Non-Fermi-Liquid Behavior in a Heavy-Fermion Alloy at a Magnetic Instability

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The specific heat C and electrical resistivity ρ of the heavy-fermion alloy $CeCu_{5.9}Au_{0.1}$ exhibit non-Fermi-liquid behavior well below 1 K, i.e., $C/T \propto -\ln(T/T_0)$ and $\rho = \rho_0 + A'T$, over more than a decade in temperature T. The magnetic susceptibility χ measured in 0.1 T shows a cusp for $T \to 0$. This behavior is attributed to the proximity to magnetic order: In contrast to $CeCu_6$, $CeCu_{6-x}Au_x$ alloys show long-range antiferromagnetic order, with $T_N \to 0$ for $x_c = 0.1$. Hence $CeCu_{5.9}Au_{0.1}$ is at the edge of a zero-temperature quantum phase transition. In a large magnetic field ($B \gtrsim 3$ T) Fermi-liquid behavior is recovered.

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Correlated electron systems exhibit a fascinating variety of behavior ranging from heavy-fermion systems with, at least in some cases, unconventional superconductivity, to high-temperature superconductors, as well as the exciting physics in dimensions lower than three, such as the integer and fractional quantum Hall effects. An issue of considerable current debate is to what extent these systems (in the normal state) can be described as Fermi liquids where the excitations have a one-to-one correspondence to those of a noninteracting Fermi gas with the well-known behavior of the specific heat $C = \gamma T$ with γ independent of temperature T in the limit $T \to 0$, a Pauli susceptibility χ independent of T, and a T-dependent electrical resistivity contribution $\Delta\rho = AT^2$ arising from particle-particle collisions. In fact, most heavy-fermion systems have been described within the framework of Fermi-liquid theory, albeit with huge effective masses m^* of the quasiparticles exceeding the free-electron mass by a factor of up to several hundred, and corresponding to huge values of γ, χ, and A with, roughly, $\gamma \propto \chi \propto \sqrt{A}$ [1]. A marginal Fermi liquid has been proposed to occur for high-temperature superconductors in order to explain phenomenologically certain features such as the linear T dependence of the electrical resistivity [2].

Recently, non-Fermi-liquid behavior has been reported for $U_{0.2}Y_{0.8}Pd_3$, with $C/T = \gamma(T) \propto -\ln(T/T_0)$, $\chi \propto T^{-\beta}$, and $\Delta\rho \propto -T$ [3,4]. Two conceptionally very different origins for this strikingly different behavior have been invoked. (i) A two-channel Kondo effect due to the electrical quadrupolar interaction might be present in U alloys where the U atoms are in a $5f^2$ atomic configuration, and also in Ce alloys of hexagonal and cubic Ce^{3+} site symmetry [3,5]. This would be effectively a single-impurity effect although it is surprising to find this for 20 at.% "impurities." However, recent experiments on more dilute samples suggest indeed a single-ion effect in this particular system [6]. (ii) On the other hand, the proximity of magnetic order and the concomitant onset of long-range correlations might lead to a breakdown of the Fermi-liquid description. This suggestion is based on the observation of scaling laws in the field and temperature dependence of C and of the magnetization M [4]. In fact, the $U_xY_{1-x}Pd_3$ system exhibits spin-glass behavior for $x \geq 0.3$ [3]. Evidence for non-Fermi-liquid behavior was found also in $UCu_{5-x}Pd_x$ for $x = 1.5$ [5], again in a U system close to spin-glass freezing. Hence, other systems have to be investigated in order to determine which scenarios might lead to non-Fermi-liquid behavior.

In this paper, we report on the observation of several features suggesting non-Fermi-liquid behavior in a Ce alloy of orthorhombic symmetry (possibly with a small low-temperature monoclinic distortion) where a quadrupolar Kondo effect should be absent [6]. Thus our results strongly support the notion that the breakdown of Fermi-liquid behavior is a quite general feature which may occur just at the onset of long-range correlations for $T \to 0$ in systems close to magnetic order. For this study, the $CeCu_{6-x}Au_x$ system was chosen. It is important to note that in these alloys the Ce atoms occupy a completely ordered sublattice, with the Au atoms occupying for $x \leq 1$ exclusively the Cu(2) site of the $CuCu_6$ structure [7]. $CeCu_6$ does not show magnetic order down to 20 mK, only short-range correlations develop below 1 K [8,9]. Upon alloying with Au, long-range antiferromagnetic order develops as evidenced by sharp maxima in $C(T)$ and $\chi(T)$ [10,11] and confirmed by elastic neutron scattering on a $x = 0.5$ alloy [12,13]. In addition, the system has been well characterized by a detailed single-crystal study [14]. Between $x = 0.2$ and 1, the Néel temperature T_N varies linearly with x with a threshold concentration $x_c \approx 0.1$ [15]. Thus, a $CeCu_{5.9}Au_{0.1}$ alloy seemed particularly well suited for the study under question.

