Hexadecapolar Kondo Effect in URu_2Si_2?

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We derive the coupling of a localized hexadecapolar mode to conduction electrons in tetragonal symmetry. The derivation can be easily adapted to arbitrary multipoles in an arbitrary environment. We relate our model to the two-channel Kondo (2CK) model and show that for an f^2 configuration a relevant crystal field splitting in addition to the 2CK interaction is intrinsic to tetragonal symmetry. We discuss possible realizations of a hexadecapolar Kondo effect in URu_2Si_2. Solving our model we find good agreement with susceptibility and specific heat measurements in $Th_{1-x}U_xRu_2Si_2$ ($x \ll 1$).

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In Ref. [1], Cox addressed important differences between U- and Ce-based heavy electron systems in terms of the atomic structure of their f shell. The most probable valence configuration in Ce-based systems has one f electron, whereas in many U-based materials, it is an f^2-many body state. In crystals where the U site has cubic symmetry, f^2 states can give rise to quadrupolar degrees of freedom which, when coupled to conduction electrons, lead to two-channel Kondo (2CK) non-Fermi liquid (NFL) behavior [1,2]. Meanwhile, various multipolar orderings have been observed [3] as well as proposed as candidates for ''hidden order'' (HO) in materials with clear phase transitions but without an obvious order parameter. A prominent example in this area is given by URu_2Si_2, for which quadrupolar [4], octupolar [5], hexadecapolar [6], and triakontadipolar [7] order parameters have all been put forth. Some recent experiments have ruled out quadrupolar order [8], whereas another might implicitly hint at it [9]. On the other hand, the hypothesis of active U hexadecapolar degrees of freedom explains numerous experiments [10]. In this Letter, we generalize the work of Ref. [1] and present a simple construction of low-energy Hamiltonians that describe the coupling between multipoles and conduction electrons in the tetragonal crystal field (TCF) of URu_2Si_2. We show that in TCF the low-energy degrees of freedom of an f^2 configuration couple symmetrically to multiple channels of conduction electrons, regardless of the f^2-coupling and -level schemes. Taking hexadecapolar degrees of freedom for U, and solving the resulting model with the numerical renormalization group (NRG), we successfully describe the properties of $Th_{1-x}U_xRu_2Si_2$ (TURS) in magnetic field [11,12] and place the measurements around the crossover between the local moment and 2CK scaling regimes. Thus, hexadecapolar fluctuations can also explain the anomalies observed in TURS.

