# Crystal Field Triplets: A New Route to Non-Fermi-Liquid Physics

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A model for crystal field triplet ground states of rare earth or actinide ions with dipolar and quadrupolar couplings to conduction electrons is studied for the first time with renormalization group methods. The quadrupolar coupling leads to a new nontrivial, non-Fermi-liquid fixed point, which survives in an intermediate valence Anderson model. The calculated magnetic susceptibility displays one parameter scaling, going as $T^{-\alpha}$ ($\alpha \approx 0.4$) at intermediate temperatures, reminiscent of the non-Fermi-liquid alloy $UCu_{5-x}Pd_x$.

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Recent data for many "heavy fermion" Ce or U based compounds and alloys display diverging low-temperature magnetic susceptibility $\chi(T) \sim T^{-\alpha}$ and electronic specific heat coefficients $C_{el}/T = \gamma(T) \sim T^{-\alpha}$ or $\sim -\ln(T)$, unlike those of a Fermi liquid [$\chi(0)$ and $\gamma(0)$ constant] [1]. A number of theoretical scenarios have arisen to explain this non-Fermi-liquid (NFL) physics, which broadly fall into two categories:

(1) *Localized.*—including models associated with peculiar symmetry allowed interactions between $f$-electron moments and conduction electrons [2–6] or a disorder induced distribution of Kondo scales [7,8]. Among the first class of models, multichannel Kondo Hamiltonians employing couplings between localized magnetic or orbital doublets [induced by crystalline electric field (CEF) splittings] and conduction electrons have been extensively studied [3–6].

(2) *Extended.*—in which the NFL behavior is driven by coupling of electrons to low-lying modes induced by intersite $f$-moment interactions in proximity to a quantum critical point [9,10].

It is of considerable interest to sort out the applicability of these differing scenarios to real materials. Of particular interest is the alloy system $UCu_{5-x}Pd_x$ [11] which displays NFL behavior in the range $1 \leq x \leq 2.5$ that has been described in terms of localized disordered Kondo physics [7,8] or Griffiths' phase theory [10]. This alloy appears to display two separate NFL regimes as a function of temperature: (1) An "impurity" regime independent of $x$ for $80 \leq T \leq 300$ $K$ with $\chi(T) \sim T^{-1/3}$ and $\chi''(\vec{Q}, \omega, T) \sim \omega^{-1/3}$ for $\hbar\omega \geq k_B T$ (there appears to be negligible $Q$ dependence to $\chi''$) [12,13]. (2) A low temperature "lattice" regime in which $\chi(T) \sim \gamma(T) \sim T^{-\alpha(x)}$ and for which some evidence exists for intersite interaction effects [13]. The impurity regime data is not compatible with any plausible multichannel Kondo model assuming ground state magnetic or orbital doublet levels on the U ions. The picture is further complicated by analysis of photoemission data which suggests the U ions are in the mixed valent regime, possessing a nearly 50-50 mix of ground state weight in the $f^3$ and $f^2$ configurations [14]. This is problematic in that the Kondo effect has been conventionally studied only for nearly integral valent ions.

In this paper, we present a Kondo model for uranium ions with a CEF triplet ground state that is allowed in cubic symmetry. This model features an effective spin 1 local moment coupled via magnetic dipole and electric quadrupole interactions to one band of effective spin 3/2 conduction electrons. The model displays a new stable NFL fixed point (FP) at low temperatures which is robust even in the mixed valent regime of the more fundamental Anderson model. We find three different power-law regions for the magnetic susceptibility $\chi(T)$: a characteristic $T^{-\alpha}$ ($\alpha \approx 0.4$) dependence in the intermediate temperature region over two decades, a Curie-Weiss law $T^{-1}$ at higher temperatures, and a universal power-law behavior $T^{-2/3}$ at lower temperatures. Despite the differing temperature regimes, a surprising one parameter scaling emerges for $\chi(T)$. The quadrupolar coupling is demonstrated to be relevant using multiplicative and numerical renormalization group (NRG) methods [6,15]. We compare the FP properties with those of the unstable FP reached in the absence of the quadrupolar coupling, and examine the FP structure in the presence of uniaxial symmetry breaking fields. We argue that the intermediate and low temperature regimes may be relevant to the physics of $UCu_{5-x}Pd_x$. The model also possibly applies to Pr, Tb, and Tm ions in some host metals.

