Tuning of Non-Fermi-Liquid Behavior with Pressure

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The Néel temperature of the antiferromagnetic heavy-fermion alloy $CeCu_{5.7}Au_{0.3}$ ($T_N = 0.49$ K for $p = 0$) can be continuously tuned to zero with increasing hydrostatic pressure p. At $p = p_c$ where $T_N = 0$ is reached, the specific heat C varies as $C/T \sim -\ln(T/T_0)$ between 0.1 and 2 K, indicative of non-Fermi-liquid behavior. The observed linear $T_N(p)$ dependence puts rigorous constraints on theories for a quantum phase transition at $T = 0$ between magnetic and nonmagnetic ground states.

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The description of metals in terms of Fermi liquids has been highly successful. The excitations of a Fermi liquid manifest themselves in well-known low-temperature thermodynamic and transport properties, such as the electronic specific heat varying as $C = \gamma T$ with γ independent of T in the limit $T \to 0$, a Pauli spin susceptibility χ independent of T, and a T dependent electrical resistivity contribution arising from particle-particle collisions varying as $\Delta\rho \sim AT^2$. Even for heavy-fermion metals with huge effective masses m^* of the quasiparticles, Fermi-liquid behavior is generally observed with correspondingly huge values of γ, χ, and A compared to conventional metals with, roughly, $\gamma \sim \chi \sim \sqrt{A}$ [1,2]. In these heavy-fermion materials which contain a large concentration of Ce, Yb, or U atoms, often on a regular sublattice, the ground state depends critically on the competition between magnetic intersite interactions, i.e., the indirect Ruderman-Kittel-Kasuya-Yoshida (RKKY) exchange interaction between magnetic moments via the conduction electrons, and the on-site Kondo exchange interaction favoring local singlet formation.

Generally, a very strong antiferromagnetic exchange interaction J between local moments and conduction electrons will result in a nonmagnetic ground state with only short-lived short-range magnetic correlations. Weakening J by increasing the lattice parameter (i.e., reducing the hybridization between conduction electrons and f electrons which enters J) can lead to long-range magnetic order. Such an increase is often brought about by alloying. One example is nonmagnetic $CeCu_6$ where Au expands the lattice and antiferromagnetic order is observed in $CeCu_{6-x}Au_x$ above a critical concentration $x_c \approx 0.1$ [3]. Starting from a long-range ordered state the breakdown of antiferromagnetic order with decreasing x at x_c, where the Néel temperature scales to zero, might signal a zero-temperature quantum phase transition. For such a magnetic-nonmagnetic transition strong deviations from Fermi-liquid behavior are expected to occur [4–6] and were indeed observed recently in a few systems [7], notably in $CeCu_{5.9}Au_{0.1}$ where $C/T \sim -\ln(T/T_0)$, $\chi \sim (1 - \alpha T^{1/2})$, and $\Delta\rho \sim A'T$ was found [8].

Non-Fermi-liquid behavior may also arise for a certain class of single-ion Kondo models. Again, in may cases the Kondo effect may be described in terms of a local Fermi liquid [9]. However, for the special case that the number of conduction-electron channels is larger than $2S$ where S is the impurity spin, non-Fermi-liquid behavior was predicted [10] and recently observed in $U_xY_{1-x}Pd_3$ [11,12], although in this case, too, the proximity to magnetic order was invoked [12]. Also, in low-dimensional metals Fermi-liquid behavior is expected to break down, most dramatically in one-dimensional metals where a Luttinger liquid is formed. Recent photoemission spectroscopy data on quasi-one-dimensional and quasi-two-dimensional metals indicated the absence of a sharp Fermi edge [13,14].

Thus, conceptionally very different scenarios may lead to non-Fermi-liquid behavior. It is therefore of crucial importance to establish a firm experimental basis which can be used to test various theoretical predictions. The present paper reports the first study where the Néel temperature T_N is continuously tuned to zero by pressure in a heavy-fermion alloy in order to look for non-Fermi-liquid behavior in the specific heat of a three-dimensional metal. This allows a clear observation of the scaling of T_N with the relevant variable, in this case pressure, which can be compared to the "concentration scaling" $T_N(x)$. For the present study a $CeCu_{5.7}Au_{0.3}$ alloy was chosen. Earlier work on an $x = 0.5$ alloy has shown that T_N decreased strongly with pressure p [15]. For the present alloy with $T_N = 0.49$ K at zero pressure we find that T_N decreases linearly with p and can indeed be forced to $T_N = 0$ within the accessible pressure range, thus reaching a quantum critical point. After suppression of T_N at a critical pressure p_c, we observe $C/T \sim a\ln(T/T_0)$ with coefficients a and T_0 which are very close to data for $x \approx x_c \approx 0.1$ and $p = 0$. This indicates that the non-Fermi-liquid state is rather unique at the magnetic instability, regardless of how is was reached. Our data allow us to explore different regions in the vicinity of a quantum critical point: (i) the ordered region where T_N scales to zero, and (ii) the "classical region" at the critical point $x = x_c$ or $p = p_c$ but, in fact, at a finite temperature where classical fluctuations dominate over quantum fluctuations.

