

## Lifshitz Transition in Kondo Alloys

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We study the low-energy states of Kondo alloys as a function of the magnetic impurity concentration per site  $x$  and the conduction electron average site occupation  $n_c$ . Using two complementary approaches, the mean-field coherent potential approximation and the strong-coupling limit, we identify and characterize two different Fermi-liquid regimes. We propose that both regimes are separated by a Lifshitz transition at  $x = n_c$ . Indeed, we predict a discontinuity of the number of quasiparticles that are enclosed in the Fermi surface. This feature could provide a scenario for the non-Fermi liquid properties that were recently observed in Kondo alloy systems around  $x = n_c$ .

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Kondo alloys are realized in broad families of strongly correlated materials where magnetic quantum impurities are embedded randomly in a crystalline system with conduction electrons [1]. The Kondo problem has been intensively studied for almost half a century [1], and the single-impurity model was solved exactly by various methods [2–6]. In the 1970s, Nozières adapted Landau’s approach [7,8], showing that a single Kondo impurity can be described universally at low energy as a local Fermi liquid (LFL) [9]. Multiple-impurity models were later introduced to describe systems with a high concentration of Kondo ions. It appeared that, unlike the single-impurity models, the multiple-impurity models did not belong to a single universality class, but could rather give rise to a large variety of ground states depending on the lattice structure, the electronic occupation, and the strength of the Kondo coupling, which can be tuned with pressure [10,11].

Putting aside the relevant issues of ordering, we concentrate here on the coherent Fermi liquid (CFL) ground state that is observed at low temperature in many dense Kondo systems [12]. The electronic exhaustion problem [13,14] suggested by Nozières remained a long-standing issue for years [15–23] and was finally understood [21,24]: the Kondo temperature  $T_K$  that characterizes the crossover to the low-temperature Kondo screening regime and the Fermi liquid energy scale  $T_{\text{coh}}$  that characterizes the CFL are indeed the same energy scale. It was shown that the ratio  $T_K/T_{\text{coh}}$  still depends on the electronic filling and lattice structure [21,25,26]. But this ratio was proven not to depend on the Kondo coupling, as was incorrectly initially thought [21,22,24].

A simple picture of the periodic dense Kondo lattice model had been provided within the strong Kondo coupling limit [14,15,27–29]. Even if this limit does not correspond to the experimental reality, its qualitative validity is supported by a poor-man’s scaling analysis [1,30],

which shows that the low-temperature Kondo physics of the single-impurity model is renormalized to a strong-coupling fixed point. In the infinite coupling limit, the ground state of a Kondo lattice made of  $N$  sites with  $N_S = N$  spins 1/2 is characterized by the formation of  $N_c$  local Kondo singlets, where  $N_c$  denotes the number of conduction electrons. The CFL is recovered from a perturbation expansion at lowest order in *hopping/coupling*. The corresponding fermionic quasiparticles correspond to the “bachelor” Kondo spins, whose double occupancy is forbidden, giving rise to strong correlation effects. This strong-coupling description also provides the correct number of quasiparticles:  $N - N_c$  free spins, which according to a particle-hole transformation, give  $2N - (N - N_c) = N_c + N$  quasiparticles (the factor 2 comes from the spin degeneracy). This result is in perfect agreement with the Luttinger theorem [31], which predicts an enlargement of the Fermi surface due to the contribution of the Kondo impurities [14,21].

Our analysis starts here: on one hand, the universal single-impurity Kondo model can be described as a LFL. This picture well describes Kondo systems with very dilute impurities. On the other hand, many realizations of dense Kondo systems with a periodic lattice of magnetic ions have a CFL ground state. Can these two Fermi liquids be continuously connected at zero temperature?

We consider a Kondo alloy model (KAM), defined by the Hamiltonian

$$H = \sum_{ij\alpha} t_{ij} c_{i\alpha}^\dagger c_{j\alpha} + J_K \sum_{i \in K} \mathbf{S}_i \cdot \boldsymbol{\sigma}_i, \quad (1)$$

where  $c_{i\alpha}^{(\dagger)}$  denotes annihilation (creation) operator of a conduction electron with spin  $\alpha = \uparrow, \downarrow$  on site  $i$  of a periodic lattice that contains  $N$  sites;  $t_{ij}$  denotes intersite hopping energy, and  $J_K$  is a local Kondo antiferromagnetic interaction between local quantum spin 1/2 denoted  $\mathbf{S}_i$  and

the local density of spin of conduction electrons  $\sigma_i$ . These  $N_S \equiv xN$  Kondo spins, with concentration  $x \leq 1$ , are located on a subpart  $K$  of the periodic lattice. We choose a frozen configuration for  $K$  sites, whose positions are distributed randomly without spatial correlation. The number of conduction electrons is fixed to  $N_c \equiv n_c N$ , and we restrict  $n_c \leq 1$  for particle-hole symmetry reasons.

