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Correlations in multicomponent systems with application to the Emery model

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Abstract. A new class of variational functions is proposed to study the correlations and charge fluctuations in multicomponent systems. It is applied to the Emery model of the CuO_2 plane of a high-temperature superconductor. A correlated state is constructed by making use of the local unitary transformations acting on the uncorrelated single-band Hartree–Fock function. The transformations perform the p–d hybridization depending on the number of holes on the clusters, each containing one d orbital and the appropriate Wannier combination of p orbitals. The non-perturbative single-band Hamiltonian is obtained. It is shown that the band approach to treating the transformed Hamiltonian is quite adequate. In this way the correlated paramagnetic state with large intercluster fluctuations and the antiferromagnetic (AF) state have been obtained. The hole localization on clusters in the AF state and the decay of AF ordering upon doping is traced.

1. Introduction

For a long time, and more recently in connection with high-temperature superconductors (HTSC), a variety of successful methods have been proposed to describe the electronic correlations in strongly correlated materials. Some of them that originated from Hubbard's work [1, 2] are now extended over various cluster-band models [3–5]. They use the decoupling of the equations of motion for electronic operators [1, 2, 6].

Direct construction of the variational many-electron wavefunction with correlations has been achieved by Gutzwiller and others [7–11] or in the slave-boson technique [12, 13]. In both methods uncontrolled approximations are introduced in calculations of matrix elements or in the account of the constraints.

A number of studies of the t – J model [14–18] have used expansions in t/U where t is the hopping integral and U is the on-site interaction. For the CuO_2 plane of an HTSC such an expansion leads to separation of the copper spin system and that of oxygen holes. Then the ground state (GS) of the undoped system corresponds to spins localized on copper and the oxygen extra holes are bounded with d holes in singlet states (Zhang–Rice singlet [15] or more complicated objects [17, 18]). Practically, the parameters of such expansions ($4t/U_d$, $4t/|\delta\varepsilon|$) are not small; here $\delta\varepsilon = \varepsilon_d - \varepsilon_p$.

An improved expansion has been proposed recently [19, 20] that is convergent even at $\delta\varepsilon \rightarrow 0$. It is based on the cluster perturbation method in which the main intracluster part of the p–d and p–p bond interactions has already been taken into account in zero approximation and the cluster (or cell) consists of the d orbital of Cu and the corresponding Wannier combination [21] of oxygen p orbitals. In the perturbative approach the construction of the effective spin Hamiltonian and the single-band Hamiltonian for additional holes requires the definition of the basis of zero approximation (the ground manifold) [19, 20]. Such a

ground manifold includes [4, 5, 19, 20] the singly occupied hybrid orbitals of clusters in the insulating case and the additional lowest singlet states of clusters with double holes at some doping. However, there is no unique self-consistent way to choose the ground manifold in the perturbation method in the case of a doped insulator, and the problem of treating the effective Hamiltonian remains unsolved as yet. So it may be useful to try an alternative approach for treating the correlations and charge fluctuations in multicomponent systems.

The present work is devoted to the variational non-perturbative study of correlations and charge fluctuations in the Emery model in the frame of a modified band approach. A new type of correlated variational wavefunction Ψ is constructed by use of local unitary transformations acting on the single-band Hartree–Fock (HF) function Φ or on the antiferromagnetic (AF) function of the unrestricted HF approach. So we take

$$\Psi = W\Phi \quad (1)$$

$$W = \prod_n W_n \quad (2)$$

where W is the product of local unitary operators W_n acting on the states of different non-overlapping clusters $\{d_n, a_n\}$. Each cluster is just the same cluster as has been used in the cell perturbation method [19, 20]. It includes the strongly correlated d orbital and the appropriate Wannier combination a_n of the p orbitals [21].

After the transformation, the total Hamiltonian $\tilde{H} = W^+HW$, consisting of the intracluster part and the pair intercluster interactions, is represented in the form

$$\tilde{H} = W^+HW = \sum_n \tilde{h}_n + \sum_{n \neq m} g_{nm}(\lambda, \lambda') \tilde{R}_{\lambda n} \tilde{Q}_{\lambda' m}. \quad (3)$$

It acts in the space of the original function Φ . Here each of the transformed operators $\tilde{L}_n = \tilde{h}_n$ or \tilde{R}_n or \tilde{Q}_n refers to the n th cluster and is connected with the corresponding components of the original Hamiltonian H by the equation

$$\tilde{L}_n = W_n^+ L_n W_n. \quad (4)$$

Our aim is to show that even at large U_d the single-band HF function Φ after the transformation can provide lower energy than the states of localized type with one hole on each cluster. This allows us to apply the very convenient and well elaborated band approach to the new problem determined by the transformed Hamiltonian \tilde{H} . In particular, the use of the single-band AF function allows us to find in a self-consistent way both the degree of hole localization and intercluster charge fluctuations. It also allows us to consider metal-like systems with doping on an equal footing with insulating undoped systems.

The function (1) differs from that of Gutzwiller's *ansatz* [7, 8]. In the latter a non-unitary transformation of the HF function suppresses the configurations with doubly occupied strongly correlated sites. In the function (1) a unitary transformation W redistributes the hole density between the weakly and strongly correlated orbitals and creates the singly and doubly occupied states of d orbitals in the necessary proportion.

The outline of this paper is as follows. In section 2 the unitary transformation operators referring to one cluster with two orbitals are defined. In section 3 the single-band mapping of the simplified Emery model by the unitary transformations is performed. The accuracy of such mapping is estimated in the limit of non-interacting holes (section 4). Section 5 presents the results for the correlated paramagnetic (PM) and AF states. The hole localization and intercluster charge fluctuations are discussed.

2. Unitary transformations for the two-orbital cluster

Consider an isolated cluster with two orbitals described by the creation operators a_σ^+ and d_σ^+ .