Assuming a dominantly tetravalent ($5f^2$) U ion for the moment, the Hund's rule ground state has total angular momentum $J = 4$, which is split into a quadrupolar ($\Gamma_3$) doublet, two magnetic triplets ($\Gamma_4$,$\Gamma_5$), and a singlet ($\Gamma_1$) under the action of the cubic CEF. The possible ground states, which are accessed by varying the two parameters of the crystal field Hamiltonian, are $\Gamma_3$, $\Gamma_5$, or $\Gamma_1$ [16]. As argued elsewhere, the $\Gamma_3$ ground state will give rise to the two-channel quadrupolar Kondo effect upon coupling to conduction electrons [3,4,6]. The two channels arise from coupling to a local quartet ($\Gamma_8$) of conduction electrons

  

which may be described as a tensor product of states with two magnetic labels and two orbital ($\Gamma_3$) labels.

In the case of a $\Gamma_5$ ground state, however, the $f^2$ triplet has both magnetic and quadrupolar moments, and can be represented by a pseudospin $S = 1$. This can couple both to the above mentioned $\Gamma_8$ quartet of conduction electrons and to the doublet channels ($\Gamma_6$ or $\Gamma_7$). For most plausible crystal field situations we find, however, that the coupling to the $\Gamma_8$ quartet is larger. Representing the conduction electrons as a single spin $S_c = 3/2$ manifold we obtain the following Kondo model:

$$H = \sum_{km} \varepsilon_k c^\dagger_{km} c_{km} + \sum_{kk'mm'} c^\dagger_{k'm'} c_{km} \times [J_{\rm D}(\vec{S}_{\rm c})_{m'm} \cdot \vec{S} + J_{\rm Q}(\vec{Q}_{\rm c})_{m'm} \cdot \vec{Q}]. \quad (1)$$

Here $m, m' = \pm 1/2, \pm 3/2$, and a potential scattering term is neglected. The quadrupolar operators are given by $\{Q^i, i = 1, \ldots, 5\} = \{S_y S_z + S_z S_y, S_z S_x + S_x S_z, S_x S_y + S_y S_x, S_x^2 - S_y^2, (2S_z^2 - S_x^2 - S_y^2)/\sqrt{3}\}$. The conduction electron with wave number $k$ and pseudospin $m$ has kinetic energy $\varepsilon_k$ and is created (annihilated) by the operator $c^\dagger_{km}$ ($c_{km}$). In the limit of small hybridization between the conduction band and the $f$ orbitals, this Kondo Hamiltonian (1) can be directly derived from an Anderson Hamiltonian where we restrict ourselves to the valence fluctuation between the $5f^1\Gamma_7$ and $5f^2\Gamma_5$ states. In this case, we obtain a coupling ratio $J_D/J_Q = 2$ and a marginally irrelevant potential scattering with amplitude $J_D$. Our NRG calculations show that even including the $5f^3$ configuration and extending the Anderson model parameters to the mixed-valent regime, the Kondo model (1) describes a stable FP [17]. Therefore, at low enough temperatures, we can use this model to study the realistic Kondo effect corresponding to this FP.

The relevance of the quadrupolar coupling $J_Q$ in Eq. (1) can be immediately seen from a multiplicative renormalization group procedure, valid in the weak coupling regime. After a straightforward but lengthy calculation we derive the following RG equations:

$$dj_D/dx = (j_D^2 + 12 j_Q^2)(1 - 5 j_D)$$
$$dj_Q/dx = 6 j_D j_Q - 36 j_Q^3 - 15 j_D^2 j_Q . \quad (2)$$

Here $x = \ln(E_{\rm F}/\omega)$ denotes the scaling variable (with $E_{\rm F}$, the Fermi energy, and $\omega$, the characteristic energy scale), and we have introduced the dimensionless couplings $j_Q = \varrho_0 J_Q$ and $j_D = \varrho_0 J_D$ with $\varrho_0$, the density of states at the Fermi surface. The flow diagrams obtained from a numerical solution of Eq. (2) are shown in Fig. 1. In the absence of quadrupolar exchange the model scales to the dipolar FP "$D$" at ($j_Q = 0$, $j_D = 1/5$). This FP has been shown to be characterized by a critical exponent $\Delta = 1/6$ associated with its spin sector, which can be mapped to the spin sector of the 10-channel Kondo problem [18,19]. Obviously, this dipolar FP is unstable to quadrupolar perturbations and for any nonzero $j_Q$ it flows to a new FP "S" at ($j_D = 1/5$, $j_Q = \sqrt{1/60}$).

