Universal Behavior in Heavy-Electron Materials

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We present our finding that an especially simple scaling expression describes the formation of a new state of quantum matter, the Kondo Fermi liquid (KL) in heavy-electron materials. Emerging at T^* as a result of the collective coherent hybridization of localized f electrons and conduction electrons, the KL possesses a non-Landau density of states varying as $(1 - T/T^*)^{3/2}[1 + \ln(T^*/T)]$. We show that four independent experimental probes verify this scaling behavior and that for $CeIrIn_5$ the KL state density is in excellent agreement with the recent microscopic calculations.

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Scaling behavior in heavy-electron materials was first suggested by Nakatsuji *et al.* [1], hereafter NPF, who found, through a careful analysis of experiments [2] on the thermal and magnetic behavior of the 115 materials, $Ce_{1-x}La_xCoIn_5$ and its Ir counterpart, that these results could be interpreted using a two-fluid model of their specific heat and bulk susceptibility. One fluid, the emergent itinerant heavy-electron component, was characterized by an order parameter, $f(T/T^*)$, that increased with decreasing temperature and scaled with the temperature, T^*, at which itinerant behavior emerged; the second component was a spin liquid, characterized by an order parameter, $[1 - f(T/T^*)]$, and made up of weakly interacting local moments, that at low temperatures could be described by a collection of noninteracting Kondo impurities. The itinerant component possessed two striking properties: its spin susceptibility and specific heat were related by a temperature independent Wilson ratio, R_W, and both exhibited scaling behavior, varying logarithmically as T^*/T. The NPF proposal was subsequently shown to extend to a broad spectrum of heavy-electron materials by Curro *et al.* [3], hereafter CYSP, who showed that the NPF two-fluid model provided a natural explanation for the hyperfine anomaly found in materials for which NMR and μSR measurements of the Knight shift (KS) do not track the measured bulk spin susceptibility. By interpreting its appearance as the onset of two-fluid behavior, their work provided an independent determination of T^* and of the heavy-electron spin susceptibility that emerges below T^*, although for $CeCoIn_5$ (Ce-115), the NMR expression for this quantity differed somewhat from its NPF thermal determination. Taken together, the work of these authors suggests that in a Kondo lattice the hybridization of the localized f-electron magnetic moments with the conduction electrons becomes a collective or global process at T^* that leads to the formation of a new state of quantum matter, the Kondo Fermi liquid, or KL, whose density of states displays distinctly non-Landau scaling behavior between T^* and a typically much lower cutoff temperature, T_0, below which another form of quantum order appears. As we shall see, it coexists there with both the spin liquid and a "light" electron Landau Fermi liquid made of those conduction electrons that do not hybridize with the local moments.

In this Letter, we present the results of an analysis that unites the thermal and magnetic determinations of the parameters that characterize KL behavior and leads to an especially simple scaling description of its density of states. We show that this KL density of states is seen in a number of other experimental probes of heavy-electron behavior, and that, quite remarkably, when our scaling expression for the KL state density is applied to the heavy-electron material $CeIrIn_5$, it yields a state density that is in excellent agreement with the recent microscopic calculations for this material by Shim *et al.* [4]. The agreement provides a double validation—of the methods used in their microscopic approach and the results obtained here using a phenomenological approach—and we discuss its implications for heavy-electron behavior above T_0, and the new quantum ordered states that emerge below T_0.

Our approach to reconciling small differences in the details of the scaling results of NPF and CYSP for the Co 115 material, and obtaining a simple form for its KL density of states, is perhaps best explained by comparing the NPF two-fluid description of the bulk susceptibility:

$$\chi = f(T)\chi_{\mathrm{KL}} + [1 - f(T)]\chi_{\mathrm{SL}}, \tag{1}$$

in which a Fermi liquid contribution from those light conduction electrons that do not hybridize is neglected, and the comparable expression for the Knight shift of a probe nucleus (apart from a constant offset K_0),

