Nature of the insulating state in the three-band Hubbard model: A tight-binding approach

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We study the three-band Hubbard model, commonly used to describe the copper-oxygen planes of high-T_c superconductors, from the point of view of electronic properties. Utilizing perturbation theory around the atomic limit to evaluate finite-temperature Green's functions, one-particle densities of states are calculated in some simple approximations. Both metallic paramagnetic and insulating antiferromagnetic solutions are obtained, depending on the hole concentration. We discuss the overall scenario obtained here in comparison with experimental results and other theoretical approaches.

I. INTRODUCTION

A great deal of theoretical work has been devoted to studying electronic properties of the CuO_2 planes that are a common feature of high-T_c superconducting materials. It is accepted that the relevant orbitals are $3d_{x^2-y^2}$ for copper and $2p_x$ or $2p_y$ for oxygen. Based on this, three-band models[1] have been proposed that vary in generality depending on the choice of parameters such as the on-site Coulomb interactions, the charge transfer (CT) gap between copper and oxygen levels, the hopping matrices, and a nonlocal Coulomb interaction.

It is still a matter of controversy whether or not the low energy excitations of such multi-band models can be reproduced by an effective one-band Hubbard Hamiltonian.[2] Also, the role of charge and spin degrees of freedom in determining the nature of the ground state has yet to be clarified. Starting from the atomic limit, with a Coulomb interaction at copper sites much larger than the bare CT gap, it is obvious that the ground state for one hole per CuO_2 cluster has this hole occupying the copper site, while all oxygen orbitals are empty. The lowest lying excitations are clearly of charge transfer nature. Corrections to second order in the copper-oxygen hybridization[3] yield an antiferromagnetic (AF) insulating ground state, with the hole still at the copper site, and the oxygen orbitals providing a superexchange mechanism for the AF interaction. On the other hand, band structure calculations[4] and Hartree-Fock treatment of the Coulomb interaction[5,6] show the hybridization leading to formation of bandlike (extended) states. In this case, the nature of the AF state also changes towards a bandlike character, and the AF gap no longer coincides with the CT gap as in the localized description.

In order to further investigate this controversial point, i.e., the nature of the insulating AF state in the three-band Hubbard model, we utilize a tight-binding approach that, although starting from the atomic limit, is able to obtain the exact band structure in the uncorrelated limit. We treat the hopping (hybridization) as a perturbation, employing a regular many-body perturbative expansion with a diagrammatic representation to calculate one-particle Green's functions. From these, the density of states is obtained and analyzed with respect to the presence and position of gaps, spin polarization, and degree of p-d admixture as the total number of holes in the system is varied. In this paper, we remain in the simplest approximation scheme, closely related to the Hubbard I decoupling of the equations of motion for Green's functions in the single-band Hubbard model.[7]

Our results indicate that there is an important transfer of spectral weight from the almost empty (and, thus, weakly correlated) p band to the lowest lying band, which was originally of pure d character. This can yield a metallic ground state if a paramagnetic solution is imposed. However, an insulating AF solution is obtained close to the stoichiometric concentration, with a gap opening inside the lowest lying band. We will discuss in some detail the relation between these results and those obtained by the Hartree-Fock approximation,[5] slave-boson theory,[8] and the local moment approach,[3] as well as a possible relationship with spectroscopic measurements.[9]

The paper is organized as follows. In Sec. II we introduce the model Hamiltonian and the relevant Green's functions, describing the perturbation method. In Sec. III we describe the approximation we will adopt, and calculate the Green's functions and densities of states for the paramagnetic case. The antiferromagnetic solution is presented in Sec. IV. Our main results are discussed in Sec. V.

