Non-Fermi Liquid Behavior within the Ferromagnetic Phase in URu$_{2-x}$Re$_x$Si$_2$

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The URu$_{2-x}$Re$_x$Si$_2$ system exhibits ferromagnetic order for Re concentrations $0.3 < x < 1.0$ and antiferromagnetic order for $0 < x < 0.15$. Non-Fermi liquid (NFL) behavior is observed in the specific heat for $0.15 < x < 0.6$, in which $C/T \propto -\ln T$, and also in the power law $T$-dependence of the electrical resistivity ($\rho(T) \propto T^n$) with an exponent $n < 2$ for $0.15 < x < 0.8$, at low temperatures, providing strong evidence that the non-Fermi liquid behavior persists within the ferromagnetic phase. The NFL behavior found in URu$_{2-x}$Re$_x$Si$_2$ is most consistent with proximity to a quantum critical point.

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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 $\chi$, specific heat $C$, and electrical resistivity $\rho$, which are at odds with those of a Fermi liquid (i.e., $C/T \sim \chi \sim \text{const.}$, and $\rho(T) \propto T^2$). Such unusual low-$T$ behavior is most often observed in close proximity to the value of a control parameter (i.e., $P$, $x$, $H$) where a magnetic phase transition is suppressed to zero temperature, or quantum critical point (QCP).

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 the general behavior of such NFL systems as Y$_3$U$_1$Fe$_3$ [1]. Since many non-Fermi liquid compounds are disordered alloys, various 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 quantum critical point 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 non-Fermi liquid physics has focussed on antiferromagnetic (AFM) quantum critical points; comparatively little is known about ferromagnetic (FM) quantum critical points by studies of heavy fermion systems such as Th$_{1-x}$U$_x$Cu$_2$Si$_2$ [12] and CeCu$_2$Ge$_2$ [13]. The largest body of evidence for a FM QCP is found in $d$-electron systems (e.g., Ni$_3$Pd$_{1-x}$ [14], MnSi [11], ZrZn$_2$ [11]), where non-Fermi liquid behavior is observed only near the QCP. In these cases, the theoretical 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 a variation of the Curie temperature with a control parameter $\alpha$, $T_C \propto |\alpha - \alpha_c|^{9/4}$ where $\alpha_c$ corresponds to the QCP, appear to describe the NFL properties reasonably well.

In this Letter, we report the first example of non-Fermi liquid behavior observed deep within the ferromagnetic phase of the heavy fermion URu$_{2-x}$Re$_x$Si$_2$ system, in which $C(T)/T \propto -\ln T$ and $\rho(T) \propto T^n$ with $n \sim 1.2$ over more than a decade in temperature below 20 K for $x = 0.6$. This result is in striking contrast to the behavior of the $d$-electron systems where the NFL characteristics are found only near the FM QCP. Furthermore, it is one of the rare examples in which the NFL behavior occurs within the magnetic phase [15]. While no theory at present can account for the coexistence of ferromagnetism and NFL behavior, some of the physical properties near the FM QCP are consistent with the renormalization-group theory of Belitz and coworkers, which explicitly includes the effects of disorder on the low-energy dynamics [9]. In addition, URu$_{2-x}$Re$_x$Si$_2$ provides an opportunity to investigate the behavior in the vicinity of both an AFM and a FM critical point, since Re substitution suppresses the AFM (or spin density wave SDW) transition that occurs in URu$_2$Si$_2$ at $T_N = 17.5$ K [16]. Substitution of Re for Ru in URu$_2$Si$_2$ rapidly suppresses superconductivity from $T_{SC} = 1.5$ K in URu$_2$Si$_2$ to $T_{SC} = 0.23$ K at $x = 0.01$ and also suppresses and broadens the AFM transition to $T_N \sim 13$ K at $x = 0.1$ [17]. Further investigations [18] of the URu$_{2-x}$M$_x$Si$_2$ (M = Re, Tc) systems revealed the first occurrence of a ferromagnetic instability in a heavy fermion system. The physical properties of the URu$_{2-x}$Re$_x$Si$_2$ system such as the saturation moment $\mu_s$, Curie temperature $T_C$, and electronic specific heat coefficient $\gamma$ exhibit maxima with Re concentration, reaching values $\mu_s \sim 0.44 \mu_B/\text{U atom}$ and $T_C \sim 38$ K at $x = 0.8$, and $\gamma \sim 160 \text{ mJ/mole K}^2$ at $x = 0.6$. The ferromagnetic ordering was confirmed by neutron scattering experiments [19] for $x = 0.8$ and NMR measurements for $x = 0.4$ [20].