At the FP "S" the ratio $j_Q/j_D$ takes the value $j_Q/j_D = \sqrt{5/12}$, and the interaction part of the Hamiltonian can be written in the following pseudo-SU(3) invariant form:

$$H_{\rm int} = J \sum_{kk'mm'} \sum_{i=1}^{8} \lambda^i c^\dagger_{km} (\lambda^i_{\rm c})_{mm'} c_{k'm'} , \quad (3)$$

where the $\lambda^i$'s denote the $3 \times 3$ Gell-Mann matrices [20] satisfying the SU(3) Lie algebra $[\lambda^i, \lambda^j] = 2 i f^{ijk} \lambda^k$, and can be easily expressed in terms of the spin one impurity operators. The $4 \times 4$ matrices $\lambda^i_{\rm c}$ are constructed from the conduction electron spin operators $S_{\rm c}$, and satisfy a "pseudo-SU(3)" Lie algebra: $[\lambda^i_{\rm c}, \lambda^j_{\rm c}] = 2 i f^{ijk} \lambda^k_{\rm c} +$ octupolar terms. While these latter terms, which arise from the commutators, spoil the SU(3) symmetry of the local triplet, they cannot couple to the impurity, and are hence irrelevant in the RG and general sense. Since both the leading (second order) and next leading (third order) logarithmic scaling equations result in the FP structure of Eq. (3), we believe that this result is universal and independent of the weak coupling approximation.

Unfortunately, the pseudosymmetry found is not strong enough for the usual characterization of the FP by boundary conformal field theory (CFT) [21]; in particular, it is impossible to absorb the impurity spin in the conduction electron currents without violating the Kac-Moody algebra of the conduction electrons. However, we can study the properties of the novel FP by using the NRG. Following Wilson [15], we rewrite Eq. (1) as

$$H_{N+1} = \Lambda^{1/2} H_N + \sum_m (f^\dagger_{N+1,m} f_{Nm} + \text{H.c.}),$$
$$H_0 = \Lambda^{-1/2} \sum_{mm'} \sum_{T=S,Q} f^\dagger_{0,m'} f_{0,m} [\tilde{J}_T (\vec{T}_c)_{m'm} \cdot \vec{T}], \quad (4)$$

where $H_0$ represents the effective exchange interaction on the impurity site, $f^\dagger_{Nm}$ ($f_{Nm}$) creates (annihilates) a conduction electron in the logarithmic discretized band, and $\tilde{J} = 2J\varrho_0/(1 + \Lambda^{-1})$ with $\Lambda$, the discretization parameter. We follow the usual procedure and iteratively diagonalize

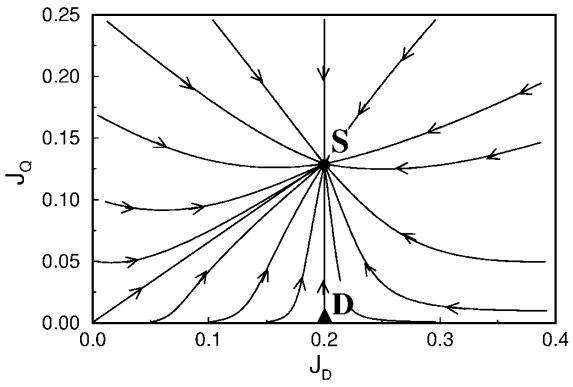


FIG. 1. Scaling trajectories obtained from Eq. (2).

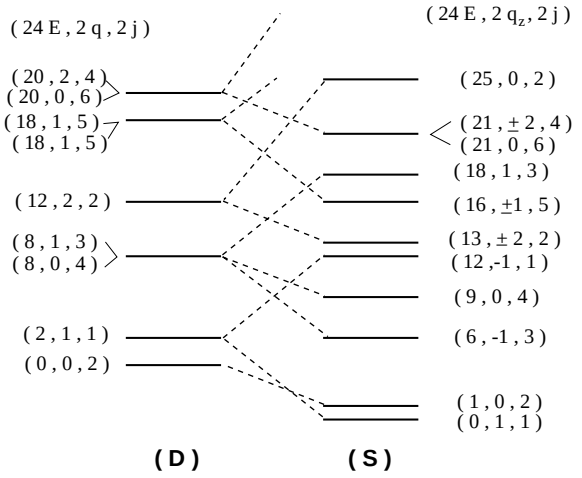


FIG. 2. Finite-size spectrum extracted from the NRG spectrum, and NRG quantum numbers of the fixed points $D$ and $S$ of Fig. 1. The energy $E$ is measured in units of $\pi/L$ ($v_F = 1$ and $L$, the system size), which we identified from the exact CFT solution at $D$. ($D$) For $J_Q = 0$, each energy level is labeled by axial charge $q$ and total spin $j$. ($S$) For $J_Q \neq 0$, $q$ is not a good quantum number since the charge SU(2) symmetry is broken by the quadrupolar interaction.

