (8) by the energy distance not smaller than the charge transfer gap Δ. So in the following we shall call the former as the upper subspace and the latter as the lower subspace. Just the lower subspace determines the low-energy physics in the system. It is worth noting also that though the three-particle state (ξ-level) lies in the lower subspace, it can be neglected in the small doping limit we treat below. A doped hole goes to the singlet state, which provides the lowest energy in the system. This state is well separated from the triplet states. Particularly, at $t^{dp}/\Delta \simeq 1/2$ one has $E(\tau)-E(\psi) \simeq 1.5\Delta$.

As the next step, we apply the canonical transformation (13) to the hopping Hamiltonian

$$\tilde{H}_{int} = \exp(\sum_i S_i)\, H_{int}\, \exp(-\sum_i S_i) \qquad (16)$$

and $\tilde{H}^{(i,j)}_{int}$ is clearly obtained from (7) by replacing

$$X_i^{\sigma 0} \to \tilde{X}_i^{\sigma 0} = e^{S_i} X_i^{\sigma 0} e^{-S_i}\ ,\ c^+_{i\sigma} \to \tilde{c}^+_{i\sigma} = e^{S_i} c^+_{i\sigma} e^{-S_i}\ . \qquad (17)$$

Let us now introduce the projection operator P_1 onto the lower subspace and P_2 onto the upper subspace. Then the hopping Hamiltonian $\tilde{H}_{int}$ involves different kinds of contributions: $P_1\tilde{H}_{int}P_1$ and $P_2\tilde{H}_{int}P_2$ correspond to the lower and upper subspaces, respectively, while $\tilde{H}_{mix} = P_1\tilde{H}_{int}P_2 + P_2\tilde{H}_{int}P_1$ mixes these subspaces.

Our aim is to obtain the effective Hamiltonian H_{eff} describing the low-energy physics in the system. Requiring $|V_{ij}|/\Delta \ll 1$, it can be written formally to the second order in V_{ij}/Δ as

$$H_{eff} = P_1\tilde{H}_0 P_1 + P_1\tilde{H}_{int}P_1 - P_1\tilde{H}_{int}P_2 \frac{1}{\tilde{H}_0 - \tilde{E}^{(0)}} P_2\tilde{H}_{int}P_1 \qquad (18)$$

where $\tilde{E}^{(0)}$ is the energy of a refering ground state. Taking into account that the largest value of $|\lambda_{ij}|$ is for the nearest (i,j)-sites and equals $|\lambda_1| \simeq 0.14$ we have $|V_{ij}|/\Delta \leqslant 2t^{dp}\lambda_1/\Delta$. Hence, the expansion (18) is valid if

$$t^{dp}/\Delta \ll 1/(2|\lambda_1|) \simeq 4\ . \qquad (19)$$

We emphasize that the requirement (19) on t^{dp} is more soft than that one commonly uses, i.e. $t^{dp}/\Delta \ll 1$, in derivation of the effective second-order Hamiltonian in the spin fluctuation regime [3,4,6,8-13]. Moreover, the expansion procedure developed here is more suitable for copper oxides where $t^{dp}/\Delta \simeq 1/2$.

4. Effective Hamiltonian: zero- and first-order contributions

To obtain effective Hamiltonian (18) explicitly, let us first note that the zero-order part $P_1\tilde{H}_0 P_1$ comes from (14) after omitting the terms acting in the upper subspace and, of course, the three-particle contribution (ξ-term) due to a small doping. Further the first-order term, $P_1\tilde{H}_{int}P_1$ is obtained from $\tilde{H}_{int}$ by the following replacement

$$\tilde{X}_i^{\sigma 0} \to P_1\tilde{X}_i^{\sigma 0}P_1 \equiv \tilde{X}^{\sigma 0}_{i11};\ \tilde{c}^+_{i\sigma} \to P_1\tilde{c}^+_{i\sigma}P_1 \equiv \tilde{c}^{+11}_{i\sigma}\ . \qquad (20)$$

