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Kondo effect in “bad metals”

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We study the low-temperature properties of a Kondo lattice using the large- \mathcal{N} formalism. For a singular density of conduction states (DOS), we generalize the single-impurity result of Withoff and Fradkin: the strong-coupling fixed point becomes irrelevant if the DOS vanishes at the Fermi level E_F . However, for E_F close enough to the singularity, and close to half-filling, the Kondo temperature, T_K , can become much smaller than the characteristic Fermi liquid scale. At $T=0$, a meta-magnetic transition occurs at the critical magnetic field $H_c \simeq (k_B/\mu_B)T_K$. Our results provide a qualitative explanation for the behavior of the YbInCu₄ compound below the valence-change transition.

The Kondo model has been introduced to explain the transport and thermodynamic anomalies of dilute magnetic alloys [1] due to weak interaction between the conduction electrons and impurity spins. The model is defined by the Hamiltonian $H_K = J\mathbf{s} \cdot \mathbf{S}$, where \mathbf{s} and \mathbf{S} are the spins of conduction and localized electrons, respectively, and J is the coupling constant. Our current understanding of Kondo systems is based on the solutions obtained by the perturbative scaling [2], numerical renormalization group [3], and Bethe Ansatz [4], which are usually derived for a featureless conduction band. The main result is the crossover from the high-temperature regime, where the impurity is magnetic and the conduction electrons are only weakly perturbed by the spins, to the strong coupling regime, where the magnetic moment is screened completely and the conduction states form a Fermi liquid [3]. The Kondo temperature separates these two regimes and is the only relevant energy scale.

Kondo systems with a non-constant DOS or a high concentration of magnetic impurities show more complicated behavior. Assuming an electron-hole symmetry and a DOS with a power-law singularity at the Fermi level E_F , Withoff and Fradkin [5] found a critical coupling J_c such that for $J > J_c$ the dilute Kondo systems scale to the strong coupling limit, while for $J < J_c$ the renormalized coupling decreases with temperature and the “usual” Kondo screening does not occur. Here, we study the low-temperature Kondo effect in bad metals and consider a lattice of magnetic ions coupled by exchange interaction to a conduction band with a minimum (or a gap or a pseudo-gap) close to E_F . We show that such a model generates more than one energy scale and explains the low-temperature behavior of Yb- and Eu-based intermetallic compounds, like YbInCu₄ [6], EuNi₂(Si_{1-x}Ge_x)₂ [7] or Eu(Pd_{1-x}Pt_x)₂Si₂ [8], which exhibit a valence-change transition. We also study the Kondo lattice with an enhanced DOS around E_F and show that it explains the properties of YbAl₃ [9] in the coherent regime.

The Kondo lattice Hamiltonian reads,

$$H = \sum_{\langle i,j \rangle, \sigma} (t_{ij} - \mu \delta_{ij}) c_{i\sigma}^\dagger c_{j\sigma} + \frac{J}{\mathcal{N}} \sum_i \mathbf{s}_i \cdot \mathbf{S}_i, \quad (1)$$

where t_{ij} is the hopping, $c_{i\sigma}^\dagger$ creates an electron of spin $\sigma = 1, \dots, \mathcal{N}$ at site i , \mathbf{s}_i is the c -electrons local spin density, μ is the chemical potential which determines the band filling, $n_c/2 = \mathcal{N}^{-1} \sum_\sigma \langle c_{i\sigma}^\dagger c_{i\sigma} \rangle$, and \mathbf{S}_i are the local spins of $SU(\mathcal{N})$ symmetry. We use the large- \mathcal{N} approach, write $\mathbf{S}_{i\sigma, \sigma'} = f_{i\sigma}^\dagger f_{i\sigma'} - \delta_{\sigma\sigma'}/2$, where $\sigma, \sigma' = 1, \dots, \mathcal{N}$, and enforce the local constraints $\sum_{\sigma=1}^{\mathcal{N}} f_{i\sigma}^\dagger f_{i\sigma} = \mathcal{N}/2$ by the Lagrange-multipliers $i\lambda_i(\tau)$ [10,11]. The quartic fermionic interactions in Eq. (1) are decoupled by introducing Hubbard-Stratanovich transformations [11], which lead in the limit $1/\mathcal{N} = 0$ to the following set of equations for the hybridization parameter r and the Lagrange multiplier λ ,

