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# Model of Interband Pairing in Mixed Valence and Heavy Fermion Systems

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Various possible types of interband pairings in mixed valence and heavy fermion systems are investigated, using a two-band model with Kondo-type interaction. In this model, the system may develop interband singlet superconducting ordering, or electron-hole pairing of excitonic type, which induces an effective $f-d$ hybridization. The conditions for the realization of each of these orderings depends crucially on the dispersion of the corresponding original bands. The mutual influence of the two pairings, as well as the effect of "external" hybridization, are investigated.

## 1. Introduction

Due to their complex and unusual properties, mixed valence and heavy fermion systems have attracted considerable attention. Up to now, there exists no consensus on the nature and microscopic origin of the quantum state associated with heavy fermions. Their appearence is ascribed to the existence of an $f$-band in the vicinity of the Fermi level $\varepsilon_F$ [1] (or to the $f$-band with "electronic polaron" narrowing [2]), as well as to the Kondo lattice (collective Kondo or Abrikosov-Suhl resonance [3]). For detailed reviews of possible pairing mechanisms the reader is referred to Refs. 3–6. In the absence of a full understanding of their nature, a phenomenological description might be useful.

In one of the most promising models, the existence of two electron bands in the close vicinity of the Fermi level is proposed [7]. In these bands, the mobilities of the electrons are assumed to be different. Associated with these different dispersions is a light mass $m_d$ and a heavy mass $m_f$, respectively. This perception is supported both by experimental evidence [8, 9] and by theoretical considerations [10, 11]. In particular, Tesanovic and Valls have shown [11], that it is (at least in some approximation) possible, to reduce Anderson or Kondo lattices to a two band structure with hybridization of the form $Vf_{k\sigma}^{+} d_{k\sigma}$ between these bands, including an interband interaction of the Kondo type. Although the Kondo lattice problem has not been solved yet, the character of this interaction is easily understandable on qualitative terms: when the starting point is the Anderson model with deep $f$-level $E_f \ll \varepsilon_F$, the Schrieffer-Wolff transformation just yields this particular type of Kondo interaction, with an associated exchange constant $J \sim V/(\varepsilon_F - E_f)$. When the $f$-level approaches $\varepsilon_F$ (in models of the type of Refs. 1 and 2), or in more sophisticated treatments [11], the exact form of the interaction is less evident, but it is quite reasonable to suspect that the main qualitative features will pertain; i.e. that there will be an effective antiferromagnetic exchange interaction between initial (bare) "heavy $f$" and "light $d$" electrons with opposite spins.

It is also quite natural to expect, and will indeed be shown below, that there may evolve singlet interband pairing either of a superconducting type of the form $\langle f_\uparrow^+ d_\downarrow^+ - f_\downarrow^+ d_\uparrow^+ \rangle$ or an electron-hole pairing of excitonic type $\langle f_\sigma d_\sigma^+ \rangle$. Such pairings give raise to a coherent singlet ground state of superconducting or of insulating type.

Anomalous averages over field operators in the above form have already been studied in early treatments of the Kondo effect [12, 13]. However, in the case of a single impurity, introduction of these averages could serve only as an approximation: in that case, no actual phase transition exists. The appearence of anomalous averages signals only a transition from the regime of weak antiferromagnetic coupling to the crossover towards strong effective coupling in a perturbation expansion in $J$ [13]. This compares with similar findings in the perturbative approach to quantum chromodynamics (QCD), or to other nonabelian gauge theories. However, in dealing with a regular, periodic system like the Anderson or Kondo lattice, one may speculate, that the same anomalous averages, which in the single impurity case only indicate a transition to the strong coupling regime, give raise to an adequate description of the (collective) phenomenology. The same argument applies even for conventional bulk superconductivity: since if the Fröhlich interaction mediated attraction of the BCS theory would act only at a singular point (e.g. at the coordinate origin of configuration space), then the introduction of the averages $\langle a^+ a^+ \rangle$ would not correspond to an actual phase transition, but would rather be an approximation. This is due to the fact, that bulk superconductivity is realized as a collective phenomenon of the manybody system of electrons.

