Heavy-Fermion Behavior in a Negative-U Anderson Model

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We show that the low-temperature fixed-point behavior of the negative-U Anderson model involves a "charge Kondo effect," where the local pair behaves as a *Heisenberg* rather than an *XY* degree of freedom. Interactions of the local pair with the conduction sea generate a highly polarizable Fermi liquid with enhanced linear specific heat and charge *and* pair susceptibilities.

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The dramatic role played by slow spin fluctuations in the development of strongly correlated electron behavior is well known, in the context of both d- and f-electron systems.[1,2] These effects are often understood within the framework of the Brinkman-Rice[2] scenario for the approach to a Mott transition, whereby the development of slow spin fluctuations gives rise to an almost incompressible Fermi liquid with a large enhancement of the linear specific heat and spin susceptibility, loosely termed, a "heavy-fermion" ground state. The essence of this phenomenon is the development of a highly degenerate manifold of local moment states: The splitting of this degeneracy at low temperatures is accompanied by the formation of a highly correlated electron fluid.

Can analogous behavior occur in response to slow charge fluctuations? Early interest in such a possibility occurred in the context of the mixed-valence phenomenon.[3,4] In this case, however, the bulk of evidence now suggests that the root of strong renormalization effects is the low-frequency *spin* fluctuations that are generated by virtual charge fluctuations.[5,6] The well-known case of polaronic conductors also fails to provide us with a charge analog of the Brinkman-Rice phenomenon, there being no intrinsic degree of freedom to play the role of a local moment. We have reexamined this question, considering the effects of slow pair fluctuations between singlet states differing by two units of charge. Our most important conclusion is that in a two-band environment, local pair formation bears close analogy to local moment formation. Internal charge degrees of freedom of a pair severely modify the nature of the fluid that results, leading to a charge analog of heavy-fermion conductors.

The dynamics and formation of pairs in a two-band environment may be described by a negative-U Anderson model,

$$H=\sum_{\mathbf{k}\sigma}(\epsilon_{\mathbf{k}}-\mu)c^{\dagger}_{\mathbf{k}\sigma}c_{\mathbf{k}\sigma}+\sum_{j\sigma}V[c^{\dagger}_{k\sigma}d_{j\sigma}e^{i\mathbf{k}\cdot\mathbf{R}_j}+\text{H.c.}] +(E_d-\mu-U/2)n_d-(U/2)(n_d-1)^2, \quad (1)$$

where $d^{\dagger}_{j\sigma}$ creates an electron localized at site j, $U>0$, and E_d is the energy of an electron in the localized "d state." The attractive U could have a variety of microscopic origins, such as a charge disproportionation in a valence-skipping compound,[7,8] exchange of a low-energy bosonic excitation, or a more complex many-body phenomenon. We are interested in the possibility of generic features that arise from strong localized pairing interaction that may be separated from the detailed origins of the interaction and the precise identification of the localized "d" state.

Consider the atomic limit of this model with one d site per unit cell. As the chemical potential μ passes through the special value $\mu_0=E_d-U/2$ the total electronic charge per unit cell $N(\mu)$ will jump by 2,

$$N(\mu_0^+)-N(\mu_0^-)=2,$$

due to occupation of the d^2 state. If the filling of the electron sea N lies in the range $N(\mu_0^-)<N<N(\mu_0^+)$, the chemical potential is pinned to the value $\mu=\mu_0$ (Fig. 1), where the d^0 and d^2 states are *degenerate*. Hybridization generates tunneling within this degenerate manifold, similar to the spin fluctuations that occur between up and down configurations of a local moment in a re-

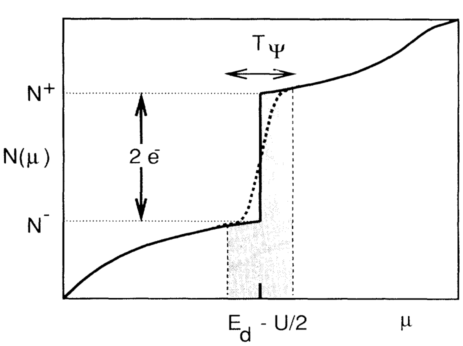

FIG. 1. The jump in the occupancy of a pair state that occurs at the symmetric point of the negative-U Anderson model. The charge Kondo effect smears this jump out over an energy scale T_Ψ.

pulsive-U Anderson model. We are thus led to consider the possibility of a "charge Kondo effect,"[9] where by analogy with the positive-U case, the charge of the local d states will become dynamically screened by low-frequency pair fluctuations, generating a highly polarizable electron fluid.