$$\left\{ -\frac{r}{J}, \frac{1}{2}, \frac{n_c}{2} \right\} = -\{G_{fc}, G_f, G_c\} (\tau = 0^-). \quad (2)$$

Here, $G_f(\tau) = -\langle T f_0(\tau) f_0^\dagger(0) \rangle$ is the f -fermion Green’s function, $G_{fc}(\tau) = -\langle T f_0(\tau) c_0^\dagger(0) \rangle$ is the mixed Green’s function, and $G_c(\tau) = -\langle T c_0(\tau) c_0^\dagger(0) \rangle$ the c -electron Green’s function, all of them being computed for an arbitrary site $i = 0$ (In the paramagnetic state the spin index has been suppressed). Local Green’s functions can be obtained from the free fermionic propagator $\mathcal{G}_f(i\omega_n) = (i\omega_n + \lambda)^{-1}$, using the relations $G_c(i\omega_n) = G_c^0(i\omega_n + \mu - r^2 \mathcal{G}_f(i\omega_n))$, $G_f(i\omega_n) = \mathcal{G}_f(i\omega_n) + r^2 \mathcal{G}_f^2(i\omega_n) G_c(i\omega_n)$ and $G_{fc}(i\omega_n) = r \mathcal{G}_f(i\omega_n) G_c(i\omega_n)$, in which ω_n are the Matsubara fermionic frequencies, and G_c^0 the non-interacting electronic Green function [10,11]. Typical band-shapes and fillings considered in this work are shown in Fig. 1.

The temperature T_K at which the second order transition with the order parameter $r(T)$ takes place is obtained from Eqs. (2) as a non trivial solution of the equation ($r = 0$, $J > J_c$),

$$\frac{2}{J} = \int_{-\infty}^{+\infty} d\omega \frac{\tanh[\omega/2T_K]}{\omega} \rho_0(\omega + \mu). \quad (3)$$

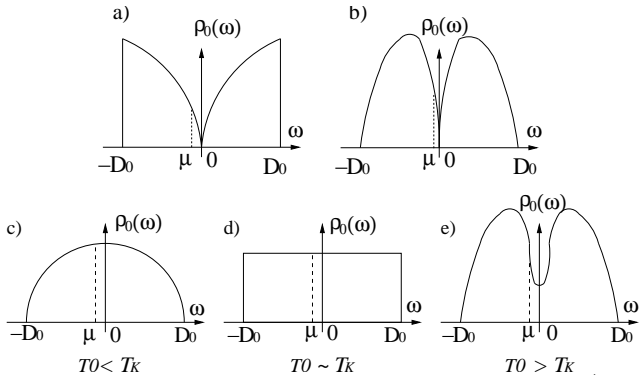


FIG. 1. Schematic non interacting DOS: singular (a and b), semi-elliptic (c), constant (d), and pseudo-gaped (e). As soon as $\rho_0(\omega = \mu) \neq 0$, the Kondo temperature T_K is finite. Close to the half-filling, the shape of the DOS around μ is crucial for determining the Fermi-liquid temperature: (c) μ is close to a maximum of ρ_0 and $T^* < T_K$. (e) μ is close to a minimum of ρ_0 , and $T^* > T_K$. (d) ρ_0 is constant and $T^* \sim T_K$.

