Superconductivity, Phase Separation, and Charge-Transfer Instability in the $U=\infty$ Limit of the Three-Band Model of the CuO_2 Planes

M. Grilli, R. Raimondi, C. Castellani, and C. Di Castro
Dipartimento di Fisica, Università di Roma "La Sapienza," I-00185 Roma, Italy

G. Kotliar
Serin Physics Laboratory, Rutgers University, P.O. Box 849, Piscataway, New Jersey 08854
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The $U=\infty$ limit of the three-band Hubbard model with nearest-neighbor repulsion V is studied using the slave-boson approach and the large-N expansion technique to order $1/N$. A charge-transfer instability is found as in weak-coupling theory. The charge-transfer instability is always associated with a diverging compressibility leading to a phase separation. Near the phase-separation, charge-transfer-instability region we find superconducting instabilities in the s- and d-wave channels. The requirement for superconductivity is that V be on the scale of the Cu-O hopping as suggested by Varma, Schmitt-Rink, and Abrahams.

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Since the discovery of high-temperature superconductivity [1] in the copper-oxide compounds, an innumerable variety of mechanisms have been proposed. In spite of considerable theoretical effort, however, the origin of this phenomenon is still not agreed upon.

Many models of the copper oxides begin with a two-dimensional Hamiltonian containing a copper-oxygen hybridization and on-site and nearest-neighbor repulsion [2,3]. Varma, Schmitt-Rink, and Abrahams [2], in particular, suggested that for the special chemistry of Cu-O compounds, where the effective copper and oxygen levels are close in energy, the nearest-neighbor repulsion can produce s-wave superconductivity through charge-transfer resonances.

These models have been extensively analyzed in weak coupling [4-6] where two important features have been found: (a) When the nearest-neighbor copper-oxygen repulsion exceeds the copper-oxygen hybridization, the system undergoes a charge-transfer (CT) instability characterized by the vanishing of the lowest excitonic particle-hole excitation. The CT instability is reached by increasing the doping. (b) Near this instability the system undergoes a superconducting instability of A_{1g} (extended s) or B_{2g} (d_{xy}) character. The existence of this attraction required the order-parameter average to extend over the complete Brillouin zone [7,8].

In this paper we would like to analyze the same model in the limit that the copper on-site repulsion is much larger than all the other energy scales in the problem, using the large-N expansion technique which is not perturbative in the coupling constants. There are several motivations for going beyond weak-coupling theory: (a) From photoemission studies it is clear that the copper on-site repulsion is the largest energy scale in the problem, and (b) at half filling the copper oxides are charge-transfer insulators. The proximity to an insulating state may be relevant to the physics and is outside the scope of weak-coupling theory.

We consider the model

$$\tilde{H}=\sum_{i,\sigma}\varepsilon_d^0\tilde{d}_{i\sigma}^\dagger\tilde{d}_{i\sigma}+\varepsilon_p^0\sum_{i,\sigma,\alpha=x,y}p_{i\sigma\alpha}^\dagger p_{i\sigma\alpha} -\frac{t_{pd}}{\sqrt{N}}\sum_{i,\sigma}[(p_{i\sigma x}^\dagger-p_{i\sigma-x}^\dagger+p_{i\sigma y}^\dagger-p_{i\sigma-y}^\dagger)\tilde{d}_{i\sigma}+\text{H.c.}] +\frac{V}{N}\sum_{i,\sigma}\tilde{d}_{i\sigma}^\dagger\tilde{d}_{i\sigma}\sum_{\sigma',\alpha=x,y}(p_{i\sigma'\alpha}^\dagger p_{i\sigma'\alpha}+p_{i\sigma'-\alpha}^\dagger p_{i\sigma'-\alpha}) \quad (1)$$

subject to single-occupancy constraint on the copper, $\sum_\sigma\tilde{d}_{i\sigma}^\dagger\tilde{d}_{i\sigma}\le q_0N$. The index σ runs from 1 to N. The system of interest has $q_0=\frac{1}{2}$ and $N=2$. We carried out a controlled large-N expansion at $q_0=\frac{1}{2}$, using the function-integral technique and the slave-boson trick [9]. $p_x^\dagger$ and $p_y^\dagger$ create a hole in the x and y oxygen orbitals, and the physical copper-hole creation operator is written as $\tilde{d}_{i\sigma}^\dagger=d_{i\sigma}^\dagger b_i$ in the hybridization term in Eq. (1). As usual, a Lagrange multiplier λ_i will be introduced to enforce the single-occupancy constraint. We decouple the copper-oxygen repulsion by introducing two Hubbard-Stratonovich fields X and Y. X is coupled to the difference in charge between a copper atom and its surrounding four

