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Comparison of CeRh$_2$Si$_2$ and CeRh$_2$–$_x$Ru$_x$Si$_2$ near their Magnetic-Nonmagnetic Boundaries

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A zero-temperature magnetic-nonmagnetic phase boundary is accessed in CeRh$_2$Si$_2$ by application of pressure and in CeRh$_2$–$_x$Ru$_x$Si$_2$ at ambient pressure for $x = 1.0$. A comparison of specific heat and resistivity measurements in the two cases emphasizes the importance of disorder in producing non-Fermi-liquid-like behavior in these as well as in other Ce-based systems. [S0031-9007(97)03104-9]

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The ground state of a periodic lattice of Kondo impurities is determined by the competition between intersite, Ruderman-Kittel-Kasuya-Yosida (RKKY), and intrasite, Kondo, interactions [1,2]. The balance between these pressure-dependent interactions is set by the magnitude of the exchange parameter $J$ with RKKY interactions increasing as $J^2$, and Kondo interactions increasing exponentially with $J$. Because of these different functional dependences on $J$, at some critical value $J_c$ there will be a zero-temperature critical point that separates a magnetically ordered phase from one with no long range order, assuming that some other phase transition does not intervene. Fluctuations around this quantum-critical point are expected to lead to non-Fermi-liquid (NFL) temperature dependences of thermodynamic and transport properties [3,4]. Other suggested origins of NFL behavior found in Ce- and U-based Kondo-lattice compounds [5] include local spin fluctuations near an antiferromagnetic instability [6,7], a multichannel Kondo effect [8], and a distribution of Kondo temperatures introduced by crystallographic disorder [9]. Attempts to describe NFL power law and logarithmic dependences of thermodynamic and transport properties by these possible mechanisms have met with varying degrees of success [7–11], and there remains no consensus for the origin of NFL behavior. However, what is common to these systems in which NFL behavior is claimed is the presence of disorder, introduced intentionally by chemical alloying on the $f$ or ligand sites or that is inherent to the material’s crystal structure, e.g., Ce$_2$Ni$_3$ [12]. In this Letter, we report results of a study in which a zero-temperature critical point in CeRh$_2$Si$_2$ is accessed in two ways: by application of pressure, which does not introduce disorder, and by chemical substitution of Ru for Rh, which does. The results are qualitatively different. They strongly suggest the important role of disorder in inducing NFL behavior in this system and, by implication, more generally in $f$-electron NFL systems.

CeRh$_2$Si$_2$ is an ideal system for this study. Its Néel temperature $T_N$ can be reduced from an ideal pressure value of 35 K to $T = 0$ at a critical pressure $P_c \sim 9$ kbar [13], a pressure readily accessible in specific heat and resistivity measurements. Substitution of Ru for Rh drives $T_N$ to zero in CeRh$_2$–$_x$Ru$_x$Si$_2$ for $x_c = 0.95$ [14]. Measurements reported below were performed on well-characterized CeRh$_2$Si$_2$ and CeRh$_2$–$_x$Ru$_x$Si$_2$ ($x = 1.0$) samples prepared by arc melting. Powder x-ray diffraction showed that all samples formed in the ThCr$_2$Si$_2$ structure with no detectable second phase. Pressure measurements were carried out in Be-Cu pressure cells with AgCl (flourinert) as the pressure-transmitting medium in specific heat (resistivity) experiments. Additional experimental details will be given later [15].

The inset in Fig. 1 shows the magnetic specific heat $C_m(T)$ divided by temperature for CeRh$_2$Si$_2$ at ambient pressure. A sharp anomaly, peaked near 35 K, signals the onset of magnetic order that is also found at this temperature in magnetic susceptibility and resistivity measurements. The magnetic entropy, represented by the solid curve,-crosses the dotted horizontal line corresponding to $S_m = R \ln R$ at a temperature just above $T_N$, implying that magnetic order develops in a ground-state doublet. In the absence of magnetic order, we can estimate the specific heat Sommerfeld coefficient $\gamma$ from the Anderson-impurity relationship $\gamma = (N - 1) \pi R/6 T_K$, where $N$ is the ground-state degeneracy and $R$ is the gas constant. Taking $T_K = 33$ K from quasielastic neutron scattering experiments [17] at $T \approx T_N$ and $N = 2$, we obtain $\gamma = 130$ mJ/mole K$^2$, much greater than the experimental value, 22.8 mJ/mole K$^2$ (see Fig. 1). This large reduction in $\gamma$ in the ordered state is found commonly in Ce-based magnets with nearly full-moment ordering and may be attributed [18] to the existence of a large internal magnetic field that quenches, at least partially, Kondo-like spin fluctuations.

