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Mean-field theory of high-T_c superconductivity: The superexchange mechanism

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We develop the simplest mean-field theory of an extended Hubbard model in the limit of a large intrasite Coulomb interaction, concentrating on the possibility of superconductivity induced by the superexchange interaction and weakened by the intersite Coulomb repulsion. We calculate the critical temperature and the coherence length as a function of filling, as well as the temperature dependence of the magnetic susceptibility and specific heat. Finally, we comment on the physics of the insulating state at half filling, and mention the probable effects of fluctuations.

Following the pioneering work of Bednorz and Müller[1] and the subsequent discovery of superconductivity above liquid-nitrogen temperatures in the quarternary compound $(Y_{1-x}Ba_x)_2CuO_{4-y}$,[2] a number of theoretical ideas have been proposed to explain this remarkable discovery. Theoretical arguments based on the conventional electron-phonon mechanism appear to rule out critical temperatures much larger than 40 K.[3] Consequently, any other mechanism involving conventional phonons, such as Bose condensation of bipolarons,[4] will most likely lead to a smaller T_c. What remains is the possibility that the superconductivity is due to an unconventional mechanism mediated by Coulomb interactions, perhaps enhanced by the presence of phonons.

One such possibility, discussed by Varma, Schmitt-Rink, and Abrahams,[5] is pairing due to charge-transfer excitations (excitons) $Cu^{2+}O^{2-} \rightarrow Cu^{+}O^{-}$, which dominate for sufficiently small intrasite Coulomb repulsion. This covalent-metallic picture should be contrasted with the modest covalency and large intrasite Coulomb repulsion characteristic of almost ionic (semi)conductors. The latter underlies Anderson's resonating valence bond (RVB) mechanism,[6] in which the superconductivity occurs as a result of spin correlations induced by superexchange between electrons on nearest-neighbor Cu sites.[7]

In this paper we further examine this possibility. We develop a simple mean-field theory of the Hubbard Hamiltonian, which describes spin-$\frac{1}{2}$ fermions on a two-dimensional (2D) square lattice, interacting through intrasite (U) and nearest-neighbor (V) Coulomb repulsions:

$$H = -t \sum_{\substack{(i,m)\\ \sigma}} c_{i\sigma}^{\dagger} c_{i+m,\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow} + \frac{V}{2} \sum_{(i,m)} (n_i - 1)(n_{i+m} - 1) , \tag{1}$$

where $n_{i\sigma} = c_{i\sigma}^{\dagger} c_{i\sigma}$ is the number operator for electrons of spin $\sigma (= \pm 1/2)$ at site i, $n_i = \sum_\sigma n_{i\sigma}$ is the number density, and (i,m) implies unrestricted summation over the nearest neighbors (of site i).

Our discussion is based on the band-structure calculations of Mattheis,[8] which suggest that the relevant physics can be modeled in terms of two-dimensional bands spanned by the O $2p$ and Cu $3d$ orbitals. It is worth noting that the fermion operators $c_{i\sigma}$ in (1) represent electrons localized on the Cu sites. These are the "quasiparticle" operators introduced by Anderson[9] in his theory of superexchange in transition-metal salts, and have the virtue of eliminating explicit reference to ligand electrons. Here we will make use of the quasiparticle picture away from half filling, and we thus rely on the large electron affinity of the oxygen. The effective interaction parameters t, U, and V can be crudely estimated by extrapolating Anderson's discussion of transition-metal salts to our case: We obtain the values $t \sim 0.25$–0.5 eV, $U \sim 3$–4 eV, and $V \sim 1$–2 eV.