Any operators acting on the function of the cluster may be expanded via the complete set of products of the Hubbard operators A_{ij} and D_{lm} [2,3] referring to a and d orbitals respectively:

$$A_{ij} = \begin{pmatrix} P_0^a & u^a & a_\alpha^1 & a_\beta^1 \\ u^{+a} & P_2^a & -a_\beta^{+2} & a_\alpha^{+2} \\ a_\alpha^{+1} & -a_\beta^2 & P_\alpha^a & a_\alpha^+ a_\beta \\ a_\beta^{+1} & a_\alpha^2 & a_\beta^+ a_\alpha & P_\beta^a \end{pmatrix}_{ij} \quad (5)$$

with

$$\begin{aligned} P_0^a &= (1 - n_\sigma^a)(1 - n_{-\sigma}^a) & P_\sigma^a &= n_\sigma^a(1 - n_{-\sigma}^a) & P_2^a &= n_\sigma^a n_{-\sigma}^a \\ u^{+a} &= a_\alpha^+ a_\beta^+ & a_\sigma^1 &= a_\sigma(1 - n_{-\sigma}^a) & a_\sigma^2 &= a_\sigma n_{-\sigma}^a. \end{aligned} \quad (6)$$

Here $\sigma = \alpha, \beta$ is a spin index; equivalent notation $\sigma = 1, 2$ will be used below; P_0, P_2 or P_σ are the projection operators to the states with empty and doubly occupied a (d) orbital or the projection to the state of one particle with spin σ at the orbital. The known properties of operators A_{ij} (or D_{ij}) are

$$A_{ij}^+ = A_{ji} \quad A_{ij} A_{kl} = \delta_{jk} A_{il} \quad \sum_{i=1}^4 A_{ii} = P_0 + P_2 + P_\alpha + P_\beta = I. \quad (7)$$

The local unitary operator W acting on the two-orbital cluster {d, a} is generated by anti-Hermitian operators Z_i :

$$W = W_1 W_2 W_3 = \exp(Z_1 + Z_2 + Z_3) \quad (8)$$

$$Z_1 = -\alpha \sum_{\sigma} [a_{\sigma}^+ d_{\sigma} (1 - n_{-\sigma}^a)(1 - n_{-\sigma}^d) - \text{HC}] \quad (9)$$

$$Z_2 = -\sum_{\sigma} \{ [u a_{\sigma}^+ n_{-\sigma}^a d_{\sigma} (1 - n_{-\sigma}^d) - v d_{\sigma}^+ n_{-\sigma}^d a_{\sigma} (1 - n_{-\sigma}^a)] - \text{HC} \} \quad (10)$$

$$Z_3 = -\beta \sum_{\sigma} (a_{\sigma}^+ d_{\sigma} n_{-\sigma}^a n_{-\sigma}^d - \text{HC}). \quad (11)$$

Each of the mutually commutative operators Z_i acts only on a state with definite number (i) of particles on a cluster and redistributes the amplitudes in that state. The operator W_2 acts on the singlet component of a state with two particles. Explicit expressions for W_i via A_{ij} and D_{ij} are given in the appendix. Operators W corresponding to the different non-overlapping clusters commute with each other.

At $\alpha = u = v = \beta \equiv \gamma$ the operator $W(\alpha, \beta, u, v)$ takes the form

$$W_0(\gamma) = \exp(Z_0) = I + (\cos \gamma - 1) \left(n^a + n^d - 2 \sum_{\sigma} n_{\sigma}^a n_{-\sigma}^d \right) + \left(\frac{\sin \gamma}{\gamma} \right) Z^0 \quad (12)$$

$$Z_0 = Z_0(\gamma) = -\gamma \sum_{\sigma} (a_{\sigma}^+ d_{\sigma} - d_{\sigma}^+ a_{\sigma}). \quad (13)$$

It transforms an arbitrary HF state into a new uncorrelated HF state with different hybridization of p and d orbitals. The use of operator $W(\alpha, u, v, \beta)$ of generalized hybridization with $\alpha \neq \beta \neq u \neq v$ is needed to obtain the correlated state.

It is important that any of the transformed operators $\tilde{L}_n = W_n^+ L_n W_n$ (and in particular the constituents $\tilde{h}_n, \tilde{R}_n, \tilde{Q}_n$ of the transformed Hamiltonian (3)) can be represented in the form

$$\tilde{L}_n = \sum \tilde{c}(ijkl) A_{ij} D_{kl}. \quad (14)$$

Here the coefficients \tilde{c} of the expansion are connected by linear relations with the same expansion coefficients of the corresponding original operator L_n , and the explicit expressions of the transformation matrix via variational parameters α, u, v, β have been obtained.

3. The one-band mapping of the Emery model

Consider a simplified model of the CuO_2 plane in which the on-site interaction of holes on Cu only is added to the linear Emery model:

$$H = H_0 + \sum_n U_d n_{n\sigma}^d n_{n-\sigma}^d \quad (15)$$

$$H_0 = \sum_{n,\sigma} [\varepsilon_d n_{n\sigma}^d + \varepsilon_p (n_{n\sigma}^x + n_{n\sigma}^y)] + t \sum_{n,m} \sum_{\sigma} (d_{n\sigma}^+ p_{m\sigma} \zeta_{nm} + \text{HC}). \quad (16)$$

Here d and p = {x or y} refer to the d orbital of copper and to the p_x, p_y orbitals of oxygen; $\zeta_{nm} = \pm 1$ are defined by the signs of the orbitals. Following [19, 20] we rewrite H via the basis $\{d_n a_n c_n\}$ of d orbitals and appropriate symmetric and antisymmetric Wannier orbitals [21]:

$$a_{n\sigma}^+ = N^{-1/2} \sum_k e^{-ikn} a_{k\sigma}^+ \quad c_{n\sigma}^+ = N^{-1/2} \sum_k e^{-ikn} c_{k\sigma}^+. \quad (17)$$

