Low-Temperature Properties of the Two-Impurity Kondo Hamiltonian

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For the two-impurity Kondo system, the low-temperature properties—impurity spin-spin correlation function, susceptibilities, specific heat, and Wilson ratio—are strongly nonuniversal functions of the ratio I_0/T_K of the Ruderman-Kittel-Kasuya-Yosida coupling to the single-impurity Kondo temperature. In particular, there is a new unstable fixed point for an antiferromagnetic coupling with $I_0/T_K \approx -2.2$. The calculations utilize a newly found symmetry of Kondo Hamiltonians, axial charge.

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The understanding of correlations in heavy-fermion systems is far from complete.[1] The behavior of a pair of spin-$\frac{1}{2}$ impurities is a start for our understanding of a lattice of localized moments.[2] The low-temperature behavior for antiferromagnetic correlations is especially complex, since the interimpurity Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction and Kondo effects compete directly in a way that has been addressed by only a few previous theories.[3] Recently two of us discussed the specific application of Wilson's numerical renormalization-group method to the two-impurity Kondo Hamiltonian and presented results primarily for fairly small ferromagnetic initial RKKY couplings.[4]

Here we present ground-state (low-temperature) properties for a full range of initial interactions and develop a two-parameter Fermi-liquid Hamiltonian for a slightly simplified case of the antiferromagnetic regime. For antiferromagnetic RKKY couplings $I_0 \sim -2T_K$ (where T_K is the single-impurity Kondo temperature) we find a new unstable fixed point characterized by diverging staggered susceptibility and coefficient of specific heat. At this new fixed point, both the ratio I_0/T_K and the impurity spin-spin correlation function $\langle \mathbf{S}_1 \cdot \mathbf{S}_2 \rangle$ are remarkably constant with T_K varying over 2 orders of magnitude. The Wilson ratio reaches a minimum value of much less than unity.

Our Hamiltonian is a natural extension of the original Kondo model to two spin-$\frac{1}{2}$ impurities $\mathbf{S}_1$ and $\mathbf{S}_2$, separated by a distance r, each impurity spin interacting with the spins $\mathbf{s}_c(\mathbf{r}_i)$ of the conduction electrons at the sites i.[4] The interaction term is

$$H_{\text{int}} = J[\mathbf{s}_c(\mathbf{r}_1)\cdot\mathbf{S}_1 + \mathbf{s}_c(\mathbf{r}_2)\cdot\mathbf{S}_2]. \quad (1)$$

This Hamiltonian introduces a new scale—the initial (high-temperature) RKKY coupling $I_0 \propto (\rho J)^2$ where ρ is the density of conduction-electron states. (In general I_0 is a function of the impurity separation r, oscillating between the ferromagnetic and antiferromagnetic values.) The other important scale is the single-impurity Kondo temperature $T_K \propto (|\rho J|)^{1/2}\exp(1/\rho J)$.

There are three regimes of low-temperature behavior, depending on the ratio of the initial RKKY interaction to the single-impurity Kondo temperature. (a) When zero $I_0/T_K > -2.2$, that is, for all ferromagnetic interactions and medium to small initial antiferromagnetic interactions, the ground state is that of impurity spins completely quenched by the Kondo effect,[2] with nonzero interimpurity spin correlations. (b) When $I_0/T_K < -2.2$, for large antiferromagnetic initial interactions, *no* Kondo effect occurs.[2] The ground state is an uncompensated singlet with strong spin correlations. (c) When $I_0/T_K \approx -2.2$, at moderate antiferromagnetic initial couplings, there occurs an unstable fixed point of complex nature, completely unlike those of the single-impurity system. Its properties are discussed in subsequent sections.

These ground states are summarized in Fig. 1 by the impurity spin-spin correlation function $\langle \mathbf{S}_1 \cdot \mathbf{S}_2 \rangle$ at zero temperature as a function of I_0/T_K.[5] Note that $\langle \mathbf{S}_1 \cdot \mathbf{S}_2 \rangle$ is a continuous function through the unstable fixed point (marked by a symbol). For I_0/T_K larger than the critical value the moments are quenched by a Kondo effect while for smaller values no Kondo effect occurs. Note that the critical value of the antiferromagnetic coupling is far larger than commonly thought would allow a Kondo effect. The ground state is a singlet for all values except $\langle \mathbf{S}_1 \cdot \mathbf{S}_2 \rangle = 0.25$.