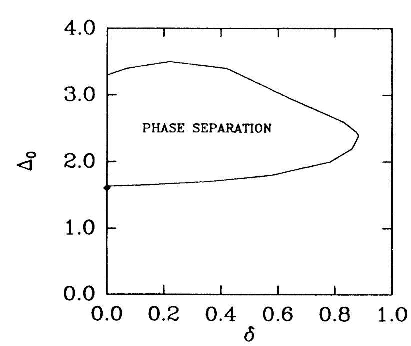

FIG. 1. Phase diagram $\Delta_0\equiv\varepsilon_p^0-\varepsilon_d^0$ vs δ for a value of $V=1.75t_{pd}$. The diamond indicates the Brinkman-Rice transition point.

oxygen orbitals and Y is coupled to the total charge of this cluster. The partition function of the model is then given by

$$Z=\int Dp^\dagger_{\alpha\sigma}Dp_{\alpha\sigma}Dd^\dagger_\sigma Dd_\sigma Db^\dagger Db\,D\lambda\,DX\,DY\exp\left(-\int_0^\beta S\,d\tau\right), \tag{2a}$$

$$S=\sum_i\left[\sum_\sigma d^\dagger_{i\sigma}\frac{\partial d_{i\sigma}}{\partial\tau}+\sum_{\sigma,\alpha}p^\dagger_{i\sigma\alpha}\frac{\partial p_{i\sigma\alpha}}{\partial\tau}+b_i^\dagger\frac{\partial b_i}{\partial\tau}\right]+\sum_i\left[i\lambda_i(b_i^\dagger b_i-q_0N)+\frac{N}{V}(X_i^2+Y_i^2)\right]+H, \tag{2b}$$

$$H=\sum_{i,\sigma}d^\dagger_{i\sigma}d_{i\sigma}(\varepsilon_d^0+i\lambda_i+X_i+iY_i)$$
$$+\sum_{i,\sigma,\alpha}p^\dagger_{i\sigma\alpha}p_{i\sigma\alpha}[\varepsilon_p^0-(X_i-iY_i)]-\frac{t_{pd}}{\sqrt{N}}\sum_{i,\sigma}[(p^\dagger_{i\sigma x}-p^\dagger_{i\sigma -x}+p^\dagger_{i\sigma y}-p^\dagger_{i\sigma -y})d_{i\sigma}b_i^\dagger+\text{H.c.}]\,. \tag{2c}$$

We studied the model of Eq. (2) at the level of mean-field theory ($N=\infty$) and of fluctuations ($1/N$ corrections) at $T=0$, to understand the effect of the nearest-neighbor copper-oxide repulsion.

Our main results are the following.

(a) There is a region (see Fig. 1) in the $(\varepsilon_p^0-\varepsilon_d^0)$-$\delta$ (doping) plane, where the system undergoes phase separation.

(b) Near but outside the phase-separation region of the phase diagram there is a superconducting instability in the s- and d-wave channels. This instability can be seen by taking Fermi-surface averages of the scattering amplitude in the Cooper channel. Hence, the superconductivity found in the weak-coupling analysis [5] persists in strong coupling.

The superconducting instabilities in the s-wave channel can be understood by observing that close to a phase-separation boundary, $dn/d\mu$ is large and therefore F_0^s is negative (but less than 1 to obey the stability requirement). The s-wave scattering amplitude in a simple potential model has a contribution proportional to A_0^s. Note that all previous studies [10,11] using the slave-boson technique and the large-N expansion have so far failed to give s-wave superconductivity because they did not include the parameter V.

(c) There is never a pure CT instability. A divergence of the CT compressibility χ is always accompanied by a divergence of the uniform susceptibility $dn/d\mu$. Therefore phase separation always occurs before the excitonic instability is reached.

(d) There is a critical value of the copper-oxygen repulsion $V_c\sim 2t_{pd}$. For $V<V_c$ the phase-separation region occurs above the metal–charge-transfer-insulator transition (Brinkman-Rice point) which remains second order. For $V>V_c$ the Brinkman-Rice transition becomes first order. Our analysis suggests that it lies on the phase-separation line.

Finally, we note that in the large-N limit the charge-density-wave instability is suppressed because the effect of V in Eq. (1) vanishes at $\mathbf{q}=(\pi,\pi)$ for the direct diagrams, while the exchange ones are higher order. The spin-density-wave instability is also suppressed, since the exchange coupling J is only generated at order $1/N^2$. The absence of a spin-density-wave instability makes our analysis inappropriate to describe the $N=2$ zero-doping case [12]. However, one expects the spin fluctuations to be less relevant by increasing doping.

