Observation of Non-Fermi-Liquid Behavior in $U_{0.2}Y_{0.8}Pd_3$

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We present data for the temperature and magnetic-field dependences of the specific heat, magnetic susceptibility, magnetization, and electrical resistivity of $U_{0.2}Y_{0.8}Pd_3$. The data demonstrate the scaling behavior of the thermodynamic quantities at temperatures up to 10 K and magnetic fields up to 14 T. The magnetic field scales as $T^{1.3}$. The zero-field specific heat varies with temperature as $-T\ln T$. The magnetic field strongly affects the thermodynamics, but the magnetoresistance is very small (-2% at $T=0.36$ K and $H=14$ T). We argue that our data are inconsistent with any single-impurity interpretation. We suggest that the system has a second-order phase transition at $T=0$.

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The cornerstone of the Fermi-liquid theory is the hypothesis that the Hilbert space of low-lying eigenstates of an interacting system is isomorphic with the Hilbert space of the noninteracting electron gas. The immediate consequence of this hypothesis is that low-temperature properties of an interacting system resemble those of the noninteracting Fermi gas: weakly temperature-dependent positive magnetic susceptibility, a specific heat linear in temperature, a well-defined quasiparticle peak in the imaginary part of the one-electron Green function, etc. Interactions reveal themselves in a renormalization of various coefficients and also in small deviations from the free-electron behavior: The quasiparticle peak at energy ϵ acquires a finite width $\gamma(\epsilon)(\lim_{\epsilon\to 0}\gamma/\epsilon=0)$, the electrical resistivity has the following temperature dependence, $\rho=\rho(0)+aT^2+\cdots$, etc. It is legitimate to say that the concept of the Fermi liquid implies that all multiparticle correlations are short ranged in both space and time. This condition is present in the derivation of the Fermi-liquid relations as the suggestion that a two-particle vertex Γ^{ω} is a smooth function of frequencies and momenta (see, for example, Refs. [1,2]). Long-range correlations between particles are known to break the Fermi-liquid behavior (the first realistic model was found in Ref. [3]).

Long-range correlations are present if a system has a second-order phase transition; at the transition point the correlations become infinitely long ranged. Therefore, if a metallic system has a phase transition at $T=0$ then its ground state is not isomorphic with the free-fermion-gas ground state. At $T=0$ the low-lying states are scale invariant; at a nonzero temperature the only scale is the temperature itself. The experimental indications of the second-order phase transitions are nonanalytic temperature and field dependences of physical quantities and, first of all, their scaling. (We remind the reader that exactly that happens in one-dimensional models of interacting electrons like the Hubbard model which has an antiferromagnetic transition at $T=0$; it is well known that the Fermi-liquid theory never works in one dimension.)

In this Letter we present the experimental data for the specific heat, magnetic susceptibility, magnetization, resistivity, and magnetoresistivity of the pseudobinary uranium cubic compound $U_{0.2}Y_{0.8}Pd_3$. These data clearly demonstrate nonanalytic temperature and magnetic-field dependences of the measured quantities and also show the scaling. We argue that this indicates a second-order phase transition taking place in this system at $T=0$.

The first zero-magnetic-field data, showing unusual $T\ln T$ dependence of the specific heat, were obtained for this compound by Seaman *et al.* [4].

Subsequently, we synthesized a compound of the same nominal stoichiometry. According to the x-ray-diffraction analysis, the investigated material is a single phase with the $AuCu_3$-type crystal structure and a lattice constant of 4.074 Å. The magnetic-susceptibility and specific-heat data presented in this Letter are normalized to U mole after subtracting the corresponding quantities for YPd_3, a nonmagnetic analog of $U_{0.2}Y_{0.8}Pd_3$.

Our specific-heat data are presented in Figs. 1 and 2. We show that there are two different regimes in the

