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Inducement of non-Fermi-liquid behavior with a magnetic field

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Using Ag doping of the nearly magnetic heavy-fermion system $CeCu_6$ to vary the onset of antiferromagnetism between $T_N=0$ and 0.8 K, we have found a large region of the phase diagram where magnetic field can reach the quantum critical point $(T_N \rightarrow 0)$ with sufficiently strong antiferromagnetic correlations remaining to produce non-Fermi-liquid (NFL) behavior. In this field and composition regime, the $CeCu_{6-x}Ag_x$ samples exhibit the typical NFL temperature dependencies for the various measured parameters over a broad range of temperature down to 100 mK. Application of higher fields to these samples causes entry into the Fermi-liquid regime. Due to the ease of changing magnetic fields when establishing the phase diagram at low temperatures, the increased efficacy of field suppression of T_N versus that achieved by pressure, and the fact that magnetic field does not change the volume as do both pressure and doping, magnetic field offers distinct advantages as a method for exploring the crossover between antiferromagnetic, non-Fermi-liquid, and Fermi-liquid behavior in the vicinity of the quantum critical point. [S0163-1829(98)51008-9]

Since the discovery[1] of the particular subset of non-Fermi-liquid (NFL) behavior characterized by a specific heat $C \propto -T \log T$ in $U_{0.2}Y_{0.8}Pd_3$, the entire field of NFL research has been quite active. A ''Fermi liquid'' (FL) is a metal where, at low temperatures, the electron-electron interactions are of short range and both C/T and the susceptibility χ are constant in a range (typically $\leqslant 1$ K) above $T=0$, while the resistivity ρ is proportional to $\rho_0 + AT^2$ in the same temperature range. If the interactions are, however, long range then the metal will show deviations from these FL temperature dependencies and may be called a non-Fermi-liquid (NFL) system. Thus, pressure experiments[2] on $ZrZn_2$ show $\rho \propto \rho_0 + T^{1.6}$, which was sufficient for the authors to apply the ''NFL'' label. A certain subclass of NFL systems exhibit a divergence in C/T at low temperatures $\propto -\log T$, a resistivity linear in $T(\rho \propto \rho_0 + BT)$, and a temperature-dependent χ (either $\propto 1-\sqrt{T}$ or $-\log T$). Due to the growing numbers of systems showing this subclass of NFL behavior, it seems possible that the underlying physical phenomena producing the requisite long-range correlations in these materials may be of some universal nature. Experiments have tended to focus on magnetic fluctuations, remaining in a system where the magnetism has been suppressed via doping or pressure, as providing the long-range interactions needed to prevent entering the FL state at low temperatures. This ''nearness to magnetism'' has long been recognized[3–4] as important for achieving NFL properties and is the reason why heavy-fermion, highly correlated, electron systems (which are, in general, near to a magnetic instability) provide such a rich research area for NFL physics. Different theoretical mechanisms have been proposed, including that of a quantum critical point (QCP),[3,5–7] single-ion quadrupolar Kondo model,[8] or a distribution of low-lying T_{Kondo} states induced by disorder.[9–12] Various predictions for the NFL temperature dependencies and critical exponents have been put forward.[13–14]

In this paper, where we investigate whether tuning $T_N \rightarrow 0$ using an applied magnetic field can produce NFL behavior, the QCP picture seems, based on symmetry[8] and the non-disorder-inducing nature of a magnetic field, to be the appropriate model. Quantum critical phenomena were first discussed in Ref. 15, followed by numerous other works (see, e.g., Refs. 5–7), and have been recently reviewed.[16] The central premise is that[15] a QCP exists, tuned to by some external parameter, when the fluctuation modes (here the magnetic correlations) in the order parameter are of (predominantly[7]) higher energy than the energy $k_B T_{\text{long-range order}}$ (here T_{Neel}). Since an applied magnetic field can suppress long-range antiferromagnetic order just as adjusting composition or pressure can, leaving the long-wavelength fluctuations called for in the theory,[3,5–7,16] this seemed a natural experiment to try. The mechanism by which the long-range order is suppressed can be different for B, x, and P—e.g., B_c overcomes the negative Ruderman-Kittel-Kasuya-Yosida exchange coupling parameter J, while doping and pressure change the hybridization to reach x_c or P_c by tuning the Kondo screening to fully screen the local moments. In all cases the local moments are still present, just prevented from long-range order.