To simplify matters, first consider an isolated symmetric negative-U Anderson impurity, where interactions between different localized pairs are neglected. For a perfectly particle-hole symmetric band, this model is isomorphic to its positive-U counterpart,[10-12] and the mapping

$$d_\downarrow^\dagger \to -d_\downarrow, \quad c_{k\downarrow}^\dagger \to c_{-k\downarrow} \tag{2}$$

transforms the symmetric repulsive-U Anderson model into the corresponding attractive-U model, interchanging pair and spin degrees of freedom:

$$\mathbf{S}_d \leftrightarrow \mathcal{T}_d .$$

Here $\mathbf{S}_d$ and $\mathcal{T}_d = \frac{1}{2}\tilde{d}^\dagger \boldsymbol{\tau} \tilde{d}$ are the impurity spin and isospin, respectively; $\tilde{d}^\dagger \equiv (d_\uparrow^\dagger, -d_\downarrow)$ is a Nambu spinor for the d electron and $\boldsymbol{\tau} \equiv (\tau^1, \tau^2, \tau^3)$ denotes Pauli matrices in particle-hole space. The third component of the isospin is the atomic charge $\mathcal{T}_d^3 = \frac{1}{2}(n_d - 1)$, while transverse components describe the pair amplitude and phase $\mathcal{T}_d^+ = d_\uparrow^\dagger d_\downarrow^\dagger$.

Properties of the negative-U Anderson model change markedly below temperatures of order U, due to a crossover associated with the suppression of charge $1e^-$ valence fluctuations (Fig. 2). In the repulsive-U model this crossover is the origin of local moment formation; for the negative-U case, a special kind of "local pair formation" occurs. Unlike the small polaron problem, here the model is symmetric, so the chemical potential for pair creation is zero: The degenerate d^2 and d^0 states are like "up" and "down" states of a local moment. At low energies, the local pair therefore behaves as a Heisenberg "isospin," interacting with the conduction sea via pair exchange: $d^2 \rightleftharpoons d^0 + 2e^-$. This low-energy regime is described by a Kondo isospin model,[8] derived by a Schrieffer-Wolff[13] transformation, as in the positive-U model,[14]

$$H = \sum E_{\mathbf{k}} c_{\mathbf{k}\sigma}^\dagger c_{\mathbf{k}\sigma} + 2J \mathcal{T}_c \cdot \mathcal{T}_d , \tag{3}$$

where $J = 2V^2/U$ and $\mathcal{T}_c = \frac{1}{2}\Psi(0)^\dagger \boldsymbol{\tau} \Psi(0)$ is the conduction-electron isospin at the impurity site, written in terms of the Nambu spinor $\Psi^\dagger(0) = \sum_k (c_{k\uparrow}^\dagger, c_{-k\downarrow})$. The most unexpected feature of this Hamiltonian is the "antiferromagnetic" interaction between conduction electrons and the localized pair. This interaction grows at low temperatures, inducing increasingly strong pair fluctuations, until ultimately the conduction-electron pairs bind to the local pair, forming a neutral object: an isospin singlet.

In a symmetric conduction band, the total isospin $\mathcal{T} = \mathcal{T}_d + \mathcal{T}_c$ is conserved, so charge and pair susceptibilities are *equal*. At high temperatures this system will exhibit Curie law charge *and* pair susceptibilities with small logarithmic corrections

$$\chi_c = \chi_p = \frac{1}{4T}\left\{1 - 2J\rho\left[1 + 2(J\rho)\ln\left(\frac{T}{U}\right)\right] + \cdots\right\} \tag{4}$$

due to the partial "quenching" of the impurity isospin. Pair fluctuations will drive a logarithmic growth of resistivity as the temperature is lowered. At the pair fluctuation temperature

$$T_\Psi = U(\pi J\rho)^{1/2} \exp[-1/2J\rho] , \tag{5}$$

the isospin of the impurity becomes coherently admixed with the isospin of the conduction band, binding a pair degree of freedom from the conduction sea to form an isospin singlet.

