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Experimental evidence for predicted universal behavior in low-temperature Kondo lattices

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We have previously shown that the $1/N$ expansion of the Kondo lattice leads to the following predictions concerning the heavy-Fermi-liquid state: (i) the low-temperature specific heat behaves as $C_V = \gamma T \propto T/\bar{T}_K$ with corrections $\Delta C_V = (T/\bar{T}_K)^3 \ln(T/\bar{T}_K)$, (ii) the zero-temperature spin susceptibility $\chi \propto 1/\bar{T}_K$, and (iii) the resistivity $\rho \propto (T/\bar{T}_K)^2$. These results all contain a unique energy scale $\bar{T}_K$. Here we analyze recent pressure-dependent specific-heat measurements on UPt_3 combined with χ and ρ data to confirm the scaling of these quantities with one strongly pressure-dependent energy scale γ^{-1}. This picture is further supported by evidence of systematic scaling of χ and ρ with γ throughout the entire class of heavy-fermion compounds. This behavior, along with other experimental observations, suggests that the low-T properties of UPt_3 do *not* derive from ferromagnetic spin fluctuations.

It is the purpose of the present paper to demonstrate the existence of a single pressure- (p) dependent energy scale in the low-temperature (T) state of the heavy-fermion system UPt_3. We analyze a variety of p-dependent data and show that the $T^3 \ln T$ contribution to the specific heat,[1] the spin susceptibility[2] χ, and the resistivity[3] ρ are all given in terms of their zero-pressure values by the ratio $(\gamma/\gamma_{p=0})^n$, where $n = 3$, 1, and 2, respectively. Here γ is the usual linear coefficient of the specific heat. This behavior is in accord with what is predicted[4] by a $1/N$ expansion of the Kondo lattice, and does not support the ferromagnetic-spin-fluctuation picture. It should be stressed that the low-T state we focus on is in the "coherent" regime which prevails only at the lowest temperatures. The notion that there is a single energy scale must necessarily break down as the temperature becomes comparable to the higher characteristic energy parameters of these systems.

In addition to this pressure-dependent scaling in UPt_3 there are a number of universal features associated with the coherent Fermi-liquid state in the general class of heavy-fermion systems. (i) The dimensionless ratio of the linear specific-heat contribution γ and susceptibility χ is close to the free-electron value of unity.[5] (ii) The specific heat[6] C_V often exhibits a rapid downturn with increasing temperature which in some cases has been fit to a function of the form $T^3 \ln T$. (iii) The resistivity[7,8] ρ behaves as T^2. (iv) The low-T susceptibility[1,8] χ_T appears to vary as T^2. What provides perhaps the most striking evidence of universal behavior in the heavy-fermion systems is Kadowaki and Woods'[7] observation that the coefficient $A \equiv \rho/T^2$ is the same multiple of γ^2 in essentially all materials.

It is generally assumed[1,3] that the presence of a $T^3 \ln T$ term in C_V, and T^2 contributions to ρ and χ_T are signatures of strong spin-fluctuation effects. However, we wish to stress here that *all these features arise naturally when a $1/N$ expansion is used to treat the Kondo lattice.* The similarity of the $1/N$ formalism and the spin-fluctuation problem is not surprising in view of the fact that both theories involve fermion-fermion interactions which take place by exchange of a screened low-energy boson. However, the microscopic physics of the two systems is very different.

On general grounds it can be shown that the existence of a boson with low characteristic energy gives rise to the "universal" features (ii)–(iv) listed above. In particular, appreciable $T^3 \ln T$ contributions to the specific heat are expected for any Fermi liquid in which the interaction between the quasiparticles is mediated by a screened boson with propagator $D(q,\omega)$ which depends on $(\omega/T_0)(k_0/q)$. Here T_0 and k_0 are the characteristic energy and wave number of the interaction. This low energy can derive from, but need not necessarily be associated with, a magnetic tendency. By contrast, a simple one-band Fermi liquid will generally not exhibit a T^2 term in the resistivity since in such systems electron-electron interactions cannot dissipate current. In the cases where an appreciable T^2 term is observed, a two-band model and/or umklapp processes are assumed to be important. Here the lighter electrons which carry the current scatter from the electrons in the heavy band or from the screened boson.[8] Umklapp processes which break down momentum conservation are important, at the very least, because they lead to separate equilibration of the boson and the current carriers. This same boson will give rise to a T^2 term in χ_T although there may be other physical origins of this effect.[8]

