Magnetic and charge-transfer phase separation in the three-band t-J model

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We investigate the physical properties of a three-band generalized Hubbard model for the CuO_2 planes of high-T_c superconducting cuprates, in the presence of Cu-Cu superexchange and Cu-O Coulombic interaction. The explicit introduction of these interaction terms emphasizes the different role of spin and charge degrees of freedom in determining the behavior of the system. In particular, they provide two different mechanisms for phase separation, which are more effective in different doping regions. Experimental indications for this to occur are discussed. We infer that magnetic and charge-transfer pairing interactions are related to the above phase-separation mechanisms and, while being simultaneously present, gradually interchange their relevance, the first one being dominant at low and intermediate doping, and the second one at high doping.

I. INTRODUCTION

Many different behaviors arise upon doping in copper oxides due to the strongly correlated nature of these systems and the interplay of the various degrees of freedom.[1] The antiferromagnetic insulator which is present in the stoichiometric and lightly doped compound turns into a *strange metal*, which at optimal doping displays a linear T dependence of the resistivity above the critical temperature up to very high temperatures. Magnetic correlations and possibly a spin gap are present in the metal at low and intermediate doping. At still higher doping the strange metal gradually turns into a normal metal which displays a Fermi-liquid behavior and strongly reduced magnetic scattering. Three qualitatively different nonsuperconducting behaviors are therefore present in copper oxides: antiferromagnetic insulator, strange metal, and normal metal.

The above scenario suggests that spin degrees of freedom play a major role at low doping, when charge degrees of freedom are essentially frozen out due to the strongly correlated nature of the system. In this regime magnetic correlations are naturally expected to be relevant, so that a description in terms of a (extended) t-J model has often been proposed to suitably describe the low-energy physics of the system.

On the other hand charge degrees of freedom become gradually more important in the intermediate- and high-doping regimes. In particular, while the undoped system has one hole per cell mostly residing in copper $d_{x^2-y^2}$ (d_x) orbitals, the holes introduced by doping have a large wave function component on oxygen sites. Well inside the metallic regime it is natural to expect that the covalent character of the copper-oxygen planes becomes more relevant, thus emphasizing the importance of the charge dynamics. Although it was sometimes proposed[2] that an effective single-band t-J model could still be appropriate in this high-doping regime, various considerations indicate that this scheme *could* be somewhat oversimplified: First of all, contrary to the single-band model, the original three-band model[3,4] exhibits a wider variety of behaviors and is able to describe the occurrence of a metal–charge-transfer–insulator transition (MCTIT) by varying the atomic-level energy difference between the oxygen $2p$ orbitals ε_p^0 and the copper d_x orbital ε_d^0.[5] A multiband model, which keeps track of this relevant energy scale, is certainly richer since it allows a clear identification of the role of this transition in the charge dynamics. Second the nearest neighbor (NN) Coulombic repulsion V between holes on copper and holes on oxygen can play a role affecting the oxygen-copper charge-transfer fluctuations.[4] In particular, it was shown in previous work,[4,6–8] that if V is sizable, charge-transfer excitations are relevant in the low-energy charge dynamics and may affect the stability of the system.

The above considerations show that the reduction to a single-band effective model could miss some important physical effects and motivate our investigation of an extended three-band Hubbard model.

Since we are interested in both the magnetic effects and the charge dynamics, by investigating the (smooth) crossover between the low-doping and the intermediate- or high-doping regimes we are going to equip our model with magnetic interaction terms,[11] as well as with NN Coulombic repulsion between holes on copper and holes on oxygen.

While the model and the technical details of our approach are contained in Sec. II, the results are presented in Sec. III and the physical implications as well as our conclusions are in Sec. IV.

II. MODEL AND TECHNICAL DETAILS

Due to the strongly interacting nature of the superconducting copper oxides, we consider a model with an infinite repulsion U_{dd} between holes on the same copper atoms. We treat the limit of very large local repulsion between holes on copper sites by means of the slave-boson technique.[12] Moreover, due to the above mentioned possible relevance of the NN Coulombic repulsion between holes on copper and holes on oxygen, we extend the standard three-band Hubbard model with a NN Coulombic interaction V. Finally, to include the effects of magnetism we also consider a direct Heisenberg interaction between spins on NN copper sites.[9,10] To be sure, superexchange magnetic effects are already contained in the extended three-band Hubbard model, so that, in principle, this latter term should not be included. However, our slave-boson treatment of the model misses this interaction in the mean-field approximation, which we are going to consider (a Cu-Cu magnetic interaction would arise only at higher order in the perturbation expansion, via the exchange of slave-boson fluctuations). Therefore, following a customary use we explicitly include a superexchange coupling J. The value of J is in principle fixed by the other parameters of the Hamiltonian, but, in the very same spirit of the t-J model, in the following we consider it to be an independent parameter. However, having in mind a description of the copper oxides, we will not assume for the magnetic coupling J values which are unrealistic for these systems ($J \leq 0.2$ eV). According to the above manyfold extensions of the three-band Hubbard model we decided to indicate the resulting Hamiltonian as a three-band t-J model.

