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## The two-channel Kondo route to non-Fermi-liquid metals

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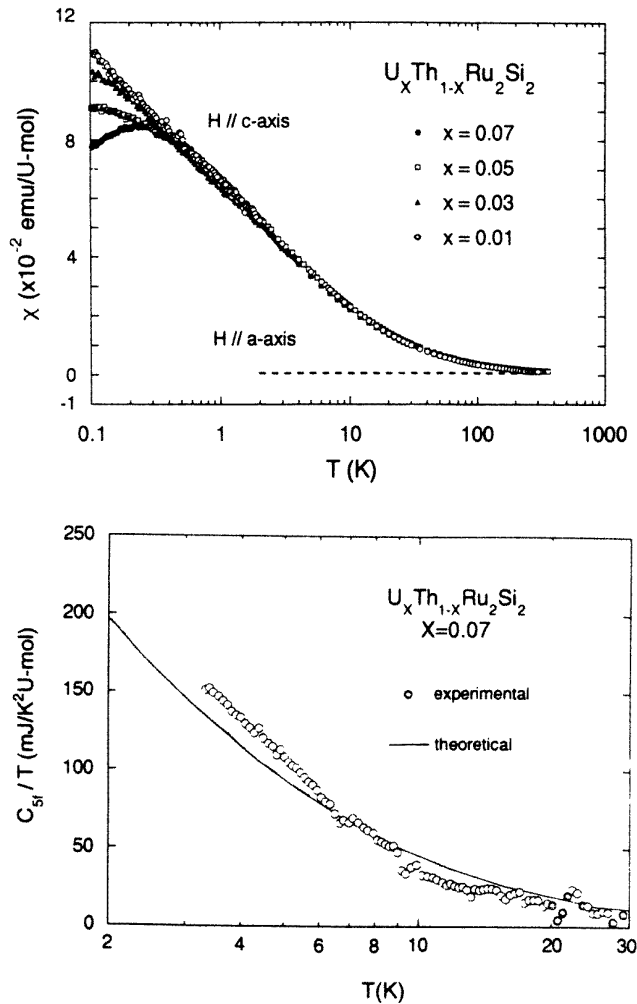
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**Abstract.** We present a pedagogical and critical overview of the two-channel Kondo model and its possible relevance to a number of non-Fermi-liquid alloys and compounds. We survey the properties of the model, how a magnetic two-channel Kondo effect might obtain for  $\text{Ce}^{3+}$  ions in metals, and a quadrupolar Kondo effect for  $\text{U}^{4+}$  ions in metals. We suggest that the incoherent metal behaviour of the two-channel Kondo-lattice model may be useful in understanding the unusual normal-state resistivity of  $\text{UBe}_{13}$  and speculate that the residual resistivity and entropy of the two-channel lattice paramagnetic phase might be removed by either antiferromagnetic (or antiferroquadrupolar) ordering or by a superconducting transition to an *odd-frequency* pairing state.

### 1. Introduction

Since the discovery of heavy-fermion superconductivity in 1979 [1], there have been numerous theoretical efforts to explain this mysterious phenomenon (for reviews see references [2–7]). The name results from a superconducting phase transition in which electrons with effective masses hundreds of times the free-electron value pair. That is to say, the electrons nearly localize (infinite effective mass) and yet at the end of the day at sufficiently low temperatures conduct electricity perfectly! Moreover, the superconductivity is clearly ‘exotic’ in character: there are compelling reasons to believe that the electron pairs are in non-trivial angular momentum states about their centre of mass, unlike the conventional superconductors such as aluminium or lead [8]. The materials which display this physics are rare-earth or actinide intermetallic compounds such as  $\text{CeCu}_2\text{Si}_2$  and  $\text{UBe}_{13}$  which possess tightly bound 4f or 5f electronic orbitals and localized moments on the Ce or U sites as well as ‘light’ conduction states contributed by the surrounding ligands. Because the f orbitals are both strongly correlated—it costs several electron volts to multiply occupy an f site—and possess strong magnetic tendencies which are normally an anathema to electron pairing, this superconductivity is truly remarkable. Indeed, in several materials superconductivity and magnetism *coexist*, unlike in the cuprate superconductors! Clearly, the intense interest in these materials over now nearly two decades of study is richly deserved.

An apparently separate area of research in strongly correlated materials which has attracted many workers is the study of non-Fermi-liquid metals, which arose recently in the context of the high-temperature superconductors [9–11]. To put this in context, the so-called quasiparticle description of Landau’s Fermi-liquid theory [12] has shaped the way we view the ordinary metallic state. In Landau’s phenomenological approach, a 1:1 mapping of non-interacting electron states to interacting electron states is assumed



**Figure 1.** The susceptibility and specific heat of  $\text{Th}_{1-x}\text{U}_x\text{Ru}_2\text{Si}_2$ . These data show clear evidence of log divergence in  $\chi$ ,  $C/T$  for low  $T$ . Single-ion scaling is also evident, above  $\sim 0.5$  K, indicating an impurity origin to the NFL physics. After references [53, 54].

asymptotically close to the Fermi energy. The states can then be described in terms of quasiparticles, or dressed electrons, which have an enhanced effective mass and magnetic moment through their interactions with other quasiparticles in the surrounding medium. Much of the intuition is then built on these coherent quasiparticle states. It is crucial, however, for the description to work, that the energy width of the quasiparticle excitations vanish more rapidly than their energy position as the Fermi level is approached. This ensures the coherence. On general grounds in such a Fermi liquid the quasiparticle decay rate vanishes as  $T^2$  in three dimensions. A test of this is to measure the electrical resistivity which is proportional to the decay rate, roughly speaking. Because all theories of superconductivity and other instabilities (such as spin-density waves) in a metal are based upon a quasiparticle assumption, the discovery of non-Fermi-liquid metals would provide a basis for a possible paradigm shift. More exciting still would be the discovery of

superconductivity in such materials!

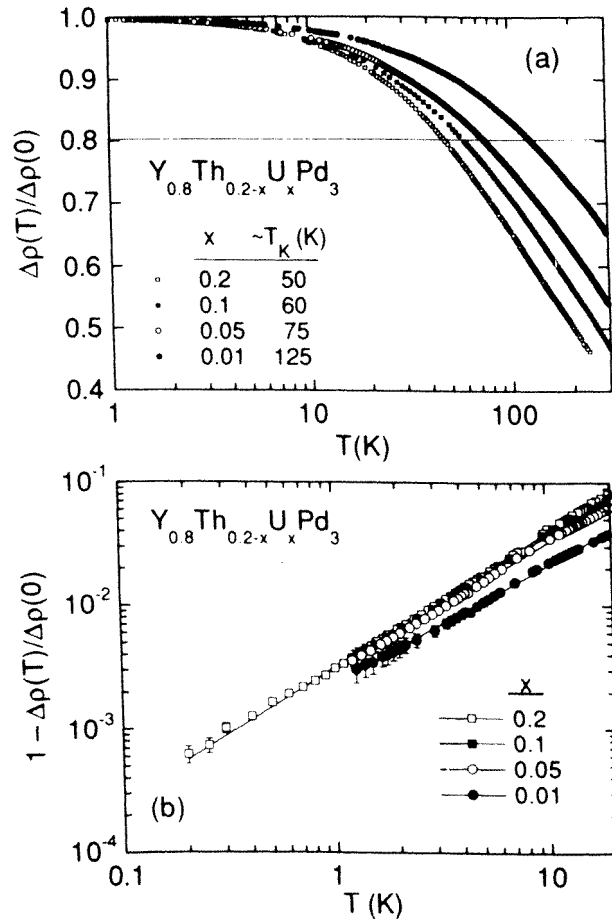
Fortunately, the high-temperature superconductors provide just such a possibility [9]! Anderson noted that in many of the cuprate superconductors (near optimal  $T_c$ -values as it turns out), the quasiparticle decay rate vanishes linearly in the temperature judging from resistivity measurements [10]. A linear-in- $\omega$  low-temperature scattering rate is also observed with optical conductivity measurements, which would not arise, for example, from the high-temperature linear-in- $T$  scattering of phonons in a conventional metal (where it merely represents the increasing population of thermal phonons). These results alone imply a breakdown in the Fermi-liquid picture. Besides Anderson's approach, the 'marginal Fermi-liquid' phenomenology [11] was developed to correlate the disparate anomalous data. In this approach, it is assumed that the spin and charge susceptibilities have an unusual form, in that they are approximately momentum independent and vanish linearly in  $\omega/T$  for frequencies  $\omega \leq k_B T/\hbar$  where  $T$  is the temperature. In contrast, the spin and charge susceptibilities of a Fermi liquid are strongly momentum dependent and have a low-frequency behaviour which becomes independent of temperature as  $T \rightarrow 0$ . This hypothesis gives rise, with certain assumptions, to the linear-in- $T$  scattering rate, concomitant with an effective mass which diverges logarithmically as the Fermi energy is approached.

**Table 1.** Non-Fermi-liquid heavy-fermion alloy systems. This table lists the relevant properties of all non-Fermi-liquid heavy-fermion alloy systems for which a two-channel Kondo model description may be appropriate. The columns for specific heat, susceptibility, and resistivity indicate the low-temperature behaviour. All have logarithmic-in- $T$  specific heat coefficients over an extended temperature range. The coefficients  $A, B$  listed in the resistivity column are assumed positive. The column under 'Single ion?' answers whether single-ion scaling has been observed.

Alloy	$T_K$	$C/T$	$\chi(T)$	$\rho(T)$	Single ion?	References
$Y_{1-x}(\text{Th}_{1-y}, \text{U}_y)_x \text{Pd}_3^{(*)}$	$\sim 40$ K	$\ln T$	$1 - aT^{1/2}$	$1 - AT$	Yes	[13, 42, 45]
$\text{Th}_{1-x}\text{U}_x\text{Ru}_2\text{Si}_2^{(*)}$	12 K	$\ln T$	$\ln T$ ( $\mathbf{H} \parallel c$ )	$1 + BT^{1/2}(?)$	Yes	[53]
$\text{Th}_{1-x}\text{U}_x\text{Pd}_2\text{Si}_2^{(*)}$	12 K	$\ln T$	$\ln T$ ( $\mathbf{H} \parallel c$ )	$1 + BT^{1/2}(?)$	Yes	[54]
$\text{La}_{1-x}\text{Ce}_x\text{Cu}_2\text{Si}_2^{(*)}$	$\sim 10$ K	$\ln T$	$\ln T$	$1 - AT$	Approximate	[55]
$\text{Th}_{1-x}\text{U}_x\text{M}_2\text{Al}_3$	$\sim 20$ K	$\ln T$	$\ln T(?)$	$1 - AT$	?	[13, 58]
$\text{Th}_{1-x}\text{U}_x\text{Be}_{13}$	$\sim 10$ K	$\ln T$	$1 - aT^{1/2}$	$1 + BT^{1/2}$ or $1 + AT$	No	[59, 60, 61]

An exciting development of the past few years is that a number of heavy-fermion alloys have been discovered to have non-Fermi-liquid properties as well [13]. These include the systems  $Y_{1-x}(\text{Th}_{1-y}, \text{U}_y)_x \text{Pd}_3$ ,  $\text{Th}_{1-x}\text{U}_x\text{Ru}_2\text{Si}_2$ ,  $\text{Th}_{1-x}\text{U}_x\text{Pd}_2\text{Si}_2$ ,  $\text{Th}_{1-x}\text{U}_x\text{Be}_{13}$ ,  $\text{La}_{1-x}\text{Ce}_x\text{Cu}_2\text{Si}_2$ ,  $\text{Th}_{1-x}\text{U}_x\text{M}_2\text{Al}_3$  ( $M = \text{Ni}, \text{Pd}$ ),  $\text{UCu}_{5-x}\text{Pd}_x$ , and  $\text{CeCu}_{6-x}\text{Au}_x$ . All of these materials display specific heat coefficients which over an extended range of temperatures appear to behave as  $-\ln(T)$ , and resistivities which have leading low-temperature variation that is often linear in  $T$ , but decidedly non-Fermi-liquid like. The magnetic susceptibility is also non-Fermi-liquid like, often varying as  $1 - AT^{1/2}$ , or logarithmically, at low temperatures. Figure 1 displays the susceptibility and specific heat of  $\text{Th}_{1-x}\text{U}_x\text{Ru}_2\text{Si}_2$ , and figure 2 displays the resistivity of  $Y_{1-x}(\text{Th}_{1-y}, \text{U}_y)_x \text{Pd}_3$ . Properties of these systems are summarized in table 1.

