Non-Fermi-Liquid Behavior within the Ferromagnetic Phase in $URu_{2-x}Re_xSi_2$

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The $URu_{2-x}Re_xSi_2$ system exhibits ferromagnetic order for Re concentrations $0.3 < x \leq 1.0$. Non-Fermi-liquid (NFL) behavior is observed in the specific heat for $0.15 \leq x \leq 0.6$ [$C/T \propto -\ln T$ (or $T^{-0.1}$)], and also in the power-law T dependence of the electrical resistivity [$\rho(T) \propto T^n$] with $n < 2$ for $0.15 \leq x < 0.8$, at low T, providing strong evidence that the NFL behavior persists within the ferromagnetic phase. Furthermore, the deviation of the physical properties of $URu_{2-x}Re_xSi_2$ from Fermi-liquid behavior is most pronounced at the ferromagnetic quantum critical point, and the NFL behavior found in the ferromagnetic phase may be consistent with the Griffiths-McCoy phase model.

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Strong electronic correlations in f-electron materials give rise to an assortment of interesting phenomena. A large number of heavy fermion intermetallic compounds containing Ce, Yb, or U have been found to have unusual low temperature properties that appear to violate the Fermi-liquid paradigm [1,2]. These non-Fermi-liquid (NFL) materials exhibit weak power-law or logarithmic T dependences in various physical properties such as magnetic susceptibility χ, specific heat C, and electrical resistivity ρ, which are at odds with those of a Fermi liquid [i.e., $C/T \sim \chi \sim$ const, and $\rho(T) \propto T^2$]. Such unusual low-T behavior is most often observed at a quantum phase transition (QPT), in close proximity to where a magnetic phase transition is suppressed to zero temperature.

A number of theories with distinct origins have been put forth to explain the NFL behavior in these heavy fermion systems. Models [3,4] based on an unconventional Kondo effect are consistent with such NFL systems as $Y_xU_{1-x}Pd_3$ [1]. Since many NFL compounds are disordered alloys, disorder-driven mechanisms have been proposed such as the Kondo disorder model [5,6] and a Griffiths-McCoy phase model [7]. Other models consider the behavior near a second-order QPT, or quantum critical point (QCP), based on renormalization-group techniques (in the clean limit [8] or with the inclusion of disorder [9]) or spin-fluctuation models [10,11].

To date, most research on NFL physics has focused on antiferromagnetic (AFM) QPTs; comparatively little is known about ferromagnetic (FM) QPTs by studies of heavy fermion systems such as $Th_{1-x}U_xCu_2Si_2$ [12]. The largest body of evidence for a FM QPT is found in the d-electron systems (e.g., Ni_xPd_{1-x} [13], MnSi [11], $ZrZn_2$ [11]). In these cases, the predictions of phenomenological 3D spin-fluctuation models [10,11], e.g., $C/T \propto -\ln T$, $\rho \propto T^{5/3}$, $\chi \propto T^{-4/3}$, and variation of the Curie temperature with control parameter α, $T_C \propto |\alpha - \alpha_c|^{3/4}$ where α_c corresponds to a QCP, describe the NFL properties reasonably well. However, experiments indicate that MnSi [14] and $ZrZn_2$ [15] undergo a first-order QPT, and general understanding of QPTs remains incomplete.