II. THE MODEL AND PERTURBATION APPROACH

The usual three-band Hubbard model[1] for the CuO_2 planes of high-T_c superconductors is described by the Hamiltonian

$$H = (\varepsilon_d - \mu)\sum_{i\sigma} d^\dagger_{i\sigma} d_{i\sigma} + U_d \sum_i n^d_{i\uparrow} n^d_{i\downarrow} + (\varepsilon_p - \mu)\sum_{j\sigma} p^\dagger_{j\sigma} p_{j\sigma} + U_p \sum_j n^p_{j\uparrow} n^p_{j\downarrow} - t \sum_{\langle ij\rangle\sigma} \left(d^\dagger_{i\sigma} p_{j\sigma} + \text{H.c.} \right) , \quad (1)$$

where ε_d (ε_p) is the atomic energy of copper (oxygen) sites, μ is the chemical potential, U_d (U_p) is the on-site Coulomb repulsion between two holes in the same copper (oxygen) site, and t denotes the hopping matrix between neighboring copper and oxygen sites. We are working in the hole representation. Thus $d^\dagger_{i\sigma}$ ($p^\dagger_{j\sigma}$) creates a hole with spin σ at the corresponding copper (oxygen) site. It is important to keep track of the kind of oxygen orbital (p_x or p_y) that corresponds to a given site. We do not distinguish between them in the Hamiltonian for economy of notation. We will not consider here the presence of a nonlocal Coulomb interaction between holes in nearest neighbor atoms as well as a direct hybridization between oxygen orbitals.

The relevant temperature (Matsubara) Green's functions are, for example,

$$G^{dd}_{ii'\sigma}(\tau) = -\left\langle \hat{T}\, d_{i\sigma}(\tau) d^\dagger_{i'\sigma}(0) \right\rangle , \quad (2)$$

where the angular brackets indicate ensemble average, and $\hat{T}$ is the time-ordering operator, here referring to the imaginary time τ, which is defined in the interval $[-\beta, \beta]$, with β representing the inverse of the temperature. There are definitions similar to Eq. (2) for $G^{dp}_{ij\sigma}$, $G^{pp}_{jj'\sigma}$, etc.

Applying the usual formalism of perturbation theory, we consider the local part of H [see Eq. (1)] as the unperturbed Hamiltonian H_0, and the hopping term is taken as the perturbation H_1. Equation (2) then becomes

$$G^{dd}_{ii'\sigma}(\tau) = -\frac{\left\langle \hat{T}\, d_{i\sigma}(\tau) d^\dagger_{i'\sigma}(0) S(\beta) \right\rangle_0}{\langle S(\beta)\rangle_0} , \quad (3)$$

with

$$S(\beta) = \sum_{n=0}^{\infty} \frac{(-1)^n}{n!} \int_0^\beta d\tau_1 \cdots \int_0^\beta d\tau_n \hat{T}\, H_1(\tau_1) \cdots H_1(\tau_n) . \quad (4)$$

The subscript zero on the angular brackets indicates that the averages are taken with respect to the unperturbed Hamiltonian. In Eqs. (3) and (4) the "time" dependence of the operators is also given by H_0, in contrast with Eq. (2).

The density of states is obtained through

$$\rho^{dd}_\sigma(\omega) = -\frac{1}{\pi} \text{Im}\, G^{dd\,+}_{ii\sigma}(\omega) , \quad (5)$$

where

$$G^{dd\,+}_{ii\sigma}(\omega) = G^{dd}_{ii\sigma}(\omega_n)\big|_{\omega_n \to \omega + i0^+} \quad (6)$$

is the Fourier transform of the retarded (real-time) Green's function. Similar relations hold for $\rho^{pp}_\sigma(\omega)$, in which case one has to sum the contributions from p_x and p_y orbitals.

III. PARAMAGNETIC CASE

In order to calculate Green's functions through Eq. (3) we utilize a diagrammatic representation for the perturbation series, similar to the one that has been presented by Metzner[10] for the one-band Hubbard model. In the present case, all possible local Green's functions may appear at each vertex. Selecting only chainlike diagrams, which can be summed up by means of a Dyson's equation, we obtain the simplest approximation, in which the diagonal Green's functions, for the paramagnetic case, assume the form

$$G^{dd}_{\mathbf{k}\sigma}(\omega_n) = \frac{g^d_\sigma(\omega_n)}{1 - g^d_\sigma(\omega_n) g^p_\sigma(\omega_n) \gamma_{\mathbf{k}}} \quad (7a)$$

and

$$G^{pp}_{\mathbf{k}\sigma}(\omega_n) = g^p_\sigma(\omega_n) + \frac{g^p_\sigma(\omega_n)}{1 - g^d_\sigma(\omega_n) g^p_\sigma(\omega_n) \gamma_{\mathbf{k}}} , \quad (7b)$$

