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Two Fluid Description of the Kondo Lattice

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We present a two-fluid description of the Kondo lattice that is based on an analysis of $CeCoIn_5$ at various levels of dilution with La. We show that thermal and transport measurements provide evidence for the survival of the Kondo impurity component in the lattice as $T \rightarrow 0$ K, and that the evolution of the low temperature properties of the Kondo lattice can be viewed as the partial condensation of the lattice of Kondo centers into a heavy fermion fluid. The resulting two-fluid model is shown to be applicable to the general problem of the ground state of the Kondo lattice.

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Remarkably rich physics is found in the vicinity of the magnetic/nonmagnetic boundary in matter, a region that includes the high T_c superconductors, giant magnetoresistance materials, and especially the heavy electron superconductors [1]. The theoretical framework for the physics of heavy electron materials is the Kondo lattice that has, at high temperatures, localized electronic magnetic moments at each lattice site weakly coupled antiferromagnetically to an itinerant electron band. The single site impurity problem for the coupled electron has been solved to describe the Kondo effect that develops at low temperatures [2,3]. Yet, for the Kondo lattice, because of complex intersite coupling effects, the general form of a solution has not been found despite over 30 years of experimental and theoretical effort.

We present here a two-fluid form for the solution that is derived from an analysis of the bulk specific heat and spin susceptibility of the heavy electron superconductor $CeCoIn_5$, a particularly interesting member of the 115 family ($CeMIn_5$) of Kondo lattice materials that exhibits an unconventional Cooper pairing with T_c of 2.3 K and non-Fermi-liquid behavior due to its proximity to an antiferromagnetic instability [4–9]. We note that $CeCoIn_5$ is a particularly clean material for analysis, having the three important energy scales of the Kondo lattice, the single ion Kondo temperature T_K (1.7 K), the intersite coupling energy scale T^* (45 K), and the crystal electric field (CEF) splitting (120 K) well separated, while the La dilution keeps essentially the same T_K and CEF parameters [9]. We find that, below a crossover temperature of the intersite coupling scale, $T^* \sim 45$ K, one can follow in detail the emergence of a coherent state, the heavy electron fluid, from the lattice of noninteracting Kondo centers. This coherent state is analogous to the superfluid in the two-fluid model for liquid HeII, while the lattice of noninteracting Kondo centers, the "Kondo impurity fluid," is analogous to the normal fluid found in HeII. The relative fraction, f, of the coherent state plays the role of an order parameter; it increases linearly with decreasing temperature until it saturates at 0.9. The resistivity is shown to be simply the product of $(1 - f)$ and that of an isolated Kondo impurity. The generality of this result is suggested by the corresponding analysis for $Ce_{1-x}La_xCoIn_5$ and $CeIrIn_5$.

We begin our analysis at low temperatures by decomposing C_{MAG}, the magnetic f-electron part of the specific heat of $Ce_{1-x}La_xCoIn_5$ alloys (obtained by subtracting the background lattice specific heat, that of $LaCoIn_5$), into two components:

$$C_{MAG}/T = [1 - f(T)](C_{KI}/T) + f(T)(C_{HF}/T), \qquad (1)$$

where C_{KI} is the experimentally measured single ion contribution of the f electrons, C_{HF} is the itinerant heavy fermion fluid contribution, and $f(T)$ is its relative weight. In Fig. 1(a), we plot C_{MAG}/T, normalized per Ce atom, versus C_{KI}/T of isolated Kondo impurity sites obtained from the measured specific heat of $Ce_{1-x}La_xCoIn_5$ alloys at low Ce concentrations ($x > 0.95$). C_{KI} agrees very well with the exact results of $S = 1/2$ Kondo impurity model with the single parameter $T_K = 1.7$ K [3,9]. In this plot, T is an implicit variable and low T corresponds to large values of C_{MAG}/T. By definition, the slope is 1 for the single impurity limit with $x = 0.97$, 0.98. Notably, even with increasing the Ce concentration, the linear relation between C_{MAG}/T and C_{KI}/T holds at low T with a slope decreasing systematically from 1. For $0.25 > x \geq 0$, the slope converges to 0.1, indicating that the variation of C_{MAG}/T at low T is a linear function of the single impurity contribution C_{KI}/T: $C_{MAG}/T = A + BC_{KI}/T$ with $A \approx 290$ mJ/mol-Ce K^2 and $B \approx 0.1$. Figure 1(b) shows that for the entire alloying range an analogous linear relation for the susceptibility, $\chi = C + D\chi_{KI}$, is found between the measured χ and χ_{KI} of isolated Kondo impurity sites, obtained at the single impurity limit. Again for $0.25 > x \geq 0$, the slope D converges to 0.1 with a constant $C \approx 0.008$ emu/mol-Ce. At low temperatures, it thus appears that the ground state has