Here, $\rho_0(\omega)$ is the non-interacting DOS and the energy is measured with respect to the center of the conduction band, which is assumed to be symmetric and of half-width D_0 . Assuming spin-rotation symmetry, we find that the large- \mathcal{N} result for the lattice (3) coincides with the solution of the scaling equations for the $SU(2)$ single-impurity Kondo model [5]. The T_K vanishes continuously at

$$\frac{2}{J_c} = \int_{-\infty}^{+\infty} d\omega \frac{\rho_0(\omega + \mu)}{|\omega|}, \quad (4)$$

and for $\rho_0(\mu) = 0$ the model admits a finite J_c and a quantum critical behavior. In the case of a DOS with a gap $\Delta_0 \ll D_0$ around E_F we find $J_c \propto D_0/\ln[D_0/\Delta_0]$, while the pseudo-gap centered at E_F and characterized by a single energy scale D_0 gives $J_c \propto D_0$. The power-law singularity $\rho_0(\omega) = A|\omega|^\gamma$ gives $J_c = 2\gamma D_0/(\gamma+1)$, which is the Whithoff and Fradkin result [5,13] (the constant A follows from the normalization condition, $\int \rho_0(\omega) = 1$). However, for $\rho_0(\mu) \neq 0$ (see Figs. 1(a) and 1(b) or 1(e)), the integral diverges logarithmically and $J_c = 0$.

Taking the non-constant DOS and solving Eq.(3) in the weak coupling limit gives [11], $T_K = F_K \exp[-1/J\rho_0(\mu)]$ where

$$F_K = \alpha \sqrt{D_0^2 - \mu^2} \exp \left(\int_{-D_0-\mu}^{D_0-\mu} \frac{d\omega}{|\omega|} \frac{\rho_0(\mu + \omega) - \rho_0(\mu)}{2\rho_0(\mu)} \right), \quad (5)$$

where $\alpha \approx 1.13$. This result is derived in ref. ([11]) for an even and analytic DOS but it also holds for $\rho_0(\omega)$ with an algebraic singularity close to E_F . The T_K defines the Kondo temperature of the lattice; it characterizes the high-temperature behavior and gives the temperature at which the weak coupling behavior breaks-down due to the

single-impurity singlet formation. The screening cloud at $T \approx T_K$ involves the electronic states in an energy interval $|\omega| \leq T_K$ around E_F (E_F is the non-interacting Fermi level of n_c electrons having a “small” Fermi surface, FS).

At low temperatures, Eqs. (2) lead to a Fermi liquid (FL) ground state which is characterized by two energy scales, T^* and T^{**} . The scale T^* is defined as the inverse of the quasi-particle DOS at the Fermi energy of a “large FS” which contains, at $T = 0$, $n_c + n_f$ quasi-particles (n_f is the occupancy of the localized f -orbitals, which is close to 1 for $SU(2)$ Kondo systems). This scale is relevant for the $T \rightarrow 0$ properties and appears in the static spin susceptibility, $\chi_{loc}(T = 0) \sim 1/T^*$, or the specific heat coefficient $C/T \sim 1/T^*$. The scale T^{**} characterizes the deformation of the “quasi-particle band” with temperature; it defines the relevant energy scale for quasi-particle excitations and can be related to the temperature at which the coherence is lost and the quasiparticles Fermi surface destroyed. This scale depends on all the electronic states between “small” and “large” FS, as well as on some hole states inside the “small” FS [11]. The system behaves as a FL provided $T/T_{FL} \ll 1$ where T_{FL} is the smallest of the three temperatures defined above. One common situation arises for $T^* \ll T^{**} \ll T_K$, such that the band of quasiparticles is “rigid” and we have the FL laws, provided this rigid band does not have too much structure within the Fermi window. The other usual situation is $T^{**} \ll T^* \ll T_K$, such that the deviations from the FL laws are due to the temperature-induced deformation of the effective DOS, i.e. the break-down of the FL is due to the loss of coherence. A different behavior is expected in bad metals with only a few states around E_F and $T_K \ll T^{**}$ or $T_K \ll T^*$.