where

$$\gamma_{\mathbf{k}} = 4t^2 \left(\cos^2 \frac{k_x a}{2} + \cos^2 \frac{k_y a}{2} \right) , \quad (8)$$

a being the lattice parameter. The lower case g's that appear in the above equations stand for the zeroth order (atomic) Green's functions, which read

$$g^\alpha_\sigma(\omega_n) = \frac{1 - \langle n^\alpha_{\bar\sigma}\rangle}{i\omega_n - \varepsilon_\alpha + \mu} + \frac{\langle n^\alpha_{\bar\sigma}\rangle}{i\omega_n - \varepsilon_\alpha - U_\alpha + \mu} , \quad (9)$$

where $\bar\sigma \equiv -\sigma$ and $\alpha = d, p$. The average number of particles in both d and p atomic orbitals is determined, together with the chemical potential, through the self-consistency relation

$$\langle n^\alpha_\sigma \rangle = \lim_{\tau \to 0^+} \frac{1}{\beta} \sum_{\omega_n} e^{i\omega_n \tau} \frac{1}{N} \sum_{\mathbf{k}\sigma} G^{\alpha\alpha}_{\mathbf{k}\sigma}(\omega_n) . \quad (10)$$

In this section we consider only the paramagnetic case. We thus set $\langle n^\alpha_{\bar\sigma}\rangle = \langle n^\alpha_\sigma \rangle$, and full translation invariance has been used in deriving Eqs. (7a) and (7b).

The *chain approximation* introduced above is equivalent to the so called Hubbard I decoupling scheme when the equations of motion for the Green's functions are utilized.[7] This approximation becomes exact in the noninteracting limit $U_d = U_p = 0$. The one-particle density of states for this case is shown in Fig. 1. There, the positions of the original atomic d and p levels are shown by arrows. We can see that the hybridization gives rise to three bands (bonding, nonbonding, and antibonding), in agreement with band structure calculations.[4] The charge transfer gap Δ is not renormalized with respect to the atomic value $\Delta = \varepsilon_p - \varepsilon_d$. The nonbonding band, of zero width and p character, is due to the combination of p_x

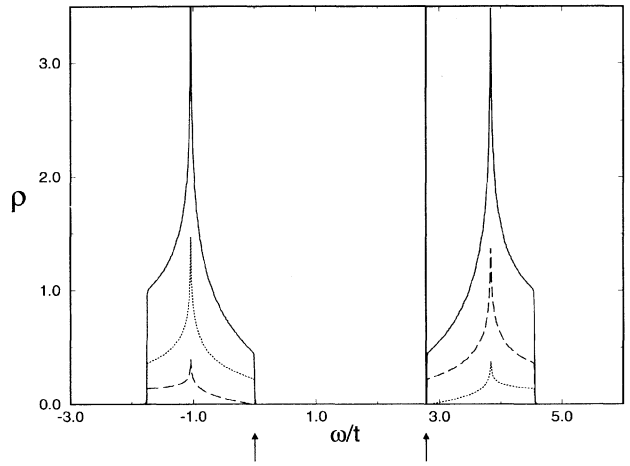

FIG. 1. Density of states in the noninteracting limit for the CuO_2 structure (with $\Delta/t = 2.8$). We also show separately the contributions from d (dotted) and p (dashed) levels. The arrows indicate the positions of the atomic levels.

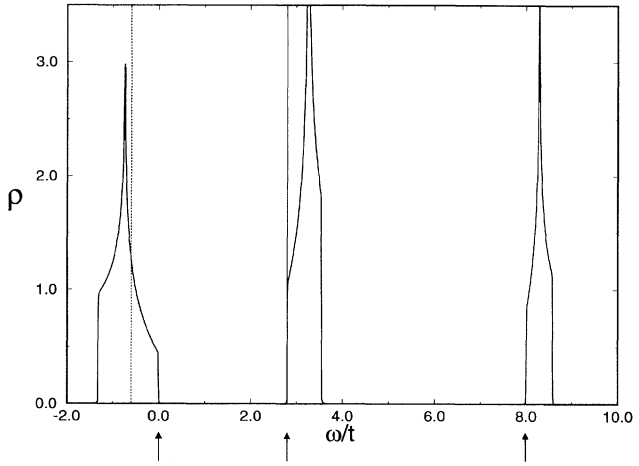

FIG. 3. Same as in Fig. 2, except that now $U_p = 0$.

and p_y orbitals that do not hybridize with the $d_{x^2-y^2}$ orbital. This band is related to the isolated g_σ^p that appears in Eq. (7b).