These arguments may justify recent attempts to describe superconductivity in heavy fermion systems as a consequence of interband ("hybrid") pairing [14, 15]. The hamiltonian is given by

$$H=\sum_{k,\sigma}\{\varepsilon_f(k) f_{k\sigma}^+ f_{k\sigma}+\varepsilon_d(k)\, d_{k\sigma}^+ d_{k\sigma} + V(f_{k\sigma}^+ d_{k\sigma}+\text{h.c.})\}+\frac{J}{2}\sum_{k,p,q}(f_{k\uparrow}^+ d_{p\downarrow}^+ - f_{k\downarrow}^+ d_{p\uparrow}^+) \cdot (d_{p-q\uparrow} f_{k+q\downarrow} - d_{p-q\downarrow} f_{k+q\uparrow}), \tag{1}$$

with $J>0$. Here, the model of Refs. 14 and 15 was generalized by the introduction of $f-d$ hybridization. If the $d$- and $f$-bands are not too narrow, that is if the bandwidths $W_d$, $W_f > T_c$, the standard BCS approximation is valid, and anomalous averages can be introduced:

$$\Delta=\frac{J}{2}\sum_{k}\langle d_{k\downarrow} f_{-k\uparrow} - d_{k\uparrow} f_{-k\downarrow}\rangle. \tag{2}$$

Then, $T_c$ is given by the expression [14]

$$T_c = 1.14(W_d\, W_f)^{1/2}\, e^{-\frac{1+\lambda}{J N_d(0)}}, \tag{3}$$

where $\lambda = W_f/W_d = m_d/m_f$, $N_d(0)$ is the density of states of the $d$-band, and the masses correspond to the bare values in the original unrenormalized Hamiltonian (1). In the opposite case of a narrow $f$-band $W_f \le T_c$ instead of (2), more general averages have to be introduced, relaxing the strict singlet condition for states in momentum space $(k, -k)$. In this case one obtains for $T_c$ the expression [15]

$$T_c \simeq W_d\, e^{-\frac{1}{J N_d(0)}}. \tag{4}$$

The hybrid pairing model described above is rather promising. It explains quite naturally many features of heavy fermion superconductivity [15]:

*(i)* the existence of superconductivity, e.g. in $CeCu_2Si_2$, can be explained, as well as the absence of superconductivity in its non-$f$ counterpart $LaCu_2Si_2$;

*(ii)* no extra and additional pairing mechanisms of superconductivity need to be introduced: the Kondo-type interaction, which is usually emloyed to explain the main properties of these compounds in the normal phase, also provide a mechanism for Cooper pairing;

*(iii)* one is naturally lead to singlet pairing despite (here due to) the strong exchange interactions and spin fluctuations. Experimentally, singlet superconductivity seems to be realized at least in $CeCu_2Si_2$ and possibly in some other heavy fermion superconductors [16];

*(iv)* one can, as yet rather speculatively, try to explain the nonexponential behaviour of different properties close to $T_c$ (as originating from the possible incomplete nesting of the $f$- and $d$-band Fermi surfaces).

The treatment of Refs. 14 and 15 however ignores the effect of induced hybridization (e.g., see Ref. 11). Besides that, these investigations account only for superconducting pairing (2). However, also nonzero "excitonic" averages $\Sigma \propto \langle f_\sigma d_\sigma^+ \rangle$ may appear as an additional contribution to "induced" hybridization. They would form as a consequence of bare hybridization. Even in the absence of initial hybridization they give raise to negative contributions to the average interaction energy: with such a decoupling, $\langle H_{\text{int}} \rangle = -J|\Sigma|^2$.

Corresponding anomalous averages were introduced long ago in the course of studies of excitonic insulators (see, e.g. Refs. 17); in the context of valence fluctuations they were introduced in Ref. 18.

Furthermore, it can be shown, that triplet interband pairing of the form $\langle f_\uparrow d_\uparrow^+ \rangle$, which yields a magnetic ground state of the spin density wave type, is unfavorable and renders a positive contribution to $\langle H_{\text{int}} \rangle$.