Below we assume the large-U limit, and make use of a canonical transformation to eliminate states with doubly occupied sites,[10] yielding

$$\begin{aligned} H = & -t \sum_{\substack{(i,m)\\ \sigma}} b_i f_{i\sigma}^{\dagger} f_{i+m,\sigma} b_{i+m}^{\dagger} + \frac{J}{2} \sum_{(i,m)} (\boldsymbol{\sigma}_i \cdot \boldsymbol{\sigma}_{i+m} - n_i n_{i+m}) \\ & - J \sum_{\substack{(i,m,m')\\ \sigma \\ m \neq -m'}} b_i (f_{i,\sigma}^{\dagger} f_{i+m,-\sigma}^{\dagger} f_{i+m,-\sigma} f_{i+m+m',\sigma} - f_{i,\sigma}^{\dagger} f_{i+m,-\sigma}^{\dagger} f_{i+m,\sigma} f_{i+m+m',-\sigma}) b_{i+m+m'}^{\dagger} \\ & + \frac{V}{2} \sum_{(i,m)} (n_i - 1)(n_{i+m} - 1) + \sum_i \lambda_i \Big(b_i^{\dagger} b_i + \sum_\sigma f_{i,\sigma}^{\dagger} f_{i,\sigma} - 1\Big) - \mu \sum_{i\sigma} n_{i\sigma} , \end{aligned} \tag{2}$$

where $J = 2t^2/(U-V)$ is the superexchange interaction. Here we have introduced the representation of the fermion operators $c_{i,\sigma}(c_{i,\sigma}^{\dagger})$ acting on states with no double occupancy, $c_{i,\sigma} = b_i^{\dagger} f_{i,\sigma}$, in terms of fermions, $f_{i,\sigma}(f_{i,\sigma}^{\dagger})$, and auxiliary Bose fields, $b_i(b_i^{\dagger})$. The latter keeps track of the occupancy at a given site, with $b_i^{\dagger} b_i = 0$ for occupied and $b_i^{\dagger} b_i = 1$ for unoccupied sites. The condition of no double occupancy (which implies that $\sum_\sigma f_{i,\sigma}^{\dagger} f_{i,\sigma}$ is at most unity) is incorporated through the constraints $b_i^{\dagger} b_i + \sum_\sigma f_{i,\sigma}^{\dagger} f_{i,\sigma} = 1$, enforced independently at each site by the Lagrange multipliers, λ_i. Above, $\boldsymbol{\sigma}_i = \sum_{\sigma\sigma'} f_{i,\sigma}^\dagger \boldsymbol{\tau}_{\sigma\sigma'} f_{i,\sigma'}$ and $n_i = \sum_\sigma f_{i,\sigma}^\dagger f_{i,\sigma}$ are the spin and number densities at site i; the sums over m and m' again correspond to unrestricted sums over nearest neighbors. The first and third terms in Eq. (2) represent, respectively, the hopping of electrons, and of pairs of electrons, away from half filling, while the second term gives rise to the superexchange interaction.

We begin by considering the simplest mean-field theory of (2), based on two approximations. First of all, the auxiliary boson is replaced by a filling-dependent c number, while the Lagrange multiplier is taken to be uniform ($\lambda_i = \lambda$).[11] In this particular case the appropriate procedure leads to a replacement of

$$b_i \rightarrow b_i \approx b(1-n/2)^{-1/2} = [2\delta/(1+\delta)]^{1/2} \equiv \tilde{\delta}^{1/2}$$

(the last equality follows from the mean field form of the constraint, $b^2 + \sum_{i\sigma}\langle f_{i,\sigma}^\dagger f_{i,\sigma}\rangle/N = b^2 + n = 1$), where $n \equiv \langle n_i \rangle \equiv 1-\delta \le 1$. This replacement is consistent with the requirement that the mean-field theory applied to a fully spin-polarized partially filled band leads to no mass renormalization.[11] We note that treating the bosons as c numbers amounts to assuming the Bose condensation of the b's. Our second assumption is that, for any value of the filling, the order parameter takes the form

$$\tilde{\Delta}(T,\delta) \equiv \sum_{i,m} \langle b_i^\dagger f_{i,\downarrow} f_{i+m,\uparrow} b_{i+m}^\dagger \rangle / zN \approx \tilde{\delta} \sum_{i,m} \langle f_{i,\downarrow} f_{i+m,\uparrow} \rangle / zN \equiv \tilde{\delta}\Delta \, ;$$

$\tilde{\Delta}$ represents the pairing of the physical electrons with opposite spins on nearest-neighbor sites, while Δ can be interpreted as the amplitude of preexisting pairs.

