Microscopic model of hybrid pairing: A common approach to heavy-fermion and high-T_c superconductivity

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Real-space pairing among hybridized electrons in a periodic system is proposed. It is mediated by the *s-d* exchange of the type which leads to the Kondo effect in impurity systems. The effective Hamiltonian in the hybridized basis is transformed into a form similar to that for a narrow-band case. The importance of the results for both the heavy-fermion and the high-T_c superconductors is briefly discussed.

In this paper we propose a novel approach that deals with singlet pairing mediated by antiferromagnetic exchange interactions between conduction (c) electrons and localized (f) electrons in hybridized systems modeled by the periodic Anderson model (PAM). Namely, we visualize explicitly how the singlet spin correlations in real space can lead to superconductivity in heavy-fermion systems.[1] We also briefly discuss the connection with the exchange-mediated pairing proposed recently for narrow-band electrons.[2-6] The present approach thus provided a synthesis of the concept of antiferromagnetic c-f interaction[7,8] with the recent concept[2] of exchange-mediated pairing in narrow bands.

The pairing mechanism is supplied by the second-order coupling V^2/U, where V is the magnitude of hybridization between c and f states and U is the magnitude of the intra-atomic (f-f) electron repulsion. However, unlike the standard treatments,[7,9] the f-c coupling neither diverges[7] nor vanishes[9] as the bare f-level position (ε_f) approaches the Fermi level. This situation arises because only a nondivergent part of higher-order processes is transformed out by an appropriate canonical transformation.[8] In effect, the PAM is divided into a part corresponding to the $U=\infty$ limit that can be treated with the help of methods devised recently,[10,11] and a part representing hybrid (f-c) pairing which is missing in the above treatments.

The hybrid f-c pairing process has been proposed recently.[12] Here, we derive explicitly the microscopic Hamiltonian containing this pairing and show how the effective two-band situation arises. The method of diagonalizing the effective Hamiltonian also differs from that of Ref. 12.

The starting Hamiltonian may be written in the Wannier representation as

$$H=\sum_{m,n,\sigma}{}' t_{mn}c^{\dagger}_{m\sigma}c_{n\sigma}+\varepsilon_f\sum_{i,\sigma}N_{i\sigma}+U\sum_i N_{i\uparrow}N_{i\downarrow}+\sum_{i,m,\sigma}V_{im}(a^{\dagger}_{i\sigma}c_{m\sigma}+c^{\dagger}_{m\sigma}a_{i\sigma}), \quad (1)$$

where the (i,j) label f states, the (m,n) label delocalized c states, and $N_{i\sigma}=a^{\dagger}_{i\sigma}a_{i\sigma}$ is the particle number operator for the localized f states. The first term describes the band energy of the delocalized c states, the second and third, single-particle and Coulomb (atomic) energies for f states, whereas the last represents the hybridization energy, involving both intra-atomic ($i=m$) and interatomic ($i\neq m$) parts. The matrix elements t_{mn} and V_{mi} represent hopping and hybridization integrals, respectively. We choose the matrix elements V_{im} as real. Also, we assume that $U>|\mu|$ and $U\gg|V_{im}|$, where μ is the Fermi energy. In other words, we consider here the large-U limit. Then, the charge transfers $f\leftarrow c$ and $f\rightarrow c$ can be divided into low- and high-energy processes. Explicitly, we introduce a decomposition

$$a^{\dagger}_{i\sigma}c_{m\sigma}+c_{m\sigma}a_{i\sigma}\equiv(1-N_{i-\sigma}+N_{i-\sigma})(a^{\dagger}_{i\sigma}c_{m\sigma}+c^{\dagger}_{m\sigma}a_{i\sigma})=(1-N_{i-\sigma})(\cdots)+N_{i-\sigma}(\cdots). \quad (2)$$