In this Letter, we report the first example of non-Fermi-liquid behavior observed deep within the ferromagnetic phase of the heavy fermion system $URu_{2-x}Re_xSi_2$, in which $C(T)/T \propto -\ln T$ (or $T^{-0.1}$) and $\rho(T) \propto T^n$ with $n \sim 1.2$ over more than a decade in temperature below 20 K for $x = 0.6$. Furthermore, it is one of the rare examples in which the NFL behavior occurs within the magnetic phase [16]. The Griffiths-McCoy phase model [7] may provide an explanation for the coexistence of ferromagnetism and NFL properties since the Griffiths singularities which give rise to the NFL power-law behavior are predicted to occur on both sides of the FM QCP [17]. The substitution of Re for Ru in URu_2Si_2 rapidly suppresses superconductivity from $T_{SC} = 1.5$ K in URu_2Si_2 to $T_{SC} = 0.23$ K at $x = 0.01$ and also suppresses the AFM (or hidden order [18]) transition to $T_N \sim 13$ K at $x = 0.1$ [19]. Further investigations [20] of the $URu_{2-x}M_xSi_2$ ($M =$ Re, Tc) systems revealed the first occurrence of a FM instability in a heavy fermion system as shown in Fig. 1. The physical properties of $URu_{2-x}Re_xSi_2$, such as the saturation moment μ_s, T_C, and electronic specific heat coefficient γ exhibit maxima with Re concentration, reaching values $\mu_s \sim 0.44\mu_B$/U atom and $T_C = 38$ K at $x = 0.8$, and $\gamma \sim 160$ mJ/mol K^2 at $x = 0.6$. The FM order was confirmed by neutron scattering experiments [21] for $x = 0.8$ and NMR measurements for $x = 0.4$ [22].

Polycrystalline samples of $URu_{2-x}Re_xSi_2$ for $0.125 \leq x \leq 0.7$ were prepared by arc melting appropriate amounts of URu_2Si_2 and URe_2Si_2 (Ref. [19]). Electrical resistivity, specific heat, and magnetic susceptibility measurements between $\sim$1 and 300 K were made in a ^{4}He cryostat, in a semiadiabatic ^{3}He calorimeter, and with a SQUID magnetometer, respectively. Measurements of $\rho(H, T)$ down to 60 mK were performed in a ^{3}He-^{4}He dilution refrigerator. The absolute magnitude of ρ could not be determined due to the presence of microcracks in the samples. Electron probe microanalysis (EPMA) was performed on a commercial spectrometer on $URu_{2-x}Re_xSi_2$ samples with $0 \leq x \leq 0.9$. The characteristic x-ray lines were measured using wavelength dispersive spectroscopy at eight to ten locations on the sample. The actual Re concentration x was within 4% (7%) for $x \leq 0.6$ ($x > 0.6$) of the nominal concentration, e.g., $x_{\mathrm{EPMA}} = 0.309(1)$ for $x = 0.3$, indicating a homogeneous distribution of Re (nominal concentrations are used hereafter).

We first focus our attention on determining the ferromagnetic critical point in the $URu_{2-x}Re_xSi_2$ system which is deduced solely from the magnetic properties since no FM anomaly is observed in either $C(T)$ or $\rho(T)$, similar to the behavior of some other ferromagnetic systems such as $Th_{1-x}U_xCu_2Si_2$ [12] and $ZrZn_2$ [23]. The Curie temperatures for $0.275 \leq x \leq 0.5$ determined from a modified Arrott plot analysis, i.e., plotting $M^{1/\beta}$ vs $(H/M)^{1/\gamma}$ [where β and γ are critical exponents defined as $M(T, H = 0) \propto T^{\beta}$, $M(T = 0, H) \propto H^{1/\delta}$, $\gamma = \beta(\delta - 1)$], are $T_C = 1.9$, 4.6, and 8.2 K for $x = 0.275$, 0.35, and 0.5, respectively, as shown in Fig. 2(a) for $x = 0.35$. This analysis is based on the Arrott-Noakes equation $(H/M)^{1/\gamma} = (T - T_C)/T_1 + (M/M_1)^{1/\beta}$, where T_1 and M_1 are normalization factors [24]. The ordering temperatures determined from an Arrott plot analysis increase from $T_C = 12$ K for $x = 0.5$ to a maximum value of $T_C \sim 38$ K for $x = 0.8$ (Fig. 1) [20]. In addition, the Curie-Weiss temperatures deduced from $\chi(T)$ measurements at $H = 20$ Oe are slightly lower than, though generally consistent with, the T_Cs obtained from the Arrott plots (Fig. 1). The ferromagnetic QCP is estimated to be $x_c^{\mathrm{FM}} = 0.30(5)$ by a linear extrapolation of the T_Cs determined from these various types of analysis of the magnetization data.