Below T_Ψ there will be a second crossover into a strongly correlated Fermi liquid with an Abrikosov-Suhl scattering resonance at the Fermi energy which elastically scatters holes and particles with unitary phase shift

$$\delta_e = \delta_h = \pi/2 . \tag{6}$$

The width of the resonance is of order T_Ψ, and charge and pair susceptibilities are

$$\chi_c = W/4\pi T_K , \tag{7}$$

where $W/4\pi = 0.1023\ldots$ is the universal Wilson number.[15] The "Wilson ratio" of pair susceptibility to linear specific-heat capacity, in dimensionless units, is

$$R = \chi_c/\gamma = 2 , \tag{8}$$

showing complete suppression of local *spin fluctuations.*

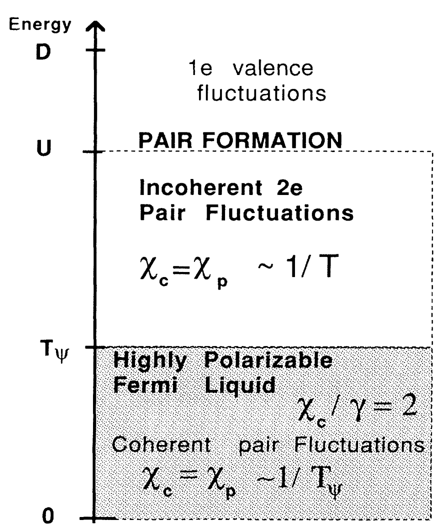

FIG. 2. The flow from high-energy valence fluctuations to a low-energy Fermi-liquid state in the negative-U Anderson model.

Since the strong-coupling fixed point is neutral, departures from perfect isospin symmetry are generally irrelevant in the one-impurity model, and can actually enhance this phenomenon. A conduction-band particle-hole symmetry can be recast as a magnetic-field term coupled to the conduction-band isospin. Providing this is small on a scale of the bandwidth, this produces no significant effects. More important still, however, a Coulomb screening will actually tend to enhance the charge Kondo effect. A Coulomb screening term $H_C = Vn_d n_c$ between the electrons and local pair enhances the Ising component of the spin exchange $J_z \to J+4V$. As in the spin Kondo effect, this term drives the initial Hamiltonian further along the scaling trajectory towards strong coupling. So actually, the local pair behaves increasingly like a Heisenberg isospin at low temperatures rather than a charged boson with no internal degree of freedom, and the charge Kondo effect in the symmetric model is robust against conduction-band asymmetries.

Can this phenomenon survive in the lattice once the interactions between local pairs are included? To study this possibility, we write the low-energy degrees of freedom of a negative-U Anderson lattice as an isospin Kondo lattice[16] model,

$$H=\sum_{\mathbf{k}\sigma}(\epsilon_{\mathbf{k}}-\mu)c_{\mathbf{k}}^{\dagger}\tau_3 c_{\mathbf{k}}+J\sum_j \mathcal{T}_d(j)\cdot c_j^{\dagger}\boldsymbol{\tau}c_j\,. \quad (9)$$

Here $c_j^{\dagger}=(c_{j\uparrow}^{\dagger},c_{j\downarrow})$ and $c_{\mathbf{k}}^{\dagger}=(c_{\mathbf{k}\uparrow}^{\dagger},c_{-\mathbf{k}\downarrow})$ are conduction-electron spinors in the position and momentum basis, respectively. Unlike the impurity model, isospin conservation cannot be taken for granted, for even in a band with a symmetric density of states, degenerate electron and hole states *never* lie in the same region of momentum space.