The resulting t-J slave-boson Hamiltonian reads

$$\mathcal{H} = \sum_{il,\sigma} \left(t_{pd}^{l} d_{i\sigma}^{\dagger} b_i p_{i+l,\sigma} + \text{H.c.} \right) + \sum_{\substack{ill' \\ \sigma}} \left(t_{pp}^{ll'} p_{i+l,\sigma}^{\dagger} p_{i+l',\sigma} + \text{H.c.} \right) + V \sum_{\substack{il \\ \sigma\sigma'}} n_{i\sigma}^{d} n_{i+l,\sigma'}^{p}$$
$$+ (\varepsilon_p^0 - \mu) \sum_{il,\sigma} p_{i+l,\sigma}^{\dagger} p_{i+l,\sigma} + (\varepsilon_d^0 - \mu) \sum_{i,\sigma} d_{i\sigma}^{\dagger} d_{i\sigma} + J \sum_{\substack{(ij) \\ \sigma\sigma'}} d_{i\sigma}^{\dagger} d_{i\sigma'} d_{j\sigma'}^{\dagger} d_{j\sigma} + \frac{U_{pp}}{4} \sum_{il,\sigma} n_{i+l,\sigma}^{p} n_{i+l,-\sigma}^{p}, \tag{1}$$

where $d_{i\sigma}^{\dagger}$ creates a hole with spin σ on the copper $d_{x^2-y^2}$ orbital on the site i, while $p_{i+l,\sigma}^{\dagger}$ (we take the Cu-Cu distance to be unit, so that $l = \pm\mathbf{x}/2, \pm\mathbf{y}/2$) creates a hole on the p_x or p_y oxygen orbitals on site $i + l$. The density operators are defined by $n_{i\sigma}^{d} = d_{i\sigma}^{\dagger} d_{i\sigma}$ and $n_{i+l,\sigma}^{p} = p_{i+l,\sigma}^{\dagger} p_{i+l,\sigma}$. Hamiltonian (1) includes atomic, hybridization, superexchange, and NN Coulombic terms. The slave-boson operator $b_i^{\dagger}$, which labels the empty sites, has been introduced in the p-d hybridization term to take care of the strongly correlated nature of copper sites. The no-double-copper-occupancy constraint $\sum_{\sigma} d_{i\sigma}^{\dagger} d_{i\sigma} + b_i^{\dagger} b_i = 1$ has to be enforced to project the Hamiltonian (1) onto the physical states. The mean-field Hamiltonian $\mathcal{H}_{\text{MF}}$ is obtained posing $\langle b_i \rangle = b_0$ (uniform all over the system) and enforcing the constraint of no double copper occupancy via a uniform lagrange multiplier λ_0, so that the constraint is fulfilled in the average.

The quartic interactions are decoupled via the Hartree-Fock factorizations

$$J \sum_{\substack{(ij) \\ \sigma\sigma'}} d_{i\sigma}^{\dagger} d_{i\sigma'} d_{j\sigma'}^{\dagger} d_{j\sigma} \rightarrow \frac{2N_s}{J} (\Delta_1^2 + \Delta_2^2)$$
$$+ \Delta_1 \sum_{im,\sigma} \left(\phi_m d_{i\sigma}^{\dagger} d_{i+m,\sigma} + \text{H.c.} \right) + \Delta_2 \sum_{im} \left[\psi_m \left(d_{i\uparrow} d_{i+m,\downarrow} - d_{i\downarrow} d_{i+m,\uparrow} \right) + \text{H.c.} \right], \tag{2}$$

with $\Delta_1 \phi_m = J \langle \sum_{\sigma'} d_{i\sigma'} d_{i+m,\sigma'}^{\dagger} \rangle$ and $\Delta_2 \psi_m = J \langle d_{i\uparrow}^{\dagger} d_{i+m,\downarrow}^{\dagger} - d_{i\downarrow}^{\dagger} d_{i+m,\uparrow}^{\dagger} \rangle$, N_s being the number of copper sites,

$$\frac{U_{pp}}{4} \sum_{il,\sigma} n_{i+l,\sigma}^{p} n_{i+l,-\sigma}^{p} \rightarrow \frac{U_{pp}}{4} \sum_{il,\sigma} n_p n_{i+l,\sigma}^{p} - \frac{N_s U_{pp}}{2} n_p^2, \tag{3}$$