Three main scenarios have been put forward in an effort to understand the physics of these systems.



**Figure 2.** The resistivity of  $Y_{0.8}(Th_{1-y}U_y)_{0.2}Pd_3$ . The resistivity is approximately linear for all concentrations, and the low-temperature behaviour is approximately independent of the concentration. After reference [45].

(1) *Proximity to a quantum critical point.* When a critical point such as that associated with a ferromagnetic or antiferromagnetic transition is tuned to zero temperature by some external parameter  $R$  such as pressure or dopant concentration, this is known as a ‘quantum critical point’. For temperatures directly above the  $T = 0, R_c$  point, non-Fermi-liquid behaviour is expected in thermodynamic and transport properties [14] (NFL phenomenology can also be present near a quantum spin-glass transition—see [15]). Phenomenologically, this seems to be the most relevant scenario for  $CeCu_{6-x}Au_x$ , as is discussed elsewhere in this Special Issue.

(2) *Distribution of Kondo temperatures.* If the material is highly disordered, this can result in a distribution of Kondo temperature scales. Each scale determines the temperature at which Fermi-liquid behaviour will set in around a single magnetic impurity antiferromagnetically coupled to conduction electrons (assuming an effective spin-1/2 impurity magnetic moment). Averaging over such a distribution can produce thermodynamic and transport properties which look non-Fermi-liquid like due to the broad range of effective

Fermi temperatures [16]. Essentially, the unquenched moments contribute the NFL physics. Based upon the experimental observation of a broad distribution of internal fields through NMR on  $\text{UCu}_{5-x}\text{Pd}_x$  ( $x = 1, 1.5$ ), this appears to be a likely scenario to explain the phenomenology of this material.

(3) *The two-channel Kondo model.* Nozières and Blandin introduced the multi-channel Kondo model in 1980 [17, 18]. In this model,  $M$  degenerate channels of conduction electrons couple with identical exchange integrals to a spin- $S_I$  impurity. When  $M > 2S_I$ , a non-trivial non-Fermi-liquid critical point governs the low-temperature properties of the system. It has been argued extensively elsewhere that the most likely observable version of this model for rare-earth and actinide systems is the  $M = 2$ ,  $S_I = 1/2$  model, and there are strong reasons to argue for its relevance to the alloys  $\text{Y}_{1-x}\text{U}_x\text{Pd}_3$ ,  $\text{Th}_{1-x}\text{U}_x\text{Ru}_2\text{Si}_2$ ,  $\text{Th}_{1-x}\text{U}_x\text{Pd}_2\text{Si}_2$ ,  $\text{Th}_{1-x}\text{U}_x\text{Be}_{13}$ ,  $\text{La}_{1-x}\text{Ce}_x\text{Cu}_2\text{Si}_2$ , and  $\text{Th}_{1-x}\text{U}_x\text{M}_2\text{Al}_3$ . The model is also relevant to various mesoscopic devices, one of which shows non-Fermi-liquid transport properties believed to be due to so-called two-level-system Kondo defects in the device [20]. The lattice version of the two-channel Kondo model has been argued to be relevant to the concentrated heavy-fermion system  $\text{UBe}_{13}$  [18], which shows clear NFL behaviour above its superconducting phase transition, as first pointed out in reference [3], and examined more recently in reference [21]. (An alternative approach to locally driven NFL behaviour for both impurities and the lattice appears in the work of Si *et al* [19].)

In this paper, intended as a pedagogical overview rather than a report of new results, we lay out the arguments and evidence for the relevance of scenario (3) above to the non-Fermi-liquid heavy-fermion systems. (A lengthy technical review of the two-channel Kondo effect and its relevance to real materials is in preparation and will be published elsewhere [22].) A brief outline is as follows: in section 2 we briefly review the two-channel Kondo model and describe how this can arise in the context of the quadrupolar Kondo effect for tetravalent uranium ions and the magnetic Kondo effect for trivalent Ce ions. In section 3 we survey the data on existing heavy-fermion alloy systems which support at least a partial interpretation in terms of the two-channel Kondo effect. In section 4, we turn to reviewing an approach to the two-channel Kondo lattice which is based upon taking the limit of infinite spatial dimensions. This gives a paramagnetic phase which is an ‘incoherent metal’, namely, a finite residual resistivity and entropy result despite a finite density of states at the Fermi energy. This result has possible relevance to the heavy-fermion superconductor  $\text{UBe}_{13}$ , which has an anomalously large residual resistivity at  $T_c$  together with strong evidence for ‘cleanliness’. A critical discussion of the oversimplifications in this model will be provided, particularly focusing on the assumption of ‘global’ channel symmetry. Finally, in section 5, we conclude and provide speculation on the possibility of superconductivity in the two-channel Kondo lattice and its possible relation to the heavy-fermion superconductors.

Before proceeding to the discussion of the two-channel Kondo physics, some general comments about non-Fermi-liquid and Fermi-liquid metals are in order. The Fermi-liquid paradigm has been hugely successful, and such noted scientists as P W Anderson have likened it to the Standard Model of particle physics [23]. This comparison not only honours the great success of the Fermi-liquid theory, but evokes depression among many condensed-matter physicists due to the intellectual tyranny exerted by that very success. In such a climate, it is no surprise that many condensed-matter physicists desire to go beyond our ‘standard model’ as eagerly as our particle physics counterparts long to move past theirs.

Enthusiasts for the Fermi-liquid paradigm are quick to point out that since no obvious order parameter characterizes the normal-state physics of high-temperature superconductors or heavy-fermion materials, there is no sharp distinction between a Fermi-liquid normal

state and a non-Fermi-liquid normal state. These can only be separated by a crossover, and therefore the most ardent Fermi-liquid practitioners will proclaim a victory of sorts over upstart advocates of non-Fermi-liquid physics. Indeed, a common point of view for  $\text{UBe}_{13}$  (and the cuprates) is that superconductivity merely intervenes before the onset of a coherent Fermi-liquid state. In the case of  $\text{UBe}_{13}$ , application of magnetic fields which completely suppress the superconductivity reveal no tendency to Fermi-liquid behaviour down to approximately 200 mK (the specific heat behaves approximately logarithmically with temperature in a 12 T field from 200 mK to 3 K [21]). While one cannot rule out Fermi-liquid behaviour setting in at some yet lower temperature, the data beg the question ‘even if a Fermi-liquid state eventually sets in below say, 100 mK, once we apply magnetic field, of what relevance is this to a superconducting transition at 0.9 K in zero field?’

It seems to us that such debates obscure a point of commonality between Fermi-liquid physics and non-Fermi-liquid physics that is best described in renormalization group language: the physically relevant meaning of a Fermi-liquid or non-Fermi-liquid normal state is that of an unstable fixed point which governs the physics of the system in an intermediate-temperature regime prior to the onset of an ordered ground state (superconducting, magnetic, etc) [24]. Such fixed points are characterized by their excitation spectra, and their conceptual power is augmented by quantitative utility: these excitations can be fed into calculations of transport properties or superconducting gap equations, for example, in the case of the Fermi liquid where electronic quasiparticles describe the spectrum. In the case of the Fermi-liquid fixed point, some forty years of experience plus recent explicit renormalization group treatments [24] have driven home the existence and usefulness of this particular *unstable* fixed point. However, even for the most apparently stable Fermi-liquid metal (e.g., copper) there is ample theoretical reason to believe that some ordering will prevail at low temperatures (e.g., the Kohn–Luttinger instability). Examined in this light, the above-mentioned view that ‘nuisance’ superconductivity obscures the Fermi-liquid fixed point in  $\text{UBe}_{13}$  or the cuprates is decidedly a case of putting the cart before the horse.

In the study of non-Fermi-liquid metals, which is a much younger sub-field of physics than that of conventional metals, we are still working to see whether such an unstable fixed point exists, with excitations that are *not* specified by electronic quasiparticles. Such new excitations could give rise to very novel superconducting or magnetic states. It seems clear in the case of the cuprate superconductors and such heavy-fermion materials as  $\text{UBe}_{13}$  and  $\text{CeCu}_2\text{Si}_2$  that the interesting physics above and below any phase transition happens in a region far from any Fermi-liquid fixed point. Thus, the data issue forth a challenge to theorists to provide descriptions of a conceptual and quantitatively useful new unstable fixed point (fixed points?) governing the normal state of the system, which matches the compelling elegance and falsifiability of Landau’s brilliant theory. It is in the spirit of attempting to answer that challenge that this manuscript proceeds—though honestly, for any of the above materials, we do not have such a complete and elegant new theory yet.

## 2. The two-channel Kondo model: non-trivial physics and the origin in real materials

### 2.1. One- and two-channel Kondo models

Before describing the two-channel Kondo model, it is helpful to briefly describe the physics of the ordinary one-channel Kondo model. This model assumes the presence of an  $S_I = 1/2$

impurity local moment in a metal with the Hamiltonian

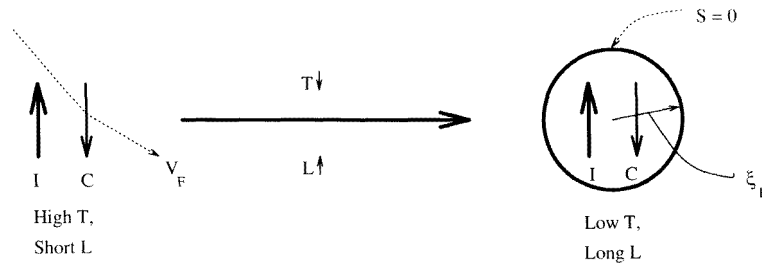
$$H = \sum_{k,\sigma} \epsilon_k n_{k,\sigma} + \mathcal{J} \mathbf{S}_I \cdot \mathbf{s}_c(0) \tag{1}$$

where the first term corresponds to the conduction electrons with energies  $\epsilon_k$  and occupancies  $n_{k,\sigma}$ ,  $\sigma$  is the spin, and the second term describes the exchange coupling between the spin-1/2 local moment ( $\mathbf{S}_I$ ) and the conduction electron spin amplitude at the impurity site  $\mathbf{s}_c(0)$ .

The Kondo effect refers to the formation of a many-body resonance about a local moment in a metal when the exchange coupling between local and conduction spins  $\mathcal{J}$  is antiferromagnetic. By using third-order perturbation theory for the conduction electron  $t$ -matrix which describes scattering off of the local moment, Jun Kondo showed in 1964 [25] that the scattering strength actually grows logarithmically as the temperature is lowered. This perturbation theory approach breaks down when successive terms in the logarithmically divergent expansion grow to the same size, which happens at a temperature scale  $T_K$  given by

$$k_B T_K \approx E_F \left( \frac{\mathcal{J}}{E_F} \right)^{1/2} \exp(-E_F/\mathcal{J}) \tag{2}$$

where  $E_F$  is the conduction electron Fermi energy. Kondo’s calculation actually foreshadowed the discovery of asymptotic freedom in quantum chromodynamics and has the same feature that systematic perturbation theory works well at high energy scales but fails at low energy scales.



**Figure 3.** A renormalization group cartoon for the single-channel Kondo model. At high temperatures  $T$  and short length scales  $L$ , the local moment labelled  $I$  is weakly aligned antiparallel to the conduction electron spin (labelled  $C$ ). At low temperatures and long length scales  $L$ , it is bound into a singlet lump of size  $\xi_K \simeq \hbar v_F/k_B T_K$  as the effective coupling strength diverges.

In the intervening decades since Kondo’s discovery, we have achieved a complete understanding of the static and dynamical properties of the simple model of equation (1). A beautiful physical picture emerged from K G Wilson’s numerical renormalization group (NRG) treatment of the problem, for which a cartoon picture is depicted in figure 3 [26]. Essentially, the NRG calculation integrates out high-energy, low-length-scale excitations to determine the effective Hamiltonian at each temperature scale. A unique feature of the Kondo Hamiltonian is that it is ‘renormalizable’—that is, the form does not change—and no new couplings are introduced. You simply must specify the effective exchange coupling at each temperature. This makes the model much like high-energy-physics theories which are always renormalizable (in fact it is generally a criterion for a theory there!) Wilson’s work showed that a many-body resonance is indeed formed—in effect, the exchange coupling



grows to infinite strength as the temperature is lowered. Thus the high-temperature local moment is wiped out at low temperatures as a many-body singlet state captures one unit of conduction spin. The size of this lump is of order  $\hbar v_F/k_B T_K = \xi_K$ , the so-called Kondo coherence length.

