Cooper Instability in the Presence of a Spin Liquid

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We examine a new two-band model which allows the coexistence of a spin liquid and conduction sea. Exchange coupling between the two fluids leads to Kondo-spin compensation of the spin liquid, generating bound states between the spin liquid and conduction electrons. When this occurs, the spin and charge degrees of freedom decouple, forming a superconductor of a novel kind.

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A central issue in the problem of oxide superconductivity is the nature of the coupling between the copper d spins and the oxygen p holes.[1-4] A commonly held view is that for the low-energy physics, the p holes become bound into singlet pairs with the d spins, forming holes whose motion is described by an almost half-filled Hubbard model with Heisenberg interactions.[5-8] In this paper we examine the idea that the spin of the p hole is actually a relevant degree of freedom, and we explore the possibility that the process of d-spin compensation by p holes is modified in a nontrivial way by the presence of an almost antiferromagnetic (AFM) background.

Beginning with a general two-band Hubbard model for d and p holes in a copper oxide plane, we assume that the d holes are localized, and that the repulsive interaction U_{dd} between d holes is sufficiently large to allow the high-lying d-charge fluctuations to be integrated away. This generates a model describing localized d spins, mutually interacting through a nearest-neighbor superexchange interaction, coupled to a dilute conduction sea of p holes via *nonlocal* AFM exchange interactions,[3,6,9]

$$H=J_H\sum_{(i,j)}\mathbf{S}_i\cdot\mathbf{S}_j+J_K\sum_j\mathbf{S}_j\cdot p^\dagger_{j\sigma}\mathbf{S}_{\sigma\sigma'}p_{j\sigma'}+\sum_{\mathbf{k}\sigma}\epsilon_{\mathbf{k}}p^\dagger_{\mathbf{k}\sigma}p_{\mathbf{k}\sigma}. \quad (1)$$

Here, $\mathbf{S}_i=d^\dagger_{i\alpha}[\mathbf{S}]_{\alpha\beta}d_{i\beta}$ represents a d spin at site i and $p^\dagger_{\mathbf{k}\sigma}$ creates a Bloch wave composed of the $p\sigma$ orbitals. The operator

$$p^\dagger_{j\sigma}=\sum_{\mathbf{k}}\gamma_{\mathbf{k}}e^{-i\mathbf{k}\cdot\mathbf{R}_j}p^\dagger_{\mathbf{k}\sigma}$$

creates a p hole with the symmetry of a $d_{x^2-y^2}$ state at site j.[6] The function $\gamma_{\mathbf{k}}=[1-(c_x+c_y)/2]^{1/2}$, where $c_l=\cos k_l$, $l=x,y$, is the form factor of a p Wannier state with $d_{x^2-y^2}$ symmetry. We take the Cu-Cu distance to be unity. We also include the weak dispersion $\epsilon_{\mathbf{k}}$ of the p holes generated by potential scattering off the d spins and overlap of p orbitals.

We study the model in the limit where $J_K,J_H\gg D$ so that the p-band dispersion can be ignored ($\epsilon_{\mathbf{k}}=-\mu$). This limit is of special interest to cuprate superconductivity and will be referred to as the "dual exchange model." Note that the p charge in a given σ orbital is *not* conserved since the Kondo interaction is nonlocal $\gamma_{\mathbf{k}}\neq 1$, and so the p holes are still mobile. We treat the ratio J_K/J_H as a variable parameter measuring the strength of p-d coupling. When $J_K\gg J_H$, the p holes bind rigidly to the d spins, forming mobile singlets that behave as holes in a half-filled infinite-U Hubbard model.[6,9]

At low hole concentration x and finite temperature, d-spin compensation is not complete. While the singlet binding energy of a p hole is of order J_K, the energy per unit cell is merely xJ_K and when $xJ_K\lesssim T\lesssim J_H$, triplet excitations of the p-d pairs will be present. In this paper we present calculations that suggest that the compensation of the d spins by the p holes is both inhibited and modified by the AFM background, so that when it occurs, it is accompanied by the development of superconductivity.

