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1996 J. Phys.: Condens. Matter 8 9689

(http://iopscience.iop.org/0953-8984/8/48/003)

J. Phys.: Condens. Matter **8** (1996) 9689–9706. Printed in the UK

Non-Fermi-liquid behaviour in the heavy-fermion system $CeCu_{6-x}Au_x$

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Received 16 September 1996

Abstract. In heavy-fermion systems with 4f or 5f atoms (such as Ce or U) the competition between the on-site moment compensation by the Kondo effect and the long-range RKKY interaction between localized magnetic moments leads to the possibility of either a non-magnetic or a magnetically ordered ground state. However, even in the case of no long-range magnetic order as exemplified by $CeCu_6$, short-range dynamic intersite correlations are observed. Yet, the thermodynamic and transport properties of this alloy at very low temperatures T resemble those of a Fermi liquid (FL). Upon alloying with Au, long-range incommensurate antiferromagnetism is observed in $CeCu_{6-x}Au_x$ for $x > 0.1$. For $x = 0.1$ where $T_N = 0$, the specific heat C depends on T as $C/T \sim -\ln(T/T_0)$, the magnetic susceptibility as $\chi \sim 1-\alpha\sqrt{T}$, and the T-dependent part of the electrical resistivity as $\Delta\rho \sim T$. This is in marked contrast to the FL behaviour $C/T \sim \chi \sim$ constant, $\Delta\rho \sim T^2$. It is suggested that low-energy spin excitations are at the origin of these non-Fermi-liquid (NFL) anomalies which occur at a zero-temperature quantum phase transition. Large magnetic fields B restore FL behaviour. The low-T range of FL behaviour in C and ρ extends towards higher T with increasing B, with the crossover temperature T_{cr} varying roughly linearly with B. Apart from changing the Au concentration x, the magnetic–non-magnetic transition can be tuned by applying pressure p to antiferromagnetic samples with $x > 0.1$. For $x = 0.3$, $T_N \to 0$ at $p_c \approx 8$ kbar and NFL behaviour is observed in the specific heat for this critical pressure. For $x = 0.2$, $p_c \approx 4$ kbar where we likewise observe a logarithmic divergence of C/T and for $p = 6.9$ kbar we recover FL behaviour. Finally, we report on a remarkable 'universality' of C/T in the system $CeCu_{6-x}M_x$ with M = Au, Pd, Pt: regardless of how $T_N = 0$ is reached in this system (alloying with different elements M, varying concentration, or applying pressure), the C/T versus $\ln T$ curves are practically identical. Possible origins of NFL behaviour are discussed.

1. Introduction

Fermi-liquid theory has been very successful in describing the low-temperature behaviour of metals. Even many heavy-fermion systems (HFS) with their very strong electronic correlations are often analysed in terms of Fermi liquids. In these materials, with (often) a regular sublattice of 4f or 5f atoms, notably Ce, Yb, or U, a change of the magnetic behaviour occurs with decreasing temperature T from that of a collection of 'free' localized 4f or 5f magnetic moments (modified by the crystalline electric field) coupled weakly to the conduction electrons, to low-T local singlets where the localized moment—under certain circumstances—is screened completely by the conduction electrons through the Kondo effect. The energy gain of the singlet formation $k_B T_K \sim \exp(-1/N(E_F)J)$ sets the temperature scale where this change occurs. Here $N(E_F)$ is the (unrenormalized) conduction-electron density of states at the Fermi level and J is the conduction-electron–f-electron exchange constant. At sufficiently low $T \ll T_K$, Fermi-liquid (FL) properties

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are observed in a number of HFS with, however, a very large effective mass m^* derived from the huge linear specific-heat coefficient $\gamma := C/T$ and a correspondingly large Pauli susceptibility, both being only weakly dependent on T. The electrical resistivity of a FL exhibits a contribution $\Delta\rho = AT^2$ arising from particle–particle collisions [1]. Another FL property which however is less accessible at least for HFS is the existence of a sharp Fermi edge for $T \to 0$. The problem of proving or disproving FL behaviour in HFS lies in the low energy scale compared to that for conventional metals where $T_F \sim 10^4$ to 10^5 K. In HFS the corresponding energy scale is set by the Kondo temperature which is of the order of 10 to 100 K (sometimes even lower). Here we use for simplicity T_K as a measure of this scale keeping in mind that T_K in HFS might be modified by interactions with respect to the single-ion Kondo temperature in dilute magnetic alloys. Despite these problems the phenomenological correlations $\gamma \sim \chi$ [2] and $A \sim \sqrt{\gamma}$ [3] observed approximately for different HFS do suggest the validity of the FL description. The Wilson ratio $R = (\chi/\gamma)(\pi^2 k_B^2/\mu_0\mu_{eff}^2)$ deviates from the free-electron value $R = 1$ [2]. The observed values of $R \sim 2$ to 5 can be accounted for in the framework of FL theory [1] by invoking a negative Landau parameter F_0^a.

The competition between on-site Kondo interaction quenching the 4f or 5f localized magnetic moments and intersite RKKY interaction between these moments via the conduction electrons allows for non-magnetic or magnetically ordered ground states in HFS. In a simple picture [4], this competition is governed by a single parameter, namely the effective exchange constant J for exchange between conduction electrons and local moments, which enters the characteristic energy scales $k_B T_K$ and $k_B T_{RKKY} \sim J^2 N(E_F)$ for Kondo and RKKY interactions, respectively. The strength of the exchange interaction is usually tuned by composition or pressure. Owing to the extremely strong dependence of the Kondo energy scale on the interatomic distance d which arises from the exponential dependence of $k_B T_K$ on J which in turn depends on the hybridization with conduction electrons, the latter depending strongly on d, volume changes are often the dominant effect in producing the magnetic–non-magnetic transition. This is also reflected in the very large Grüneisen parameter in these materials [5].

Recently, striking deviations from FL behaviour have been found in several HFS, notably a specific-heat coefficient γ that diverges logarithmically towards low T, i.e. $C/T \sim -\ln(T/T_0)$. This non-Fermi-liquid (NFL) behaviour may have different microscopic origins such as the single-ion multichannel Kondo effect or a collective effect caused by the incipient antiferromagnetic order. The NFL behaviour in the former case is believed to arise from the 'overscreening' of the spin S of the 4f or 5f atom owing to the presence of several conduction-electron channels N, with $N > 2S$ [6]. Hence it is essentially a single-ion effect. This scenario is also possible when an internal degree of freedom other than the spin couples to a corresponding degree of freedom of the conduction electrons. Such an internal degree of freedom ('pseudospin') is provided in certain Ce or U alloys by the 4f or 5f quadrupole moment, and the true spin of the conduction electron accounts for the two channels ($s_z = \pm 1/2$) [7]. $U_xY_{1-x}Pd_3$, the first HFS where NFL behaviour was found [8], was suggested to be a candidate for this 'quadrupolar Kondo effect'. However, fluctuations in the U concentration, the proximity to magnetic ordering of the U ions, uncertainty about the ground state, and the absence of a strongly temperature-dependent quadrupolar susceptibility (which can be measured by ultrasonics) indicate that the situation is much more complex in this system.