We now proceed with the Hartree-Fock factorization of (2), which leads to the following BCS-like Hamiltonian:

$$H_{\mathrm{BCS}} = \sum_{k,\sigma} \xi(k) f_{k,\sigma}^\dagger f_{k,\sigma} - I \sum_k \tau(k) (\Delta^* f_{-k,\downarrow} f_{k,\uparrow} + \mathrm{H.c.}) \, , \qquad (3)$$

where $\tau(k) = 2[\cos(k_x a) + \cos(k_y a)]$ (a is the lattice spacing), $\xi(k) = -\tilde{\delta} t \tau(k) - \mu + \lambda$, and $I \equiv 4J(1+3\tilde{\delta}) - V$. Both hopping terms—for single electrons and for pairs—are linear in the concentration of bosons, since both processes require the presence of a *single* empty site. It is important to note that the fully screened Coulomb repulsion V must appear in the effective interaction I on equal footing with J, since in this case, in contrast with BCS theory, both the pairing mechanism *and* the screening of V occur on the same frequency scale.

The Hamiltonian (3) is diagonalized by the Bogoliubov canonical transformation by introducing the quasiparticle operators, $\gamma_{k0} = u_k f_{k,\uparrow} - v_k f_{-k,\downarrow}^\dagger$ and $\gamma_{k1} = u_k f_{-k,\downarrow} + v_k f_{k,\uparrow}^\dagger$; in thermal equilibrium u_k and v_k can be chosen as real, and are given by

$$u_k^2 = [1 + \xi(k)/E(k)]/2, \quad v_k^2 = [1 - \xi(k)/E(k)]/2 \, ,$$

where $E(k) = [\xi^2(k) + I^2\Delta^2\tau^2(k)]^{1/2}$ is the quasiparticle energy; without loss of generality we take $u_k > 0$, and $\mathrm{sgn}(v_k) = \mathrm{sgn}[\tau(k)]$. The self-consistent equations for the gap parameter Δ and the chemical potential $\tilde{\mu}(=\mu-\lambda)$ are then obtained from the defining relations, $\Delta = \sum_k \tau(k) \langle f_{-k,\downarrow} f_{k,\uparrow} \rangle / z$, $n = \sum_{k,\sigma} \langle f_{k,\sigma}^\dagger f_{k,\sigma} \rangle = 1 - \delta$:

$$2z/I = \sum_k \frac{\tau^2(k)}{E(k)} \tanh\left[\frac{\beta E(k)}{2}\right] \, ,$$
$$\delta = \sum_k \frac{\xi(k)}{E(k)} \tanh\left[\frac{\beta E(k)}{2}\right] \, . \qquad (4)$$

We can now proceed to a brief discussion of our results.

(1) The critical temperature. T_c [obtained by setting $\Delta(T_c) = 0$ in Eq. (4)], starts out with a finite value $T_c(\delta=0) \approx J - V/4$ (see Fig. 1). Away from half filling, T_c first increases to a maximum determined by the competition between pair hopping, which is linear in $\tilde{\delta}$, and the pair breaking due to single-particle hopping, an effect of order $\tilde{\delta}^2$ for small $\tilde{\delta}$. T_c then decreases rapidly until superconductivity is destroyed through the unbinding of pairs for $\tilde{\delta} \sim I/(2zt)$, where the binding energy I becomes of the order of the effective bandwidth $2zt\tilde{\delta}$. For higher values of $\tilde{\delta}$ the gap equation yields nontrivial solutions for T_c, not shown in Fig. 1. These solutions are, however, ignored as they are associated with values of $[4t + \tilde{\mu}(\delta)] < I$ for which true bound states split off below the bottom of the band. The critical temperature in this regime is always smaller than that for $[4t + \tilde{\mu}(\delta)] \approx I$ and is expected to be a smoothly decreasing function with increasing $\tilde{\delta}$; its value is controlled by the center-of-mass degrees of freedom of the pairs, and is outside the scope of our BCS-like mean-field theory.