Fully fledged "isospin conservation" actually develops in the vicinity of a nesting instability of a half-filled band. Consider the special case of a bipartite lattice with strong nearest-neighbor hybridization. To a good approximation, near half filling, the degenerate electron and hole states are separated by an amount $\mathbf{Q}$ in momentum space over most of the Brillouin zone, so $\epsilon(\mathbf{k}) \approx -\epsilon(\mathbf{k}-\mathbf{Q})$, where $\mathbf{Q}$ is the zone center. Exact equality occurs in the case of perfect nesting, and, in this special case, the staggered component of the transverse isospin $\mathcal{T}^{\perp}(\mathbf{Q})$ is conserved. By applying the gauge transformation $c_{j\sigma}\to e^{i\theta_j/2}c_{j\sigma}$, $d_{j\sigma}\to e^{i\theta_j/2}d_{j\sigma}$, where $\theta_j = \mathbf{Q}\cdot\mathbf{R}_j$, the x and y isospin axes are rotated through 180° about the z axis, so the staggered component of the transverse isospin transforms into the uniform transverse isospin $\mathcal{T}'^{\perp}(\mathbf{q})=\mathcal{T}^{\perp}(\mathbf{q}+\mathbf{Q})$. After this transformation, the Hamiltonian becomes

$$H=\sum_{\mathbf{k}\sigma}c_{\mathbf{k}}^{\dagger}(\tilde{\epsilon}_{\mathbf{k}}-\mu_{\mathbf{k}}\tau_3)c_{\mathbf{k}}+2J\sum_j \mathcal{T}_d'(j)\cdot\mathcal{T}_c'(j)\,, \quad (10)$$

where

$$\tilde{\epsilon}_{\mathbf{k}}=[\epsilon_{\mathbf{k}-\mathbf{Q}/2}-\epsilon_{\mathbf{k}+\mathbf{Q}/2}]/2\,,\quad \mu_{\mathbf{k}}=\mu-[\epsilon_{\mathbf{k}-\mathbf{Q}/2}+\epsilon_{\mathbf{k}+\mathbf{Q}/2}]/2 \quad (11)$$

are the symmetric and asymmetric components of the band structure. This is now precisely a Kondo lattice model, where isospin replaces spin. The term $\mu_{\mathbf{k}}$ plays the role of a momentum-dependent magnetic field that breaks an otherwise perfect isospin conservation. Fortunately, we do not require that this term be identically zero, which would imply perfect nesting and probably the formation of an insulator. The condition for a large Kondo energy T_Ψ is significantly weaker, and demands merely that the average magnitude of $\mu_{\mathbf{k}}$ be small compared with the bandwidth: a condition that is satisfied quite far away from perfect nesting. In the discussion that follows we accordingly take the delicate point of view that $\mu_{\mathbf{k}}$ can be treated as a finite, but irrelevant perturbation.

In this lattice, as in magnetic Kondo systems, superexchange processes occur, which here induce exchange pair coupling (SPC) between local pairs,

$$H'=\tfrac{1}{2}\sum_{i,j}K(\mathbf{R}_i-\mathbf{R}_j)\mathcal{T}_i'\cdot\mathcal{T}_j'\,. \quad (12)$$

This is the analog of the RKKY interaction, and contains a long-range oscillatory component of the form $(J^2\rho)\cos\mathbf{Q}\cdot\mathbf{R}/R^3$. When the SPC is large compared with T_Ψ an ordered charge-density wave $[\langle\mathcal{T}'^3(\mathbf{Q})\rangle = \tfrac{1}{2}\rho_{\mathbf{Q}}]$ or superconducting ground state $[\langle\mathcal{T}'^+(\mathbf{Q})\rangle = \langle\mathcal{T}^+(0)\rangle=\Psi]$ will result. In this case the low-energy isospin dynamics are anisotropic due to Coulomb interactions and band effects, falling into the XY class in the superconductor, and the Ising class for charge-density wave (CDW).

When the frequency of pair fluctuations T_Ψ becomes large, the properties of the conductor become severely modified. As the Kondo fixed point is approached, the renormalized isospin interactions will tend to become more *isotropic*: The system of local pairs will behave more like a fluid with a *Heisenberg* charge degree of freedom, in which the charge and pairing amplitudes behave as *three components of one fluctuating vector*.