Here

$$a_{k\sigma}^+ = (-e_x s_x x_{k\sigma}^+ + e_y s_y y_{k\sigma}^+) / F_k \quad c_{k\sigma}^+ = (e_x s_y x_{k\sigma}^+ + e_y s_x y_{k\sigma}^+) / F_k \quad (18)$$

with

$$e_{x(y)} = i \exp(ik_{x(y)}/2) \quad (19)$$

$$F_k = (s_x^2 + s_y^2)^{1/2} \quad s_{x(y)} = \sin(k_{x(y)}/2) \quad \delta\varepsilon = \varepsilon_d - \varepsilon_p.$$

In the basis $\{d_n a_n c_n\}$ the Hamiltonian (15) takes a form that is convenient for local transformations:

$$H = \sum_{n,\sigma} [\varepsilon_d n_{n\sigma}^d + \varepsilon_p n_{n\sigma}^a + \frac{1}{2} U_d n_{n\sigma}^d n_{n-\sigma}^d + 2t (d_{n\sigma}^+ a_{n\sigma} + \text{HC})] + 2t \sum_{n \neq m} \sum_{\sigma} F(n-m) (d_{n\sigma}^+ a_{m\sigma} + \text{HC}) \quad (20)$$

with

$$F(l) = F_l = N^{-1} \sum_k e^{ikl} F_k.$$

Here F_k is determined by equation (19). The term $\varepsilon_p n_{n\sigma}^c$ is already omitted in H since the c band decouples from the a - d problem of interest; it refers to the non-bonding p band.

In order to obtain an approximate single-band description of problem (20), we make the transformation of the single-band function Φ . For example, let Φ be defined in the subspace of weakly correlated a_n orbitals, so that

$$\Psi = W\Phi^a \quad \Phi^a = |\Phi(a^+)0\rangle. \quad (21)$$

This is one possible way to map the multicomponent multiband system by the single-band model.

Note that each of the possible components $|a_{n\sigma}^+0\rangle$ or $|a_{n\uparrow}^+a_{n\downarrow}^+0\rangle$ in the original function Φ^a is transformed by W_n into the whole spectrum of all the possible single-hole states or the singlet double-hole states of the n th cluster, in particular the states with the occupied d orbital.

The transformed Hamiltonian W^+HW acting in the space of arbitrary functions Φ^a in the a subspace is a projection of the modified Hamiltonian (3) on the states with empty d sites:

$$\hat{H}^a = Sp_d\{D_{11}\tilde{H}\} = \sum_n \tilde{h}_n^a + \sum_{n \neq m} g_{nm}(\lambda, \lambda') \tilde{R}_{\lambda n}^a \tilde{Q}_{\lambda' m}^a. \quad (22)$$

Here each of the operators $\tilde{L}^a = \{\tilde{h}_n^a, \tilde{R}_n^a \text{ or } \tilde{Q}_n^a\}$ in the a subspace is derived from the representation (14) of the corresponding transformed operators \tilde{L} in the full a - d space. It is expressed in the form

$$\tilde{L}_n^a = \sum_{i,j} \tilde{c}(ij11) A_{ij} \quad (23)$$

via the same coefficients \tilde{c} as in equation (14).

As a result the transformed Hamiltonian in the subspace of the arbitrary function Φ^a takes the form

$$\begin{aligned} \hat{H}^a = & \sum_{n,\sigma} (E_a n_{n\sigma}^a + \frac{1}{2} V_0 n_{n\sigma}^a n_{n-\sigma}^a) \\ & + 2t \sum_{\sigma} \sum_{n \neq m} F(n-m) (a_{n\sigma}^+ a_{m\sigma} + \text{HC}) (\xi_0 + \xi_1 n_{n-\sigma}) (\eta_0 + \eta_1 n_{n-\sigma}^a). \end{aligned} \quad (24)$$

Here

$$\begin{aligned} V_0 &= vU_d + \kappa_1 \delta\varepsilon + 8tF_0 h_1 \\ E_a &= \frac{1}{2} [(\varepsilon_d + \varepsilon_p) + \kappa_0 \delta\varepsilon + 8tF_0 h_0] \end{aligned}$$

and the expressions for $\kappa_0, \xi_0, \xi_1, \eta_0, \eta_1, h_0, h_1$ and v via the parameters α, u and v of the transformation are given by equations (A11) in the appendix.

Thus, the local unitary transformations being common for all the clusters allow us to reduce the Hamiltonian of multicomponent system (15), (16) to an analogue of the Hubbard one without using the expansion in $4t/U, 4t/\delta\varepsilon$ or in intercluster interaction. The density of particles for the new problem is the same as in the original one. We expect that, choosing the optimal variational parameters, one can get good accuracy of the approach. In the next section we study the accuracy of the single-band mapping of the Emery model with the use of local transformations in the limit of non-interacting particles.

4. Estimation of the accuracy in the non-interacting holes limit

The exact wavefunction Ψ^{ex} and the energy H^{ex} of the problem (16) with $U_d = 0$ can be represented in the form

$$\Psi^{\text{ex}} = \prod_k \exp[Z_0(\gamma_k, a_k, d_k)] \prod_{\sigma} \prod_k^F a_k^{\pm} |0\rangle \quad (25)$$

$$H^{\text{ex}} = 2N^{-1} \sum_k^F [\bar{\varepsilon} - (\delta\varepsilon^2/4 + 4t^2 F_k^2)^{1/2}] \quad (26)$$

$$\cot(2\gamma_k) = \delta\varepsilon/(4t F_k) \quad \bar{\varepsilon} = \frac{1}{2}(\varepsilon_d + \varepsilon_p). \quad (27)$$

The index F in (25), (26) defines the range of k variations inside the Fermi boundary and $Z_0(\gamma, a, d)$ is defined by equation (13).