First, the KAM is studied using the mean-field approximation for the Kondo interaction, [11,32,33] and considering the random position of  $K$  sites within a CPA-DMFT method [25], i.e., a matrix version of dynamical mean field theory (DMFT) [34,35], which is equivalent to the matrix version of the coherent potential approximation (CPA) [36,37]. Kondo alloy models had been studied earlier using CPA methods, but focusing mostly on the high concentration of impurities [38–40]. In Ref. [41], an Anderson alloy model was also studied using CPA and DMFT methods, but with a choice of parameters which enforces the system to be always in the CFL regime. The matrix CPA-DMFT formalism that is presented in detail in Ref. [25] treats Kondo and non-Kondo sites separately. For the sake of simplicity, the numerical calculations were performed assuming that the conduction electrons move on a Bethe lattice, characterized by an elliptic noninteracting local density of states (DOS), as depicted on Fig. 1. The local DOS on a  $K$  site is analyzed as a function of  $x$ ,  $n_c$ , and the temperature  $T$ . At high  $T$  (left column on Fig. 1), it corresponds to the noninteracting conduction band, and for  $T \approx T_K$  (second column) a Kondo resonance forms

around the Fermi level  $\omega = 0$ . The position of the resonance depends on  $n_c$ , but its shape at  $T \approx T_K$  does not depend on  $x$ . This is fully consistent with the interpretation that  $T_K$  characterizes the incoherent local Kondo singlet formation. At lower temperature (right column), when a Fermi-liquid regime sets in, the local DOS can exhibit two qualitatively different features: for  $x < n_c$  [Figs. 1(a) and 1(c)], it remains roughly similar to the noninteracting one, with just a small deformation around the Fermi level, due to the Kondo resonance. But for  $x > n_c$  [Figs. 1(b) and 1(d)], the Kondo resonance is split by a hybridization gap. A similar reduction of the DOS has already been suggested in Refs. [38,42]. This splitting can be explained by the standard two-band picture, which is commonly used for Kondo lattices. A systematic analysis of the DOS at low  $T$  for different impurity doping and electronic filling clearly shows that the crossover between CFL (with a gap) and LFL (without gap) regimes occurs when  $x = n_c$ . The dense system remains a Fermi liquid since the gap occurs at  $\omega > 0$ , i.e., above the Fermi level. However, this mean-field result suggests that the Kondo-interacting (at low  $T$ ) and the Kondo-decoupled (at high  $T$ ) Fermi liquids can be smoothly connected to each other only for  $x < n_c$ . At larger impurity concentrations (or equivalently, at lower electronic filling), a singularity may disconnect the two Fermi-liquid regimes, related to this gap opening.

Studying the strong-coupling limit  $J_K \gg |t_{ij}|$  provides clear evidence of the analytical disconnection between the CFL ( $N_S > N_c$ ) and the LFL ( $N_S < N_c$ ). Hereafter, we generalize to the KAM the exact strong-coupling treatment of Ref. [15]. We start with the extreme limit  $t_{ij} = 0$ . Depending on  $N_c/N_S = n_c/x$ , we can distinguish two different situations, as illustrated in Fig. 2. For  $N_S < N_c$ , the ground state is characterized by  $N_S$  local Kondo singlets on each Kondo site.  $N_S$  electrons are thus localized on the  $K$  sites that effectively become “forbidden sites” for the remaining  $N_c - N_S$  electrons. The latter can freely occupy the left  $N - N_S$  non-Kondo sites. This degenerate ground state has a finite entropy per site  $\mathcal{S}_{\text{LFL}} = (1-x)\ln(1-x) - (n_c-x)\ln(n_c-x) - (1-n_c)\ln(1-n_c)$ . For  $N_S > N_c$ , the ground state is characterized by the formation of  $N_c$  Kondo singlets. The non-Kondo sites are empty, but a degeneracy results from the various energetically equivalent possible positions of the Kondo screened impurities on  $K$  sites. The resulting entropy per site is