The specific heat C/T vs $\ln T$ of $URu_{2-x}Re_xSi_2$ for $0.15 \leq x \leq 1.0$ is shown in Fig. 3. Below $\sim$5 K, C/T exhibits a logarithmic temperature dependence indicative of non-Fermi-liquid behavior for $0.15 \leq x \leq 0.6$. It is interesting to note that this logarithmic behavior persists well into the FM region of the phase diagram, and, in particular, the $x = 0.6$ sample displays such behavior over the largest temperature interval ($0.6 \leq T \leq 7.3$ K). For $x = 0.8$ and $x = 1.0$, however, C/T is (nearly) independent of temperature for $0.6 < T < 6$ K; this return to FL behavior is supported by a T^2 dependence of the electrical resistivity [20]. The C/T data for $0.15 \leq x \leq 0.6$ were fit by $C/T = \gamma_0 - c_0 \ln T$ where γ_0 and c_0 are constants. The upper T range of these fits is extended by 1–3 K, if, as an approximation, the nonmagnetic contribution of $ThRu_2Si_2$ [25] is subtracted from the $URu_{2-x}Re_xSi_2$ data. The value of c_0 reaches a maximum at $x \sim 0.3$ as shown in the inset of Fig. 3, close to the FM QCP; similar behavior is also observed in $Ni_{1-x}Pd_x$ [13]. Fits of the data to a power law ($C/T \propto T^{-n}$) over a similar temperature range yield an exponent $n \sim 0.2$ for $x = 0.15$–0.4 and $n \sim 0.1$ for $x = 0.6$. Extrapolation of the logarithmic (or power-law) T dependence of C/T to $T \rightarrow 0$ K for $x = 0.15$–1 gives an entropy $S \sim 1$ J/mol K, in agreement with the expected entropy for a Kondo system with a large $T_K \sim 100$ K [26]; this is taken as evidence that the NFL behavior in $URu_{2-x}Re_xSi_2$ is a bulk phenomenon and not an impurity or inhomogeneity effect.

The normalized electrical resistivity $\rho/\rho(300\text{ K})$ vs T of $URu_{2-x}Re_xSi_2$ for $0.2 \leq x \leq 0.8$ is shown in the inset of Fig. 4. The resistivity for all x exhibits a weak T dependence above $\sim$100 K, followed by a maximum, then decreases rapidly due to coherence effects in the f-ion sublattice. Below $\sim$20 K, $\rho(T)$ of $URu_{2-x}Re_xSi_2$ for

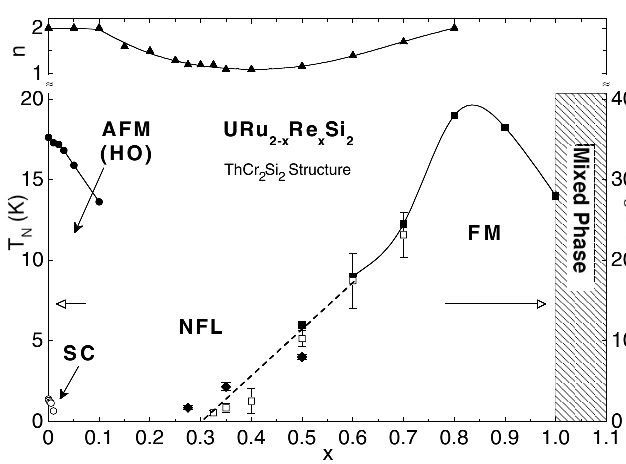

FIG. 1. Magnetic phase diagram of $URu_{2-x}Re_xSi_2$ showing the antiferromagnetic or hidden order (AFM or HO, filled circles) [19], superconducting (SC, open circles) [19], and ferromagnetic [FM, filled squares [20]: Arrott plot analysis; filled diamonds: modified Arrott plots; open squares: low-field $\chi(T)$ data] regions. The power-law exponent of $\rho(T)$, n, is shown in the top part of the diagram.