For the same case $U_d = 0$ the optimal locally transformed single-band function and corresponding minimal energy per unit cell are

$$\Psi = \prod_n \exp[Z_0(\gamma, d_n, a_n)] \prod_{\sigma} \prod_k^F a_{k\sigma}^{\pm} |0\rangle \quad (28)$$

$$H(\gamma) = \frac{1}{2}(\varepsilon_d + \varepsilon_p)n - (A^2 + B^2)^{1/2} \quad (29)$$

$$A = \frac{1}{2}n\delta\varepsilon \quad B = 2tN^{-1} \sum_k^F F_k. \quad (30)$$

Here F_k is defined by equation (19), \sum^F is the sum over k inside the Fermi boundary, and n is the hole concentration. The optimal local transformation here corresponds to parameters

$$\alpha = u = v \equiv \gamma = \frac{1}{2} \arccot(A/B).$$

The error $\delta H = H(\gamma) - H^{\text{ex}}$ for the energy appears to be sufficiently small. For example, for the hole concentrations $n = 1$ and 1.5 one gets $\delta H/t = 0.0018$ and 0.0055 at $\delta\varepsilon = -2t$, or $\delta H/t = 0.004$ and 0.012 at $\delta\varepsilon = -4t$. At $\delta\varepsilon = 0$ the approximate function (28) and energy (29) coincide with exact quantities (25) and (26) since in that case $\gamma_k = \text{const} = \gamma_0 = \pi/4$. Even at large hole concentration $n \simeq 1$ the δH values are much less than the energy gain due to the correlations at $U_d \neq 0$. Note that the transformed single-band function cannot describe the admixture of the triplet double-hole states and of the states with three holes on the cluster. But the weights P^T and P_3 of these states in exact function (25) are also negligibly small. For example, at $n = 1$ these weights are $P^T = 0.13 \times 10^{-3}$ and $P_3 = 0.9 \times 10^{-4}$ at $\delta\varepsilon = -2t$ and they vanish at $\delta\varepsilon \rightarrow 0$.

Such a high accuracy of the locally transformed single-band state is due to the multicomponent character of the model and to the choice of the cluster basis with appropriate Wannier orbital. A concentration n of about one hole per unit cell means in fact $n_{\sigma} \simeq 0.25$ of holes with definite spin per orbital. This leads to slight dependence of γ_k on k inside the Fermi boundary in equation (25) and provides high accuracy of the local transformation approach.

5. The results for the correlated PM and AF states: hole localization and intercluster charge fluctuations

The aim of further study is to answer the question of whether the band approach can be adequate in treating the locally transformed single-band Hamiltonian (22) or it is necessary to start from the localized limit and corresponding perturbation approach. This question is very important since the band mean-field (or HF) approach is a well elaborated one and it allows us to obtain quantitative results by a non-perturbative self-consistent procedure as opposed to the perturbative methods [19, 20] based on the wavefunction of localized type.

Consider first the undoped system with $n = 1$ and compare the average energies H calculated with the functions of both types (localized versus non-localized ones).

A function of the first type Ψ_{loc} is composed of singly occupied local orbitals a_n with some spin distribution

$$\Psi_{\text{loc}}(\alpha) = W \prod_n a_{n,\sigma_n}^+ |0\rangle. \quad (31)$$

Since each cluster in Ψ has exactly one particle, the energy H_{loc} per unit cell depends on the parameter α only. Minimization over α yields

$$H_{\text{loc}}/N = \frac{1}{2}(\varepsilon_d + \varepsilon_p) - [(\frac{1}{2}\delta\varepsilon)^2 + 4t^2 F_0^2]^{1/2}. \quad (32)$$

Here F_0 and $\delta\varepsilon$ are defined by equations (19).

At a particular value $\alpha = -\pi/2$ the function (31) and corresponding energy turn into

$$\Psi_{\text{loc}}(\alpha = -\pi/2) = \prod_n d_{n,\sigma_n}^+ |0\rangle \quad H_{\text{loc}}/N = \varepsilon_d. \quad (33)$$

Just such a state has been used for the undoped case in early works [14–18]. However H_{loc} has a minimum at the value α determined by the equation $\tan(2\alpha) = -4tF_0/\delta\varepsilon$. The degenerate states (31) with $W = W(\alpha)$ form the ground manifold in the cell perturbation approach [19, 20]. A large part of the p-d bond energy is already taken into account here since each hole is localized on the lower hybrid orbital of the cluster with appropriate Wannier function a_n . This provides a considerable energy gain in comparison with the energy of state (33) with localization of holes on d orbitals only.

The second type of solution is based on the HF function Φ^a in the correlated function of paramagnetic state

$$\Psi^{\text{PM}} = W \Phi_{\text{HF}}^a \quad \Phi_{\text{HF}}^a = \prod_{\sigma} \prod_k^{\text{F}} a_{k\sigma}^+ |0\rangle. \quad (34)$$

Index F means the product over k inside the Fermi boundary.

Note that for the state (34) the probabilities of each cluster to have one, two or zero holes are $P_1 = n(2-n)/4$, $P_2 = n^2/4$ and $P_0 = (2-n)^2/4$, which are 0.5, 0.25 and 0.25, respectively, for the undoped case. It means that the state (34) implies large intercluster charge fluctuations forbidden in the localized state.

The energy $H^{\text{PM}} = \langle H \rangle$ corresponding to trial function (34) is

$$\frac{H^{\text{PM}}}{N} = 2E_a\rho_0 + V_0\rho_0^2 + 8t \left(q_1 \sum_{m \neq 0} F_m \rho_m - \xi_1 \eta_1 \sum_{m \neq 0} F_m \rho_m^3 \right). \quad (35)$$

Here

$$\rho_m = N^{-1} \sum_k^F e^{ikm} \quad \rho_0 = n/2 \quad q_1 = (\xi_0 + \rho_0 \xi_1)(\eta_0 + \rho_0 \eta_1)$$

n is the hole density and \sum_k^F means summing over k inside the Fermi boundary. We have verified that the spectrum E_k of linearized Hamiltonian $(H^a)_L$ exhibits perfect nesting, i.e. $E_k = E(z)$ depends on the 'radial' characteristics of momentum k only:

$$z = (2 + \cos k_x + \cos k_y)/2. \quad (36)$$

The Fermi boundary is uniquely determined by the hole density according to the equation $z(k_F) = z_F(n)$. Thus, the problem is reduced to the minimization of H^{PM} (34) over α, u, v .

