Magnetic, nonmagnetic, and non-Fermi-liquid ground state of an impurity fluctuating between two magnetic configurations

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The ground state of a generalized Anderson model for valence fluctuations between two magnetic p, d, or f configurations is discussed. In the isotropic limit, in contrast to the ordinary Anderson model, the localized problem (limit of zero bandwidth) does not correspond to a stable fixed point, except in the extreme case of $j-j$ coupling. Taking the simplest possible case (fluctuations between p^4 and p^3 or p^5) under a tetragonal (C_{4v}) crystal field, depending on the retained low-energy multiplets, the model can be mapped into an ordinary Anderson model, another model with a magnetic ground state, and for particular parameters, a two-channel Kondo model.

The unusual normal state properties of high-T_c superconductors can be explained by a non-Fermi liquid (NFL).[1] Also several strongly correlated f-electron materials display NFL behavior in their low-temperature specific heat, magnetic susceptibility, and electrical resistivity.[2–5] Thus, the study of microscopic models with NFL behavior, particularly if they are relevant to real systems, is of great interest at present.[6–9] The spin-1/2 two-channel Kondo model[10] (TCKM) has been solved exactly[11] and is consistent with several properties observed in $Y_{0.8}U_{0.2}Pd_3$ (Refs. 2 and 12) and other U alloys.[4] Renormalization-group calculations show that the low-temperature properties of the overscreened multichannel Kondo model are characterized by an intermediate-coupling fixed point[13,14] with NFL behavior.[14,11,15] It is interesting that an analytic approach valid for large spin and number of channels reproduces the leading critical exponent in all cases.[15]

The Anderson model for dilute magnetic alloys and its integer valent limit, the Kondo model, were used to explain several properties of strongly correlated f-electron materials. The scaling behavior of these models is known, and in contrast to the TCKM, the low-temperature properties are described by the strong-coupling fixed point (SCFP) (infinite value of the coupling constant V or J) with a nonmagnetic ground state and Fermi-liquid behavior.[16–19] The SCFP is characterized by a Hamiltonian $H_0+H'_{\rm band}$, where H_0 contains the impurity and the innermost Wannier function of the conduction electrons and $H'_{\rm band}$ describes the rest of the conduction electrons.[17–19] Exact solutions of the Kondo and Anderson impurity models and several generalizations were obtained using the Bethe ansatz.[20–23] All Bethe ansatz solvable models for valence fluctuations between two magnetic configurations have a magnetic ground state,[22] which corresponds to a SCFP similar to that described above, but with a degenerate ground state of H_0.[24] However, no rigorous results exist for the generalized Anderson model which describes an impurity in an isotropic medium fluctuating between two *realistic magnetic* configurations.

In this paper we study the possibility that at very low temperatures, the behavior of the above-mentioned generalized Anderson model is described by a stable SCFP of the form $H_0+H'_{\rm band}$. If this were the case, the degeneracy of the ground state of the local Hamiltonian H_0 would determine the magnetic or nonmagnetic character of the ground state of the system, and the latter would necessarily be a Fermi liquid at low temperatures. However, it turns out that in general, the model for infinite hybridization V is highly non trivial, and an eventual "local" SCFP of the form $H_0+H'_{\rm band}$ is unstable. As a result it cannot be concluded for the general isotropic case whether the ground state is magnetic or not, or whether the low-temperature excitations are those of a Fermi liquid. We show, however, mapping the model into simpler effective Hamiltonians, that the simplest case for valence fluctuations between two magnetic configurations under a strong tetragonal (C_{4v}) crystal field can lead to different behaviors, including a NFL one, depending on the parameters. Previously, Cox has derived selection rules for the applicability of the TCKM to U^{4+} and Ce^{3+} ions in metals.[25]

The generalized Anderson model for an impurity ion in an isotropic medium has the form:[26,27,21]

$$H=H_{\rm band}+H_{\rm ion}+H_{\rm mix}. \tag{1}$$

$H_{\rm band}$ describes the extended states with the same orbital angular momentum l as that of the impurity shell. Calling k,μ, and σ the radial momentum, orbital angular momentum, and spin, respectively, one has

$$H_{\rm band}=\sum_{k\mu\sigma}\epsilon_k c^\dagger_{k\mu\sigma}c_{k\mu\sigma}=\sum_{kjm}\epsilon_k c^\dagger_{kjm}c_{kjm}. \tag{2}$$