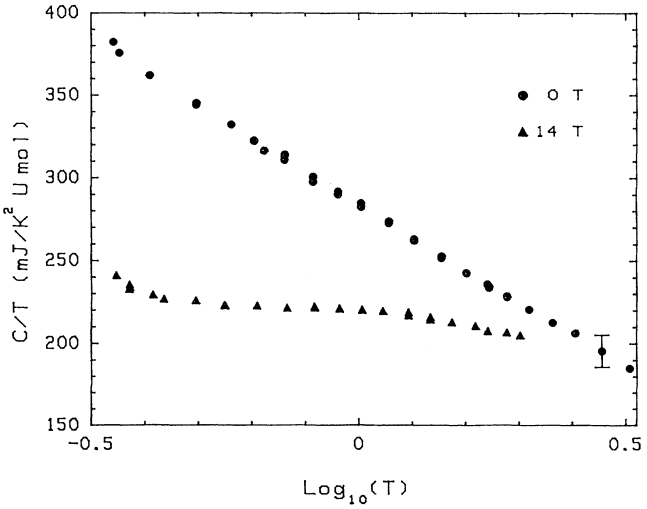

FIG. 1. Specific heat divided by temperature (C/T) vs $\log_{10}T$ in 0- and 14-T magnetic fields for $U_{0.2}Y_{0.8}Pd_3$. The absolute error of C (mainly instrumental error) is about the same for all measurements (5%) and therefore does not significantly affect an analysis of the data.

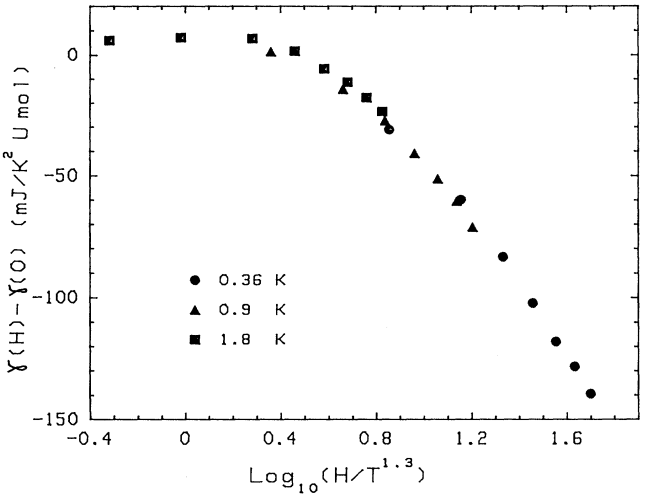

FIG. 2. The crossover function $C(T,H=0)/T - C(T,H)/T$ vs $\log_{10}(H/T^{1.3})$ measured at 0.36, 0.9, and 1.8 K.

specific heat:

$$C(T,0)/T = -1.3A\log_{10}T + B\,, \qquad (1)$$

$B = 480\ \mathrm{mJ/K^2(U\,mol)}$, $A = 150\ \mathrm{mJ/K^2(U\,mol)}$,

and

$$C(T,H)/T = A\log_{10}H + D \quad (H \gg T)\,, \qquad (2)$$

$D = 410\ \mathrm{mJ/K^2(U\,mol)}$,

with a crossover between them,

$$\frac{C(T,H)}{T} - \frac{C(T,0)}{T} = f\left(\frac{H}{T^{\beta}}\right), \quad \beta = 1.3 \pm 0.05\,, \qquad (3)$$

given by the function $f(x)$ represented in Fig. 2.

The ratio of slopes, $[\gamma(T,0)/\log_{10}T] : [\gamma(0,H)/\log_{10}H]$, is equal to 1.3 and coincides with the scaling dimension of the magnetic field as it should for any crossover.

The magnetic susceptibility and magnetization were measured with a SQUID magnetometer at temperatures down to 1.8 K and in fields up to 5.5 T. At low temperatures and in low fields [Fig. 3(a)] the susceptibility has a power-law temperature dependence:

$$\chi = T^{-\eta}, \quad \eta = 0.3 \pm 0.05\,.$$

The magnetization data, presented in Fig. 3(b), also reveal the scaling:

$$M/H = T^{-\eta} g(H/T^{\beta}), \quad \eta = 0.3, \quad \beta = 1.3\,. \qquad (4)$$

This scaling is less perfect than the scaling for the specific-heat data. This in part, at least, can be attributed to the high sensitivity of magnetization to always unavoidable small inclusions of secondary phases and magnetic impurities. Nevertheless, β determined from the specific heat and η from the susceptibility data give the best description of the magnetization in the form of Eq. (4). A change of η by 0.05 or β by 0.2 in either direction results in a much larger spread of data points than in Fig. 3(b). Therefore, the magnetization results serve as an important check point for the self-consistency of the postulated scaling.