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two fluids present; one is the residual (10%) single ion component, the Kondo impurity fluid, and the other is the dominant (90%) coherent state, the heavy fermion fluid, brought about by intersite coupling. This indicates that each f electron of the dense Kondo lattice acts as though it were a mixture of 10% single ion Kondo impurity fluid and 90% coherent heavy fermion fluid. We further find that the low temperature Wilson ratio for the heavy fermion fluid, $R_W = \alpha\chi/(C/T)$, where $\alpha = (\pi^2 k_B^2/3\mu_B^2)$, is 2.0, a result identical to that for the Kondo impurity case [2,3].

In general, C_{HF}/T, C_{KI}/T, and f all have temperature dependence. C_{KI}/T is known experimentally, as is C_{MAG}/T. To obtain the temperature dependence of both f and C_{HF}, we first write the magnetic susceptibility in a form analogous to Eq. (1):

$$\chi(T) = [1 - f(T)]\chi_{KI}(T) + f(T)\chi_{HF}(T), \qquad (2)$$

and then make the ansatz (we refer to this as the local Wilson ansatz below) that for all temperatures below T^* the Wilson ratio of the specific heat to the spin susceptibility of the coherent heavy fermion component of Eqs. (1) and (2) is always 2, i.e., $\chi_{HF}(T) = 2C_{HF}(T)/\alpha T$. We can then eliminate χ_{HF} from the two equations and determine $f(T)$ and $C_{HF}(T)/T$ from experiment, with the result shown in Fig. 2 and its insets. In Fig. 2, we see that $f(T)$, which plays a role of an order parameter for the coherent heavy fermion fluid, increases linearly with T as T is reduced below T^*, becoming constant around $T = 2$ K. For $CeCoIn_5$, we find that $f(0) \sim 0.9$ and $T^* \sim 45$ K. As noted above, this continuous increase in the fraction of the coherent heavy fermion part in the homogeneous mixture with the Kondo impurity part is qualitatively similar to the condensation of the normal fluid into the