It must be noted that this solution (and other approaches, such as conformal field theory and the Bethe *ansatz* method) rest upon a mapping to an effective one-dimensional problem. This mapping follows from the fact that only the s-symmetry partial waves of plane-wave states couple to the impurity in equation (1); hence, the effective spatial dimension is radial.

This singlet ‘lump’ is polarizable at any finite temperature or energy, and may be excited to a triplet state. The virtual polarizations of the singlet mediate an effective electron–electron interaction for the states outside, which are then described by a local version of Landau’s Fermi-liquid theory, as explained elegantly by P Nozières [27]. The effective Fermi temperature is set by the Kondo scale, which may be exponentially small compared to the Fermi temperature of the conduction band alone. The extra specific heat coefficient and extra magnetic susceptibility per impurity are then proportional to  $1/T_K$ . In this light, the origin of heavy fermions is no mystery: the local moments are provided by partially filled 4f- or 5f-electron shells, and the Kondo effect gives a small effective Fermi temperature  $T_F^* \sim T_K \sim 1/m^*$ .

In addition, the Fermi-liquid behaviour shows up in the electrical resistivity. The low-temperature value is finite since the impurity breaks translational invariance and thus always disrupts the current; the value reflects a fully resonant  $\pi/2$  phase shift for scattering off the ‘lump’. Indeed, the low-lying excitations are in a 1:1 map with those of a  $\pi/2$  phase-shifted one-dimensional free-electron gas, confirming the central *ansatz* of the Landau phenomenology microscopically here. As  $T$  is raised from zero, the resistivity diminishes with a  $T^2$ -coefficient, the sign change relative to a periodic system reflecting the impurity scattering. Apart from the trivial fall-off of the scattering resonance on moving away from the Fermi level, this  $T^2$ -coefficient does indeed reflect the quasiparticle relaxation in the local Fermi liquid.

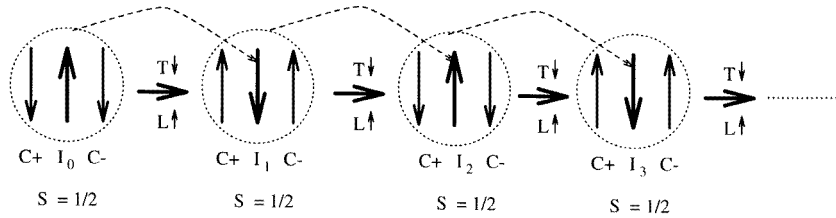
A simple modification to this single-channel Kondo model produces non-Fermi-liquid behaviour. That of course is the two-channel Kondo model, introduced by Nozières and Blandin [17] and given by

$$H = \sum_{k,\sigma,\alpha} \epsilon_k n_{k,\sigma,\alpha} + \mathcal{J} S_I \cdot \sum_{\alpha} s_{\alpha}(0) \quad (3)$$

where  $\alpha = \pm$  is the channel index. Essentially, we have added an extra copy of the conduction band in equation (1) to the problem. Putting aside the question of whether this is physical for the moment, we can view the solution to the model as an interesting theoretical issue in its own right.

Nozières and Blandin [17] argued that there must be a non-trivial ground state in this model, something more interesting than a singlet ‘lump’. The technical details of their argument rest upon a perturbative renormalization group theory regulated by an expansion in the inverse of the number of channels. However, the physical content may again be made clear by a numerical renormalization group cartoon (see figure 4). We begin by considering high temperatures, looking at length scales of the order of one lattice spacing about the impurity spin. As the temperature is lowered the effective coupling grows, so at the next length scale, the two channels of conduction spin and impurity bind to form a new effective spin 1/2 which serves as an impurity to conduction states outside that length scale.

How do these conduction states couple? By a superexchange process, an electron outside the first length scale may tunnel in, exchanging spin with an electron inside (that tunnels

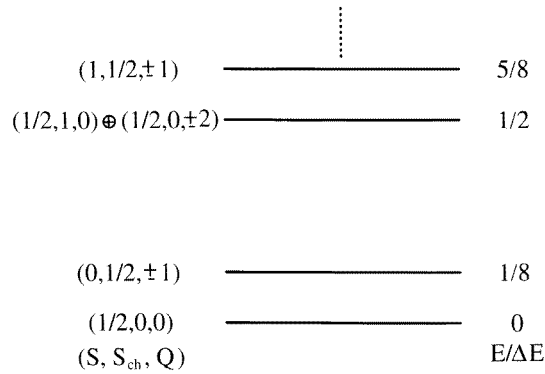


**Figure 4.** A renormalization group cartoon for the two-channel Kondo model. At high temperatures and short length scales  $L$ , the local moment ( $I$ ) is weakly aligned antiparallel to the two conduction electron channels ( $C_{\pm}$ ) of spin. However, the binding process of figure 3 leads to another doublet which is antiferromagnetically coupled to the two channels of conduction spin outside that length scale. Eventually, this process continues till a fixed-point finite coupling strength is attained.

out) provided that the inner electron has opposing spin. But the spin of the electrons within that length scale determine the overall spin of the bound impurity/conduction complex, and hence the net coupling is antiferromagnetic. Thus the picture at the new length scale is the same as that of the original. If you lower the temperature again, you get the same result. If you lower the temperature again, you get the same result again. As long as the effective exchange coupling  $\mathcal{J}(T)$  tunes to some particular value (which it does) we get a true critical state as  $T \rightarrow 0$ : as in iron at its Curie temperature, the physics appears the same at all length scales at  $T = 0$ ! The effective size of the critical state grows as  $1/T$  for  $T \rightarrow 0$ . Also, unlike the ordinary Kondo problem, the degeneracy of the impurity spin is never lifted. At each length scale, a bound spin-1/2 complex sits at the centre and plays the role of the effective impurity.

Given the development of a non-trivial critical state, it is perhaps no surprise that the physical properties of the electronic states surrounding this impurity are not of a Fermi-liquid character. Indeed, from exact treatments based upon the Bethe *ansatz* method [28] and conformal field theory [29], it is clear that the extra specific heat coefficient  $\gamma$  and spin susceptibility per impurity diverge as  $\ln(T_K/T)/T_K$  as the temperature tends to zero. That is, the effective mass tends to infinity at zero temperature! Also, the resistivity saturates to its zero-temperature limit not with a  $T^2$ -law, but with a  $\sqrt{T}$ -law [30]. It should be noted that the square-root law shows only below about  $0.05T_K$  [31], while the  $\ln(T)$  divergence in the specific heat coefficient and susceptibility appears below about  $0.5T_K$ . For a range of intermediate temperatures  $0.05T_K \leq T \leq T_K$  the resistivity is approximately linear in temperature. Finally, the residual degeneracy shows itself in a net residual entropy per impurity of  $(k_B/2)\ln 2$ , that is a kind of ‘fractional degeneracy’!

In the impurity limit, a crossover to Fermi-liquid behaviour can be induced in two ways by application of external fields [17, 32–34]. When a ‘spin field’  $H_s$  is applied which couples linearly to the spin of the impurity and conduction electrons, the system will cross over to the fixed point of a Fermi liquid in the presence of a polarized scatterer [33]. The crossover scale will be  $T_s = H_s^2/T_K$  [32]. In an applied ‘channel field’ which linearly splits the spin of the conduction electrons *and* more importantly the degeneracy of the exchange integrals, the system will crossover to the ordinary Kondo effect for the more strongly coupled channel and the other channel will decouple. The crossover scale here is  $T_{ch} = \Delta\mathcal{J}^2/T_K$  where  $\Delta\mathcal{J}$  is the splitting of the exchange integrals [33]. Concomitant with these crossovers will be a shoving out of the residual entropy into a Schottky-like peak in  $C/T$  [32, 34]. Physical properties will be universal functions of  $T/T_{\alpha}$  with  $\alpha = s, ch$ .



**Figure 5.** The finite-size spectrum of the two-channel Kondo model. Here  $S$  denotes spin,  $S_{ch}$  channel spin, and  $Q$  charge. Energies are measured in units of  $\Delta E = \hbar\pi v_F/L$ , where  $L$  is the system size. The low-lying splittings and quantum numbers are not that of a Fermi liquid (after reference [33]); for example, the charge- $\pm$  excitation has zero spin independent of boundary conditions, and the level spacings are fractions of  $\Delta E$ .

The excitation spectrum of the two-channel model is decidedly not that of a Fermi liquid; this point is illustrated in figure 5, where finite-size spectra for one-dimensional chains of radius  $L$  and hard-wall boundary conditions are illustrated for a two-channel Fermi gas (the s-wave channel) and for the corresponding two-channel-model fixed point. Such spectra are obtained by conformal field theory or numerical renormalization group calculations [29, 33, 35]. The most striking features are: (i) a fractional spacing of the two-channel model energy levels relative to that of the free-electron gas; (ii) a ‘separation’ of spin, channel, and charge quantum numbers. In the free Fermi case, addition of an electron must correspond to addition of charge 1, spin 1/2, and channel spin 1/2. This is obviously not the case for the two-channel model. There is no 1:1 correspondence of excitations to a free Fermi gas.

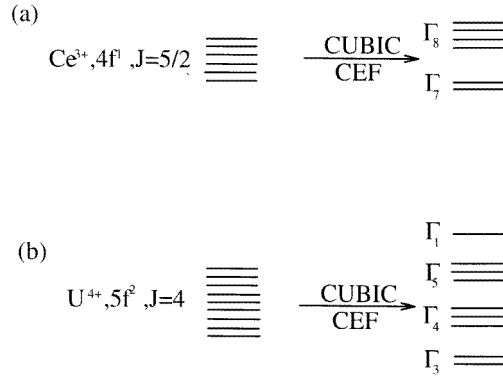
## 2.2. Possible realizations of two-channel Kondo physics for heavy-fermion materials

This section contains a pedagogical overview of the results of references [18, 36, 37].

The two-channel Kondo effect requires two key ingredients: (i) the existence of local two-level systems (which may or may not be magnetic in practice) providing an impurity *pseudo*-spin 1/2 ( $S_j$ ); (ii) the existence of local symmetries affording a twofold degeneracy (apart from magnetic or Kramers’ degeneracy) to conduction electron states which couple to the impurity. Practically, this means that there must be a symmetry-allowed local quartet of conduction states to couple to the impurity.

The non-Fermi-liquid physics in heavy-fermion systems is exclusively associated for now with Ce- and U-based materials. Crystal-field physics tells us how we may obtain local two-level systems for these ions, as illustrated in figure 6. Consider first a Ce ion embedded in a host of cubic symmetry. In a metal, Ce tends to be trivalent; that is, the dominant ground-state quantum weight occurs for states containing the  $4f^1$  configuration.

In free space, the  $\text{Ce}^{3+}$  ion would have, according to Hund’s rules, a ground-state angular momentum  $J = 5/2$  which transforms according to the matrices of the corresponding irreducible representation (or irrep) of the full rotation group. In the 24-element point group of the cubic host, this sixfold degeneracy cannot be maintained, and the  $J = 5/2$  multiplet



**Figure 6.** The crystal-field splitting of  $\text{Ce}^{3+}$  and  $\text{U}^{4+}$  ions in cubic symmetry. (a) illustrates the lifting of the sixfold degeneracy of the  $4f^1$   $J = 5/2$  Hund's rules ground multiplet of the  $\text{Ce}^{3+}$  ion to yield a magnetic doublet ground state and quartet excited state. (b) illustrates the lifting of the ninefold-degenerate  $5f^2$   $J = 4$  Hund's rules ground multiplet of the  $\text{U}^{4+}$  ion to yield a ground quadrupolar doublet, two excited magnetic triplets, and an excited singlet. In the  $\text{Ce}^{3+}$  case, the doublet lies lowest over half the one-parameter crystal-field space, and in the  $\text{U}^{4+}$  case, the doublet lies lowest over nearly half of the two-parameter space [39].

splits into a doublet (usually labelled  $\Gamma_7$ ) and a quartet (labelled  $\Gamma_8$ ). This splitting is said to arise from the 'crystalline electric field', i.e., the external potential of cubic symmetry arising from the presence of the surrounding ions. In practice, however, hybridization with surrounding (ligand) electronic orbitals is likely to produce a substantial amount of the splitting through a second-order perturbation theory shift. If the doublet lies lowest (which holds, trivially, over half of the one-parameter crystal-field parameter space), then an effective *magnetic* pseudo-spin 1/2 is obtained to describe the low-lying degrees of freedom of the  $\text{Ce}^{3+}$  ion. The magnetic character follows from Kramers' theorem, which asserts that odd-electron configurations must have an extra degeneracy due to time-reversal symmetry. Practically, this means that the degeneracy is lifted and the doublet split linearly under application of a magnetic field  $\mathbf{h}$ .