To study the relative magnitude of T_K , T^* , and T^{**} , we use the analytic expressions obtained from Eqs. (2) and evaluate them close to half-filling [11,12]. This gives $T^* \approx T^{**} = F_0 \exp[-1/J\rho_0(\mu)]$, and

$$F_0 = (D_0 + \mu) \exp \left(\int_{-D_0-\mu}^{\epsilon_F^>-\mu} \frac{d\omega}{|\omega|} \frac{\rho_0(\mu + \omega) - \rho_0(\mu)}{\rho_0(\mu)} \right), \quad (6)$$

where $\epsilon_F^>$ is the Fermi level corresponding to $(n_c + 1)/2$ non-interacting fermions per spin component. The break-down of the FL laws is now due to quasi-particle excitations and a simultaneous destruction of the coherent state. Note, we assume that the system is not at half-filling exactly (where the “large” FS coincides with the Brillouin zone and we would get a Kondo insulator). But we also assume that the system is not too far from the particle-hole symmetry, because for small filling the ‘exhaustion’ gives rise to additional effects (see discussion by Nozières [14] and [15]). Both assumptions are fulfilled for $n_f \neq 1$ but not for $n_f = 1$ or $n_f \simeq 0$. The expressions for F_K and F_0 can be sim-

plified to $F_K \approx \alpha D_0 \exp\left(\int_{-D_0}^{D_0} \frac{d\omega}{|\omega|} \frac{\rho_0(\mu+\omega) - \rho_0(\mu)}{2\rho_0(\mu)}\right)$ and $F_0 \approx D_0 \exp\left(\int_{-D_0}^{D_0} \frac{d\omega}{|\omega|} \frac{\rho_0(\mu+\omega) - \rho_0(\mu)}{\rho_0(\mu)}\right)$, such that $F_0 \approx (F_K/\alpha)^2/D_0$. Thus, we have the relationships between the Kondo temperature and the FL scale

$$T^* = \frac{F_K}{\alpha^2 D_0} T_K, \quad (7)$$

regardless of the specific form of ρ_0 . A constant ρ_0 gives $F_K/D_0 = 1$ and $T^* \sim T_K$, which explains the T/T_K scaling observed in many heavy fermion compounds.

We consider now in more detail the effects due to the shape-variation of $\rho_0(\omega)$. If μ is close to a local maximum of $\rho_0(\omega)$ the integrand in F_K is mainly negative, such that $F_K/D_0 < 1$ and $T^* \leq T_K$, as found in systems showing ‘protracted screening’ [16,9] (exhaustion effects can also reduce T^*/T_K [14] and see [15]). However, if μ is close to a local minimum (see Fig. 1(e)) one finds $T_K \ll T^*$. Note, the relative magnitude of T^* and T_K does not depend on T_K (or J) but is related to the convexity of $\rho_0(\omega)$ around $\omega = \mu$. An intuitive interpretation is provided by the following argument. The incoherent Kondo cloud forms at $T \approx T_K$ and involves only a few states around E_F . But the energy scale T^* depends on the density of quasiparticle states at the Fermi level of the ‘‘large’’ FS, and the energy T^{**} involves all the states between the ‘‘small’’ and the ‘‘large’’ FS, and some additional holes inside the ‘‘small’’ FS [11,12]. Thus, for μ close to the minimum of $\rho_0(\omega)$, the integrals for T^* and T^{**} involve the DOS which is much larger than the one used to evaluate T_K , and $T_K \ll T^*$ follows.