Making some simple algebra one obtains

$$(\tilde{X}^{\sigma 0}_{i11};\ \tilde{c}^{+11}_{i\sigma}) = 2\sigma A^{\psi}_{x;c}|\psi_i\rangle\langle f_{i\bar{\sigma}}| + \qquad (21)$$
$$+ A^{\tau}_{x;c}\left(\frac{1}{\sqrt{2}}|\tau_{i0}\rangle\langle f_{i\bar{\sigma}}| + |\tau_{i2\sigma}\rangle\langle f_{i\sigma}|\right)$$

where

$$A^{\psi}_{c;x} = \left(\frac{1}{\sqrt{2}}\cos\theta_1\cos\theta_2 + \sin\theta_1\sin\theta_2\ ;\ -\frac{1}{\sqrt{2}}\sin\theta_1\cos\theta_2\right),$$

$$A^{\tau}_{c;x} = (-\cos\theta_1\ ;\ -\sin\theta_1)\ . \qquad (22)$$

To present the results in a more convenient form, from now we introduce the new type of Hubbard operators defined as follows

$$X_i^{\sigma\sigma'} \equiv |f_{i\sigma}\rangle\langle f_{i\sigma'}|\ ;\ X_i^{\psi\psi} \equiv |\psi_i\rangle\langle\psi_i|\ ;\ X_i^{\psi\sigma} \equiv |\psi_i\rangle\langle f_{i\sigma}|; \qquad (23)$$

$$X_i^{\alpha\alpha'} \equiv |\tau_{i\alpha}\rangle\langle\tau_{i\alpha'}|\ ;\ X_i^{\psi\alpha} \equiv |\psi_i\rangle\langle\tau_{i\alpha}|;\ X_i^{\alpha\sigma} \equiv |\tau_{i\alpha}\rangle\langle f_{i\sigma}|;$$

$(\alpha = 0, \pm 1)$, etc.

For example, in terms of these operators the zero-order effective Hamiltonian is written as

$$P_1\tilde{H}_0 P_1 = E(f)\sum_{i\sigma} X_i^{\sigma\sigma} + E(\psi)\sum_i X_i^{\psi\psi} \qquad (24)$$

and the triplet sector in our notation is located at the zero energy $E(\tau)=0$. For the first-order contribution we obtain finally the form

$$P_1\tilde{H}_{int}P_1 = \sum_{i\neq j,\sigma} V_{ij}\Big\{ K_{\psi\psi} X_i^{\psi\sigma} X_j^{\sigma\psi} + \qquad (25)$$

$$+\tfrac{1}{2}K_{\tau\tau}\Big[X_i^{0\sigma} X_j^{\sigma 0} + 2\, X_i^{2\sigma,0} X_j^{\sigma,2\sigma} + \sqrt{2}\,(X_i^{2\sigma,\sigma} X_j^{\bar{\sigma},0} + \text{h.c.})\Big] -$$

$$- 2\sigma K_{\psi\tau}\Big[\frac{1}{\sqrt{2}} X_i^{\psi\sigma} X_j^{0\sigma} + X_i^{\psi,\sigma} X_j^{\bar{\sigma},2\bar{\sigma}} + \text{h.c.}\Big]\Big\}$$

where

$$K_{\psi\psi} = 2\,\tilde{A}_x^{\psi} A_c^{\psi}\,, \quad K_{\tau\tau} = 2A_x^{\tau} A_c^{\tau}\,, \quad K_{\psi\tau} = \tilde{A}_x^{\psi} A_c^{\tau} + A_c^{\psi} A_x^{\tau}\,. \qquad (26)$$

Here $\tilde{A}_x^{\psi} = A_x^{\psi} - \sqrt{2}\gamma\cos\theta_1\cos\theta_2$ and all coefficients $A_{x;c}^{\mu}$ are defined in (22). Several contributions are involved in Hamiltonian (25): the first term $\sim K_{\psi\psi}$ corresponds to singlet-singlet hoppings, subsequent terms $\sim K_{\tau\tau}$ represent the hoppings from triplet to triplet states and the hybridization between them, and, at last, the terms $\sim K_{\psi\tau}$ represent the hybridization between singlet and triplet states.