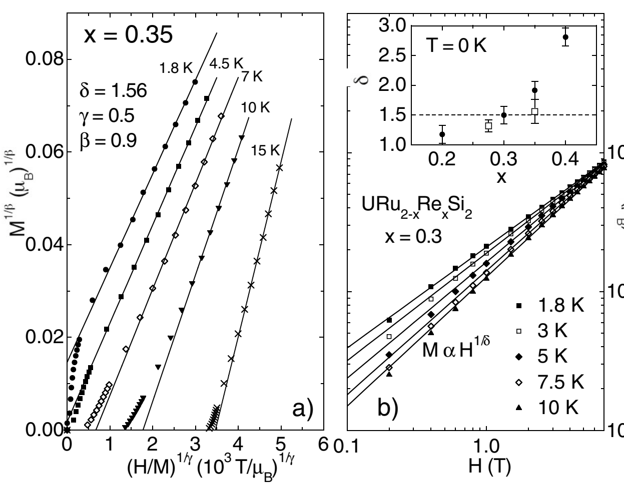

FIG. 2. (a) Modified Arrott plot $M^{1/\beta}$ vs $(H/M)^{1/\gamma}$ for $x = 0.35$. (b) M vs H of $URu_{2-x}Re_xSi_2$ for $x = 0.3$. The lines are fits to $M = H^{1/\delta}$. Inset: $\delta(0)$ vs x obtained from a linear extrapolation of the $\delta(T)$ data (closed symbols) or by the modified Arrott plot analysis (open symbols) discussed in the text.

$0.15 \leq x \leq 0.7$ follows a power-law T dependence $[\rho - \rho_0]/(\rho(300\text{ K})) \propto T^n$, where ρ_0 is a constant, as displayed in Fig. 4. The exponent n decreases from 1.6(1) for $x = 0.15$ to 1.1(1) for $0.2 < x < 0.5$ before increasing to 1.7(1) for $x = 0.7$, as shown in Fig. 1. Thus, NFL behavior coexists with ferromagnetism in the range $0.3 < x \leq 0.7$, consistent with the $C(T)$ measurements. Furthermore, magnetoresistivity measurements for $x = 0.6$ (required to suppress possible impurity superconductivity at $T_{SC} \sim 1$ K) reveal NFL power-law behavior for $H \geq 40$ kOe with $n = 1.4$ from $0.08 \leq T \leq 13$ K ($n = 1.2$ above 1 K for $H < 40$ kOe) [27], suggesting the NFL characteristics are robust in magnetic fields and extend to the lowest temperatures. The power-law exponent for $x = 0.2$ is $n = 1.2$ ($0.15 \leq T \leq 6.2$ K) with an apparent crossover to FL behavior below $T^* \sim 0.15$ K. A return to a FL ground state occurs for $x \leq 0.1$ and $x \geq 0.8$.

The magnetic susceptibility of $URu_{2-x}Re_xSi_2$ in the paramagnetic region ($0.15 \leq x \leq 0.3$) also exhibits NFL properties in which $\chi(T)$ can be described by a power law $\chi(T) \propto T^{-n}$ below 5 K (not shown). The exponent n increases from $\sim$0.2 for $x = 0.2$ to $\sim$0.4 for $x = 0.3$.

The magnetic phase diagram of $URu_{2-x}Re_xSi_2$ is shown in Fig. 1. There is evidence for non-Fermi-liquid behavior in the $URu_{2-x}Re_xSi_2$ system from $\rho(T)$, $\chi(T)$, and $C(T)$ measurements in both the paramagnetic and the FM regions of the phase diagram ($0.15 \leq x \leq 0.3$). Both the logarithmic divergence in $C(T)$ and the power-law behavior in $\rho(T)$ observed in the $x = 0.4$ and 0.6 samples indicate that the NFL region extends well within the FM phase. The region of the phase diagram over which $URu_{2-x}Re_xSi_2$ exhibits NFL characteristics in $\rho(T)$ and $C(T)$ is made apparent by the power-law exponent n of the resistivity as shown in the top part of the phase diagram. The exponent appears to approach a minimum value $n \simeq 1$ at $x \sim 0.35$ with a return to FL behavior ($n = 2$) and at the concentration where the FM is strongest ($x = 0.8$).