The polycrystalline samples were arc melted under high-purity Ar atmosphere from the constituents (Ce 99.999%, Cu 99.999%, Au 99.99%) and annealed for two weeks under Ar atmosphere at 700 °C. The single crystal was grown (with the same quality of the constituents) with the Czochralski method in a W crucible under Ar atmosphere. All samples are single phase and at room temperature exhibit the orthorhombic $CeCu_6$ structure with slight changes of the lattice constants [14]. The sample preparation as well as the measuring techniques

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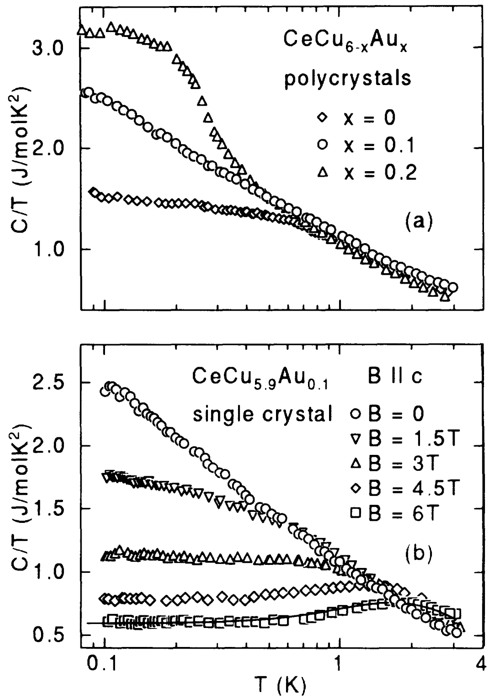

FIG. 1. (a) Specific heat C plotted as C/T vs temperature T (semilog) of $CeCu_{6-x}Au_x$ polycrystals. (b) C/T vs T (semilog) for a $CeCu_{5.9}Au_{0.1}$ single crystal for different magnetic field B applied to the easy direction. Solid line for $B=6$ T indicates fit of the resonance-level model (see text).

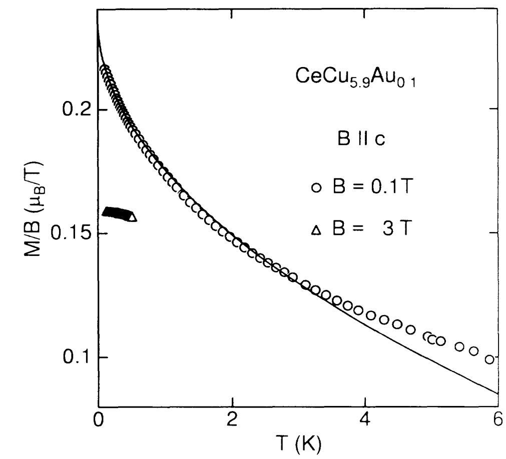

FIG. 2. Susceptibility $\chi=M/B$ of $CeCu_{5.9}Au_{0.1}$ in $B=0.1$ T (applied to the easy direction) as a function of temperature T (circles). Solid line indicates a fit of $\chi=\chi_0(1-\alpha\sqrt{T})$ to the data. Also shown is M/B measured in $B=3$ T (triangles).

for specific heat, electrical resistivity, and magnetization techniques are briefly described elsewhere [14].