where $n_p = 2 \langle n_{i+l,\sigma}^{p} \rangle$ is the p density per unit cell and per spin,

$$V \sum_{il,\sigma\sigma'} n_{i\sigma}^{d} n_{i+l,\sigma'}^{p} \rightarrow 2X_0 \sum_{i,\sigma} n_{i\sigma}^{d} + 2Y_0 \sum_{il,\sigma} \frac{n_{i+l,\sigma}^{p}}{2} - \frac{2N_s}{V} X_0 Y_0 + \frac{8N_s Z_0^2}{V} + Z_0 \sum_{il,\sigma} \left(\theta_l d_{i\sigma}^{\dagger} p_{i+l,\sigma} + \text{H.c.} \right)$$
$$- \frac{8N_s \Delta_{pd}^2}{V} + \Delta_{pd} \sum_{il} \left[\chi_l \left(p_{i+l,\uparrow} d_{i\downarrow} - p_{i+l,\downarrow} d_{i\uparrow} \right) + \text{H.c.} \right], \tag{4}$$

where $X_0 = Vn_p$, $Y_0 = Vn_d$, and $n_d = \langle n^d_{i\sigma}\rangle$ is the d density per unit cell and per spin; $Z_0\theta_l = V\langle d_{i\sigma}p^\dagger_{i+l,\sigma}\rangle$ is the Fock decoupling term, and $\Delta_{pd}\chi_l = V\langle d^\dagger_{i\downarrow}p^\dagger_{i+l,\uparrow}\rangle$ is the anomalous decoupling term. An anomalous p-d order parameter can be factorized since it is indirectly generated by the attraction mediated by J; it would otherwise vanish since the V interaction is purely repulsive.

The on-site p-p anomalous average needs not to be considered in the U_{pp} term because it is not generated in the attractive channel mediated by J; nor it can arise due to the decoupling of the repulsive U_{pp} term.

In the above formulas ϕ_m, ψ_m, θ_l, and χ_l ($m = \pm\mathbf{x}, \pm\mathbf{y}$, $l = \pm\mathbf{x}/2, \pm\mathbf{y}/2$) are phase factors which take into account the proper symmetry of the order parameters. We mainly specialize our analysis to the case $\phi_m = 1$ (uniform symmetry); $\theta_l = 1$ for $l = \mathbf{y}/2, -\mathbf{x}/2$; $\theta_l = -1$ for $l = -\mathbf{y}/2, \mathbf{x}/2$ (p-d bonding symmetry). For the anomalous d-d order parameter we take $\psi_m = 1$ for m parallel to $\mathbf{x}$ and $\psi_m = -1$ for m parallel to $\mathbf{y}$ corresponding to a d-wave pairing since we never found an s-wave solution with lower energy. As far as the anomalous p-d term is concerned we have $\chi_l = i$ for $l = \mathbf{x}/2, \mathbf{y}/2$ and $\chi_l = -i$ for $l = -\mathbf{x}/2, -\mathbf{y}/2$. An order parameter displaying this symmetry arises indeed as a consequence of the attractive superexchange interaction only. Note, instead, that in the mean-field treatment of our model no pairing is present due to the repulsive charge-transfer interaction V. This is a shortcoming of our mean-field analysis. Indeed previous analyses[6–8] have shown that a charge-transfer-mediated attraction can appear when fluctuations are considered.

The mean-field free energy $\mathcal{F}_{\rm MF}$ is required to be stationary with respect to the mean-field parameters b_0, λ_0, Δ_1, Δ_2, Δ_{pd}, X_0, Y_0, and Z_0. The self-consistency equations have then the general form

$$\frac{\partial\mathcal{F}_{\rm MF}}{\partial P_i} \equiv \frac{\partial\mathcal{F}_0}{\partial P_i} + \sum_{\substack{k,\alpha\\ \sigma}} f(E^\alpha_k - \mu)\frac{\partial E^\alpha_k}{\partial P_i} = 0, \qquad (5)$$

where

$$\mathcal{F}_0 \equiv N_s\Bigg[\lambda_0(b_0^2-1) - \frac{2}{V}X_0Y_0 + \frac{8}{V}Z_0^2 - \frac{8}{V}\Delta_{pd}^2 + \frac{2}{J}(\Delta_1^2+\Delta_2^2) - \frac{U_{pp}}{2}n_p^2\Bigg];$$

$f(E^\alpha_k-\mu)$ is the Fermi function, E^α_k are the eigenvalues of the mean-field Hamiltonian, and P_i represents a generic mean-field parameter. The condition of particle number conservation $-\partial\mathcal{F}_{\rm MF}/\partial\mu = N_s(1+\delta)$, where δ is the hole doping with respect to half-filling, has to be numerically solved together with the self-consistency equations to obtain the phase diagram of the model in terms of the physical parameters.