Below, we shall study the consequences of the introduction of "external" hybridization and also excitonic pairing ("induced" hybridization), in addition to superconducting pairing.

## 2. Simultaneous Treatment of Different Types of Pairing

In the following the two-band model, described by the Hamiltonian (1) is used. The dispersion laws for the $f$- and $d$-bands are modeled by

$$\varepsilon_f(k)=\frac{k^2-k_F^2}{2m_f}, \quad \varepsilon_d(k)=\frac{k^2-k_F^2}{2m_d}. \tag{5}$$

The most straightforward case of a complete nesting of the Fermi-surfaces of the two bands is assumed. We do not specify the exact nature of these bands. This model can be considered as a phenomenological one, which describes the properties of valence fluctuation materials reasonably well.

It is assumed, that both bands are not very narrow, such that $W_d, W_f > T_c$. The mean field approximation (see discussion in the last section) is used. In addition to the superconducting averages we introduce excitonic averages $\langle f_{k\sigma} d_{k\sigma}^+\rangle$, which combine in the Hamiltonian (1) with the (bare) hybridization $V$ to give

$$\tilde{V}_\sigma = V + \frac{J}{2}\sum_k \langle f_{k\sigma} d_{k\sigma}^+\rangle. \tag{6}$$

As we study the nonmagnetic case, $\tilde{V}_\uparrow = \tilde{V}_\downarrow$. Hence the spin index in $\tilde{V}$ can be omitted.

After the corresponding decouplings, the Hamiltonian (1) takes on the form

$$\begin{aligned} H = \sum_{k,\sigma} \{&\varepsilon_f(k) f_{k\sigma}^+ f_{k\sigma} + \varepsilon_d(k)\, d_{k\sigma}^+ d_{k\sigma} \\ &+ (\tilde{V} f_{k\sigma}^+ d_{k\sigma} + \text{h.c.})\} \\ &+ \sum_k \{\Delta(f_{k\uparrow}^+ d_{-k\downarrow}^+ - f_{k\downarrow}^+ d_{-k\uparrow}) + \text{h.c.}\} + \text{const}. \end{aligned} \tag{7}$$

This hamiltonian can be treated in a usual manner, either by applying a Bogolyubov transformation to rotate the fields to eigenstates of (7), or by introducing normal and anomalous Green's functions:

$$G_{dd}(x,x') = -\langle T d_\uparrow(x)\, d_\uparrow^+(x')\rangle, \tag{8a}$$
$$G_{fd}(x,x') = -\langle T f_\uparrow(x)\, d_\uparrow^+(x')\rangle, \tag{8b}$$
$$F_{dd}^+(x,x') = \langle T d_\uparrow^+(x)\, d_\downarrow^+(x')\rangle, \tag{8c}$$
$$F_{fd}^+(x,x') = \langle T f_\downarrow^+(x)\, d_\uparrow^+(x')\rangle. \tag{8d}$$

Here, $G_{ff}$, $G_{dd}$ are normal Green's functions, $G_{fd}$ describes the effect of (bare) hybridization and excitonic pairing, $F_{fd}$ and $F_{dd}$ are the Gor'kov Green's functions corresponding to the singlet inter- and intraband superconducting pairings. The definition of the Green's functions for interchanged spin and band indices is straightforward.

In the Matsubara representation, the system of equations for the Green's functions at $T\neq 0$ has the following form (see Ref. 17):

$$\begin{pmatrix} (i\omega_n-\varepsilon_d) & -\tilde{V}^* & 0 & \Delta \\ -\tilde{V} & (i\omega_n-\varepsilon_f) & \Delta & 0 \\ 0 & \Delta^* & (i\omega_n+\varepsilon_d) & \tilde{V} \\ \Delta^* & 0 & \tilde{V}^* & (i\omega_n+\varepsilon_f) \end{pmatrix} \begin{pmatrix} G_{dd} \\ G_{fd} \\ F_{dd}^+ \\ F_{fd}^+ \end{pmatrix} = \begin{pmatrix} 1 \\ 0 \\ 0 \\ 0 \end{pmatrix}. \tag{9}$$