The last member is an alternative expression in terms of the total angular momentum $j=l\pm(1/2)$ and its projection m. The impurity shell is described by $H_{\rm ion}$. We retain only the ground state multiplets of two neighboring l^n and l^{n+1} configurations,

$$H_{\rm ion}=E\sum_{M_0}|J_0M_0\rangle\langle J_0M_0|+(E+\Delta)\sum_{M_1}|J_1M_1\rangle\langle J_1M_1|, \tag{3}$$

where J_i,M_i are the total angular momentum and projection of the l^{n+i} configuration. The hybridization part can be written in the following equivalent forms:

$$\frac{H_{\rm mix}}{V}=\sum_{k\mu\sigma} c^{\dagger}_{k\sigma} f_{\mu\sigma}+\text{H.c.}=\sum_{kjm} c^{\dagger}_{kjm} f_{jm}+\text{ H.c.}$$
$$=\sum_{kjmM_0M_1} a_j\langle J_0 j M_0 m|J_1M_1\rangle(c^{\dagger}_{kjm}|J_0M_0\rangle\langle J_1M_1| +\text{H.c.}) \quad (4)$$

where the $\langle J_0 j M_0 m|J_1M_1\rangle$ are Clebsch-Gordan coefficients and the a_j are two numbers which depend on the particular form of the highly correlated ionic states.[26,27,21] When $J_0\neq 0$ and $J_1\neq 0$, both $a_j\neq 0$ except in the extreme (unrealistic) case of pure j-j coupling, where for a configuration with n electrons, $a_{l+1/2}=0$ if $0\leq n\leq 2l-1$ and $a_{l-1/2}=0$ if $2l\leq n\leq 4l+2$. For the more realistic LS coupling and Hund rules, the expressions of the a_j are given and their values are tabulated for all cases of valence fluctuations between two magnetic p,d, or f configurations in Ref. 21.

The model cannot be solved with the Bethe ansatz except when only $a_{1/2}$ is different from zero[28] and the resulting ground state is magnetic if both configurations are magnetic.[21,22] For the interesting case in which $J_0,J_1,a_{l-1/2}$, and $a_{l+1/2}$ are all different from zero, renormalization-group calculations have not been done, probably due to the fact that the Hilbert space is enlarged by a factor 2^{4l+2} in Wilson's renormalization group.[17–19] The narrow-band limit H_0 [ϵ_k equal to the chemical potential for all k, and any Δ (Ref. 29)] for Hund rules multiplets has been solved by Baliña and me using exact diagonalization.[21,29,30] The results can be summarized as follows. For configurations p^n and p^{n+1}, only for $n=3$ and $n=4$ are both configurations magnetic, with $J=2$ ($J=3/2$) for an even (odd) number of electrons according to Hund rules. Since J_0 and J_1 can be interchanged by a special electron-hole transformation[21,22] both cases are equivalent. For $n=4$ the ground state consists of two singlets with total number of particles $N=n+2$ and $n+4$ and one doublet with the number of conduction electrons at the innermost Wannier function $n_c=N-n=3$.[28] The results for two magnetic d^n and d^{n+1} configurations (n = 1,2 and 5 to 8) are qualitatively similar: there are $2l+1=5$ consecutive values of the n_c in the ground state and the total angular momentum of each of them is J_t = 0, 3/2, 2, 3/2, and 0, respectively. The minimum value of n_c is $n_c^{\rm min}=n+2$ for $n<5$ and $n_c^{\rm min}=10-n$ for $n\geq 5$. The case of Tm ($l=3,n=12$) is also similar and all values of n_c between 7 and 13, and $J_t=0,3/2,2,5/2,4,9/2$, are present in the ground state.[29] However, the cases of $l=3$ and $n=1$ or $n=2$ (which might be relevant for Pr and U systems) have only $n_c=8$ in the ground state and J_t = 15/2 and $J_t=8$, respectively.[30] A common feature of all ground eigenstates with $J_t\neq 0$ is that the impurity is overscreened: the impurity component of J_t is small and points in the opposite direction as the projection of J_t.[21,29,30]

Unfortunately, the local Hamiltonian H_0 does not describe the zero-temperature properties of the system because an eventual SCFP with H_0 decoupled from the rest of the system is unstable, if crystal fields are neglected.[31] When there is degeneracy in n_c, the Hamiltonian near this possible SCFP has the form (eliminating eventual potential scattering as in Ref. 19, Appendix A, Sec. 4)

$$H\cong H_0(V)+H'_{\rm band}+\sum_{\mu\sigma}(c^{\dagger}_{0\mu\sigma}c_{1\mu\sigma}+\text{H.c.}), \quad (5)$$

