Spin-cluster states in CuO_2 planes

G. Seibold and E. Sigmund
Institut für Theoretische Physik der Universität Stuttgart, Pfaffenwaldring 57, 7000 Stuttgart 80, Federal Republic of Germany

V. Hizhnyakov
Institute of Physics, Estonian Academy of Sciences, Riia 142, Tartu, Estonia
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When doping the antiferromagnetically ordered high-T_c cuprates with holes magnetic polarons (spin-polarized clusters) are formed. We discuss two different mean-field approaches of the three-band Hubbard model to calculate the binding energy and polarization of the clusters. At higher hole concentrations and due to cluster diffusion a phase separation is formed by establishing large fractal or percolative clusters. This percolation picture allows us to understand experimentally obtained results for phase separation as well as magnetic and conductive phase diagrams.

I. INTRODUCTION

Doping or oxidation of the antiferromagnetically (AF) ordered perovskite compounds like La_2CuO_4 and $YBa_2Cu_3O_6$, which are the parent materials for high-T_c superconductors, leads to the creation of holes in the CuO_2 planes. The dynamics of the charge carriers is governed by strong correlation effects which leads to a redistribution of spin and charge densities in the vicinity of the excess holes. Due to these effects the antiferromagnetic order in the CuO_2 planes disappears quickly with doping and at higher doping concentrations superconductivity is established below the critical temperature.

A large number of experiments show that upon doping the high-T_c cuprates a separation into small charge-carrier enriched domains and carrier depleted regions takes place.[1-4] The results of these investigations give support to an electronic mechanism of the phase separation in contrast to a chemical phase separation where the excess oxygen ions should cluster in domains. The simultaneous observation of a diamagnetic signal below T_c and a Néel temperature of $T \approx 250$ K in $La_2CuO_{4+\delta}$ samples[1] strongly indicates a separation mechanism of percolative type as proposed in the model of percolative phase separation.[5] This model is based on the idea that doping of the CuO_2 planes with holes leads to the creation of small ferromagnetically ordered clusters (magnetic polarons, ferrons). These clusters only have low mobility whereas the holes inside the clusters can move freely. As a result, when increasing the hole concentration and, due to cluster diffusion, the clusters start to overlap and a (fractal) percolation network is built up. This leads to the destruction of the AF order and to the appearance of metallic-like conductivity, or below T_c to superconductivity, within the percolation network.[5] Such a mechanism allows us to explain the phase diagrams of $La_{2-x}Sr_xCuO_4$ and $YBa_2Cu_3O_{6+\delta}$ in quantitative agreement with experiment[6] giving support to spin-cluster formation in these materials. The spin-cluster model also allows us to explain recent experimental data on percolative phase separation in weakly doped $La_2CuO_{4+\delta}$ and $La_{2-x}Sr_xCuO_4$ samples.[1,7] Also, the magnetic susceptibility measurements[1] show the existence of small ferromagnetic (superparamagnetic) particles associated with holes. Recently, the existence of spin-polarized clusters in $La_2CuO_{4+\delta}$ has been demonstrated by electron paramagnetic resonance (EPR) experiments.[8] In addition, magnetic-resonance signals measured in $RBa_2Cu_3O_x$ (R = Y,Gd) have been attributed to clusters with magnetic ordering.[9] A review on experiments about phase separation is given in Ref. 10.

To relate the foregoing experimental results with a theoretical model it is necessary to investigate the electronic structure of the spin-cluster states. In the present paper we study the formation of spin-polarized clusters in the CuO_2 planes within two different mean-field approximations of the three-band Hubbard model.[11] First we follow the Hartree-Fock approach which neglects all fluctuations around the electronic mean field (MF) values. However, for large values of the Hubbard repulsion which are believed to be realized in the considered systems these fluctuations are small so that the Hartree-Fock approximation should yield reasonable results. The second approach is based on the slave-boson method in the saddle-point approximation[12] which corresponds to the Gutzwiler variational approximation. Regarding the metal-insulator transition (MIT) in the high-T_c compounds, previous calculations using the above-mentioned MF approximations of the three-band Hubbard Hamiltonian always led to a critical dopant concentration which was about ten times larger than the experimentally observed one.[13,14] The reason is that these calculations preserved the translational invariance of the system whereas in our model the MIT transition is based on the fractal percolation of ferromagnetic clusters. In this context it was recently shown within the scope of the slave-boson method that a magnetically disordered system drastically reduces the critical concentration of the MIT.[15]

We note that in the weak-coupling limit the concept of spin-polarized clusters leads to the formation of spin bags introduced by Schrieffer, Wen, and Zhang.[16] The transition between spin-bag and spin-polarized cluster behavior has already been studied numerically by Su and Chen[17] in a Hartree-Fock approximation of the one-band Hubbard model on a 8×8 lattice. They have found that for small values of U an additional hole causes a small depression of the staggered magnetization whereas above a critical value of the Hubbard repulsion a ferromagnetic core at the center of the polaron is formed. As shown in Ref. 5 the attraction of rather heavy spin-polarized clusters due to a pure electronic mechanism cannot lead to superconductivity with high-T_c values. On the other hand, high-T_c superconductivity may be provided by the attraction of highly mobile holes moving along the percolation net.[18]

This paper is organized as follows. After introducing the three-band Hubbard model we calculate the Green's functions in the Hartree-Fock approximation. It is shown that spin-polarized clusters are created upon doping whose polarization, binding energy, and effective mass are obtained by a Lifshitz formalism. In the last section we study the influence of fluctuations within the scope of the slave-boson method in the saddle-point approximation.