Polycrystalline samples of URu$_{2-x}$Re$_x$Si$_2$ for $0.125 \leq x \leq 0.7$ were prepared by arc melting as described in Ref. 17. Electrical resistivity, specific heat, and magnetic sus-
ceptibility measurements between \( 1 \) K and 300 K were made in a \(^4\)He cryostat, in a semi-adiabatic \(^3\)He calorimeter, and with a Quantum Design SQUID magnetometer, respectively. Measurements of \( \rho(T) \) down to 60 mK were performed in a \(^3\)He-\(^4\)He dilution refrigerator. The absolute magnitude of the resistivity could not be determined due to the presence of microcracks in the URu\(_2\)Re\(_x\)Si\(_2\) samples.

We first focus our attention on determining the AFM and FM quantum critical points in URu\(_2\)Re\(_x\)Si\(_2\). The AFM (SDW) transition is suppressed and significantly broadened with increasing Re concentration to \( T_N \sim 13 \) K for \( x = 0.1 \) and is no longer observed (above 0.6 K) for \( x = 0.15 \) as shown in Fig. 1. Similar behavior is found in the electrical resistivity. A hump-like feature corresponding to the AFM transition occurs for \( x = 0 \) at \( T_N \sim 17.5 \) K and by \( x = 0.1 \) the transition is substantially broadened and centered at \( T_N \sim 5 \) K [17]. No sign of an AFM transition could be observed in \( \rho(T) \) for \( x = 0.125 \) which may be due to the fact that the transition is too broad and/or occurs below 1.8 K. From these measurements, the AFM QCP is estimated to be \( x_c^{AFM} = 0.15 \).

The ferromagnetic critical point in the URu\(_2\)Re\(_x\)Si\(_2\) system is determined solely from the magnetic properties since no FM anomaly is observed in either \( C(T) \) or \( \rho(T) \) measurements, similar to the behavior of some other ferromagnetic systems such as Th\(_{1-x}\)U\(_x\)Cu\(_2\)Si\(_2\) [12] and Ni\(_{1-x}\)Pd\(_x\) [14]. The Curie temperatures determined from an Arrrott 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. 3) [18]. A ferromagnetic ordering temperature could not be determined from this type of analysis for \( 0.3 \leq x \leq 0.4 \). The Curie temperatures deduced from \( \chi(T) \) measurements (Fig. 3) in a field of \( H = 20 \) Oe are slightly lower than, though generally consistent with, those obtained from the Arrrott plots. No FM order could be determined for \( x = 0.3 \) by either method; therefore, the FM QCP is estimated to be \( x_c^{FM} = 0.3 \).

The specific heat \( C/T \) vs \( \ln T \) for \( 0.15 \leq x \leq 1.0 \) is shown in Fig. 1. At temperatures 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 K \( < T < 6 \) K; this return to Fermi liquid behavior is supported by a \( T^2 \) dependence of the electrical resistivity [18]. The \( C/T \) data for 0.15 \( \leq x \leq 0.6 \) were fit by the expression \( C/T = \gamma_0 - c_0 \ln T \) where \( \gamma_0 \) 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\(_2\)Si\(_2\) [21] is subtracted from the URu\(_2\)Re\(_x\)Si\(_2\) data. The value of \( c_0 \) reaches a maximum at \( x \sim 0.3 \) as shown in the inset of Fig. 1, close to the FM QCP; similar behavior is also observed in Ni\(_{1-x}\)Pd\(_x\) [14]. Least-squares fits of the data to a power law \( (C/T \propto T^{-n}) \) over a similar temperature range yield an exponent \( n \sim 0.2 \) which is (nearly) independent of \( x \).