We note that the finite value of T_c at $\delta = 0$ is an artifact of our mean-field approximation. First, unlike the gap parameter Δ the physical order parameter $\tilde{\Delta} \approx \tilde{\delta}\Delta$ vanishes for $\delta = 0$, even within this simple mean-field theory. More importantly, true off-diagonal long-range order implies

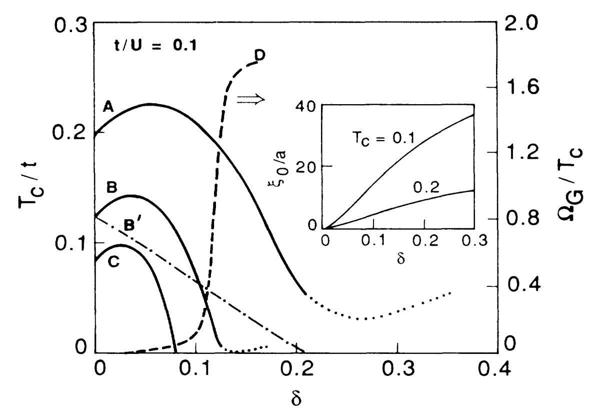

FIG. 1. Critical temperature (T_c) vs δ for $t/U = 0.1$, with $t/V = \infty$, 3.0, 2.0 for curves A, B, and C, respectively. Excitation gap (Ω_G) vs δ for $t/U = 0.1$ and $t/V = 3.0$ (curve D). Inset: $T = 0$ coherence length vs δ for $T_c = 0.1$ and $T_c = 0.2$. The dotted segments of curves A and B denote the non-BCS-like regime $4t + \tilde{\mu}(\delta) < I$. Curve B' represents T_c vs δ for d-wave-like pairing with $t/U = 0.1$ and $t/V = 3.0$.

long-range coherence of the Bose fields b in

$$\tilde{\Delta}(T,\delta)=\sum_{i,m}\langle b_i^\dagger f_{i,\downarrow} f_{i+m,\uparrow} b_{i+m}^\dagger\rangle/zN\ ,$$

which is clearly absent at $\delta=0$ due to the "incompressibility" constraint, $b_i^\dagger b_i+\sum_\sigma f_{i,\sigma}^\dagger f_{i,\sigma}=1$. Close to half filling T_c is determined by the Bose fluctuations. These can be very crudely incorporated into our mean-field theory by explicitly accounting for the thermal depletion of the b condensate, in which case $\tilde{\Delta}(T,\delta)\propto b_0^2(T,\delta)\Delta(T,\delta)$ (b_0^2 is the b condensate fraction). Thus, near $\delta=0$, T_c is limited by the condensation temperature of the bosons, $T_{BE}\sim 8\tilde{\delta}^\nu J\Delta^2a^2$, $\nu\sim\frac{2}{3}$ in 3D, whereas $\nu\sim 1$ in 2D; with increasing $\tilde{\delta}$, $T_c(\delta)$ is expected to reach a maximum for $\tilde{\delta}<I/2zt$. This point of view is, however, too naive: The off-diagonal long-range order of the superconducting state is associated with a particular value of a *single* phase variable, namely, that of the physical order parameter $\tilde{\Delta}(T,\delta)$, rather than *two* phases (one of the superconducting gap and one of the b condensate) as implied by the above mean-field picture. These two points of view are reconciled by a precise treatment of the double occupancy constraint, which in turn restores the gauge symmetry with respect to one combination of the phases.