Figure 1 also shows the effect of pressure on $C_m/T$ for CeRh$_2$Si$_2$ at $T \leq 10$ K. With the application of pressure, there is a qualitative increase in $C_m/T$ at all temperatures, but an uncertainty of 0.5% in the heat capacity of the pressure cell limits a detailed interpretation of $C_m(T)$ for temperatures near and above 10 K. However, a specific heat anomaly with a magnitude of 10% or more of the $P = 0$ anomaly would be clearly observable.
The absence of a phase transition in these data can be understood from the pressure dependence of $T_N$ established by resistivity measurements and shown in the inset of Fig. 2: For pressures less than 7.1 kbar, $T_N(P) \approx 0.75T_N(0) = 26.2$ K, which is outside the temperature range of pressure-dependent specific heat measurements; for $P \approx 11.0$ kbar, $T_N = 0$. Overall, $C_m/T$ data shown in Fig. 1 are fully consistent with the $T_N(P)$ phase diagram determined resistively.

In direct contrast with the behavior of nonordering Ce heavy-fermion systems, in which $\gamma$ decreases with pressure [19], $\gamma$ increases initially as pressure is applied, which is shown in Figs. 1 and 2. However, for higher pressures, $\gamma$ does decrease, approximately linearly, with increasing pressure. The linear-in-pressure constructions in Fig. 2 show that the crossover from $\partial \gamma/\partial P > 0$ to $\partial \gamma/\partial P < 0$ occurs very near $P_c$. Qualitatively, this behavior of $\gamma(P)$ can be understood on the basis of Doniach’s model [2] for competing Kondo and RKKY interactions. With increasing pressure, the balance between Kondo and RKKY interactions shifts in favor of increasing dominance of Kondo-spin compensation of the localized 4f moments, redistributing magnetic entropy both to higher temperatures (because $T_K$ increases with pressure) and from localized degrees of freedom to Kondo-like spin fluctuations. It is this latter process that produces the initial increase in $\gamma$ which is allowed because the internal magnetic field is also suppressed through the Kondo effect. Once the ground state is paramagnetic ($P > P_c$), $\gamma$ decreases as in nonordering Ce heavy-fermion compounds.

Sufficiently near $P_c$, quantum-critical fluctuations should dominate the low-temperature specific heat, producing a logarithmic or stronger divergence of $C_m/T$ and very large values of this ratio [3]. Neither of these effects are found in our measurements. Inspection of the data in Fig. 1 shows the absence of any significant temperature dependence of $C_m/T$ at low temperatures. Therefore, we conclude that there is no evidence in these data for non-Fermi-liquid behavior. This conclusion is supported as well by our resistivity measurements (not shown) that give, for $T \approx 10$ K, $\rho \propto T^\beta$ with $\beta = 2.80 \pm 0.5$ at $P = 7.3$ kbar and $\beta = 2.03 \pm 0.01$ at $P = 12.4$ kbar.