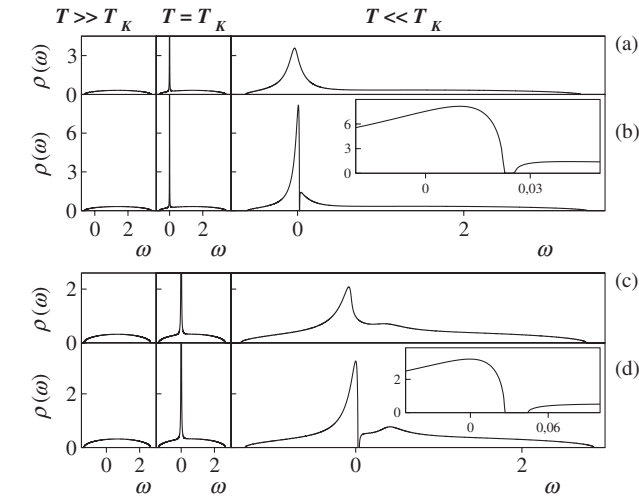


FIG. 1. Local DOS on a Kondo site for  $n_c = 0.20$  [(a) and (b)] and  $n_c = 0.60$  [(c) and (d)] and impurity concentrations (a)  $x = 0.10$ , (b)  $x = 0.30$ , (c)  $x = 0.50$ , and (d)  $x = 0.70$ . Left column: elliptic DOS obtained at  $T \gg T_K$ . Second column: formation of the Kondo resonance, at  $T \approx T_K$ . Right column: comparison between the dilute LFL [(a) and (c)] and dense CFL [(b) and (d)] regimes. Insets: hybridization gaps above the Fermi level, which characterize the CFL regime. Parameters:  $J_K = 1.5$  and electronic bandwidth  $W = 4$ , with  $T_K = 0.099$  [(a) and (b)] and  $0.18$  [(c) and (d)].

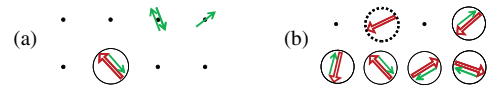


FIG. 2 (color online). Schematic picture of the ground state of the KAM for large  $J_K$  values. Kondo spins are depicted by red double-line arrows, and electrons are green single-line arrows. Black points refer to non-Kondo sites. Kondo singlets are marked by a solid circle. (a) Dilute case. (b) Dense case: dotted circles indicate unscreened Kondo spins (see text).

$\mathcal{S}_{\text{CFL}} = x \ln(x) - n_c \ln(n_c) - (x - n_c) \ln(x - n_c)$ . Both  $\mathcal{S}_{\text{LFL}}$  and  $\mathcal{S}_{\text{CFL}}$  are analytical functions of  $x$  and  $n_c$ , but they cannot be analytically connected to each other. Indeed, both vanish like  $\mathcal{S} \approx -|x - n_c| \ln|x - n_c|$  when  $x \rightarrow n_c$ . The presence of an absolute value in this expression is clearly identified as a singularity separating the two regimes. Furthermore, the vanishing of  $\mathcal{S}$  at  $x = n_c$  suggests that this point does not correspond to a Fermi liquid even when considering perturbations in electronic hopping  $t$ .

Then, we consider the lowest order in perturbation expansion, which is linear in  $t$ . Fermi liquids characterize both dilute and dense regimes, but a singular point separates the LFL from the CFL.

**LFL regime.** The first perturbative correction generates an effective model where the fermionic quasiparticles are  $N_{\text{qp}}^{\text{LFL}} = N_c - N_S$  free electrons moving with hopping energy  $t_{ij}$  on the lattice made of the  $N - N_S$  non-Kondo sites [see Fig. 2(a)]. Of course this is valid only if  $1 - x$  is bigger than the percolation threshold  $x_p$ . The only correlation effect in this effective model results from the frozen random depletion of the Kondo sites. This picture recovers the early approaches of Nozières to the single-impurity model, which was mapped onto a problem of light conduction electrons scattering on the Kondo singlet [9,13,14]. Here, light conduction electrons scatter on the  $K$  sites, which are effectively excluded. From CPA approach, we find that the site depletion leads to an effective conduction band for the  $N_{\text{qp}}^{\text{LFL}}$  quasiparticles, with a renormalized hopping term  $t_{ij}\sqrt{1-x}$ . The local DOS is mostly renormalized by the site depletion, and its total spectral weight is reduced by a factor of  $1-x$ . The missing weight  $x$  is transferred to a higher energy peak, which corresponds to excitations where one of the  $K$  sites has 0 or 2 conduction electrons. Note that the occupation  $N_{\text{qp}}^{\text{LFL}}$  corresponds to  $(n_c - x)/2$  quasiparticles per site and per spin component; this filling is smaller than  $1-x$ , since  $n_c < 1$ .