To construct a tractable model, valid at very low energies, we take into account only the two lowest-lying $5f$ configurations with double occupancy ($n_{5f} = 2$). This is motivated by recent local-density approximation plus dynamical mean-field theory (LDA + DMFT) calculations, which indicate that, while U has mixed valence in URu_2Si_2 with $2 \leq n_{5f} \leq 3$, two TCF singlets with $n_{5f} = 2$ and different symmetries have the highest probability [6]. The U degrees of freedom are then described by a $J = 4$ multiplet, split by the TCF. The ground state and the nearest excited level are, respectively, time reversal and parity even, A_{2g} and A_{1g} basis states of the group $\mathcal{J} \times D_{4h}$. Here $\mathcal{J} \equiv \{I, \mathcal{T}\}$ is the time-reversal symmetry group, I the identity, $\mathcal{T}$ the time-reversal operator, and D_{4h} the tetragonal point group including parity. Viz., the lowest-lying singlets are $|A_{2g}\rangle \equiv \frac{i}{\sqrt{2}}(|4\rangle - |-4\rangle)$ and $|A_{1g}\rangle \equiv \frac{\cos\phi}{\sqrt{2}}(|4\rangle + |-4\rangle) + \sin\phi|0\rangle$, given in terms of the eigenvectors $|J_z\rangle$ of the operator $\hat{J}_z$ in the $J = 4$ multiplet with the quantization axis chosen parallel to the c axis of the crystal. To keep the equations short, we follow Refs. [1,2] and assume the f shell of the U atom hybridizes mostly with $l = 3, J = \frac{5}{2}$ conduction electrons. Conduction electrons at the local site $\psi^\dagger_{l\,J\,J_z}$ can be classified into the four double-valued or spinor irreducible representations (irreps), Γ_{6p}, Γ_{7p} of the tetragonal double point group, $\bar{D}_{4h}$, with $p = g/u$ for parity even (odd) irreps [i.e. for l even (odd)]. Under time-reversal symmetry $\mathcal{T}\psi^\dagger_{l\,J\,J_z}\mathcal{T}^{-1} = (-)^{l-J+J_z}\psi^\dagger_{l\,J\,(-J_z)}$. We set up a basis so that the $\alpha = \pm$ components of the Kramers doublets $\Psi^\dagger_{\Gamma^{(n)}_{jp}\alpha}$ (where n enumerates doublets of the same type within one J multiplet, and $j = 6, 7$) comply with our convention: $\mathcal{T}\Psi^\dagger_{\Gamma^{(n)}_{jp}+}\mathcal{T}^{-1} = \Psi^\dagger_{\Gamma^{(n)}_{jp}-}$, implying the same for annihilation operators. For the local conduction electron basis, we choose the following two independent Γ_{7u} Kramers doublets for creation operators: $\begin{bmatrix}\Psi^\dagger_{\Gamma^{(1)}_{7u}+}\\ \Psi^\dagger_{\Gamma^{(1)}_{7u}-}\end{bmatrix} \equiv \begin{bmatrix}\psi^\dagger_{\frac{5}{2}}\\ -\psi^\dagger_{-\frac{5}{2}}\end{bmatrix}$, $\Psi^\dagger_{\Gamma^{(2)}_{7u}} \equiv \begin{bmatrix}\psi^\dagger_{-\frac{3}{2}}\\ -\psi^\dagger_{\frac{3}{2}}\end{bmatrix}$, and one Γ_{6u}: $\Psi^\dagger_{\Gamma^{(1)}_{6u}} \equiv \begin{bmatrix}\psi^\dagger_{1/2}\\ -\psi^\dagger_{-\frac{1}{2}}\end{bmatrix}$, on using the compact notation: $\psi^\dagger_{J_z} \equiv \psi^\dagger_{3\,\frac{5}{2}\,J_z}$. Adjoint doublets with the same transformation properties in the same basis are

$\Xi_{\Gamma_{ju}^{(n)}\alpha} \equiv \sum_{\beta\in\{+,-\}} \epsilon_{\alpha\beta} (\Psi^\dagger_{\Gamma_{ju}^{(n)}\beta})^\dagger$ with $\epsilon_{\alpha\beta}$ the two-by-two antisymmetric matrix with entry $\epsilon_{+-} = 1$.

Kondo Hamiltonians are made up of spin-flip and diagonal processes: $\mathcal{H}_K = \mathcal{H}_\perp + \mathcal{H}_z$. When constructing these two parts connecting the two singlets, the only relevant, nontrivial tensor products of irreps are $\Gamma_{6u} \otimes \Gamma_{6u} = \Gamma_{7u} \otimes \Gamma_{7u} = A_{1g} \oplus A_{2g} \oplus E_g$ and $A_{2g} \otimes A_{2g} = A_{1g}$ [13]. Taking the appropriate tetragonal Clebsch-Gordan coefficients, symmetry thus binds the form of the spin-flip and diagonal parts to be [13]

$$\mathcal{H}_\perp = i \sum_{n,m=1}^{2} \mathcal{J}_\perp^{n\,m} (\Psi^\dagger_{\Gamma_{7u}^{(n)}+} \Xi_{\Gamma_{7u}^{(m)}-} + \Psi^\dagger_{\Gamma_{7u}^{(n)}-} \Xi_{\Gamma_{7u}^{(m)}+}) \times |A_{1g}\rangle\langle A_{2g}| + \text{H.c.}, \tag{1}$$

$$\mathcal{H}_z = \sum_{n,m=1}^{2} \sum_{i\in\{1,2\}} \mathcal{J}_z^{n\,m\,i} (\Psi^\dagger_{\Gamma_{7u}^{(n)}+} \Xi_{\Gamma_{7u}^{(m)}-} - \Psi^\dagger_{\Gamma_{7u}^{(n)}-} \Xi_{\Gamma_{7u}^{(m)}+}) \times |A_{ig}\rangle\langle A_{ig}|. \tag{2}$$