![Fig. 1](image1.png)

**Fig. 1:** Specific heat \( C \) divided by temperature \( T \) vs \( T \) for \( 0 \leq x \leq 1.0 \) on a semi-log scale. The solid lines are linear fits to the data. Data for the \( x = 1 \) sample are from Ref. 18. Inset: Coefficient of the logarithmic contribution \( c_0 \) vs \( x \). The dashed line is a guide to the eye. The location of the AFM \( (x_c^{AFM} = 0.15) \) and FM \( (x_c^{FM} = 0.3) \) QCPs are indicated by the vertical dotted lines.

![Fig. 2](image2.png)

**Fig. 2:** \( \rho/(\rho(300 \text{K})) \) vs \( T \) of URu\(_2\)Re\(_x\)Si\(_2\) for 0.2 \( \leq x \leq 0.8 \). Inset: \( [\rho - \rho(0)]/\rho(300 \text{K}) \) 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.

The normalized electrical resistivity \( \rho/(\rho(300 \text{K})) \) vs \( T \) of URu\(_2\)Re\(_x\)Si\(_2\) for 0.2 \( \leq x \leq 0.8 \) is shown in Fig. 2. 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 \( J \)-ion sublattice, typical of many heavy fermion compounds. Below \( \sim 20 \)
K, $\rho(T)$ of URu$_2$Re$_x$Si$_2$ for $0.1 < x \leq 0.7$ follows a power law $T$-dependence of the form $[\rho - \rho_0]/[\rho(300\text{ K})] \propto T^n$, where $\rho_0$ is a constant, as displayed in the inset of Fig. 2. The exponent $n$ decreases from 1.6 for $x = 0.15$ to 1.1 for $0.2 < x < 0.5$ before increasing to 1.5 for $x = 0.7$, as shown in Fig. 3. Thus, the 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 K $\leq T \leq 13$ K [$n = 1.2$ above 1 K for $H \leq 40$ kOe] [22], 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 K $\leq T \leq 6.2$ K) with an apparent crossover to Fermi liquid behavior below $T^* \sim 0.15$ K. A return to a Fermi liquid ground state occurs for $x \leq 0.1$ and $x \geq 0.8$.

The magnetic susceptibility of URu$_2$Re$_x$Si$_2$ 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$_2$Re$_x$Si$_2$ is shown in Fig. 3 (similar to previous results [18]). There is evidence for non-Fermi liquid behavior in the URu$_2$Re$_x$Si$_2$ system from $\rho(T)$, $\chi(T)$, and $C(T)$ measurements between the AFM and FM phases of the phase diagram (0.15 $\leq x \leq 0.3$). Both the logarithmic divergence in $C(T)$ and power law behavior in $\rho(T)$ observed in the $x = 0.4$ and $x = 0.6$ samples indicate that the NFL region extends well within the FM phase. The variation of $T_C$ and $T_N$ with Re concentration near the QCPs is difficult to determine from the present measurements and future experiments are planned to elucidate the nature of $T_{mag}(x)$ near the critical points.

It is compelling to ascribe the non-Fermi liquid behavior in the URu$_2$Re$_x$Si$_2$ system to proximity to a quantum critical point in light of the phase diagram presented above. The work of Belitz and coworkers 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 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 $M(t,H = 0) \propto t^\beta$, $M(t = 0,H) \propto H^{1/\delta}$), $\gamma = \beta(\delta - 1) = 1$, where $t = |x - x_c|$, which differ from the classical values of $\beta_{class} \simeq 0.37$ and $\delta_{class} \simeq 4.86$ [23]. These predictions can be applied to the physical behavior of the URu$_2$Re$_x$Si$_2$ system near the FM QCP with reasonable success. The exponent $\delta$ was determined by a power law fit to the magnetization isotherms at low temperatures for $x = 0.3$ as shown in Fig. 4. A linear extrapolation of the $\delta(T)$ data to $T = 0$ K for different Re concentrations (Fig. 4a) yields the zero temperature value $\delta(0)$ (Fig. 4b). 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.