In the case of U ions, controversy arises over whether they are predominantly trivalent or tetravalent, or whether a localized point of view is useful at all [38]. Here we assume the utility of a localized point of view, and further assume that U ions in metals are predominantly tetravalent. A  $\text{U}^{4+}$  ion has a  $5f^2$  configuration which by Hund's rules gives a ground-state angular momentum  $J = 4$ . When embedded into a host of cubic symmetry, the  $J = 4$  multiplet will be split into a doublet (labelled  $\Gamma_3$ ), two magnetic triplets (labelled  $\Gamma_4$  and  $\Gamma_5$ ), and a singlet ( $\Gamma_1$ ). Straightforward analysis of the two-parameter crystal-field Hamiltonian shows that over about half of the parameter range the  $\Gamma_3$  doublet lies lowest in energy [39]. In contrast to the Kramers' doublet of the preceding paragraph, however, this doublet is *not* split linearly in an applied magnetic field. The magnetic moment operator has vanishing expectation value within the doublet. Instead, the electric quadrupole tensor has a non-vanishing expectation value. The  $zz$ -component of this operator is given by matrix elements:

$$Q_{zz} = e \int d\mathbf{r} \rho_U(\mathbf{r}) [3z^2 - r^2] \quad (4)$$

and the component  $Q_{zz} \sim 3J_z^2 - J(J+1)$  has the expectation value  $\pm 8$  in this  $\Gamma_3$  manifold. (The proportionality of  $Q_{zz}$  to  $3J_z^2 - J(J+1)$  follows from the Wigner-Eckart theorem.)

Note that if this state lies lowest the magnetic susceptibility arises from Van Vleck processes, i.e., virtual excitations to the excited magnetic triplets.

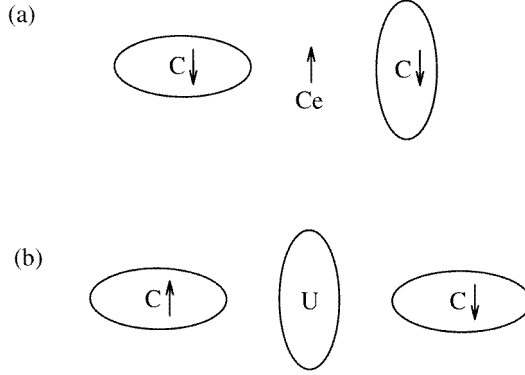
Next, we turn to the existence of the quartet of conduction states. In this cubic case, such local quartets will indeed exist. To conceptualize this, imagine hybridizing plane-wave conduction states with the f orbitals of the  $\text{Ce}^{3+}$  or  $\text{U}^{4+}$  ion. As a first approximation, we use the spherically averaged potential, so that only  $l = 3$  partial waves of the conduction states couple to the f states. These  $l = 3$  partial waves must further be spin-orbit coupled into  $j = 5/2$ ,  $j = 7/2$  partial waves. For simplicity, consider only the  $j = 5/2$  manifold (which may be shown to have the dominant coupling to the  $\text{Ce}^{3+}$  or  $\text{U}^{4+}$  ions). Under the crystal field, this is split into a doublet ( $\Gamma_7$ ) and quartet ( $\Gamma_8$ ) of partial waves. The quartet has the property that it decomposes into the tensor product  $\Gamma_3 \otimes \Gamma_7$ ; that is, it is described by a combination of ‘orbital’ ( $\Gamma_3$ ) and ‘spin’ ( $\Gamma_7$ ) indices. The orbital indices correspond loosely to ‘stretched’ and ‘squashed’ conduction orbitals about the f site.

To complete the logic, we must consider coupling of the conduction states to the impurity. This involves considering tensor operators of impurity and conduction states. In the  $\text{Ce}^{3+}$  case, the tensor product  $\Gamma_7 \otimes \Gamma_7 = \Gamma_1 \oplus \Gamma_4$ , where the operator transforming as a  $\Gamma_1$  irrep is just the electrical charge operator, and the  $\Gamma_4 \sim \mathcal{S}_I$  triplet of operators is the effective spin of the  $\text{Ce}^{3+}$  ground doublet. The  $\Gamma_1$  charge operator gives rise to uninteresting potential scattering terms. The conduction  $j = 5/2$ ,  $\Gamma_7$  doublet clearly also has  $\Gamma_4 \sim \mathcal{S}_{c7}(0)$  tensors, where  $\mathcal{S}_{c7}(0)$  represents the net  $\Gamma_7$  symmetry conduction spin at the origin. The quartet state has the tensor spectrum  $\Gamma_8 \otimes \Gamma_8 = \Gamma_1 \oplus \Gamma_2 \oplus \Gamma_3 \oplus 2\Gamma_4 \oplus 2\Gamma_5$ . The two sets of  $\Gamma_4$  triplet operators can couple to the impurity spin. One of the  $\Gamma_4$  triplets may be viewed as a direct sum of two spin-1/2 operators  $\mathcal{S}_{c8\alpha}(0)$  labelled by the orbital ( $\Gamma_3$ ) index  $\alpha$ ; the other may be viewed as a triplet of spin-3/2 operators ( $\mathcal{S}$ ). We shall discuss the physical origin of these couplings below. Practically, if we for the moment neglect this second  $\Gamma_4$  triplet, we see that the spin-dependent, symmetry-allowed coupling from the operators discussed so far is

$$H_{\text{Kondo}} = \mathcal{J}_7 \mathcal{S}_I \cdot \mathcal{S}_{c7}(0) + \mathcal{J}_8 \mathcal{S}_I \cdot \sum_{\alpha} \mathcal{S}_{c8\alpha}(0) \quad (5)$$

which has a one-channel coupling ( $\mathcal{J}_7$ ) and a two-channel coupling ( $\mathcal{J}_8$ ). These couplings compete, and the Fermi-liquid one-channel fixed point wins as  $T \rightarrow 0$  if  $\mathcal{J}_7 > \mathcal{J}_8 > 0$ , a non-Fermi-liquid two-channel fixed point wins as  $T \rightarrow 0$  for  $\mathcal{J}_8 > \mathcal{J}_7 > 0$ , and a non-Fermi-liquid three-channel fixed point wins if  $\mathcal{J}_7 = \mathcal{J}_8 > 0$  [36]. This may be thought of as a ‘dynamical’ selection rule on the feasibility of the two-channel Kondo effect for  $\text{Ce}^{3+}$  ions. Physically, the two-channel coupling means that the  $\Gamma_8$  electrons may screen the  $\text{Ce}^{3+}$  magnetic moment equally well in degenerate ‘stretched’ or ‘squashed’ orbitals. We illustrate the two-channel screening picture in figure 7(a) for Ce ions.

For the  $\text{U}^{4+}$  ion, the product  $\Gamma_3 \otimes \Gamma_3 = \Gamma_1 \oplus \Gamma_2 \oplus \Gamma_3$ , where the  $\Gamma_1$  operator is again just the  $\text{U}^{4+}$  charge, and gives rise to potential scattering, while the  $\Gamma_2 \oplus \Gamma_3$  operators describe the pseudo-spin of the  $\Gamma_3$  doublet. Note that an intrinsic anisotropy exists in this case: physically, the  $\Gamma_2$  operator is a magnetic octupole moment (transforming as  $J_x J_y J_z$ ) and the  $\Gamma_3$  doublet corresponds to the electric quadrupole operator doublet  $\sim [J_x^2 - J_y^2, 3J_z^2 - J(J+1)]$ . It turns out that this anisotropy is irrelevant in a renormalization group sense. We thus denote  $\Gamma_2 \oplus \Gamma_3$  as practical pseudo-spin triplet  $\tau_I$ , where the  $\tau_I^i$  are spin-1/2 matrices in the orbital space of  $\Gamma_3$  states. This pseudo-spin triplet couples to the  $\Gamma_2 \oplus \Gamma_3$  pseudo-spin triplet from the  $\Gamma_8 \otimes \Gamma_8$  tensor spectrum, which may be viewed as a direct product of two pseudo-spin-1/2 operators labelled by *magnetic index*  $\mu$  (corresponding to the  $\Gamma_7$  labels in the  $\Gamma_3 \otimes \Gamma_7 = \Gamma_8$ ), namely  $\tau_{c8\mu}(0)$ . Hence, the effective Kondo coupling



**Figure 7.** The two-channel screening of  $\text{Ce}^{3+}$  and  $\text{U}^{4+}$  ground doublets. (a) shows the  $\text{Ce}^{3+}$  case in which electrons in stretched and squashed orbitals screen the  $\text{Ce}^{3+}$  spin; these orbitals are degenerate in cubic symmetry, and the orbital index serves as the channel index. (b) shows the  $\text{U}^{4+}$  case in which electrons with up or down spin sit in squashed orbitals to screen the stretched orbital of the  $\text{U}^{4+}$  site. In this case the channel degree of freedom is the magnetic spin, and the orbital motion of the electrons produces the screening of the  $\text{U}^{4+}$  orbital fluctuations.

is of the two-channel form, given by

$$H_{Kondo} = \mathcal{J}_Q \tau_I \cdot \sum_{\mu} \tau_{c8\mu}(0). \quad (6)$$

Physically, the two-channel form means that conduction electron orbital motion can screen the  $\text{U}^{4+}$  quadrupole moment equally well for magnetic spin-up and magnetic spin-down electrons. We illustrate this in figure 7(b).

The fundamental origin of these antiferromagnetic couplings is through the Schrieffer–Wolff transformation [40] applied to the Anderson model, in which conduction– $f$  coupling only arises through hybridization. This transformation eliminates virtual charge fluctuations to excited configurations through second order in the hybridization matrix element  $V$ . The coupling  $\mathcal{J}_7$  is dominated by virtual  $f^0$ – $f^1$  fluctuations, and goes as  $V^2/(E(f^0) - E(f^1))$ , where  $E(f^n)$  is the energy of the locally charge neutral state containing the  $f$  configuration  $f^n$ . The coupling  $\mathcal{J}_8$  is dominated by virtual fluctuations to the nine  $\Gamma_3$  doublets of the  $f^2$  configuration, and is approximately  $V^2/(E(f^2) - E(f^1))$ . The coupling  $\mathcal{J}_Q$  is dominated virtual fluctuations from the  $f^2$  ground  $\Gamma_3$  doublet to excited magnetic doublets in either  $f^1$  or  $f^3$ . Note in the  $\text{Ce}^{3+}$  case that  $E(f^2) - E(f^1) \sim U_{ff} - [E(f^0) - E(f^1)]$ , where  $U$  is the Coulomb repulsion (of order 6 eV). Although  $E(f^0) - E(f^1) \approx 2$  eV, it is still possible for the two-channel coupling to exceed the one-channel coupling in principle due to the large number of intermediate  $f^2$  states.

We close this subsection with some remarks about generalizing the above models for  $\text{Ce}^{3+}$  and  $\text{U}^{4+}$  ions.

(1) *Additional couplings.* In addition to the couplings of equations (5) and (6), additional interactions are possible [37]. First, mixed couplings scattering between  $\Gamma_7$  and  $\Gamma_8$  conduction states are possible for the  $\text{U}^{4+}$   $\Gamma_3$  case; these are irrelevant in a renormalization group sense. For the  $\text{Ce}^{3+}$  ion, as alluded to above, a trio of effective spin-3/2 operators may couple to the impurity spin [37, 41]. This interaction is mediated by virtual excitations to  $f^2$ -triplet  $\Gamma_4, \Gamma_5$  triplet states, and gives rise to a pair of new fixed points if added to equation (5), both of which are non-Fermi liquid in character. With the inclusion of

couplings that mix  $\Gamma_6, \Gamma_8$  states, the only stable fixed points may be shown to be single-channel, two-channel, and three-channel ones (remarkably, this latter fixed point *is* stable in that it has a stability region in coupling space of finite measure [37]).