It should be emphasized that the energy H^{PM} obtained in the HF approximation for optimal modified Hamiltonian $\tilde{H}(\alpha \neq u \neq v)$ appears to be considerably lower than the energy of the original problem in the HF approximation. The latter is very close to energy

$$H(\gamma) = \langle \Psi(\gamma) | H | \Psi(\gamma) \rangle \quad (37)$$

for the optimal function of type (28) without correlations ($\alpha = u = v \equiv \gamma$).

Now we can find the energy H^{AF} of the correlated AF state

$$\Psi^{AF} = W \Phi^{AF}(a) \quad (38)$$

constructed on the basis of the one-determinant AF state $\Phi^{AF}(a)$ of the band approach in the a subspace,

$$H^{AF} = \langle \Psi^{AF} | H | \Psi^{AF} \rangle = \langle \Phi^{AF} | \tilde{H} | \Phi^{AF} \rangle.$$

The usual mean-field treatment of the modified Hamiltonian gives

$$\begin{aligned} H^{AF} = & H^{PM}(\rho_m) - \delta_0^2 \left(V_0 - 8t\xi_1\eta_1 \sum_{m \neq 0} \zeta_m F_m \rho_m \right) \\ & - 4t(\xi_1\eta_0 + \eta_1\xi_0 + 2\xi_1\eta_1\rho_0)\delta_0 \sum_{m \neq 0} F_m \delta_m (1 + \zeta_m) + 4t\xi_1\eta_1 \sum_{m \neq 0} F_m \rho_m \delta_m^2 (1 + \zeta_m). \end{aligned} \quad (39)$$

Here H^{PM} depends on ρ_m only and is determined by equation (35), $\zeta_n = (-1)^{n_x + n_y}$, $\zeta_\sigma = \mp 1$ for $\sigma = \pm 1/2$ and the values $\rho_m = \langle \hat{\rho}_m \rangle_a$ and $\delta_m = \langle \hat{\delta}_m \rangle_a$ are the averages over Φ^{AF} of the corresponding operators

$$\hat{\rho}_m = \frac{1}{2} N^{-1} \sum_{k, \sigma} e^{ikm} a_{k\sigma}^+ a_{k\sigma} \quad (40)$$

$$\hat{\delta}_m = -\frac{1}{2} N^{-1} \sum_{k, \sigma} e^{ikm} \zeta_\sigma a_{q\sigma}^+ a_{k\sigma} \quad q = (\pi, \pi) + k. \quad (41)$$

In the site representation these operators are

$$\hat{\rho}_m = \frac{1}{2} N^{-1} \sum_{n, \sigma} a_{n\sigma}^+ a_{n+m, \sigma} \quad \hat{\delta}_m = -\frac{1}{2} N^{-1} \sum_{n, \sigma} \zeta_\sigma \zeta_n a_{n\sigma}^+ a_{n+m, \sigma}. \quad (42)$$

The values $2\rho_0$ and $2\delta_0$ determine the mean density of holes and the spin density on the cluster.

The linearized Hamiltonian of the correlated AF problem

$$\hat{H}_L = \sum_m \frac{\partial H^{AF}}{\partial \rho_m} (\hat{\rho}_m - \rho_m) + \sum_m \frac{\partial H^{AF}}{\partial \delta_m} (\hat{\delta}_m - \delta_m) + H^{AF} \quad (43)$$

determines the self-consistent equations for ρ_m and δ_m . A typical property of the system with nesting is a dependence of Fermi surface on one variable (36) only. This facilitates the numerical solution of self-consistent equations. For undoped AF insulator ($n = 1$) there is additional symmetry: all ρ_m with even $m_x + m_y$, except ρ_0 , are equal to zero.

Figure 1 presents the dependence of the optimal parameters α, u, v of local transformation in correlated PM wavefunction (34) on the on-site interaction U_d for various values of $\delta\varepsilon = 0, -2t, -4t$ at $n = 1$. The values α, u, v appear to be very close to the intracluster parameters α_0, u_0, v_0 that minimize the intracluster part h_n of the Hamiltonian; the corresponding operators $W_1(\alpha_0)$ and $W_2(u_0, v_0)$ transform the states $|a_{n\sigma}^+ 0\rangle$ or $|a_{n\uparrow}^+ a_{n\downarrow}^+ 0\rangle$ to the lowest states of the cluster with one and two holes respectively. At $\delta\varepsilon = -2t$ the values α_0, u_0, v_0 are presented in figure 1 by broken curves. At $U_d \rightarrow 0$ the optimal parameters $\alpha = u = v$ correspond to the uncorrelated state (28). At $U_d \rightarrow \infty$ one gets $v \rightarrow 0$ and the weight of doubly occupied d orbitals

$$P_2^d = \langle n_\sigma^d n_{-\sigma}^d \rangle = \nu \langle n_\sigma^a n_{-\sigma}^a \rangle_a \quad (44)$$

tends to zero. Here ν is given by equation (A11) and $\langle \dots \rangle_a$ means the averaging over $\Phi_{HF}(\mathbf{a})$. The dependences of P_2^d on U_d for the correlated and uncorrelated states (34) and (28) are presented in figure 2. Correlations suppress the doubly occupied d states but produce little change of the total density of holes on Cu. Note that the weights of the double-hole and empty clusters (as opposed to the weights for the d orbitals) are constants and equal to 0.25 for function (34) at $n = 1$: the correlations in the state (34) do not suppress the intercluster charge fluctuations.

The ground-state energies for various trial functions are presented in figure 3 for $\delta\varepsilon = -2t$, $n = 1$ and $0 < U_d < 10t$. The full curves marked by PM and 1 correspond to energies $H(\alpha, u, v)$ and $H(\gamma)$ for states with and without PM correlations, respectively; the broken curves AF+PM and AF correspond to the same quantities for the correlated or uncorrelated AF states ($\alpha \neq u \neq v$ or $\alpha = u = v \equiv \gamma$), respectively. The horizontal line refers to the energy H_{loc} corresponding to function (31), which describes localization of holes on lower hybrid orbitals of clusters. It is worth noting that the ground state (32) of zero order in t corresponds to much larger energy $H = \varepsilon_d = -2$ at $t = 1$. Absolute values of H are given for $\varepsilon_p = 0$.