The results of numerical analysis for the coefficients $K_{\mu\nu}$ from (26) are presented in Fig.2 . However, more representative quantities are the hopping integrals between neighbouring $\vec{i}$ and $\vec{j}$ sites, $V_{ij}K_{\mu\nu} = 2t^{dp}\lambda_1 K_{\mu\nu}$. Particularly, at $t^{dp}/\Delta \simeq 1/2$ one has for the singlet-singlet hopping $2t^{dp}\lambda_1 K_{\psi\psi} \simeq 0.16\Delta$ (0.48eV. at $\Delta\simeq 3$eV) and for triplet-triplet hopping $2t^{dp}\lambda_1 K_{\tau\tau} \simeq -0.12\Delta$. We note these values to be by an order of magnitude smaller than the relevant energy distance $E(\tau)-E(\psi)\simeq 1.5\Delta$ (see Fig.1). Moreover, we obtain (at $t^{dp}/\Delta\simeq 1/2$) that the singlet-triplet mixing parameter is quite small $2t^{dp}\lambda_1 K_{\psi\tau}\simeq -0.03\Delta$. These findings are consistent with the ZR result and hence make plausible further reduction of

the singlet-triplet description to the one-band t-J model [10]. In this case $t=2t^{dp}\lambda_1 K_{\psi\psi}$.

Recently, Hybersten et al. [11] have used a cluster method to reduce a multi-band description to the one-band, the so-called t-t'-J, model. They added a t'-term describing the next nearest neighbour hopping of the singlet and obtained the following estimations: $t\simeq 0.44$eV. and $t'\simeq -0.06$eV. In our consideration $t'=2t^{dp}\lambda_2 K_{\psi\psi}$ with $-\lambda_2\simeq 0.02$ (see Table 1); hence $t'=(\lambda_2/\lambda_1)t\simeq +0.07$eV. It is remarkable that both t and $|t'|$ parameters obtained here are very close to that of Ref.[11].

Now we demonstrate more clearly that results obtained by ZR [10] and others [12] are contained in ours as a limiting case. Actually, for small on-site hybridization $2V_0=4t^{dp}\lambda_0 \ll \Delta$, let us expand the $\cos\theta_{1;2}$ and $\sin\theta_{1;2}$ functions to the second order in $t^{dp}/\Delta \ll 1/(4\lambda_0) \simeq 1/4$. Then the singlet binding energy relative to the triplet states is

$$E(\tau)-E(\psi) = 8\,\lambda_0^2\,(\,t_1+t_2\,) \qquad (27)$$

which is very close to the ZR result (here $t_1=(t^{dp})^2/\Delta$ and $t_2=(t^{dp})^2/(U_d-\Delta)$). Analogously, for the nearest neighbours hopping and mixing parameters in Hamiltonian (25) we obtain (i,j=n.n.)

$$V_{ij}K_{\psi\psi} = -8\lambda_0\lambda_1\left(\tfrac{1}{2}t_1+t_2\right);\quad V_{ij}K_{\tau\tau} = 4\lambda_0\lambda_1 t_1;$$

$$V_{ij}K_{\psi\tau} = 4\sqrt{2}\,\lambda_0\lambda_1 t_2. \qquad (28)$$

Taking into account that $8\lambda_0\lambda_1\simeq -1$ we again come to the ZR result for the singlet-singlet hopping parameter. Moreover, all the parameters (28) coincide with that of Ref [12] obtained in the spin fluctuation regime $t^{dp}/\Delta \ll 1$, $t^{dp}/(U_d-\Delta) \ll 1$.

However, we emphasize that the estimations (27) and (28) follow from our consideration in a more strong limit, $t^{dp}/\Delta \ll 1/4$, while $t^{dp}/(U_d-\Delta) \ll 1$. Just the exact diagonalization of the on-site Hamiltonian (6) establishes this limit as a real weak hybridization (or spin fluctuation) regime.

Strictly speaking, this limit is not realized in copper oxides, where $t^{dp}/\Delta \simeq 1/2$. The more general approach valid for rather strong hybridization $t^{dp}/\Delta \ll 1/(2|\lambda_1|) \simeq 4$ is developed here, which gives effective Hamiltonian (24) and (25).