II. HOLE DYNAMICS IN CuO_2 PLANES

Our considerations are based on the three-band Hubbard model (see, e.g., Emery[11]) which takes the $d_{x^2-y^2}$ band of the Cu ions and the p_σ bands of the oxygen ions into account. The model Hamiltonian reads

$$H=\sum_\sigma H_{0\sigma}+H_{\rm int}\ , \tag{1}$$

where

$$H_{0\sigma}=\epsilon_d\sum_n n^d_{n\sigma}+\epsilon_p\sum_m n^p_{m\sigma}+T^{\pm}\sum_{nm}(d^\dagger_{n\sigma}p_{m\sigma}+\text{H.c.})\ , \tag{2}$$

$$H_{\rm int}=U\sum_n n^d_{n\sigma}n^d_{n-\sigma}\ . \tag{3}$$

$n^d_{n\sigma}$ and $n^p_{m\sigma}$ are the electronic occupation numbers of the $d_{x^2-y^2}$ and p_σ orbitals, respectively, and $d^\dagger(d)$ and $p^\dagger(p)$ are electronic creation (annihilation) operators in the corresponding Cu and O orbitals obeying the usual commutation rules for fermion operators. The spin index σ indicates the spin directions ($\sigma=\uparrow\downarrow$). Only the transfer between Cu and the four nearest oxygen ions is taken into account. The transfer integral $T^{\pm}$ changes sign due to the two possibilities of overlap. $H_{\rm int}$ describes the Coulomb repulsion for two electrons on the same Cu site with opposite spins. Band-structure calculations indicate the following parameter values: $U\approx8$ eV, $\epsilon=\epsilon_p-\epsilon_d\approx3$ eV, $T\approx1$ eV.

The effect of the strong particle-particle (Hubbard) repulsion is treated by two different approaches.

A. Hartree-Fock approach

In our first approach the calculation is based on a Hartree-Fock approximation of the original three-band Hubbard Hamiltonian which within this approximation reads as

$$H^{\rm MF}=\sum_\sigma H^{\rm MF}_{0\sigma}+H^{\rm MF}_{\rm int}\ , \tag{4}$$

where

$$H^{\rm MF}_{0\sigma}=\sum_n(\epsilon_d+U\langle n^d_{n-\sigma}\rangle)n^d_{n\sigma}+\epsilon_p\sum_m n^p_{m\sigma}+T^{\pm}\sum_{nm}(d^\dagger_{n\sigma}p_{m\sigma}+\text{H.c.})\ , \tag{5}$$

$$H^{\rm MF}_{\rm int}=-U\sum_n\langle n^d_{n\sigma}\rangle\langle n^d_{n-\sigma}\rangle\ . \tag{6}$$

In order to describe the AF ordered state we introduce two Cu sublattices (the magnetic unit cell contains two CuO_2 units). The components of the state vector corresponding to the two different Cu ions in a unit cell are denoted by d_1 and d_2. The corresponding diagonal elements of the Hamiltonian (4) read

$$\epsilon_{1\sigma}=\epsilon_d+U\langle n_{1-\sigma}\rangle\ , \tag{7}$$

$$\epsilon_{2\sigma}=\epsilon_d+U\langle n_{2-\sigma}\rangle\ . \tag{8}$$

From the symmetry of the AF state one immediately obtains the following relations:

$$\langle n_{1\uparrow}\rangle=\langle n_{2\downarrow}\rangle\ , \tag{9}$$

$$\langle n_{1\downarrow}\rangle=\langle n_{2\uparrow}\rangle\ . \tag{10}$$

Therefore, we can use the notations $\langle n_\uparrow\rangle$ and $\langle n_\downarrow\rangle$ and consider these quantities as modulated with twice the lattice period.