The couplings are real and must satisfy $\mathcal{J}_z^{1\,2\,i} = \mathcal{J}_z^{2\,1\,i}$ to ensure Hermiticity, but otherwise arbitrary. We omitted processes including Γ_6 electrons as they decouple from the impurity. The hexadecapolar, i.e., "spin-flip," fluctuations are thus coupled to four species of conduction electrons, namely, to the two independent Γ_{7u} Kramers doublets. $\mathcal{H}_\perp$ has the structure of the 2CK model where the role of spin index is played by the index distinguishing the two different Γ_{7u}'s, and the channels are distinguished by the Kramers indices. To make this correspondence more explicit, we introduce the operators $\left[\begin{smallmatrix}\eta_a\\ \eta_b\end{smallmatrix}\right] \equiv \left[\begin{smallmatrix}\psi_{-\frac{3}{2}}\\ \psi_{\frac{5}{2}}\end{smallmatrix}\right]$, $\eta_b \equiv \left[\begin{smallmatrix}\psi_{\frac{3}{2}}\\ -\psi_{-\frac{5}{2}}\end{smallmatrix}\right]$, and perform the unitary transformation, $|A_{1g}\rangle \to |A'_{1g}\rangle \equiv i|A_{1g}\rangle$, which allows us to rewrite $\mathcal{H}_\perp$ in the standard notation: $\mathcal{H}_\perp = \frac{\mathcal{J}_\perp^{2\,1} - \mathcal{J}_\perp^{1\,2}}{2} \eta^\dagger_{q\mu}\, \sigma^+_{\mu\nu}\, \eta_{q\nu} S^- + \text{H.c.} + \mathcal{O} S^x$, with $q \in \{a, b\}$, $\mu, \nu \in \{\uparrow, \downarrow\}$. Here and in the following, repeated channel (q) and spin (μ, ν) indices are to be summed over, $\mathcal{O} = \mathcal{O}^\dagger$ contains only conduction electrons [14], $\sigma^+ \equiv \sigma^x + i\sigma^y$ is composed of Pauli matrices, and $S^+ \equiv S^x + iS^y \equiv |A_{2g}\rangle\langle A'_{1g}|$, $S^z \equiv (|A_{2g}\rangle\langle A_{2g}| - |A'_{1g}\rangle\langle A'_{1g}|)/2$, $S^- \equiv S^{+\dagger}$, $\mathbb{1} \equiv |A_{2g}\rangle\langle A_{2g}| + |A'_{1g}\rangle\langle A'_{1g}|$.

Channel symmetry follows from time-reversal symmetry. The operator $\mathcal{O} S^x$ is irrelevant around the 2CK fixed point (and marginal in the free fermion scaling regime), as shown by NRG calculations or using conformal field theory results [15,16]. It does not destroy the 2CK state, as it neither breaks channel symmetry nor lifts the spin degeneracy. Thus we must have $\mathcal{J}_\perp^{1\,2} \neq \mathcal{J}_\perp^{2\,1}$ for overscreening to occur. This asymmetry comes up naturally, e.g., if we start off with a spherical symmetric Anderson Hamiltonian, perform the Schrieffer-Wolff transformation to arrive at a Kondo-type of interaction, and then project to the TCF states $|A_{2g}\rangle, |A_{1g}\rangle$ at strong spin orbit (i.e., jj) coupling [1,2,15].