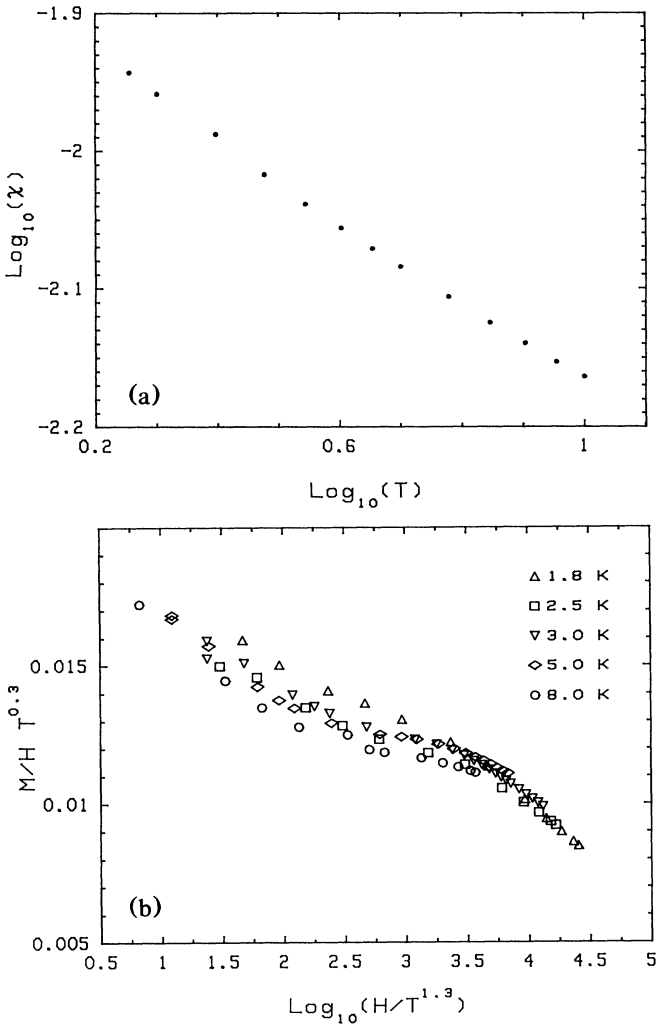

FIG. 3. (a) $\log_{10}(\chi)$ vs $\log_{10}(T)$ for temperatures $1.8 \leq T \leq 10$ K and in $H = 0.1$ T. χ is expressed in emu/(U mol) and T in K. (b) The magnetization $M/HT^{0.3}$ vs $\log_{10}(H/T^{1.3})$. M/H is expressed in emu/mol, H in Oe, and T in K.

In principle, a scaling is possible for single-impurity systems, but we argue that the observed phenomenon is not of a single-impurity nature. In fact, there is only one single-impurity theory which predicts a low-temperature scaling: the multichannel Kondo model formulated by Nozières and Blandin [5]. Later, Cox provided arguments that this model describes diluted uranium alloys [6]. This model was used in Ref. [4] to explain the observed $T\ln T$ specific heat of the $U_{0.2}Y_{0.8}Pd_3$ compound.

The fact that the magnetic field has a scaling dimension greater than 1 is the first indication that any single-impurity interpretation is not valid in this case; single-impurity fixed points have scaling dimensions smaller than 1. For any single-impurity theory the scaling dimension β is related to the scaling dimension of the spin operator, Δ, in the following way: $\beta=1-\Delta$ [7]. The form of spin-spin correlation functions, $\langle S(t)S(0)\rangle=1/|t|^{2\Delta}$, restricts Δ to values greater than zero.

The data disagree with the single-impurity theory also in the following important point. According to this theory, an application of a magnetic field (or, in fact, of any relevant field) should initially lead to a sharp increase of the specific heat and only when the field is strong enough ($H\gg T^{\Delta}T_K^{1-\Delta}$; Δ is its scaling dimension and T_K is the Kondo temperature) should the specific heat decrease. The physical reason for this scenario is that in the absence of external relevant fields the ground state of the multichannel Kondo model possesses some finite entropy. The applied field destroys the non-Fermi-liquid fixed point and turns the scaling trajectory towards the strong-coupling fixed point where this residual degeneracy is absent. Naturally, to consume the entropy difference between the two ground states the system should increase its specific heat. The corresponding calculations for the two-channel Kondo model where performed by Sacramento and Schlottmann [8]. It is clear that such a subtle feature as the residual ground-state degeneracy can suffer from an interimpurity interaction. So, the absence of the discussed feature is a clear indication that this interaction is important.