Upon transforming (5) into $\mathbf{k}$ space one finds that two oxygen states are decoupled from all other states while in the basis of the four remaining states the Hamiltonian (5) reads as

$$H_{\mathbf{kk}}=\begin{pmatrix}\epsilon_1 & 0 & -TV_k & -2T\sqrt{1-\frac{1}{4}V_k^2}\\ 0 & \epsilon_2 & 2T & 0\\ -TV_k & 2T & \epsilon_p & 0\\ -2T\sqrt{1-\frac{1}{4}V_k^2} & 0 & 0 & \epsilon_p\end{pmatrix}, \tag{11}$$

with $V_k=\cos k_x+\cos k_y$. The secular equation reads

$$\mathrm{Det}(E-H_{\mathbf{kk}})=D_1(E)D_2(E)-4T^4V_k^2\ , \tag{12}$$

where

$$D_i(E)=(E-\epsilon_p)(E-\epsilon_i)-4T^2\ . \tag{13}$$

Equation (12) describes four bands where in the case of CuO_2 only the lowest band is filled with holes. A peculiar property of the model is the appearance of nonanalytical critical points along the line $k_x+k_y=\pi$. These critical points lead to a divergent behavior in the density of states

of the type $(1/\sqrt{E})\ln(1/E)$, while normal logarithmic singularities disappear.

B. Green's functions

The diagonal elements of the one-particle Green's functions for the two Cu sites in the doubled unit cell are given by

$$G_{ii}=\sum_{\mathbf{k}}(E-H_{\mathbf{kk}})^{-1}_{ii}=\sum_{\mathbf{k}}\frac{|\{E-H_{\mathbf{kk}}\}|_{ii}}{\det|E-H_{\mathbf{kk}}|}\ , \tag{14}$$

where $|\{A\}|_{ii}$ denotes the adjoint subdeterminant of element (ii) of the matrix A. We define the imaginary part of the complex energy to be negative: $E\rightarrow E-i\epsilon$. Making use of Eqs. (9) and (10) we write the Green's function on Cu site 1 as $G_{11}=G_{\uparrow}$ and on Cu site 2 as $G_{22}=G_{\downarrow}$, respectively. After a straightforward calculation one can express G_{σ} in terms of the Green's function G^{2-D} of a simple quadratic lattice which yields

$$G_{\uparrow}=(E-\epsilon_p)\sqrt{(D_2/D_1)}G^{2-D}[\sqrt{D(E)}]\ , \tag{15}$$

$$G_{\downarrow}=(E-\epsilon_p)\sqrt{(D_1/D_2)}G^{2-D}[\sqrt{D(E)}]\ , \tag{16}$$

where $D(E)=D_1(E)D_2(E)$. G^{2-D} is derived in Ref. 20 and is given by a simple elliptic integral of the first kind K_1. For the Green's functions we finally get

$$G_{\uparrow}=2\frac{E-\epsilon_p}{\pi D_1}K_1\left[\frac{4T^2}{\sqrt{D(E)}}\right]\ , \tag{17}$$

$$G_{\downarrow}=2\frac{E-\epsilon_p}{\pi D_2}K_1\left[\frac{4T^2}{\sqrt{D(E)}}\right]\ , \tag{18}$$

where $D(E)/16T^4>1$. To obtain the spectral functions in the band and gap regions one has to use the analytical continuations of the elliptic integral (cf. Ref. 21).

In Fig. 1 we have plotted the real and imaginary part of $G_{\uparrow}$ and $G_{\downarrow}$ up to the second hole band. The expectation values $\langle n_{\uparrow}\rangle$ and $\langle n_{\downarrow}\rangle$ are immediately found from the self-consistent equations

$$\langle n_{\uparrow}\rangle=\frac{1}{\pi}\int dE\ \mathrm{Im}[G_{\uparrow}(E)]\ , \tag{19}$$

$$\langle n_{\downarrow}\rangle=\frac{1}{\pi}\int dE\ \mathrm{Im}[G_{\downarrow}(E)]\ , \tag{20}$$

where the integration is restricted to the lowest hole band. As can be seen from Fig. 2, for real values of parameters the ground state is almost completely polarized although the total Cu spin is considerably smaller due to the p-d hybridization. The ground-state properties of the totally AF ordered lattice have been intensively discussed in Ref. 13.

C. Doped system

With doping, two main scenarios have to be considered.

(1) The ideal AF order remains unchanged and the additional holes fill up the second band. However, since the Coulomb repulsion depends on the filling, doping leads to the decrease of the total spin at every site which results in a charge redistribution and particularly in the decreasing of the charge-transfer gap. This scenario ends up with a metal-insulator transition (MIT) at $\langle n_{\uparrow}\rangle=\langle n_{\downarrow}\rangle$. As found by Oleś and Zaanen,[13] such a MIT would occur at a hole concentration of $\delta\approx 0.24$.