When Coulomb interactions are taken into account, the bands are split, similar to what occurs in a strongly correlated single-band system. The positions of the new subbands depend on the relative values of the energy parameters. Their widths depend also on the overall filling factor, i.e., the total number of particles in the system. We have chosen our parameters such that $\Delta/t = 2.8$, $U_d/t = 8$, and $U_p/t = 2.4$. These values lie in the range generally considered as consistent with experimental observations.[11] The resulting density of states is shown in Fig. 2 for the stoichiometric situation (one hole per unit cell).[12] In the absence of hybridization one would have only δ functions in the positions indicated by arrows in the figure. The outer ones would have purely d character while the inner ones would be purely of p character. In this case, one would have the leftmost "band" completely filled (one hole per copper atom) while all the others would be empty. With the hybridization included, we can see from Fig. 2 that the chemical potential (dotted line) falls within the lowest lying band. The other bands are still empty. However, the occupied band is not completely filled. There has been a transfer of spectral weight from the upper subbands to the lowest one.[13] This is due to the fact that the p levels in the atomic limit would be empty, which strongly diminishes correlation effects on oxygen sites. This can be seen by the narrowness of the band that lies close to $\varepsilon_p + U_p$, while the rest of the density of state (DOS) does not differ significantly with respect to the case $U_p = 0$, shown in Fig. 3. This weakly correlated nature of the p orbitals yields a hybrid band whose character is intermediate between the strong correlation limit (one particle per subband) and the uncorrelated limit (two particles per band). Only for a strongly hole doped system ($n \sim 1.27$) do we find a CT insulator, with the chemical potential lying inside the first gap.

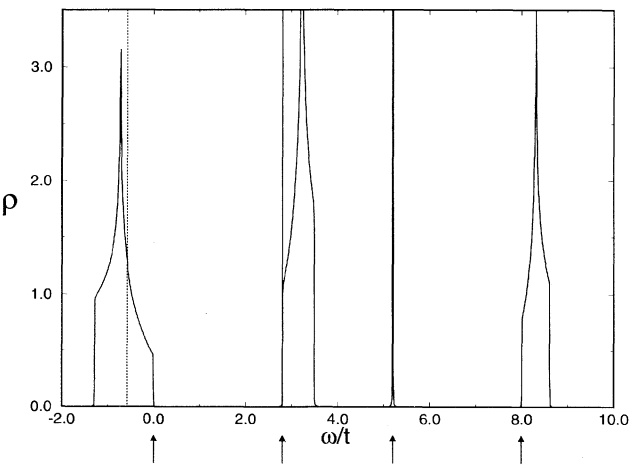

FIG. 2. Density of states in the chain approximation for the paramagnetic case and zero doping ($n = 1$). The arrows indicate the positions of the atomic levels, and the dotted vertical line shows where the chemical potential is. The values of the parameters are $\Delta = 2.8$, $U_d = 8$, and $U_p = 2.4$, in units of t.

This picture is very different from what we would expect on the basis of experimental results. However, we must look for magnetic solutions before we can compare our results with experiment.

IV. ANTIFERROMAGNETIC SOLUTION

In order to look for an antiferromagnetic solution we divide the lattice in two sublattices, A and B, such that copper atoms nearest to each other belong to different sublattices. Next we impose the antiferromagnetic condition, i.e., that the average number of holes in a given site of sublattice A with a given spin orientation is equal to the corresponding average for sublattice B with *oppo-*

site spin. We also consider that there will be no moment formation on oxygen sites, imposing the average occupation of these sites for both spin orientations to be the same. With these conditions, all equations of the chain approximation can be written in terms of quantities related to only one of the copper sublattices.