An alternative interpretation in terms of a collective effect was suggested early on for $U_xY_{1-x}Pd_3$ [9] and probably is a viable explanation for other systems [10, 11] where the quadrupolar Kondo effect can be ruled out. In these systems, long-range magnetic order

(or sometimes only short-range spin-glass-like magnetic order) is observed for a certain concentration range of alloy constituents and the magnetic ordering temperature is driven to zero by varying some parameter (usually the composition of the alloy). The delicate balance between magnetic order and the non-magnetic ground state, the latter being due to the nearly perfect screening of the moments by the Kondo effect, is of interest in its own right. In the vicinity of this magnetic–non-magnetic transition, NFL behaviour appears.

The present article focuses on $CeCu_{6-x}Au_x$ where a number of different investigations have been carried out. An advantage of this system is that the magnetic–non-magnetic transition can be tuned by changing the concentration of the non-magnetic constituents or by changing the hydrostatic pressure, thus leaving the stoichiometric Ce sublattice basically intact. In $CeCu_6$, the quadrupolar Kondo effect can be ruled out for symmetry reasons. Before presenting the results on $CeCu_{6-x}Au_x$, we will review the salient features of $CeCu_6$ as a starting point.

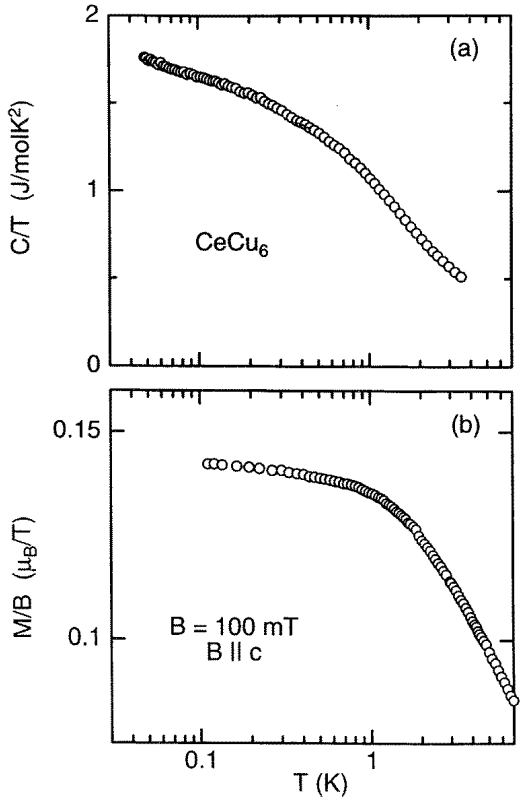

Figure 1. (a) The specific-heat C plotted as C/T versus T for $CeCu_6$ (after [15]). (b) The magnetic susceptibility $\chi \approx M/B$ of $CeCu_6$ (measured in an applied magnetic field $B = 0.1$ T parallel to the c-direction) (after [15]).

2. $CeCu_6$—a Fermi liquid with magnetic correlations

$CeCu_6$ has been established as a HFS showing no long-range magnetic order down to the range of ∼20 mK [12, 13]. $CeCu_6$ crystallizes in the orthorhombic *Pnma* structure and undergoes an orthorhombic–monoclinic distortion at around 200 K [14]. The change of the orthorhombic angle is only small (∼1.5°). In order to avoid confusion, we use the orthorhombic notation for the direction of the lattice vectors throughout this paper. $CeCu_6$ exhibits a pronounced magnetic anisotropy in the magnetization ratios along the three axes:

M_c:M_a:$M_b \approx 10$:2:1 at low T [13]. In this paper, measurements on $CeCu_{6-x}Au_x$ single crystals only, grown by the Czochralski method, are reported. Unless stated otherwise, magnetic fields are applied parallel to the easy direction (the c-direction). Figure 1 shows the T-dependencies of the specific-heat coefficient C/T and of the magnetic susceptibility $\chi \approx M/B$ measured in 0.1 T [15]. Very recently, Schuberth *et al* [16] have extended the measurements of C down to 10 mK and of χ to even below 1 mK. Their analysis of χ (after subtraction of an impurity contribution attributed to Gd) suggests magnetic order at around 5 mK [16]. This is backed by NQR measurements which likewise hint at (possibly nuclear) magnetic order [17]. μSR measurements have put an upper limit on the static moment of 10^{-2} to 10^{-3} μ_B/Ce atom (depending on the assumption of long-range magnetic versus spin-glass order) above 40 mK [18].

Although the non-magnetic ground state is usually considered a generic feature of HFS, $CeCu_6$ is about the only system where magnetic order is absent except for the anomalies below 5 mK just mentioned. All other HFS show some kind of static magnetic order with often very tiny ordered moments ($10^{-2}\,\mu_B$). A particularly intriguing example is $CeAl_3$, long considered the archetypal HFS without magnetic order. Recently, $CeAl_3$ has been shown to order antiferromagnetically when produced in single-crystalline form [19]. This is probably due to very subtle structural differences, i.e. strains, between polycrystals and single crystals. An inhomogeneous distribution of Kondo temperatures was inferred from NMR measurements for this material [20].

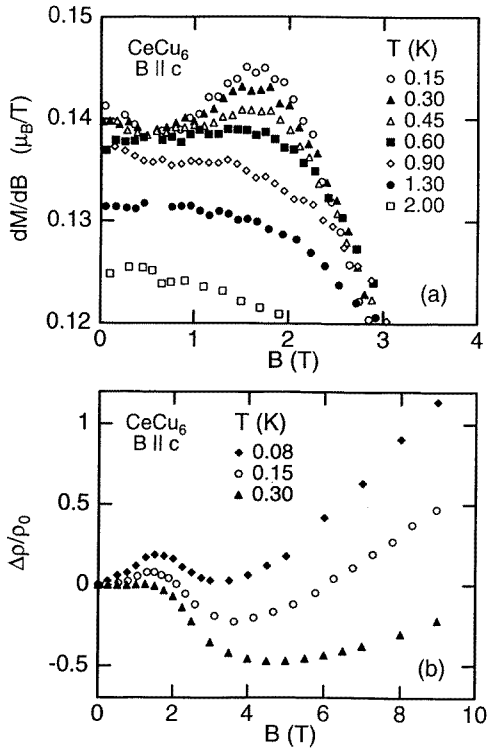

Figure 2. (a) The differential susceptibility dM/dB versus applied magnetic field for $CeCu_6$ (after [23]). (b) The magnetoresistivity $\Delta\rho/\rho_0 = (\rho(B) - \rho(0))/\rho(0)$ for $CeCu_6$.

Although $CeCu_6$ does not order magnetically above 5 mK, the expectation $C/T \approx$ constant for a FL is not met very well (cf. figure 1(a)). The single-ion Kondo model with $T_K = 6.2$ K does not fit the data below $\sim$0.4 K [15]. Instead C/T increases towards low

T which might be a precursor of the order at 5 mK. On the other hand, the susceptibility does tend to a constant for $T \to 0$ (see figure 1(b)) and the T^2-dependence of the electrical resistivity is rather well obeyed between 40 and 200 mK [13]; see also figure 8 below.

A possible reason for the deviations from FL behaviour already in $CeCu_6$ (besides the magnetic order at very low T) might be the existence of intersite antiferromagnetic correlations. These have been first inferred from ultrasonic-attenuation measurements $\alpha(B)$ where a maximum at $B \approx 2$ T was found [21] with the field applied along the easy direction (the c-direction). More directly, they were observed in the dynamic structure factor $S(Q, \omega)$ of inelastic neutron scattering from peaks in $S(Q, \omega)$ for energy transfer $\hbar\omega = 0.3$ meV at $\boldsymbol{Q} = (1\ 0\ 0)$ and $(0\ 1 \pm 0.15\ 0)$ [22]. The rather large widths of these peaks correspond to correlation lengths extending roughly only to the nearest Ce neighbours. These correlations vanish at a field of $\sim$2 T.