Our model is defined within the subspace constrained by the (Gutzwiller) requirement $n_d=1$ at each site. This constraint manifests itself as a local SU(2) gauge invariance of the Heisenberg spin operator $\mathbf{S}_j$,[5] a feature that can be exploited in a path integral treatment to impose the constraint.[10] In terms of Nambu spinors for the d holes $d^\dagger_j=(d^\dagger_{j\uparrow},d_{j\downarrow})$, the local SU(2) gauge transformation is written $d_j\to g_jd_j$, where $g_j=\exp(i\mathbf{W}_j\cdot\boldsymbol{\tau})$ is a unitary 2D matrix.

The partition function for our model is $Z=\mathrm{Tr}[P_Ge^{-\beta H}]$ where P_G is the Gutzwiller projection for one d spin per site. Rewriting it as an integral over the SU(2) group[5]

$$P_G=\prod_j\int\frac{d^3W_j}{8\pi^2}\hat{g}_j, \quad (2)$$

where $\hat{g}_j=\exp[id^\dagger_j(\mathbf{W}_j\cdot\boldsymbol{\tau})d_j]$ permits us to incorporate the constraint into a Lagrangian $\mathcal{L}=\mathcal{L}_0+H$,

$$\mathcal{L}_0=\sum_{\mathbf{k}}p^\dagger_{\mathbf{k}}\partial_\tau p_{\mathbf{k}}+\sum_j d^\dagger_j(\partial_\tau-iW_j)d_j,$$

where $W_j=\mathbf{W}_j\cdot\boldsymbol{\tau}$, and we have also introduced Nambu spinors for the p electrons. Finally, factorizing the interactions, we obtain

$$Z=\int\mathcal{D}[d,p;W,V,U]\exp-\left(\int_0^\beta(\mathcal{L}_0+H)d\tau\right),$$
$$H=\sum_{\mathbf{k}}\epsilon_{\mathbf{k}}p^\dagger_{\mathbf{k}}\tau_3p_{\mathbf{k}}+\sum_{(i,j)}[d^\dagger_iU_{ij}d_j+\mathrm{H.c.}]+\frac{1}{J_H}\mathrm{Tr}[U^\dagger_{ij}U_{ij}]+\sum_i[d^\dagger_iV_ip_i+\mathrm{H.c.}]+\frac{1}{J_K}\mathrm{Tr}[V^\dagger_iV_i], \quad (3)$$

where the matrices U_{ij} and V_j are proportional to unitary matrices, and $U_{ji}=U_{ij}^\dagger$. Our model has the following time-dependent SU(2) gauge invariance:

$$\begin{aligned} &d_j \to g_j d_j, \quad V_j \to g_j V_j , \\ &U_{ij} \to g_i U_{ij} g_j^{-1}, \quad W_j \to g_j (W_j + i\partial_\tau) g_j^{-1} , \end{aligned} \tag{4}$$

associated with the absence of d-charge fluctuations. There is also the usual electromagnetic U(1) gauge symmetry associated with the charged p holes, introduced via $\gamma_{\mathbf{k}} \to \gamma_{\mathbf{k}-e\mathbf{A}}$ and $\epsilon_{\mathbf{k}} \to \epsilon_{\mathbf{k}-e\mathbf{A}}$, where $\mathbf{A}$ is the electromagnetic field.

We interpret our SU(2) Lagrangian as describing electrons propagating in a fluctuating pairing field generated by the AFM interactions. The V field describes the compensation of the d spins by the p holes, and as in the case of heavy-fermion systems,[11] we expect condensation of this field to preempt formation of a Néel phase.

In the mean-field (MF) approximation, the fields U_{ij} and V_j are treated as classical variables, which only need to be translationally invariant up to a gauge transformation. Actually, each choice of saddle-point solution for U_{ij} and V_j is a point on the orbit of all gauge equivalent solutions. Here, we partially fix the gauge with the choice

$$\begin{aligned} &U(\mathbf{R}_i+\hat{\mathbf{l}},\mathbf{R}_i) = U_l = -i\Delta e^{i\mathbf{W}_l\cdot\tau} , \\ &V_j = V_0 e^{i\alpha\cdot\tau} = V, \quad W_j = \mathbf{W}_0\cdot\tau , \end{aligned} \tag{5}$$

where $\mathbf{W}_l = \theta\hat{\mathbf{n}}_l$ and $\hat{\mathbf{n}}_l$ $(l=x,y)$ are unit vectors.