The most remarkable fact is the near constancy of the ratio I_0/T_K for the unstable fixed point, especially since T_K varies 2 orders of magnitude between $|\rho J| = 0.15$ and $|\rho J| = 0.35$. Unlike other fixed points of the one- and two-impurity problems, this fixed point apparently cannot be obtained by the scaling of variables of the original Hamiltonian to special values such as infinity, since T_K arises from a many-body low-temperature effect, and does not appear as a parameter in the original Hamiltonian (1). Also remarkable is the near coincidence of the curves, especially for the two smaller $|J|$'s, for antiferromagnetic couplings up to even moderate sizes. For ferromagnetic couplings we find no such scaling of the correlation function.

At low temperatures we can quantitatively express the deviations from the fixed point in regions (a) and (b) above in terms of a Fermi-liquid–like effective Hamiltonian[4] with six parameters:

$$\Delta H_{12}^{\text{eff}} = -\sum_{p=e,o} t_p(f_{0p\mu}^{\dagger} f_{1p\mu} + \text{H.c.}) + \sum_{p=e,o} U_p(n_{0p}-1)^2 + U_{eo}4\mathbf{j}_{0e}\cdot\mathbf{j}_{0o} - J_{eo}4\mathbf{s}_{0e}\cdot\mathbf{s}_{0o}. \quad (2)$$

The subscripts $p=e,o$ refer to states that are even and odd combinations of the state about the origin between the two impurities. Here, as in Ref. 4, f_0 and f_1 are momentum-shell operators for the conduction electrons, with f_0 localized about the impurity center and f_1 orthogonal to f_0 and slightly less localized. The number and spin of the f_0 electrons are $n_{0p} \equiv f_{0p\uparrow}^{\dagger} f_{0p\uparrow} + f_{0p\downarrow}^{\dagger} f_{0p\downarrow}$, and $\mathbf{s}_{0p} \equiv f_{0p\mu}^{\dagger} \frac{1}{2} \sigma_{\mu\mu'} f_{0p\mu'}$, respectively. Finally, the axial charge, $\mathbf{j}_{0p}$, is a (newly found) conserved vector quantity whose components[6] commute like those of angular momentum and which is a result of particle-hole symmetry in Hamiltonian (1). Axial charge is also conserved in the single-impurity Kondo and Anderson Hamiltonians. The z component of $\mathbf{j}_{0p}$ is one-half the charge as defined by Wilson[7] and Krishna-murthy, Wilkins, and Wilson[8]: $j_{0e}^z \equiv \frac{1}{2}(n_{0e}-1)$, for example.

The coefficients t_p, U_p, U_{eo}, and J_{eo} are deduced from the energy levels generated from the numerical renormalization-group approach. We can then treat the effective Hamiltonian as a perturbation on the fixed-point energy levels and analytically calculate thermodynamic quantities such as the susceptibility, uniform and staggered, and the coefficient of specific heat. Figure 2[9] shows low-temperature impurity contributions to χ, χ_s(staggered)$\propto \beta\langle (S_1^z - S_2^z)^2\rangle$, and $\gamma \equiv C/T$. The new unstable fixed point (marked by a bar on the I_0/T_K axis) is characterized by diverging values of staggered susceptibility and specific-heat constant while the uniform susceptibility remains fairly small.