Thus the existence of T^2 contributions in ρ and χ_T and the $T^3 \ln T$ term in C_V does not provide detailed information about the quasiparticle interactions. It is our contention that it is the dependence of the various thermodynamic and transport properties on the linear coefficient of the specific heat γ which helps most to differentiate between the various microscopic mechanisms which govern the interactions in the Fermi liquid. Thus the observations that $A \propto \gamma^2$ and $\chi \propto \gamma$ discussed above are most significant. General Fermi-liquid considerations do not dictate a simple dependence of χ, A, δ, χ_T, etc. on γ. For example, χ depends on γ as well as on an additional (enhancement) parameter $1/(1+F_0^a)$. Thus as a function of pressure, χ is not expected to scale only with γ. Nor is this observed to be the case in the prototypical Fermi liquid ^{3}He. In the paramagnon model γ is inversely proportional to the Fermi energy and depends logarithmically on the spin-fluctuation energy. However, A, δ, etc. vary as powers of the spin-fluctuation temperature. By contrast in the Kondo-lattice picture a single energy parameter γ^{-1} sets the scale for the characteristic "coherence temperature." The Landau parameters such as F_0^a are essentially independent of γ. Other thermodynamic and transport variables then depend on simple powers of γ only.

In order to differentiate between the bosons of the $1/N$ expansion (called "Kondo bosons") and paramagnons, we have studied a variety of pressure-dependent data in UPt_3 as functions of γ. In Fig. 1 are plotted the pressure-dependent spin susceptibility,[2] the T^2 coefficient in the resistivity[3] A, and the coefficient δ for the $T^3 \ln T$ contribution[1] to C_V as functions of $\gamma/\gamma_{p=0}$. These quantities are all normalized to their zero-pressure values. The solid lines correspond to the functional dependences $(\gamma/\gamma_{p=0})^n$. Here the values of γ are obtained by interpolating the data[1] over the entire pressure regime. This interpolation is reasonably straightforward since $1/\gamma$ appears to vary linearly with p in this pressure interval.[9] As can be seen $n=1,2$, and 3 fit the data extremely well for χ, A, and δ, respectively. The only appreciable deviation from the solid curve is the highest-pressure value of δ which may be in error by as much[10] as a factor of 2.0. The data suggest that UPt_3 can be described by a universal energy scale $\bar{T}_K(p)=\gamma^{-1}$ such that $\chi \propto \gamma$, $\delta \propto \gamma^3$, and $A \propto \gamma^2$. It should be noted that the fact that χ and A scale with the same characteristic temperature has been previously recognized.[3] The recent specific-heat measurements of Brodale *et al.*[1] allow us to deduce that γ, δ, χ, and A all are determined by this same temperature scale.

In Fig. 2 are replotted χ and A data[5,7] for a variety of heavy-fermion systems (at ambient pressure) also in terms of the parameter γ. Again the scaling of χ and ρ/T^2 with γ and γ^2, respectively, is quite striking. Figures 1 and 2 together suggest that there exists a kind of universality associated with the low-temperature coherent heavy-Fermi liquid.

To gain theoretical insight into these results we summarize our earlier work[4] on the Kondo lattice with N degenerate levels at each site and fractional occupation Q_0. The essential bare parameters of the theory are J, the antiferromagnetic exchange between conduction and valence electrons, and $N(0)$, the conduction-band density of states. Here the Kondo limit $N(0)J \ll 1$ is assumed. The quasiparticles are described by a mean-field ($N=\infty$) renormalized band structure[11] so that to leading order

$$\chi \propto \gamma \; , \qquad (1)$$

where

$$\gamma = \pi^2 N/(3T_k) \approx N(0)\exp[1/JN(0)] \; . \qquad (2)$$

In the Kondo limit the f charge fluctuations are suppressed by a factor which varies as T_k. In Ref. 1 the Grüneisen parameters have been shown to be anomalously large, e.g., $\Gamma_\gamma = d\ln\gamma/d\ln V = 57$. Equation (2) provides microscopic understanding of this observed large volume and pressure sensitivity of γ through the exponential dependence on $N(0)$. Because the Fermi level lies in a narrow peak in the density of states, mean-field theory already accounts for the large enhancements observed in γ and χ. We have shown[4] how the leading order interactions between the mean-field quasiparticles are mediated by the Kondo boson (KB) propagator[12] which though formally resembling the dressed random-phase-approximation (RPA) susceptibility of paramagnons represents distinctly different physics. The KB propagator consists of coherent hybridization fluctuations between conduction and nearly localized f electrons dressed by quasiparticle density fluctuations. The $O(1/N)$ corrections[13] to γ/χ are independent of T_K and thus are not strongly pressure dependent. In our $O(1/N)$ calculation, the energy scale $\bar{T}_K$ emerges from the KB propagator frequency dependence, where $\bar{T}_K = Q_0 T_k$. Thus we have found that[14]