5. Second-order contributions. Superexchange interaction

In this section our aim is to obtain the contributions of second order in V_{ij}/Δ to the effective Hamiltonian H_{eff}, i.e. the last term in (18). However, instead of the operator form, given by(18), we have chosen an equivalent way of derivation based on the canonical Schriffer-Wolff transformation. This transformation is more convenient because it allows one to take into account all the necessary virtual processes on an equal footing. Requiring that the final effective Hamiltonian should not include the term linear in $\tilde{H}_{mix}=P_1\tilde{H}_{int}P_2+$ h.c.,we obtain the following equation for the generator W of the canonical transformation

$$[\ \tilde{H}_0\ ,\ W\ ] = \tilde{H}_{mix}\ . \qquad (29)$$

Then the second order contribution $H^{(2)}_{eff}$ is formally given by

$$H^{(2)}_{eff} = \frac{1}{2} P_1 [\ W\ ,\ \tilde{H}_{mix}\ ]\ P_1\ . \qquad (30)$$

To present it explicitly, we decompose $\tilde{H}_{int}$, defined in (5), (7), and (16), into two parts: $\tilde{H}_{int}=\tilde{H}'_{int}+\tilde{H}''_{int}$, which is obvious. Then, particularly, the part $P_1\tilde{H}'_{int}P_2$ can be written as

$$P_1\tilde{H}'_{int}P_2 = \sum_{i\neq j} V_{ij}\Big[\ \tilde{X}^{\sigma 0}_{i12}(\tilde{c}^{12}_{j\sigma} + \tilde{c}^{11}_{j\sigma}) + \tilde{c}^{+12}_{i\sigma}\ (\tilde{X}^{0\sigma}_{j12} + \tilde{X}^{0\sigma}_{j11})\ +$$

$$+\ (\ \tilde{X}^{\sigma 0}_{i11}\tilde{c}^{12}_{j\sigma} + \tilde{c}^{+11}_{i\sigma}\ \tilde{X}^{0\sigma}_{j12})\Big] \qquad (31)$$

and the operators entering into (31) are defined by (21) and the following formulas

$$(\tilde{X}^{00}_{i12};\ \tilde{c}^{12}_{i0}) = 2\sigma F_{x;c}|f_{i\bar{\sigma}}\rangle\langle\varphi_i|\ ,$$

$$(\tilde{X}^{00}_{i12};\ \tilde{c}^{+12}_{i0}) = L_{x;c}\Big[|f_{i0}\rangle\langle 0| + \frac{1}{\sqrt{2}}|\tau_{i0}\rangle\langle g_{i\bar{\sigma}}| + |\tau_{i2\sigma}\rangle\langle g_{i\sigma}|\Big] +$$

$$+ 2\sigma\ M_{x;c}|\psi_i\rangle\langle g_{i\bar{\sigma}}| \qquad (32)$$

$$F_{c;x} = \Big(\frac{1}{\sqrt{2}}\cos\theta_1\sin\theta_2 - \sin\theta_1\cos\theta_2;\ -\frac{1}{\sqrt{2}}\sin\theta_1\sin\theta_2\Big)\ ,$$

$$L_{c;x} = (\ -\sin\theta_1\ ;\ \cos\theta_1)\ , \qquad (33)$$

$$M_{c;x} = \Big(\frac{1}{\sqrt{2}}\sin\theta_1\cos\theta_2 - \cos\theta_1\sin\theta_2;\ \ \frac{1}{\sqrt{2}}\cos\theta_1\cos\theta_2\Big).$$

The part $\gamma\ P_1\tilde{H}''_{int}P_2$ can be represented in the same manner, which we omit for clarity. After some lengthy calculation one can check that the second order contributions can be finally written as

$$H^{(2)}_{eff} = \sum_{i\neq j} \frac{2V^2_{ij}}{\Delta}\Big[\ a\ \tilde{S}_i\ \tilde{S}_j - \frac{b}{4} N_i\ N_j\ \Big] + \qquad (34)$$

$$+ \sum_{i\neq j} \frac{2V^2_{ij}}{\Delta}\Big[\ d^{\alpha}_{\mu\nu} S^{\alpha}_i\ X^{\mu\nu}_j + c^{\gamma\delta}_{\mu\nu}\ X^{\gamma\delta}_i\ X^{\mu\nu}_j\ \Big] + \sum_{i\neq j\neq k} H^{(2)}(ijk).$$