The MF Hamiltonian for our *Ansatz* is $H_{\mathrm{MF}} = \sum_{\mathbf{k}} \psi_{\mathbf{k}}^\dagger h_{\mathbf{k}} \psi_{\mathbf{k}}$, with the corresponding Green's function $\mathcal{G}_{\mathbf{k}}(i\omega_n) = (i\omega_n - h_{\mathbf{k}})^{-1}$, where

$$h_{\mathbf{k}} = \begin{bmatrix} \epsilon_{\mathbf{k}}\tau_3 & V_{\mathbf{k}}^\dagger \\ V_{\mathbf{k}} & W+U(\mathbf{k}) \end{bmatrix} , \tag{6}$$

$U(\mathbf{k}) = \sum_l U_l \exp(ik_l) + \mathrm{H.c.}$, and we defined $\psi_{\mathbf{k}}^\dagger = (p_{\mathbf{k}}^\dagger, d_{\mathbf{k}}^\dagger)$, $V_{\mathbf{k}} = V\gamma_{\mathbf{k}}$.

The MF free energy per unit cell,

$$F[U,V,W] = T \sum_{\mathbf{k},i\omega_n} \mathrm{Tr}\ln[\mathcal{G}_{\mathbf{k}}(i\omega_n)] + \frac{2V_0^2}{J_K} + \frac{4\Delta^2}{J_H} , \tag{7}$$

must then be stationary with respect to variations in U, V, and W, which generates three MF equations,

$$\begin{aligned} &0 = \langle d_i^\dagger \tau d_i \rangle = T \sum_{\mathbf{k},i\omega_n} \mathrm{Tr}[\tau \mathcal{G}_{\mathbf{k}}^{dd}(i\omega_n)] , \\ &U_l = J_H \langle d_{i+l} d_i^\dagger \rangle = -TJ_H \sum_{\mathbf{k},i\omega_n} \mathcal{G}_{\mathbf{k}}^{dd}(i\omega_n) e^{ik_l} , \\ &V = J_K \langle d_i p_i^\dagger \rangle = -TJ_K \sum_{\mathbf{k},i\omega_n} \gamma_{\mathbf{k}} \mathcal{G}_{\mathbf{k}}^{dp}(i\omega_n) , \end{aligned} \tag{8}$$

where the superscripts on $\mathcal{G}$ label the block components.

We now proceed to discuss our results. Two nontrivial "normal" phases and a superconducting phase occur within this class (see Fig. 1).

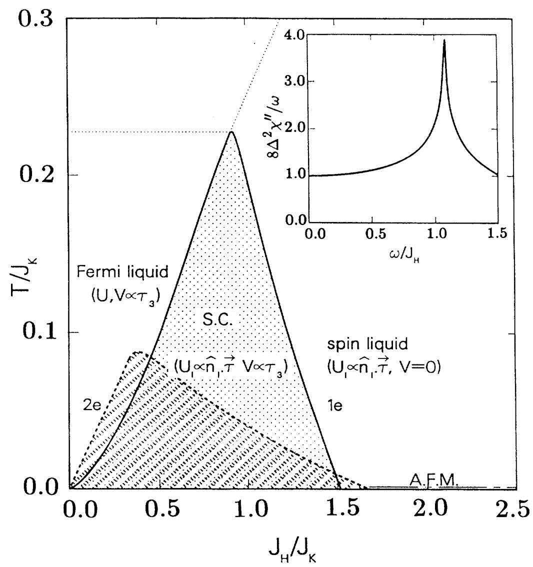

FIG. 1. Mean-field phase diagram, assuming zero p-band dispersion $\epsilon_{\mathbf{k}} = -\mu$. The chemical potential μ is adjusted to maintain the p-hole concentration x per unit-cell constant at (i) $x=1$ (solid line) and (ii) $x=0.05$ (dashed line), showing how T_c drops with hole density, broadening the range over which a spin-liquid to superconductor transition occurs. Dotted lines indicate the crossover to the spin-liquid and Fermi-liquid phases for the case $x=1$. "$2e$" and "$1e$" refer to the charge associated with the superconducting fluctuations. The dashed line labeled "AFM" indicates the *expected position of the AFM phase* in an uncompensated Heisenberg model. Inset: Calculated power spectrum of the AFM spin fluctuations in the "$s+id$" spin-liquid state $\chi''/\omega = \mathrm{Im}\chi(\mathbf{G},\omega)/\omega$.