$H_{N+1}$ to probe the system size on a scale of order $k_F^{-1}\Lambda^{N/2}$ and temperature of order $T_F\Lambda^{-N/2}$. In the NRG calculations we exploited the full spin and axial charge symmetry of the model, with the axial charge operator $\vec{q}$: $q_+ = \sum_{n=0}^{\infty}(-1)^n(f^\dagger_{n,3/2}f^\dagger_{n,-3/2} - f^\dagger_{n,1/2}f^\dagger_{n,-1/2})$, and $q_z = \frac{1}{2}\sum_{n=0}^{\infty}\sum_m(f^\dagger_{nm}f_{nm} - \frac{1}{2})$. These latter satisfy $[q_z, q_\pm] = \pm q_\pm$ and $[q_+, q_-] = 2q_z$ [18]. Figure 2 shows the finite-size energy spectrum obtained at FP's "$D$" and "$S$". The $J_Q = 0$ spectrum in the sector with dipolar coupling only ("$D$") coincides with the exact CFT spectrum for the model of the impurity spin coupled to spin 3/2 electrons [18]. In sector ($S$) we show the finite size spectrum of the new fixed point obtained from the NRG calculations. This should coincide with the spectrum of a yet unknown boundary CFT. As easily shown, the relevant quadrupolar coupling breaks the axial charge SU(2) symmetry down to U(1), while it conserves that of the total spin $j$. Apart from some marginally irrelevant trivial potential scattering that only induces a phase shift, and can be easily subtracted, this symmetry breaking is *universal* and cannot be described by a simple phase shift: Some originally degenerate SU(2) axial charge multiplets are split at the new FP "$S$" by some quadrupolar charge operator, while others are split by a dipolar charge operator.

To determine the dimension of the leading irrelevant operator that governs the new FP, we carried out a finite-size analysis of the NRG levels. Within the NRG scheme the finite-size energy $1/L$ corresponds to $\sim\Lambda^{-N/2}$ and the levels relax to their FP values as

$$E_{\mathrm{NRG}} - E^* \propto \Lambda^{-\Delta N/2}, \tag{5}$$

where $E^*$ is the FP energy and $\Delta$ denotes the scaling dimension of the leading irrelevant operator [22]. For $J_Q \neq 0$ almost the same energy level relaxation is found as for $J_Q = 0$ over the whole parameter space of Fig. 1, in agreement with an exponent $\Delta = 1/6$ (see Fig. 3). This implies that $\chi(T) \sim \gamma(T) \sim T^{2\Delta-1} = T^{-2/3}$ at low $T$.

To determine the impurity susceptibility $\chi(T)$ we calculated the temperature dependent magnetization induced by a small local field at the impurity site. The resulting curves are plotted in Fig. 3. Each curve has an interesting region where it behaves like $T^{-\alpha}$ over approximately two decades, and $1/3 < \alpha \approx 0.4 < 2/3$ slightly depending on the magnitude of the Kondo couplings. After adjusting the overall scale of $\chi(T)$, as shown in Fig. 3, we can scale all $\chi(T)$ data on a single universal curve using a single temperature scale $T^*$ (though strong coupling induces some deviations from scaling at higher temperatures—see the discussion below). This one parameter scaling, also observed in the quadrupolar susceptibility [17], strongly suggests that the intermediate temperature regime behavior reflects the new low temperature FP rather than some unstable FP, similarly to the universal single channel Kondo susceptibility in the regime $T > T_K$. For small to intermediate Kondo couplings, $\tilde{J}_Q, \tilde{J}_D \lesssim 0.5$, $\chi$ behaves according to the Curie-Weiss law $T^{-\alpha}$ ($\alpha = 1$) at large temperatures. For smaller temperatures an intermediate region appears