The first term represents the processes which do not involve U in any order, while the second does involve U and hence leads to higher-order mixing processes. The basic idea introduced at this point[9] is to canonically transform out the second term only and to replace it by an effective interaction incorporating higher-order virtual processes. In this manner we avoid singularities that are present in the original Schrieffer-Wolff formulation.[7] The effective Hamiltonian obtained in that fashion reads

$$\tilde{H}=\sum_{m,n,\sigma}\left(t_{mn}+\sum_i V_{mi}V_{in}\frac{N_{i-\sigma}}{U+\varepsilon_f}\right)c^{\dagger}_{m\sigma}c_{n\sigma}+\varepsilon_f\sum_{i,\sigma}N_{i\sigma}+\sum_i\left(U+\frac{2}{U+\varepsilon_f}\sum_m V^2_{mi}\right)N_{i\uparrow}N_{i\downarrow}+\sum_{i,m,\sigma}V_{im}(1-N_{i-\sigma})(a^{\dagger}_{i\sigma}c_{m\sigma}+c^{\dagger}_{m\sigma}a_{i\sigma})+\sum_{i,m}\frac{2V_{im}V_{mi}}{U+\varepsilon_f}(\mathbf{S}_i\cdot\mathbf{S}_m-\tfrac{1}{4}N_in_m)+\frac{1}{U+\varepsilon_f}\sum_{i,m,n,\sigma}V_{mi}V_{in}S_i^{-\sigma}c^{\dagger}_{m-\sigma}c_{n\sigma}, \quad (3)$$

where

$$S_i^q \equiv a_{i\sigma}^\dagger a_{i-\sigma},\ N_i = \sum_\sigma N_{i\sigma},\ \text{and}\ n_m = \sum_\sigma n_{m\sigma}.$$

The Hamiltonian contains renormalizations of the conduction-band width and of U. More important are the last two terms: the antiferromagnetic f-c coupling and the spin-flip scattering of c electrons by the f moments.

The Hamiltonian (2) may be rewritten in a more compact form by introducing the pairing operators in real space

$$b_{im}^\dagger = \frac{1}{\sqrt{2}}(a_{i\uparrow}^\dagger c_{m\downarrow}^\dagger - a_{i\downarrow}^\dagger c_{m\uparrow}^\dagger),$$
$$b_{im} = \frac{1}{\sqrt{2}}(a_{i\downarrow} c_{n\uparrow} - a_{i\uparrow} c_{m\downarrow}). \tag{4}$$

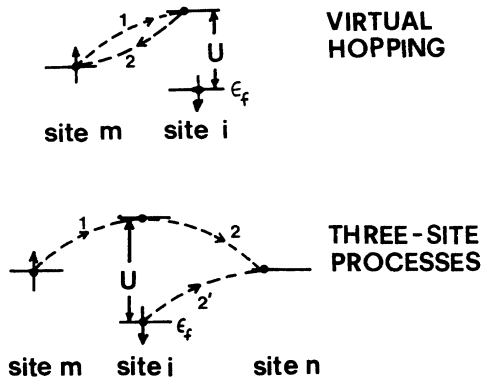

FIG. 1. Schematic representation of the hopping processes to second order in V_{im}. The hoppings labeled as 2 and 2′ are alternative processes.

Then, the expression (3) has a closed form

$$\tilde{H} = \sum_{m,n,\sigma}{}' t_{mn} c_{m\sigma}^\dagger c_{n\sigma} + \varepsilon_f \sum_{i,\sigma} N_{i\sigma} + U \sum_i N_{i\uparrow} N_{i\downarrow} + \sum_{i,m,\sigma} V_{im}(1 - N_{i-\sigma})(a_{i\sigma}^\dagger c_{m\sigma} + c_{m\sigma}^\dagger a_{i\sigma}) - \sum_{i,m,n} \frac{2V_{mi}V_{in}}{U + \varepsilon_f} b_{im}^\dagger b_{in}, \tag{5}$$

where the irrelevant renormalization of U in (3) has been ignored. The first four terms represent the PAM in the $U = \infty$ limit while the last provides a real-space singlet pairing of f and c electrons. The pairing part is composed of two-site ($m = n$) and three-site terms. The former represents pair binding, whereas the latter represents a hopping of the conduction electron from m to n via the localized paired $|i\uparrow\downarrow\rangle$ intermediate state (i.e., exhibits *resonant behavior of the hybrid f-electron–conduction-electron bond*),[6] as shown schematically in Fig. 1. The range of the pairing interaction depends on the nature of hybridization matrix element $V(R_i - R_m)$. In the simplest case of intra-atomic hybridization $V_{im} = V\delta_{im}$ we obtain the hybrid intra-atomic pairing postulated before.[12] However, if $\varepsilon_f \to 0$ one encounters additionally a residual hybridization which cannot be transformed out within the canonical perturbation expansion formalism[8] if $\varepsilon_f \lesssim V$. The itinerant character of bare atomic (f) states is provided by the presence of this residual hybridization.