Fermi-liquid phase.—$J_K/J_H \gtrsim 1$; $U=\Delta\tau_3$, $W_0=\lambda\tau_3$, $V=V_0\tau_3$ with wave function of the form

$$|\psi\rangle = P_G \prod_{\mathbf{k}n\sigma} (\alpha_{\mathbf{k}n} p_{\mathbf{k}\sigma}^\dagger + \beta_{\mathbf{k}n} d_{\mathbf{k}\sigma}^\dagger) |0\rangle .$$

In this phase, the p holes compensate the d *spins* ($V \neq 0$) forming a metallic phase with d-like quasiparticles, reminiscent of heavy-fermion metals.[11] This phase occurs at weak Heisenberg couplings, when compensation develops before strong AFM correlations have formed amongst the d spins. This phase is always unstable to superconductivity at low temperatures.

Doped spin-liquid phase.—$J_K/J_H \lesssim 1$; $V=0$, $W_0=0$, $U \neq 0$, $\theta=\pi/2$, $\hat{\mathbf{n}}_x \cdot \hat{\mathbf{n}}_y = 0$, where the d spins are uncompensated ($V=0$), but condense into a singlet state with strong local AFM correlations. The eigenvalues H_{MF} are $\epsilon_{\mathbf{k}}$ and $E_{\mathbf{k}} = 2\Delta(c_x^2+c_y^2)^{1/2}$ where

$$\Delta = \tfrac{1}{4} J_H \sum_{\mathbf{k}} (c_x^2+c_y^2)^{1/2} \tanh[\beta E_k/2] . \tag{9}$$

Were this phase to remain uncompensated down to zero temperature, then in the gauge $(\hat{\mathbf{n}}_x, \hat{\mathbf{n}}_y) = (\hat{\mathbf{x}}, \hat{\mathbf{y}})$, the MF ground state would have the form $|\Psi_{\mathrm{SL}}\rangle = P_G |\tilde{\Psi}_{\mathrm{SL}}\rangle$, where

$$|\tilde{\Psi}_{SL}\rangle = \prod_{k'<k_F,\sigma} p^\dagger_{k'\sigma} \prod_{\mathbf{k}} (1 - e^{-i\phi(\mathbf{k})} d^\dagger_{\mathbf{k}\uparrow} d^\dagger_{-\mathbf{k}\downarrow}) |0\rangle ,$$

with quasiparticle creation operator

$$a^\dagger_{\mathbf{k}} = (d^\dagger_{\mathbf{k}\sigma} - \mathrm{sgn}\sigma d_{-\mathbf{k}-\sigma} e^{-i\phi(\mathbf{k})})/\sqrt{2} ,$$

where $\tan\phi(\mathbf{k}) = c_y/c_x$. This phase occurs for large Heisenberg coupling, when the development of AFM d-spin correlations occurs before d-spin compensation. The d-spin component has been discussed by Affleck and Marston,[12] who called it a flux state, and also Kotliar,[8] who called it as an $s+id$ state.

While the MF description of this doped spin liquid is a form of resonating-valence-bond (RVB) state,[1] its application here differs in some important ways from its past use in one-band models. There are two fluids, which are decoupled only at the MF level. The low-lying neutral fermions appearing in the MF theory have a relativistic spectrum, $E_{\mathbf{q}+\mathbf{G}/2} = 2\Delta q + O(q^2)$, $\mathbf{G} = (\pm\pi, \pm\pi)$ with no Fermi surface. Beyond MF theory, the spin exchange between p holes and d spins generates inelastic scattering that is a major source of mobility for the p holes. However, without compensation of the d spins there is no common Fermi surface for the p and d fluids, and so p holes at the Fermi energy have finite lifetimes and this state is consequently a poor metal.