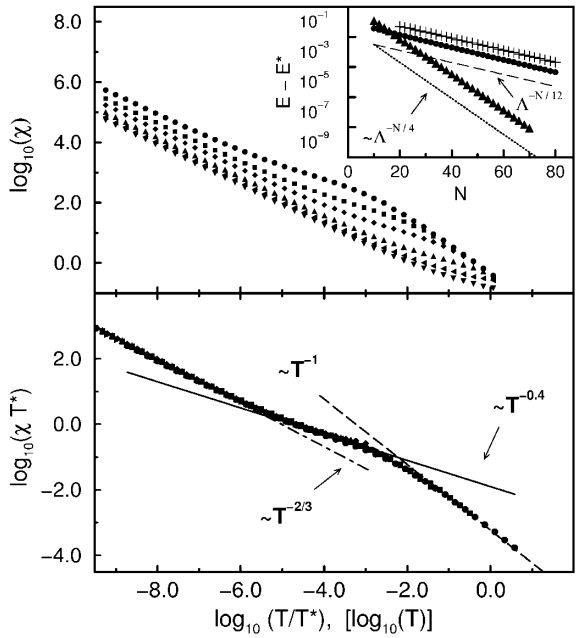


FIG. 3. Magnetic susceptibility: (Top) From top to bottom, the couplings are $4\tilde{J}_D = 8\tilde{J}_Q = 0.5$, 0.7, 1.0, 2.0, 4.0, and 8.0. (Bottom) Rescaled magnetic susceptibility. All the data collapse to a universal curve. Inset: Relaxation of NRG levels near to the fixed points. The initial couplings were $\tilde{J}_D = \tilde{J}_Q = 0.1$ (crosses) and $\tilde{J}_D = 1.0$ and $\tilde{J}_Q = 0$ (circles). For comparison, data for the two-channel Kondo fixed point are also given (triangles), scaling as $\Lambda^{-N/4}$ with $\Lambda = 3$.

where $\alpha \approx 0.4$. As the temperature decreases further, $\chi(T)$ turns up and behaves as $\sim T^{-2/3}$ in the vicinity of the novel FP. For larger couplings the Curie-Weiss part is absent, and $\chi$ starts as $\sim T^{-1/3}$ at high temperatures and then the exponent $\alpha$ gradually goes up to $2/3$ at low $T$. The appearance of the intermediate region with $\chi(T) \sim T^{-\alpha}$ ($\alpha \approx 0.4$) is specific to nonzero quadrupolar coupling $J_Q$. When $J_Q = 0$, the dipolar coupling $J_D$ gives only monotonic behavior $T^{-1} \rightarrow T^{-2/3}$ with decreasing temperatures. This is also a clear difference between the Kondo effect for local triplet and doublet states.

Finally, we discuss the stability of the novel FP against a uniaxial (tetragonal) lattice distortion. The distortion lifts the triplet degeneracy, giving either a singlet or doublet ground state split by a value $h_Q$, and destabilizes the novel triplet FP. Instead, we have two possible stable FP's: when the singlet lies lowest, a Fermi liquid FP arises, and a ground doublet experiences a NFL FP, associated with the two-channel quadrupolar Kondo effect for tetragonal symmetry [4]. According to our NRG calculation, the crossover temperature below which the system flows away from the cubic FP to one of the above two varies as $h_Q^2$ for a small distortion [17]. This power implies that the operator with $j = 2$ corresponding to lattice distortions has the same scaling dimension $1/2$ at "$S$" as at the unstable FP "$D$" of Fig. 1. The two tetragonal FP's are separated by a boundary line in coupling space on which the novel cubic FP resides. The features associated with this cubic symmetric FP are expected to appear in some U-based compounds with uniaxial anisotropy close to this boundary, too.

In conclusion, we investigated a new Kondo Hamiltonian describing the dynamics of a local triplet. The quadrupolar exchange interaction drives the model to a new fixed point, characterized by a universal charge SU(2) symmetry breaking, a leading irrelevant operator with dimension $\Delta = 1/6$, and a pseudo-SU(3) symmetry. For small and intermediate couplings the magnetic susceptibility is universal and has an intermediate temperature range, where it scales as $\chi \sim T^{-\alpha}$ with $\alpha \approx 0.4$. In the strong coupling case this reduces to a power with $\alpha \approx 1/3$. Since the extreme mixed valence of $UCu_{5-x}Pd_x$ suggested by photoemission is compatible with a strong coupling limit of the Kondo model, we suggest that the intermediate temperature range susceptibility may correspond to the impurity range identified for this material [12]. While this material is strongly disordered [8], our estimations show that a considerable fraction of the U ions has a perfect *local* surrounding, and for these the crossover temperature generated can easily be below the lower limit of the universal scaling observed ($\sim$20 K). The surprising increase of the power law at lower temperatures will give the concentrated system a greater tendency towards intersite interaction effects, qualitatively compatible with the suggested interaction driven low temperature physics. To test the idea further, we strongly urge an experimental study of this system with uranium diluted away by thorium. The model may prove relevant to the alloy $Y_{1-x}U_xPd_3$ as well [23]. Triplet ground states have been identified in the concentrated Pr ($4f^2$) compounds $PrPd_3$ [24] and $PrB_6$ [25], so that dilution of Pr by La would be an important experimental study. Dilute Pr in Pd [26] shows unambiguous Kondo behavior, but the existing data do not resolve the relevance of our model for this system.

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