To draw physical conclusions from the processes contained in the form (5) we can apply in principle any of the methods[10–12] proposed for the PAM in the $U \to \infty$ limit and treat the pairing part (the last term) in the Hartree-Fock approximation. Having in mind the application to the heavy-fermion case we use the simple ansatz introduced by Rice and Ueda,[10] i.e., we renormalize the residual hybridization term in (5) according to

$$V_{mi}(1 - N_{i-\sigma})(a_{i\sigma}^\dagger c_{m\sigma} + c_{m\sigma}^\dagger a_{i\sigma}) \equiv \tilde{V}_{mi}(a_{i\sigma}^\dagger c_{m\sigma} + c_{m\sigma}^\dagger a_{i\sigma}), \tag{5'}$$

with $q_\sigma = (1 - n)/(1 - n_\sigma)$. For the nonmagnetic state considered here we have $q_\sigma = q = (1 - n)/(1 - n/2)$. Such renormalization leads to an effective single-particle hybridization by assigning to the residual hybridization the principal qualitative function of promoting the effective f-f hopping. In other words, the itineracy of f electrons is provided by the residual hybridization which is nonzero as long as $n_f < 1$. This procedure is justifiable when the width W of the c band greatly exceeds $|V|$, and near the limit $n_f \to 1$ (we have in mind the application to heavy fermions). Incorporating (5′) into (5) and taking the space Fourier transform we obtain

$$\tilde{H} = \sum_{k,\sigma} \varepsilon_k n_{k\sigma} + \varepsilon_f \sum_{k,\sigma} N_{k\sigma} + \sum_{k,\sigma} \tilde{V}_k (a_{k\sigma}^\dagger c_{k\sigma} + c_{k\sigma}^\dagger a_{k\sigma}) - \frac{2}{U + \varepsilon_f} \frac{1}{N} \sum_{k,q} V_k V_q b_k^\dagger b_q, \tag{6}$$

where $\tilde{V}_k = q^{1/2} V_k$, and ε_k and V_k are the Fourier transforms of t_{mn} and V_{im}. Also,[13]

$$b_k^\dagger = (a_{k\uparrow}^\dagger c_{-k\downarrow}^\dagger - a_{k\downarrow}^\dagger c_{-k\uparrow}^\dagger)/\sqrt{2}.$$

Note that the term $U\sum N_{i\uparrow}N_{i\downarrow}$ in (5) has been neglected since the renormalization $V_{im} \to \tilde{V}_{im}$ is caused by the circumstance that, for $U \to \infty$, we have that $\langle N_{i\uparrow}N_{i\downarrow}\rangle = 0$.

Next, we decouple the last term of (6) using the Hartree-Fock (BCS-type) approximation in real space

$$b_{im}^\dagger b_{in} \simeq \langle b_{im}^\dagger \rangle b_{in} + b_{im}^\dagger \langle b_{in}\rangle - \langle b_{im}^\dagger\rangle\langle b_{in}\rangle + \tfrac{1}{2}\sum_\sigma [(a_{i\sigma}^\dagger c_{n\sigma} + c_{m\sigma}^\dagger a_{i\sigma}) S_{-\sigma} - S_\sigma S_{-\sigma}]$$
$$+ \tfrac{1}{2}\sum_\sigma (\bar{N}_{i-\sigma} c_{m\sigma}^\dagger c_{n\sigma} + N_{i\sigma}\langle c_{m-\sigma}^\dagger c_{n-\sigma}\rangle - \bar{N}_{i-\sigma}\langle c_{m\sigma}^\dagger c_{n\sigma}\rangle), \tag{7}$$