The sample was a cylinder cut from a $CeCu_{5.7}Au_{0.3}$ single crystal grown by the Czochralski method from the

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constituents (Ce $5N$, Cu $5N$, Au $4N$) as described in detail elsewhere [3]. X-ray diffraction showed a single-phase orthorhombic $CeCu_6$ structure with slight changes of the lattice constants [3]. The specific heat was measured with the sample mounted in a Cu-Be pressure cell with a methanol-ethanol mixture as the pressure-transmitting medium. The pressure was determined at low T via the known $T_c(p)$ relation [16] of a superconducting Sn platelet also mounted in the pressure cell. T_c was measured with the mutual induction method with coils outside the pressure cell. Pressure homogeneity was checked by mounting two Sn platelets ~1 cm apart inside the cell. No difference in the T_c width (~20 mK) could be detected compared to a single platelet. This indicates that the pressure was homogeneous to ~0.1 kbar. The heat capacity of the whole cell was measured with the standard heat-pulse technique. In order to ensure thermal equilibration between cell and sample rather long thermal relaxation times to the bath (~10 min) were chosen. Thus, a lowest measuring temperature of ~0.1 K was reached. The cell contribution to the heat capacity was determined in separate runs for several pressures (0, 3.4, 6, and 8 kbar), showing a negligible pressure dependence, and was subtracted to yield the heat capacity of the sample, which amounted to between 25% and 50% of the total heat capacity.

Figure 1 shows the specific heat C of $CeCu_{5.7}Au_{0.3}$ vs temperature T on a linear scale for several pressures. The rather pronounced specific heat maxima indicate the antiferromagnetic transition. $T_N(p)$ decreases strongly. For $p = 7.1$ and 8.2 kbar T_N has been suppressed to

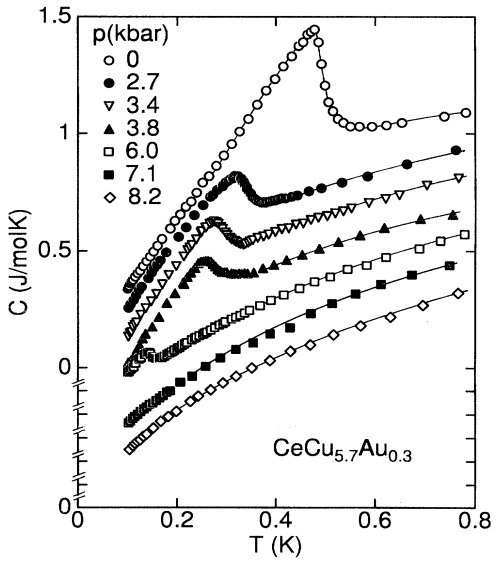

FIG. 1. Specific heat C of $CeCu_{5.7}Au_{0.3}$ vs temperature T for various hydrostatic pressures p. Data sets for each p are shifted downward consecutively by 0.1 J/mol K for clarity. Solid lines are a guide to the eye.

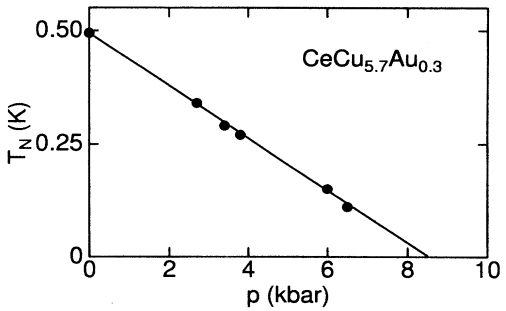

FIG. 2. Néel temperature T_N of $CeCu_{5.7}Au_{0.3}$ vs pressure p. Solid line is a linear fit to the data.

below ≈80 mK. Figure 2 shows T_N vs p. We take the temperature of the inflection point of $C(T)$ as the Néel temperature, rather than the maximum which is somewhat rounded, because experience in the determination of critical exponents tells us that rounding effects always shift the maximum of $C(T)$ toward lower temperatures compared to the "true" T_N. For $p = 0$ this gives $T_N = 0.495$ K compared to 0.48 K obtained from the maximum. However, we verified that the same results for the functional $T_N(p)$ dependence are obtained when taking the C/T maxima. From Fig. 2, we obtain $T_N(p) = 0.495 - 0.058\ \text{kbar}^{-1}/p$ K yielding critical pressure $p_c = 8.5$ kbar where $T_N = 0$.