$$\delta \propto \gamma^3 \; , \qquad (3)$$

and the coefficient of the T^2 term in the resistivity was found to scale as

$$A \propto \gamma^2 \; . \qquad (4)$$

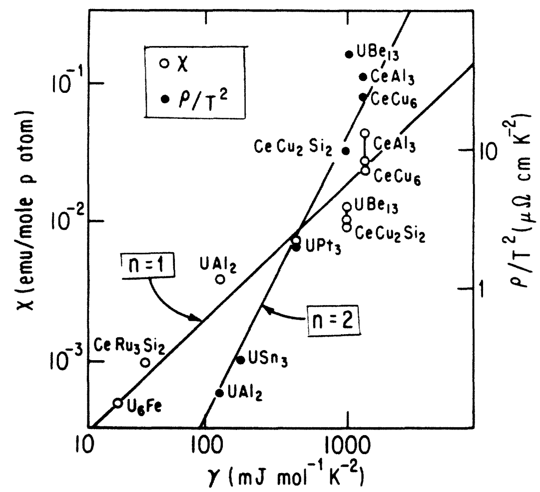

FIG. 2. Universal ratios in the heavy-Fermion compounds. χ/γ data are from Ref. 5 and $A=\rho/T^2$ data are from Ref. 7. The solid lines are theoretical results of Eqs. (1) and (3).

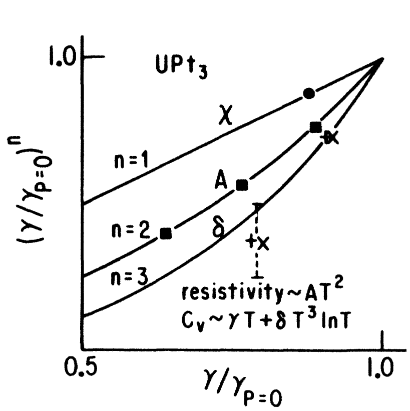

FIG. 1. Scaling of thermodynamic and transport coefficients with pressure-dependent γ. (For the latter, see Ref. 1.) χ are from Ref. 2, resistivity from Ref. 3. The symbols + and × correspond, respectively, to the coefficient δ of $T^3\ln T$ term in C_V and the coefficient ϵ of T^3 term in C_V (from Ref. 1). For the latter see Ref. 14.

It can also be shown that the temperature-dependent susceptibility varies as

$$\chi_T \propto T^2 \gamma^2 \ . \qquad (5)$$

$\bar{T}_K$ can therefore be labeled the "coherence" energy scale of the Kondo lattice. It should be stressed that this Fermi-liquid energy scale is simply related to the mass enhancement $\bar{T}_k \sim \gamma^{-1}$. Equations (1), (3), and (4) are represented by the solid lines in Fig. 1. It is clear that $\bar{T}_K$ governs the dynamics at low frequencies and temperatures. The details of the bare band structure, finite f-charge fluctuations, and crystal-field splitting all enter at higher-energy scales. The surprisingly general behavior in a wide variety of heavy-fermion systems evident in Fig. 2 also corresponds to the predictions of the Kondo-Anderson-lattice Hamiltonian [Eqs. (1) and (4)].