The energy spectrum (the dispersion) is given by the roots of the determinant of (9): substituting real frequencies yields

$$\begin{aligned} \omega_\pm^2(k) = &\frac{\varepsilon_+^2(k)+\varepsilon_-^2(k)}{4} + |\Delta|^2 + |\tilde{V}|^2 \pm \tfrac{1}{2}[\varepsilon_+^2(k)\,\varepsilon_-^2(k) \\ &+ 4|\Delta|^2\varepsilon_-^2(k) + 4|\tilde{V}|^2\varepsilon_+^2(k)]^{1/2}, \end{aligned} \tag{10}$$

where

$$\varepsilon_\pm(k) = \varepsilon_d(k) \pm \varepsilon_f(k).$$

The energy gap is a combination of the superconducting and excitonic gaps. For instance, at the Fermi momentum $k=k_F$ and $\varepsilon_f=\varepsilon_d=0$, $\omega^2=|\Delta|^2+|\Sigma|^2$. It can also be seen, that for $\Delta$ or $\tilde{V}=0$, the spectrum (10) reduces to the well known spectrum of hybridized bands and an ordinary superconductor respectively. One should bear in mind however, that for $m_f \neq m_d$, the actual gap in the energy spectrum is an indirect one, and smaller than $[|\Delta|^2+|\Sigma|^2]^{1/2}$ [14, 18].

In the following, the parameters $\Delta$ and $\tilde{V}$ will be determined selfconsistently. The corresponding selfconsistency equations can be obtained from the respective Green's functions $F_{fd}^+$, $G_{fd}$ and have the form

$$\Delta^* = -\frac{J}{2}\Delta^* T\sum_{\omega_n}\sum_k \cdot\left\{\frac{[i\omega_n+\varepsilon_d(k)][i\omega_n-\varepsilon_f(k)]-|\tilde{V}|^2-|\Delta|^2}{[\omega_n^2+\omega_+^2(k)][\omega_n^2+\omega_-^2(k)]} + (f\leftrightarrow d)\right\}, \tag{11}$$

$$\tilde{V} = V - \frac{J}{2}\tilde{V} T\sum_{\omega_n}\sum_k \cdot\frac{[i\omega_n+\varepsilon_f(k)][i\omega_n+\varepsilon_d(k)]-|\tilde{V}|^2-|\Delta|^2}{[\omega_n^2+\omega_+^2(k)][\omega_n^2+\omega_-^2(k)]}. \tag{12}$$

The concrete results depend in an essential way on the dispersion laws of the $f$- and $d$-bands. We shall study the cases of two electron bands or of an electron and a hole band separately.

### *A. Two Electron Bands*

First, we study the case of two electron bands, i.e. $m_f, m_d > 0$ (see Fig. 1a). The self-consistency equations (11) and (12) will be analyzed near $T_c$, where $\Delta \ll 1$. Furthermore, it is assumed that $V$ and $\tilde{V}$ are small. Developing (11) and (12) in powers of $\Delta$ and $\tilde{V}$, one obtains after summation over $\omega_n$ and $k$

$$\Delta^* = \Delta^* \left\{ \frac{JN_d(0)}{1+\lambda} \ln \frac{2\gamma\lambda^{1/2} W_d}{\pi T} - \frac{JN_d(0)}{2} \frac{7\zeta(3)}{4\pi^2 T_c^2} \frac{\lambda}{(1+\lambda)^3} |\Delta|^2 - JN_d(0) \frac{\lambda}{(1+\lambda)^2} \frac{7\zeta(3)}{4\pi^2 T_c^2} |\tilde{V}|^2 \right\}, \tag{13}$$

$$\tilde{V} = V - \tilde{V} \left\{ \frac{JN_d(0)}{4(1-\lambda)} \ln \lambda + \frac{JN_d(0)}{2} \frac{\lambda}{(1+\lambda)^2} \frac{7\zeta(3)}{4\pi^2 T_c^2} |\Delta|^2 \right\}. \tag{14}$$

From (13) and (14) the following conclusions can be drawn:

*(i)* For $\tilde{V}=0$ one obtains from (13) the usual expression (3) for $T_c$ (remember that $\lambda = W_f/W_d = m_d/m_f$);

*(ii)* In contrast to the equation for $\Delta$, coefficients of the linear terms in $\tilde{V}$ in (14) are not divergent. This means, that for a weak coupling (and very small $\lambda$), there is no spontaneous excitonic pairing in the absence of "external" hybridization. At the same time, when $V \neq 0$, there always occurs a mixing of the bands. This mixing becomes stronger due to the excitonic contribution $(J/2)\langle df^+ \rangle$ to $\tilde{V}$. If $\lambda \to 0 (m \to \infty$, flat $f$-band), the approximation breaks down, and one has to use parquet techniques (see below and Ref. 19). We also note, that the coefficient of the term $|\tilde{V}|^2$ in (14) is exactly zero (all the poles in the corresponding expression obtained from (12) lie in the same half-plain);

*(iii)* As can be see from (13) and (14), the mutual influence of different order parameters $\Delta$ and $\tilde{V}$ is destructive and they suppress one another (an increase of $\tilde{V}$ leads to the decrease of $\Delta$ and vice versa). From (13) one can determine the decrease of the superconducting transition temperature by hybridization:

$$\frac{\Delta T_c}{T_c} = -\frac{\lambda}{1+\lambda} \frac{7\zeta(3)}{4\pi^2 T_c^2} |\tilde{V}|^2. \tag{15}$$

It is easy to understand this result qualitatively. In our case of complete nesting, the hybridization admix states near $\varepsilon_F$ suitable for pairing with the "wrong" states. Then, part of the averages $\langle f^+ d^+ \rangle$ are changed for intraband terms $\langle f^+ f^+ \rangle$ and $\langle d^+ d^+ \rangle$ which, with our choice of interaction (1), are inefficient for superconductivity. This conclusion correlates with that of Menezes [20].

As a collorary it can be inferred, that for $\tilde{V}=0$, there exists only interband pairing, whereas nonzero $\tilde{V}$ induces intraband pairing: the respective Gor'kov functions $F_{ff}$, $F_{dd}$ are nonzero, although of higher order in $\tilde{V}\Delta$.

The above results can be presented more transparently by writing down the Landau free energy functional:

$$\mathscr{F} = \tfrac{1}{2} a \Delta^2 + \tfrac{1}{4} b \Delta^4 + \tfrac{1}{2} a' \tilde{V}^2 + \tfrac{1}{4} b' \tilde{V}^4 + \tfrac{1}{2} c \Delta^2 \tilde{V}^2 - d V \tilde{V}. \tag{16}$$

Here, $\Delta$ and $\tilde{V}$ are assumed real. This functional (and the free energy for arbitrary $T$), can be obtained from the Green's functions in a standard way. For our present purposes, the exact form of is unnecessary. It suffices to note, that *(i)* as follows from the results presented above (see (13) and (14)), the coefficient $a$ passes through zero, $a = \alpha(T - T_c)/T_c$, with $T_c$ given by (3); *(ii)* as follows from (14), for weak coupling, the coefficient $a' > 0$; and, most important of all, *(iii)* the coefficient $c$ describing the coupling of the two order parameters $\Delta$ and $\tilde{V}$ is positive. Moreover *(iv)*, the coefficient $b' = 0$.

Minimizing the free energy with respect to $\Delta$ and $\tilde{V}$, yields

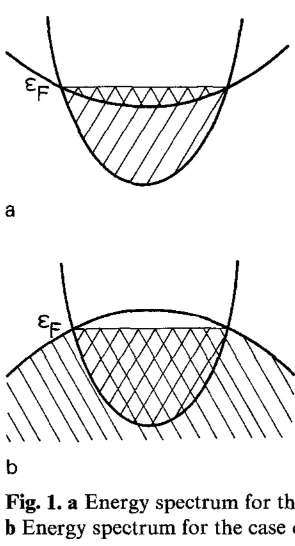


**Fig. 1. a** Energy spectrum for the case of two electron bands. **b** Energy spectrum for the case of electron and hole bands