The breaking of the antiferromagnetic correlations is also observed in the differential magnetic susceptibility $\mathrm{d}M/\mathrm{d}B$ as a shallow maximum at 2 T at very low T; see figure 2(a) [23]. It vanishes quickly upon increasing T. This maximum indicating the breaking of short-range correlations has been identified with the 'metamagnetic transition' in loose analogy to the metamagnetic transition in strongly anisotropic antiferromagnets. In other systems such as $CeRu_2Si_2$ this maximum, occurring here at 8 T, is much more pronounced [24]. It is interesting to note in the present context that all the way through this metamagnetic transition $CeRu_2Si_2$ exhibits well-defined FL behaviour as evident from the prevalence of de Haas–van Alphen oscillations [25].

A maximum is also seen in $\rho(B)$ at 2 T for low T—see figure 2(b)—as observed previously [13]. This might be interpreted as arising from scattering of low-lying excitations present at the metamagnetic transition, although the observation of a similar maximum for $x = 0.1$ where a metamagnetic transition is not observed in $\mathrm{d}M/\mathrm{d}B$ makes this assignment questionable (see below).

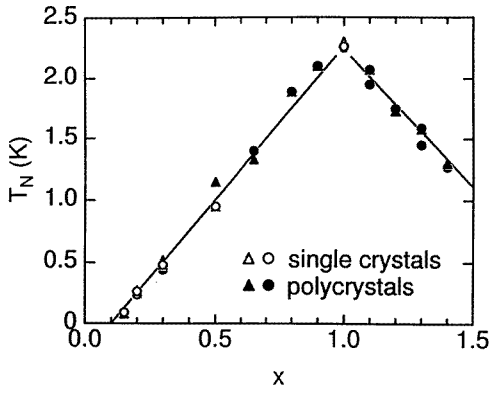

Figure 3. The Néel temperature T_N of $CeCu_{6-x}Au_x$ versus Au concentration x as determined from the specific heat (triangles) and magnetic susceptibility (circles) (after [27]).

3. Magnetic order in $CeCu_{6-x}Au_x$

Upon alloying with Au the $CeCu_6$ lattice expands while retaining the orthorhombic (at room temperature) $Pnma$ structure. Thus the hybridization between Ce 4f electrons and conduction electrons and hence J decreases, leading to a stabilization of localized magnetic moments which can now interact via the RKKY interaction. The result is antiferromagnetic order in $CeCu_{6-x}Au_x$ beyond a threshold concentration $x_c \approx 0.1$ as is evident from the sharp

maxima in the specific heat and magnetization. Some examples can be seen in figures 6 and 7 below. For $0.1 \leqslant x \leqslant 1$ where Au occupies exclusively the Cu(2) position in the $CuCu_6$ structure the Néel temperature T_N varies linearly with x. For the stoichiometric compound $CeCu_5Au$ a complex magnetic phase diagram has been mapped out [26]. Beyond $x = 1$, T_N decreases again (figure 3). This is due to a subtle change within the orthorhombic structure: for $x < 1$ the lattice parameters a and c increase while b decreases with growing Au content, whereas for $x > 1$ all three lattice parameters $a, b,$ and c increase [27]. For all x up to 1.5, the unit-cell volume increases.

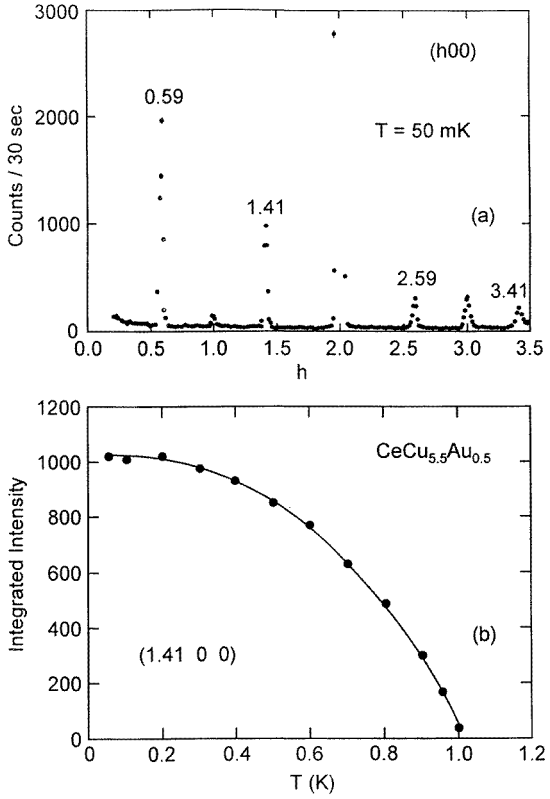

Figure 4. An (h 0 0) elastic neutron scattering scan (a) and the integrated intensity of the (1.41 0 0) magnetic reflection versus temperature T (b) for $CeCu_6$ (after [28]). The line is a guide to the eye.

Figure 4(a) shows an elastic neutron scattering scan along the (h 0 0) direction for $x = 0.5$ [28]. Below 1 K incommensurate reflections are observed. Figure 4(b) shows that the integrated intensity of the (1.41 0 0) reflection decreases smoothly to zero at $T_N \approx 1$ K, indicating a second-order phase transition. The magnetic ordering wave vector $\boldsymbol{Q} = (0.59\ 0\ 0)$ is different from the wave vector of the short-range correlation in pure $CeCu_6$. However, for $x = 0.2$ a change to $\boldsymbol{Q} = (0.79\ 0\ 0)$ is found for the ordering wave vector [29]. Figure 5 gives a preliminary scenario of the evolution of the dynamic short-range correlations for $CeCu_6$ (neutron energy transfer $\hbar\omega = 0.3$ meV) [22] and for $CeCu_{5.9}Au_{0.1}$ ($\hbar\omega = 0.25$ meV) [29] to the long-range magnetic ordering wave vector. Of course, more measurements are necessary to corroborate this simple scenario. A further

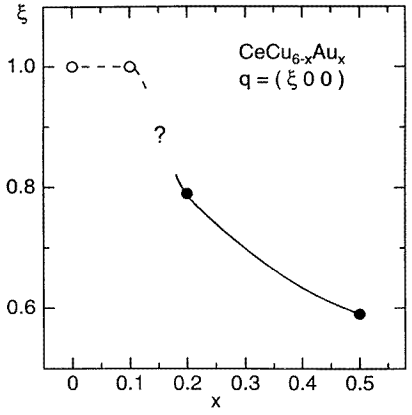

Figure 5. The magnetic ordering wave vector (closed circles) and the wave vector of dynamic correlations (open circles) along the a-direction of $CeCu_{6-x}Au_x$.