Figure 1(a) shows the specific heat C of polycrystalline $CeCu_{6-x}Au_x$ with $x=0$, 0.1, and 0.2 plotted at C/T vs $\log T$. While C/T for $x=0$ is roughly constant at low T and in very good agreement with previous results [16], a broad maximum is observed for $x=0.2$ indicative of magnetic ordering as corroborated by the maximum in the ac susceptibility at $T_N=0.27$ K [15]. The alloy with $x=0.1$ exhibits an unusual specific heat with a $C/T \propto -\ln(T/T_0)$ dependence over more than one decade in T (with slight deviations above 1 K due to the onset of crystal-field excitations). This is clearly an indication of non-Fermi-liquid behavior. Figure 1(b) displays C/T of a single crystal of the same composition. In zero applied magnetic field the data follow again the $C/T = a\ln(T/T_0)$ behavior. The data for both samples with $x=0.1$ agree within 5% for $T<1$ K. The corresponding fits to the data yield $a=-0.59$ and -0.63 J/mol K^2, and $T_0=6.67$ and 5.27 K for the polycrystal and single crystal, respectively. In an applied magnetic field B parallel to the c direction, which is the easy direction in $CeCu_6$ [16] and also in $CeCu_{6-x}Au_x$ [14], the specific heat decreases strongly [Fig. 1(b)]. [Here we have already subtracted the uninteresting hyperfine contribution, mostly due to ^{63}Cu and ^{65}Cu, $C_N=b_N(B_{\rm eff}/T)^2$. For $B=6$ T, we obtain $B_{\rm eff}=7.02$ T. For smaller fields, the same ratio $B_{\rm eff}/B=1.17$ was assumed.] For large magnetic fields C can be described in terms of the single-ion resonance-level model of Zeeman-split quasiparticle levels (energy splitting $E=\mu B$) broadened with a Lorentzian of width δ [17]. The fitting parameters yielding the solid line in Fig. 1(b) are the Kondo temperature $T_K=\delta/k_B=5.5$ K and $E=7.1$ K. These values are in good agreement with those of other $CeCu_{6-x}Au_x$ alloys [14]. In particular, T_K falls between the values for $x=0$ and $x=0.5$ [14]. Independent of any model, it is evident from Fig. 1(b) that Fermi-liquid behavior with $C/T=$const over an appreciable T range (0.1 to 0.7 K) is recovered in a large magnetic field $B\geq 3$ T. The entropy change ΔS for $CeCu_{5.9}Au_{0.1}$ between 0 and 3 K for $B=0$ appears to be much larger than for $CeCu_6$. Extrapolating C/T linearly to $T=0$ (on a linear C/T vs T plot) yields $\Delta S=2.8$ J/mol K which is roughly 50% of the doublet ground-state entropy.

Figure 2 shows the magnetic susceptibility χ vs T. Here χ is taken as M/B in a rather small field $B=0.1$ T. χ increases towards lower T, albeit more slowly than any power law, as a plot of χ vs T on a log-log scale (not shown) reveals. Likewise, χ when plotted vs $\log T$ levels off for $T\rightarrow 0$. Interestingly, the data can be described quite accurately by $\chi=\chi_0(1-\alpha\sqrt{T})$ with $\chi_0=0.237\mu_B/T$ and $\alpha=0.262$ K$^{-1/2}$ [18]. The breakdown of Fermi-liquid behavior can be directly inferred from a comparison of C and χ. The ratio of $\chi T/C$ increases by $\approx 45\%$ between $T=0.1$ K and $T=0.5$ K, in clear disagreement with a temperature-independent Wilson ratio expected for a Fermi liquid. However, in $B=3$ T, M becomes almost constant at low T, increasing by only 1.3% between 0.5 and 0.1 K (see Fig. 2). This again indicates recovery of Fermi-liquid behavior in high magnetic field.

Finally, Fig. 3 shows the electrical resistivity ρ vs T in $B=0$, 3, and 6 T. For $B=0$, we observe $\rho=\rho_0+A'T$ from our lowest measuring temperature of 20 mK up to 0.5 K, i.e., over more than one decade in T, with $A'=27.6\ \mu\Omega\,\mathrm{cm\,K^{-1}}$. Similar to the specific heat, typical heavy-fermion behavior, $\rho=\rho_0+AT^2$, is recovered in $B=3$ and 6 T up to 0.7 and 1.7 K, as indicated by the fit, with $A=13.2$ and $3.7\ \mu\Omega\,\mathrm{cm\,K^{-2}}$, respectively. Indeed, perfect Fermi-liquid scaling is observed with $\gamma/\sqrt{A}=3.1\times10^2\ \mathrm{J\,mol^{-1}\,K^{-1}\,\Omega^{-1/2}\,cm^{-1/2}}$ independent of B.

The positive linear T dependence of ρ for $B=0$ over such a large T range is observed here for the first time in a heavy-fermion system, while in $U_{0.2}Y_{0.8}Pd_3$ [3] and also in $UCu_{5-x}Pd_x$ [5] a negative linear T dependence was found. The decrease of ρ towards low T in our system, as opposed to the increase in the single-ion case, is, of course, due to the onset of coherence and correlated scattering, even though the system does not behave as a Fermi liquid. The fact that the magnetoresistance is positive at the lowest temperatures up to $B\approx3$ T (see Fig. 3) underscores this novel behavior for a heavy-fermion system. It is, of course, well known that the resistivity of high-T_c superconducting oxides varies linearly with T over a wide temperature range which led to the proposal of a marginal Fermi liquid [2]. It should also be mentioned that $\Delta\rho\propto T$ was observed over a very limited temperature range (less than a factor of 2) in dilute RhFe alloys in a narrow concentration region [19].