$$K - K_0 = Af(T)\chi_{\mathrm{KL}} + B[1 - f(T)]\chi_{\mathrm{SL}}, \tag{2}$$

in which the spin liquid component is coupled to the probe nucleus by a transferred hyperfine coupling constant B, while the itinerant KL component is coupled by a direct hyperfine coupling A. The emergent anomalous component of the KS considered in CYSP is then

$$K_{\mathrm{anom}} = K - K_0 - B\chi = (A - B)f(T)\chi_{\mathrm{KL}}. \tag{3}$$

On combining Eqs. (1) and (3), one gets a very simple formula for the order parameter $f(T)$,

$$f(T) = 1 - \frac{\chi(T) - \alpha K_{\rm anom}(T)}{\chi_{\rm SL}(T)}, \tag{4}$$

in which $\alpha = 1/(A - B)$ and the only unknown quantity is $\chi_{\rm SL}$. The behavior of $\chi_{\rm SL}$ is known in two limits: at high temperatures, $T > T^*$, $\chi_{\rm SL}$ is given by the Curie-Weiss law, $\chi_{\rm CW}(T) \sim (T + T^*)^{-1}$; at low temperatures, $T < T^*/4$, say, because the local f-electron moments are strongly screened by the weakly hybridized conduction electrons, it reduces to a collection of noninteracting single Kondo impurities, an assumption born out by the NPF results. A simple linear interpolation between these limits then yields $\chi_{\rm SL}$, while α is determined by comparing the results obtained with above approach with those found by a self-consistent determination of $f(T)$ based on the NPF approach. The result we obtain for the order parameter,

$$f(T/T^*) = f(0)\left(1 - \frac{T}{T^*}\right)^{3/2}, \tag{5}$$

can be combined with our result for the KL quasiparticle effective mass determined from the specific heat fits,

$$m^*_{\rm KL} = m_h\left(1 + \ln\frac{T^*}{T}\right), \tag{6}$$

seen in $\chi_{\rm KL}$, where m_h is the quasiparticle effective mass at T^*, to yield the reduced density of states for the KL,

$$\rho_{\rm KL} = \left(1 - \frac{T}{T^*}\right)^{3/2}\left(1 + \ln\frac{T^*}{T}\right). \tag{7}$$

Figure 1 shows that our result provides an excellent fit to $K_{\rm anom}$ obtained from NMR experiments for both the 115 family of heavy-electron materials and all others that display a KS anomaly. Note that T^* is anisotropic for materials with anisotropic electronic structures.

Three recent experiments on the 115 materials provide additional evidence for the presence of the Kondo liquid and a description of its density of states using Eq. (7): the anomalous Hall effect, point-contact spectroscopy, and Raman scattering.

Anomalous Hall effect.—Hundley *et al.* [5] have found that the Hall effect in the Ce-115 compounds does not follow the standard skew scattering theory, based on single Kondo impurity scattering, that explains the behavior of many heavy-electron materials [6]. In this theory, the influence of applied external fields on the local Kondo impurities produces a left-right asymmetry that is seen as a skew scattering contribution to the Hall effect. To understand the departure of their results from this theory, Hundley *et al.* introduce the dimensionless function,

$$\alpha(T) = [R^{\rm Ce}_H(T) - R^{\rm skew}_H(T)]/R^{\rm La}_H(T), \tag{8}$$

to describe additional effects originated from the localized Ce ions. Here $R^{\rm Ce}_H(T)$ is the Hall coefficient of the Ce-115 compound and $R^{\rm La}_H(T)$ is the Hall coefficient of the corresponding La compound. Hundley *et al.* find that the skew scattering term $R^{\rm skew}_H(T)$ makes a minor contribution to the total Hall effect and that while $\alpha(T)$ is nearly constant at high temperatures, it increases sharply below a characteristic onset temperature that we identify with T^*. On analyzing their results, we find that $\alpha(T)$ exhibits exactly the following scaling behavior:

$$\alpha(T) - \alpha(300\ {\rm K}) \sim \rho_{\rm KL}(T/T^*). \tag{9}$$

As may be seen in Fig. 2(a), the scaled $\alpha(T) - \alpha(300\ {\rm K})$ for all three Ce-115 materials, even under external pressure [7], falls upon the KL density of states when one takes the onset temperature T^* for the anomalous Hall effect at ambient pressure to be 53 K for $CeCoIn_5$, 31 K for $CeIrIn_5$, and 20 K for $CeRhIn_5$, results that are comparable to the values of T^* for these materials obtained from an analysis of their magnetic and thermal behavior. As shown in Fig. 2(b), similar scaling behavior is also found in $Ce_{1-x}La_xCoIn_5$ [8]. In these materials, the onset temperature of the Hall anomaly again agrees very well with the earlier results from the magnetic susceptibility and the specific heat and displays clearly the linear decrease of T^* with increasing La doping that was found by NPF. We note that while spin-fluctuation scattering is an attractive explanation for the low temperature transport behavior of the 115 materials and has been suggested as the physical

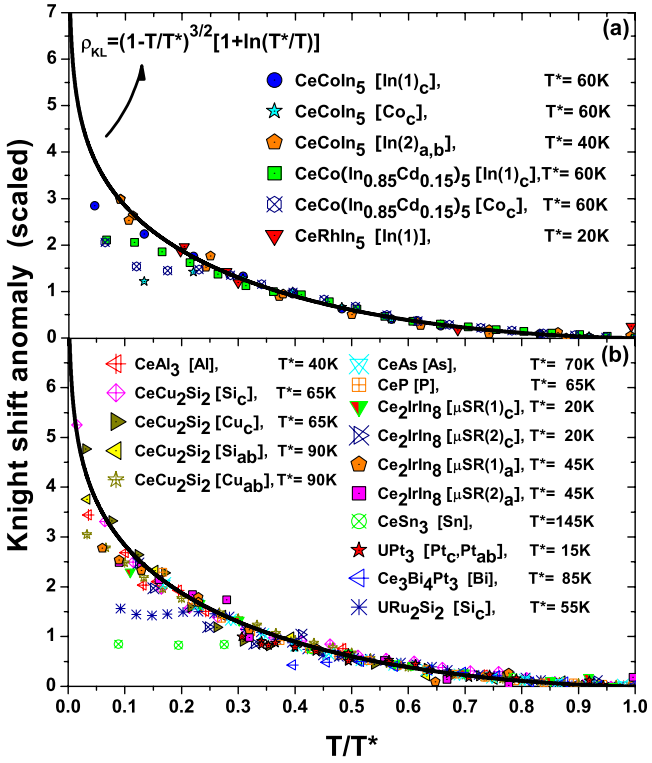

FIG. 1 (color online). Scaling behavior of the anomalous Knight shift for Ce-115 and some other heavy-electron materials (data from Refs. [13] and references in Ref. [3]).

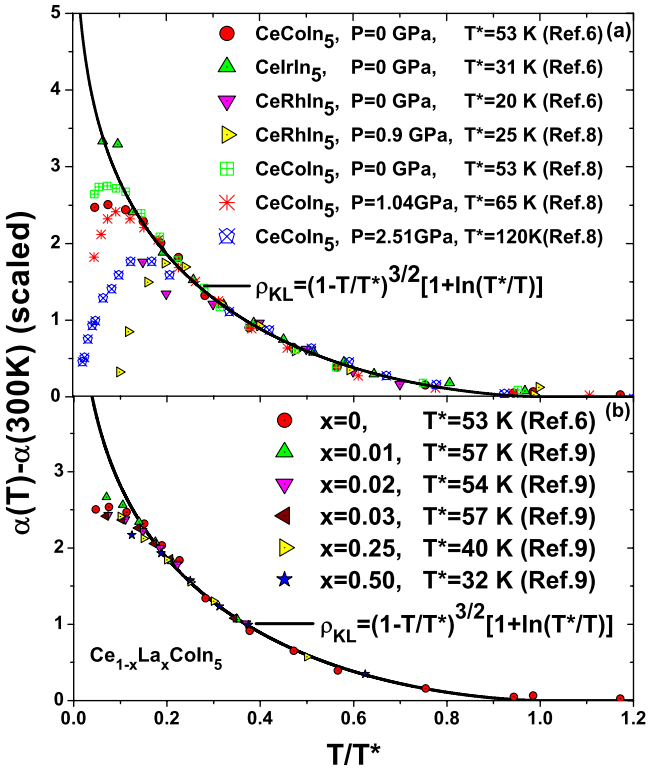

FIG. 2 (color online). Scaling behavior of the anomalous Hall coefficient for (a) Ce-115 materials [5,7] and (b) $Ce_{1-x}La_xCoIn_5$ [8].