The above discussion applies to the regime in which (a) J is sufficiently large that T_c is nonzero, and (b) the BCS-like pairing theory is valid, i.e., $4t+\tilde{\mu}(\delta)>I$. We note that the results are extremely sensitive to small changes in the parameters. In fact, for the most realistic estimates quoted above, we find that the second condition is not satisfied, thus requiring a treatment of the crossover between Cooper (i.e., BCS-like) pairing and Bose condensation, a regime outside the scope of this paper. More importantly, for the (t and U) parameter values discussed above we find that superconductivity survives only for unrealistically small values of V, suggesting that even within the mean-field theory help from another attractive interaction (e.g., phonons) may be required to fully stablize the superexchange mechanism. For the sake of illustration, in the figures we have chosen parameter values consistent with both (a) and (b) for the range of fillings specified in the captions.

(2) Coherence length. The absence of coherence in the insulating phase is further substantiated by a calculation of the $T=0$ coherence length ξ_0 based on the usual derivation of the Ginzburg-Landau free energy. As seen in Fig. 1, ξ_0 vanishes at half filling, as expected in the case of the incompressible ($F_0^s=\infty$) Fermi liquid[12] implied by our mean-field theory. Note that ξ_0 is a measure of the correlation between *different* pairs but that, in the contrast to BCS theory, it does not coincide with the extent of a *single* pair (here taken to be of the order of the lattice spacing).

(3) Excitation spectrum and tunneling density of states. At half filling, our mean-field theory gives a realization of state discussed by Anderson.[7] The Bogoliubov quasiparticle excitations define a "pseudo-Fermi surface" $E(k)=I\Delta|\tau(k)|=0$ across which $\tau(k)$ changes sign. In close agreement with Anderson's ideas,[6] the low-lying excited states with a fixed average number of particles are of the form $|k',s';k,s\rangle\equiv\gamma_{k',s'}^\dagger\gamma_{k,s}^\dagger|G\rangle$ ($s,s'=0,1$), and correspond to the gapless excitations of a Fermi liquid. The nature of the excitations can be further understood by considering the effect of an arbitrary external potential on $|G\rangle$. More precisely, the coherence factors associated with the coupling of these excitations to external density (or charge) and spin fluctuations are $(u_{k'}v_k+v_{k'}u_k)$ and $(u_{k'}v_k-v_{k'}u_k)$, respectively. Since at half filling $v_k=\text{sgn}[\tau(k)]u_k=\pm 1/\sqrt{2}$, it is easy to see that whenever k and k' are "on the same or opposite sides of the pseudo-Fermi surface" [i.e., $\text{sgn}\tau(k)=\text{sgn}\tau(k')$ or $\text{sgn}\tau(k)=-\text{sgn}(k')$] the states $|k',s';k,s\rangle$ represent spinless charge excitations or chargeless spin excitations, respectively. The fact that some excited states carry charge even in the limit of infinite U is easily understood since within our mean-field theory the double occupancy constraint is only satisfied on the average, rather than independently at each site. A proper treatment of the constraints will eliminate all charge fluctuations, but in that case the precise nature of the spin excitations remains to be determined. We have also calculated the single-particle density of states, which is gapless for $\delta=0$ as expected. Away from $\delta=0$ an excitation gap, $\Omega_G=I\Delta|\tilde{\mu}|/(I^2\Delta^2+\tilde{\delta}^2t^2)^{1/2}$, opens at the true Fermi surface, which no longer coincides with the "pseudo-Fermi surface." An important quantity, accessible in tunneling experiments, is the ratio Ω_G/T_c, which we find to increase with $\tilde{\delta}$ from zero to a value of order of unity for $\tilde{\delta}\sim I/(2zt)$.

(4) Magnetic susceptibility and specific heat. The spin susceptibility and specific heat were calculated from the expressions appropriate for a gas of noninteracting quasiparticles (for illustration we show plots of the specific heat in Fig. 2). In the "normal state" (i.e., $\Delta=0$), by increasing δ from zero at a fixed temperature T, we find a crossover from localized spins fluctuations (at $\delta=0$), to a Boltzmann gas with effective mass $m_{\text{eff}}\sim 1/\tilde{\delta}$ for $\tilde{\delta}<T/2zt$, and finally to a degenerate noninteracting Fermi gas when $2zt\tilde{\delta}\sim T$. Consistent with the discussion above, for finite Δ and $\delta=0$, the thermodynamics is essentially that expected of a Fermi liquid with an effective mass $m^*(\delta=0)\propto(I\Delta a^2)^{-1}$, and crosses over to that of a