The $T_N$ phase boundary is very steep as $P$ approaches $P_c$, unlike that for CeCu$_{6-x}$Au$_x$ with $x = 0.2$ and 0.3 [11,20] but more like Ce$_7$Ni$_3$ [10]. Taking $P_c = 9$ kbar for CeRh$_2$Si$_2$, we have specific heat data at 0.79$P_c$ and 1.22$P_c$. However, $C_m/T \propto -\ln T$ in Ce$_7$Ni$_3$ for $P \approx 1.5P_c$ and in CeCu$_{5.8}$Au$_{0.2}$ to a least $P = 1.2P_c$, in both cases remarkably far from the critical point. If NFL signatures arise from quantum or local spin fluctuations in those cases, they should be observed clearly in our data as well, but they are not. A possible explanation for the lack of a well-defined logarithmic temperature dependence of $C_m/T$ in CeRh$_2$Si$_2$ is that the transition is first order near $P_c$. However, there is no indication of a first-order transition in the specific heat data. We also have looked for hysteretic behavior in $T_N(P)$ near $P_c$ by resistivity measurements, and within uncertainties in determining $T_N (\pm 1$ K) and $P (\pm 0.5$ kbar), there is no evidence for hysteresis that might be taken as indicative of a first-order transition.

It is instructive to compare these pressure results to those obtained at ambient pressure when CeRh$_2$Si$_2$ is alloyed with Ru. Substituting Ru in CeRh$_{2-x}$Ru$_x$Si$_2$ initially suppresses $T_N$ rapidly to a plateau, where $T_N \sim 11$ K (for $0.2 < x < 0.8$). With additional Ru substitution, $T_N$ drops abruptly to zero at $x_r = 0.95$ [14], similar to the drop of $T_N$ near $P_c$ in CeRh$_2$Si$_2$. The results of specific heat and susceptibility measurements are shown in Fig. 3 for one representative sample with nominal $x = 1.0$ [21]. For this
sample, $C_m/T$ increases logarithmically from $\sim 11$ K to near 1 K before rolling over to an approximately constant value of $\sim 700$ mJ/mole K$^2$ for $T \lesssim 0.2$ K. Interestingly, a linear extrapolation of $C_m/T$ vs $T^2$ from above 15 K to $T = 0$ gives $\gamma(T = 0) = 130$ mJ/mole K$^2$, a value remarkably close to our estimate of $\gamma$ from the neutron quasielastic linewidth of CeRh$_2$Si$_2$ at $T \approx T_K$. As shown in the inset to Fig. 3, $\chi \propto \ln T$ for $2 \lesssim T \lesssim 30$ K. Although there is some evidence in $C_m/T$ and $\chi$ for minor sample inhomogeneity, these data are generally consistent with NFL behavior in an interval spanning at least one decade in temperature above $\sim 1$ K. However, specific heat measurements below 1 K suggest that the NFL behavior does not persist in the ground state of CeRh$_2$Si$_2$ but that $C_m/T$ approaches more conventional Fermi-liquid behavior at the lowest temperatures. This is corroborated by resistivity measurements (not shown) that find $\rho = \rho_0 + \alpha T^2$ for $0.04 \lesssim T \lesssim 0.20$ K, where $\rho_0 = 83 \mu\Omega$ cm and $\alpha = 24 \mu\Omega$ cm/K$^2$. The large $T^2$ coefficient is also consistent with the large $T \rightarrow 0$ Sommerfeld coefficient.

There are two notable differences between conditions under which NFL behavior is found in CeRhRuSi$_2$ and those that fail to show NFL signatures in CeRh$_2$Si$_2$. The first difference is the nearness to a $T = 0$ critical point. Defining nearness as $\Delta = |A - \delta|/|\delta|$, where $\delta$ is pressure in the case of CeRh$_2$Si$_2$ or $x$ in CeRh$_{2-x}$Ru$_x$Si$_2$, then $\Delta \approx 0.2$ for CeRh$_2$Si$_2$ and $\Delta = 0.05$ for CeRh$_{0.95}$Ru$_{0.05}$Si$_2$. The second difference is that CeRh$_2$Si$_2$ is crystallographically ordered, whereas alloying with Ru introduces disorder. Recent model calculations [22] of a disordered Anderson lattice demonstrate that NFL behavior is a robust feature, provided that disorder is sufficient to produce a distribution of Kondo temperatures, which includes $T_K \rightarrow 0$, and that RRK interactions are negligible. Because this model is related closely to the phenomenological approach used by Bernal et al. [9], we follow Ref. [9] by assuming that alloying with Ru produces a distribution of Kondo temperatures through a Gaussian distribution $P(\lambda)$, where $\lambda$ is defined by the relationship $T_K(\lambda) = E_F \exp(-1/\lambda)$. The average value of $\chi$ or $C_m$ is given by

$$\langle X \rangle = \int_0^\infty P(T_K)X(H, T; T_K) dT_K.$$  \hspace{1cm} (1)