where

$$S_\sigma \equiv \langle a_{i\sigma}^\dagger c_{m\sigma} \rangle .$$

The first three terms in Eq. (7) provide a singlet paired state, the next three renormalize the residual hybridization

$$\tilde{V}_{im} \rightarrow \tilde{V}_{im} - \sum_n \frac{V_{mi} V_{in}}{U + \varepsilon_f} S_{-\sigma} ,$$

while the last three renormalize both the position of the atomic level

$$\varepsilon_f \rightarrow \varepsilon_f - \sum_{m,n} \frac{V_{mi} V_{in}}{U + \varepsilon_f} \langle c_{m-\sigma}^\dagger c_{n-\sigma} \rangle ,$$

and the hopping matrix element

$$t_{mn} \rightarrow t_{mn} - \sum_i \frac{V_{mi} V_{in}}{U + \varepsilon_f} \bar{N}_{i-\sigma} .$$

All the additional contributions are of second order and should not change the essence of the problem. Hence, we neglect them hereafter and assume that the system has no spin polarization.

Performing the transformation to the hybridized basis for the operator $b_k^\dagger$ we have

$$b_k^\dagger = -\frac{s}{2}(\sin 2\theta_k)(a_{k\uparrow}^\dagger a_{-k\downarrow}^\dagger - a_{k\downarrow}^\dagger a_{-k\uparrow}^\dagger)/\sqrt{2} + (\cos^2\theta_k)(a_{k\uparrow}^\dagger \beta_{-k\downarrow}^\dagger - a_{k\downarrow}^\dagger \beta_{k\uparrow}^\dagger)/\sqrt{2}$$
$$- s(\sin^2\theta_k)(\beta_{k\uparrow}^\dagger a_{-k\downarrow}^\dagger - \beta_{k\downarrow}^\dagger a_{-k\uparrow}^\dagger)/\sqrt{2} + \tfrac{1}{2}(\sin 2\theta_k)(\beta_{k\uparrow}^\dagger \beta_{-k\downarrow}^\dagger - \beta_{k\downarrow}^\dagger \beta_{-k\uparrow}^\dagger)/\sqrt{2} .$$

On substituting this expression into the Fourier transform of (7) and the resultant in to (6), we arrive at the effective Hamiltonian

$$\tilde{H} = \sum_{k,\sigma} (E_{k-} a_{k\sigma}^\dagger a_{k\sigma} + E_{k+} \beta_{k\sigma}^\dagger \beta_{k\sigma}) - \frac{s\Delta}{U + \varepsilon_f} \sum_k V_k \sin 2\theta_k (a_{k\uparrow}^\dagger a_{-k\downarrow}^\dagger - a_{k\downarrow}^\dagger a_{-k\uparrow}^\dagger)/\sqrt{2} + 2|\Delta|^2 N/(U + \varepsilon_f) + \cdots , \quad (8)$$

where $\Delta \equiv -(1/N)\sum_q V_q \langle b_q \rangle$ is the gap parameter; the angle of mixing is determined from the condition

$$\tan(2\theta_k) = 2\tilde{V}_k/(\varepsilon_k - \varepsilon_f) ,$$

and the energy of quasiparticles in the two hybridized bands (α and β) are given by

$$E_{k\pm} = \frac{\varepsilon_k + \varepsilon_f}{2} \pm \left[\left(\frac{\varepsilon_k - \varepsilon_f}{2} \right)^2 + \tilde{V}_k^2 \right]^{1/2} .$$

Additionally, we have assumed that $V_{-k} = sV_k \equiv \pm V_k$. Hence, the transformation to the hybridized basis leads to a two-band situation.[14] The pairing takes place in the α band if $n_f < 1$ and the overlap between the hybridized bands is small (e.g., when there is a hybridization gap or pseudogap between the bands). In that case Eq. (8) reduces to the BCS Hamiltonian for the α band

$$\tilde{H} = \sum_{k,\sigma} E_{k-} a_{k\sigma}^\dagger a_{k\sigma}$$
$$+ \Delta \sum_k \gamma_k (a_{k\uparrow}^\dagger a_{-k\downarrow}^\dagger - a_{k\downarrow}^\dagger a_{-k\uparrow}^\dagger)/\sqrt{2} + \text{H.c.} , \quad (9a)$$

with

$$\gamma_k = \frac{-2s}{U + \varepsilon_f} \frac{V_k \tilde{V}_k}{(\varepsilon_k - \varepsilon_f)[1 + 4\tilde{V}_k^2/(\varepsilon_k - \varepsilon_f)^2]^{1/2}} . \quad (9b)$$