It is surprising that there is a wide range of parameters where

$$H^{PM} < H_{loc}. \quad (45)$$

For example, it is true for all U_d up to $10t$ at $\delta\varepsilon = 0$, or for $U_d < 6t$ at $\delta\varepsilon = -2t$, or for $U_d < 2t$ at $\delta\varepsilon = -4t$. This means that the energy per unit cell of the charge-transfer intercluster fluctuations,

$$\Delta \simeq 0.25(E_2 + E_0 - 2E_1) > 0$$

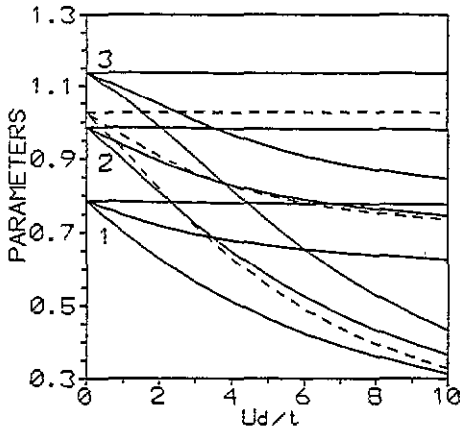


Figure 1. The U_d dependences of generalized hybridization parameters α, u, v of correlated wavefunction (34). In each bunch of curves the higher, central and lower curves refer to $-\alpha, -u, -v$ respectively and bunches 1-3 correspond to $\delta\epsilon = 0, -2, -4$ for the hole density $n = 1$ per site. Broken curves correspond to parameters $-\alpha_0, -u_0, -v_0$ minimizing the intracluster energy only.

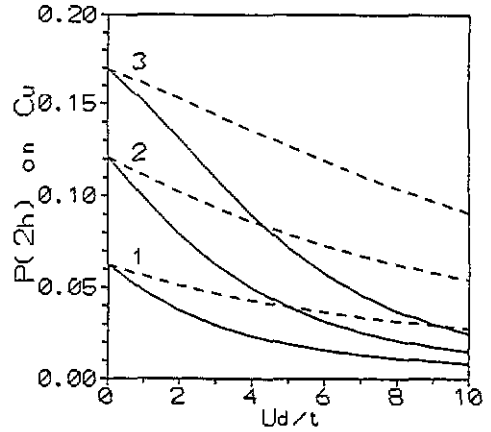


Figure 2. The probabilities (P_2) of double occupation of Cu by holes as a function of U_d calculated with the PM correlated states (34) (full curves) and with approximate uncorrelated functions (28) (broken curves) at $n = 1$. Curves 1, 2, 3 correspond to $\delta\epsilon/t = 0, -2, -4$.

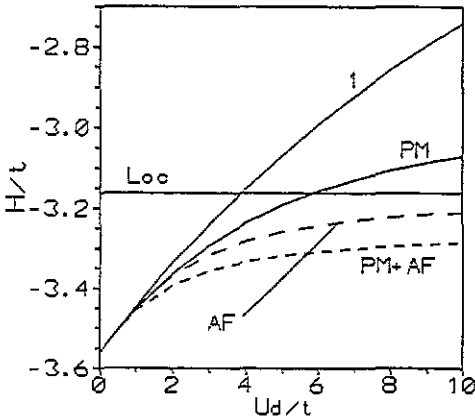


Figure 3. The total energy per unit cell for model $\delta\epsilon/t = -2, n = 1$. Curves 1 and 1 correspond to states (34) or (28) with or without PM correlations. Curves AF+PM and AF refer to similar states with AF ordering. The horizontal line is the energy of localized state (31) after minimization over α .

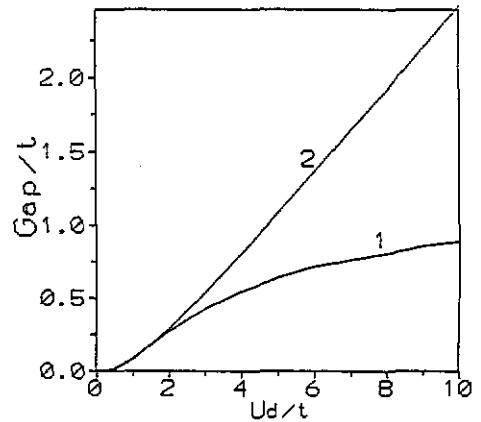


Figure 4. The U_d dependences of the dielectric AF gap at $n = 1$ for $\delta\epsilon/t = -2$. Curves 1 and 2 refer to the correlated AF+PM or uncorrelated AF states.

may be less than the absolute value of negative intercluster part of the p-d bond energy,

$$T^{\text{PM}} \simeq 8t v_0 (\xi_0 + \xi_1 \rho_0) (\eta_0 + \eta_1 \rho_0) \quad v_0 = N^{-1} \sum_k^F (F_k - F_0).$$

Here E_i are the lowest energies of the cluster states with i holes and the parameters α, u, v in the correlated PM state are close to their intracluster values α_0, u_0, v_0 .

Thus we come to the conclusion that the band approach for treating the transformed single-band Hamiltonian can be used successfully as a basis for constructing the insulating AF state of an undoped system. It has some advantage over the cell perturbation method [19,20]. It allows us to obtain in a self-consistent way the degree of localization and can be easily applied to metal-like doped systems.