Studies of NFL behavior induced via doping are quite numerous (see Ref. 17 for a review). Studies of NFL behavior induced with pressure, due to the difficulty of the experiments, have been carried out on antiferromagnets only for Au-doped $CeCu_6$,[18] Ce_7Ni_3 (Ref. 19) and $CePd_2Si_2$.[20] Due to our experience[21] with $CeCu_{6-x}Ag_x$, which has similar properties to $CeCu_{6-x}Au_x$, since T_N for doped $CeCu_6$ is a factor of 10 lower than in $CePd_2Si_2$, and since Ce_7Ni_3, which has a complex hexagonal structure, requires[22] long-term annealing, we chose $CeCu_{6-x}Ag_x$ for our first attempt to induce NFL behavior with field.

Samples of $CeCu_{6-x}Ag_x$, $x=0.09$, 0.48, 1.2, were prepared via arc melting together stoichiometric amounts of the pure elements, using Ames Ce and six 9's Cu and Ag, under a purified argon atmosphere. Samples were x-rayed; lattice parameters are shown in Table I. All samples were single phase according to the x-ray results. Magnetic susceptibility measurements down to 1.8 K and up to 7 T were made using a Quantum Design magnetometer. Measurements of the re-

TABLE I. Parameters for $\mathrm{CeCu}_{6-x}\mathrm{Ag}_x$.

	$x=0.09$ $B=0$ T	$x=0.48$ $B=4$ T	$x=1.2$ $B=3$ T
a[Å]	8.11	8.19	8.36
b[Å]	5.10	5.09	5.09
c[Å]	10.16	10.28	10.44
V[Å^3]	420.4	428.4	443.7
T_N[K]	0	0.5	0.8
$\rho=\rho_0+Bt^{\alpha}$			
with α	1.1 ± 0.1		1.0 ± 0.1
T range of validity [K]	0.05–0.50		0.1–0.4
$\chi\propto 1-\sqrt{T}$			
T range of validity [K]	1.8[a]–5	1.8[a]–5	1.8[a]–6 ($\chi\propto 1-\log T$)
$C/T\propto -\ln T$			
T range of validity [K]	0.3[a]–3	0.4[a]–3	0.3[a]–3
β[b]	0.85	1.35	1.7

[a] Lowest temperature of measurement.

[b] From scaling.

sistivity in field were performed down to 0.030 K in a dilution refrigerator, while specific heat in fields to 13 T was performed in a ^{3}He rig using established methods.[4,21]

$\mathrm{CeCu}_{5.91}\mathrm{Ag}_{0.09}$ corresponds[21] to x_c in $\mathrm{CeCu}_{6-x}\mathrm{Ag}_x$ where T_N, which is 0.8 K for $x=1.2$, is first suppressed to $T=0$. As expected from the similar composition tuning experiments[23–24] for $\mathrm{CeCu}_{6-x}\mathrm{Au}_x$, at this critical concentration the low-temperature dependencies of the zero-field resistivity ρ (Fig. 1), dc magnetic susceptibility χ (not shown), and specific heat divided by temperature C/T (not shown) exhibit non-Fermi-liquid behavior. (See also Table I.)

As the Ag doping is increased, T_N increases to 0.5 K for $x=0.48$ and to 0.8 K for $x=1.2$ (see the phase diagram in Fig. 2 and Table I). Can magnetic field depress the antiferromagnetism back to $T=0$ and leave sufficient magnetic fluctuations to achieve the NFL behavior found in the neighborhood of the QCP via pressure tuning or doping? We consider first the maximum Ag concentration, i.e., $\mathrm{CeCu}_{4.8}\mathrm{Ag}_{1.2}$, where T_N is highest and one can expect the greatest resistance of these fluctuations to field suppression. As can be seen in Fig. 3 for the specific heat, in 0 and 2 T, $\mathrm{CeCu}_{4.8}\mathrm{Ag}_{1.2}$ is magnetic, while for 3 T ($=B_c$) the magnetism is just suppressed and C/T is in fact linear in $\log T$ over a decade in temperature. Thus, $\mathrm{CeCu}_{4.8}\mathrm{Ag}_{1.2}$—via the usual criterion that $C/T\propto -\log T$ over a decade of temperature—does in fact exhibit NFL behavior induced via the application of a certain critical magnetic field.