origin of the anomalous Hall effect [7], NMR spin-lattice relaxation measurements [9] show that these build up at temperatures that are significantly lower than T^* in these materials; moreover, such a buildup cannot explain the many other physical signatures of KL behavior considered here. We therefore conclude that Hall measurements not only provide a direct measure of the KL state density, but offer a reliable independent estimate of T^*.

Point-contact spectroscopy.—Kondo liquid behavior has also been found to play a role in point-contact conductance. In the recent experiment by Park *et al.* on $CeCoIn_5$ [10], a conductance asymmetry, defined as the ratio of the differential conductance at -2 mV and 2 mV, is found to develop at the characteristic KL temperature T^* and to increase with decreasing temperature down to the superconducting transition temperature T_c (2.3 K). As Curro [11] has pointed out, their measured asymmetry follows the KS anomaly measured by CYSP and as may be seen in Fig. 3, both follow the universal KL density of states for temperatures above the cutoff temperature, $T_0 \approx 10$ K, that marks the end of Kondo scaling behavior, while their mutual agreement continues down to even lower temperature. This tells us that the breakdown of universality does not necessarily indicate the breakdown of the two-fluid description but signals instead a transition of the KL state to some other low temperature quantum state.

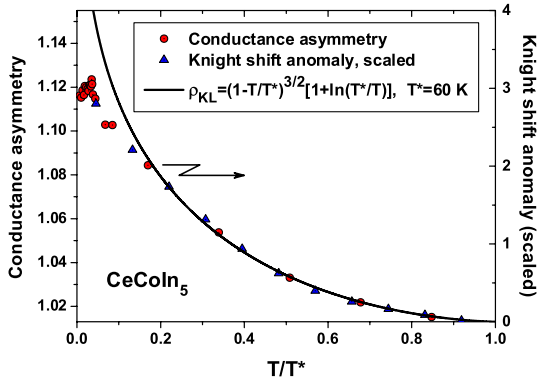

FIG. 3 (color online). Point-contact conductance asymmetry of $CeCoIn_5$ [10] compared to its c-axis KS anomaly and the universal density of states of the Kondo liquid.

Raman scattering.—Very recently, Martinho *et al.* [12] have used Raman scattering to probe the behavior of $CeCoIn_5$. They note that the Fano line shape of the Raman spectral function probes the coupling of phonons to itinerant electrons and that the Fano asymmetry parameter q becomes markedly temperature dependent below a temperature of order T^*. This led us to compare it with our KL state density, with the results shown in Fig. 4. While not as convincing as the anomalous Hall effect or tunneling conductance asymmetry, these results suggest that in the future Raman scattering might offer a useful probe of KL behavior.

At this stage it is natural to inquire what, if any, theoretical basis exists for our phenomenological description of Kondo liquid behavior. The answer comes in the very recent work of Shim *et al.* [4], hereafter SHK, who carry out a first principles calculation of the localized to itinerant transition in $CeIrIn_5$ using dynamical mean field theory in combination with local density approximation (LDA + DMFT). In Fig. 5 we compare their calculated density of states at the quasiparticle peak with our universal KL

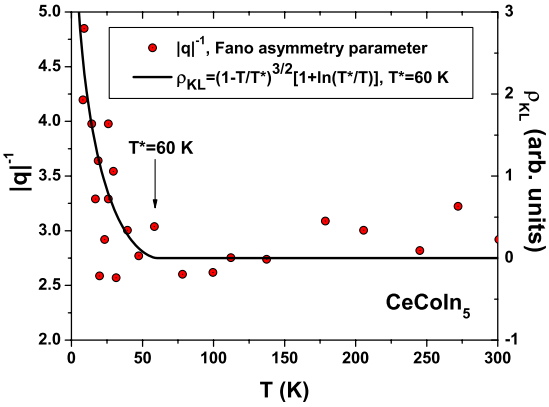

FIG. 4 (color online). Fano asymmetry parameter $|q|^{-1}$ from the Raman spectra of $CeCoIn_5$ [12] compared to the Kondo liquid density of states.