(2) *Additional partial waves.* In principle, all conduction partial waves of appropriate crystalline symmetry may couple to the impurity. In practice, only a ‘bonding’ combination of these partial waves will possess a relevant coupling; all non-bonding combinations have irrelevant couplings [37].

(3) *Other crystal symmetries.* The above arguments may be generalized to any  $U^{4+}$  doublets in hexagonal or tetragonal symmetry, and to the  $\Gamma_9(S_z = \pm 3/2)$  magnetic doublet of  $Ce^{3+}$  ions in hexagonal symmetry. A slight trickiness for these cases is that the conduction quartet is a direct sum of two doublets. However, the crucial point in each instance is that *flipping* the impurity ‘spin’ requires a transition between conduction doublets. In these instances, the  $U^{4+}$  ion will have a  $c$ -axis magnetic moment, and thus can couple to the  $c$ -axis channel spin. However, there is no transverse coupling so the interaction is practically irrelevant. Note that the operators flipping the pseudo-spin must be quadrupolar in character. Hence, the quadrupolar Kondo effect is rather robust, in that for any ground doublet in cubic, hexagonal, or tetragonal symmetry the two-channel Kondo model applies. However, the magnetic two-channel effect for  $Ce^{3+}$  ions is highly restricted by symmetry, and further by the ‘dynamic’ selection rule mentioned above [18].

(4) *Spin and channel fields.* For the quadrupolar Kondo effect in cubic symmetry, the ‘spin’ field will be a uniaxial stress or applied electric field gradient, and the ‘channel’ field will be magnetic. One subtlety exists here: at higher fields, the induced magnetostriction will act as a uniaxial stress and lift the degeneracy of the ground non-Kramers doublet. For simplicity, assume a lowest  $f^3$  excited configuration. The crossover between the channel and spin field for applied magnetic field at a field  $H^*$  can be estimated by equating the exchange splitting  $\Delta J(H) \approx \mu_B H V^2 / [E(f^2) - E(f^3)]^2$  with the magnetostriction-induced splitting of the quadrupolar doublet given by  $\Delta E_{vv} = \chi_{vv} H^2$  where  $\chi_{vv}$  is the Van Vleck susceptibility. This gives  $H^* \approx \mu_B V^2 / \{ [E(f^2) - E(f^3)]^2 \chi_{vv} \}$ . The splitting of the exchange integrals is induced in this case by applying the Schrieffer–Wolff transformation to the Zeeman-split excited magnetic configuration. These labels will be reversed for the magnetic two-channel Kondo effect in cubic symmetry. In hexagonal and tetragonal symmetry, a  $c$ -axis magnetic field will act as a spin field in all cases, while in-plane uniaxial stresses or electric field gradients will act as spin fields for the quadrupolar Kondo case, and in-plane magnetic fields will act as channel fields for the quadrupolar Kondo case. The lone  $Ce^{3+}$  two-channel Kondo possibility has ‘in-plane’ spin operators which are actually *octupolar*! Hence, in-plane uniaxial stresses or electric field gradients will act as channel fields, and suitable in-plane combinations of magnetic field with uniaxial stress will act as spin fields.

### 3. Experimental support for the two-channel Kondo effect in heavy-fermion materials

The early days of the single-channel Kondo effect produced a wealth of data, and frightened away many workers from studying dilute alloys who believed the field to be thoroughly mined out. Recently, the potential for non-Fermi-liquid behaviour from a dilute alloy has motivated a surging re-examination of alloy systems, particularly those based upon U ions. As we shall see, these studies provide some support for the two-channel Kondo physics argued as possible in the previous section, as well as a number of data which disagree with at least the simplest version of the model.

*$Y_{1-x}U_xPd_3$  and related alloys.* The key discovery in this direction was that the cubic system  $Y_{1-x}U_xPd_3$  displays non-Fermi-liquid behaviour in a concentration range from

$0.0 < x < 0.2$  [42]. This discovery helped to ignite the study of NFL physics in heavy-fermion systems. The NFL physics was demonstrated in the specific heat coefficient, which shows a logarithmic temperature dependence over a decade and a half of temperature for  $x = 0.2$ . Also, the resistivity shows a nearly linear temperature dependence over a wide range, and the magnetic susceptibility displays a  $\sqrt{T}$  temperature dependence. Moreover, integration of the specific heat for  $x = 0.2$  above 0.5 K reveals an entropy plateau near  $R/2 \ln 2$  per U ion, strongly suggesting that  $R/2 \ln 2$  entropy is present below the lowest temperatures measured, supported further by the observation of a total of  $R \ln 2$  entropy in the same temperature range for higher  $x$ -values. The uranium ion is clearly tetravalent in this compound on the basis of the discovery of a phenomenon known as ‘Fermi-level tuning’ [43]: as the uranium is substituted for the trivalent Y ions, the f level moves further away from the Fermi surface as measured in photoemission. This corresponds beautifully to donation of one extra electron per U ion. This produces something of a complication in the understanding of the data since the Kondo scale then depends crucially on the concentration. By holding the supply of tetravalent donors fixed through substituting inert Th ions for U ions, it is possible to go to lower concentrations with essentially the same Kondo scale. In this way, it has been determined that the non-Fermi-liquid physics is a single-ion effect.

Given the cubic site symmetry, the tetravalence of the U ions, the clear non-Fermi-liquid behaviour, the apparent two-channel Kondo character of the specific heat coefficient, and the differing temperature behaviour of the magnetic susceptibility and specific heat coefficient, an identification of this material as a quadrupolar Kondo system was made. This identification remains controversial, but received additional experimental support through two kinds of study: (i) unpolarized neutron scattering which showed two inelastic lines possessing the right intensity to be described by excited magnetic triplets above a non-magnetic  $\Gamma_3$  doublet [44]; (ii) alloying experiments on, e.g.,  $\text{La}_{1-x}\text{U}_x\text{Pd}_3$  [45]. The La ion, also trivalent, is considerably larger than the Y ions, so the expanded lattice is expected to have lower hybridization and a much smaller Kondo scale. Indeed, no Kondo anomaly is seen in the resistivity in this material. On the other hand, for low concentration, a broadened Schottky anomaly with  $R \ln 2$  entropy is observed.

Below about 0.5 K, the specific heat coefficient of  $\text{Y}_{0.8}\text{U}_{0.2}\text{Pd}_3$  samples increases [13, 42, 46], consistent with the presence of large entropy below this temperature. Assuming the validity of the quadrupolar Kondo picture, this upturn can have a quite natural interpretation. Namely, the random placement of  $\text{U}^{4+}$  ions produces random electric field gradients due to the charge difference with the  $\text{Y}^{3+}$  ions. These field gradients will produce a random splitting of the quadrupolar doublets. Estimating the average magnitude of this field gradient gives about 5 K, and this yields a spin-field crossover temperature  $T_s \approx (5 \text{ K})^2/40 \text{ K} = 0.63 \text{ K}$  [47], where we used  $T_K = 40 \text{ K}$  estimated from experiment. Hence it seems plausible that this upturn in  $C/T$  represents a spin-field crossover due to the random field gradients.

Standing against this interpretation are several experimental data. (i) The resistivity is indeed NFL in character, but has a linear-in- $T$  low-temperature behaviour rather than saturating with a  $\sqrt{T}$ -law [13, 42]. (ii) In an applied magnetic field, the specific heat behaves contrary to expectation, dropping with increased field and displaying a scaling behaviour incompatible with the simple two-channel Kondo effect [48]. (iii) Ultrasonic measurements on polycrystalline samples, which can measure the quadrupolar susceptibility through sound velocity sampling, shows no significant temperature-dependent softening below the Kondo scale of 40 K in the  $x = 0.2$  samples [49]. (iv) Polarized neutron scattering experiments do suggest the presence of a quasielastic line corresponding to a magnetic ground state for  $x = 0.2$  samples and  $x = 0.45$  samples [50].



In response to (i), as mentioned in section 2.1, the one-impurity resistivity is approximately linear in an intermediate temperature regime. However, the observed linearity only extends to temperatures of order  $0.05T_K$  [31]. With regard to (iii), it should be noted that grain boundary effects and misorientation may obfuscate straightforward observation of ultrasound data in polycrystalline samples [51]. Obviously, single-crystal samples would be preferred here, but it has not proven possible so far to produce these. Finally, with regard to (iv), while the polarized data for  $x = 0.45$  appear to unambiguously support a magnetic triplet ground state, such fits are within error bars for  $x = 0.2$ . Indeed, the assignment of a triplet ground state is problematic, because its width would be less than the 4 meV energy scale obvious from thermodynamics, and it would produce a static susceptibility at least twenty times larger than the measured values. It therefore seems prudent to investigate other possibilities, such as a crystal-field-level crossing as a function of  $x$  [52].

Item (iii) is in many respects the strongest argument against a single-ion quadrupolar Kondo interpretation of the data. On this basis, it was proposed that collective magnetic effects induced the non-Fermi-liquid behaviour. This received bolstering from the fact that for  $x > 0.3$  a spin-glass state is indeed observed [13, 42]. Moreover, as mentioned above, neutron scattering data at higher concentrations support a ground  $\Gamma_5$  magnetic triplet state [44, 50]. However, the Th-doping experiments mentioned above indicate that single-ion physics dominates the non-Fermi-liquid behaviour here, and given that the  $\text{La}_{1-x}\text{U}_x\text{Pd}_3$  system which shows no Kondo effect and no non-Fermi-liquid behaviour displays spin-glass behaviour at the same concentration of U ions, the collective magnetic effects are probably irrelevant to the NFL physics [13, 42, 45].

$\text{Th}_{1-x}\text{U}_x\text{Ru}_2\text{Si}_2$  and  $\text{Th}_{1-x}\text{U}_x\text{Pd}_2\text{Si}_2$ . These are tetragonal systems from which single crystals can be made.  $\text{Th}_{1-x}\text{U}_x\text{Ru}_2\text{Si}_2$  has been studied for four concentrations between  $x = 0.01$  and  $x = 0.07$  [53]. Above about 0.5 K, the  $c$ -axis magnetic susceptibility per U ion is independent of concentration, and log divergent in  $T$ , compatible with the two-channel quadrupolar Kondo effect in tetragonal symmetry. The  $c$ -axis susceptibility is flat in temperature, consistent with Van Vleck susceptibility. A fit to the Bethe *ansatz* calculation of  $\chi$  is good over four decades of temperature. Using the Kondo-scale value of  $T_K = 12$  K from this fit, a reasonable zero-parameter fit to the specific heat is obtained which then shows evidence for the  $R/2 \ln 2$  residual entropy since the theoretical curve integrates to this value. However, a problem exists in that the electrical resistivity shows no Kondo upturn, and while large (of order  $40 \mu\Omega$  cm per U ion), shows a downturn below  $T_K$ . This downturn is roughly logarithmic in temperature, which at least is non-Fermi-liquid like. The properties of  $\text{Th}_{1-x}\text{U}_x\text{Pd}_2\text{Si}_2$  are essentially similar [54].

$\text{La}_{1-x}\text{Ce}_x\text{Cu}_{2.2}\text{Si}_2$ . This system is a plausible candidate for the two-channel magnetic Kondo effect, given the description above. For  $x = 0.1$ , this system shows logarithmic divergences in both  $\chi(T)$  and  $c/T$  [55]. Moreover, taking the logarithmic slopes to compute a Wilson ratio, and using the cubic  $\Gamma_7$  effective moment, one obtains the value of 2.7, to be compared with the theoretical expectation of  $8/3$  [36]. The reason for taking this value despite the tetragonality of the crystal is that susceptibility measurements for  $x = 1$  (fully concentrated  $\text{CeCu}_2\text{Si}_2$ ) are isotropic (cubic) in the best superconducting and most non-Fermi-liquid-like samples [5]. The resistivity is roughly compatible with the two-channel Kondo effect, though measurements at lower temperatures would be desirable since calculations suggest that the  $\sqrt{T}$ -behaviour should set in only below about  $0.05T_K$  [31]. Application of a magnetic field increases the entropy at low temperatures [55], in accord with the simple expectation that polarization of the local moment should lift the residual degeneracy. Although the measured enhancement falls below that expected for the two-channel Kondo effect, it is qualitatively different to the drop of  $C/T$  in magnetic field

expected from the single-channel Kondo effect. Approximate single-ion scaling behaviour is observed in the specific heat for  $x = 0.025, 0.1, 0.15$ , and in the magnetic susceptibility for  $x = 0.025, 0.1$  [55].