The slave-boson approach here adopted to deal with the $U_{dd} = \infty$ limit has often been used in the investigation of strongly interacting systems like heavy fermions and superconducting cuprates. It has the major advantage of being nonperturbative in the interaction, so that, already at the mean-field level, it allows a suitable description of the strongly correlated metallic phase and of the metal-insulator transition.[13] In particular, as it is easily recognized from an inspection of Eq. (1), the mean-field value b_0 of the slave-boson field multiplicatively renormalizes the Cu-O hopping t_{pd}, leading to a bandwidth reduction ($b_0 < 1$). On the other hand, the mean-field value of the Lagrange multiplier λ_0 additively shifts the value of the copper atomic-level energy. For positive doping, the shifted copper level $\varepsilon_d = \varepsilon_d^0 + \lambda_0$ approaches the oxygen levels so as to allow the occupied band to acquire enough oxygen character in order to accomodate $n = 1+\delta$ holes per cell without violating the no-double-occupancy constraint.[13]

At finite doping the system is always metallic for any value of the bare charge-transfer energy $\varepsilon_p^0 - \varepsilon_d^0$: The bandwidth renormalization b_0 is finite and, by varying doping or $\varepsilon_p^0 - \varepsilon_d^0$, the renormalized copper level is continuously shifted together with the chemical potential. For small values of $\varepsilon_p^0 - \varepsilon_d^0$ the system stays metallic even at half-filling, whereas, for values of $\varepsilon_p^0 - \varepsilon_d^0$ larger than a critical value, the system becomes insulating when $n = 1$. Specifically, the bandwidth vanishes ($b_0 = 0$) and the chemical potential discontinuously jumps in going from negative to positive doping.

The present slave-boson technique provides, therefore, a mean-field description of the system in the metallic phase in terms of self-consistently renormalized quasiparticle bands. The insulating phase is characterized by an infinite quasiparticle mass (zero bandwidth) and a jump in the chemical potential in the limit of infinitesimally small negative or positive doping. The above scheme results from the limit of infinite U_{dd} and underlines the role of the energy-level difference $\varepsilon_p^0 - \varepsilon_d^0$ in determining the charge dynamics and particularly the metallic versus charge-transfer-insulating character of the system.

For the treatment of the other interaction terms of the model (1) we adopted a standard decoupling approach. This choice is justified by the relatively small value of the couplings involved, so that a Hartree-Fock-like scheme should be appropriate in order to obtain a qualitatively correct picture.

As far as the magnetic interaction is concerned, while real systems display antiferromagnetic spin long-range order in the insulating phase, we considered a magnetic phase with dimer order, in which the Δ_1 order parameter is real and has different values on the various bonds ($\Delta_1^x \neq \Delta_1^{-x} \neq \Delta_1^{\pm y}$). This magnetic singlet phase has no spin long-range order, although it mimics the antiferromagnetic (AF) insulator with the same doubling of the unit cell. An explicit spin-spin decoupling could in principle be considered, allowing for the introduction of antiferromagnetic spin long-range order. However, in the physical systems, the antiferromagnetic long-range order is rapidly destroyed by doping, giving rise to a metallic phase with magnetic correlations. Since we are focusing on the behavior of the metallic phase of the cuprates and to its smooth crossover from the intermediate-doping to the high-doping regimes, we neglected any decoupling leading to a possible spin long-range order. The introduction of a spin-symmetry breaking, in fact, would only be needed to describe the insulating phase and its doping-

induced disruption. This latter phenomenon involves in a crucial way the role of fluctuations and it consitutes a difficult, still unsettled, problem, which is beyond the scope of our work and which hardly could be reasonably approached within our mean-field scheme. This is why we directly focused on the singlet decoupling which is more suited to describe the magnetic correlations in the metallic phase already in the mean-field approximation. Of course one should be aware of the fact that our treatment of the magnetic correlations is not suitable for a complete analysis of the insulating phase and it only provides a qualitative description of the magnetic correlations. The phase diagram obtained in this way can be qualitatively trusted at not too low doping, corresponding to a physical situation when the spin long-range order has already disappeared.