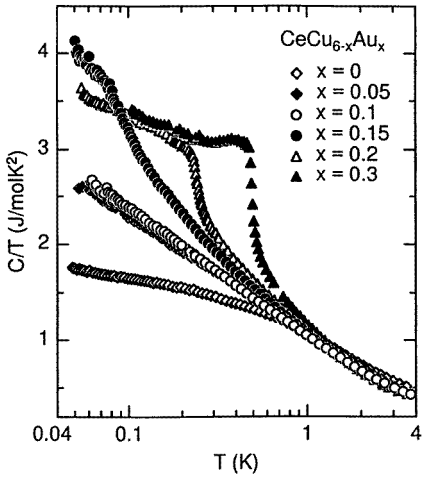

Figure 6. The specific heat C of $CeCu_{6-x}Au_x$ plotted as C/T versus $\log T$ (after [35]).

point worth noting is that the magnitude of the ordered moment decreases drastically from $\sim 1\,\mu_B$/Ce atom for $x = 0.5$ to $0.02\,\mu_B$/Ce atom for $x = 0.2$. The q-dependence of the incommensurate reflection for $x = 0.5$ can be approximated by $f^2 = \exp(-(q/q_f)^2)$ with $q_f = 2.8$ Å^{-1} [28] which is slightly smaller than the theoretical value $q_f = 4.3$ Å^{-1} for Ce^{3+} [30]. This slightly narrower form factor (in q-space) might indicate a tendency towards more delocalized moments as expected for a spin-density wave formed out of heavy-fermion quasiparticles. On the other hand, the strong x-dependence of the magnetic ordering wave vector $\boldsymbol{Q}$ is difficult to understand within a spin-density-wave scenario of magnetic order.

In order to microscopically understand the origin of the long-range antiferromagnetism in $CeCu_{6-x}Au_x$, a detailed investigation of the dependence of T_N on hydrostatic pressure p was conducted [27]. Over the whole range up to $x = 1.3$, T_N decreases under pressure. One can compare the volume dependence of $T_N^{(p)}(V)$ obtained from the p-dependence via the known compressibility κ of pure $CeCu_6$ [32] with the concentration-dependent volume dependence $T_N^{(x)}(V)$ inferred from room-temperature x-ray diffraction. With increasing x, $T_N^{(p)}(V)$ and $T_N^{(x)}(V)$ differ more and more [27]. This suggests that besides the volume expansion upon alloying which is the dominant effect in establishing magnetic order, one or more of the following mechanisms might be operative: (i) changes of the electronic structure upon alloying with Au, (ii) anisotropy of the volume change under compression and upon alloying, and (iii) decreasing compressibility κ of $CeCu_{6-x}Au_x$ with increasing x. However, recent measurements of κ at room temperature for $x = 0.5$ yield results similar to those for $x = 0$, rendering (iii) unlikely [33].

Of course, the $T_N(x)$-dependence beyond the maximum at $x = 1$ cannot be attributed to a volume effect since, as mentioned above, $\mathrm{d}T_N/\mathrm{d}p < 0$ also for $x > 1$. On the other hand, the volume dependence of T_N for $x < 1$ is consistent with the Doniach model [4] of a competition of on-site Kondo compensation and intersite RKKY interaction. Further light might be shed on the mechanism by uniaxial-pressure experiments. For $x = 0.2$ it was found that while for p parallel to the b- and c-directions T_N decreases as might be expected from the hydrostatic-pressure dependence, T_N *increases* for p parallel to the

a-direction [34]. Apparently, a decrease of the *a*-axis of the unit cell stabilizes the net antiferromagnetic component of the RKKY interaction.

In conclusion to this section, the long-range magnetic order in $CeCu_{6-x}Au_x$ has been well characterized by a number of different measurements and, in addition, yields a surprising richness of phenomena. The most exciting features are, however, observed right at the onset of antiferromagnetic order, i.e. at around $x = 0.1$.

4. Non-Fermi-liquid behaviour at the magnetic instability

4.1. Concentration tuning

Figure 6 shows specific-heat data for concentrations in the vicinity of the critical concentration $x_c \approx 0.1$ plotted as C/T versus log T [35]. The Néel temperature T_N manifests itself as a sharp kink in C/T which becomes less pronounced as T_N decreases. It is, however, still clearly visible for $x = 0.15$ where $T_N = 0.080$ K as confirmed by a maximum in the ac susceptibility. For $x = 0.1$ we observe the non-Fermi-liquid behaviour $C/T = a\ln(T/T_0)$ between 0.06 K and ~2.5 K, i.e. over almost two decades in T. These data represent at present the best example of a logarithmic divergence of C/T for $T \to 0$. (The positive deviations above 2.5 K can be attributed to phonon and crystal-field contributions to C.) It is important to verify that the critical NFL behaviour at x_c does not arise from some inhomogeneity of the alloys, i.e., a distribution of magnetic ordering temperatures. A recent μSR study has shown that there is no ordered magnetic moment in a $x = 0.1$ sample, with the detection limit $\mu < 10^{-3}\,\mu_B$ [36]. On decreasing the concentration further, we find for $x = 0.05$ an almost identical behaviour, i.e. here, too, the specific heat shows NFL behaviour. Finally, for $x = 0$ we recover the approximate FL behaviour of $CeCu_6$ discussed in section 2 above.

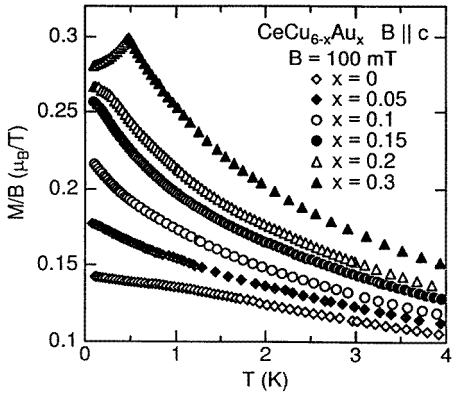

Figure 7. The magnetic susceptibility $\chi \approx M/B$ versus temperature T (after [35]).

It is interesting to note that the NFL behaviour does not extend 'symmetrically' from $x = x_c$ into the FL and magnetically ordered regions. While there is hardly any difference between $x = 0.1$ and 0.05, the 'departure' from NFL behaviour for $x = 0.15$ is occurring already at around 0.5 K, or at $T \approx 6\,T_N$. This different behaviour might be related to the fact that very low temperatures (or a very large distance from the NFL critical point) are needed on the disordered side to enter the FL state. In the phase diagram near a quantum critical point proposed by Millis [37] the crossover temperature from NFL to FL behaviour (with logarithmic corrections) away from the critical point (in the disordered region) varies as

$T_{cr} \sim |\delta - \delta_c|^{z/2}$ where δ is some parameter driving the magnetic–non-magnetic transition (in our case the Au concentration x) and z is the dynamic critical exponent, while the Néel temperature varies as $T_N \sim |\delta - \delta_c|^{z/(z+1)}$. Hence with $z = 2$ as expected for an antiferromagnet, T_N initially increases much faster with increasing $|\delta - \delta_c|$ than T_{cr}, in grossly qualitative agreement with the observed behaviour. On the other hand, for the related system $CeCu_{6-x}Pd_x$ with also $x_c \approx 0.1$, a clear tendency towards FL behaviour was observed in C/T for $x = 0.05$ [38]. For $z = 2$, a $-\sqrt{T}$-correction to FL behaviour is predicted [37] while a logarithmic divergence of C/T is expected to occur for a ferromagnet with $z = 3$.