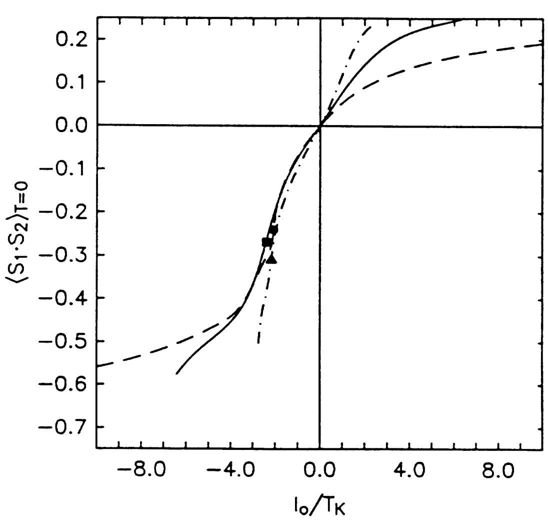

FIG. 1. Zero-temperature impurity spin-spin correlation function as a function of the ratio of the initial RKKY coupling I_0 to the Kondo temperature T_K for three values of $\rho J = -0.15$ (dashed line), -0.25 (solid line), and -0.35 (dash-dotted line). The unstable fixed points—indicated for decreasing ρJ by the circle, square, and triangle, respectively—are remarkably close even though T_K varies over 2 orders of magnitude. Note there is a limiting value of the antiferromagnetic correlation that can be achieved without explicitly adding to Eq. (1) an $\mathbf{S}_1 \cdot \mathbf{S}_2$ term.

Even the stable fixed points are surprising. In the strong antiferromagnetic limit, in which of course there is no Kondo effect, the loss of degrees of freedom suggests a ground-state singlet, but $\langle \mathbf{S}_1 \cdot \mathbf{S}_2 \rangle$ never achieves the singlet value of $-\frac{3}{4}$. In fact, as Fig. 1 indicates, over the range of ρJ examined, $\langle \mathbf{S}_1 \cdot \mathbf{S}_2 \rangle$ never drops below -0.66. For large antiferromagnetic couplings all thermodynamic quantities tend toward zero, as expected for nearly free electrons. For zero initial RKKY interaction $\chi = \chi_s \approx 2\gamma$. As ferromagnetic interactions increase, the three thermodynamic quantities diverge, with $\chi \approx 2\gamma \approx 2\chi_s$.

Figure 3 shows the dependence of the Wilson ratio

$$R = \tfrac{1}{3}\pi^2(k_B/\mu_B)^2 \chi/\gamma \quad (3)$$

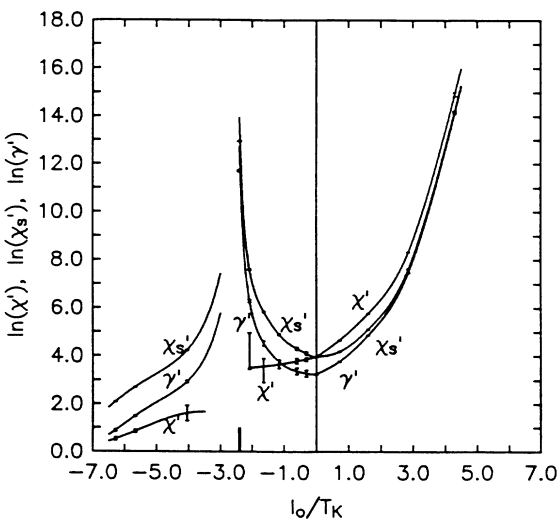

FIG. 2. Logarithms of impurity contributions to susceptibilities (uniform and staggered) scaled by $1/4\rho\mu_B^2$, and to specific-heat coefficient scaled by $3/4\rho\pi^2 k_B^2$, as functions of the scaled RKKY coupling I_0/T_K with $\rho J = -0.25$. (The dimensionless thermodynamic quantities are denoted by primes. Since these plots are for a single value of J, the scaling of I_0 is merely for comparison with Fig. 1.) Approaching the (hitherto unmentioned) stable ferromagnetic fixed point occurring at $r=0$ (for $I_0/T_K = 6.42$) $\chi_s' \approx \gamma'$ and $\chi' \approx 2\chi_s'$ diverge sharply. In the antiferromagnetic regime, χ_s and γ diverge at the unstable fixed point, which occurs at $I_0/T_K \approx -2.3$, as bracketed by the vertical lines. χ appears to be decreasing, but large error bars make the unstable fixed-point value uncertain. For large antiferromagnetic couplings, all properties tend toward zero, with $\chi' \approx \gamma'$ (i.e., Wilson ratio $R=1$.)