III. PHASE DIAGRAM: PHASE SEPARATION AND SUPERCONDUCTIVITY

We numerically solved the self-consistency equations for the b_0, λ_0, Δ_1, Δ_2, Δ_{pd}, X_0, Y_0, and Z_0 mean-field parameters at fixed doping and for various values of the Hamiltonian parameters. We need to clarify the different roles of these mean-field parameters. According to the discussion in the previous section, b_0, λ_0, X_0, Y_0, and Z_0 determine the renormalized band structure: b_0 and Z_0 act on the t_{pd} hopping, whereas λ_0, X_0, and Y_0 shift the copper and oxygen atomic levels. On the other hand, Δ_1, Δ_2, and Δ_{pd}, although they also affect the band structure, are more directly related to the existence of magnetic correlations (Δ_1) or to the anomalous superconducting order (Δ_2 and Δ_{pd}). In this sense they directly characterize the various phases of the system. Threfore, the phase diagram will mainly be concerned with their behavior only. Moreover, due to hybridization, the establishment of superconducting long-range order in the *pd* sector (Δ_{pd}) is strictly related to the appearance of the same long-range order in the *dd* sector (Δ_2), so that only this latter order parameter needs to be considered.

In Fig. 1 the phase diagram is given in the plane of the bare charge-transfer gap $\varepsilon_p^0 - \varepsilon_d^0$ and the doping δ. At $T = 0$ a metal-insulator transition is found at half-filling for a critical value of the bare charge-transfer gap. In the presence of a sufficiently large *p-d* Coulombic interaction V, this transition is first order and takes place between a metal with uniform singlet decoupling of the superexchange term ($\Delta_1 \sim J, \Delta_2 = 0$) and an insulating magnetic phase.

At low doping, above the critical value of $\varepsilon_p^0 - \varepsilon_d^0$, the system is unstable due to the presence of the magnetic phase, which tends to eject the holes introduced by the doping. In the absence of long-range Coulombic interactions the system is led to phase separation,[10,14,15] between hole-poor and hole-rich regions.

At intermediate doping, if V is sufficiently large, a different instability occurs as a consequence of charge-transfer processes. To our knowledge the charge-transfer mechanism is the only purely electronic mechanism providing phase separation in the intermediate- and high-

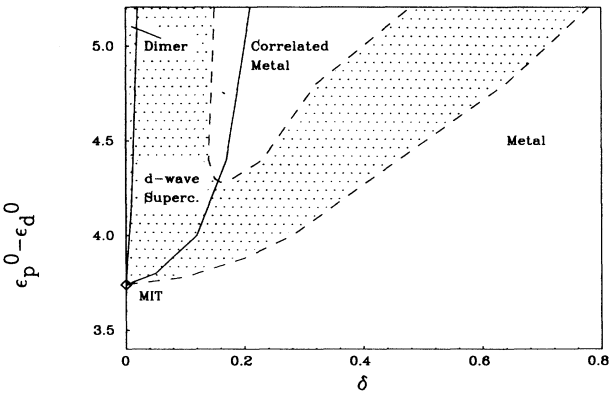

FIG. 1. Phase diagram $\varepsilon_p^0 - \varepsilon_d^0$ vs the doping δ with T=0.002 eV, t_{pd}=1.4 eV, t_{pp}=0.4 eV, J=0.2 eV, V=3.0 eV, and U_{pp}=0.5 eV. Solid lines indicate the phase transition lines. The shaded area delimited by the dashed lines indicates the phase-separation region determined via the Maxwell construction. The diamond indicates the metal-insulator transition (MIT).

doping regimes.[6–8] It must be emphasized that *these two mechanisms for phase separation do not prevent each other, but play their roles in different regions of doping.* The two phase separations merge in the vicinity of the metal-insulator transition.

In the intermediate-doping regime, a *d*-wave superconducting phase ($\Delta_1, \Delta_2 \sim J$) is stable at large charge-transfer gaps.

The metallic phase stable at higher doping changes continuously from a metal with local magnetic correlations ($\Delta_1 \sim J, \Delta_2 = 0$) to a standard metal with no magnetic correlations ($\Delta_1 \simeq 0, \Delta_2 = 0$).

As mentioned above, direct formation of superconductivity induced by the repulsive charge-transfer interaction V cannot be achieved within our mean-field approach. Nevertheless, we stress that in the stable region near the phase separation driven by the charge-transfer mechanism, an attractive residual interaction among the Fermi quasiparticles is present. A general Fermi-liquid analysis may, indeed, be useful in this respect. This analysis allows, in fact, to find that in the high-doping stable regime the system is still characterized by a negative (i.e., attractive) dynamical scattering amplitude

$$\Gamma_\omega = \lim_{\omega \to 0} \lim_{q \to 0} \Gamma\left(k_F, k_F'; q = k_F - k_F', \omega\right), \qquad (6)$$

where $\Gamma\left(k_F, k_F'; q = k_F - k_F', \omega\right)$ is the residual interaction between the quasiparticles, which arises from the exchange of the fluctuations of the bosonic fields. In our mean-field approach, where no exchange of bosons has been considered, we can evaluate only the small q and ω limits of this residual interaction from the direct calculation of the compressibility[16]