Here S^{α}_i (α = x,y,z) are the 1/2-spin operators to be defined through the Hubbard operators as

$$S^{\alpha}_i = 1/2 \sum_{\sigma\sigma'} \sigma^{\alpha}_{\sigma\sigma'} X^{\sigma\sigma'}_i\ , \qquad N_i = \sum_{\sigma} X^{\sigma\sigma}_i$$

where σ^{α} are the Pauli matrices. The operators $X^{\mu\nu}_j(\mu,\nu,\gamma,\delta = \psi,0,\pm 1)$ act in the space of singlet and triplet states. Summation over equal indices in (34) is implied.We deliberately subdivide boson-like operators into two classes, $\{S^{\alpha}\}$ and $\{X^{\mu\nu}\}$, to extract explicitly superexchange interaction between localized spins given by the first sum in (34). Three-

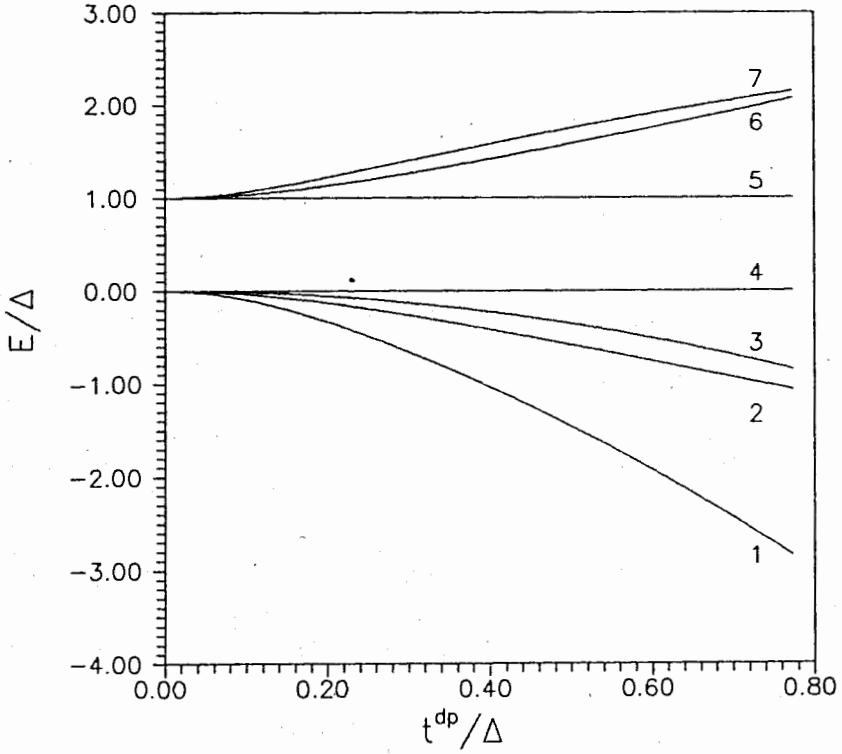


Fig.1. Calculated energies of the localized states as functions of the radio t^{dp}/Δ. Curves from 1 to 7 correspond to $E(\psi)$,$E(f)$,$E(\xi)$,$E(\tau)$,$E(0)$,$E(g)$ and $E(\varphi)$,respectively. The singlet state energy $E(\psi)$ is the lowest.

Table 1. The values of the coefficients $\lambda(i-j)$ as functions of the intersite distance $(\vec{i}-\vec{j}) = n_x\vec{a}_x + n_y\vec{a}_y$

$n_y \backslash n_x$	0	1	2
0	0.96	-0.14	-0.013
1	-0.14	-0.02	-0.07
2	-0.013	-0.07	-0.003

site interaction $H^{(2)}(ijk)$ is out of our consideration. In this paper we also do not explicitly specify the c-number matrix factor $d^{\alpha}_{\mu\nu}$ and $c^{\gamma\delta}_{\mu\nu}$ and restrict ourselves to an analysis of the magnetic interaction between localized spins. The magnetic term that is of the second order in V_{ij} in our consideration, is analogous to the fourth-order (in t^{dp}) term in the conventional expansion and, hence, to the J-term in the t-J model.