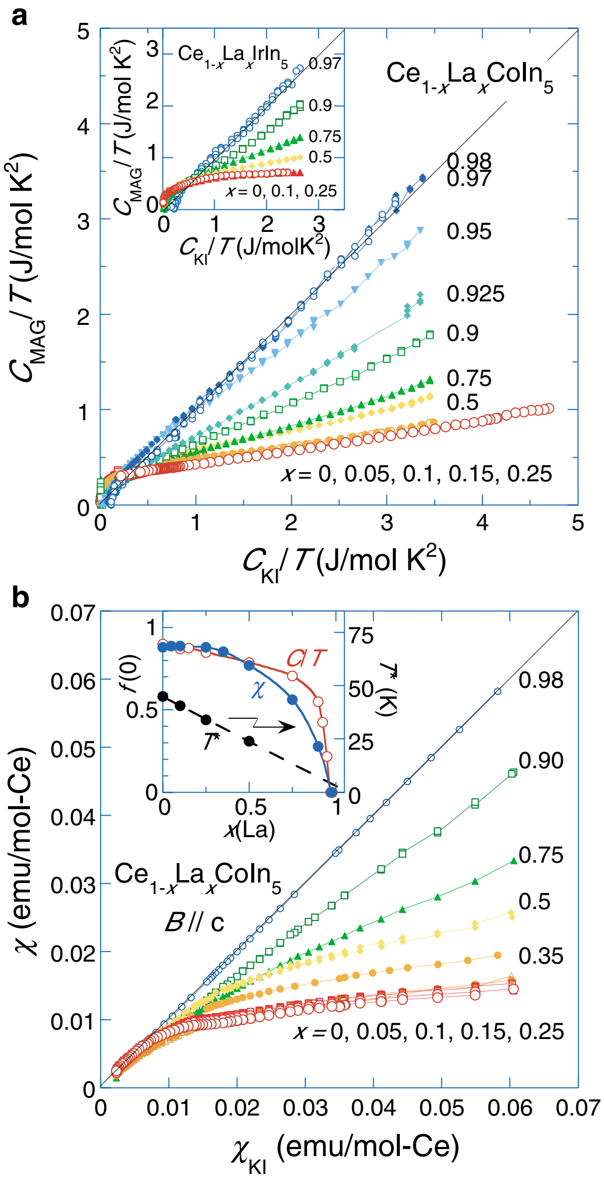

FIG. 1 (color). (a) The f-electron contribution to the specific heat divided by temperature C_{MAG}/T vs its single impurity limit counterpart C_{KI}/T for $Ce_{1-x}La_xCoIn_5$ with temperature as an implicit variable. Inset: The same plot for $Ce_{1-x}La_xIrIn_5$. The specific heat was measured by a relaxation method using high-quality single crystals grown by an In self-flux method [4,9]. (b) χ vs χ_{KI} for $Ce_{1-x}La_xCoIn_5$ with temperature as an implicit variable. Here, χ is the magnetization per mole Ce along the c axis of the single crystals divided by the field, and χ_{KI} is its value in the single impurity limit. The magnetization was measured down to 1.8 K with a quantum design MPMS SQUID magnetometer. Inset: The left axis shows $f(0)$ derived from C_{MAG}/T in (a) (red open symbol) and from χ in (b) (blue solid symbol) for $Ce_{1-x}La_xCoIn_5$. T^* is given on the right axis. The broken line is a linear fit.

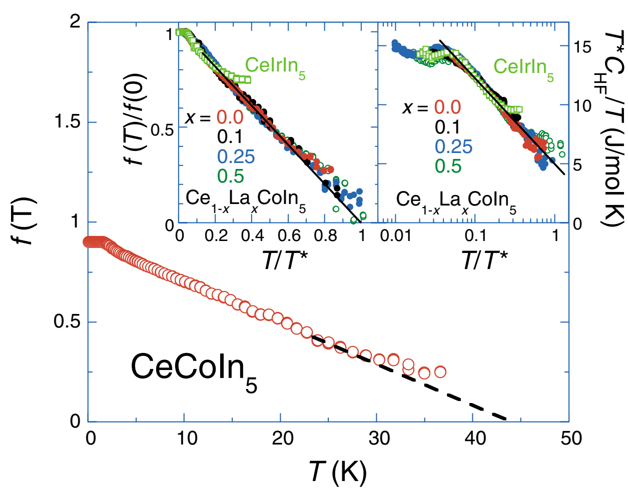

FIG. 2 (color). The relative weight of the heavy fermion component $f(T)$ for $CeCoIn_5$ as a function of T. The broken line shows a linear fit. Left inset: The scaling behavior with T^* of $f(T)/f(0)$ for $Ce_{1-x}La_xCoIn_5$ and $CeIrIn_5$. The solid line represents a fit to the linear increase of $f(T)$ that universally appears below $\sim T^*$. Right inset: T^*C_{HF}/T as a function of $\ln(T/T^*)$ for $Ce_{1-x}La_xCoIn_5$ and $CeIrIn_5$ under $B = 0$ T. Here, C_{HF}/T represents the heavy fermion component of C_{MAG}/T. The solid line is a fit to the $\ln(T/T^*)$ behavior that universally appears for $0.05T^* < T < T^*$.

superfluid in the two-fluid model of HeII. The right inset of Fig. 2 shows the T dependence of the decomposed heavy fermion part C_{HF}/T determined uniquely by our local Wilson ansatz. $C_{HF}(T)/T$, corresponding to an average effective mass, m^*, develops as ln$T$ below T^*.

There are three independent checks on our analysis of $CeCoIn_5$. First, if we make the ansatz that the magnetic part of the electrical resistivity of $CeCoIn_5$ comes solely from the Kondo impurity component, corresponding to that part of electron-electron scattering coming from scattering off the local Kondo centers, one can use our two-fluid decomposition to calculate the resistivity of $CeCoIn_5$. We thus take the measured Ce single impurity resistivity in $LaCoIn_5$, scale this to 100% Ce, and multiply by $1 - f(T)$, which is the time averaged fraction of such centers. We plot without any adjustment this expression and compare with the measured curve in Fig. 3. Experiment is remarkably well reproduced; in particular, the so-called coherence peak is now seen to be due to the loss of the local Kondo centers, as the heavy fermion fluid develops, and the peeling away of the resistivity from the single impurity result reflects the onset of coherent behavior at T^*. That the fit is not perfect may reflect many-body modifications in the scattering at low temperatures.