The Green's functions are again obtained by summation of all chainlike diagrams. In contrast to the paramagnetic case, these functions are now spin dependent. There are two copper sites (A and B) and four oxygen sites per unit cell. It is convenient to perform a partial renormalization that takes into account all possible hybridization processes between a copper site and its neighboring oxygens. These partially renormalized local Green's functions are

$$G^A_\sigma(\omega_n) = G^B_{\bar\sigma}(\omega_n) = \frac{g^A_\sigma(\omega_n)}{1 - g^A_\sigma(\omega_n)\ 4t^2_{dp}\ g^p_\sigma(\omega_n)} \ . \quad (11)$$

With this, the total Green's functions are given by

$$G^A_{\mathbf{k}\sigma}(\omega_n) = \frac{G^A_\sigma(\omega_n)}{1 - G^A_\sigma(\omega_n) t_{\mathbf{k}} g^p_\sigma(\omega_n) G^A_{\bar\sigma}(\omega_n) t_{\mathbf{k}}\ g^p_\sigma(\omega_n)} \quad (12)$$

for copper sites belonging to sublattice A, and

$$G^{pp}_{\mathbf{k}\sigma}(\omega_n) = 2g^p_\sigma(\omega_n) + \frac{2g^p_\sigma(\omega_n) + 4t^2_{dp}\ g^p_\sigma(\omega_n)\ [G^A_\sigma(\omega_n) + G^A_{\bar\sigma}(\omega_n)]\ g^p_\sigma(\omega_n)}{1 - G^A_\sigma(\omega_n)\ t_{\mathbf{k}}\ g^p_\sigma(\omega_n)\ G^r_{A\bar\sigma}(\omega_n)\ t_{\mathbf{k}}\ g^p_\sigma(\omega_n)} \ , \quad (13)$$

which includes the contributions of all four oxygens in the unit cell. Here we have defined

$$t_{\mathbf{k}} = 4t^2 \cos\frac{k_x a}{2} \cos\frac{k_y a}{2} \ . \quad (14)$$

The self-consistency process [see Eq. (10)] involves adjusting $\langle n^d_\uparrow\rangle$, $\langle n^d_\downarrow\rangle$, and $\langle n^p\rangle \equiv 2\langle n^p_\sigma\rangle$, together with the chemical potential μ for a given total number of holes $n \equiv \langle n^d_\uparrow\rangle + \langle n^d_\downarrow\rangle + 2\langle n^p\rangle$. We reproduce the paramagnetic solution obtained before if we impose the equality $\langle n^d_\downarrow\rangle = \langle n^d_\uparrow\rangle$ at the start of the self-consistency process. However, depending on the total number of holes, this solution may be unstable, in the sense that an arbitrarily small difference between the two initial occupation numbers for different spin orientations will lead to a magnetic solution ($\langle n^d_\downarrow\rangle \neq \langle n^d_\uparrow\rangle$) when self-consistency is achieved. In this case, a gap opens in the density of states around the chemical potential as well as in the other subbands, as shown in Fig. 4. The spin polarization is evidenced in the detailed plot of the low energy region shown in Fig. 5.

In Fig. 6 we show the variation of the sublattice magnetization $m = \langle n^d_\uparrow\rangle - \langle n^d_\downarrow\rangle$ as a function of the total number of holes. We can see that the stability region of the AF solution lies asymmetrically around the stoichiometric condition $n = 1$, in (at least) qualitative agreement

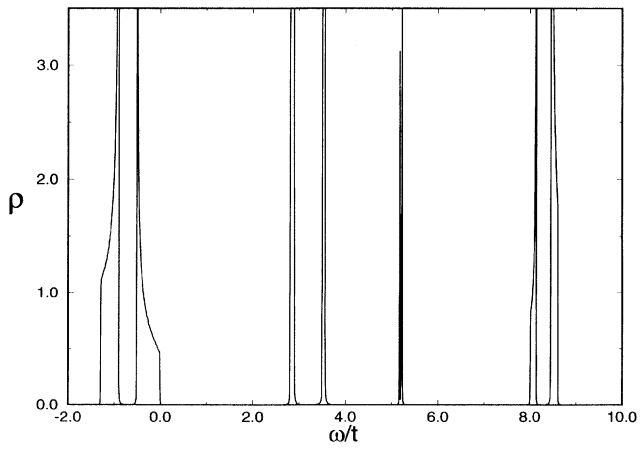

FIG. 4. Density of states in the chain approximation for the antiferromagnetic case and n =1, with the same parameters as in Fig. 2.