(2) Due to the nonlinear nature of the mean-field equations one can expect (in analogy with, e.g., electron-

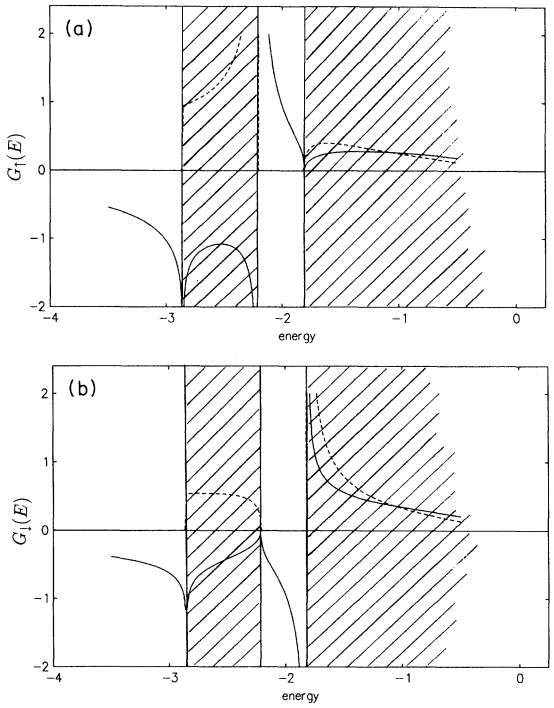

FIG. 1. Local Green's functions for (a) spin-up (b) spin-down projections of the wave function on Cu sites. The shaded areas indicate the lower Hubbard band and the lower part of the bonding oxygen band. Solid line: real part; dashed line: imaginary part of the Green's functions.

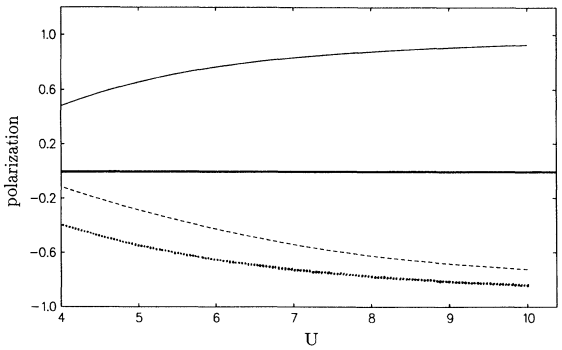

FIG. 2. Spin-polarization $\langle n_{\uparrow}\rangle-\langle n_{\downarrow}\rangle$ on Cu sites vs the Hubbard repulsion parameter U ($\epsilon=3, T=1$). Solid line: undoped case; dashed line: polarization on the perturbed Cu site (one turned spin, one additional hole); dotted line: polarization on the perturbed Cu sites (two turned spins, one additional hole).

lattice polaron theory) a spontaneous symmetry breaking, leading to the creation of localized excitations. The new states arising from such symmetry breaking should be determined self-consistently.

We have found that the second scenario leads to a lower ground-state energy, a result which was already obtained in an earlier work by use of unitary transformations.[5,6] In this section we will illustrate the calculation in the case of the perturbation of one Cu spin upon doping the system with one additional hole. The generalization to perturbations of more than one Cu site is straightforward (see, e.g., Ref. 20). In the following we assume that the Cu spin at site $\mathbf{m}=0$, which originally has a spin direction of, for example, $\uparrow$, can spontaneously fluctuate, changing the spin values to $\langle n_{\uparrow}^{l} \rangle$ and $\langle n_{\downarrow}^{l} \rangle$. These values are treated self-consistently. This problem is mathematically similar to the problem of magnetic impurities (e.g., the Wolff model) and can be treated within the standard localized perturbation theory. The perturbation of the Hamiltonian (5) is given by

$$V=V^{\uparrow}+V^{\downarrow} , \tag{21}$$

where

$$V^{\downarrow}_{mm^l}=U\delta_{m0}\delta_{m^l0}(\langle n_{\uparrow}^{l} \rangle-\langle n_{\uparrow} \rangle)d^{\dagger}_{0\downarrow}d_{0\downarrow} , \tag{22}$$

$$V^{\uparrow}_{mm^l}=U\delta_{m0}\delta_{m^l0}(\langle n_{\downarrow}^{l} \rangle-\langle n_{\downarrow} \rangle)d^{\dagger}_{0\uparrow}d_{0\uparrow} . \tag{23}$$

The new Green's functions for each spin direction can be found from Dyson's equation

$$G^{\sigma}(mm^l)=G_0^{\sigma}(mm^l)+G_0^{\sigma}(m0)V^{\sigma}G^{\sigma}(0m^l) , \tag{24}$$

where G_0 denotes the Green's function of the undisturbed lattice. The solution of (24) is

$$G^{\sigma}(mm^l)=G_0^{\sigma}(mm^l)+V^{\sigma}\frac{G_0^{\sigma}(m0)G_0^{\sigma}(0m^l)}{1-V^{\sigma}G_0^{\sigma}(00)} . \tag{25}$$