The RVB MF theory provides an approximate representation of the AFM background of d spins at finite temperature. Computing the spin-spin correlations at the mean-field level, we find there are gapless AFM spin fluctuations at $\mathbf{q} = (\pi,\pi)$ (see Fig. 1).[13] The short-range spin correlations predicted by this picture are similar to those in an undoped Heisenberg antiferromagnet. (For a single plaquet, the $s+id$ Gutzwiller state is the ground state of the Heisenberg model.[14] The magnetic correlation length diverges as a power rather than an exponential function of the inverse temperature, and so this does not provide a good description of the long-range AFM order that develops in an undoped Heisenberg antiferromagnet. However, since the coupling between the conduction and d spins is short ranged, this RVB picture is sufficient for a consideration of the spin-compensation process that preempts magnetism in the presence of doping. We now discuss the nature of the phase that develops.

Cooper instability.—The presence of the short-range AFM correlations severely modifies the motion of the p holes. First, consider the artificial case of an isolated p electron propagating in a spin-liquid background. Introduce a trial two-particle wave function $|\psi\rangle = P_G|\tilde{\psi}\rangle$ where

$$|\tilde{\psi}\rangle = \sum_{k>k_F} \eta_{\mathbf{k}} p^\dagger_{\mathbf{k}\sigma} a^\dagger_{-\mathbf{k}-\sigma} \mathrm{sgn}\sigma |\tilde{\Psi}_{SL}\rangle$$

describes a p hole paired with a neutral fermion. Let us write our MF Hamiltonian in a form reminiscent of BCS theory,

$$H_{MF} = \sum_{\mathbf{k}\sigma} E_{\mathbf{k}} a^\dagger_{\mathbf{k}\sigma} a_{\mathbf{k}\sigma} + \epsilon_{\mathbf{k}} p^\dagger_{\mathbf{k}\sigma} p_{\mathbf{k}\sigma} - \tfrac{1}{2} J_K \sum_{\mathbf{k}\sigma,\mathbf{k}'\sigma'} (\gamma_{\mathbf{k}} p^\dagger_{\mathbf{k}\sigma} s^\dagger_{-\mathbf{k}-\sigma})(\gamma_{\mathbf{k}'} s_{-\mathbf{k}'-\sigma'} p_{\mathbf{k}'\sigma'}) ,$$

$$s^\dagger_{-\mathbf{k}-\sigma} = \alpha d_{\mathbf{k}\sigma} + \beta \mathrm{sgn}\sigma d^\dagger_{-\mathbf{k}-\sigma}, \quad |\alpha|^2 + |\beta|^2 = 1 ,$$

where the, initially undetermined, coefficients α and β are allowed by SU(2) symmetry. Minimizing the energy in the MF approximation $\langle P_G H\rangle \approx \langle H_{MF}\rangle = E_{SL} - E$ where $-E$ is the bound-state energy, leads to the bound-state equation $J_K^{-1} = g(E)$, where

$$g(E) = \sum_{k>k_F} \frac{\gamma^2_{\mathbf{k}}}{2(E_{\mathbf{k}} + \epsilon_{\mathbf{k}} + E)} \left[1 - \frac{C(c_x + c_y)}{[2(c_x^2 + c_y^2)]^{1/2}}\right] ,$$

and $C = \sqrt{2}[\mathrm{Re}(\alpha^*\beta) + \mathrm{Im}(\alpha^*\beta)]$. The bound-state energy is maximized by $C=1$. For $J_K < J_c = 1/g(0)$, it is not possible for bonds inside the spin liquid to be broken, and a bound state does not form. For smaller values of J_H d-spin compensation is not complete at $T=0$, and we expect the development of AFM long-range order, for which this approach is inappropriate.

Quite unlike the conventional Cooper instability, the presence of a Fermi surface of the conduction electrons does not play a central role, and J_c is finite even when k_F becomes zero. An isolated conduction electron can hence bind to the spin liquid, transferring its spin to free neutral quasiparticles in the spin liquid.