Recently, experimental evidence has been reported [13] on the presence of phase separation in superconducting oxides. In particular, phase separation appears to occur between the magnetic and the superconducting phase in La_2CuO_{4+x} and $Nd_{2-x}Ce_xCuO_4$ (case 1) or between the superconducting phase and the large-doping metallic phase in $La_{2-x}Sr_xCuO_4$ (case 2). Whereas the occurrence of case 1 is likely to be related to magnetic interactions [14–17], case 2 can be explained by the model discussed in the present Letter [18] (upper branch of the curve in Fig. 1) and indicates the relevance of the charge degrees of freedom at intermediate doping. Our analysis and the experimental findings of Ref. [15] also support the idea that superconductivity and phase separation are possibly related phenomena, both arising from attractive forces [19].

We sketch here the derivation of our results. The saddle-point equations for $X_0=\langle X_i\rangle$, $Y_0=i\langle Y_i\rangle$, $\lambda_0=i\langle\lambda_i\rangle$ [20], and $r_0=\langle b_i\rangle/\sqrt{N}$ are obtained by differentiating the free energy per site and per spin

$$F=\lambda_0(r_0^2-q_0)+\frac{1}{V}(X_0^2-Y_0^2)-\frac{T}{N_{\text{sites}}}\sum_{k,l}\ln[1+e^{-\beta[E_l(k)-\mu]}]\,, \tag{3}$$

where $E_l(k)$ are the three eigenvalues of the Hamiltonian (2c) with saddle-point values of the bosonic fields in it. As said above, for $V<V_c$ the Brinkman-Rice transition is of second order and the mean-field equations have a unique solution. However, an instability occurs as the compressibility $dn/d\mu$ becomes infinite. The phase diagram of Fig. 1 is derived by calculating μ vs n and using a Maxwell construction. Near the lower instability line the almost soft mode has a strong CT component and a small but finite density-fluctuation component. Near the upper border line the two components appear instead with almost equal weight.

For $V>V_c$ the Brinkman-Rice transition is first order,

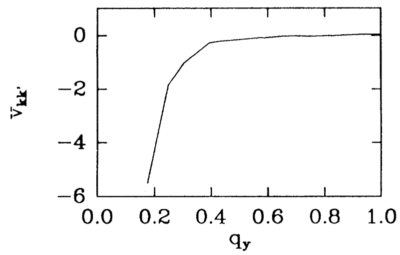

FIG. 2. Effective interaction between quasiparticles on the Fermi surface $v(k,k')$ plotted vs momentum transfer $q=k-k'$ for $\Delta_0=3.5t_{pd}$, $V=1.75t_{pd}$, and $\delta=0.2$. The quasiparticles have equal x component of the momentum ($k_x=k_x'$) and opposite component in the y direction ($k_y=-k_y'$).

that is, r_0 jumps to zero as $\Delta_0 \equiv \varepsilon_p^0 - \varepsilon_d^0$ approaches $\Delta_{0c}(V)$. The mean-field equations have three solutions in some range of parameters. This signals the CT instability of the system. However, since this mode is coupled to the density fluctuations, phase separation also takes place.

Having established the region of the phase diagram which is free from instabilities, we turn to the study of the effective interaction between the quasiparticles. For $N=\infty$ the quasiparticles are given by the eigenvectors of H in Eq. (2c) which is diagonalized by a unitary transformation $\Phi_{k,\alpha}=\sum_\beta U_{\alpha\beta}(k)\Psi_{k,\beta}$, where we introduced three-component rotations for the orbitals $\Psi_{k,\alpha}=(ip_{k,x}, ip_{k,y}, d_k)$ to get the quasiparticle operators $\Phi_{k,\alpha}$. In the new basis $H_{\rm MF}=\sum_{k,\alpha}E_\alpha(k)\Phi^\dagger_{k,\alpha}\Phi_{k,\alpha}$. The quasiparticles interact with the fluctuations of the four-component field $A^\mu=(\delta r,\delta\lambda,\delta X,\delta Y)$ via

$$H_{\rm int}=\sum_{k,q,\sigma}\Psi^\dagger_{k+q\sigma/2}\Lambda^\mu(k,q)\Psi_{k-q\sigma/2}A^\mu(q)$$
$$=\sum_{k,q,\sigma}\Phi^\dagger_{k+q\sigma/2}\tilde\Lambda^\mu(k,q)\Phi_{k-q\sigma/2}A^\mu(q)\,. \qquad (4)$$

