LETTERS

Scaling the Kondo lattice

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The origin of magnetic order in metals has two extremes: an instability in a liquid of local magnetic moments interacting through conduction electrons, and a spin-density wave instability in a Fermi liquid of itinerant electrons. This dichotomy between 'local-moment' magnetism and 'itinerant-electron' magnetism is reminiscent of the valence bond/molecular orbital dichotomy present in studies of chemical bonding. The class of heavy-electron intermetallic compounds of cerium, ytterbium and various 5*f* elements bridges the extremes, with itinerant-electron magnetic characteristics at low temperatures that grow out of a high-temperature local-moment state[1]. Describing this transition quantitatively has proved difficult, and one of the main unsolved problems is finding what determines the temperature scale for the evolution of this behaviour. Here we present a simple, semi-quantitative solution to this problem that provides a basic framework for interpreting the physics of heavy-electron materials and offers the prospect of a quantitative determination of the physical origin of their magnetic ordering and superconductivity. It also reveals the difference between the temperature scales that distinguish the conduction electrons' response to a single magnetic impurity and their response to a lattice of local moments, and provides an updated version of the well-known Doniach diagram[2].

The physics of a single local-moment spin coupled to the conduction electrons in a simple metal—the single-ion Kondo problem—is now well understood and completely characterized by the strength, J, of that coupling and ρ, the density of electronic states of the host metal at the Fermi surface[3]. Until now no comparable understanding has been achieved for Kondo lattice materials—dense intermetallic compounds containing a chemically ordered lattice of local moments that are Kondo coupled to the compound's conduction electrons[1]. Such materials have low-temperature properties characterized by large effective electronic masses (whence the nomenclature 'heavy electron') deriving from collective local-moment deconfinement—a 4*f* or 5*f* local-moment–itinerant-electron transition that emerges below a characteristic temperature scale T^* (ref. 4). Above T^*, the local moments exhibit a Curie–Weiss-type magnetic susceptibility that begins to change at T^*. The deconfinement evolving below T^* becomes visible in Knight shift experiments[5] that show the emergence of a second component of the spin susceptibility, the heavy-electron Kondo liquid, whose logarithmic increase in density of states with decreasing temperature can be measured in these and other experiments[6]. It is generally accompanied by a drop in electrical resistivity, often called the development of coherence, as well as the development of a low-frequency Drude peak in the optical conductivity and signatures in other properties that we discuss in the Supplementary Information. The interplay of their single-site Kondo physics with intersite coupling effects gives the study of these intermetallic compounds a remarkable richness.

In the collective deconfinement, the entropy, $R\ln 2$ (where R denotes the universal gas constant), carried by the local-moment degeneracy becomes associated with the itinerant-electron system. This suggests that T^* is the temperature at which this entropy is developed in the lattice. Thus, with the entropy associated with the lattice vibrational degrees of freedom and excited crystal field contributions subtracted, we might find that $S(T^*) \approx R\ln 2$, assuming a ground-state crystal field doublet for the *f* electrons, typical of cerium and ytterbium heavy-electron materials (other situations have a simple generalization). We find that this is observed experimentally: a magnetic entropy of approximately $R\ln 2$ develops at temperature T^*, supporting the viewpoint that coherence represents the entanglement of the local-moment spin degree of freedom with those of the conduction electrons.

The unusual and interesting properties of Kondo lattice materials can thus be viewed as deriving from a large transfer of entropy from the high-temperature local-moment system, making T^* a primary parameter for characterizing such systems. We note that the heavy-electron superconductors in this class of materials typically develop an entropy of order $(0.1–0.2)R\ln 2$ at the superconducting transition temperature, T_c, with T_c/T^* values of order 1/20, suggesting a possible connection between T_c and T^*. It is of fundamental and practical importance to learn what determines the magnitude of T^*. We suggest a very simple solution to this problem: the Kondo coupling J governs both the single-ion screening (hybridization) that proceeds independently above T^* and the collective hybridization that produces coherent behaviour below T^*.