The dimensionless factors a and b in (34) are given by

$$a = \frac{\Delta}{2}\left[\frac{\cos^2 2\theta_1}{2E(f)-E(0)-E(\tau)} - \frac{(\cos\theta_2 + 1/\sqrt{2}\,\sin 2\theta_1\,\sin\theta_2)^2}{2E(f)-E(0)-E(\psi)} - \right.$$

$$\left. - \frac{(\sin\theta_2 - 1/\sqrt{2}\,\sin 2\theta_1\,\cos\theta_2)^2}{2E(f)-E(0)-E(\varphi)} + \frac{\sin^2 2\theta_1}{(U_d-\Delta)}\right], \qquad (35)$$

$$b = a - 2\cos^3 2\theta_1 .$$

The form of the denominators in (35) shows which kinds of virtual processes contribute to the superexchange constant. Namely,as the first step, one of two particles should be excited from the f-level to the initially empty level (with the energy E(0)), while the other goes either to a triplet τ-level ($E(\tau)=0$ in our notation), or a singlet ψ-, or φ-level starting from the same f-level. The second step should restore their position. It is worth noting that the second order (in $t^{dp}/(U_d-\Delta)$) contribution from the upper Hubbard sublevel is taken into account in the starting Hamiltonian (1). This contribution enters additively into the exchange constant as the last term in (35).

We have numerically analyzed the factors a, that is positive, and b as functions of t^{dp}/Δ . These factors allows one to estimate the superexchange antiferromagnetic integral $J_{dd}(i,j) \equiv 2a(2t^{dp}\ \lambda_{ij})^2/\Delta$, and the additional integral $I_{dd}(i,j) \equiv 2b\ (2t^{dp}\ \lambda_{ij})^2/\Delta$, entering into Hamiltonian (34), for any distance between i and j sites. For the neighbouring sites (i,j=n.n.) these quantities are presented in Fig.3.

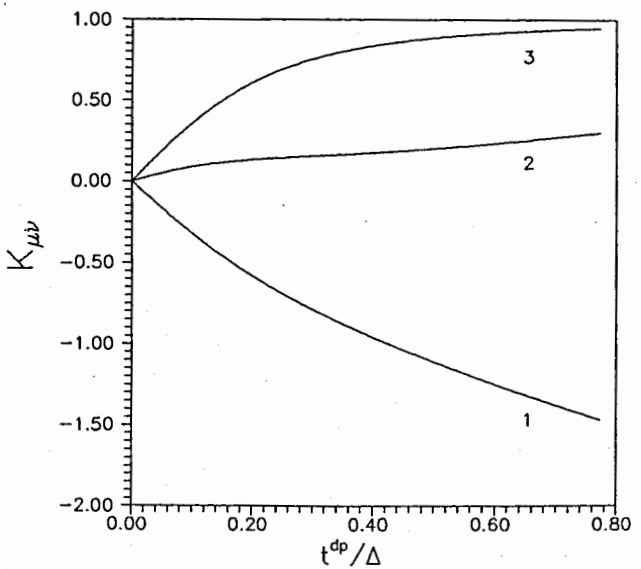


Fig.2. Calculated scale factors $K_{\mu\nu}$ entering into Hamiltonian (25): (1) $K_{\psi\psi}$,(2) $K_{\psi\tau}$,(3) $K_{\tau\tau}$.

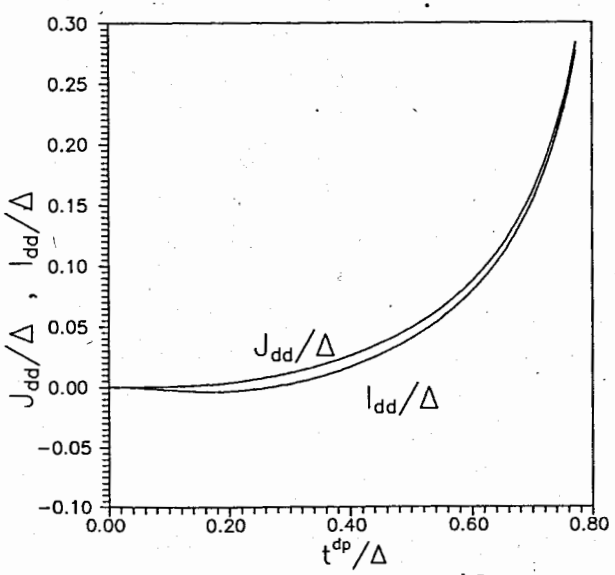


Fig.3. Calculated superexchange antiferromagnetic integral J_{dd} and additional integral I_{dd} taken for n.n. sites . The definitions are given in the text.