Next, we study the effects of an external magnetic field H applied to the spin system at $T = 0$. Within the large- \mathcal{N} approach, we obtain a transition between the Kondo phase ($r \neq 0$, ‘‘large FS’’) and the ferromagnetic spin lattice decoupled from the paramagnetic metal ($r = 0$, ‘‘small’’ FS). The critical field H_c is given by the relation

$$\frac{2}{J} = \int_{-\infty}^{+\infty} d\omega \tanh\left[\frac{\omega}{2T}\right] \frac{\omega}{\omega^2 - H_c^2} \rho_0(\omega + \mu), \quad (8)$$

which shows that the logarithmic divergence of Eq.(3) disappears, and $J_c \neq 0$, as soon as $H_c \neq 0$. In the weak coupling limit, $J \ll D_0$, we find $H_{c0} = \alpha^{-1} F_K \exp[-1/J\rho_0(\mu)]$ and obtain, at $T = 0$, the universal relation $H_{c0} = k_B T_K / \mu_B \alpha$. Thus, in Kondo lattices with a non-constant DOS we can expect two different types of the zero-temperature magnetization $m(H)$. For $T^* \ll T_K$ and $H \ll H_c$, the magnetization rises initially as $m(H) \propto H/T^*$ but for higher fields the slope is reduced and eventually, around $H^* \propto k_B T^* / \mu_B \ll k_B T_K / \mu_B$, the $m(H)$ saturates. For $H^* \ll H \ll H_c$ we are dealing with a saturated FL ($r \neq 0$). In the opposite case, $T_K \ll T^*$ and $H_c \ll H^*$, the low field limit still gives $m(H) \propto H/T^*$. But for higher fields the slope of $m(H)$ rises rapidly and at about $H_c \propto k_B T_K / \mu_B$ there

is a meta-magnetic transition into the spin-disordered ($r=0$) phase. Of course, this simple considerations should be corrected for direct and indirect effects due to the conducting sea.

Solving Eq. (8) for a constant DOS we find the critical line $[H_c(T)/H_{c0}]^2 + [T/T_K]^2 = 1$, which holds for any J and n_c . The same relation is also found at half-filling, for any DOS. In general, we expect a ‘nearly’ universal phase boundary, with some small deviations due to the structure of the DOS *and* the particle-hole asymmetry.

We can use these results to discuss the anomalous behavior of the Yb- and Eu-based intermetallics that we have mentioned before. In YbInCu₄, which is a typical example [6], the valence-change transition takes place, at ambient pressure, at $T_v \approx 40K$. Above T_v , one finds the Yb ions in a 3^+ state, behaving as ‘almost free spins’, and the transport properties of a bad metal. That is, the linear and non-linear magnetic response of the paramagnetic phase are very well explained by the crystal field theory of independent f-states, while the electrical resistance is very high, there are no logarithmic terms, and the fields of up to 40 Tesla do not produce any significant effects. The Kondo scale determined from the high-temperature properties seems to be very small. Below T_v , the valence state of Yb ions changes to 2.9^+ and the system behaves as a FL with a high characteristic scale, $T^* \approx 500K$ [17], such that the physical quantities are nearly temperature-independent. The optical conductivity [18], Hall effect [19] and the thermoelectric power [20] indicate a major reconstruction of the conduction band: a bad metal with chemical potential close to the pseudo-gap (or small gap) transforms at T_v into a good metal with a large FL scale. The FL ground state can be destroyed by a magnetic field of about $H_{c0} \leq 40$ Tesla, which induces a metamagnetic transition and restores a bad metal. The valence-change transition in Eu-based intermetallics is similar [7,8], except the transition temperatures and the critical fields are higher ($T_v \geq 100$ K and $H_c \geq 50$ Tesla), and the Eu ions undergo an almost complete valence change from 2^+ (f^7) to 3^+ (f^6) state.

We explain such a behavior of YbInCu₄-like intermetallics by the proximity of μ to the pseudo-gap, which reduces T_K and gives $T^* \gg T_K \propto H_c$. If we take $T_v \propto T_K$, the Kondo lattice model describes the valence-change as a transition from a bad metal with a ‘‘small FS’’ to a good metal with hybridized f-states and ‘‘large FS’’. Above T_v , the Yb ions form a disordered spin-lattice which is decoupled from the conduction band, while below T_v the f-electrons participate in the ‘‘heavy’’ quasiparticle band with a ‘‘large’’ FS. Since T^* is not affected by the pseudo-gap, and $T^* \gg T_v$, all the properties of the system below T_v are nearly temperature-independent. If we estimate the FL corrections to the $T = 0$ value of the magnetic susceptibility or the electric resistivity up to $\mathcal{O}[(T/T^*)^2]$ and assume $T^*/T_K \leq 10$ (as indicated by the data), the maximum relative deviation at $T \sim T_v$