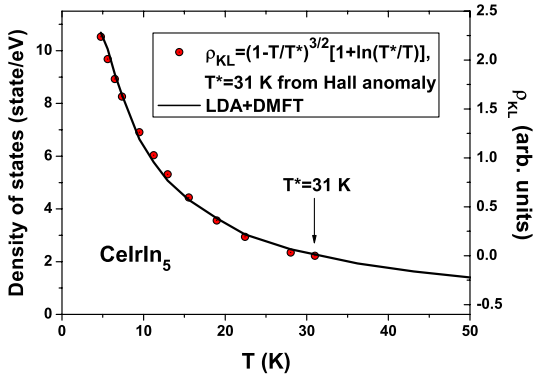

FIG. 5 (color online). LDA + DMFT density of states of the quasiparticles in $CeIrIn_5$ [4] compared to the KL density of states. We take the onset temperature $T^* = 31$ K from the Hall anomaly.

density of states; T^* is chosen to be 31 K, as suggested by the anomalous Hall data. Since in the SHK result there is no clear onset temperature at which the density of states starts to increase, we simply compare the low temperature ($T < T^*$) portions and find a linear relation between them.

The excellent agreement between the two results indicates that SHK have captured the Kondo liquid state density without having to resort to a cluster calculation below T^*, a truly remarkable finding that provides quite strong evidence for the validity of both their microscopic approach to hybridization and our phenomenological scaling expression for the KL state density. Since there is no experimental evidence for a change in the specific heat at the hyrbridization phase transition (i.e., nothing special happens at T^*), and since local hybridization can influence the density of states up to very high temperatures, it is to be expected that an exact calculation of the state density would do just what their calculation does, i.e., go smoothly through T^* and yield our scaling results below T^*.

It is useful to make a distinction between the local (i.e., single Kondo impurity) hybridization that must begin at temperatures well above T^* and the universal global hybridization behavior that begins at T^*. The local hybridization is expected to be highly anisotropic and complex because the Kondo coupling, J, of the single Kondo impurities to different parts of the Fermi surface will be different; this is just what SHK find. T^* is the temperature at which, as a result of feedback effects on the coupling between individual hybridization channels, hybridization becomes a ''simple'' collective coherent universal phenomenon, characterized by an order parameter, $f(T/T^*)$, that goes as $(1 - T/T^*)^{3/2}$, and a logarithmic increase in the average effective mass, $m^*_{\mathrm{KL}} = m_h[1 + \ln(T^*/T)]$, where m_h incorporates the hybridization occurring above T^*. We further note that NPF argue that the physical origin of the energy scale T^* that characterizes KL behavior is the nearest neighbor coupling between the localized electrons, a view that is supported by its appearance as the Curie-Weiss parameter in the high temperature susceptibility for $CeCoIn_5$ and other materials in which crystal field effects play out at substantially higher temperatures.

We conclude that once one has identified T^* for a given heavy-electron material, the interacting components necessary to understand its transport behavior at all temperatures as well as the appearance of quantum order below T_0 can now be specified. There will in general be three: a heavy-electron non-Landau Kondo Fermi liquid whose quasiparticles have the temperature dependent effective mass given by Eq. (6), a light-electron Landau Fermi liquid corresponding to electrons on those parts of the expanded Fermi surface that have not hybridized individually or collectively, and a residual spin liquid made up of the screened interacting local moments. So to understand the behavior of heavy-electron materials between T^* and T_0 one needs first of all to combine our scaling description of the Kondo liquid behavior and the two-fluid model to follow the temperature evolution of all three components, while it is their mutual interaction, in the presence of global hybridization, that determines transport properties above T_0, and the nature of the ordered state below T_0.

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