The equations determining the spin distribution at the disturbed lattice site then read as

$$\langle n_{\sigma}^{l} \rangle=\frac{1}{\pi}\int_{-\infty}^{E_F} dE\ \mathrm{Im}G^{\sigma}(00,E) , \tag{26}$$

where E_F is the chemical potential, depending on the number of additional holes. The localized states are the solution of the equations:

$$1=V^{\sigma}G_0^{\sigma}(E), \quad \sigma=\uparrow,\downarrow . \tag{27}$$

As can be concluded from the real part of $G(E)$ in Fig. 1, Eq. (27) has only solutions with $V^{\uparrow}>0$ and $V^{\downarrow}<0$ (beside the trivial solution $V^{\sigma}=0$). It is seen for any values of parameters that three localized states exist, one originating from the change of the $\langle n_{\uparrow} \rangle$ value and two due to the perturbation of $\langle n_{\downarrow} \rangle$. For one additional hole the Fermi level coincides with the upper localized level in the gap. Figure 2 shows the dependence of the polarization at site $\mathbf{m}=0$ on the repulsion parameter U in comparison with the polarization at the unperturbed sites. Obviously, there exists a reversed spin polarization the value of which is increasing with U. Thus we showed that the mean-field Hamiltonian (5) for a doped system has solutions with local fluctuations (mainly reversion) of Cu spins.

The total change of energy caused by a spin-flip process consists of two parts.

(1) Due to the perturbation (21) of the Hamiltonian (5) the energy change is given by

$$\Delta E^{\mathrm{loc}}=\sum_{\sigma}\int_{-\infty}^{E_F} dE\ E\ \Delta N_{\sigma}(E) . \tag{28}$$

$\Delta N_{\sigma}(E)$ is the change in the density of states. We have to consider two contributions to $\Delta N_{\sigma}(E)$. The first is the change of density in the bands mainly from the band edges; the second is the appearance of singularities in $\Delta N_{\sigma}(E)$ outside the bands due to the formation of localized levels. The expression for the change in the density of states is derived in Ref. 22 and reads

$$\Delta N_{\sigma}(E)=\frac{1}{\pi}\frac{d}{dE}\,\mathrm{Im}\ln[1-V^{\sigma}G_0^{\sigma}(E)] . \tag{29}$$

Evaluating (29) one finds that every local state gives a contribution equal to 1, besides the change of the local density in the bands. Inserting Eq. (29) into (28) immediately leads to

$$\Delta E^{\mathrm{loc}}=\sum_{l}E_l+\frac{1}{\pi}\sum_{\sigma}\int_{E_0}^{E_1}\arctan\frac{V^{\sigma}\mathrm{Im}G^{\sigma}(00)}{1-V^{\sigma}\mathrm{Re}G^{\sigma}(00)} , \tag{30}$$

where E_l denote the energies of the occupied localized states measured from the band edges.

(2) The energy change resulting from the perturbation of (6) reads as

$$\Delta E^{\mathrm{int}}=U(\langle n_{\uparrow} \rangle\langle n_{\downarrow} \rangle-\langle n_{\uparrow}^{l} \rangle\langle n_{\downarrow}^{l} \rangle) . \tag{31}$$

D. Results

In Fig. 3 we have plotted the binding energy of one additional hole in a CuO_2 lattice with one flipped spin $-(\Delta E^{\mathrm{int}}+\Delta E^{\mathrm{loc}})$ versus the repulsion parameter U for a value of $\epsilon=\epsilon_p-\epsilon_d=3$ eV. The behavior in the large U limit coincides with the results obtained in an earlier

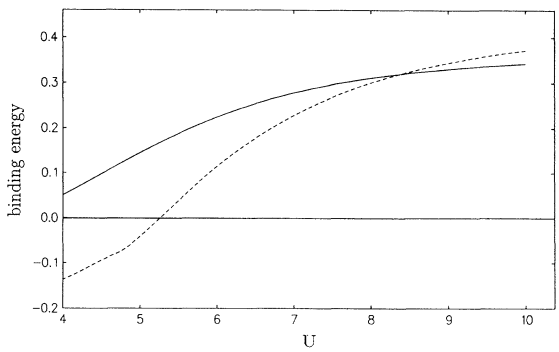

FIG. 3. Binding energy for one additional hole in dependence of the Hubbard repulsion parameter U ($\epsilon=3, T=1$). Solid line: one turned spin; dashed line: two turned spins.

work by use of unitary transformations.[6] In fact, for large U the mean-field ground state is described by a well-polarized lattice so that fluctuations around the expectation values can be neglected. Thus in antiferromagnetically ordered lattices a MF calculation not only approaches the correct ground-state energy in the small U limit, but also in the strongly correlated case $U \rightarrow \infty$.[23] The wave function of the additional hole is extremely localized in space and has a very small amplitude on next-nearest four Cu atoms only. However, one should in principle (at least for smaller values of U) take a larger area in the perturbation (21) into account. Calculations which incorporated the next-nearest four Cu spins have been done and gave a small increase of the polarization and binding energy.[24]

Figure 4 shows the one-particle spectrum of a spin-down polarized cluster. Only the two lowest bands are plotted. There are two localized states appearing in the charge-transfer (CT) gap. The upper one belongs to the spin-up spectrum and splits from the lower band edge of the first oxygen band whereas the other stems from the lower Hubbard band and belongs to the spin-down spectrum. It is the latter level which carries most of the amplitude of the additional hole resembling the fact that the polarization of the corresponding Cu ion has changed its sign. There is one additional level appearing about 10^{-2} eV below the lower Hubbard band whose amplitude has a maximum at the four surrounding Cu ions of the polaron center. This level is the consequence of our limitation of the perturbation to one single site. However, as long as its contribution to binding energy and polarization is small this approximation is justified.