The 3×3 vertices $\Lambda^\mu(k,q)$ can be read off Eqs. (2), and the quasiparticle vertices $\tilde\Lambda^\mu_{\alpha\beta}(k,q)$ are defined as

$$\tilde\Lambda^\mu(k,q)=U(k+q/2)\Lambda^\mu(k,q)U^\dagger(k-q/2)\,.$$

The propagators of the A field are given by

$$D^{\mu,\nu}(q)=\langle A^\mu(q)A^\nu(-q)\rangle=[2NB^{\mu,\nu}+N\Pi^{\mu,\nu}(q)]^{-1}$$

with

$$\Pi^{\mu,\nu}(q)=\sum_{k,\alpha,\beta}\frac{f(E_\alpha(k+q/2))-f(E_\beta(k-q/2))}{E_\alpha(k+q/2)-E_\beta(k-q/2)}\tilde\Lambda^\mu_{\alpha\beta}(k,q)\tilde\Lambda^\nu_{\beta\alpha}(k,-q)\,. \qquad (5)$$

The matrix $B^{\mu,\nu}$ has zeros everywhere except for the elements $B^{1,1}=r_0^2\lambda_0$, $B^{1,2}=B^{2,1}=ir_0^2$, $B^{3,3}=B^{4,4}=1/V$. With this notation one can write the static effective interaction among the quasiparticles in the Cooper channel as

$$v(k,k')=-\sum_{\mu,\nu}\tilde\Lambda^\mu\left(\frac{k+k'}{2},q=k-k'\right)D^{\mu,\nu}(q=k-k')\tilde\Lambda^\nu\left(-\frac{k+k'}{2},q=k'-k\right)$$
$$+\tilde\Lambda^\mu\left(\frac{k-k'}{2},q=k+k'\right)D^{\mu,\nu}(q=k+k')\tilde\Lambda^\nu\left(\frac{k'-k}{2},q=-k-k'\right)\,. \qquad (6)$$

A typical shape of the effective interaction among the quasiparticles on the Fermi surface is shown as a function of the momentum transfer in Fig. 2. Notice that the interaction is attractive for a wide region of q. The superconducting coupling constants are calculated as Fermi-surface averages [21],

$$\lambda_l=\frac{\int\int dk\,dk'\,\delta(E(k)-\mu)\delta(E(k')-\mu)g_l(k)v(k,k')g_l(k')}{\int dk\,\delta(E(k)-\mu)g_l(k)^2}\,. \qquad (7)$$

TABLE I. s- and d-wave coupling constants for $V=1.75t_{pd}$ and various values of Δ_0 and δ.

Δ_0 \ δ	0.1	0.15	0.2	0.25	0.3	0.35	0.4	0.45	0.7
					s-wave				
1.6			0.5226		0.3470		0.216		
1.65		0.5615		0.3967		0.2606		0.1299	
1.76									−0.24
3.5	0.729		−1.1226		−0.2413		−0.082		
					d-wave				
1.6			−0.037		−0.092		−0.111		
1.65		−0.074		−0.121		−0.139		−0.137	
1.76									−0.162
3.5	0.012		−2.623		−0.902		−0.574		

We studied Fermi-surface averages with

$$g_0(k) = \cos(k_x) + \cos(k_y),$$

$$g_1(k) = \cos(k_x) - \cos(k_y).$$

The λ_0 and λ_1 couplings are found to be generally attractive near the phase-separation region. They are tabulated in Table I for $V = 1.75t_{pd}$.

The previous analysis indicates the existence of s-wave pairing in specific regions of model (2) in the limit of large N near phase separation. To obtain a substantial attraction for realistic values of V (several studies estimate $V < V_c$) the system has to operate close to the metal-charge-transfer-insulator transition. This feature is certainly present in the copper-oxygen system and absent in the weak-coupling approach.

We want to warn that, since pairing appears near phase separation, it is important to understand if and how our results will be modified by the presence of the long-range Coulomb interaction. This requires the introduction of a dynamical screening and a subsequent solution of the Eliashberg equations. It is clear, however, from this and earlier work [2-5] that the three-band model with Cu-O repulsion has physics, in the metallic state, which is different from the one-band repulsive Hubbard model.

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[19] An analogous relation between phase separation and superconductivity appears in the Kondo-like spin-hole-coupled model as shown in Ref. [17].

[20] The paths for λ and Y in the functional integral have to be deformed away from the real axis to meet the saddle-point values.

[21] While a negative value of λ_l indicates the presence of a Cooper instability, the solution of the full gap equation would require a suitable average over the entire Brillouin zone (BZ). This last procedure was employed in the weak-coupling approach [7]. However, since in Ref. [7] no instability was revealed when Fermi-surface averages were performed (the instability appeared only after the entire BZ averages), we expect that a similar procedure in our strong-coupling regime would result in a strengthening of the superconducting instability.