As the magnetic field is raised to 4 T, the low-temperature C/T data in Fig. 3 show a distinct departure from $\log T$, with a bend at 1 K. Further increases in field suppress the long-wavelength magnetic fluctuations which exist at low temperatures at B_c and recover (see phase diagram, Fig. 2) the FL behavior, i.e., C/T in 5 and 13 T remains essentially temperature independent at low temperature. This behavior with field (NFL behavior at B_c and entry into a FL regime for higher fields) is echoed in the resistivity data for $\mathrm{CeCu}_{4.8}\mathrm{Ag}_{1.2}$ as a function of field shown in Fig. 1 for 3 and 7 T. Thus, $\rho=\rho_0+BT$ between 0.1 and 0.4 K for $B_c=3$ T, but already for 7 T, $\rho=\rho_0+AT^2$, i.e., FL behavior, over a decade in temperature. As B_c is approached from above, the region where $\rho=\rho_0+AT^2$ shrinks (at 4 T, the resistivity is quadratic with temperature only up to 0.120 K) while the coefficient of the quadratic term increases dramatically—see the inset in Fig. 1. At 13 T, $\rho=\rho_0+AT^2$ up to 1.5 K, while $A(4\,\mathrm{T})$ is twenty times larger than $A(13\,\mathrm{T})$—dramatic evidence that increasing field suppresses the long-range magnetic correlations above $B(\mathrm{QCP})$, thus enhancing the temperature extent of the FL behavior monotonically with increasing field.

The $1-\sqrt{T}$ behavior found in χ_{dc} (not shown) below 5 K for $\mathrm{CeCu}_{5.91}\mathrm{Ag}_{0.09}$, which is at the QCP in zero field, changes to ($-\log T$) for $\mathrm{CeCu}_{4.8}\mathrm{Ag}_{1.2}$ at B_c, a temperature dependence seen[8] for a number of NFL systems. Below B_c, χ vs T rises at low temperature due to the magnetic fluctuations above $T_N(B)$; above B_c there is a distinct tendency towards saturation in χ at low T and 7 T causes χ to bend over and trend towards a constant at the lowest temperature of measurement, i.e., FL behavior is also achieved with $B>B_c$ for the susceptibility of $\mathrm{CeCu}_{4.8}\mathrm{Ag}_{1.2}$.

To compare to other, nondoping experiments near the QCP, these specific heat, resistivity, and susceptibility results as a function of magnetic field are very analogous to results as a function of pressure[25] found by Umeo, Kadomatsu, and Takabatake[19] for $\mathrm{Ce}_7\mathrm{Ni}_3$ [$T_N(P=0)=1.8$ K, $P_c\cong 5.4$ kbar], where pressure suppresses an antiferromagnetic transition reaching NFL behavior at P_c with further pressure reaching the Fermi-liquid regime, $C/T\propto$const and $\rho=\rho_0+AT^2$. In fact, our $C(B)/T$ data in Fig. 3 are very reminiscent of the $C(T)/T$ data for $\mathrm{Ce}_7\mathrm{Ni}_3$ in Fig. 6, Ref. 19. Additionally, in good agreement with our resistivity data (Fig. 1) as a function of field, Umeo, Kadomatsu, and Takabatake see an increase of more than a factor of 30 in the resistivity quadratic term "A" coefficient upon decreasing the pressure in the FL regime from 15 kbar down towards $P_c=5.4$ kbar. At the same time the temperature range where $\rho=\rho_0+AT^2$ decreases dramatically as $P\rightarrow P_c$.