The diagonal part $\mathcal{H}_z$ cannot lead to NFL behavior by itself, but it can quite possibly destroy it. Channel symmetry is preserved by time-reversal symmetry. However, the level degeneracies are lifted by the TCF, both between the $|A_{2/1g}\rangle$ states and also in each screening channel between $\Gamma_{7u}^{(n)}$ electrons with different n's. The dangerous terms are

$$\mathcal{H}_{zi}^{\text{rel}} = \Delta_{\text{imp}}\, \eta^\dagger_{q\,\mu}\, \eta_{q\,\mu}\, S_z, \tag{3}$$

$$\mathcal{H}_{zc}^{\text{rel}} = \Delta_{\text{cond}}\, \eta^\dagger_{q\,\mu}\, \sigma^z_{\mu\,\nu}\, \eta_{q\,\nu}\, \mathbb{1}. \tag{4}$$

Both TCF splittings are relevant around the 2CK fixed point with scaling dimension $\frac{1}{2}$ [15,16] and present in $\mathcal{H}_z$ with amplitudes $\Delta_{\text{imp}} = (\mathcal{J}_z^{2\,2\,1} + \mathcal{J}_z^{1\,1\,1} - \mathcal{J}_z^{2\,2\,2} - \mathcal{J}_z^{1\,1\,2})/2$ and $\Delta_{\text{cond}} = (\mathcal{J}_z^{2\,2\,1} - \mathcal{J}_z^{1\,1\,1} + \mathcal{J}_z^{2\,2\,2} - \mathcal{J}_z^{1\,1\,2})/4$. In fact, they are of the only type of relevant perturbation, when channel symmetry is intact [16]. Thus for this model to exhibit 2CK scaling in some temperature range, Δ_{imp} and Δ_{cond} must fall below the Kondo temperature T_K. This necessarily requires fine-tuning, and the basic assumption of the $A_{2g} - A_{1g}$ scenario—and, as we show below, of any other doublet-ground state scenarios—is that this accidental degeneracy is responsible for the unique behavior of URu_2Si_2 among the large number of U-based heavy fermions. We note that the ordinary TCF splitting $\Delta_{\text{ord}} S_z$ is of the same type, and thus has the same scaling properties as Eqs. (3) and (4) in all scaling regimes, as $\eta^\dagger_{q\mu}\, \eta_{q\mu}$ is marginal. Moreover LDA + DMFT calculations for URu_2Si_2 found $\Delta_{\text{ord}} < T_K$, confirming the accidental degeneracy on the scale of T_K [6].

A local, z-directed magnetic field results in two leading additions to the Hamiltonian: $\mathcal{H}_{\text{mag}i} \propto \mu_B\, i\,(S^+ - S^-)$ and $\mathcal{H}_{\text{mag}c} \propto \mu_B \sum_{J_z\in\{\pm\frac{3}{2},\pm\frac{5}{2}\}} J_z\, \psi^\dagger_{J_z}\, \psi_{J_z}\, \mathbb{1}$ for the two-singlet part and for the local conduction electrons, respectively. These terms have similar effects as $\mathcal{H}_{zi/c}^{\text{rel}}$. Namely, the impurity part, $\mathcal{H}_{zi}^{\text{rel}} + \mathcal{H}_{\text{mag}i}$, amounts to an effective magnetic field (or TCF splitting) pointing into other than the z direction. The same holds true for the conduction electrons with the effective magnetic field or TCF splitting being different in the two channels. Thus, while $\mathcal{H}_K$ is not identical to the 2CK Hamiltonian, it flows to the same fixed point when the relevant perturbations, which split apart the two different Γ_7 irreps, or the two local singlets, vanish; and the application of magnetic field thus breaks both the channel and the spin symmetry of the 2CK model.