Up to now we have only considered the symmetry breaking of the ground state by one flipped spin. Nevertheless, for a critical value of U the situation with two turned spins (diagonally neighbored) gives a larger binding energy as can be seen in Fig. 3. This behavior is in agreement with calculations of Auerbach and Larson[19] for the t-J model. In the mean-field picture two turned spins maximally produce four localized states in the CT gap and two below the first hole band [Fig. (4)]. These levels can be filled up with one or two holes, respectively. The results are summarized in Table I. Obviously the polarization of the perturbed spins is much larger in the case of two turned spins and one additional hole than in the case of one turned spin only. The reason is the same as discussed before: enlarging the perturbation leads to an increase of the polarization. Doping a system with

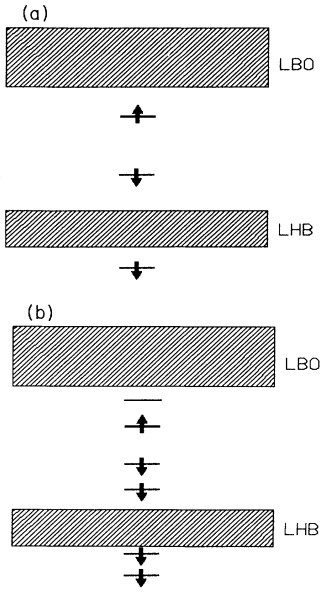

FIG. 4. Sketch of the band scheme for one additional hole in the case of one (a) and two (b) turned spins which originally had mainly spin-up mean values. The additional hole occupies a localized level of the spin-up subsystem with mainly oxygen character. LHB: lower Hubbard band; LBO: lower part of the bonding oxygen band.

two holes and additionally turning two Cu spins leads to a polarization of the perturbed spins comparable to the situation of one flipped spin and one hole. The energy of this two-turned-spin cluster with two holes is at least for larger U the same as for two separated clusters with one turned spin and one hole. In the limit $U \rightarrow \infty$ and one additional hole the ground state should be totally ferromagnetically ordered according to Nagaoka's theorem.[25]

So far we have confined ourselves to a symmetry-breaking solution of the translational invariant three-band Hubbard Hamiltonian. However, as in the theory of strong-coupling polarons (Pekar-type solutions) one should in principle perform a Bloch-superposition of these wave functions to guarantee the translational invariance of the solution. This ends up with a dispersion relation for the quasiparticles, i.e., in our case the spin-polarized clusters. To keep the calculation tractable we

TABLE I. The binding energy and the Cu-spin polarization for the undoped $\langle n_\uparrow \rangle$, $\langle n_\downarrow \rangle$ and doped $\langle n_\uparrow^l \rangle$, $\langle n_\downarrow^l \rangle$ cases with one and two turned spins in dependence of U and ϵ (Hartree-Fock approach).

				1 turned spin, 1 hole			2 turned spins, 1 hole			2 turned spins, 2 holes		
U/T	ϵ/T	$\langle n_\uparrow^0 \rangle$	$\langle n_\downarrow^0 \rangle$	$\langle n_\uparrow^l \rangle$	$\langle n_\downarrow^l \rangle$	$-\Delta E$	$\langle n_\uparrow^l \rangle$	$\langle n_\downarrow^l \rangle$	$-\Delta E$	$\langle n_\uparrow^l \rangle$	$\langle n_\downarrow^l \rangle$	$-\Delta E$
4	2	0.66	$3E$-2	0.31	0.57	0.12	0.12	0.65	0.04	0.32	0.55	0.38
6	2	0.84	$5E$-3	0.29	0.76	0.30	0.11	0.82	0.24	0.29	0.75	0.63
8	2	0.91	$2E$-3	0.25	0.86	0.37	0.11	0.89	0.37	0.26	0.86	0.74
6	3	0.77	$8E$-3	0.25	0.68	0.22	0.10	0.75	0.11	0.25	0.67	0.51
8	3	0.88	$2E$-3	0.21	0.83	0.31	0.09	0.86	0.30	0.21	0.83	0.64
10	3	0.93	$8E$-4	0.18	0.90	0.34	0.08	0.92	0.37	0.18	0.90	0.68

have confined ourselves to hopping processes of a magnetic polaron with one turned spin to nearest and next-nearest neighbors. The dispersion relation of the polaron is strongly anisotropic in k space and yields the maximum of the effective mass in the $\langle 11 \rangle$ direction. For $U=8T$ and $\epsilon=3T$ we obtain a value of 10 bare masses.