$$\kappa \equiv \frac{\partial n}{\partial \mu} = \frac{\nu^*}{1 + F_0^s},$$

where ν^* is the quasiparticle density of states at the Fermi level and F_0^s is the usual effective Landau parameter $F_0^s \equiv 2\nu^*\Gamma_\omega$.[17] We numerically evaluated the compressibility $\frac{\partial n}{\partial \mu}$ from the μ-vs-δ self-consistent behavior, as well as the quasiparticle density of states ν^*. From their comparison, it turns out that F_0^s, i.e., the dynamical limit of the interaction between the quasiparticles Γ_ω, is negative ($-1 < F_0^s < 0$; see Fig. 2) in a large region close, but outside, the phase-separation region, where the Landau-Pomeranchuk criterion for stability is violated ($F_0^s < -1$). Additional insight can be obtained by recasting the expression of the compressibility in terms of the static limit of the interaction between the quasiparticles,

$$\kappa = \nu^* \left(1 - 2\nu^*\Gamma_q\right), \tag{7}$$

where

$$\Gamma_q = \frac{\Gamma_\omega}{1 + 2\nu^*\Gamma_\omega} = \lim_{q\to 0}\lim_{\omega\to 0} \Gamma\left(k_F, k_F'; q = k_F - k_F', \omega\right) \tag{8}$$

is the static effective interaction between two quasiparticles at the Fermi level once the screening due to all the quasiparticles is taken into account. From the expression of Eq. (7) one can see that a divergent compressibility can be related to a negatively divergent total effective scattering amplitude between the quasiparticles at large distance ($q \to 0$).

The presence of attractive forces in the particle-hole channel makes plausible (and indeed it allows one to show in a random-phase-approximation) the existence of attractive forces in the particle-particle channel, eventually leading to Cooper instability. An additional mechanism for pair formation in the high-doping regime is in this way provided, which extends to higher doping and lower bare charge-transfer gaps, the superconducting phase shown in the phase diagram of Fig. 1.

The tendency towards phase separation could therefore provide the attractive forces leading to superconductivity.[6–8] Moreover, superconductivity could stabilize the system with respect to phase separation,[18] thus reducing the phase-separation region.

We note that, when the strength of the local Coulomb repulsion on oxygen sites U_{pp} is increased above 1.5 eV, the phase-separation instability due to the charge-transfer mechanism is suppressed. However, even in the absence of this second phase separation, a sizable doping interval persists, where an attraction is present in the particle-hole channel as a remnant of the instability occurring at lower U_{pp}. Keeping all the others parameters fixed at the values reported in Fig. 1, this attraction disappears for U_{pp} larger than about 3 eV [density functional calculations in the local density approximation (LDA) usually report values between 0 and 4 eV for this quantity in the cuprates]. Exact cluster diagonalization calculations,[19] providing indications of superconductivity occurring close to a charge-transfer-induced phase separation, were reported in the absence of U_{pp} only. Thus they do not help us in testing our mean-field results on the disappearance of the phase separation with U_{pp}.

In Fig. 3 a phase diagram in the T-vs-δ plane is shown. The low-doping instability separates a fully dimerized insulating phase and a d-wave superconducting phase at low temperature. At higher temperature the phases which separate are the dimerized phase and a magnetically correlated metal. The intermediate-doping instability separates instead a magnetically correlated hole-poor metal ($\Delta_1 \sim J, \Delta_2 = 0$) and a normal hole-rich metal ($\Delta_1 \simeq 0, \Delta_2 = 0$).

In Fig. 4 we show a different T-vs-δ phase diagram, which was obtained for a lower value of the bare charge-transfer gap. In this case only one phase-separation region is present. Both the magnetic and charge-transfer degrees of freedom are relevant in the same region of doping. The small value of $\varepsilon_p^0 - \varepsilon_d^0$ allows the charge-transfer mechanism to be effective already at low doping. Phase separation takes place between a dimer insulator at $\delta = 0$ and a magnetically correlated metallic phase at intermediate doping, which gradually turns into a normal metal without any further instability. We remark that in the present case strong magnetic correlations persist up to quite high doping due to the presence of droplets of a zero doping insulator in the system.

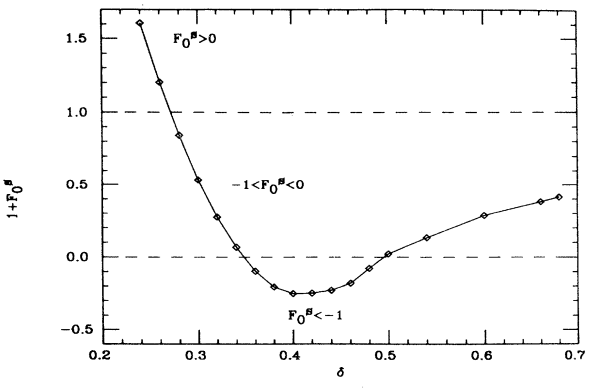

FIG. 2. Compressibility denominator $1 + F_0^s$ as a function of doping in the proximity of the charge-transfer-mediated instability.