where $H_0(V)$ is the model including only the innermost conduction-electron Wannier functions $c_{o\mu\sigma}$ discussed above, for large V, and $H'_{\rm band}$ is the band without $c_{0\mu\sigma}$ states, discussed in Sec. III B 3 of Ref. 18. The $c^{\dagger}_{0\mu\sigma}$ and $c_{0\mu\sigma}$ connect different states with the ground state energy. If the first and second terms of Eq. (5) describe a stable fixed point, the last term should be negligible after a sufficiently large number of iterations. However, in this case, the operators $c_{1\mu\sigma}$, $c^{\dagger}_{1\mu\sigma}$ are affected by a factor $\Lambda^{-1/4}$ in each iteration, where $\Lambda>1$ is the parameter of the logarithmic discretization [see Eq. (4.23) of Ref. 18]. This, together with the overall factor $\Lambda^{1/2}$ of each iteration, gives a factor $\Lambda^{1/4}$ for the last term of Eq. (5), indicating that it is a relevant operator.[31] Thus, in the case that $H_0(V)+H'_{\rm band}$, $V\rightarrow\infty$ actually corresponds to a fixed point; it is unstable. In fact, in this case, the limit $V\rightarrow\infty$ does not lead to a trivially solvable model as for the ordinary Anderson or Kondo impurity.[17–19] For the two cases of H_0 solved before in which there is no degeneracy in n_c,[30] we expect an analogy with the TCKM in which overscreening makes the SCFP unstable.[13,14] However, in contrast to the TCKM, there is not a "self-similarity" by which the Hamiltonian near the strong-coupling fixed point takes the same form as the original one. Thus, it is possible that the low-temperature physics of the model is described by a Fermi-liquid fixed point in which more than one of the innermost Wannier functions of the conduction electrons is coupled with the impurity. This is the case of a recently proposed simplified model for Tm impurities under a cubic crystal field, in which the $4f^{12}$ configuration is represented by an A_1 singlet and a T_1 triplet, while only the lowest doublet of the $4f^{13}$ configuration and the conduction-electron doublets (to make Wilson's renormalization-group calculations tractable) are retained.[32] Depending mainly on the relative strength of the A_1-doublet and T_1-doublet hybridizations, the ground state is a singlet or a doublet, and in an intermediate region the SCFP is unstable but the ground state is a singlet with Fermi-liquid behavior.[32] Also, in the simplest case $p^4\leftrightarrow p^5$ (or equivalently $p^3\leftrightarrow p^4$) the effective Hamiltonian Eq. (5) for $V\rightarrow\infty$ has a form similar to the ordinary Anderson model,[16,18,19] but with hybridization dependent on the occupation of the localized level. Similar models have been solved with the Bethe ansatz and have a singlet ground state.[23] Thus, one might expect that the ground state for valence fluctuations between two magnetic isotropic p configurations is a singlet. However, we show below that under an appropriate strong crystal field, this case can display magnetic or NFL behavior at low temperatures.

Both cases for valence fluctuations between two magnetic p configurations are equivalent (they are related by a special electron-hole transformation.[21,22]) In one of them, the configurations with $J_1=2$ and $J_0=3/2$ are hybridized through conduction electrons with $j=1/2$ and $j=3/2$. Under a tetragonal (C_{4v}) crystal field the states with total angular momentum 3/2 (J_0 and j) split into two doublets E'_1 and E'_2. The states with $j=1/2$ transform like E'_1, and $J_1=2$ splits into three singlets with symmetries A_1,B_1,B_2, and an E doublet. Let us assume that one can neglect all states of each p^n

configuration except those of lowest energy. For definiteness, let us assume that E_1' is the lowest lying doublet of the $J_0=3/2$ configuration. In the other case all following arguments are equivalent. From the decomposition of the direct product of representations,

$$E_i'\otimes E_i'=A_1+A_2+E;\quad E_1'\otimes E_2'=B_1+B_2+E, \tag{6}$$

one realizes that if the lowest state of the $J_1=2$ configuration is a singlet only one of the conduction-electron doublets E_i' ($i=1,2$) mixes both configurations. Thus, the problem becomes equivalent to the ordinary Anderson model (in the limit of infinite repulsion at the impurity site) and the ground state is a singlet.[16–19]

In the case in which the doublet E is the lowest in energy of the $J_1=2$ configuration, both conduction-electron doublets contribute to the hybridization. The eigenstates of E (E_2') under a rotation of 90° (C_4) have eigenvalues $e^{i\pi/2},e^{-i\pi/2}$ ($e^{i3\pi/4},e^{-i3\pi/4}$). We denote these states by $|1\rangle,|-1\rangle(|+\rangle,|-\rangle)$, respectively. The states of E_1' are denoted by $|\uparrow\rangle,|\downarrow\rangle$ in obvious notation (corresponding to the two states of $j=1/2$). In terms of these states the Hamiltonian takes the form