For the order of TCF levels in URu_2Si_2, other scenarios have also been put forth in the literature. It turns out that the structure of the effective low-energy Hamiltonian for any two quasidegenerate states is rather similar to that of the two-singlet case considered above. This also applies to a proposed E_g (or Γ_5) doublet ground state [11,17,18], formed by $|E_g\, x\rangle, |E_g\, y\rangle$. Fluctuations within this doublet

can couple to two products of irreps of $l = 3$ electrons: $\Gamma_7 \otimes \Gamma_7$ and $\Gamma_7 \otimes \Gamma_6$. In the 2CK language, the two different irreps play the role of spin, while their Kramers indices, connected by time reversal, play the role of channel index again. 2CK scaling would occur down to $T = 0$ if the two lower-lying irreps were degenerate, i.e., if the spin symmetry of the 2CK model were unbroken. As the degeneracy is approximate, the system will flow to a Fermi liquid (FL) fixed point eventually.

In the HO phase of URu_2Si_2, the Ref. [6] proposed order parameter, $\langle |A_{2g}\rangle\langle A_{1g}|\rangle$, is nonvanishing due to its real part which, in highest order of the multipole expansion, contains the expectation value of the hexadecapolar A_{2g} tensor: $[(\hat{J}_x^2 - \hat{J}_y^2)(\hat{J}_x\hat{J}_y + \hat{J}_y\hat{J}_x) + (\hat{J}_x\hat{J}_y + \hat{J}_y\hat{J}_x)(\hat{J}_x^2 - \hat{J}_y^2)]$ [6]. However, the same reasoning can be repeated for $\langle |E_g\, x\rangle\langle E_g\, y|\rangle$, whose real part also contains the same hexadecapolar ordering. These points are substantiated by the Hamiltonian construction for an E_g ground state in the Supplemental Material [19].

It has long been recognized that χ_c, the (magnetic) dipole susceptibility of $Th_{1-x}U_xRu_2Si_2$ along the c axis, shows log$T$ behavior at low T (see Refs. [11,18] and Fig. 1) in accord with the 2CK descriptions corresponding to both scenarios. However, susceptibility and resistivity measurements find that the magnetic field (H) induced crossover scale to a FL depends on H linearly, i.e., $T_H \propto H^\eta$ with $\eta = 1$, which does not agree with the $\eta = 2$ behavior corresponding to the 2CK scaling regime [12]. To make contact with these experiments, we solved the model, Eq. (1), by NRG and confirmed that it indeed flows to the 2CK fixed point where $\mathcal{O}S^x$ is irrelevant. Then we added a magnetic field, mimicked only by Eq. (3), to the 2CK model, and solved this model using an upgraded version of our density matrix-NRG code [20]. The values of the magnetic field and the Kondo coupling were adjusted to fit the experimental data of Refs. [11,12].

Invoking ω/T scaling [21], we fitted the T dependence of χ_c by the real part of the dynamic susceptibility of the 2CK model in magnetic field, as we trust our dynamic correlation functions (produced by the density matrix algorithm at $T = 0$) better than the thermodynamic quantities. Figure 1 shows convincing agreement between theory and experiment apart from the small discrepancy for $T > 30$ K, i.e., for large energies where the resolution of NRG is limited. We obtained $T_K \approx 1.3$ K from the fit (see the caption of Fig. 1 for further details on T_K). This finding places the measurements in magnetic fields around the crossover region between the local moment and 2CK scaling regimes. In both regimes, scale invariance entails the hyperscaling relation, $\eta + \nu = 2$ with ν the critical exponent defined by $\chi \propto H^{-\nu}$. Thus for $H = 0$, the observed $\nu = 0$ gives $\eta = 2$; i.e., for T between 0.1 and 10 K, the system is in the 2CK scaling regime. In contrast, for $H = 1$–5 T, the experiments measure $\eta = 1$ resulting in $\nu = 1$. Thus, we conclude that these magnetic fields, in addition to the ubiquitous, relevant TCF splitting, are (slightly) larger than T_K and the system flows directly from the local moment regime to a one-channel Kondo fixed point without traversing the 2CK scaling regime. In Fig. 1(a), the ratios of magnetic fields to T_K, fitting the susceptibility, further illustrate this point.