Finally, it has been argued based on thermoelectric power measurements for the concentrated ( $x = 1$ ) system that support for a greater  $f^2$  ground-state weight is present in this system (c.f. the dynamic selection rule of section 2.2) [36]. The measured thermopower goes negative well above  $T_K$  for this system and attains a large value of about  $-30 \mu\text{V K}^{-1}$  [5]. The relatively high temperature at which this sign changes suggests that it is not a coherent lattice effect. Within a single-band picture and ignoring excited crystal-field excitations, a positive thermopower, which is most typical for  $\text{Ce}^{3+}$  ions, indicates dominance of particle scattering and strong  $f^1$ - $f^0$  charge fluctuations. A negative thermopower is possible if hole scattering dominates at low temperatures, which is consistent with strong  $f^1$ - $f^2$  charge fluctuations. However, direct thermopower measurements on the dilute system indicate that the thermopower is positive [56, 57]. The only possible ‘loopholes’ in the above theoretical argument concern the relevance of excited crystal-field states in determining the sign of the thermopower and whether multi-band carrier effects may play a role. These require further detailed examination.

Since clear evidence for irreversibility in the magnetization appears for  $x = 0.15$ , it has been suggested that proximity to a spin glass generates the NFL physics [55, 57]. This is further bolstered by analysis of the low-temperature specific heat and resistivity which are claimed to show crossover behaviour consistent with a mean-field theory of a metallic  $T = 0$  spin-glass transition [57].

$\text{Th}_{1-x}\text{U}_x\text{Pd}_2\text{Al}_3$  and  $\text{Th}_{1-x}\text{U}_x\text{Ni}_2\text{Al}_3$ . These two hexagonal systems reveal  $-\ln T$  specific heat coefficients at low temperatures and low concentrations ( $x \simeq 0.1$ ) ([13] (Pd), [58] (Ni)). The susceptibility in the Pd-based system for a polycrystalline sample can be fitted to either  $-\ln T$  or  $1 - A\sqrt{T}$  behaviour at low temperatures. In each case the resistivity apparently saturates with a linear-in- $T$  law. The U ions in these systems are probably tetravalent so they are candidates for the quadrupolar Kondo effect in hexagonal symmetry arising from a non-Kramers doublet. The low-concentration data clearly show single-ion scaling. Kim *et al* [58] argue that in the Ni case there is a proximity to a spin-glass ordering. Nevertheless, given the same crystal structure and the single-ion scaling observed in  $\text{Th}_{1-x}\text{U}_x\text{Pd}_2\text{Al}_3$  [13], it is clear that these systems deserve further careful study as quadrupolar Kondo candidates.

$\text{Th}_{1-x}\text{U}_x\text{Be}_{13}$ . Aliev *et al* have studied this system extensively [59–62]. At the value  $x = 0.9$  which is certainly far from dilute, they find  $C/T \sim -\ln T$ ,  $\chi(T) \sim 1 - AT^{1/2}$ ,  $\rho(T) \sim 1 + BT^{1/2}$  (with  $B > 0$ ), all of which fit the two-channel cubic quadrupolar Kondo picture as discussed in section 2.2. A complication is that in this crystal structure no dopants appear to leave the volume unchanged which means that the hybridization is strongly affected by the doping. (Indeed, since the Th ions are larger they expand the lattice and diminish the hybridization which will lower  $T_K$ . The data appear to suggest that this happens relative to the bulk  $x = 1$  system.) An extensive study of  $\text{M}_{1-x}\text{U}_x\text{Be}_{13}$  alloys by Kim *et al* [63] revealed that while the specific heat could be significantly altered by doping, the magnitude of the low-temperature magnetic susceptibility was hardly affected. This suggests further that the origin of the specific heat and susceptibility are different, consistent with an interpretation in terms of Van Vleck susceptibility which is important for the two-channel quadrupolar Kondo effect in cubic symmetry [18]. This point is further bolstered by the pressure dependence seen in the specific heat being much stronger than that seen in the magnetic susceptibility [64].

A further consistency with the quadrupolar Kondo effect is the non-linear susceptibility

(Aliev *et al* [62]). This was motivated in part by earlier measurements of Ramirez *et al* [65] on UBe<sub>13</sub> which will be discussed below. In theory, the non-linear susceptibility  $\chi^{(3)}(T)$  is defined from the magnetization via

$$\chi^{(3)}(T) = 6[M(H, T) - \chi(T)H]/H^3. \quad (7)$$

For a magnetic doublet ground state,  $\chi^{(3)}$  is expected to be large and negative, as is easily seen from straightforwardly expanding the Brillouin-function magnetization to obtain  $\chi^{(3)} \sim 1/T^3$  for localized moments. This would be modified, at low temperature, to  $\sim 1/T_0^3$  for a Kondo system. In a more general situation,  $\chi^{(3)}$  depends upon the orientation of  $H$ . For a purely localized quadrupolar moment system with a cubic non-Kramers  $\Gamma_3$  ground doublet, Morin and Schmitt [66] have shown that for a field along a principal axis,  $\chi^{(3)}$  will display a *positive* Curie-law divergence, while for a field along a body diagonal  $\chi^{(3)}$  will be of Van Vleck character at low temperature and *negative*. This result is easily understood in terms of the magnetoelastic coupling of the  $\Gamma_3$  ground state—principal-axis fields induce tetragonal distortions which are quadratic in  $H$  and split the doublet. There is no linear coupling however to strains along the body diagonal (matrix elements do exist for excited states). Hence, the non-linear susceptibility for a principal-axis field is essentially a measure of the quadrupolar susceptibility. While the quadrupolar Kondo effect would modify this from a  $1/T$  divergence to  $-\ln T$ , the divergence would still be present, and the characteristic anisotropy provides an excellent test of the applicability of the quadrupolar Kondo model [65].

Aliev *et al* [62] performed measurements only on polycrystalline samples. For  $x = 0.1$  they found that the powder-averaged  $\chi^{(3)}$  is predominantly negative at high temperatures but passes through a minimum with decreasing temperature and tends towards a sign change as the temperature is lowered, which is in accord with the expectations of the previous paragraph. In contrast, when pure UBe<sub>13</sub> is measured for similarly prepared polycrystalline samples,  $\chi^{(3)}$  is relatively large, negative, and decreases with decreasing temperature, qualitatively in agreement with a magnetic ground state. Indeed, the polycrystalline data agrees excellently with the Ramirez *et al* single-crystal data [65], which exclude the possibility of large-moment paramagnetic impurities giving rise to the  $x = 0$   $\chi^{(3)}$ -results.

On the basis of this work and the *positive* coefficient of  $\sqrt{T}$  in the  $x = 0.1$  resistivity, Aliev *et al* [61, 62] put forward an interesting set of hypotheses. First, the  $x = 0.1$  samples are in the strong-coupling regime. That is, the coupling strength exceeds that of the non-trivial fixed point. This can explain the positive coefficient of the resistivity. Second, the ionic ground state changes as a function of Th doping, being an  $f^3\Gamma_6$  doublet for  $x = 0$ , and an  $f^2\Gamma_3$  doublet for  $x = 0.1$ . This would require the U ions to be strongly mixed valent between 3+ and 4+, which is not implausible. This second hypothesis checks with the first hypothesis because it is precisely in the mixed-valence regime where strong coupling might plausibly occur (the dimensionless Schrieffer–Wolff exchange can grow to order unity). The hypotheses are very interesting because the different symmetry ground states would seem to imply a novel quantum critical point at the precise point in  $x$  where the levels cross.

There are three major concerns with the hypotheses.

(1) Taking the Th ions as tetravalent, and the U ions as intermediate between 3+ and 4+ valence, the substitution would add electrons, and this would actually drive the uranium ions more towards trivalence.

(2) While the non-linear susceptibility for pure UBe<sub>13</sub> is strongly temperature dependent, the susceptibility is not, and hence this interpretation is problematic.

(3) It is not clear that a sufficiently small energy scale can be generated in the mixed-valent regime for the uranium ions, though some variational treatments of the ion with full

spherical symmetry suggest that this is possible [67].

(4) It remains to be seen whether strongly broadened excited magnetic triplet levels on the  $U^{4+}$  site can produce such a non-linear susceptibility result. The idea is that at  $x = 0$ , the broad levels strongly overlap with the ground state and dominate the non-linear susceptibility; reduced hybridization resulting in narrower excited levels allows the system to move closer to the ionic limit and the modified Morin–Schmitt results discussed above. A similar narrowing of the  $f^3$  excited configuration in the extreme mixed-valence limit could yield qualitatively similar effects.

Regardless of these concerns, the hypotheses of Aliev *et al* [61, 62] are very intriguing and deserve further exploration.

#### 4. The two-channel Kondo lattice

##### 4.1. The model and results for the normal state

The existence of concentrated and ordered heavy-fermion compounds such as  $UBe_{13}$  [21] and  $CeCu_2Si_2$  [68] which display non-Fermi-liquid behaviour together with superconductivity makes consideration of the lattice generalization of the two-channel Kondo model important. The most straightforward extension of equation (3) is

$$H = \sum_{k\alpha\sigma} \epsilon_k c_{k\alpha\sigma}^\dagger c_{k\alpha\sigma} + \mathcal{J} \sum_{\mathbf{R}} \mathbf{S}_I(\mathbf{R}) \cdot \sum_{\alpha} \mathbf{S}_{c\alpha}(\mathbf{R}) \quad (8)$$

where we assume two degenerate bands throughout the lattice labelled by index  $\alpha$  and the conduction spin operator is

$$\mathbf{S}_{c\alpha}(\mathbf{R}) = \frac{1}{2N_s} \sum_{k,k',\sigma,\sigma'} e^{i\mathbf{R}\cdot(\mathbf{k}-\mathbf{k}')} \mathbf{S}_{\sigma\sigma'} c_{k\alpha\sigma}^\dagger c_{k'\alpha\sigma'}. \quad (9)$$

Obviously, it is unrealistic to expect two conduction bands to remain degenerate throughout the entire Brillouin zone; our model is only relevant to the so-called ‘local approximation’ or infinite-dimensional limit [69–77]. In this limit which we have employed for calculating properties of the model [78], the self-energy becomes rigorously independent of momentum. We shall discuss the potential problems with this limit for real materials in the next subsection.

It is reasonable to ask what expectations one might have for the lattice model. In the normal state of the model, one can expect very different behaviour from that of the single-channel Kondo lattice. For simplicity, restrict consideration to half-filling. Then the ordinary Kondo lattice is expected to be an insulator. This can be seen from strong coupling where bound singlets form at every site, to intermediate coupling where one may view (in a slave-boson theory) the system as a ‘band’ insulator with two electrons per cell (local and itinerant). Indeed, in the symmetric Anderson lattice this band insulator description is accurate [79]. Finally, for weak coupling, intersite interactions drive the model to an antiferromagnetic insulator, the intersite coupling being a mixture of conduction-spin-polarization-mediated interactions (RKKY) and superexchange (virtual charge fluctuations). Moving away from half-filling it is possible to find ‘Fermi-liquid’ regions in the asymmetric Anderson lattice model relatively close to half-filling [80]. If we raise the degeneracy and shove the Kondo resonance structure off the Fermi energy, we expect a metal for sufficiently large  $J$ .

We can understand these results within the ‘average  $T$ -matrix approximation’ [81, 82], which seeks to estimate the lattice self-energy from the single-ion  $T$ -matrix. This

approximation is an exact version of the local approximation for the unphysical limit of a Lorentzian density of states. By combining the Lippmann–Schwinger equation relating the conduction electron Green’s function and  $T$ -matrix with the Dyson equation, and enforcing the local approximation in keeping only frequency dependence at most, we obtain

$$\Sigma_c(\omega) = \frac{t_1(\omega)}{1 + G_0(\omega)t_1(\omega)} \quad (10)$$

where  $t_1$  is the one-particle on-site  $t$ -matrix, and  $G_0$  is the local (on-site) unperturbed conduction electron Green’s function. In the special case of particle–hole symmetry,  $G_0(0) = -i\pi N(0)$ , while for phase-shift scattering only,

$$t_1(\omega) = -\sin \delta(\omega) \exp[i\delta(\omega)]/[\pi N(\omega)].$$

In this case, the imaginary part of the self-energy at the Fermi energy cancels out and we have only  $\Sigma_c(0) = -\tan \delta(0)/[\pi N(0)]$ . For  $\delta(0) = \pi/2$  expected for  $S_I = 1/2$ , the divergence signals formation of an insulating state, while for  $\delta \neq \pi/2$ , the phase shift is simply absorbed in an overall energy shift and a normal-metal state prevails. The latter situation may be realized in an applied magnetic field in the  $S_I = 1/2$  case, or by moving to higher spin degeneracy (the Coqblin–Schrieffer limit). We note that this is a manifestation of Bloch’s theorem: at the Fermi energy we simply form coherent quasiparticle states in a metallic or band insulator phase. The phase shift at every magnetic site simply renormalizes the underlying lattice potential.