$$\Delta^2 = -\frac{a(T)+c\tilde{V}^2}{b}; \tag{17}$$

$$\tilde{V}^2 = \frac{dV}{a'+c\Delta^2}. \tag{18}$$

The expressions (15) and (16) exactly correspond to (13) and (14) and all the qualitative conclusions obtained above may be easily translated into this formulation. Hence it can be inferred, that superconducting and excitonic averages suppress each other. This is due to the fact, that the coefficient $c$ in (16)–(18) is positive. There exists a spontaneous superconducting ordering, since $a(T)$ passes through zero, but no such ordering in $\tilde{V}(a'>0)$. Furthermore, nonzero $\tilde{V}$ decreases $T_c$.

### B. Electron and Hole Bands

Let us suppose now, that the Fermi level lies near the top of the $f$-band (see Fig. 1b). In this case the energy spectrum may be of the form (5), but with $m_f<0$. This is the canonical configuration of the excitonic insulator problem.

It is easy to verify, that compared to the situation in the preceding section, the interband superconducting and excitonic pairings change place: now there exists an instability in the excitonic channel, since the coefficient $a'(T)$ in (16) passes through zero at a temperature $T_{c,\mathrm{exc}}$

$$T_{c,\mathrm{exc}} = 1.14(W_f\, W_d)^{1/2}\, \mathrm{e}^{-\frac{2(1+\lambda)}{JN_d(0)}}. \tag{19}$$

The superconducting channel is finite and regular. As follows from the results presented above, the superconducting and excitonic pairings suppress each other respectively. In the presence of a (bare) hybridization $V$, the excitonic phase with an insulating gap will survive even for temperatures $T>T_{c,\mathrm{exc}}$, very similar as for the case of a ferromagnet in a magnetic field. As there is no external field associated with the superconducting order parameter (as is the (bare) hybridization $V$ with respect to the excitonic averages), pure excitonic ordering without superconducting admixtures occurs [17].

There actually exist compounds which display properties similar to such a phase. These are mixed valence compounds of the type $SmB_6$ and the gold phase of SmS. In these systems, the integral valence phase would be (and in the case of black SmS actually is) semiconducting. In the mixed valence phase, all the $d$-electrons have been promoted to the $d$-band from the $f$-level. Hence, the necessary condition for a complete excitonic pairing $N_{f,\,\mathrm{holes}}=N_{d,\,\mathrm{electrons}}$ is automatically satisfied. In fact it turns out, that exactly those compounds for which above requirement is fulfilled, exhibit a small gap (of the order of 50 K) in the mixed valence ground state. We believe, that these compounds are to be treated as excitonic insulators. The origin of the energy gap in their excitation spectrum is not just the effect of direct (bare) hybridization, but the combined action of both bare hybridization and excitonic pairing [21, 22]. Experimental confirmation of this perception can be found in [23].

## 3. Concluding Remarks

Summarizing it can be said, that depending on the details of the spectrum, the model (1) is capable of explaining the formation of different types of orderings in valence fluctuation systems. These orderings include interband ("hybrid") superconductivity, as well as excitonic semiconducting ones. In the introduction we have already discussed some appealing features for a description of heavy fermion superconductivity. Similarly, this approach seems quite appropriate for the description of small gap mixed-valence systems. It permits the explanation of recent experiments on such structures [23].

As to the mutual influence of these two possible types of ordering, we have shown, that in our model they are competitive and in general suppress each other.

It should however be stressed, that these findings were obtained for a simplified model of wide enough bands with complete nesting, using a mean field approximation. When the $f$-band is nearly flat (which corresponds to $m_f\to\infty$), then both the superconducting and the excitonic channels will be singular. In this case, the mean field approximation is inappropriate. A more appropriate formalism would then be the parquet approximation or even more sophisticated techniques [13, 19, 24]. In this case, superconducting and excitonic pairings may coexist and their mutual influence may be different. This situation needs a very subtle treatment. Although complicated, such calculations could provide the most appropriate approach to heavy fermion superconductivity. The considerations presented above could be understood as a preliminary step towards a comprehensive understanding of this fascinating phenomenon.

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