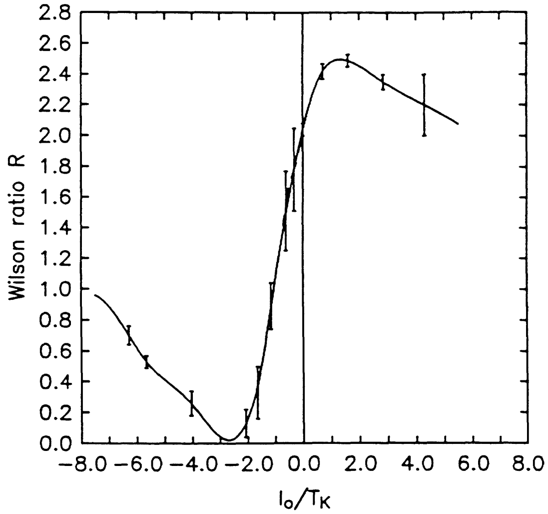

FIG. 3. Wilson ratio R [Eq. (3)] vs scaled initial RKKY coupling (for $\rho J = -0.25$). For reference, $R = 1$ for free-electron and strong antiferromagnetic limits; $R = 2$ for single-impurity (Kondo) limit and, perhaps, also for the strong ferromagnetic limit. At the unstable fixed point ($I_0/T_K \sim -2.3$) R is quite small. The behavior for $\rho J = -0.35$ is indistinguishable from that of $\rho J = -0.25$ for small antiferromagnetic couplings.

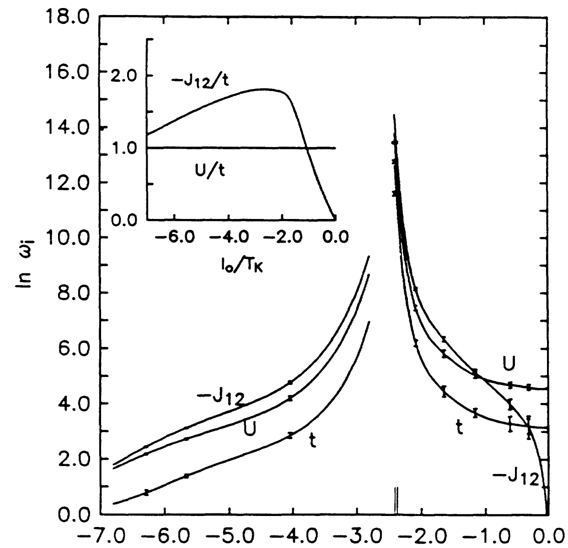

FIG. 4. Logarithms of coupling constants of the simplified effective Hamiltonian (4) vs scaled antiferromagnetic initial RKKY. For the single-impurity problem, U/t is independent of ρJ. For the two-impurity problem, the extra coupling constant $-J_{12}$, while zero for independent impurities, rapidly grows antiferromagnetically to diverge with U and t at the unstable fixed point. For large initial antiferromagnetic couplings all three coupling constants tend toward the noninteracting quasiparticles value of zero. Inset: Schematic plot of scaled ratios (see text) U/t and $-J_{12}/t$ such that the Wilson ratio is $R = 1 + (U + J_{12})/t$ in these units. (Error bars have been suppressed for clarity.) As for the single-impurity problem, we find the ratio U/t to be unity. J_{12}, however, varies widely, giving the nonuniversal Wilson ratio shown in Fig. 3. The unstable fixed point is characterized by a maximum in the ratio $-J_{12}/t$.

on the ratio of I_0/T_K for $\rho J = -0.25$. Note the very dynamic range of R about the (universal) single-impurity value $R = 2$, quantitatively confirming its nonuniversal nature for the two-impurity Kondo problem.[4] In particular, at the new unstable fixed point R reaches a minimum value much less than 1; indeed within our numerical accuracy it is zero.

It is possible to characterize the Wilson ratio in terms of a simple Fermi-liquid model. The complex interactions in the antiferromagnetic regime near the unstable fixed point are not dependent on any asymmetry between the strengths of the even- and odd-parity interactions, and so we set them equal for simplicity. The effective Hamiltonian (2) in this case, written in terms of operators at sites $i = 1$ and 2, is[10]

$$\Delta H^{\mathrm{eff}}_{(\mathrm{AFM})} = -t \sum_{i=1,2} (f^\dagger_{0i\mu} f_{1i\mu} + \mathrm{H.c.}) + U \sum_{i=1,2} (n_{0i} - 1)^2 + U_{12} 4\mathbf{j}_1 \cdot \mathbf{j}_2 - J_{12} 4\mathbf{s}_1 \cdot \mathbf{s}_2. \qquad (4)$$