In a study of heavy-electron $Ce_{1-x}La_xCoIn_5$ alloys, it has been argued that T^* corresponds to the nearest-neighbour intersite coupling[7]. Because this coupling may reasonably be expected to arise primarily from the coupling of adjacent cerium ions through the conduction electrons (namely the Ruderman–Kittel–Kasuya–Yosida (RKKY) interaction), which increases as $k_B T_{RKKY} \sim J^2\rho$ (where k_B denotes Boltzmann's constant and T_{RKKY} the RKKY temperature), on general grounds we would expect T^* to take the form[1]

$$T^* = cJ^2\rho \quad (1)$$

where J is the local Kondo coupling, ρ is the density of states of the conduction electrons coupled to the local spins and c is a constant determined by the details of the hybridization and the conduction electron Fermi surface. Here and in the following we set $k_B = 1$ for simplicity. The density of states ρ can be estimated from the Sommerfeld coefficient γ of the corresponding non-magnetic lanthanum (or yttrium, lutetium or thorium) compounds[3]:

$$\rho = \frac{3\gamma}{\pi^2}$$

On the other hand, the single-ion Kondo temperature, T_K, is given approximately by[3]:

$$T_K = \rho^{-1} e^{-1/J\rho}$$

Because both temperature scales are determined by the single-ion Kondo coupling J, we have the following equation:

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$$J\rho = -1/\ln(T_K\rho) = \sqrt{c^{-1}T^*\rho} \quad (2)$$

From a plot of $\sqrt{T^*\rho}$ against $-1/\ln(T_K\rho)$ it is therefore possible to determine whether equation (1) provides a useful expression for T^*, that is, one in which the value of c changes little from one material to another.

We have accordingly examined a wide variety of heavy-electron materials for which both T^* and T_K are already known or can be determined, with the results presented in Table 1. In constructing Table 1, we used the fact that the deconfinement of the local moments has a number of direct physical consequences, any of which can be used to estimate T^*: the evolution of the magnetic entropy towards $R\ln 2$, the development of a low-frequency Drude peak in the optical conductivity, the onset of a Knight shift or Hall anomaly, the departure of the magnetic susceptibility from Curie–Weiss behaviour, and a coherence peak in the magnetic resistivity. These different methods gave roughly the same results as long as other physical effects are not present or can be taken into account. We note that, in general, crystal field effects may have a role, so the actual behaviour may be more complicated than a simple picture based on the dominance of T^* would indicate. Determining T_K also requires some care. The most straightforward way to determine T_K for a given material is to use measurements of single-ion Kondo behaviour in the corresponding dilute compounds with a very low cerium (or uranium or ytterbium) concentration in the non-magnetic host. Because J and ρ are volume dependent, differences between the unit-cell volumes of the non-magnetic reference host and the corresponding fully dense Kondo lattice must be taken into account. As discussed in the Supplementary Information, an equivalent volume can be achieved either by applying external pressure or by proper chemical substitution.

Using the results for T_K and T^* given in Table 1, we obtain the result shown in Fig. 1, where a choice of $c \approx 0.45$ is seen to provide an excellent account of T^* for a broad range of materials that have cubic, tetragonal or hexagonal crystal structures and a magnetically ordered, superconducting or paramagnetic ground state. It is remarkable that a simple relation between T^*, the single-ion Kondo scale T_K, and the unenhanced density of states ρ should be semi-quantitatively valid for such a variety of materials. Significant variation in the material-specific factor, c, is expected, yet this seems a weak effect for this set of materials. Moreover, the dominance of single-ion, local-moment compensation when $T_K > T_{RKKY}$, which was expected to occur frequently in heavy-electron materials, is not present in this group of materials, although it could still appear in a larger data set. The present compilation suggests that there is a single dominant energy scale in the physics of heavy-electron materials—namely the Kondo coupling J—that determines both T_K and T^*. We note that the value of the structure factor c will be slightly different if a different formula is used for T_K, but the overall behaviour is not affected.