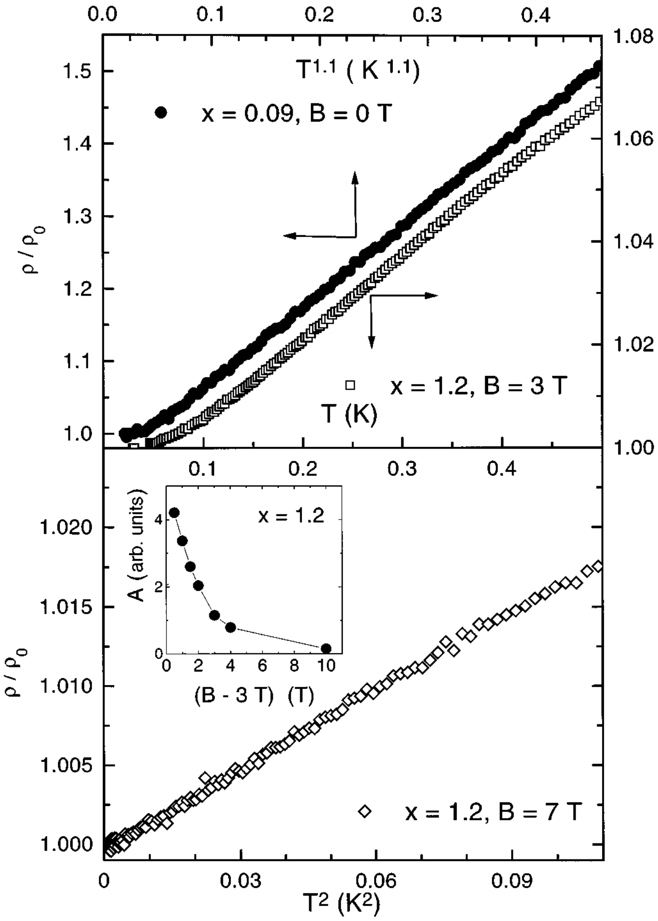

FIG. 1. The resistivity normalized by $\rho(T\rightarrow 0)$, ρ_0, plotted vs $T^{1.1}$ for $CeCu_{5.91}Ag_{0.09}$ in zero magnetic field shows linear, i.e., NFL, behavior up to 0.5 K. The resistivity for $CeCu_{4.8}Ag_{1.2}$ in $B=3$ T normalized by ρ_0 (3 T) is shown plotted vs T and displays no simple $\rho\propto T^{\alpha}$ over any appreciable range. Between 0.1 and 0.4 K the data are approximately linear in T, and then bend over at higher temperatures while between 0.03 and 0.13 K the data can be fitted to $T^{2\pm0.2}$. As the field is increased, the resistivity of $CeCu_{4.8}Ag_{1.2}$ begins to follow $\rho=\rho_0+AT^2$ over a broader temperature range (for $B=5$ T, $0.04\ \mathrm{K}<T<0.2$ K, for $B=6$ T, up to 0.28 K) until, as shown here for $B=7$ T, $\rho=\rho_0+AT^2$ over a decade in temperature. As the region where $\rho-\rho_0\propto T^2$ grows with increasing field, the coefficient A decreases (see inset).

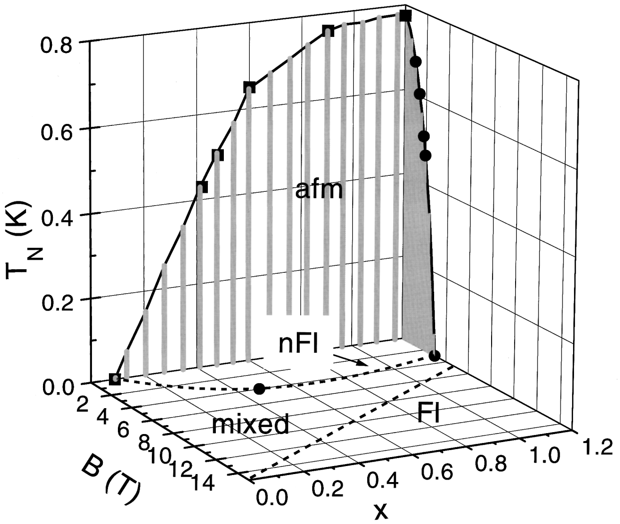

FIG. 2. Shown is a phase diagram detailing the dependence in $CeCu_{6-x}Ag_x$ of T_{Neel} on both Ag content (x) (data partly from Ref. 21) and field. Also, in the $T_N=0,B,x$ plane the line of $B_c(x)$ where NFL behavior is observed in C, ρ, and χ is shown defining the QCP's reached via tuning $T_N\rightarrow 0$ as a function of x and B. In this plane for $B>B_c$ a mixed regime is entered where C/T is neither $\propto -\log T$ (NFL) nor $\propto$const (FL). At higher fields, $C/T\propto$const, χ trends toward being const, and $\rho\propto\rho_0+AT^2$ as discussed in the text—defining FL behavior. For $x=1.2$, this regime is reached by 5 T, whereby for pure $CeCu_6$, $C/T\propto$const at low temperatures first at 14.5 T (Ref. 32).