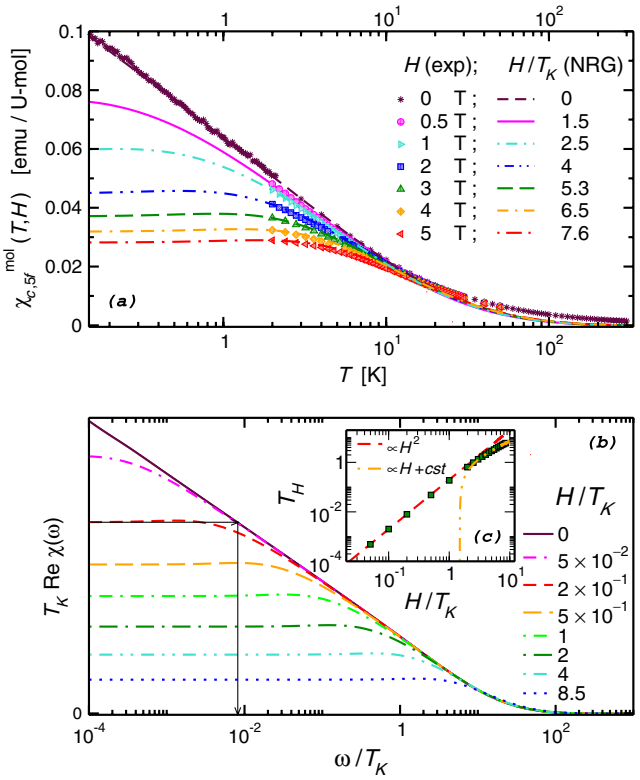

FIG. 1 (color online). (a) Symbols: Molar susceptibility $\chi^{\rm mol}_{c,5f}$ of the $5f$ electrons in $Th_{1-x}U_xRu_2Si_2$ at $x = 0.03$ vs T in magnetic fields between $H = 0$ and 5 T ($H \parallel c$) reproduced from Ref. [12]. (a),(b) Curves: The real part of the local, dynamic susceptibility of the 2CK model in the presence of magnetic field at $T = 0$ computed with density matrix-NRG at Kondo coupling $\mathcal{J}_{\rm 2CK} = 0.15D$ (in units of bandwidth) with discretization parameter $\Lambda = 2$ and keeping at most 2100 multiplets of $\mathrm{U(1)_{spin}} \times \mathrm{SU(2)_{charge1}} \times \mathrm{SU(2)_{charge2}}$ at each NRG step [20]. (c) The crossover scale T_H [defined as the intersect of the low-ω and high-ω asymptotes of Re$\chi(\omega)$, see plot (b)] shows quadratic and linear H dependence in the 2CK and local moment scaling regimes, respectively; T_K, the crossover scale between the local moment and 2CK scaling regimes, is ≈ 1.3 K. Notice that the experimental data for $H = 0$ T can be fitted equally well with every NRG curve where $H \lesssim 0.2T_K$.

By taking a closer look at the specific heat coefficient in Fig. 2(a), we can reinforce these statements and get another estimate for T_K. Two regimes for the given magnetic field values are clearly visible: At low T's, the curves for $H = 0.5$ and 1 T slightly overshoot the curve at $H = 0$ T, in contrast to the curves for $H \geq 2$ T, which exhibit a bump at $T \approx H$. The rise of $\gamma \equiv C_{p,5f}/T$ for low fields at low T is reminiscent of the 2CK scaling regime, except that the measured T dependence of γ at low T for $H = 0$ is