The correlated AF state has been obtained by the solution of the corresponding self-consistent equations at any parameters α, u, v and by the subsequent minimization of energy (39) over them. Optimal values α, u, v for the AF state are close to those for the PM state. For example, at $\delta\varepsilon = -2t, U = 7t$ the values α, u, v are 1.012, 0.775, 0.493 and 0.980, 0.774, 0.475 for the AF+PM and PM states respectively, and they are close to the intracuster parameters $(\alpha_0, u_0, v_0) = (1.025, 0.765, 0.439)$ that minimize the cluster Hamiltonian h_n . Thus the PM and AF correlations can coexist.

At $n = 1$ the energy H^{AF+PM} of the correlated AF state is always lower than both H_{loc} and H^{PM} for the states (31) and (34) (see figure 3). For example, at $U_d = 7t$ the values $\{H_{loc}, H^{PM}, H^{AF+PM}\}$ are $\{-1.916, -2.149, -2.170\}$ at $\delta\varepsilon = 0, \{-3.161, -3.129, -3.300\}$ at $\delta\varepsilon = -2t$ and $\{-4.770, -4.375, -4.843\}$ at $\delta\varepsilon = -4t$. For the same systems in the AF state the spin densities on the cluster are $2\delta_0 = 0.578, 0.864, 0.954$ for $\delta\varepsilon = 0, -2t$ or $-4t$ respectively (δ_0 is determined by equation (42)). For the undoped system, $2\delta_0 \rightarrow 1$ along with the increase of $|\delta\varepsilon|$, i.e. the correlated AF state approaches the state of localized type with alternating spins. Another attribute of the hole localization is the decrease of the second derivatives of energy over u, v at the optimal values α, u, v for the AF state with increase of U_d . All the above are in line with the result of second-order consideration of the cell perturbation approach [19,20] based on the localized states.

For an undoped insulator the value of the gap in the one-particle spectrum of the linearized Hamiltonian of the AF+PM state is considerably less than the similar gap obtained for the AF state $\Psi^{AF}(\alpha = u = v \equiv \gamma)$ without PM correlations at γ providing the minimum of $H(\gamma)$ (see figure 4). However, the actual states of extra holes and excitations may differ from those of the linearized Hamiltonian, and special study of the problem is needed.

Now we use the correlated variational function of the form (1) to study the doped system with hole concentration $n > 1$. Such metal-like systems are the most difficult ones to study by the standard [14–18] or even improved [19,20] perturbation methods, since for this case it is difficult to validate any simple choice of the ground manifold in perturbation analysis. At the same time the transformed band approach remains adequate in the case of doping. The calculations have been done for the PM state (34) and for the similar AF+PM one. The latter corresponds to the double magnetic unit cell.

The difference in energies of the AF and PM states appears to be small in comparison with the energy of the intercluster charge transfer $\Delta E = (E_0 + E_2 - 2E_1)$. At the same time the gap in the one-particle spectrum of linearized Hamiltonian is sufficiently large (figure 4). In the above approximation the phase transition from AF to PM state occurs at $n \simeq 1.2$ or 1.4 for $\delta\varepsilon = 0$ or $-2t$.

Figure 5 presents the doping dependence of δ_0 and of the probabilities $P_2 = \rho_0^2 - \delta_0^2$ and $P_0 = (1 - \rho_0)^2 - \delta_0^2$ of the double-hole or empty clusters. The value $2\delta_0$ is the staggered spin density on clusters. The values P_2 and P_0 show the degree of hole localization and of intercluster charge fluctuations. The weight P_1 of the single-hole clusters differs from unity even for the undoped case: $P_1 = 0.667$ or 0.875 at $\delta\varepsilon = 0$ or $-2t$.

6. Conclusions

The method of unitary transformations seems to be very useful for constructing the correlated

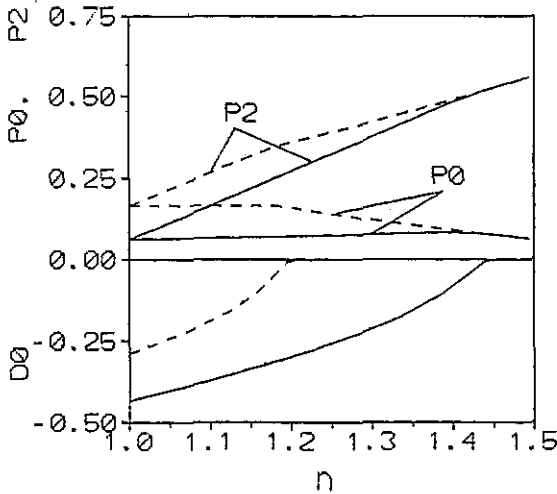


Figure 5. The doping dependences of δ_0 ($\equiv D0$), half of the spin density on cluster, and the weights of the empty and double-hole clusters P_0 and P_2 ($\equiv P0$ and $P2$) at $U_d/t = 7$, $\varepsilon_p = 0$. Broken and full curves refer to $\varepsilon_d/t = 0$ or -2 respectively.

ground state of multicomponent systems at sufficiently high density (up to $n \leq 1.5$). It allows us to take into account two important physical features of strongly correlated systems, namely, the localization of holes on lower orbitals of clusters and the band character of the hole carriers displaying significant charge fluctuations. The proposed variational correlated wavefunction possesses correct permutative and translational symmetry. Similar to Gutzwiller's *ansatz* it describes the local paramagnetic correlations, but the unitarity of the transformation permits us to calculate the ground-state energy without any further approximation, which is needed in Gutzwiller's approach. The PM correlations are described by the operators of the generalized p-d hybridization depending on the number of holes on clusters (each cluster with one strongly correlated d orbital and appropriate Wannier combination of the p orbitals). The PM correlations suppress considerably the probabilities of double occupancy of d orbitals.

Consideration of the undoped antiferromagnetic state of the simplified Emery model of the CuO_2 plane confirms the ideas about its ground state arising from the cell perturbation method [19, 21]: the holes appear to be localized on the lower hybrid orbitals of clusters rather than on the copper site. The variational method determines the degree of localization in a self-consistent non-perturbative way. For the undoped case the coexistence of the PM and AF correlations is proved and the PM correlations lead to a considerable decrease of the dielectric gap. The transition from AF to PM state with doping can be described by the method. Unlike other approaches, the method of optimal unitary transformations provides well founded correlated hopping terms in the modified Hamiltonian and therefore the method can be used to study the possibility of superconductivity of kinematic nature.