The question of how the order vanishes at a quantum critical point is of primary importance in the field of quantum phase transitions. For such transitions, static and dynamic correlations are linked by quantum effects. For a magnetic-nonmagnetic transition as in the present case, we write $T_N = |\delta - \delta_c|^{\mu}$ where δ is a relevant scaling variable. Several different theoretical scenarios exist for such a transition. Lacking a microscopic theory, we will assume that, close to δ_c, δ is tuned linearly by pressure or concentration, i.e., $|\delta - \delta_c| \sim |p - p_c|$ or $|\delta - \delta_c| \sim |x - x_c|$. In view of the small volume changes induced by p or x, this assumption is plausible. For the explicit model of competition between Kondo and RKKY interactions [4] the control parameter is assumed to be $\delta = J/W$ where W is the condition-electron bandwidth. The Néel line is suggested to vary as $T_N \sim |\delta - \delta_c|^{\nu z}$ where ν is the correlation-length exponent and z the dynamical critical exponent. From an empirical analysis of several heavy-fermion systems, Continentino concluded that the relation $2 - \alpha = \nu z$ holds instead of the hyperscaling relation $2 - \alpha = \nu(d + z)$ expected for a quantum phase transition [4]. Using this empirical relation and classical tricritical exponents, he obtained $\nu = 1/2$ and $z = 3$. However, this would lead to $\mu = \nu z = 1.5$, i.e., a *superlinear* dependence of $T_N(\delta)$ instead of the experimentally observed linear behavior $T_N(p) \sim p_c - p$ suggesting $\mu = 1$. We note

that our previous finding $T_N(x) \sim x - x_c$ [3] is also compatible with $\mu = 1$. On the other hand, Millis [5] suggests that $T_N \sim |\delta - \delta_c|^{z/(z+1)}$ provided that the temperature dependence of δ is rather weak. Depending on whether the dynamical critical exponent z is 2 or 3 this would lead to $\mu = 2/3$ or $3/4$, i.e., a *sublinear* dependence of $T_N(\delta)$. The sublinear $T_N(\delta)$ dependence would be more pronounced for the antiferromagnetic case where $z = 2$ is expected [5]. However, because of the strong-coupling limit of J for $T \to 0$, an effective $\delta(T)$ taken at $T = T_N$ would be smaller leading to an upward curvature compared to T_N when plotted against $\delta(T \to 0)$. Although it is difficult to estimate this effect quantitatively, it would be surprising if it would exactly cancel with the nonlinear $z/(z + 1)$ exponent.

Figure 3 shows C/T vs $\ln T$ for several pressures. As the magnetic order is suppressed the specific heat gives way to a $C/T = a \ln(T/T_0)$ behavior which is most clearly seen for the sample measured in $p = 8.2$ kbar $\approx p_c$, with $a = -0.65$ J/mol K^2 and $T_0 = 4.83$ K. (The slight upturn for $p = 7.1$ kbar probably indicates $T_N > 0$ but below our lowest measuring temperature.) This clearly shows that the non-Fermi-liquid behavior can be tuned with pressure and constitutes—apart from alloying experiments—the first tuning experiment of the specific heat in a magnetic-nonmagnetic quantum phase transition in a heavy-fermion system, although a number of pressure studies (including studies where the Néel temperature varied under pressure [17]) have revealed the interplay between Kondo compensation and magnetic order [18]. Recent measurements of the resistivity ρ on MnSi, with a long-wavelength helical magnetic structure resembling a ferromagnet for short distances, have shown that $T_c \sim |p - p_c|^{1/2}$ [19]. Also, the T dependence of ρ close to p_c is much weaker than the Fermi-liquid T^2 law and attributed to scattering from spin fluctuations [19].

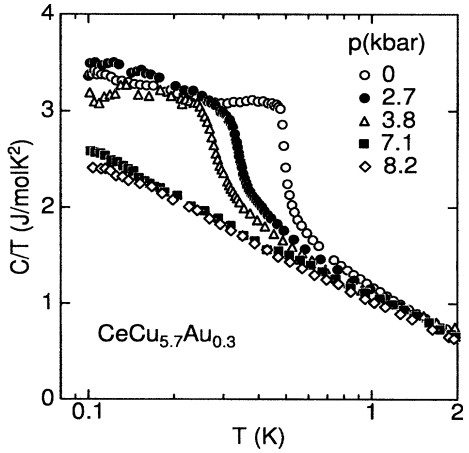

FIG. 3. Specific heat C of $CeCu_{5.7}Au_{0.3}$ plotted as C/T vs $\ln T$ for various hydrostatic pressures p.