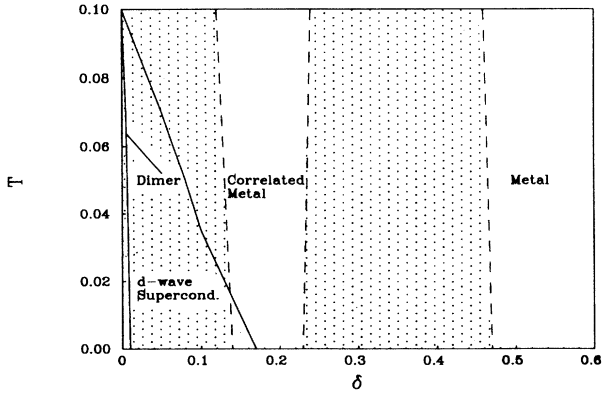

FIG. 3. Phase diagram T vs the doping δ with $\varepsilon_p^0 - \varepsilon_d^0$=4.4 eV; all other parameters are as in Fig. 1.

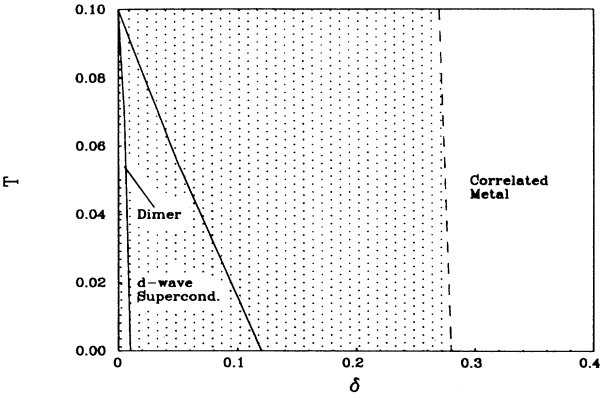

FIG. 4. Phase diagram T vs the doping δ with $\varepsilon_p^0 - \varepsilon_d^0 = 4.0$ eV; all other parameters are as in Figs. 1 and 3.

IV. DISCUSSION AND CONCLUSIONS

The results reported in the previous section are characterized both by the presence of phase separation in substantial regions of the phase diagram and by its connection with the occurrence of superconductivity.[24]

The location of the phase-separation regions in the phase diagram of Fig. 1, which start immediately above the metal–charge-transfer–insulator transition, clearly indicates the physical and generic origin of the phase separation in strongly interacting systems. A particularly intuitive explanation of this phenomenon can be achieved if one thinks at the competitive effects of the kinetic energy and the short-range inteactions (magnetic and NN Coulombic) present in the system: Whereas the kinetic energy would favor the formation of extended Bloch states, the other interactions tend to induce localizing-attractive interactions between the quasiparticles. In ordinary (nonstrongly interacting) metals the kinetic energy is dominant and a uniform, homogeneous phase is formed. On the other hand, in the presence of strong local repulsion, the kinetic energy is strongly depressed (in particular it vanishes in the proximity of the insulating phase) and it is no longer able to succesfully oppose the destabilizing effects of the short-range interactions. This remark allows a natural interpretation on why phase separation extends in the low-doping and high charge-transfer—energy-difference regions of the phase diagram.

Of course all the above arguments, far from being a proof of phase separation in all strongly interacting systems, only provide a rather natural rational to this phenomenon, the explicit occurrence of it remaining to be proved in each specific model. Phase separation in the various models still remains a debated and open issue. In particular, whereas numerical calculations seem to exclude its occurrence in the single-band Hubbard model,[20] the presence of phase separation in the single-band t-J model has been repeatedly claimed. In this latter case it still remains debated whether phase separation occurs for any value of J (Refs. 14, 15, 21) or a J larger than a critical value is needed.[22,23] In this context, without claiming to provide ultimate answers to this problem, we simply stress that our mean-field slave-boson approach suggests the presence of phase separation even for small values of the magnetic coupling J in the three-band Hubbard model. Moreover, in accordance with previous findings in the analysis of the three-band extended Hubbard model,[6–8] we confirm the presence of a phase separation induced by a NN-Coulombic interaction. Rather remarkable it appears the fact that, being based on different mechanisms, mostly effective in different doping regions, the magnetically induced and the NN Coulombic-induced phase separations peacefully cohexist in the phase diagram of our generalized three-band t-J model. This latter result particularly emphasizes the generic robustness of the phase-separation concept. However, a word of caution is needed in relating our results to the physics of the superconducting copper oxides. In particular, whereas we find that the magnetically induced phase separation may well occur for $J = 0.1$–0.2 eV, which corresponds rather well to commonly accepted values for the magnetic coupling in the copper oxides, too large values of V are needed to induce phase separation in our model. Indeed, whereas the commonly accepted values of V, as calculated from density functional calculations in the LDA, are smaller than 1 eV, we only observe a Coulombic-induced phase separation for values of $V > 1.75$ eV. Nevertheless, one should keep in mind that the theoretical estimates of V are obtained within a LDA approach and that our model was solved within a mean-field approximated scheme. Therefore requiring a really quantitative matching of our parameter V with the commonly accepted value for the cuprates may be too restrictive.