Once T_Ψ actually exceeds the SPC, this system will form the charge analog of a Kondo lattice ground state, with a highly polarizable narrow band of quasiparticles of width $\sim T_\Psi$. The mass of the heavy quasiparticles will scale approximately with the charge susceptibility,

$$m^*/m \sim \chi_c\rho\,. \quad (13)$$

Unlike a heavy-fermion compound, charge currents in the isospin Kondo lattice contain a new, pair hopping component. By gauge invariance, the uniform pair current is then

$$\mathbf{J}=-\partial H'[\mathbf{A}]/\partial\mathbf{A} = -2e\sum_{\mathbf{q}}\nabla K(\mathbf{q})[\hat{\mathbf{z}}\cdot(\mathcal{T}_{\mathbf{q}}\times\mathcal{T}_{-\mathbf{q}})]\,, \quad (14)$$

where $K(\mathbf{q})$ is the Fourier transform of $K(\mathbf{R})$. High-temperature pair motion in the "paramagnetic" regime will be diffusive, with conductivity given by the Einstein relation $\sigma_p \sim \mathcal{D}\chi_c$, where $\mathcal{D}$ is the pair diffusion constant.

Pair conductivity will therefore scale like the Curie pair susceptibility $\sigma_p \sim 1/T$. Kondo scattering of the conduction current will generate an additional logarithmic component to the conductivity[1] $\rho \sim \ln(U/T)$, so we expect the high-temperature resistance to contain a linear and a logarithmic term $\rho(T) \sim T - \ln T$.

From our knowledge of the low-temperature phases of heavy-fermion systems we may use the duality between spin and isospin to discuss possible low-temperature phases of our model. Just as *s*-wave superconductivity is suppressed in favor of exchange-driven anisotropic *d*-wave superconductivity in a Kondo lattice, conventional magnetism is suppressed in an isospin Kondo lattice, in favor of off-diagonal *spin nematic* order with order parameter $\langle d_{\mathbf{k}}^\dagger \boldsymbol{\sigma} d_{\mathbf{k}} \rangle \sim \Delta_{\mathbf{k}} \hat{\mathbf{n}}$, where $\Delta_{\mathbf{k}}$ has d symmetry.[17] Similarly, the analog of magnetic heavy-fermion phases will be superconducting or CDW states that coexist with the highly polarizable heavy-fermion phases. More detailed considerations of these phenomena can in fact be carried out within a functional integral. The low-energy Lagrangian for this model is almost identical with the spin Kondo lattice,[18] the only difference being that the constraint acts on the d spin, setting $\mathbf{S}_d = 0$ at each site. For the lattice the mean-field ground state is a Fermi liquid,

$$|\Psi\rangle = P \prod_{\mathbf{k}\sigma} (\alpha_{\mathbf{k}} c_{\mathbf{k}\sigma}^\dagger + \beta_{\mathbf{k}} d_{\mathbf{k}\sigma}^\dagger)|0\rangle , \qquad (15)$$

where the hybridization coefficients are those appropriate to a renormalized resonant d band; P projects the d-spin singlet component at each site.

Lastly, we mention some potential realizations of this phenomenon. We have specifically discussed the case of degenerate, highly localized charge-fluctuation states. In the strictest interpretation, such a state of affairs can probably only occur close to a chemical instability, as in the group-III or group-V elements, which have a propensity towards valence skipping.[7,8] For instance, Kondo scattering may play a central role in the saturated resistance of the $BaBiO_3$-based superconductors. Equally interesting, however, is the relationship of our example to a more general class of problem where high-energy processes force a flow towards a particle-hole symmetric fixed point. It may be possible to regard the *A*15 superconductors[19] in this vein, following the observation of Yu and Anderson, that the high-temperature saturated resistance is reminiscent of Kondo scattering.

To conclude, we have analyzed a scenario for the formation of a highly polarizable heavy-fermion state: the antithesis of the Brinkman-Rice liquid. In this fluid, strong correlations are generated amidst a fluid of slow charge and pair fluctuations, giving rise to an enhanced linear specific heat and charge and pair susceptibilities. In contrast to the Brinkman-Rice fluid, spin fluctuations are almost entirely suppressed.

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