For more than two electrons per site this Hamiltonian should be understood as representing hybridized hole states. The first basic conclusion one may draw from the form (9a) of the effective Hamiltonian is that the heavy quasiparticles provide pairing if the Fermi energy lies in the peak region of the density hybridized states.[14] The factor γ_k introduces, in general, an anisotropic pairing; it vanishes in the localized-moment limit ($n_f = 1$), as does the pairing. Additionally, if $V_k = V$ (i.e., if the hybridization is intra-atomic), then in the mixed-valence limit $|\varepsilon_k - \varepsilon_f| \ll |V|$ the Hamiltonian (9a) reduces to the BCS Hamiltonian with almost isotropic s-wave pairing among the occupied hybridized states. Moreover, it is crucial to note that *Eq. (9a) is of the same form as that derived for exchange-mediated superconductivity in a strongly correlated narrow band,* [2-6] where the pairing is supplied by the kinetic exchange term[15] t^2/U. Here, the role of d-d exchange is played by the f-c kinetic exchange V^2/U. In other words, we have proved a *formal* equivalence of the previous approaches[3-6] to high-T_c superconductivity (which start from the Hubbard Hamiltonian and include the kinetic exchange in the metallic phase[15] for $n < 1$) with the present approach starting from the periodic Anderson-Wolff Hamiltonian (1). The difference is in the k dependence of band energies ($q\varepsilon_k$ and E_{k-}, respectively), in the k dependence of γ_k, and in that for the present case the pairing vanishes as $n_f \rightarrow 1$.[16] One should also note that in the fourth-order terms ($\sim V^4$), which were neglected in the derivation of (3), an f-f exchange-mediated pairing appears that corresponds directly to that considered by Anderson[2] and others.[3-6] In physical terms, *we have demonstrated the relevance of the exchange-mediated pairing for the heavy-fermion systems.* The hybrid pairs move as long as the residual hybridization $\tilde{V}$ does not vanish.

There is, however, one difference between the narrow-band formalisms[2-6] and the present one. Namely, the former lead to a Mott insulating state in the limit $n = 1$. Hence the superconducting phase evolves into antiferromagnetic or other[2,4] insulating phases. In the present model, the ground state for $n = 1$ is metallic or semimetallic, depending on the position of ε_f with respect to the bottom edge of the conduction band. It is not yet clear whether the concept of Mott-Hubbard subbands can be

applied to the hybridized band (see, however, Ref. 17).

The Hamiltonian (1) is also a good starting point in the treatment of high-T_c superconductors, where the quantities V_{im} play the role of intersite hybridization between the $d_{x^2-y^2}$ state of the Cu^{2+} ion and the neighboring p_x and p_y states of O^{2-}. If we believe that for this case the direct p-p overlap between neighboring anions is small compared to V_{im} then neither the renormalization $V_{im} \rightarrow q^{1/2} V_{im}$ nor the Hartree-Fock approximation of the second-order term seems to be appropriate. In other words, the present paper may be regarded as dealing with a regime ($W \gg |V|$) complementary to that discussed by Zhang and Rice,[18] who ascribe the main role to hybridization in forming the bare hybridized bands, and calculate the d-d pairing to fourth-order terms in V. However, one can see a qualitative difference between the present pairing mechanism represented by (5) and those of either Hirsch[19] or Emery.[20] Namely, here we have an admixture of $2p^6$ and $2p^5$ states to the essentially $3d^9$ states of Cu. In effect, the hybrid p-d states are those of p holes hopping from one oxygen to another via the Cu $3d^{10}$ intermediate configurations. The $3d^8$ states are not involved. A more quantitative analysis requires inclusion of the fourth-order terms in (5) as well.