The Griffiths-McCoy phase model is consistent with the NFL properties in the paramagnetic phase of $URu_{2-x}Re_xSi_2$ ($0.15 \leq x \leq 0.3$) in which power-law behavior is observed, i.e., $C/T \propto \chi \propto T^{-n}$ ($n \sim 0.2$–0.4) and may also provide an explanation for the occurrence of the NFL behavior within the FM phase. In this model, competition between the RKKY interaction and the Kondo effect in a disordered, anisotropic material leads to the formation of magnetic clusters. Quantum mechanical tunneling between different states in these clusters gives rise to Griffiths singularities and, hence, to power-law behavior in the physical properties at low temperatures [7]. Griffiths has shown [17] that, in the FM phase, the sum of the contributions to the critical behavior from the local magnetic clusters may exceed that of the infinite cluster describing the long-range magnetic order, thus leading to the possibility of NFL characteristics within the FM phase. Measurements in magnetic field, as has been proposed for $Ce_{1-x}La_xRhIn_5$ [28], may provide further support for the applicability of the Griffiths-McCoy theory to $URu_{2-x}Re_xSi_2$.

It is compelling to ascribe the non-Fermi-liquid behavior in $URu_{2-x}Re_xSi_2$ to proximity to a quantum critical point in light of the phase diagram presented above, although no single QCP theory correctly predicts all of the NFL properties observed in $URu_{2-x}Re_xSi_2$. The work of Belitz and co-workers on the quantum critical behavior of itinerant ferromagnets explicitly incorporates the effects of nonmagnetic disorder [9]. In this theory, effective long-range interactions of the order parameter (spin) fluctuations are produced by the diffusive dynamics of the conduction

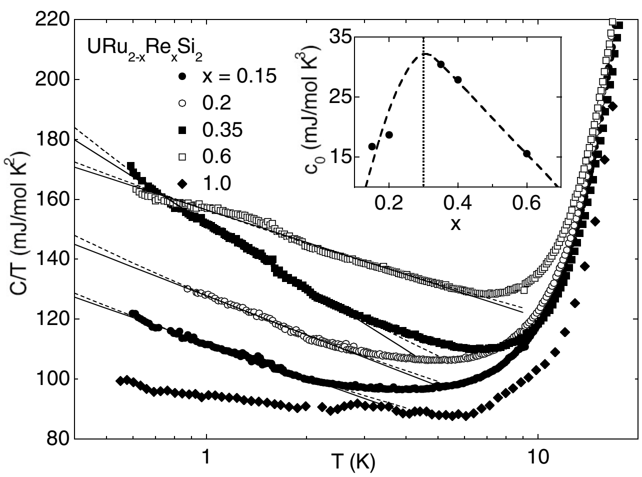

FIG. 3. Specific heat C divided by temperature T vs T for $0 \leq x \leq 1.0$ on a semilog scale. The solid (dashed) lines are linear (power-law) fits to the data. Data for the $x = 1$ sample are from Ref. [20]. Inset: Coefficient of the logarithmic contribution c_0 vs x. The dashed line is a guide to the eye. The location of FM ($x_c^{FM} = 0.3$) QCP is indicated by the vertical dotted lines.

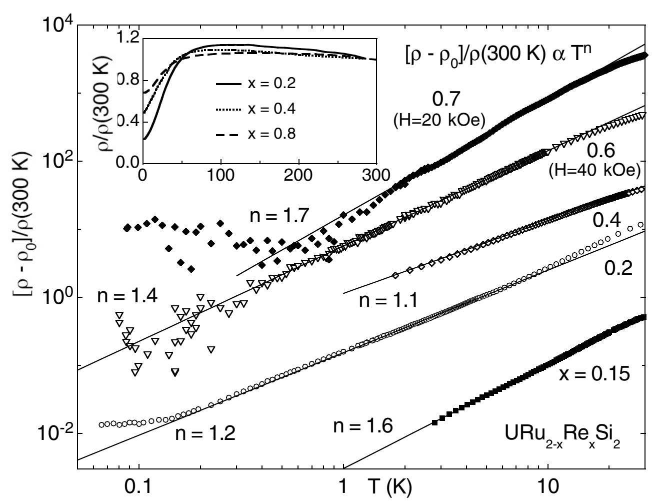

FIG. 4. $[\rho - \rho_0]/\rho(300\text{ K})$ vs T for $0.15 \leq x \leq 0.7$, plotted on a log-log scale. The solid lines are power-law fits to the data yielding the exponents n. Each of the curves has been shifted up by one decade from the curve below it for clarity. Inset: $\rho/\rho(300\text{ K})$ vs T of $URu_{2-x}Re_xSi_2$ for $0.2 \leq x \leq 0.8$.