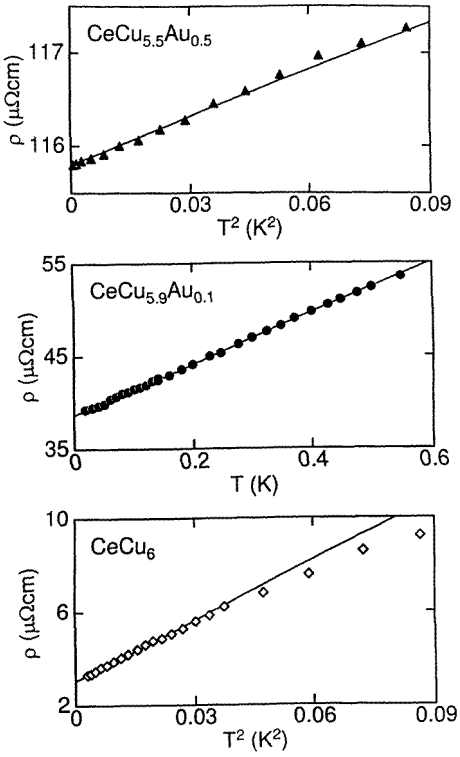

Figure 8. The temperature dependence of the electrical resistivity ρ. For $x = 0$ and $x = 0.5$, ρ is plotted versus T^2, with the straight line indicating FL behaviour. For $x = 0.1$, the linear ρ versus T dependence signals NFL behaviour (data for $x = 0.5$ after [15], for $x = 0.1$ and 0 after [11]). In all cases the current is parallel to the b-direction.

The magnetization data of the same set of samples are shown in figure 7 plotted as M/B versus T. The measuring field $B = 0.1$ T was always applied parallel to the easy direction (the c-axis) of the samples. Again, $\chi \approx M/B$ for the rather low measuring field. The transition to antiferromagnetic ordering, visible as a sharp maximum for $x = 0.3$ only, presents itself as a kink in $M(T)$ for $x = 0.2$ and 0.15. The temperatures where these features occur are in good agreement with T_N from the specific heat. ac susceptibility measurements have shown sharp maxima for $x = 0.15$ and 0.2 at temperatures which coincide with those of the kinks of the dc magnetization. For $x = 0.1$ the magnetization exhibits a cusp for $T \to 0$ which can be modelled as $M/B \sim 1 - \alpha\sqrt{T}$ between ~80 mK and 3 K. The same T-dependence of M/B is found upon reduction of the field to 0.01 T.

Upon decreasing x beyond $x = 0.1$ where the NFL behaviour $M/B \sim 1-\alpha\sqrt{T}$ is observed, M/B is quickly suppressed towards $M/B \approx$ constant for $CeCu_6$. In fact, the data for $x = 0.05$ lie almost exactly halfway between those for $x = 0.1$ and $x = 0$. This is in marked contrast to the specific-heat behaviour discussed above. Further investigations, in particular towards even lower fields and lower temperatures, are necessary to resolve the origin of the difference in C and M, which might be related to the apparent difference in the NFL behaviour at x_c, i.e. logarithmic divergence of C/T versus a cusp in M for $T \to 0$. In the current theories of NFL behaviour arising from a quantum critical point at $T = 0$, the same leading-order critical behaviour should be expected for C/T and M [39]. However, this would apply to the staggered magnetization for the magnetic order which is accessible with neutron scattering. The uniform magnetization that we have measured might have different behaviour as $T \to 0$ is approached. In order to check for thermodynamic consistency, we measured the magnetocaloric effect $\Delta T/\Delta B$, i.e. the temperature change ΔT following a field change ΔB. Since $(\Delta T/\Delta B)C/T \approx -(\partial S/\partial B)_T \equiv -(\partial M/\partial T)_B$, a $T^{-1/2}$-dependence is expected for the l.h.s. from $M/B \sim 1 - \alpha\sqrt{T}$, which is indeed observed experimentally [40].

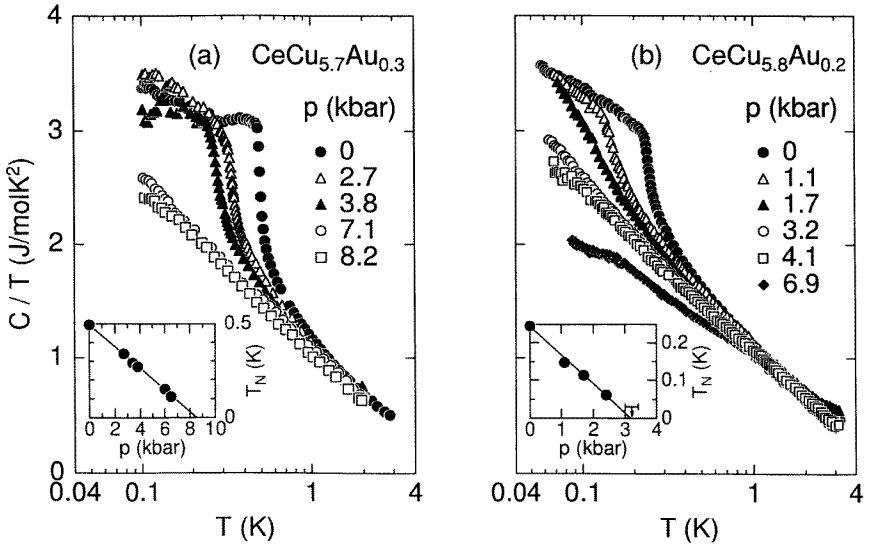

Figure 9. The specific heat C of $CeCu_{5.7}Au_{0.3}$ (after [41]) (a) and $CeCu_{5.8}Au_{0.2}$ (after [34]) (b) plotted as C/T versus $\log T$ for various hydrostatic pressures p. Insets show the $T_N(p)$-dependence.

Figure 8 shows the resistivity $\rho(T)$ for current flow parallel to the b-direction for $x = 0$, 0.1 and 0.5 (note the different horizontal scales). $CeCu_6$ exhibits $\Delta\rho \sim T^2$ as mentioned above. Likewise $CeCu_{5.5}Au_{0.5}$ exhibits a quadratic FL-like T-dependence of ρ well inside the ordered region. However, $CeCu_{5.9}Au_{0.1}$ at the magnetic instability shows $\Delta\rho = A'T$ over an appreciable T-range between 20 mK and 0.6 K. The factor-of-two difference of A' between the a-direction on the one hand and the b- or c-direction on the other hand (cf. figure 11 below) is the same as is found in pure $CeCu_6$ [13]. Again, this difference reflects the large amount of scattering in the direction of the magnetic correlations or the incipient magnetic ordering wave vector. Of course, the linear T-dependence of ρ is reminiscent of the linear T-dependence of high-T_c superconductors at optimum doping.

A final interesting point concerns the ultrasonic behaviour. For $CeCu_6$ a FL-like variation of the longitudinal elastic constants c_{22} and c_{33} with T is observed below 1 K, $\Delta c/c_0 \sim T^2$, arising from $\Delta c \sim \Omega U(T)$ where U is the internal energy and Ω the appropriate Grüneisen parameter [5]. For $CeCu_{5.9}Au_{0.1}$, Δc increases as T^2 between ~0.08 and 0.2 K only, but then turns over to a linear increase towards higher T up to 0.9 K [40]. Clearly, a simple Grüneisen parameter fit does not work since $U(T)$ is very similar for $x = 0$ and $x = 0.1$. Whether this linear T-dependence is a general feature at a magnetic instability and related to the other NFL anomalies remains to be investigated.