Now consider a finite hole density. Pair condensation, as opposed to the single pair instability, occurs at the point where the Gaussian coefficient of V in the free energy $F(U,V)$ acquires a negative eigenvalue. This occurs at $T_c = \beta_c^{-1}$ determined by

$$\frac{1}{J_K} = \sum_{\mathbf{k},\zeta=\pm} \gamma^2_{\mathbf{k}} \left[1 - \zeta \frac{c_x + c_y}{[2(c_x^2 + c_y^2)]^{1/2}}\right] \frac{1 - f(E_{\mathbf{k}}) - f(\zeta\epsilon_{\mathbf{k}})}{2(E_{\mathbf{k}} + \zeta\epsilon_{\mathbf{k}})} , \tag{10}$$

with the choice $V^\dagger[\tau_1 + \tau_2]V = \sqrt{2}V_0^2\tau_3$ which maximizes T_c. For the pure dual-exchange model, $f(\epsilon_{\mathbf{k}}) = f(-\mu) = x/4$, where x is the number of p holes per unit cell. The figure shows T_c determined by this equation at low and high hole concentration. T_c for pair condensation is higher than for single pair formation. This condensation process then stabilizes the ground state against antiferromagnetism.

Let us briefly consider the properties of the state formed by this charge $1e$ "Cooper instability" below T_c. The presence of the spin-liquid background induces a self-energy

$$\Sigma_{\mathbf{k}}(\omega) = V^{\dagger}[\omega - W - E_{\mathbf{k}}(c_x \tau_1 + c_y \tau_2)]^{-1} V$$

in the p-electron propagator $\mathcal{G}^{pp}(\omega) = [\omega - \epsilon_{\mathbf{k}} \tau_3 - \Sigma_{\mathbf{k}}(\omega)]^{-1}$, which is off diagonal in Nambu space. This resembles resonant Kondo scattering familiar in the theory of heavy fermions,[11] with the exception that it contains a *pairing component*. Since the self-energy is a locally SU(2) invariant quantity, we are forced to conclude that Kondo compensation in an AFM background generates superconductivity in the conduction of p holes, *bypassing* the intermediate development of a Fermi liquid.

More complete analysis of the quasiparticle spectrum in this phase shows there are *two* gaps[13] in which the upper gap has mainly d composition and the lower is of mainly p character. Beyond the upper gap the density of states $\rho(\omega) = \omega/\pi\Delta^2$ is linear due to the residual spin-liquid background.

As in heavy Fermi liquids, these quasiparticles *all* carry charge $-e$. Energy is minimized when the V fields are adjusted so that the original zeros of the gap in the spin liquid are located at $\mathbf{G}/2$. In a field, $\gamma_{\mathbf{k}} \rightarrow \gamma_{\mathbf{k}-e\mathbf{A}}$, and the positions of the gap's zeros shift to $\mathbf{G}/2 + e\mathbf{A}$, causing $E_{\mathbf{k}} \rightarrow E_{\mathbf{k}-e\mathbf{A}}$. This phenomena gives the quasiparticles charge $-e$ and the order parameter charge $-2e$ corresponding to a conventional superconducting flux quantum $h/2e$.

To conclude, we mention some effects of fluctuations. First, including the effects of fluctuations has the same effect as reducing the p-hole concentration: The superconducting transition temperatures T_c^{MF} are depressed, and the range of J_H/J_K where the spin liquid is stable is extended (see Fig. 1). Although we treated J_H/J_K as a variable, its unrenormalized value in cuprate superconductors is fixed by the ratios of hybridization to charge-transfer energies, probably to a value rather less than 1. The MF theory would then predict that at moderate doping values the system would enter the superconducting phase from the Fermi liquid. However, since fluctuations will extend the range of J_H/J_K where a spin liquid is stable, our scenario may also occur at moderate doping levels in a more realistic two-band model. Last, above T_c the superconducting order parameter associated with V carries charge e, due to the formation of virtual bound pairs of p holes and d spins. Since a dispersionless p band would be insulating, we have the unusual situation where these supercurrent fluctuations determine the bulk of the conductivity. We are currently examining whether this transport mechanism can be identified with the unusual high-temperature conductivity of cuprate superconductors.

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