III. SLAVE-BOSON APPROACH

In the previous section we have used a self-consistent Hartree-Fock mean-field approach to determine the polarization and binding energy of the spin-polarized clusters. However, the renormalization of the bandwidths due to the strong correlation effects in the considered systems is not taken into account by this approach. This effect can be incorporated by using the slave-boson method in the saddle-point approximation, introduced by Kotliar and Ruckenstein,[12] which is similar to the Gutzwiler variational approach.

We enlarge the original Hilbert space by introducing four boson fields for every copper site, respectively. $e_i^\dagger$ creates an empty state, $s_{i,\uparrow}$ and $s_{i,\downarrow}$ are singly occupied states with spin up and down, and finally $d_i^\dagger$ stands for the creation of a double occupied state. Since there are only four possible states per Cu site, the unphysical states are eliminated by the following conditions:

$$e_i^\dagger e_i+\sum_\sigma s_{i,\sigma}^\dagger s_{i,\sigma}+d_i^\dagger d_i=1 , \tag{32}$$

$$c_{i,\sigma}^{d\dagger}c_{i,\sigma}^{d}-s_{i,\sigma}^\dagger s_{i,\sigma}-d_i^\dagger d_i=0 , \tag{33}$$

where $c_{i,\sigma}^{d\dagger}$ stands for the creation of a hole with spin σ on the Cu site i.

In the saddle-point approximation the constraints (32) and (33) are only satisfied on the average by the Lagrange parameters λ_σ and κ, respectively.

In analogy to the previous section we double the unit cell by introducing two operators $c_{n,\sigma}^{1d}$ and $c_{n,\sigma}^{2d}$ which create holes with spin σ on the neighboring Cu sites of the corresponding unit cell n. The symmetry of the AF ordered ground state provides the following relations:

$$\lambda_\uparrow^1=\lambda_\downarrow^2 , \tag{34}$$

$$\langle n_{1,\sigma}\rangle=\langle n_{2,-\sigma}\rangle . \tag{35}$$

This ends up with the Gutzwiler variational expression for the total energy

$$NE=\sum_\sigma \langle H_\sigma^{\rm eff}\rangle+UNd^2-N[\lambda_\uparrow\langle n_\uparrow\rangle+\lambda_\downarrow\langle n_\downarrow\rangle] , \tag{36}$$

$$\begin{aligned} H_\sigma^{\rm eff}=&\sum_n \epsilon_1 c_{n,\sigma}^{\dagger 1d}c_{n,\sigma}^{1d}+\sum_n \epsilon_2 c_{n,\sigma}^{\dagger 2d}c_{n,\sigma}^{2d}+\sum_m \epsilon_p c_{n,\sigma}^{\dagger p}c_{n,\sigma}^{p} \\ &+T_\sigma\sum_{n,m}\left[c_{n,\sigma}^{\dagger 1d}c_{m,\sigma}^{p}+\text{H.c.}\right] \\ &+T_{-\sigma}\sum_{n,m}\left[c_{n,\sigma}^{\dagger 2d}c_{m,\sigma}^{p}+\text{H.c.}\right] , \end{aligned} \tag{37}$$

with

$$\epsilon_1=\epsilon^d+\lambda_\uparrow , \tag{38}$$

$$\epsilon_2=\epsilon^d+\lambda_\downarrow , \tag{39}$$

$$T_\sigma=T\frac{\sqrt{(1-\langle n^d\rangle+d^2)(\langle n_\sigma^d\rangle-d^2)}+d\sqrt{\langle n^d\rangle-\langle n_\sigma^d\rangle-d^2}}{\sqrt{\langle n_\sigma^d\rangle(1-\langle n_\sigma^d\rangle)}} . \tag{40}$$

The summation in the transfer terms is restricted to nearest neighbors. The diagonalization of (37) can now be performed in the same way as in the first section. The Green's functions keep the same analytical form as (17) and (18), where additionally the substitution

$$T^2\rightarrow T_\uparrow T_\downarrow \tag{41}$$

has to be made.

A. Doped system

We will add now an additional hole to the system and show that a spin-flip process at site, e.g., 0 will lead to a lower ground-state energy. Such a spin reversion affects the Lagrange parameters $\lambda_{0,\sigma}$, the double occupancy parameter d_0^2, and additionally the transfer to the next-nearest oxygen atoms. Introducing symmetric combinations of these oxygen states the problem reduces to a 2×2 perturbation matrix for each spin direction

$$V_\sigma=\begin{bmatrix} V_\sigma^\lambda & V_\sigma^T \\ V_\sigma^T & 0 \end{bmatrix} \tag{42}$$

in the basis of $|c_{0,\sigma}\rangle$ and $|q\rangle=\frac{1}{2}\sum_{i=1}^4|p_i\rangle$ where the summation is restricted to the four next-nearest oxygen ions of site 0. The perturbations are denoted by