U_{12} does not appear in either the susceptibility or the specific heat, and numerically it is zero.[11] Figure 4[9] displays the other three coefficients U, $-J_{12}$, and t as functions of scaled antiferromagnetic RKKY interaction. For independent impurities ($I_0 = 0$) $J_{12} = 0$, and ΔH^{eff} reduces to the same two terms as found for the single-impurity problem. As the initial RKKY interaction is increased, the high-temperature interactions between the moments are mirrored at low temperatures by increased (antiferromagnetic) interactions between the quasiparticles: $-J_{12}$ increases sharply. All three terms diverge rapidly (note the logarithmic scale) at the unstable fixed point. In the limit of strong antiferromagnetic initial couplings the deviations from the free-electron fixed point tend toward zero.

In terms of the Hamiltonian (4), the Wilson ratio can be rewritten

$$R = 1 + (U/t + J_{12}/t) C(\Lambda). \qquad (5)$$

Here $C(\Lambda)$ is a constant of order unity which depends on details of the renormalization procedure.[12] In the inset of Fig. 4 the ratios U/t and $-J_{12}/t$ scaled by C are plotted versus I_0/T_K. We find that U/t takes the universal value one,[13] so that all the deviations from the single-impurity value $R = 2$ are governed by the behavior of J_{12}. The unstable fixed point is thus characterized by a maximum in antiferromagnetic quasiparticle spin interactions.

In summary, we find complex behavior in the antiferromagnetic regime of the two-impurity Kondo problem, including an unstable fixed point at moderate initial RKKY couplings. Many heavy fermions appear to have antiferromagnetic correlation or are close to a magnetic instability,[14] which raises the interesting question as to how close they may be to the antiferromagnetic instability found in the two-impurity problem. Although one would expect that a direct application of the two-impurity results here would have to be renormalized for a lattice, we nonetheless note the intriguing correlation between the uniformly small values of the Wilson ratio found experimentally[1,15] in heavy-fermion superconductors, and the very low values we find in the vicinity of the unstable fixed point.

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[5]$\langle \mathbf{S}_1 \cdot \mathbf{S}_2 \rangle$ is calculated numerically as an expectation value for each eigenstate, based on the ratios of singlet and triplet impurity spin that existed at high temperature. At low enough temperatures $\langle \mathbf{S}_1 \cdot \mathbf{S}_2 \rangle$ takes the same value for all states, and it is this value we plot in Fig. 1.

[6]In terms of the $f_0, f_1, \ldots$ operators of Eq. (2), $j_p^+ \equiv \sum_n (-1)^n f^\dagger_{np\uparrow} f^\dagger_{np\downarrow}$; $j_p^- \equiv (j_p^+)^\dagger$; and $j_p^z \equiv \frac{1}{2} \sum_n (n_{np} - 1)$.

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[10]The coefficients of the unsymmetrized effective Hamiltonian (4) are related to those of even-odd parity H^{eff}, (2), by $t = \frac{1}{2}(t_e + t_o)$; $U = \frac{1}{4}(U_e + U_o + 3U_{eo} + 3J_{eo})$; $U_{12} = \frac{1}{4}(U_e + U_o + U_{eo} - 3J_{eo})$; $J_{12} = \frac{1}{4}(U_e + U_o + 3U_{eo} + J_{eo})$.

[11]$U_{12} = 0$ is expected. With no asymmetry between the even and odd interactions the Hamiltonian (4) now conserves charge at each site. Since, for example, $j_1^+ j_2^- + j_1^- j_2^+$ does not commute with n_1 (or n_2), the term involving U_{12} must be absent from (4).

[12]$C(\Lambda) = (1 - \Lambda^{-1})^{3/2} / (1 - \Lambda^{-3})^{1/2} 2 \ln \Lambda$. For the results presented here we chose $\Lambda = 3$.

[13]This current ratio between U and t is expected based on Nozières's "weak universality" argument, as noted in Ref. 4. See P. Nozières, J. Low Temp. Phys. **17**, 31 (1974); P. Nozières and A. Blandin, J. Phys. (Paris) **41**, 193 (1980).

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