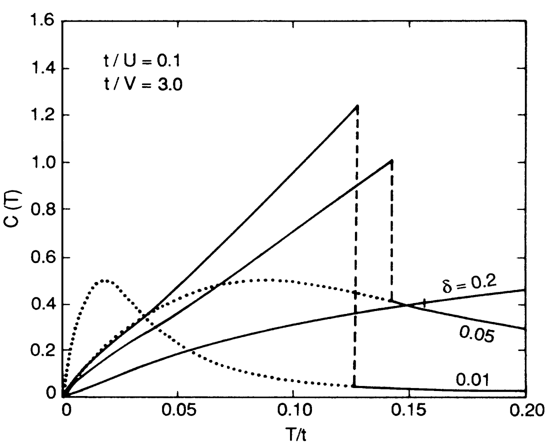

FIG. 2. Specific heat vs T for $t/U=0.1$, and $t/V=3.0$ for $\delta=0$, 0.05, 0.2. Dotted lines represent normal behavior with $\Delta=0$.

collection of localized spins for $T > T_c(\delta=0)$. In the superconducting state $(\Delta\neq0,\delta\neq0)$, the physics is qualitatively similar to that at half filling (for $\Delta\neq0$), with the associated Fermi-liquid-like behavior for both susceptibility and specific heat. The exponential falloff associated with the gap in the excitation spectrum is visible only at the lowest temperatures, since within the mean-field theory for most fillings $T_c > \Omega_G$. We further note that for reasonable values of the parameters the effective bandwidth defined by the spin excitations, of order $zI\Delta$, does not imply an appreciable enhancement of either Sommerfeld's γ or the magnetic susceptibility, as one might have expected near a Mott metal-insulator transition.

Above we have discussed the simple mean-field theory of high-T_c superconductivity based on the high U/t limit of an extended Hubbard model. A number of important problems remain open. We have ignored the antiferromagnetic long-range order predicted by the model at half filling. This is sensible only provided the antiferromagnetism is destroyed for sufficiently small δ. The interplay between superconductivity and antiferromagnetism will be considered in a further publication.[13] We have also not addressed the crucial problem of the stability of our mean-field theory with respect to fluctuations, a point particularly worrisome in two dimensions. Order parameter fluctuations, thermal phonons,[14] as well as inelastic electron-electron scattering, should lead to a decrease of T_c. Since the excitation gap is most likely not drastically affected, the ratio Ω_G/T_c will also be increased in the direction of better agreement with experiment.[15] Finally, as already mentioned above, a correct treatment of the (auxiliary) boson fluctuations is crucial in enforcing the "no double occupancy" constraint, which in turn determines the nature of the physics close to half filling. It is, in fact, not clear that a systematic treatment of the Hubbard model with purely repulsive interactions can lead to a stable superconducting state. We expect, however, that phonons or another exclusively attractive interaction acting together with the superexchange mechanism will lead to a superconducting state, qualitatively similar to that described by the above mean-field theory. In the final stages of writing, we received a paper by Baskaran, Zou, and Anderson[16] which overlaps considerably with the present work, and reaches similar conclusions.

Note added. We have also considered the possibility of d-wave pairing with an order parameter proportional to $d(k)=2[\cos(k_xa)-\cos(k_ya)]$, the only representation consistent with the nearest-neighbor hopping assumed above. (We note that odd parity pairing does not occur in this model.) The resulting $T_c(\delta)$ is indicated in Fig. 1 as curve B′. We note that for finite δ, $T_c^d < T_c^s$, for a large range of fillings, in contrast to an independent calculation of Kotliar.[17] The latter author ignored the pair hopping term which is important for the stabilization of the s-wave-like state. Kotliar makes, however, the important observation that, since the s- and d-wave-like states are degenerate at $\delta=0$ (at least in the absence of phonons), a mixed state will lead to a gap in the excitation spectrum.

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