As far as the relation between phase separation and superconductivity is concerned, the general Fermi-liquid analysis reported in the previous section clearly indicates that a divergent compressibility not only marks the presence of phase separation, but it is also related to strong attractive forces in the small-q particle-hole effective potential. This strong attraction can then originate the occurrence of superconductivity.[6–8,18]

Long-range Coulomb forces, neglected in model (1), would oppose the phenomenon of phase separation.[25] However, the diffusional motion of negatively charged oxygen ions[26] in some copper oxides provides a natural way to balance the electrostatic repulsion arising from the hole segregation. This mechanism could be present in overoxygenated La_2CuO_{4+y} and allows to attribute an electronic origin to the phase separation observed therein.[27] When the counter-ion diffusion is ineffective long-range forces prevent a true thermodynamic phase separation. However, we expect that the long-range Coulomb interactions, being strongly effective only at small q's, while suppressing the $q=0$ thermodynamic phase separation, may leave the way open to dynamical (slow) charge fluctuations[25] or to charge density waves.[8]

Some evidence of the double-phase-separation scenario is provided by neutron scattering experiments[28] in $ErBa_2Cu_3O_{6+x}$. The spectra of crystal field excitations of Er^{3+} show the existence of three distinct environments of these ions. By varying the doping three absorption

peaks coexist, having their maximum at x=0, $x \approx 0.6$, and $x \approx 0.95$, respectively. This may indicate the tendency of the system to phase separate between an insulating O_6 phase and an intermediate-doping metal at $O_{6.6}$ and between this $O_{6.6}$ metal and the high-doping metal with $O_{6.95}$. The overdoped phase ($x > 0.95$) seems to be the pure high-doping metal. It is important to realize that this analysis was carried out with a local dynamical probe. It was therefore able to provide information on the tendency to phase separation regardless to the actual occurrence of a true long-range thermodynamical segregation of different phases.

This scenario would, therefore, confirm the presence of two distinct "phase separations" (the quotes indicate the possibly local dynamical nature of the separations) as in Fig. 3. However, the experiments, showing a small region of doping where the three peaks may simultaneously coexist, still allow for a description in terms of the diagram of Fig. 4. In this latter case a possible interpretation of our result is that the slow density fluctuations arising from the Coulomb-frustrated phase separation can simultaneously occur along two distinct channels according to the two distinct driving mechanisms hypothesized in this paper. A first channel involves density fluctuations between the AF insulator and the intermediate-doping metal mainly driven by magnetism, whereas the charge-transfer driving force would be responsible for the slow fluctuations between the intermediate-doping metal and the high-doping metal. The fluctuating bubbles of the intermediate-doping metal would then be present at various doping concentrations as a result of the overlap between the two merged "phase separations." This second interpretation would account for both the large width of the peak related to the intermediate-doping metal shown in the experiments and the persistence of some magnetic effects at very large doping. In particular it would also naturally explain the presence of broad maxima at $T^* > T_c$ in the ^{63}Cu relaxation rate $(T_1T)^{-1}$ in the NMR experiments in highly doped (up to $x = 0.92$) 123 samples.[29] The (pseudo) gap in the spin excitations usually advocated to explain this behavior would then be associated to the finite-size spin waves in the AF density fluctuations[25] arising from the first fluctuation channel.

In conclusion the simultaneous presence of magnetic and charge-transfer relevant energy terms in the model leads to a possible explanation of the normal phase and of the superconductivity pairing mechanism. For the first one a gradual change with doping of the characteristics of the metal involved arise from the presence of the two distinct fluctuation channels. The superconductivity pairing mechanism would then be driven over the entire region of doping by the attractive forces produced by two different tendencies towards phase separation. This leads us to suggests that in strongly correlated systems the superconductivity pairing arises nearby a phase-separation region irrespective of the nature of the forces leading to the instability.

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