For the two-channel model, application of similar reasoning indicates a big contrast to the one-channel case. The single-particle  $T$ -matrix has the Fermi-level value  $-i/[2\pi N(0)]$  [30]. Plugging this into equation (10) within the ATA gives

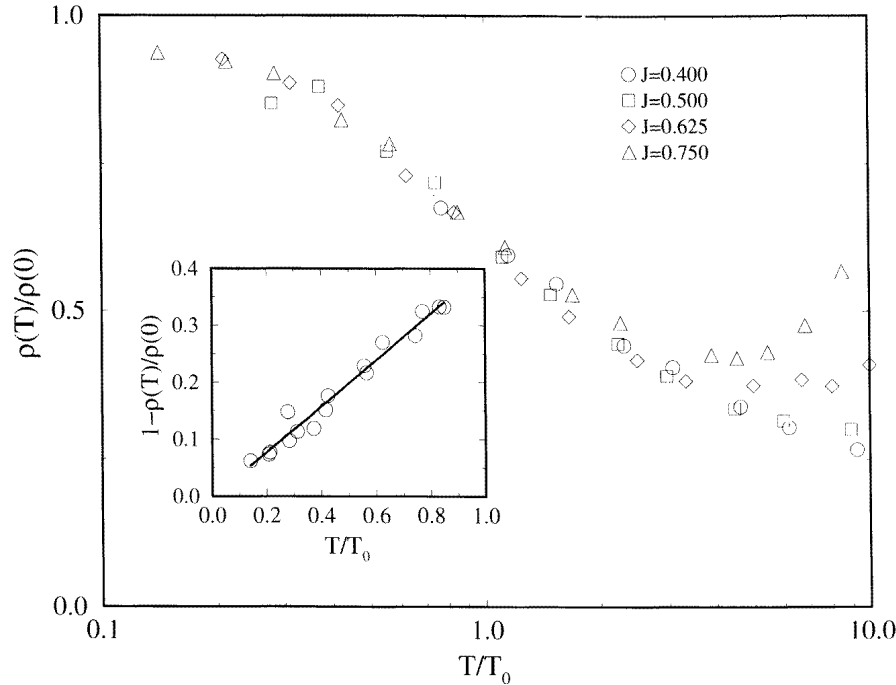
$$\Sigma_c(0) \approx \frac{-i}{\pi N(0)} \quad (11)$$

which is purely imaginary! Hence, we would expect a metallic phase (the density of excitations at the Fermi energy is finite) but with a finite residual resistivity—the electronic quasiparticles have an *intrinsic* linewidth. We call such a state an ‘incoherent metal’ [78, 82] for the clear reasons that Bloch’s theorem is violated for this state.

The physical understanding of this violation is in terms of the degeneracy of the two-channel Kondo cloud discussed in section 2.1: until some phase transition arises which locks in coherence of the degenerate screening clouds around each magnetic site, a residual entropy persists with a concomitant residual scattering due to disorder in the spin system. The self-energy is indeed the same at every site, which reflects translational invariance. Because of the disorder it is clear that this incoherent metal phase is not the true ground state of the system, and we would expect some ordering to intervene which can restore a zero-resistance metal at  $T = 0$  or an insulating phase. The situation is, in this sense, not unlike that of gadolinium metal: the large and temperature-independent resistivity above the Curie point of this ferromagnet is well understood as arising from spin-disorder scattering off the disordered Gd moments, which give rise to an  $R \ln 8$  entropy per site in the paramagnetic state. Below the Curie point, as the Gd moments order coherence is restored and the resistivity is driven to zero.

The ATA is sufficient to guide our thinking, but is not a rigorous approximation for any reasonable lattice picture due to the lack of self-consistent feedback of the self-energy. More appropriate is the local approximation mentioned above [77], which becomes rigorous in infinite spatial dimensions. The infinite-dimensional limit is taken for a nearest-neighbour hopping model of the conduction states by holding  $t^* = 2t\sqrt{d}$  fixed, where  $t$  is the hopping

matrix element and  $d$  the spatial dimensionality. In this limit, it may be shown that non-site-diagonal contributions to the self-energy fall off at least as quickly as  $1/\sqrt{d}$  with increasing  $d$ . This means that the problem maps to an effective-impurity model: by selecting a single site of the lattice, solving for the self-energy with a medium density of states specified by the site-excluded lattice, and self-consistently feeding back the impurity solution into the lattice a rigorous thermodynamic limit solution is obtained.



**Figure 8.** The resistivity of the two-channel Kondo lattice in infinite dimensions. The resistivity is calculated in the paramagnetic phase and is finite due to the spin-disorder scattering effect described in the text. In the temperature region below  $T_K$ , the resistivity is approximately linear in  $T$ , though a different behaviour at lower temperatures cannot be ruled out. This set of calculations was performed at particle-hole symmetry, and is taken from reference [78]; a maximum appears possible away from particle-hole symmetry [84].

By employing quantum Monte Carlo methods to simulate the effective-impurity model, studies have been carried out on the resistivity [78] and conductivity [83] of the two-channel lattice in infinite spatial dimensions and find just such an incoherent metal phase in the normal state. We show the resistivity results of reference [78] in figure 8. Over a wide range of temperatures below  $T_K$  (which is determined for the lattice by fitting the local susceptibility to the exact results of the Bethe *ansatz* [32]), it is found that the resistivity is approximately linear in temperature, and the residual resistivity (extrapolated to zero temperature) is finite. Using the free-energy formulae in reference [77], it is straightforward to show that this phase has a residual entropy, which is  $R \ln 2/2$  per site in the half-filled limit. It is found at low temperatures that the electrical conductivity is non-Drude like, with a peak at a finite frequency [83, 84]. We note that the lattice  $T_K$  is significantly enhanced with respect to the single-impurity value; in particular, at half-filling, to leading exponential order it is found numerically that  $\ln[T_K^{\text{lattice}}]/\ln[T_K^{\text{impurity}}] \approx \sqrt{\pi}$  [78].

Because of the residual entropy, we do not expect the true ground state of the system to be the paramagnetic one. It represents an unstable fixed point, but an unstable fixed point in which the excitation spectrum is decidedly not Fermi-liquid like: the slope of the real part of the conduction electron self-energy near the Fermi energy is positive, which is related through the Hilbert transform to the finite imaginary part of the self-energy at the Fermi level. The standardly defined quasiparticle mass, proportional to  $1 - \partial \text{Re } \Sigma / \partial \omega|_0$ , is *reduced* (it can even go negative), and the width of any possible electronic quasiparticle peak is hopelessly broad compared to its position. In short, the quasiparticle picture breaks down [78]. Studies of the phase diagram [85] near half-filling of each channel (band) confirm that for  $J \leq 0.75t^*$ , an antiferromagnetic phase is likely which vanishes as a function of doping near  $n_c = 0.8$ , where  $n$  is the occupancy per site of each band. The interaction driving this is a mix of RKKY and superexchange couplings, as in the single-channel case. However, here the superexchange is between adjacent doublet clouds and so scales as  $1/J$  for large  $J$ . This phase is commensurate except near  $n \approx n_c$ . Hence, at half-filling, we expect an antiferromagnetic insulator over most physical coupling strength values. Away from half-filling we have recently found evidence for a novel superconducting ground state, which we discuss somewhat further in the conclusions. Full details of the calculation of the phase diagram of the two-channel Kondo lattice are presented elsewhere [85].

Anders and Jarrell [84] have carried out a study of the effects of applied spin field on the two-channel lattice in infinite spatial dimensions. To do this they utilized the non-crossing approximation (NCA) method for solving the effective-impurity problem (for a description, see references [86, 87]). In accord with the crossover to impurity Fermi-liquid behaviour, it is found that the resistivity drops significantly and a Drude-like peak is restored in the optical conductivity. Moreover, an approximate scaling behaviour is found for the magnetoresistance, which is that  $[\rho(H_s, T) - \rho(0, T)]/\rho(0, T) \sim f(H_s/(T + T^*)^\beta)$ , with  $\beta = 0.39$ ,  $T^* = 0.006T_K$ . Interestingly, for zero spin field, this study performed away from particle-hole symmetry finds a resistivity peak below the temperatures accessible to the particle-hole-symmetric quantum Monte Carlo results.

#### 4.2. Possible relevance to $UBe_{13}$

The normal-state resistivity of the heavy-fermion superconductor  $UBe_{13}$  is very unusual [88, 89]. First, the value at the transition is reproducibly large for the best single crystals (of the order of  $100 \mu\Omega \text{ cm}$ ). The best samples also have sharp nuclear magnetic resonance lines for  $^9\text{Be}$  nuclei, enough so that the one of thirteen sites which is cubic can be readily observed [90]. Bolstering this result are data which show sharp resistive superconducting transitions that persist even in applied magnetic field. These NMR results contrast with those for the strongly disordered NFL material  $UCu_{5-x}Pd_x$  [16]. Second, the resistivity is strongly and reversibly depressed with the application of pressure, which is difficult to understand if it corresponds to any ordinary ‘dirt’ that might be squeezed out irreversibly with pressure [89]. Pressure also restores curvature to the resistivity and a clear window of  $T^2$ -coefficient opens up [89]. Third, a non-trivial scaling law describes the negative magnetoresistivity, specifically  $\rho(T, H)/\rho(T, 0) \sim f(H/[T + T^*]^\beta)$  where different groups have found  $\beta = 1$ ,  $T^* = 0$  [88] and  $\beta = 0.6$ ,  $T^* = -0.6 \text{ K}$  [91].

The large residual resistivity of  $UBe_{13}$  in an evidently clean system finds a natural explanation in terms of the two-channel Kondo-lattice results described in the previous paragraph [78]. A likely description is in terms of the quadrupolar Kondo effect, though a two-channel magnetic Kondo effect cannot be ruled out and would appear more consistent with non-linear susceptibility data [65]. The fact that the resistivity has a maximum is

compatible with the NCA results for the two-channel lattice away from half-filling [84]. We note also that inclusion of quadrupolar/magnetic fluctuations is generically expected to reduce the resistivity (barring special conditions in proximity to an insulating phase), since these fluctuations will provide a hint of order at lower temperatures which will remove the residual entropy.

In either the magnetic or quadrupolar two-channel lattice case, a qualitative explanation of the reduction of the residual resistivity with pressure goes as follows: pressure increases the hybridization, which in turn increases the overlap between distinct crystal-field states, driving the uranium ions towards the limit of a single Hund's rule multiplet. In this case, variational studies which become exact in the limit of large degeneracy support the formation of a singlet ground state [67]. This hypothesis may in principle be at least partially tested by performing Raman scattering in a diamond-anvil cell on a  $\text{UBe}_{13}$  sample. Raman scattering has detected magnetic dipole transitions within the low-lying f-electron states that agrees with neutron scattering observations [92]. These transitions should display substantial broadening with the application of pressure.

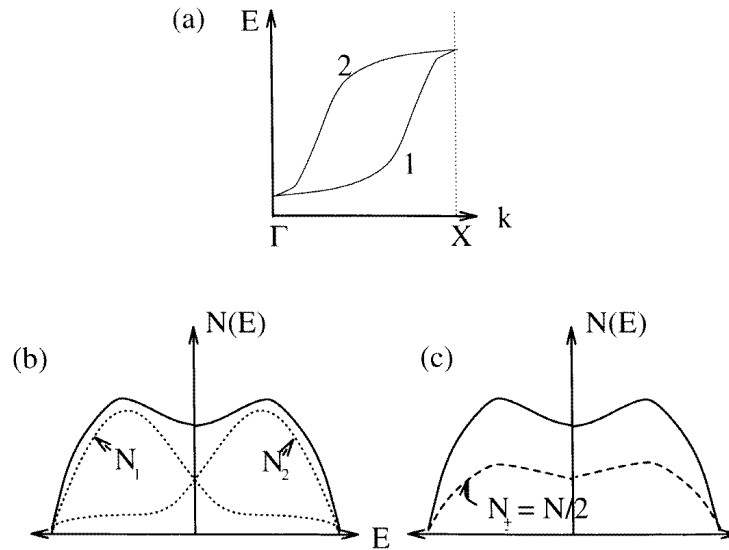
With regard to the magnetoresistance, it is tempting to compare the approximate scaling law discerned from the NCA [84] with the  $\text{UBe}_{13}$  data [88, 91], but caution is in order since in the quadrupolar case the proper *low-field* interpretation of the magnetic field is that it is a *channel field*, for which no calculations yet exist. Assuming  $V/|E(f^2) - E(f^3)| \approx 0.3$  as is reasonable for an 0.3 eV hybridization strength and 1 eV interconfiguration splitting, and given a measured susceptibility of about  $0.012 \text{ emu mol}^{-1}$  (interpreted here as Van Vleck susceptibility), the crossover field  $H^*$  at which channel-field physics is overtaken by spin-field physics for the quadrupolar case is about 4 T.