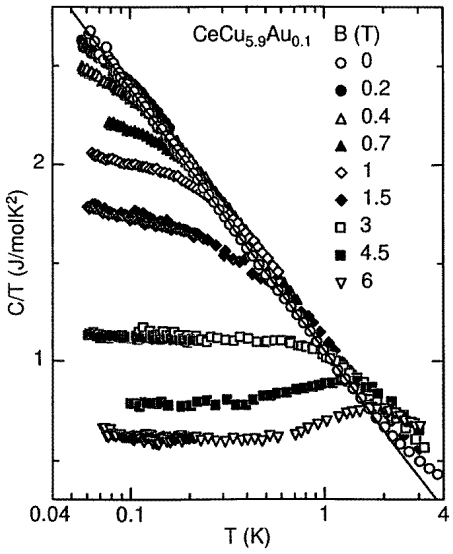

Figure 10. The specific heat C of the NFL alloy $CeCu_{5.9}Au_{0.1}$ plotted as C/T versus $\log T$ in various magnetic fields B applied parallel to the c-direction. The nuclear contribution has been subtracted (after [40]).

4.2. Pressure-tuning the magnetic transition to $T = 0$

As mentioned above, the onset of magnetic order in the $CeCu_{6-x}Au_x$ system is attributed to a weakening of the conduction-electron–4f-electron exchange constant J because of the increase of the molar volume upon alloying with Au, and indeed, T_N of $CeCu_{6-x}Au_x$ decreases under hydrostatic pressure. Figure 9 shows C/T versus $\log T$ for $x = 0.3$ and 0.2 under various hydrostatic pressures p [41, 34]. The Néel temperature (again as determined from the inflexion point of $C(T)$ above the maximum) decreases linearly with increasing p for $x = 0.3$. For $x = 0.2$, a linear $T_N(p)$-decrease is also compatible with the data. $T_N \approx 0$ is reached for 7–8 kbar or 3.2–4 kbar for $x = 0.3$ and 0.2, respectively. At these pressures both alloys exhibit NFL behaviour with, surprisingly, the same coefficients a and T_0 for both, and additionally for the NFL alloy with $x = 0.1$ and $p = 0$. The data for 7.1 and 8.2 kbar for $x = 0.3$ and for 3.2 and 4.1 kbar for $x = 0.2$, respectively, are rather close, indicating again a finite width of the critical region. However, at 6.9 kbar for $x = 0.2$ the clear suppression of the low-T increase of C/T towards C/T for $CeCu_6$ indicates restoration of the FL, i.e. we have pressure-tuned $CeCu_{5.8}Au_{0.2}$ all the way from antiferromagnetic order through the quantum critical point to the Fermi liquid. The roughly

linear $T_N(p)$-dependence is at variance with the expectation $T_c \sim |p - p_c|^{2/3}$ for $z = 2$ [37].

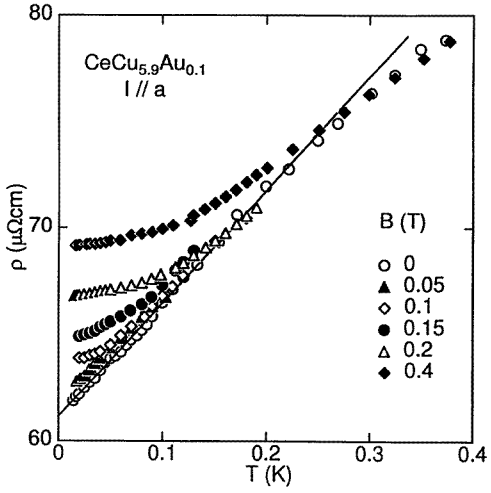

Figure 11. The electrical resistivity ρ versus temperature T of $CeCu_{5.9}Au_{0.1}$ (current parallel to the a-direction) in various magnetic fields B applied parallel to the c-direction (after [31]).

4.3. The crossover to Fermi-liquid behaviour in magnetic fields

A simple qualitative explanation for NFL behaviour at a magnetic instability is the abundance of low-energy magnetic excitations when the Néel temperature is tuned to just zero, because the magnetic excitation spectrum of an antiferromagnet is 'compressed' to a very small region around $\hbar\omega = 0$ when $T \to 0$. This speculation is supported by the recovery of FL behaviour in high magnetic fields B [11, 42]. Figure 10 shows that the tendency towards FL behaviour $C/T =$ constant is gradually weakened as the field decreases [40]. Here the hyperfine contribution to C due to the nuclear Zeeman splitting was subtracted appropriately. A negative deviation from the $C/T \sim -\ln T$ divergence is seen for all fields $B \geqslant 0.2\,\mathrm{T}$. Defining arbitrarily the temperature where the C/T data in a certain field cross the $B = 0$ straight line in figure 10 as a crossover temperature T_{cr} we find roughly $T_{cr} \sim B$. This roughly linear field dependence of T_{cr} should receive closer theoretical scrutiny. A qualitatively similar crossover is observed in the electrical resistivity where the low-T data in finite fields $\geqslant 0.1\,\mathrm{T}$ are compatible with a T^2-dependence as opposed to the zero-field linear T-dependence which is seen in finite fields only at elevated temperatures, if at all (figure 11).

Since a sufficiently large magnetic field can suppress antiferromagnetic ordering one might ask whether NFL behaviour might arise at a magnetic-field-induced instability in $CeCu_{6-x}Au_x$. In the light of the preceding discussion, however, the (negative) answer is immediately at hand: an applied magnetic field B suppresses low-lying spin excitations, and hence FL behaviour should prevail. This is indeed qualitatively observed. Figure 12 shows C/T for $CeCu_{5.8}Au_{0.2}$ in several fields. T_N first decreases with increasing B until at $B = 0.5$ T the 'knee' indicating the magnetic ordering in the C/T versus T plot has become too broad to identify T_N. From the (B, T) phase diagrams for $x = 0.3$ and $x = 0.5$ [15] we estimate that for $x = 0.2$ with $T_N = 0.25$ K the critical field is $B_c = 0.40$ T for $T \to 0$. Beyond B_c, C/T immediately shows negative deviations towards low T from

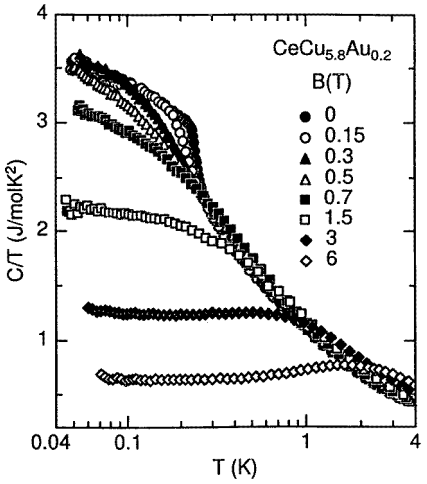

Figure 12. The specific heat C of $CeCu_{5.8}Au_{0.2}$ plotted as C/T versus $\log T$ in various magnetic fields B applied parallel to the c-direction (after [35]).

a straight line in figure 12, i.e. NFL behaviour is never observed. With further increase of B, C/T develops towards a temperature-independent plateau which presumably occurs also for the lower fields below the present T-range, i.e. when $k_BT < g\mu_BB$. (Again the hyperfine contribution due to the nuclear Zeeman splitting has been subtracted.) We add that the high-field specific heat of all $CeCu_{6-x}Au_x$ alloys including that with $x = 0.1$ can be reasonably well described [15, 35] within a single-ion Kondo model, i.e. the resonance-level model.