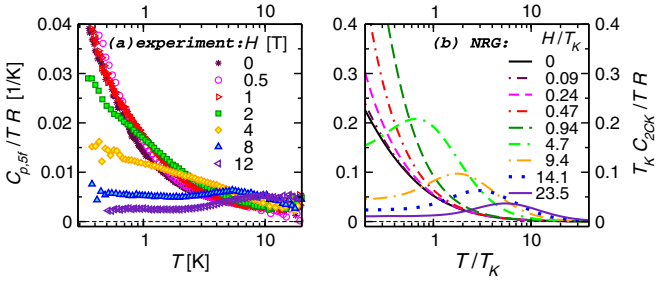

FIG. 2 (color online). (a) $5f$ electronic specific heat coefficient in units of R ($C_{p,5f}/TR$) of $Th_{1-x}U_xRu_2Si_2$ at $x = 0.07$ vs T for H between 0 and 12 T, reproduced from Ref. [18]. (b) 2CK specific heat coefficient for different H's from NRG averaged over 10 z values [25] at $\mathcal{J}_{2CK} = 0.1D$ (in units of bandwidth) with $\Lambda = 2.5$ and keeping at most 3200 multiplets of $U(1)_{spin} \times SU(2)_{charge1} \times SU(2)_{charge2}$ at each NRG step. The two sets of curves display the same trend, but there is a factor of ≈ 4 difference in the magnitudes (depending on the precise value of T_K). We ascribe this difference to experimental inaccuracy, as we observe a factor of ≈ 2 difference between the magnitudes of $C_{p,5f}/T$ for the same material published in Refs. [11,18].

not quite logarithmic. These findings can be explained by the presence of an effective TCF splitting, $0 < \Delta < T_K$, already at $H = 0$, and placing T_K between 1 and 2 T. From the susceptibility fit we estimate that Δ is anywhere below about $0.2T_K$ [cf. Fig. 1(b)]. These assertions are further confirmed by our NRG calculations for the specific heat coefficient [see Fig. 2(b) and cf. Ref. [22]). For $H > T_K$, the bumps at $T \approx H$ correspond to the Schottky anomaly due to the Zeeman splitting between the two local states.

In the 2CK model, if $\Delta < T_K$, there is a NFL region over $2\log(\frac{T_K}{\Delta})$ decades, since the splitting induced crossover scale to a Fermi liquid depends on Δ quadratically. Susceptibility measurements find a NFL region over at least one decade, putting an upper bound on the ratio $\Delta/T_K < 0.6$ and giving the conservative estimate $T_K < 5$ T and $\Delta < 3$ T.

Conclusions.—Motivated by recent findings on the electronic structure of URu_2Si_2 [6], we derived the Kondo coupling between localized hexadecapolar fluctuations and conduction electrons in TCF. The derivation can easily be adapted to arbitrary situations, as further illustrated in the Supplemental Material [19]. In each case, the local degrees of freedom are symmetrically coupled to two different irreps of conduction electrons. The coupling has the form of the 2CK model plus relevant, spin symmetry breaking perturbations present even in zero magnetic field. Solving the model with NRG we showed that the hypothesis introduced in Ref. [6] to describe the HO in URu_2Si_2 can consistently account for the behavior of TURS. Nonetheless, this behavior does not discriminate between different competing scenarios for the ground state–excited state sequence in this material. As there are always two irreps of conduction electrons involved in the coupling, and they are not connected by symmetry, we expect a FL to emerge at sufficiently low T's. Hence the intermediate NFL regime, observed in URu_2Si_2, is a result of accidental degeneracy, and is responsible for the unique properties of this compound in the dilute and dense limits, among the hundreds of known U-based heavy fermions. We found that the scale of the TCF splitting and T_K is smaller in TURS than in URu_2Si_2 [6]. The splitting between the two Γ_7 irreps should be sensitive to the conduction electron filling, and we expect it to be larger in $La_{1-x}U_xRu_2Si_2$ where clear FL behavior is observed [23]. In this context it is also worth pointing out that, in TURS, the resistivity follows an approximate $\log T$ behavior with a negative coefficient suggesting a crossover to FL behavior at sufficiently low T's. The study of the resistivity, however, will likely require a more realistic model for TURS including all bands present in the solid, and also a more sophisticated approach to calculating the resistivity in NFL quantum impurity models than the ones presently available [24].

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