The Hamiltonian (9a) may be easily diagonalized to yield quasiparticle energies in the superconducting phase of the form

$$\tilde{E}_k = [(E_{k-} - \mu)^2 + (\sqrt{2}\Delta\gamma_k)^2]^{1/2} . \tag{10}$$

We can also calculate the self-consistent equation for the gap parameter

$$1 = \frac{-s}{2N} \sum_k \gamma_k V_k \sin 2\theta_k \frac{1}{\tilde{E}_k} \tanh\left(\frac{\beta \tilde{E}_k}{2} \right) . \tag{11}$$

This equation differs from the corresponding BCS equation for the isotropic case by the presence of the factor $(\sqrt{2})\gamma_k V_k^2$. For a simple estimate of the critical temperature [it is a solution of Eq. (11) for $\Delta = 0$] this factor can be approximated by considering the case $V_k = V$ and replacing $\varepsilon_k - \varepsilon_f$ with a constant equal to $|\varepsilon_f| \equiv |\varepsilon_f - \mu|$. Then, Eq. (11) reads

$$1 = \frac{2q}{U + \varepsilon_f} V^4 \frac{1}{(\Delta\varepsilon)^2 + 4V^2} \int_{E_L}^{E_U} \frac{d\varepsilon}{\varepsilon} \tanh\left(\frac{\beta_c \varepsilon}{2} \right) \rho(\varepsilon) , \tag{12a}$$

where $\beta_c = (k_B T_c)^{-1}$ and where the integration is carried out over the lower hybridized band stretching from E_L to E_U. The above is formally a BCS-type equation. Therefore,

$$k_B T_c = 1.14 \frac{2V^2}{U + \varepsilon_f} \exp\left[-\frac{U + \varepsilon_f}{4q\rho(\mu)V^4} [(\Delta\varepsilon)^2 + 4V^2] \right] , \tag{12b}$$

where $\rho(\mu)$ is the density of hybridized states (per site per spin) at the Fermi energy. In this equation we have assumed that the characteristic energy corresponding to the Debye temperature in the ordinary BCS case is $J = 2V^2/(U + \varepsilon_f)$, the f-c coupling constant (exchange integral). One should notice that $T_c \rightarrow 0$ if either $U \rightarrow \infty$, or $q \rightarrow 0$ ($n_f \rightarrow 1$), or $V \rightarrow 0$. Also, a maximal value of $T_c \simeq J/k_B$ is achieved for $\Delta\varepsilon = 0$. However, the weak-coupling approximation made in deriving (12b) may then be violated as illustrated below.

For numerical estimate of Eq. (12b) we take $U = 10$ eV, $\Delta\varepsilon = 1$ eV, $q = 0.04$ (i.e., $n_f = 0.98$) and $\rho(\mu) = 200$ states/(eV spin) (i.e., $\gamma \sim 1$ J/mol K^2). Then $T_c \simeq 0.1$ K for $V = 0.5$ eV ($\tilde{V} = 20$ meV). The value $T_c \simeq 0.6$ K is obtained, e.g., for $U = 8$ eV, $\Delta\varepsilon = 2$ eV, and $V = 0.6$ eV. Hence, we obtain a reasonable value of T_c for heavy-fermion materials taking typical values of the parameters. Additionally, T_c grows by more than two orders of magnitude when $|V|$ increases toward the value 1 eV. This rapid increase of T_c may, however, be an artifact of the method of deriving (12b), since for the first set of parameters $J \simeq 735$ K, a value comparable to the width of the lower hybridized band if the ε_f level is closer to the bottom edge of the bare c band. For a better evaluation of the weak-coupling approximation one requires a self-consistent determination of n_f for the other specified parameters.

Summarizing, we have provided a microscopic model of pairing between hybridized electrons. It is caused by antiferromagnetic interactions between the two sets of electrons, each occupying partially filled orbitals. The hybrid (interband) pairing may be represented via an effective BCS Hamiltonian representing paired states in a single hybridized band. In this manner we provide a framework for understanding both heavy-fermion and high-T_c superconductors within the same (exchange-mediated) mechanism of pairing, even though the method of solution of the effective Hamiltonian (5) may be different for the two situations. A more complete analysis requires inclusion of the fourth-order processes, as well as a more detailed discussion of importance of the Kondo-lattice effects.

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