The increase of C/T for $T\rightarrow0$ might indicate a zero-temperature magnetic phase transition as suggested before [4] to occur in $U_{0.2}Y_{0.8}Pd_3$. However, in that system the quadrupolar Kondo effect might also produce a logarithmic divergence for $T\rightarrow0$ [3,6]. In orthorhombic $CeCu_{6-x}Au_x$ the quadrupolar Kondo effect where the aspherical charge distribution of the $4f$ ion plays the role of a pseudospin and the two channels are provided by

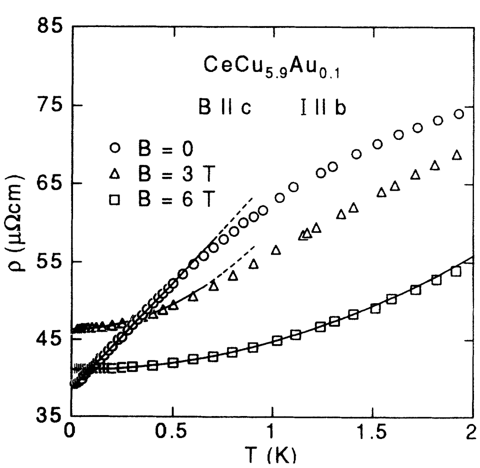

FIG. 3. Electrical resistivity ρ vs temperature T of $CeCu_{5.9}Au_{0.1}$ for different magnetic fields B applied to the easy direction. Solid lines indicate fits with a T-linear ($B=0$) and T^2 dependence ($B=3$ and 6 T).

spin-up and spin-down conduction bands is excluded because the sixfold degenerate $^2F_{5/2}$ ground state splits into three spin-degenerate doublets. Another possibility would be a genuine (magnetic) Kondo effect with the multichannel nature arising, e.g., from different conduction electron bands (s and d bands). However, s-d mixing of the conduction band in $CeCu_6$ derived from the Ce $6s$ and $5d$ and from Cu $4s$ states would mix the two channels. In fact, rather dilute $(La,Ce)Cu_6$ alloys exhibit the usual single-channel Kondo effect with the resistivity approaching the unitary limit for $T\rightarrow0$ with a $-T^2$ dependence [20]. Also, the specific heat shows nice single-ion concentration scaling [21]. Therefore, the anomalous behavior reported here can be unambiguously attributed to a cooperative effect.

Our results can be qualitatively interpreted as follows. The long-range magnetic order found in $CeCu_{6-x}Au_x$ for $x>0.1$ is suppressed to zero temperature for $x=0.1$ due to the competition with the Kondo effect. One can then view the observed anomalies as originating from the approach to a zero-temperature quantum phase transition with a weak divergence of C/T. The possibility of such a transition in Kondo lattice systems has been investigated theoretically [22]. This transition between a state of long-range magnetic order and a nonmagnetic state where the Kondo effect suppresses the long-range magnetic order occurs as a function of increasing J/W where J is the exchange interaction between f moments and s-d conduction elections (with band width W). However, up to now only scaling relations between various physical quantities have been obtained away from the critical point in the Fermi-liquid regime. Also, it is not clear if the linear T dependence of the resistivity can be understood within this model.

It has been shown some time ago that a long-range coupling of electronic quasiparticles by transverse boson-like excitations leads to a specific heat proportional to $T\ln T$ and a scattering rate that varies as $|E-\mu|$ instead of $|E-\mu|^2$ for quasiparticle-quasiparticle interactions, yielding a resistivity linear in T [23]. In this picture incipient collective excitations near the $T=0$ phase transition may lead to temperature dependent effective Landau parameters which enter the (conventional) Fermi-liquid theory where usual Fermi-liquid behavior results from Landau parameters independent of T. More theoretical work is needed to relate the various anomalous physical properties observed here in a coherent way. In particular, it must be marked out in detail which of the above scenarios will hold, as well as possible relationships between them. As a first step, a phenomenological theory of non-Fermi-liquid behavior in heavy-fermion alloys implying a critical point at $T=0$, has been proposed very recently [24]. A final important result is that non-Fermi-liquid behavior is obtained in our homogeneous system (with a stoichiometric Ce sublattice) whenever $T_N\rightarrow0$, regardless of how this is achieved. Thus non-Fermi-liquid behavior in $CeCu_{6-x}Au_x$, instead of looking at a sample with $x = x_c$ is also observed in a sample with $x = 0.3$ under pressure. This sample exhibits nicely developed antiferromagnetic ordering with very pronounced maxima in C and χ at T_N at zero pressure [14]. Pressure literally suppresses the magnetic order [10], with the result that $C/T \propto -\ln(T/T_0)$ is observed at the pressure of ≈ 8 kbar where $T_N \rightarrow 0$, again over more than a decade in T [25]. This shows that the non-Fermi-liquid behavior in our system is independent of how the transition $T_N \rightarrow 0$ is tuned.

In conclusion, our results suggest that non-Fermi-liquid behavior is a rather general feature which occurs when the nonmagnetic (Kondo compensated) and magnetic ground states are nearly degenerate, i.e., at the proximity to long-range magnetic order.

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