It is apparent that the entropy between $T = 0.1$ and 2 K is strongly reduced with pressure. Extrapolating C/T linearly from the data between 0.2 and 0.1 K to $T = 0$ for the magnetically ordered state ($p = 0$) we have $S(2\text{ K}) = 3.23$ J/mol K corresponding to $0.561R \ln 2$. With increasing p, $S(2\text{ K})$ decrease roughly linearly until $S(2\text{ K}) = 2.36$ J/mol K or $0.410R \ln 2$ for $p \approx p_c$. This may indicate that some degrees of freedom are shifted to higher temperature by the hydrostatic pressure, opposite to the naive expectation that the suppression of magnetic ordering enhances the low-T entropy. Also, at higher T crystal-field excitations come into play [20]. For $CeCu_6$ it is known that the Kondo temperature strongly increases under pressure leading to a transition from a heavy-fermion to an intermediate-valent system [21]. The corresponding decrease of the low-T specific heat has indeed been observed, where the entropy between $T = 0$ and 1.4 K as inferred from these data [22] decreases smoothly from 1.7 to 1.0 J/mol K between $p = 0$ and 8.8 kbar.

We now turn to a more detailed discussion of the behavior of the specific heat at the critical point where $T_N(p) = 0$, i.e., $p = p_c$ for the present $CeCu_{5.7}Au_{0.3}$ sample, and compare it in Fig. 4 to our earlier result of C for $T_N(x) = 0$, i.e., for the single crystal with $x = x_c$ [8]. The striking result is that both data sets obey perfect scaling with respect to each other regardless of how $T_N = 0$ was achieved. This "universality" is far from trivial, since the pressure dependence $T_N(p)$ yields a volume dependence $dT_N^{(p)}/dV$ (obtained via the compressibility of $CeCu_6$) different from the concentration dependence $dT_N^{(x)}/dV$ (obtained via the lattice constants of $CeCu_{6-x}Au_x$) [23]. This difference can probably be attributed to band-structure and/or anisotropy effects induced by alloying with Au, besides the lattice expansion which is the prime mechanism establishing magnetic order when starting from $CeCu_6$ [15,23]. Thus, the two non-Fermi-liquid critical points $x = x_c \approx 0.1$, $p = 0$ and $x = 0.3$, $p \approx p_c = 8.5$ kbar

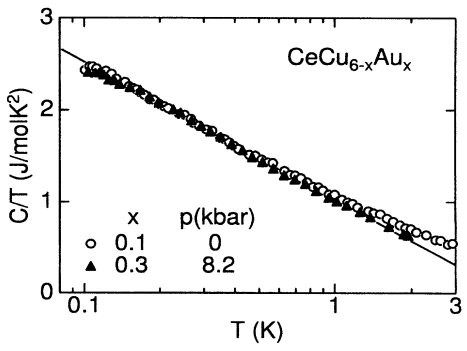

FIG. 4. Specific heat C of two $CeCu_{6-x}Au_x$ alloys for which $T_N = 0$ is reached by alloying or pressure, plotted as C/T vs $\ln T$. Solid line indicates fit of $C/T = a \ln(T/T_0)$ to the data for $x = 0.3$, $p = 8.2$ kbar.

are microscopically distinct but show the same critical behavior.

The apparent $C/T \sim -\ln(T/T_0)$ behavior is predicted in both classical ($\delta = \delta_c$) and quantum-critical regimes (no long-range magnetic order) for $d = 3$ and $z = 3$, i.e., the case expected of a ferromagnet [5]. For $z = 2$, one expects $C/T \sim 1 - \alpha T^{1/2}$ and $C/T \sim 1 - \alpha' T^2$ for classical and quantum critical regimes, respectively (again neglecting a possible temperature dependence of δ for the latter case). Clearly, our data cannot be described by a power-law behavior. A log-log plot of C/T vs T (not shown) exhibits a continuous downward curvature in the whole T range. Also, as discussed above, our $T_N(p)$ data are more consistent with a larger z. We also note that $C/T \sim -\ln(T/T_0)$ is consistent with $d = 3$, $z = 3$ in the more general approach by Sachdev [6]. Thus, our work puts important constraints on the theory of quantum critical points. In particular, from the temperature dependence of the specific heat we infer $z = 3$. Also, the pressure dependence of T_N close to the quantum critical point is more compatible with a larger z. Inelastic neutron scattering studies are underway to probe directly the dynamics at this unique quantum phase transition.

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