A second check is provided by calculating the magnetic field response of the magnetization M. While the heavy fermion component should give a linear increase of M, the local moment part should generate the sum of a van Vleck linear part and a Brillouin-type nonlinear response due to the saturation of the local moments. The total magnetization is thus decomposed as $M = fM_{HF} + (1 - f)M_{KI}$, where $M_{HF} = \chi_{HF}H$ and $M_{KI} = g\mu_B\{2a(g\mu_B H) + b\tanh[bg\mu_B H/k_B(T + T_K)]$. Here, $a = 0.0127$ and $b = 0.80330$ are constants for the CEF scheme obtained at the single impurity limit [9]. We can then calculate the field dependence of M without any tuning parameters. The inset of Fig. 3 shows that the experimental results from Ref. [10] are well reproduced by our calculation.

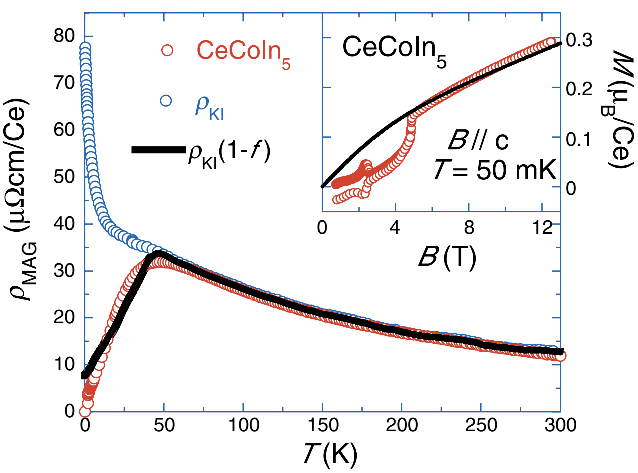

FIG. 3 (color). The magnetic part of the in-plane resistivity ρ_{MAG} defined as the difference of the temperature dependence of the resistivity of $CeCoIn_5$ and $LaCoIn_5$. The red open symbol is for $CeCoIn_5$, while the blue open symbol corresponds to ρ_{MAG} measured at the single impurity limit, ρ_{KI}. The black line is the calculated resistivity, $(1 - f)\rho_{KI}$, for $CeCoIn_5$. Here, $f(T)$ is given in Fig. 2 with its linear extrapolation to 0 around T^*. The resistivity was measured by the standard four-probe dc technique down to 0.4 K. Inset: The field dependence of the c-axis magnetization of $CeCoIn_5$ at 50 mK. The calculated magnetization based on our two-fluid model (black line) is compared with the experimental results (red open circle) presented in Ref. [10]. The agreement is good in the normal state for $B > B_{C2} = 5$ T.

Third, there is strong indirect evidence that, when the Co material goes superconducting, the local moments become part of the superconducting condensate. The usual weak coupling BCS expression for the specific heat jump at T_c is $\Delta C/\gamma T_c = 1.43$. Recognizing that γT_c is the electronic entropy at T_c, we can rewrite the BCS expression as $\Delta C/S(T_c) = 1.43$. Integrating the low temperature entropy out to $T_c = 2.3$ K for $CeCoIn_5$ gives $\Delta C/S(T_c) = 2.2$ with $S(T_c) \approx 0.2R\ln 2$. Given that $S(T_c)$ should have a large contribution from the single ion Kondo center part of about $[1 - f(0)]R\ln 2 = 0.1R\ln 2$, this strong coupling value of $\Delta C/S(T_c)$ indicates that the superconducting ground state incorporates fully the local Kondo center component into the superconducting condensate. This also shows that the local moment part is an intrinsic property of the material.

In order to test the generality of our scheme found for $CeCoIn_5$, we adopt a similar approach in analyzing the experimental results for $Ce_{1-x}La_xCoIn_5$ and $CeIrIn_5$. The inset of Fig. 1(b) shows $f(0)$ for $Ce_{1-x}La_xCoIn_5$ obtained from the linear slope of C_{MAG}/T in Fig. 1(a) and χ in Fig. 1(b). They both increase systematically from 0 with increasing the Ce content from the single impurity limit, converging to essentially the same values for the dense Kondo regime with $x \leq 0.5$. This indicates that it is the intersite coupling that drives the condensation of the lattice of Kondo centers. Interestingly, as shown in the left inset of Fig. 2, when $f(T)$ is normalized by $f(0)$, $f(T)/f(0)$ scales with T^*; it appears around T^* and linearly increases with decreasing T, indicating that T^* should give the condensation energy scale. Scaling behavior with T^* is also found for $C_{HF}(T)/T$; $C_{HF}(T)/T = A/T^* – B/T^* \ln(T/T^*)$ with $A \approx 4.5$ J/mol-Ce K and $B \approx 3.3$ J/mol-Ce K above $T \sim 0.05T^*$, and $C_{HF}(T)/T = D/T^*$ with $D \approx 14.5$ J/mol-Ce K below $T \sim 0.05T^*$, as shown in the right inset of Fig. 2. Interestingly, these scaling functions possess the same form as that found for the single impurity Kondo problem with T_K [2,3]. The T linear dependence of $f(T)$ and the ln$T$ dependence of $C_{HF}(T)/T$ are so robust that they survive up to 50% La dilution. In this range, T^* increases linearly with the Ce concentration, as shown in the inset of Fig. 1(b). Since other characteristic energy scales of the Kondo lattice, T_K and the CEF splitting, are kept constant with La dilution [9], this linear increase in the condensation energy scale