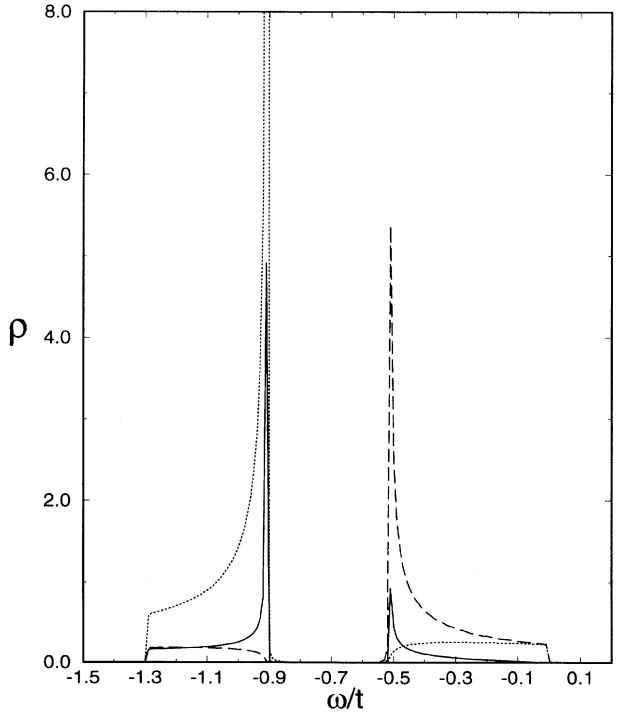

FIG. 5. Low energy part of the DOS in the AF state, showing separately the contributions from d levels with up (dotted) and down (dashed) spins, and from the p levels (continuous line).

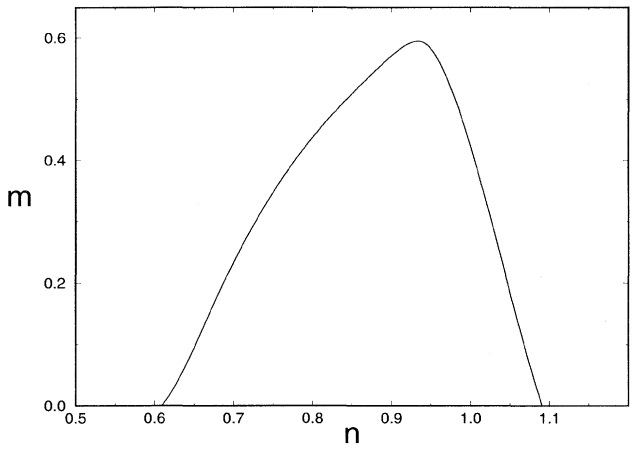

FIG. 6. Copper sublattice magnetization as a function of the total density of holes in the AF solution.

with the experimentally observed asymmetry of the AF stability region under electron or hole doping. Also, the value of the copper moment at $n = 1$ is quantitatively in close agreement with observations.[14] Furthermore, analyzing the behavior of the magnetization as a function of temperature we obtain a mean-field-like curve with a Néel temperature that points to a value of the effective exchange interaction of about 1400 K. This is very close to current estimates.[15]

V. CONCLUSIONS

We have presented here one-particle densities of states for the three-band Hubbard model described by Eq. (1), utilized as a model for the CuO_2 planes of the high-T_c superconductors. These densities of states have been obtained from the corresponding Green's functions, which have been calculated through perturbation theory around the atomic limit, in the simplest approximations.