**CFL regime.** A perturbation expansion at lowest order provides an effective model where fermionic quasiparticles correspond to the unscreened Kondo impurities. The mapping is very similar to the one described for a periodic Kondo lattice [15]. Here, we assume that the  $K$  sites' concentration exceeds the percolation threshold, i.e., that  $x > x_p$ . The unscreened impurities move on a depleted lattice made of the  $K$  sites. There are  $N_S - N_c$  unscreened spins moving on  $N_S$  sites, and since their motion is holelike, the number of quasiparticles is  $N_{\text{qp}}^{\text{CFL}} = 2N_S - (N_S - N_c) = N_S + N_c$ . This suggests that the Luttinger theorem applies in the CFL regime, both electrons and Kondo spins contributing to the Fermi surface. Here, the correlations have two origins: the random depletion, which also characterizes the LFL regime, and a supplementary infinite repulsion which prevents an unphysical double impurity occupation on a same site. Since the moving holes are Kondo singlets, the lattice hopping is also renormalized

by a factor of  $1/2$  as explained in Ref. [15]. An extra renormalization of the hopping results from the lattice site depletion. Invoking a CPA approach, the intersite hopping for the correlated quasiparticles is  $t_{ij}\sqrt{x}/2$ . The local DOS is renormalized in two steps: first, the site depletion reduces the bandwidth by a factor of  $\sqrt{x}$  and the spectral weight by a factor of  $x$ . The missing spectral weight here corresponds to excitations moving an electron from a Kondo singlet to a non-Kondo site (with 0 or 2 conduction electrons). Then, the peak at lowest energy is split again due to the effective Hubbard repulsion. We may interpret the upper forbidden Hubbard band as corresponding to excitations of local Kondo singlets into local triplets. This singlet-triplet excitation mode remains gapped (or pseudogapped) in the opposite limit,  $J_K \ll t$ , as indicated by mean-field calculations (see Fig. 1). Note that the occupation  $N_{\text{qp}}^{\text{CFL}}$  corresponds to  $(x + n_c)/2$  quasiparticles per site and per spin component; this filling is smaller than  $x/2$ , and the associated chemical potential is thus inside the lowest Hubbard band. This mapping to an effective Hubbard model is exact in the strong Kondo coupling limit, but it is interesting to observe that the numerical results obtained from the mean-field approximation with weak coupling parameters also provide a splitting of the DOS with a gap opening in the dense regime (see Fig. 1).

The characteristics of the strong-coupling effective models obtained in the CFL and LFL regimes are compared in Table I. The impossibility of analytically connecting these two families of models thus survives to finite  $t$  corrections. We identify  $N_c = N_S$  to a critical point with the following singularities: the number of quasiparticles is discontinuous as well as the number of effective sites, the effective hopping, and the Hubbard repulsion, which changes from 0 to  $\infty$ . For  $N_c = N$ , this singular point corresponds to a Kondo insulator and is physically accessible only from the LFL side. For  $N_c < N$ , the singularity of this point remains, and it is accessible from both LFL and CFL sides. The critical point is not associated to a symmetry breaking, but to a singular change of the Fermi surface known as a Lifshitz transition (LT) [43,44].

The number of quasiparticles is plotted in Fig. 3 as a function of  $N_S$  for  $J_K \gg t$ . The depicted situation

TABLE I. Characteristics of dense and dilute regimes for large  $J_K$  values.

Characteristic	LFL regime	CFL regime
	$N_S < N_c$	$N_S > N_c$
Number of quasiparticles	$N_c - N_S$	$N_c + N_S$
Effective intersite hopping	$t_{ij}$	$t_{ij}/2$
Number of effective sites	$N - N_S$	$N_S$
CPA rescaled hopping	$t_{ij}\sqrt{1-x}$	$t_{ij}\sqrt{x}/2$
Effective Hubbard repulsion	0	$\infty$
Spectral weight of the lower band	$1-x$	$x/2$

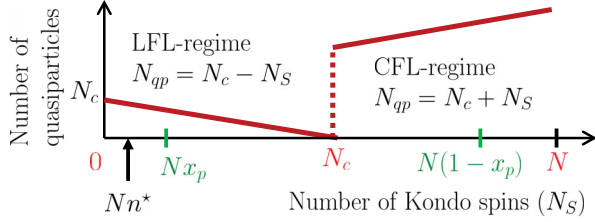


FIG. 3 (color online). Number of quasiparticles as a function of  $N_S$  for fixed  $N_c$ . A transition is expected at  $N_S = N_c$ , when  $x_p < 1/2$ . For realistic (i.e., small) Kondo coupling, only a small part of the conduction electrons is involved in the Kondo screening of the LFL regime, resulting in an intermediate third regime  $Nn^* < N_S < N_c$ .