It is on the basis of the recent pressure-dependent specific-heat measurements of Brodale *et al.*[1] that one can construct, perhaps the strongest evidence against the standard spin-fluctuation picture of UPt_3. In the simple paramagnon model[8] the linear coefficient of the specific heat γ depends on the Fermi energy T_F as well as logarithmically on the characteristic boson energy T_{SF}, whereas A, χ, and δ vary with $(T_{SF})^{-n}$, where $n=2$, 1, and 3, respectively. Thus Eqs. (1), (3), and (4), which evidently agree with experiment, are not consistent in any simple way with the paramagnon picture. Brodale *et al.* have argued that their pressure-dependent specific-heat data can be fit within the paramagnon model provided all the pressure dependence enters through the "bare" Fermi temperature T_F, so that the enhancement factor is p independent. It should be stressed that these observations of pressure-independent enhancement factors are not easily reconciled with the picture that UPt_3 is on the verge of a ferromagnetic instability. Presumably if the Fermi temperature T_F is strongly pressure sensitive, then the enhancement factor, particularly when it is large, would be even more so. An equivalent or similarly artificial assumption is needed to explain why in a nearly ferromagnetic system the dimensionless ratio of χ/γ is of order unity not only at ambient pressures but up to 4 kbar. As expected, in the Fermi-liquid state of ^3He, where spin fluctuations are known to be important, Eqs. (1) and (3) are in clear disagreement with pressure-dependent measurements.

It is important to note that on the basis of the magnitude of the measured χ/γ and $A\gamma^{-2}$, UPt_3 does not appear to be particularly distinct from the other heavy-fermion compounds (see Fig. 2). This rather strikingly general behavior would appear to be at odds with spin-fluctuation theories which single out UPt_3 from the other heavy-fermion compounds. Other experiments which do not support the ferromagnetic spin-fluctuation picture for UPt_3 are neutron[15] and magnetoresistance measurements[16] which yield evidence for antiferromagnetic spin fluctuations. It is known[17] that these shorter-wavelength fluctuations do not give rise to significant $T^3 \ln T$ contributions to the specific heat. This observation strongly suggests that the $T^3 \ln T$ term in UPt_3 derives from a low characteristic energy associated with the coherence temperature. It is interesting to observe that the canonical[6,18,19] "spin fluctuators" UPt_3,UAl_2 and (sometimes) USn_3, which cluster together in Fig. 2, all correspond to heavy-fermion systems with intermediate values of γ (ranging from about 100 to 500 in units of $\mathrm{mJ\,mole^{-1}K^{-2}}$). It is in these materials that the coherence temperature $\bar{T}_K$ is at an "optimal" value so that there is a substantial range of temperatures below $\bar{T}_K$ over which coherence effects are observable. At the same time $\bar{T}_k$ is not so large that it exceeds any nonuniversal energy scale (e.g., crystal fields or f-charge fluctuation) γ is not so low that these many-body effects are completely negligible. For the higher-γ systems $\bar{T}_K$ may be too small so that phase transitions often obscure the effects of the coherent Fermi liquid. It, therefore, may be no accident that these three uranium compounds most clearly display the effects of low-characteristic-energy bosons, effects which have previously been attributed to spin fluctuations.

In summary, the pressure-dependent scaling in UPt_3 of χ, A, and δ with $\gamma^{-1} \sim \bar{T}_K$ is evidence in favor of the Kondo-boson picture in the lattice. This boson which describes fluctuations in a "ferromagnetically inert" system is distinct from the boson of spin-fluctuation theory. Additional resistivity and χ/γ measurements suggest that at low T, UPt_3 is qualitatively similar to most other heavy-fermion systems. To further test this model it is essential that similar low-temperature, pressure-dependent studies be made on other (normal-state) heavy-fermion compounds. Temperature-dependent magnetization studies are needed to test Eq. (5). Additional pressure-dependent data on UPt_3 are also desirable.

Finally, it should be noted that in the context of superconducting pairing the fact that this boson is not associated with spin fluctuations suggests that the pairing interaction will not necessarily be in the triplet channel. Rather the pairing will take place in the (presumably lowest) angular momentum channel 1 for which the boson-mediated interaction is attractive. Within the context of a weak-coupling theory (which is probably not adequate[4]), this can be shown to correspond to the l for which A_l^s is first positive.[20] In this rather oversimplified theory, this cannot occur[4] for l equal to 0 or 1. The first allowed pairing would then be in the d channel. Arguments in favor of $l=2$ pairing have been presented elsewhere.[21]

Note added in proof: A. J. Millis and P. A. Lee have independently computed γ, χ, and ρ by a similar approach using the Cartesian Bose coordinates. Their results agree to leading order with ours.

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[13]It should be noted that there appears to be a systematic deviation of γ/χ from the value of the Wilson ratio $R=1$. This systematic deviation is consistent with the results of the $1/N$ expansion discussed in Ref. 4.

[14]There also exists a εT^3 term in the specific heat which includes corrections from variations of the saddle point parameters which were not calculated.

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