Table 1 | Experimental T^*, T_K and γ values for a variety of Kondo lattice compounds

Compound	T^* (K)	T_K (K)	γ (mJ mol^{-1} K^2)	$J\rho$	J (meV)	c	Reference
$CeRhIn_5$	20 ± 5	0.15	5.7	0.10	40	0.45	6, 8, H.-O.L.*
$CeCu_6$	35 ± 5	3.5	8	0.15	43	0.49	9, 10
$CeCu_2Si_2$	75 ± 20	10	4	0.15	90	0.47	6, 11, 12
$CePb_3$	20 ± 5	3	13	0.15	28	0.41	13, 14
$CeCoIn_5$	50 ± 10	6.6	7.6	0.16	49	0.55	4, 6, 7
$CePd_2Si_2$	40 ± 10	9	7.8	0.17	51	0.41	15, 16
$CePd_2Al_3$	35 ± 10	10	9.7	0.18	43	0.40	17, 18, 19
$CeRu_2Si_2$	60 ± 10	20	6.68	0.19	66	0.42	20, 21
U_2Zn_{17}	20 ± 5	2.7	12.3	0.15	29	0.41	22, 23
URu_2Si_2	55 ± 5	12	6.5	0.17	62	0.45	6, 24, 25
UBe_{13}	55 ± 5	20	8	0.19	57	0.43	26, 27
UPd_2Al_3	60 ± 10	25	9.7	0.21	51	0.48	19, 28
$YbRh_2Si_2$	70 ± 20	20	7.8	0.19	58	0.53	Z.F.†
$YbNi_2B_2C$	50 ± 5	20	11	0.21	44	0.47	29

* H.-O. Lee, unpublished resistivity and specific heat measurements on $Ce_{1-x}La_xRhIn_5$.
† Z. Fisk, unpublished resistivity and specific heat measurements on $Yb_{1-x}Lu_xRh_2Si_2$.

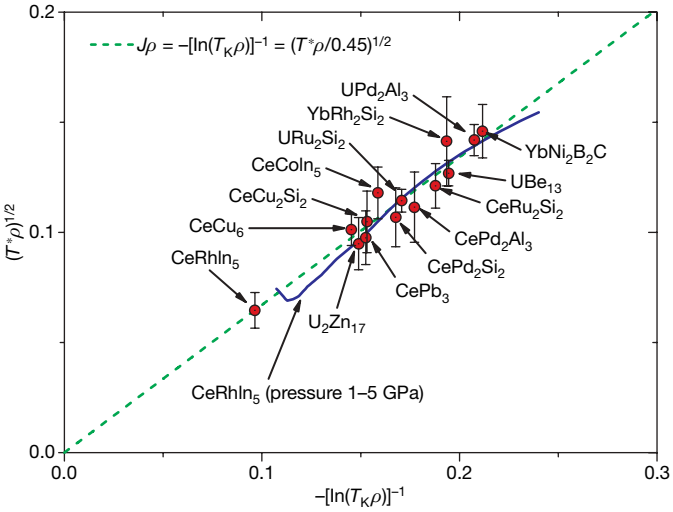

Figure 1 | Confirmation of T^* given by the intersite RKKY interaction for a variety of Kondo lattice materials. The solid line shows T^* (resistivity peak) of $CeRhIn_5$ under pressure from 1 GPa (lower left) to 5 GPa (upper right). Each error bar indicates a typical range of uncertainties for estimating T^* (see the Supplementary Information). The dashed line is a guide to all the materials. The evident correlation between T^*, T_K and ρ provides verification of equation (2) and the RKKY nature of T^*. The small deviation from the $c = 0.45$ line may result from details of the local hybridization and the conduction electron Fermi surface.