corresponds to a KAM, which is realized by substitution of non-Kondo (example La) with Kondo (example Ce) atoms, with fixed light electron structure and filling. In the CFL regime, the Kondo spins contribute to the volume of the Fermi surface, which is consistent with the general idea of Luttinger theorem. The formation of this large Fermi surface requires a sufficiently high amount of Kondo impurities, in order to form a CFL. But in the LFL regime, the Luttinger theorem is expected to break down. There, Kondo impurities are too diluted for contributing to the Fermi surface. Instead, their main effect is rather to capture conduction electrons for the formation of localized Kondo singlets. Only the resulting  $N_c - N_S$  electrons contribute to the Fermi surface and scatter on the localized Kondo singlets. The discontinuity of  $N_{qp}$  is related to a LT: the Fermi surface is not defined at the critical point  $N_c = N_S$ . This is true for  $N_c = N_S = 1$  because the system is in this case a Kondo insulator, but we predict this situation to occur more generally for  $N_c = N_S$  not necessarily equal to 1. Approached from the LFL side, this critical point corresponds to a continuous collapse of the Fermi surface. But this collapse is discontinuous from the CFL side.

Of course, this analysis does not consider possible issues of cluster formations and we also assumed percolation of the depleted effective lattices. An intermediate Kondo disorder regime with cluster formation is expected for lattices with a big percolation threshold  $x_p > 1/2$  (which is usual in two-dimensional systems with only nearest neighbor hopping). The most realistic cases are three-dimensional systems with low percolation threshold  $x_p < 1/2$  and intermediate electronic filling. Two situations can thus occur: the LT should be observed for  $x_p < n_c < 1 - x_p$ , whereas a Kondo disorder phase is expected for other values of  $n_c$ . Here, we did not analyze the Kondo disorder regime, where inhomogeneities may control the physical properties as discussed in Ref. [45]. We also did not consider the issue of magnetic ordering, but it is expected to be weakened by a factor of  $x^2$  in a Kondo alloy. Moreover, the magnetic interaction in the strong-coupling limit is small ( $t^2/J_K$  [15]).

Furthermore, it appeared that Kondo screening in the CFL is a collective effect that involves all electrons and all impurities, whereas in the LFL only a small part of conduction electrons, of the order of  $T_K/t \equiv n^*$ , contribute to the Kondo screening at small coupling [21,24]. This problem does not occur in the strong-coupling limit, but it gives rise to a critical concentration  $n^*$  for more realistic coupling. In these cases, the LT at  $N_S = N_c$  may become a crossover spread from  $x \approx n^*$  to  $x = n_c$ . It is not clear whether signatures of a Fermi surface were observed experimentally in this whole crossover regime, and the NFL regime may spread as well along a large range of intermediate concentrations separating the LFL from the CFL. We propose that in the heavy-fermion compound  $Ce_xLa_{1-x}Ni_2Ge_2$  the system goes from LFL to non-Fermi liquid (NFL) for  $x \approx n^* \ll 1$  and then from NFL to CFL for  $x = n_c \approx 0.6$  (see Fig. 4 in Ref. [46]). This mechanism may also provide a possible scenario for the origin of the NFL properties observed recently in  $Ce_xLa_{1-x}PtIn$  [47]. A difference (transition or crossover) between a dilute Kondo system and a dense heavy fermion was also suggested by other theoretical methods, including finite size calculations [29], quantum Monte Carlo simulations [48,49], and Gutzwiller approximation [50]. We propose to analyze more systematically Kondo alloys to find other examples of NFL behavior of this type. These studies should be completed by measurements that analyze the Fermi surface (angle-resolved photoemission spectroscopy, quantum oscillations) in order to test the predicted validity and violation of the Luttinger theorem, respectively in the dense and dilute regimes. One difficulty that may be faced in some compounds would come from a complex electronic band structure. Usually, several conduction bands are involved, and the criteria  $N_c = N_S$  might be not always reachable. The Lifshitz critical point could also have signatures in other experimental realizations of Kondo alloys, in bulk materials, or in artificial atom systems such as quantum dot arrays or optical lattices.

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