$$V_\sigma^\lambda=\lambda_\sigma^l-\lambda_\sigma , \tag{43}$$

$$V_\sigma^T=2(T_\sigma^l-T_\sigma) . \tag{44}$$

Following again the Green's-functions formalism we can calculate the disturbed Green's functions at the perturbed Cu site from Eq. (24):

$$G_{00}^\sigma=\frac{G_{00}^{0,\sigma}}{D_\sigma(E)} , \tag{45}$$

$$\begin{aligned} D_\sigma(E)=&1+\frac{V_\sigma^T}{T_\sigma}\left[1+\frac{V_\sigma^T}{4T_\sigma}\right] \\ &-G_{00}^{0,\sigma}\left[V_\sigma^\lambda+\frac{V_\sigma^T}{T_\sigma}(E-\epsilon_\sigma)\left[1+\frac{V_\sigma^T}{4T_\sigma}\right]\right] , \end{aligned} \tag{46}$$

Then we have to minimize the change of energy due to the three parameters $\lambda_{0,\uparrow}$, $\lambda_{0,\downarrow}$, and d_0^2. For every param-

TABLE II. The binding energy and the Cu-spin polarization for the undoped $\langle n_\uparrow \rangle$, $\langle n_\downarrow \rangle$ and doped $\langle n_\uparrow^l \rangle$, $\langle n_\downarrow^l \rangle$ cases with one turned spin in dependence of U and ϵ (slave-boson approach).

				1 turned spin, 1 hole		
U/T	ϵ/T	$\langle n_\uparrow^0 \rangle$	$\langle n_\downarrow^0 \rangle$	$\langle n_\uparrow^l \rangle$	$\langle n_\downarrow^l \rangle$	$-\Delta E$
6	2	0.83	$8E$-3	0.34	0.66	0.28
7	2	0.88	$4E$-3	0.35	0.71	0.36
8	2	0.91	$2E$-3	0.35	0.73	0.41
6	3	0.76	$2E$-2	0.35	0.55	0.23
7	3	0.84	$6E$-3	0.34	0.64	0.30
8	3	0.88	$2E$-3	0.34	0.68	0.37

eter set we have to additionally calculate the new Cu polarization values using Eq. (26). As in the first calculation we obtain two localized levels in the CT gap corresponding to the zeros of $D_t(E)$, one arising from the lower Hubbard band whereas the other comes from the first oxygen-type band. Unlike in the first approach these two levels are now both located in the upper region of the CT gap and only separated by ≈ 0.2 eV. The level below the lower Hubbard band is localized very near the band edge and disappears for larger values of U.

Table II shows the results obtained for various values of U and $\epsilon = E_p - E_d$. As one can readily see, the incorporation of fluctuations within the scope of a slave-boson approximation reduces the polarization of the spin-cluster state to 10–20 % in comparison to the Hartree-Fock approach. On the other hand, we now obtain a larger value of the binding energy. The remark in the previous section about an enlarged perturbation in space holds also here. The polarization of the spin-flipped state will increase if we would incorporate the next-nearest four Cu ions in the perturbation. However, the aim of the slave-boson calculation was to show that in the presence of correlation effects the spin-polarized state still persists. As already remarked by Olés and Zaanen[13] the MF and Gutzwiler approach yield the same results in the strong-coupling limit when charge fluctuations can be neglected. In the intermediate U regime, which is interesting for real systems, the slave-boson approach in the MF approximation leads to a much better description of the ground state.

IV. CONCLUSION

We have shown that in the Hartree-Fock and slave-boson MF approximations of the three-band Hubbard model doping with holes leads to the creation of ferromagnetically ordered clusters. This process is more pronounced when taking the influence of dopant ions (e.g., Sr, Ba, O, etc.) into account. The occurring attractive interaction between these charge compensators and the holes, in addition, supports the hole localization. The size of the clusters depends on the Coulomb repulsion on the Cu sites. Doping of the CuO_2 planes leads to a electronic phase separation of the system into hole-rich areas (fractally connected spin clusters) and hole-free AF ordered regions. The metal-insulator transition (MIT) is provided by the percolation of single-hole clusters. The percolation picture also explains the coexistence of long-range AF order and bulk superconductivity as shown with nuclear magnetic resonance (NMR) (Ref. 26) and neutron-scattering experiments.[27] Measurements which show a strong dependence of the superconducting fraction of $La_2CuO_{4+\delta}$ samples on the thermal treatment[1] clearly support the formation of spin-polarized clusters. When quenching the samples to temperatures lower than about 200 K one can "freeze in" the cluster diffusion, the formation of larger percolative structures is suppressed, and, consequently, a further slowly cooling below T_c leads to a very small diamagnetic fraction of the sample. In contrast, when the initial quenching ends up at temperatures higher than 200 K the diamagnetic fraction becomes maximal. For completeness we mention that recently the existence of spin-polarized clusters was directly observed by EPR[8] and magnetic resonance experiments.[9]

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