The exponent $\delta$ approaches 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 theoretical model, however, do not agree with the empirical 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 $T = 0$ phase transition in 3D and spin fluctuation models [11], mentioned above, do not appear to describe the physical properties of URu$_2$Re$_x$Si$_2$ near the FM QCP ($x_{c}^{FM} = 0.3$), in which $C(T)/T \sim -\ln T$ (or $T^{4.2}$) and $\rho(T) \sim T^{1.2}$ for $x = 0.2$ and $x = 0.35$. The data appear to be more consistent with 2D FM critical behavior, i.e., $C(T)/T \sim T^{-1/3}$, $\rho(T) \sim T^{4/3}$ [8, 11], although $\chi(T)$ does not follow the expected $T^{-1}$ [11] or $1/T\ln T$ [10] $T$-dependences near $x_{c}^{FM}$. Therefore, it appears that the NFL behavior in the vicinity of the ferromagnetic critical point can at least qualitatively be described by the quantum critical point model of Belitz and coworkers [15], or perhaps by 2D FM spin-fluctuation models [10, 11], although they cannot explain the NFL characteristics within the FM phase.

At the antiferromagnetic critical point $x_{c}^{AFM} = 0.15$, URu$_2$Re$_x$Si$_2$ exhibits the following $T$-dependences:

![FIG. 3: Magnetic phase diagram of URu$_2$Re$_x$Si$_2$ showing the antiferromagnetic (AFM/SDW, filled circles), superconducting (SC, open circles), and ferromagnetic (FM, filled squares): Arrott plot analysis: 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. Some data from Refs. 17, 18.](image-url)
\( C/T \propto -\ln T, \rho \propto T^{1.0}, \) and \( \chi(T) \propto T^{-0.1}. \) These temperature dependences are at odds with the predicted behavior in proximity to an AFM QCP in 3D \( (C/T \propto -\sqrt{T}, \rho \propto T^{3/2}, \chi(T) \propto T^{-3/2}) \) [8, 10] and 2D \( (C/T \propto -\ln T, \rho \propto T, \chi(T) \propto -T \log T) \) [10].

Other non-Fermi liquid theories may also be relevant to URu2-ReSi2. A quadrupolar two-channel Kondo model [3] applies to U in tetragonal symmetry; however, many of the predictions, such as \( \rho \propto \chi \propto T^{1/2}, \) are not consistent with the measurements on URu2-ReSi2. Disorder-driven NFL theories may apply to URu2-ReSi2 since disorder is certainly present. Some of the Griffiths’-McCoy phase model predictions [7] are consistent with the data in the paramagnetic region, e.g., \( C/T \propto \chi \propto T^{-n} \) [24], but other predictions, such as a monotonic change of \( n \) with \( T \), are not satisfied. Further measurements in magnetic field would be helpful to determine if the Griffiths’-McCoy theory is valid for URu2-ReSi2 as has been proposed for Ce1-xLa_xRhIn5 [25]. The specific heat of URu2-ReSi2 agrees with the predictions of the Kondo disorder model [5, 6], but the power law behavior of \( \rho(T) \) does not follow the expected (constant with \( x \)) linear-T behavior.

In summary, the antiferromagnetic \( (x_{AM} = 0.15) \) and ferromagnetic \( (x_{FM} = 0.3) \) quantum critical points have been determined in URu2-ReSi2. Measurements of specific heat and electrical resistivity on URu2-ReSi2 provide convincing evidence for the occurrence of non-Fermi liquid behavior over at least a decade in temperature well within the ferromagnetic state (\( x = 0.6 \)). The NFL properties near \( x_{FM} = 0.3 \) are in qualitative agreement with a FM QCP, although a Griffiths’-McCoy scenario cannot be ruled out. Further theoretical work is clearly needed to explain the coexistence of non-Fermi liquid behavior with ferromagnetism in URu2-ReSi2.

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[15] NFL behavior has been observed in antiferromagnets such as \( \text{U(Pt}_{0.04}\text{Pd}_{0.06}) \) [J. S. Kim et al., Phys. Rev. B 45, 12081 (1992)] and \( \text{YbRh}_2\text{Si}_2 \) [Draneneli et al., Phys. Rev. Lett. 85, 626 (2000)].


[24] The power law exponent $n$ of $C/T$ and $\chi$ for a given $x$ should be equal [7], which is not the case in $\text{URu}_{2-x}\text{Re}_x\text{Si}_2$; however, differences in $n$ have been observed in other NFL systems [de Andrade et al., Phys. Rev. Lett. 81, 5620 (1998)].