is 1%. The large- \mathcal{N} solution explains the metamagnetic transition at the critical field, $H_c \propto T_v \propto T_K$, and gives the universal ratio $H_c/T_v = \mathcal{O}(1)$ but can not describe the properties of YbInCu₄-like systems above T_v . The proper description of the high-temperature phase might require a Coulomb repulsion between the f-states and the conduction band, i.e. the free electron states of the high-temperature phase should be replaced by the over-damped states of a small-gap Mott-Hubbard insulator. A gap or a pseudo-gap is also required to explain the absence of the Kondo effect in the high-temperature phase, despite the presence of a local moment at each lattice site [21].

The YbAl₃ anomalies are of a different type but also exhibit more than one low-temperature energy scale. The data [9] show that for $T \geq T^* \simeq 40$ K the coherence is lost but the strong coupling features persist up to the highest temperatures measured, i.e., the magnetic moment remains quenched ($T_K \geq 650$ K). The de Haas - van Alphen experiments in the coherent regime show [9] that the high-field dHvA mass is greatly reduced with respect to the low-field mass, and that the critical field is about $H^* \simeq k_B T^*/\mu_B$. However, there is hardly any change in the Fermi surface for $H \leq H^*$. Using the Kondo lattice model close to half-filling and assuming that the chemical potential is close to the maximum of the DOS, we find $T^* \ll T_K$ and $H^* \ll k_B T^*/\mu_B$. Thus, we have $m(H) \simeq H/T^*$ for low fields and $m(H) \propto \text{const}$ for $H \geq T^*$, i.e. the saturation magnetization is approached in the usual Fermi liquid fashion. Since $H^* \simeq T^* \ll T_K$, the order parameter is finite and the system can be viewed as a polarized heavy FL with a “large FS”.

In summary, we studied the low-temperature properties of the Kondo lattice model with a non-constant DOS, using large- \mathcal{N} formalism. Close to the particle-hole symmetry, the ground state is a Fermi-liquid, except for $\rho_0(\mu) = 0$, in which case $T_K = 0$ for $J \leq J_c$, where J_c is a quantum critical point. This generalizes the work of Withoff and Fradkin [5] which found the quantum critical behavior for a Kondo impurity model with a singular DOS. We find that T_K is finite but reduced for a conduction band with only a few states around to μ . Since the FL parameter T^* of the Kondo lattice is much less affected by the pseudo-gap than T_K , the values of T_K and T^* can differ by more than one order of magnitude. If that is the case, the physical properties of the FL phase are nearly constant. We also find that a DOS with a peak close to μ would reverse the relative magnitude of T_K and T^* . Considering the magnetic field effects, we find the critical value $H_c \simeq (k_B/\mu_B)T_K$ which separates the Kondo phase (Fermi liquid with a large Fermi surface and hybridized bands) from the Kondo-free phase (bad metal with a pseudo-gap and a “small FS”, decoupled from the f-spins). The low-field response of the Kondo lattice is linear and proportional to $1/T^*$, while the high

field behavior sets in at about $H_c \propto T_K$ or $H^* \propto T^*$, whichever is smaller. However, the non-linear corrections go in the opposite directions for $T^* \gg T_K$ and $T^* \ll T_K$ systems. In the former case there is a metamagnetic transition at H_c , while in the latter one finds at H^* the usual paramagnetic saturation. These Kondo lattice results explain different behavior and various energy scales observed in a valence fluctuator such as YbAl₃, and in systems with a valence-change transition, such as YbInCu₄, EuNi₂(Si_{1-x}Ge_x)₂ or Eu(Pd_{1-x}Pt_x)₂Si₂.

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