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Appendix. Unitary transformations and transformed operators

To obtain the explicit expressions of the operators $W_i = \exp(Z_i)$ it is convenient to rewrite equations (9)–(11) via the Hubbard operators

$$Z_1 = \alpha \sum_{\sigma} (D_{1,2+\sigma} A_{2+\sigma,1} - A_{1,2+\sigma} D_{2+\sigma,1}) \quad (A1)$$

$$Z_2 = \sum_{\lambda, \sigma} \varphi_{\lambda} \zeta_{\sigma} \zeta_{\lambda} (D_{\lambda,2+\sigma} A_{3-\lambda,5-\sigma} - A_{5-\sigma,3-\lambda} D_{2+\sigma,\lambda}) \quad (A2)$$

$$Z_3 = \beta \sum_{\sigma} (D_{2,2+\sigma} A_{2+\sigma,2} - A_{2,2+\sigma} D_{2+\sigma,2}) \quad (A3)$$

where $\sigma = 1, 2$ correspond to the up and down spin projections, $\lambda = 1, 2$, and

$$\zeta_{\sigma} = \mp 1 \text{ at } \sigma = 1, 2 \quad \zeta_{\lambda} = \mp 1, \varphi_{\lambda} = \mu, \nu \text{ at } \lambda = 1, 2. \quad (A4)$$

Using equations (7) one can verify that

$$\begin{aligned} (Z_i)^{2n} &= (C_i)^{2n-2} (-1)^n (Z_i)^2 & (Z_i)^{2n+1} &= (-1)^n (C_i)^{2n+1} Z_i \\ C_i &= \{\alpha, r, \beta\}_i, i = 1, 2, 3 & r &= [2(u^2 + v^2)]^{1/2}. \end{aligned} \quad (A5)$$

A summation of all degrees of Z_i in $\exp(Z_i)$ leads to

$$W_1 = I + (\cos \alpha - 1) \sum_{\sigma} (D_{11} A_{2+\sigma,2+\sigma} + A_{11} D_{2+\sigma,2+\sigma}) + (\sin \alpha / \alpha) Z_1 \quad (A6)$$

$$W_2 = I + (\cos r - 1) \left(\sum_{\lambda, \lambda'} \Gamma_{\lambda \lambda'} D_{\lambda \lambda'} A_{3-\lambda,3-\lambda'} + \sum_{\sigma, \sigma'} \vartheta_{\sigma \sigma'} D_{2+\sigma,2+\sigma'} A_{5-\sigma,5-\sigma'} \right) + (\sin r / r) Z_2 \quad (A7)$$

$$W_3 = I + (\cos \beta - 1) \sum_{\sigma} (D_{22} A_{2+\sigma,2+\sigma} + A_{22} D_{2+\sigma,2+\sigma}) + (\sin \beta / \beta) Z_3 \quad (A8)$$

$$\Gamma_{\lambda \lambda'} = (2/r^2) \begin{pmatrix} u^2 & -uv \\ -uv & v^2 \end{pmatrix}_{\lambda \lambda'} \quad \vartheta_{\sigma \sigma'} = \frac{1}{2} \begin{pmatrix} 1 & -1 \\ -1 & 1 \end{pmatrix}_{\sigma \sigma'}. \quad (A9)$$

The transformed elements of the original Hamiltonian (15), (16) are now easily obtained (the cell index n is omitted here):

$$\begin{aligned} \tilde{n}_{\sigma}^a + \tilde{n}_{\sigma}^d &= n_{\sigma}^a & \tilde{n}_{\sigma}^d - \tilde{n}_{\sigma}^a &= \kappa_0 n_{\sigma}^a + \kappa_1 n_{\sigma}^a n_{-\sigma}^a \\ \tilde{a}_{\sigma} &= \xi_0 a_{\sigma} + \xi_1 a_{\sigma} n_{-\sigma}^a & \tilde{d}_{\sigma} &= \eta_0 a_{\sigma} + \eta_1 a_{\sigma} n_{-\sigma}^a \\ \tilde{a}_{\sigma}^+ \tilde{d}_{\sigma} &= h_0 n_{\sigma}^a + h_1 n_{\sigma}^a n_{-\sigma}^a & \tilde{P}_2^d &= \tilde{n}_{\sigma}^d \tilde{n}_{-\sigma}^d = \nu n_{\sigma}^a n_{-\sigma}^a. \end{aligned} \quad (A10)$$

All coefficients are expressed via parameters α, u, v :

$$\begin{aligned}
 \kappa_0 &= -1 + 2s_\alpha^2 & \kappa_1 &= -2s_\alpha^2 - 2C_r\Gamma_{11} - C_r^2(\Gamma_{11}^2 - \Gamma_{21}^2) \\
 h_0 &= s_\alpha c_\alpha & h_1 &= S_r + S_r C_r (\Gamma_{11} + \Gamma_{21}) - h_0 \\
 \xi_0 &= c_\alpha & \xi_1 &= c_\alpha C_r \Gamma_{11} + s_\alpha S_r \\
 \eta_0 &= s_\alpha & \eta_1 &= -s_\alpha(1 - C_r\Gamma_{21}) + c_\alpha S_r & \nu &= C_r^2\Gamma_{12}^2.
 \end{aligned} \tag{A11}$$

Here $s_\alpha = \sin \alpha$, $c_\alpha = \cos \alpha$, $C_r = -1 + \cos r$, $S_r = (u/r) \sin r$, and r, Γ_{ij} are defined by equations (A5) and (A9). Using equations (A10) one obtains the transformed Hamiltonian (24). At $\alpha = u = v$ one has $h_1 = \xi_1 = \eta_1 = \kappa_1 = 0$ and kinematic interactions of the correlation nature in the Hamiltonian are removed.

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