T^* must arise from the intersite coupling. In fact, T^* is basically the same as the energy scale for intersite interactions determined in Ref. [9].

Our analysis carries over to the isostructural heavy fermion superconductor, $CeIrIn_5$, with $T_c = 0.4$ K [11]. We show the plot of C_{MAG} vs C_{KI} in the inset of Fig. 1(a). Again it is seen that the low temperature specific heat of the pure Ir material is linear in that of its dilute Ce counterpart, while T_K and T^* are found to be 2.7 and 20 K, respectively. Thus, the $T \sim 0$ K normal ground state is well described by the sum of Kondo impurity fluid and coherent heavy fermion fluid parts, the latter being here 0.95 of the whole. Furthermore, $f(T)$ and $C_{HF}(T)/T$ scale with T^* at low temperatures and collapse on top of the respective T linear and $\ln T$ behavior obtained for the $Ce_{1-x}La_xCoIn_5$ system, as shown in the insets of Fig. 2. Neutron experiments show that the next higher lying doublet of the $J = 5/2$ $5f$ Hund's rule multiplet lies at 70 K, compared to 120 K for $CeCoIn_5$ [12]. This difference may be the origin of the deviations seen for $(T/T^*) > 0.2$ for the Ir material. It is further worth noting that here again the Kondo impurity fluid remnant at the superconducting T_c is incorporated into the superconducting condensate.

Our picture is one of remarkable simplicity. The Kondo lattice at high T is a gas of noninteracting Kondo centers that starts condensing into a heavy fermion fluid below a characteristic temperature T^* corresponding to the intersite coupling scale. This process represents a redistribution of the spectral weight of the f electrons, a transfer of spectral weight from an incoherent local high frequency part to the coherent itinerant low frequency part. It is well represented by the universal T linear increase of $f(T)$ and $\ln T$ increase of $C_{HF}(T)/T$ that scale with T^* for $Ce_{1-x}La_xCoIn_5$ and $CeIrIn_5$ systems.

Experiment suggests that these 115 materials are located close to an antiferromagnetic quantum critical point [7–9,11,13,14]. Our results indicate that the condensation here of the coherent heavy fermion component is incomplete at the superconducting T_c. The temperature evolution of the remaining Kondo impurity component as $T \rightarrow 0$ K remains hidden. It seems plausible that this condenses into the heavy fermion ground state for T sufficiently below the single ion T_K, albeit giving the excitation spectrum of this heavy fermion ground state unusual features. More generally, where quantum critical behavior is absent, we expect that the heavy fermion component by itself will be a Fermi liquid, a conjecture that is supported by preliminary results on the intermediate valence compound $CeSn_3$ which find that its Fermi liquid ground state can be described by a full condensation ($f = 1$) of the heavy fermion component [15].

Our model differs from the usual qualitative picture of heavy fermion materials in which the dense lattice of f moments is characterized by a "Kondo" temperature appropriate to the lattice and distinct from the single impurity Kondo scale. We see here the survival of the single impurity scale side by side with the lattice scale characterizing the strength of the interaction between the Kondo centers that is responsible for the condensation of the lattice of the Kondo centers. The "coherence" feature so prominent in the temperature dependent electrical resistivity of heavy fermions is found to come simply from the low temperature loss of scattering from the Kondo impurity component.

Our phenomenology suggests the form that the low temperature solution to the Kondo lattice must take. It provides an unexpected framework within which to view the Kondo lattice and heavy electron behavior, and has the potential to provide a new unifying view of correlated electron materials more generally. It provides an experimental algorithm for analyzing the development of coherence in heavy fermion systems, and represents a new tool for investigating quantum critical behavior and the occurrence of superconductivity in highly correlated electron systems.

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