Our approach also enables us to update the well-known Doniach diagram[2], using our experimental values of T^* in place of estimates of T_{RKKY} and comparing their variation with J to that of T_K for a specific choice of c. The result is shown in Fig. 2. Remarkably, for $c = 0.45$, we find that $T^* = T_K$ for $J\rho \approx 0.35$, a value very close to that obtained for the one-dimensional Kondo necklace[30]. We call attention to the fact that the heavy-electron materials considered here that appear to exhibit quantum critical behaviour have J values such that $J\rho$ clusters between 0.15 and 0.20, values that are low in comparison with those (0.35 or greater) at which the single-ion Kondo coupling may be expected to play a significant role. Furthermore, the cerium-based heavy-electron superconductors $CeCu_2Si_2$ and $CeCoIn_5$, as well as

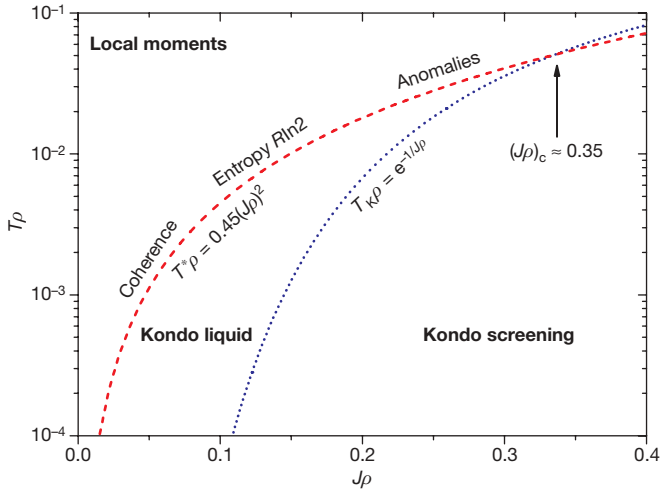

Figure 2 | Updated Doniach diagram for Kondo lattice materials. The dashed line indicates the intersite RKKY temperature, T^*, with $c = 0.45$. Below T^*, the local $4f$ (or $5f$) moments become partially itinerant, giving rise to the universal Kondo liquid behaviour and rich phase diagrams. The dotted line is the single-ion Kondo temperature T_K, below which the remaining local moments are screened. T_K reaches T^* at the critical value $(J\rho)_c \approx 0.35$.

$CeRhIn_5$ and $CePd_2Si_2$ under pressure, cluster even more tightly near $J\rho = 0.16$, implying the possibility of a necessary but not sufficient condition on where new examples might be found.

Our results suggest an answer to an interesting piece of the problem of the dense Kondo lattice, which, by inference, provides a route to similar organizing principles in correlated electron materials more broadly defined. The dense Kondo materials are in many ways the simplest highly correlated electron materials, as charge fluctuations are unimportant. In moving from 4*f*- to 5*f*-electron materials, we encounter new low-temperature ground states that condense from the heavy-electron fluid and whose nature remains a mystery. A first step into the physics of these highly correlated electron materials in which charge fluctuations come into play may be the study of how T^* evolves from the simple limiting behaviour described here. Progress in understanding these materials should apply analogously to strongly correlated 3*d*-electron systems, whose physics is in many respects that of 5*f*-electron systems but with correspondingly higher energy scales.

METHODS SUMMARY

For a broad spectrum of heavy-electron materials, we determine three fundamental physical quantities: the local Kondo coupling, *J*, between a single local moment and the background conduction electrons, and the two distinct temperature scales, T_K and T^*, that characterize the magnetic response of the conduction electrons to the introduction of local moments. T_K is the Kondo temperature that governs their magnetic response to a single magnetic impurity, and T^* is the coherence temperature that governs their collective response to a lattice of local moments.

T_K is usually estimated from measurements on dilute impurities in an appropriate non-magnetic host material that typically has lattice parameters which are different from those of the dense Kondo lattice. It is therefore necessary to apply pressure or make chemical substitutions to arrive at isovolumetric values for T_K and *J* in the Kondo lattice material. A knowledge of T_K and the electronic density of states, ρ, which is obtained from the Sommerfeld coefficient, γ, of the host material, then makes it possible to determine *J*.

The emergence of the Kondo liquid at T^* that is accompanied by a loss in strength of the local moments leads to observable physical effects in a number of phenomena. The determination of T^* from these effects, including changes in the optical conductivity, the magnetic entropy, the magnetic resistivity, and point-contact spectroscopy, as well as from the emergence of a Knight shift anomaly, is described in detail in the Supplementary Information.

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Supplementary Information is linked to the online version of the paper at www.nature.com/nature.

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