4.4. The origin of non-Fermi-liquid behaviour of $CeCu_{6-x}Au_x$

As has been discussed in the previous sections, the NFL behaviour in the $CeCu_{6-x}Au_x$ system is attributed to the proximity to a magnetic instability for $x = 0.1$ at $T = 0$. It is remarkable that virtually the same specific-heat behaviour is seen in the system $CeCu_{6-x}M_x$ (M = Au, Pd, Pt) when these alloys are close to a magnetic instability, i.e. $T_N \approx 0$, as determined from measurements with different concentrations [38]. In addition, the pressure-tuned NFL Au alloys with $x = 0.3$ and 0.2 show nearly identical specific heats at their respective critical pressures. These data are shown in figure 13 where indeed a universal straight line corresponding to $C/T = a\ln(T/T_0)$ is found with $a \approx -0.56$ J mol^{-1} K^{-2} and $T_0 \approx 7$ K. The individual data sets are also shown in figure 13, offset by 0.5 J mol^{-1} K^{-2} each for clarity [43]. This universal behaviour is unusual. In usual second-order phase transitions driven by thermal fluctuations, universality implies that in a given universality class the critical exponent and the amplitude ratio of the critical specific heat above and below the critical point are constants. This would translate to the logarithmic divergence of C/T and to a constant value a, respectively. However, the constant absolute magnitude of C/T given by the constant value of T_0 calls for special attention. This indicates some additional universality in the $CeCu_6$-derived systems, regardless of how $T_N \to 0$ is reached. It would be of interest to look for this type of universality in other systems. We should mention that this universality has up to now only been observed in the specific heat. The samples with M = Pd and Pt were polycrystals where a determination of M along the easy

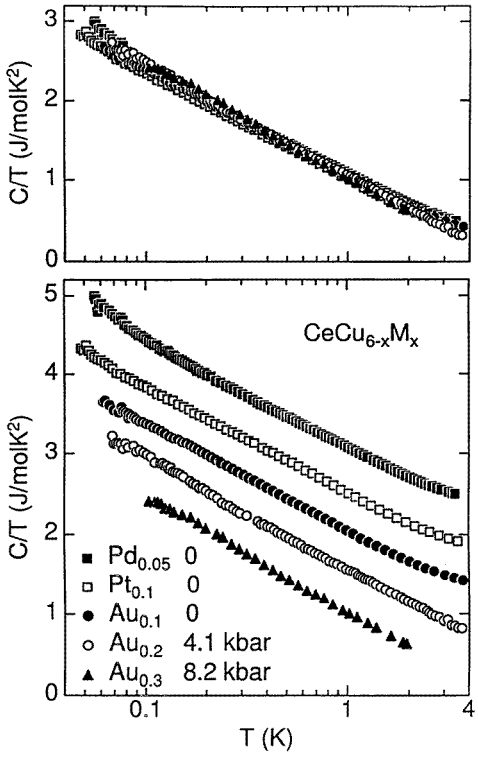

Figure 13. C/T versus $\log T$ for various $CeCu_{6-x}M_x$ samples and pressures as indicated. The upper frame shows the universality of the logarithmic divergence of C/T for $T \to 0$. The lower frame shows the same data shifted by 0.5 J mol^{-1} K^{-2} each for clarity (after [43]).

axis was not possible, but magnetization measurements under pressure are under way.

It has been suggested that NFL properties can also be understood in terms of more conventional spin-fluctuation theories [44]. Indeed it has been suggested that our specific-heat and resistivity data fit into this model [45]. Figure 14 shows the best fit of the spin-fluctuation theory to our data with the fit parameters $y_0 = 0.003$, $y_e = 16.7$ and $T_0 = 3.44$ K given by Kambe *et al* [45]. At first sight, the fit does give a satisfactory representation of our data, although an important qualitative feature is different. While the data do fall on a straight line in $C/T \sim -\ln(T/T_0)$, this strict linearity is not followed by the fit. It 'wiggles' around the data reflecting the fact that a $-\ln(T/T_0)$ behaviour of the specific heat is never truly found in the spin-fluctuation theory; rather a crossover to FL behaviour at very low T should occur with a $-\sqrt{T}$-deviation from the value of C/T at $T = 0$. It is therefore important to extend the present measurements (which actually show the NFL behaviour $C/T \sim -\ln T$ over the largest T-range yet seen) to even lower T. These measurements are currently in progress. In any case, the linear T-behaviour of the resistivity found over a large T-range in $CeCu_{5.9}Au_{0.1}$ cannot be accounted for in this model which predicts $\Delta\rho \sim T^{1.5}$ for antiferromagnets at $T_N = 0$ [44, 45].

An important point that has to be mentioned is the possible role of disorder. For ordinary phase transitions it is well known that disorder can have a decisive influence on the critical behaviour. Disorder is expected to also play a role, possibly even more important, in quantum phase transitions [37]. Clearly, the amount of disorder in the various samples of

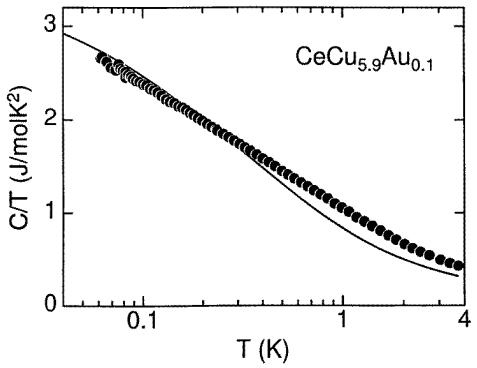

Figure 14. A comparison of the measured C/T versus $\log T$ of $CeCu_{5.9}Au_{0.1}$ with the best fit in terms of the spin-fluctuation theory (see the text for details).

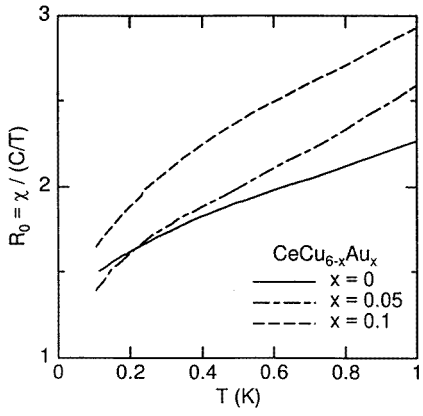

Figure 15. The temperature dependence of the Wilson ratio R for $CeCu_{6-x}Au_x$.

the present study is quite different, as suggested by their different residual resistivities ρ_0. Yet, the same NFL temperature dependence is observed in the specific heat. Also, single crystals and polycrystals show the same behaviour in C [42]. Furthermore, the pressure-tuned single crystals with higher Au concentration have larger ρ_0 than the $CeCu_{5.9}Au_{0.1}$ single crystal at the critical concentration at ambient pressure. This suggests the amount of disorder at the level of the present samples has only a minor influence on C. In order to resolve these issues, a search for NFL properties in stoichiometric HFS where the magnetic ordering temperature can be pressure-tuned to zero is highly desirable.

Experiments on non-HFS such as MnSi and $ZrZn_2$ have recently been carried out where T_N was pressure-tuned to zero [46]. For $ZrZn_2$, $\Delta\rho \sim T^{1.6}$ was found, which is close to $\Delta\rho \sim T^{1.67}$ expected for ferromagnets at the quantum critical point. In addition, the HFS $CePd_2Si_2$ shows NFL behaviour in the resistivity, i.e. an almost linear T-dependence, at the magnetic instability where antiferromagnetism gives way to superconductivity with a second-order phase transition [47]. The most comprehensive study of a stoichiometric HFS at a magnetic instability was undertaken on Ce_7Ni_3 by Umeo *et al* [48]. Although the system is very complicated because of three different Ce sites, clear indications for NFL behaviour at the critical pressure for the disappearance of antiferromagnetism were found in the specific heat, magnetization and resistivity, in rather close analogy to the present work.