The electrical resistivity of $U_{0.2}Y_{0.8}Pd_3$ (Fig. 4) is larger by 2 orders of magnitude than that of pure YPd_3,

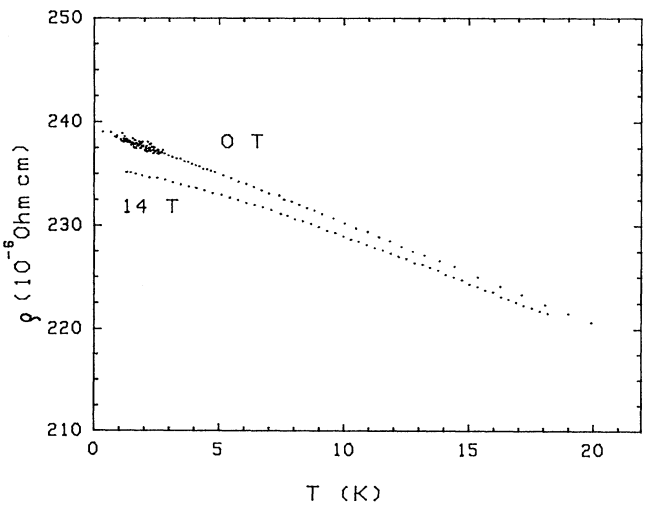

FIG. 4. Electrical resistivity of $U_{0.2}Y_{0.8}Pd_3$ measured in $H=0$ and $H=14$ T fields and at temperatures from 0.36 to 20 K. The absolute error of ρ is $\pm 25\%$ due to unfavorable geometry of the resistivity sample.

and the scattering cross section is close to the unitary limit. The resistivity is approximately linear in T:

$$[R(0)-R(T)]/R(0)\sim T\,.$$

The influence of the 14-T magnetic field is extremely weak:

$$\frac{R(T=0.36\text{ K},H=0)-R(T=0.36\text{ K},H=14\text{ T})}{R(T\approx 0.36\text{ K},H=0)}\approx 0.02\,.$$

At the same time, the similar ratio for the specific heat is about $\frac{1}{3}$. This again implies a departure from the single-impurity situation: The magnetic field has a different influence on the transport properties and the thermodynamics.

If our data, as we suggest, do correspond to a phase transition at $T=0$ we can use them to draw certain conclusions about correlation functions. The spin-spin correlation function is the most suitable for this analysis. At the point of the phase transition there is no scale and therefore the correlation function has the following form:

$$\langle\langle S^{+}(\mathbf{r},t)S^{-}(0,0)\rangle\rangle=(1/|\mathbf{r}|^{2\Delta})g(|t|/|\mathbf{r}|^{a})\,. \tag{5}$$

At finite temperatures a scale appears as a correlation length $\xi(T)$. The expression (5) is valid only at

$$|t|<T^{-1},\ |\mathbf{r}|<\xi\sim T^{-1/a}\,. \tag{6}$$

From (5) and (6) we conclude that the magnetic susceptibility is proportional to

$$\chi\sim T^{-\eta}\sim T^{-1}\xi^{3-2\Delta}\sim T^{-1-(3-2\Delta)/a} \tag{7}$$

and that the scaling dimension of the magnetic field is

$$\beta=1+(3-\Delta)/a\,. \tag{8}$$

Using the experimentally found values for β,η we find from (7) and (8),

$$a\approx\Delta\approx 2.25\pm 0.1\,. \tag{9}$$

At finite temperatures, the spin excitations are overdamped and, as it follows from Eq. (9), have the following spectrum: $\omega\sim i|\mathbf{q}|^{2+\delta}$, $\delta=0.25\pm 0.1$. This spectrum is close to the diffusive one, which can be anticipated for a disordered system.

Thus, we consider our data for $U_{0.2}Y_{0.8}Pd_3$ as the first observation of the system which, while remaining a metal down to all accessible temperatures, exhibits a non-Fermi-liquid behavior. We relate this behavior to a $T=0$ phase transition in this compound as evidenced by the observed scaling of its thermodynamic properties. A phase transition at some finite but small T_0 with extremely wide critical temperature region ΔT, $\Delta T\gg T_0$, is highly improbable.

The question why this transition happens only at $T=0$ remains unresolved. Full accounting for it requires more information on the nature of the order parameter.

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[7] This result is a simple consequence of a dimensional analysis. The action (I) of the spin (S) can be expressed as $I=H\int_0^{1/T}S(t)dt\sim HT^{-1+\Delta}$. For I to be dimensionless β has to satisfy the relation $\beta=1-\Delta$.

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