Particularly, at $t^{dp}/\Delta \simeq 1/2$ we obtain $J_{dd}(i,j=n.n.) = J_{dd} \simeq 0.05\Delta$. Then if $\Delta = 3$ eV., one has $J_{dd} \simeq 0.15$ eV. that is very close to the experimental value [15] and to the estimation given in Ref. [11]. We note that in the typical approach [6,16] one has as a rule a larger value of J_{dd} in the same range of model parameters.

For the second neighbours (diagonal bond) the corresponding superexchange integral is

$$J_{dd}(i,j=n.n.n.) \equiv J'_{dd} = J_{dd}(\lambda_2/\lambda_1)^2 \simeq 0.001\Delta .$$

If $\Delta \simeq 3$ eV., then $J'_{dd} \simeq 0.003$ eV. which is again close to that of Rev.[11]. We note that in our approach there is an obvious strict relation between the singlet-singlet hopping parameters t, t' and the exchange integrals J_{dd}, J'_{dd}, namely

$$\frac{J'_{dd}}{J_{dd}} = \left[\frac{t'}{t}\right]^2 .$$

The values for these parameters obtained numerically by Hyberstsen et.al. [11] on the basis of a cluster method approximately obey this relation as well.

6. Conclusion

It this paper we developed an approach to reduction of the two-band p-d-model of the CuO_2 plane to an effective Hamiltonian giving a low-energy behaviour of the system. Following the main idea of ZR [6] we eliminate the operators associated with oxygen coordinates and describe the system in terms of local singlet and triplet states. However, our consideration is valid in a more wide range of the p-d hybridization parameter t^{dp}, including the charge-fluctuation regime, in comparison with ZR who treated the limit of relatively weak hybridization, i.e. the spin-fluctuation regime. Two successive steps are used in our approach: first, the on-site interactions involving the hybridization term $V_0 = 2t^{dp}\lambda_0$ are kept exactly and, second, intersite interactions $\sim V_{ij} = 2t^{dp}\lambda_{ij} \ll V_0$ are treated perturbatively. This approach is developed to the second order in V_{ij}/Δ and the effective Hamil-

tonian is presented in terms of Hubbard operators associated with singlet and triplet local states. We analyzed the resulting Hamiltonian in some limiting cases and estimated the relevant quantities for the most representative values of the model parameters. Along this way we obtained a good agreement with other considerations [6,10-12]. So a further reduction of the effective Hamiltonian to the one-band t-J, or t-t'-J, model [6,11] seems to be very plausible. However, to justify this reduction, some subtle questions remain to be examined carefully.

References

1. T.M.Rice, Phys. Scripta T29 (1989) 72
2. P.W.Anderson , Science 235 (1987) 1196
3. C.Gros, R.Joynt and T.M.Rice, Ptys Rev. B36 (1987) 381
4. K.Huang and E.Manousakis, Phys.Rev. B36 (1987) 8302
5. V.J.Emery, Phys.Rev. Lett, 58 (1987) 2794
6. F.C.Zhang and T.M.Rice, Phys.Rev. B37 (1988) 3759
7. F.Mila, Phys. Rev. B38 (1988) 11358
8. H.Eskes and G.A.Sawatzky, Phys.Rev. Lett.,61 (1988) 1415
9. V.J.Emery and G.Reiter, Phys.Rev. B38 (1988)11938; ibid.,B41(1990) 7247
10. F.C.Zhang and T.M.Rice, Phys.Rev. B41 (1990) 7243
11. M.S.Hybertsen, E.B.Stechel, M.Schluter and D.R.Jennison, Phys.Rev. B41 (1990) 11068
12. J.L.Shen and C.S.Ting, Phys.Rev. B41 (1990) 1969
13. H.B.Pang, T.Xiang, Z.B.Su and L.Yu., Phys.Rev. B41 (1988) 7209
14. M.W.Long, Z.Phys. B71 (1988) 23
15. R.P.Singh et.al., Phys.Rev. Lett. 62 (1989) 2736
16. T.M.Rice, F.Mila F.C.Zhang, Preprint ETH-TH/90-27.

Received by Publishing Department
on May 23, 1991.