electrons. These long-range interactions lead to quantum critical behavior in three dimensions that is quite different from classical mean field theory behavior. Some of the predictions of this model include a logarithmic divergence in C/T close to the critical concentration x_c and critical exponents $\beta = 2$, $\gamma = 1$, $\delta = 1.5$ [defined as $M(t, H = 0) \propto t^{\beta}$, $M(t = 0, H) \propto H^{1/\delta}$, $\gamma = \beta(\delta - 1)$, where $t = |x - x_c|$], which differ from the classical values of $\beta_{\rm class} \simeq 0.37$ and $\delta_{\rm class} \simeq 4.86$ [29]. These predictions can be applied to the physical behavior of the $URu_{2-x}Re_xSi_2$ system near the FM QCP with some success. The exponent δ was determined by a power-law fit to the magnetization isotherms at low temperatures for $x = 0.3$ as shown in Fig. 2(b). A linear extrapolation of the $\delta(T)$ data to $T = 0$ K for different Re concentrations yields the zero temperature value $\delta(0)$ [Fig. 2(b)]. The value $\delta = 1.5$ at $x = 0.3$ is in agreement with the expected value of the Belitz-Kirkpatrick theory providing further support that a FM QCP occurs at this concentration. It is interesting to note that a similar critical exponent δ is obtained from the modified Arrott plot analysis (Fig. 2). The exponent δ appears to approach the classical value predicted by mean field theory with increasing Re concentration. The logarithmic behavior of C/T near the FM QCP is consistent with this model. Other predictions of this model, however, do not agree with the experimental data. For instance, the specific heat is predicted to have a cusp at T_C, and a logarithmic divergence is not expected within the FM phase. The theoretical predictions of a renormalization-group analysis [8] of a FM $T = 0$ K phase transition in 3D and spin-fluctuation models [11], mentioned above, do not appear to describe the physical properties of $URu_{2-x}Re_xSi_2$ near the FM QCP ($x_c^{\rm FM} = 0.3$), in which $C(T)/T \propto -\ln T$ (or $T^{-0.2}$) and $\rho(T) \propto T^{1.2}$ for $x = 0.2$ and 0.35. Furthermore, while the data are somewhat more consistent with 2D FM critical behavior, i.e., $C(T)/T \propto T^{-1/3}$, $\rho(T) \propto T^{4/3}$ [8,11], $\chi(T)$ does not follow the expected T^{-1} [11] or $1/T \ln T$ [10] T dependences near $x_c^{\rm FM}$. Therefore, it appears that while some of the physical properties can be described by the QCP models of Belitz and co-workers [9], or by 2D FM spin-fluctuation models [10,11], they cannot explain the NFL behavior observed within the ferromagnetic phase of $URu_{2-x}Re_xSi_2$.

In summary, the behavior of $URu_{2-x}Re_xSi_2$ in the vicinity of the ferromagnetic quantum critical point ($x_c^{\rm FM} = 0.3$) has been investigated. Measurements of specific heat and electrical resistivity on $URu_{2-x}Re_xSi_2$ provide convincing evidence for the occurrence of non-Fermi-liquid behavior over at least a decade in temperature well within the ferromagnetic state ($x = 0.6$). A Griffiths-McCoy scenario is consistent with the NFL behavior observed in both the paramagnetic and the ferromagnetic phases of $URu_{2-x}Re_xSi_2$. It is hoped that these experimental results will stimulate further theoretical work on quantum criticality near a ferromagnetic instability.

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