Another aspect of disorder is the following. In a disordered alloy a distribution of Kondo temperatures is possible. This might be due to variations in composition or internal strains which lead to a spatially varying conduction-electron–f-electron hybridization and hence different values of J and T_K. In fact, the most prominent representative of this scenario is a strongly disordered metal just above the metal–insulator transition. This is exemplified in heavily doped semiconductors where a distribution of hopping energies (due to varying donor distances) naturally leads to a distribution in T_K [49–51]. In Si:P, this T_K-distribution leads to an *algebraic* divergence of C/T for $T \to 0$ [51, 11]. It is important to realize, however, that this divergence is not a genuine NFL effect but rather originates from an incoherent superposition of Kondo temperatures with a wide $P(T_K)$. Of course, in any alloy, in particular in HFS with their often complicated metallurgy, one has to be aware of possible inhomogeneities with a resulting $P(T_K)$ which could mimic NFL behaviour. Indeed, it is has been shown recently that the low-T thermodynamic properties

of $UCu_{5-x}Pd_x$ which had been interpreted in terms of NFL behaviour [52] can alternatively be described by a $P(T_K)$ [53]. Our present data could arguably be attributed to a distribution of T_K (assuming an inhomogeneous alloy) if the fluctuation of the susceptibility inferred from μSR measurements were to occur on a rather long-range scale [54]. On the other hand, the alloying of $CeCu_6$ with Au induces just short-range fluctuations of the local susceptibility because the hybridization changes statistically around the Ce atoms due to different local environments.

It is of interest to check whether the NFL behaviour of $CeCu_{6-x}Au_x$ is reflected in distinct changes of the Wilson ratio $R = (\pi^2 k_B^2/3\mu_{eff}^2)\chi T/C$. Figure 15 shows $R(T)$ for $x = 0$, 0.05 and 0.1. (For the magnetically ordered alloys the determination of R is ambiguous because of the contribution of magnetic degrees of freedom to C/T and χ.) For χ we took the susceptibility (measured at 0.1 T) averaged over directions with the assumption $\chi_c{:}\chi_a{:}\chi_b = 10{:}2{:}1$ as determined experimentally. For simplicity, we have taken $g = 2$ and $\mu_{eff} = \mu_B$ to calculate R. Although there is no decisive difference in $R(T)$ for the three concentrations, the faster decrease of $R(T)$ for $T \to 0$ reflects the logarithmic divergence of C/T compared to the finite χ for $T \to 0$. Again, measurements are under way to determine R at even lower temperatures.

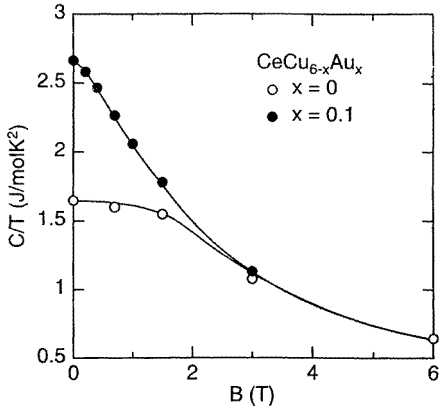

Figure 16. The magnetic field dependence for C/T of $CeCu_6$ (measured at 0.1 K) and $CeCu_{5.9}Au_{0.1}$ (measured at 0.06 K) (after [40]). Lines are guides to the eye.

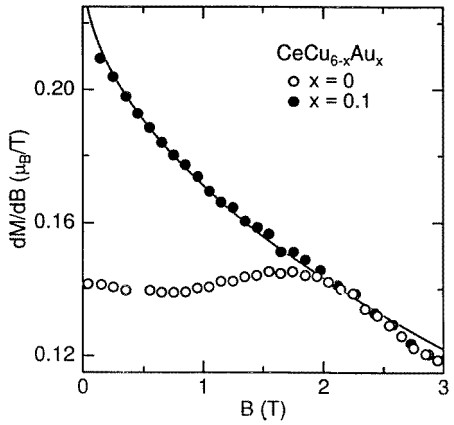

Figure 17. The differential susceptibility dM/dB versus B for $CeCu_6$ and $CeCu_{5.9}Au_{0.1}$ (measured at 0.15 K) (after [40]). The solid line indicates a square-root dependence of dM/dB.

As a final point, we return to the destruction of NFL behaviour in magnetic fields. Here, C/T and dM/dB [40] behave quite differently for $CeCu_{5.9}Au_{0.1}$ where both quantities decrease rapidly, and for $CeCu_6$ where both remain fairly constant up to moderate fields, except for the shallow maximum at 2 T arising from the metamagnetic transition (see figures 16 and 17). In fact, the dM/dB data for the NFL alloy with $x = 0.1$ can be described quite well by a $\sqrt{B}$-dependence. This underscores our interpretation of NFL behaviour in terms of low-lying spin excitations at a $T = 0$ critical point.

5. Conclusions

The present comprehensive study shows that $CeCu_{6-x}Au_x$ exhibits the most striking non-Fermi-liquid features of all systems at a magnetic instability studied thus far, i.e. the T-linear

electrical resistivity ρ and the logarithmic divergence of C/T. Up to now, there has been no indication for a tendency towards saturation of C/T or for a $T^{1.5}$-resistivity as predicted by spin-fluctuation theories. On the other hand, these models appear to work well for transition metal itinerant magnets such as $ZrZn_2$ and MnSi, and also for heavy-fermion systems with comparatively high Kondo temperatures. $CeCu_{6-x}Au_x$ where the formation of the heavy quasiparticles and the observed NFL properties occur on the same low-T scale, i.e. $T_K \approx 6$ K, seems to present a different case. The question of itinerant (spin-density-wave) versus local moment antiferromagnetism is actually intimately related to this problem. Definitely, more studies are needed to resolve this problem. In particular, more neutron scattering experiments, both elastic and inelastic, are needed. Here, the strong variation of the ordering wave vector needs to be investigated in more detail, in particular in the vicinity of the magnetic instability. Another issue which definitely has to be studied further is the role of disorder. It is hoped that our detailed investigation of the $CeCu_{6-x}Au_x$ system helps in developing an understanding of the NFL phenomena at a magnetic instability.

Acknowledgments

I thank B Bogenberger, F Huster, S Mock, A Neubert, T Pietrus, G Portisch, H G Schlager, A Schröder, M Sieck, C Speck, O Stockert, T Trappmann and M Waffenschmidt for a fruitful collaboration. Their contributions to the work presented in this article can be identified from the references cited. Collaborations with J W Lynn, R W Erwin, M Loewenhaupt and N Pyka on neutron scattering, with A Amato, A Schenk, D E MacLaughlin and their colleagues on μSR measurements, and B Lüthi and his colleagues on ultrasonic experiments are gratefully acknowledged. I thank P Wölfle, A Ruckenstein and A Rosch for stimulating discussions. The hospitality of the Institute for Theoretical Physics, University of California, Santa Barbara, supported by the National Science Foundation under Grant No PHY94-07194, is grateful acknowledged. This work was supported by the Deutsche Forschungsgemeinschaft, Grant No Lo 250/9-2.

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