We wish to emphasize some points concerning the results presented here that may differ from current expectations. Starting from the uncorrelated case, one would have a lowest lying (hybrid) band that can hold up to two holes. Qualitatively, one could think that a Hubbard-like splitting of this band due to correlations would yield an insulating state when this band was half filled ($n = 1$). However, as we have shown, the fact that this is a hybrid band, and that correlation effects are different on the two kinds of initial orbitals, makes the effective correlation weaker than in the single-band Hubbard model. The result is that the lowest lying band can hold more than one hole, even after correlation effects have been taken into account. Nevertheless, an insulating antiferromagnetic state is obtained for a hole density close to 1, with a gap opening inside the lowest band.

The picture we have is, thus, the following. For a small number of holes (strong electron doping), the lowest band is partially filled and the system is metallic. When the number of holes approaches unity (from weak electron doping up to a little above the stoichiometric situation) an antiferromagnetic gap opens inside the lowest lying band, giving rise to a magnetically ordered insulating ground state. When the number of holes is further increased the AF state becomes unstable, the gap closes, and we go back to a situation in which the lowest lying band is partially filled, yielding a metallic state. Only for strong hole doping do we reach again an insulating condition, now due to the presence of the CT gap.

We would like to mention an important point related to experiment that can possibly support these results. Photoemission studies[9] show that the Fermi level for both electron or hole doping remains in the same energy region, close to the value that one would expect from band theory calculations, and the original gap is *completely filled* when the AF order disappears. This is in qualitative agreement with our calculations. In contrast, it could not be understood in terms of an effective Hubbard-like one-band model, where a jump of the Fermi level across the correlation gap should be expected when going from hole to electron doping.[16]

In a certain sense, our results agree with those obtained by the Hartree-Fock (HF) approximation,[5] at least with respect to the nature of the insulating state, which presents a bandlike AF gap that does not coincide with the CT one. However, in the HF solution the AF instability of the paramagnetic (PM) solution is due to a perfect nesting property of the Fermi surface,[5] which can be viewed as related to the van Hove singularity of the DOS. This is not the case in our treatment, although the form of the DOS could suggest an interpretation on the same lines. We checked this point by repeating the calculations with a model (rectangular) density of states. Despite the absence of the two-dimensional van Hove logarithmic singularity in this DOS, we still obtained the AF state. It is important to mention that the same approximation is not able to find the AF phase in the half-filled single-band Hubbard model. Thus in the present case the method incorporates correctly the role of p orbitals in a superexchange mechanism that explains antiferromagnetism in high-T_c compounds.

A metallic character of the paramagnetic solution has also been found in a slave-boson treatment of the problem by Riseborough and Hanggi.[8] They actually suggest that the insulating behavior could be due to the opening up of an AF gap at the Fermi surface, as obtained here. In addition, it has been argued[17] that it is possible to explain effects of diamagnetic substitutions on the Néel temperature of La_2CuO_4 by the assumption of a metallic state in the PM phase.

A comparison with local moment approaches, in particular the Zhang-Rice[3] picture, is not as easy. Although our starting point is the same (the atomic limit), and the effective Heisenberg exchange interaction that we found is in the expected range, as we mentioned in the end of Sec. IV, the charge transfer nature of the ground state seems to be preserved in the Zhang-Rice results. In contrast, we find that the holes in the stoichiometric case do not reside on cooper sites, but present a hybrid nature, with an important admixture of copper and oxygen levels in the occupied part of the spectrum. We could suggest

that our partial renormalization of the local Green's functions that led to Eq. (11) contemplates, at least in part, the hybridization mechanism that leads to formation of the so called Zhang-Rice singlet. However, our decoupling of the local averages contributing to these Green's functions is probably the weakest point of the present treatment, and may explain the weak-coupling-like character of our final results. An extension of the work to include many-loop corrections to the local Green's functions is now in progress.

A complete three-band model for the copper-oxygen planes of high-T_c superconductors should contain direct hybridization between oxygen orbitals, as well as nonlocal Coulomb interaction between holes in nearest neighbor atoms. We have investigated the effect of including a hopping term connecting oxygen sites within our approach. Preliminary results indicate that the main effect is a broadening of the nonbonding band, with consequent reduction of the CT gap. A nonlocal Coulomb interaction is not easy to include in our formalism because, in contrast to the local one, it has to be treated as a perturbation. Thus we are faced with a double perturbation expansion, with respect to both hopping and nonlocal interaction. Work on these lines is also currently in progress.

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