The inducing of NFL behavior in $CeCu_{6-x}Ag_x$ in C, ρ, and χ with the application of magnetic field, followed by entry into the Fermi-liquid state with increasing field, reported here, has no direct comparison with previous field work on $CeCu_2Si_2$ (Ref. 26) and UBe_{13}.[27] This is because (a) $CeCu_2Si_2$ and UBe_{13} exhibit no decade of temperature where $C/T\propto -\log T$ is achieved, (b) higher (12 vs $B_c=4$ T) field does not[26] flatten out C/T as $T\rightarrow 0$ in $CeCu_2Si_2$ or, in the case[27] of UBe_{13}, has not ($B>B_c=12$ T) been measured. However, the entry into the FL state in $CeCu_{6-x}Ag_x$ above B_c observed in the resistivity reported here is analogous to the resistivity results[26] on $CeCu_2Si_2$, where field first suppresses superconductivity to reveal $\Delta\rho\propto T^{3/2}$ which changes to $\Delta\rho\propto T^2$ above 6 T. Similarly, there are numerous examples of the use of field to *destroy* NFL behavior (after $T_N\rightarrow 0$, sufficient field suppresses the remaining magnetic fluctuations as also seen in the present work) and achieve the FL state, primarily in doped systems (see, e.g., Refs. 18, 28–29) but also in the work[26–27,30] on pure $CeNi_2Ge_2$.

In order to fill in our phase diagram (Fig. 2) between $x=0.09$ and $x=1.2$, we also measured the specific heat of $CeCu_{5.52}Ag_{0.48}$ and found that $C/T\propto -\log T$ in 4 T (not shown) rather than 3 T as found for $x=1.2$.[31] (See Table I.)

As a final important probe of the phase diagram near the QCP, it has been shown[3,28] experimentally (see also Ref. 5 for a more detailed theoretical analysis) for several NFL systems that in the crossover regime near the QCP that $\Delta C(B)/T$ scales with B/T^{β} and that β is a determinant for

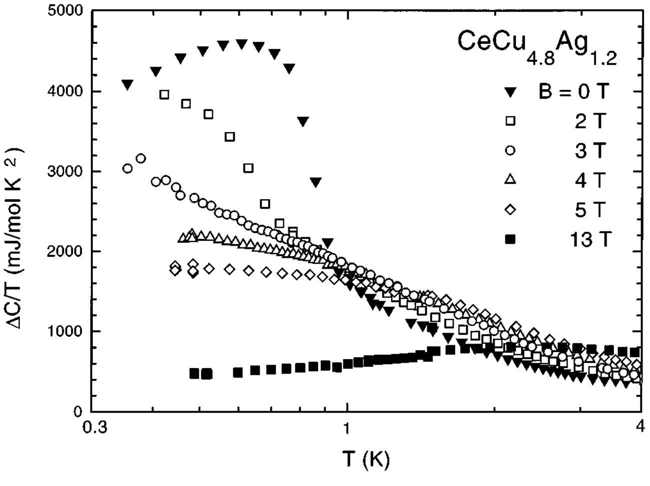

FIG. 3. Specific heat C of $CeCu_{4.8}Ag_{1.2}$ minus $C(LaCu_6)$ vs $\log T$ in magnetic fields between 0 and 13 T. For $B=3$ T, $\Delta C/T$ vs $\log T$ is a straight line over a decade of temperature.

whether the interactions in the critical regime are not of single-ion nature ($\beta > 1$). These scaling results were always for a system where the QCP was reached *via doping*. We find in the present work for $\mathrm{CeCu_{5.91}Ag_{0.09}}$, and using field to tune $T_N \rightarrow 0$ for $\mathrm{CeCu_{5.52}Ag_{0.48}}$ and $\mathrm{CeCu_{4.8}Ag_{1.2}}$ that indeed $[C(B) - C(0)]/T$ scales with B/T^{β}, with $\beta = 0.85$, 1.35, and 1.7, respectively, when B is replaced by $\Delta B (= B - B_c)$ and $C(0)$ is replaced by $C(B = B_c)$. Although we are not aware of a theory thereto, it should also be possible in experiments where the QCP is reached, and exceeded, by pressure to scale the results for $P > P_c$ to $\Delta P/T^y$, where y is of course a different exponent than β.

In conclusion, resistivity, susceptibility, and specific heat results as a function of field and doping in the antiferromagnet $\mathrm{CeCu_{6-x}Ag_x}$ have shown, for a broad range of doping, that rather modest magnetic fields are an effective tool in mapping out NFL behavior in the neighborhood of the QCP, with higher fields—just as for pressure—producing FL behavior. The application of magnetic field to reach the QCP allows a determination of the critical scaling exponent β over a much broader range of composition in the phase diagram than previously when scaling could only be investigated for the critical concentration, x_c, where T_N is just suppressed to $T = 0$. It is to be expected that other antiferromagnetic systems will display similar behavior, although complexities in the phase diagram certainly exist.

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