$$\begin{aligned}H=&\sum_{k\sigma}\epsilon_k^1 c_{k\sigma}^\dagger c_{k\sigma}+\sum_{k\sigma}\epsilon_k^2(c_{k+}^\dagger c_{k+}+c_{k-}^\dagger c_{k-})\\&+E\sum_\sigma|\sigma\rangle\langle\sigma|+(E+\Delta)(|1\rangle\langle 1|+|-1\rangle\langle -1|)\\&+V_1\sum_k(c_{k\uparrow}^\dagger|\uparrow\rangle\langle 1|+c_{k\downarrow}^\dagger|\downarrow\rangle\langle -1|+\text{H.c.})\\&+V_2\sum_k(c_{k+}^\dagger|\downarrow\rangle\langle 1|+c_{k-}^\dagger|\uparrow\rangle\langle -1|+\text{H.c.}).\end{aligned} \tag{7}$$

If one of the conduction-electron doublets is neglected (or if $V_1V_2=0$), this model is equivalent to that proposed by Balseiro and Alascio as a simplified model for Tm systems.[33,34] The model was solved exactly by the same authors and the ground state is a doublet.[33] In the general case, the properties of the model Eq. (7) are difficult to predict. For $\Delta'=\Delta-\epsilon_F\gg V_i$, where ϵ_F is the Fermi energy, the states $|1\rangle,|-1\rangle$ can be eliminated through a canonical transformation[35] and the interaction between localized and conduction electrons takes the form

$$\begin{aligned}H_{\text{int}}=&-\frac{V_1^2}{\Delta'}\sum_{k\sigma}c_{k\sigma}^\dagger c_{k\sigma}|\sigma\rangle\langle\sigma|\\&-\frac{V_2^2}{\Delta'}\sum_k(c_{k+}^\dagger c_{k+}|\downarrow\rangle\langle\downarrow|+c_{k-}^\dagger c_{k-}|\uparrow\rangle\langle\uparrow|)\\&-\frac{V_1V_2}{\Delta'}\sum_k[(c_{k\uparrow}^\dagger c_{k+}+c_{k-}^\dagger c_{k\downarrow})|\uparrow\rangle\langle\downarrow|+\text{H.c.}].\end{aligned} \tag{8}$$

Defining the states $\gamma_{k+\downarrow}=(c_{k\uparrow}+c_{k-})/\sqrt{2}$, $\gamma_{k+\uparrow}=(c_{k\downarrow}+c_{k+})/\sqrt{2}$, and $\gamma_{k-\downarrow}=(c_{k\uparrow}-c_{k-})/\sqrt{2}$, $\gamma_{k-\uparrow}=(c_{k\downarrow}-c_{k+})/\sqrt{2}$, the spin-flip part of Eq. (8) takes the same form as the corresponding term of the two-channel xxz model,[36] but with opposite signs of the coupling constants. For $V_1=V_2$, the model takes the form of two different xxz models and following the arguments of the appendix of Emery and Kivelson,[36] or those of Nozières and Blandin,[13] we expect that only the antiferromagnetic xy couplings are relevant for the low-temperature physics and the ground state is then a singlet.

Finally, let us assume that the system is under a strong cubic field and a smaller tetragonal (C_{4v}) field, such that the states A_1 and B_1 (degenerate under a cubic field) lie close in energy and are the lowest lying of the corresponding multiplet. Let us also assume hybridizations V_A and V_B of these states with the ground state E_1' are similar, and that the energies Δ_A',Δ_B' necessary to take an electron from the Fermi energy and put it at the impurity leaving it in the state A_1 or B_1 are much larger than the hybridizations. In this case a canonical transformation leads to the TCKM, but with slightly different antiferromagnetic couplings V_A^2/Δ_A' and V_B^2/Δ_B' for the two channels. This means that the ground state is a singlet.[13] However, a small difference of the couplings means that NFL behavior exists down to very low temperatures.[13]

In summary, we have shown that the low-temperature behavior of the appropriate generalization of the Anderson impurity model to two magnetic configurations in an isotropic medium is not characterized by a strong-coupling fixed point (infinite hybridization), in which a local Hamiltonian at the impurity site H_0 is decoupled from the rest of the system, except in the extreme unrealistic case of j-j coupling, where the ground state is a singlet. This feature of the model is shared with the multichannel Kondo model[10,11,13–15] and other impurity models displaying non-Fermi-liquid behavior,[6–9] but in the present case we are not able to predict the low-temperature behavior of the system. In the case of Tm, a cubic crystal field might render the strong-coupling limit stable.[31]

The importance of taking realistic configurations and the effect of crystal field are illustrated in the simplest case of valence fluctuations between two magnetic p configurations. Depending on the crystal-field parameters, the model displays magnetic, nonmagnetic, or non-Fermi-liquid behavior at low temperatures. The latter behavior extends to zero temperature only for a particular ratio of the parameters.

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