A serious criticism of the application of the simplified lattice model of the previous subsection to a real material such as  $\text{UBe}_{13}$  concerns the unphysical assumption of two degenerate bands throughout the Brillouin zone. It is well known from the early days of electronic structure theory that such degeneracies can only arise at special points and generically only a twofold degeneracy associated with time-reversal symmetry (Kramers' theorem) can obtain at an arbitrary point in the Brillouin zone. Indeed, we expect the Kramers' degeneracy to hold for the electrons in  $\text{UBe}_{13}$  though the strong spin-orbit coupling will make the effective moment dependent upon  $\mathbf{k}$ . This latter point is only relevant in the presence of an applied magnetic field. However, it is easy to show with a simple tight-binding formulation that even in the limit of zero spin-orbit coupling, a lattice of orbitals with  $\Gamma_3$  symmetry (assumed to be  $x^2 - y^2, 3z^2 - r^2$  for simplicity) cannot yield degenerate bands except at the cubic zone centre and zone corner points. The origin of this is that the degeneracy of the local orbitals obtains from the point group symmetry which assumes a particular local quantization axis. This quantization axis then depends upon the site, and mixing of orbitals from site to site is possible which produces  $k$ -space matrix elements that vanish only at the zone centre and the zone corner.

It turns out that these effects vanish in infinite spatial dimensions or in a local approximation in finite spatial dimensions. That is, any effect of the shift of the orbital quantization axis from site to site must be felt through the momentum dependence of the self-energy in ways which are not as yet clear, and therefore enters in  $1/d$  corrections to the theory.

The way to understand this qualitatively is straightforward, as is schematically illustrated in figure 9. The densities of states of the two bands are distinct and coincide only at the band edges. However, no splitting can occur for the local-orbital-projected density of states about a single site, which rigorously must be half of the total density of states. If any differences occurred, it would indicate a breaking of the point group symmetry. Such a spontaneous





**Figure 9.** A schematic view of bands and densities of states for a realistic two-channel Kondo lattice. We assume a simple cubic lattice in three dimensions and local orbitals of  $3z^2 - r^2, x^2 - y^2(\pm)$  symmetry. (a) shows the two diagonal bands (labelled as 1, 2) in  $k$ -space which can only be degenerate at the zone centre ( $\Gamma$ ) and the zone corner ( $X$ ). (b) shows the density of states for the two diagonal bands of  $k$ -space which are distinct in energy. The total density of states, however, must add to give the total density of states of (c) which is taken from the locally (on-site-) projected  $\pm$  orbitals. Here, the exact equality of the two different densities of states is assured by point group symmetry.

symmetry breaking is indeed possible and may be searched for in the numerical calculations (the possibility of a channel-density-wave ordered state, for example, exists at quarter-filling in the infinite-dimensional lattice). But it cannot describe the paramagnetic phase. Because the local density of states determines the entire local Green's function through spectral relations, it is clear that in the absence of an applied stress or spontaneous orbital ordering the banding effects cannot alter the local self-energy. (Note also that the interband matrix elements of the aforementioned two-orbital tight-binding model will have vanishing weight in infinite spatial dimension—they have a d-wave symmetry and hence vanish for the same reason that form factors for d-wave pairing do, for example.)

We offer a speculation that in a three-dimensional lattice the band-splitting effects may be relevant. The crucial comparison is of the magnitude of the renormalized splitting (dressed by the self-energy corrections) to the intersite interaction energy and Kondo scale. If the renormalized band splitting is smaller than the intersite interaction energy, then presumably some kind of ordering will dictate the ground state, possibly magnetic or quadrupolar (or superconducting). In the context of applying this model to  $\text{UBe}_{13}$ , it would appear that the renormalized band splitting is quite small and that superconductivity dominates intersite ordering. In the case of  $\text{UPt}_3$ , if one tries to apply this model, intersite coupling probably wins (as is evident from the magnetic order at 5 K) [93]. If the renormalized band splitting is larger than the intersite coupling but smaller than the Kondo scale, it may be possible to develop a heavy Fermi-liquid state, as is observed in the compound  $\text{PrInAg}_2$  [94]. This cubic material has  $4f^2$  Pr ions with clear evidence for non-Kramers ground doublets.

## 5. Conclusions and directions

In conclusion, we have presented a pedagogical discussion of the two-channel Kondo model and its possible appearance in heavy-fermion systems. We have provided a critical review of the relevance of the model to a number of diluted heavy-fermion alloys. We have overviewed recent results from the theory of the two-channel Kondo-lattice model in infinite spatial dimensions and pointed out the possible relevance to the heavy-fermion compound  $\text{UBe}_{13}$ .

In closing, we would like to offer some brief speculations concerning superconductivity in heavy-fermion systems. First, we note that the two-channel impurity model has strong on-site odd-frequency pair correlations leading to a log-divergent local pair-field susceptibility in the channel-singlet, spin-singlet sector [30] as first stressed by Emery and Kivelson [95]. In such a state electrons avoid each other through a wave function which has a *temporal* node [96–99]. In the lattice setting, odd-frequency pairing has been argued to favour a *staggered* superconducting state in which the phase alternates from site to site [98]. Evidence for just such a tendency has been found in calculations for a diluted one-channel Kondo lattice in one dimension where left- and right-moving electrons are approximately decoupled so that the system almost behaves as a two-channel model [100]. This study finds that the staggered odd-frequency pairing correlation functions have long-distance algebraic decays significantly enhanced over the non-interacting limit.

Next, we note that the two-channel Kondo model supports a first-order phase transition to an odd-frequency pairing state in infinite spatial dimensions [85]. Details of this work are presented elsewhere. The critical temperature of this superconductivity tracks the lattice Kondo scale almost perfectly, occurring at approximately half of  $T_K$ . This state is a singlet in both spin and channel indices, and the requirement of a temporal node follows simply from the Pauli principle. The possibility of such a state is anticipated in part by the enhancement of the corresponding local pair-field susceptibility in the impurity model. However, we stress that this superconductivity requires a finite concentration of two-channel sites and proceeds only by an unusual first-order transition in the model. The transition temperatures throughout the Brillouin zone appear to be degenerate.

Clearly, the transitions of the heavy-fermion superconductors appear to be second order, in contradiction with the above. Further investigation of the strength of the first-order transition in the above model, together with some estimation of the effects of finite dimensionality, are required to help resolve this issue. However, we note the following properties of the heavy-fermion superconductors which support an interpretation in terms of a two-channel Kondo-lattice model.

(1) *‘Ubiquity’*. All Ce-based heavy-fermion superconductors [1, 101–103] are found in the same 1–2–2 crystal structure and in fact two ( $\text{CeCu}_2\text{Si}_2$  and  $\text{CeCu}_2\text{Ge}_2$  [101]) are basically identical under pressure. This suggests a common origin to the physics of these materials. In contrast, there are four different U-based systems in hexagonal ( $\text{UPt}_3$ ,  $\text{UPd}_2\text{Al}_3$ , and  $\text{UNi}_2\text{Al}_3$ ), tetragonal ( $\text{URu}_2\text{Si}_2$ ), and cubic ( $\text{UBe}_{13}$ ) symmetries. It is clear that the relative difficulty of making diverse Ce-based systems compared with the relative ease of making U-based systems fits nicely with the picture of robust two-channel Kondo physics in U-based systems compared with Ce-based systems.

(2) *Non-Fermi-liquid alloys and compounds*. As compounds,  $\text{CeCu}_2\text{Si}_2$  [68] and  $\text{UBe}_{13}$  [21] clearly have a superconducting instability arising in a non-Fermi-liquid normal state. The alloys  $\text{La}_{1-x}\text{Ce}_x\text{Cu}_2\text{Si}_2$  [55],  $\text{Th}_{1-x}\text{U}_x\text{Ru}_2\text{Si}_2$  [53],  $\text{Th}_{1-x}\text{U}_x\text{Pd}_2\text{Si}_2$  [54],  $\text{Th}_{1-x}\text{U}_x\text{Be}_{13}$  [59, 60, 61, 62],  $\text{Th}_{1-x}\text{U}_x\text{Pd}_2\text{Al}_3$  [13], and  $\text{Th}_{1-x}\text{U}_x\text{Ni}_2\text{Al}_3$  [58] all show non-Fermi-

liquid behaviour with at least some overlap with the two-channel Kondo-impurity model as discussed in section 3. This strong correlation of non-Fermi-liquid physics with superconductivity is certainly intriguing and must be explained by any successful theory of these materials.

(3) *The general absence of a spin response in U-based superconductors.* The spin- and channel-singlet character of the pair-field instability mentioned above suggests that the magnetic field should break up pairs in these materials if the model is relevant. However, in  $\text{UBe}_{13}$ ,  $\text{UPt}_3$ ,  $\text{URu}_2\text{Si}_2$  there is in fact no strong change of the susceptibility in passing through the transition [90, 104–106], and  $\text{UBe}_{13}$  in particular shows no strong evidence of Pauli limiting in the upper critical field [107], consistent with essentially zero magnetic moment of the conduction states. In detail, some change of  $c$ -axis susceptibility is observed in  $\text{UPt}_3$  which is larger than any change in the basal plane [105]. Also, the upper-critical-field curve for  $c$ -axis fields actually crosses that for in-plane fields at low temperatures. This has been interpreted in terms of a triplet-pair-field state that has the feature of Pauli limiting along the  $c$ -axis and none in the basal plane [108]. For the quadrupolar Kondo model applied to these materials, you would expect just these behaviours. In the cubic case of  $\text{UBe}_{13}$ , there is no Pauli susceptibility associated with the quadrupolar Kondo ground state, only a Van Vleck susceptibility associated with transitions to damped excited states. The Van Vleck susceptibility is dominated by energy transfers at the 15 meV scale, compared with the 0.1 meV scale of the superconductivity, and so is unlikely to change on passing through the transition. On the other hand, for the hexagonal material  $\text{UPt}_3$ , it is expected that the quadrupolar Kondo effect will have a  $c$ -axis magnetic response, so pair breaking and susceptibility reduction (along with Pauli limiting) are possible for  $c$ -axis fields. At zeroth order, we expect no change in the in-plane susceptibility which will be of Van Vleck type [109]. However, the crystal-field levels are damped in this case, so to the extent that the density of states of electrons is reduced on entering the superconducting state we can expect some change of the in-plane susceptibility. Indeed,  $\text{UPd}_2\text{Al}_3$  for both directions of field displays a Knight-shift drop on entering the superconducting state as sampled by Al Knight-shift experiments [110]. However, the relative drop for the  $c$ -axis (28%) is substantially larger than that of the basal plane (13%) which is qualitatively consistent with the above expectations. In  $\text{CeCu}_2\text{Si}_2$  where a magnetic description should apply, and Knight-shift drops are expected, sizeable drops are indeed seen [104].

(4) *Multiple phases.* Here we simply remark that the possibility of multiple superconducting phases for staggered odd-frequency pairs has already been discussed phenomenologically elsewhere [111]. This may apply to the multiple phases of  $\text{UPt}_3$  and  $\text{U}_{1-x}\text{Th}_x\text{Be}_{13}$ . Two other possibilities arise in a study of the phase diagram of the model in infinite dimensions: (i) there may be multiple phases with different  $q$ -vectors, and (ii) there may be a low-temperature transition to a spin-triplet, channel-spin-triplet odd-frequency pair state. Detailed numerical investigations will be necessary to answer the question of which scenario if any can apply to the real materials.

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