When $X$ is the susceptibility, we take $X(H, T; T_K) = g \mu_B H/\exp(BJ(x)/H)$, with the argument of Brillouin function $B_J$ given by $x = g \mu_B H/k_B(T + \sqrt{2}T_K)$; for specific heat, we assume the resonant-level model for a Kondo impurity (Eq. 6 of Ref. [23]). Because the ground state is a crystal-field doublet, we use expressions for a spin-$\frac{1}{2}$ impurity in both cases, and set $g = 2$ and $E_F = 1$ eV.

A two parameter fit (mean value of $\lambda$ and width of the Gaussian distribution $\Delta \lambda$) to $\gamma(T)$ data is shown by the solid line in the inset in Fig. 3. Using this same distribution ($\lambda_{\text{mean}} = 0.175$ and $\Delta \lambda = 0.021$), we then calculate the temperature-dependent specific heat. The calculated curve agreed reasonably well with the observed $T$ dependence but fell uniformly below the measured curve. The magnitudes of the calculated and measured curves could be reconciled, as shown by the solid line in Fig. 3, by simply redefining the Kondo temperature in the Schotte and Schotte model [23] as $T_K \rightarrow aT_K$, where $a = 0.65$. This is not unreasonable, given the qualitative definition of $T_K$ in the expressions for $\chi$ and $C_m$. Given that there are no adjustable parameters in the distribution function $P(\lambda)$ used to calculate $C_m(T)$, the model of disorder provides a relatively good description of the data [24]. The mean $T_K$'s obtained from these procedures are 32.4 K for $\chi$ and 21.1 K for $C_m$, both close to the spin-fluctuation temperature derived from quasielastic neutron scattering in CeRh$_2$Si$_2$. The width of the Gaussian distribution in $\lambda$ is 0.12$\lambda_{\text{mean}}$ and, consequently, $P(T_K)$ falls rapidly to small values as $T_K \rightarrow 0$. That is, the ligand disorder in CeRhRuSi$_2$ is not sufficient to give $P(T_K)$ enough weight at small $T_K$ to produce NFL behavior as $T \rightarrow 0$, and thereby allows Fermi-liquid temperature dependences in $C_m/T$ and $\rho$ to emerge at temperatures well below 1 K.

The model of Miranda et al. [22] predicts $\rho \propto -bT$ in the limit of strong disorder, contrary to our observations at very low temperatures. Therefore, the NFL behavior at $T \approx 1$ K and Fermi-liquid behavior at $T \ll 1$ K may be understood as a consequence of “moderate” disorder [25] which produces a distribution of Kondo temperatures that cuts off at some low but finite $T_K$ which, in turn, sets the scale ($T_K = 6$ K) for $\gamma(T \rightarrow 0)$ and $T^2$ coefficient of resistivity. In the absence of intersite interactions, the Gaussian distribution $P(\lambda)$ would always have finite weight as $T_K \rightarrow 0$ and, consequently, a NFL ground state would be expected. In analogy to conventional heavy-fermion system, we speculate that this cutoff arises from the competition between intersite (RKKY) interactions, which may be weakened by disorder, and the distribution of Kondo interactions. Any complete model
of a disordered Anderson lattice must include intersite, as well as intrasite, interactions before a quantitative test of this model is possible.

The contrast in temperature dependences of $C_m/T$, between CeRh$_2$Si$_2$ near $P_c$ and CeRh$_2$$_{-1.2}$Ru$_{1.2}$Si$_2$ near $x_c$, is striking. We find no evidence for non-Fermi-liquid behavior in the specific heat or resistivity of CeRh$_2$Si$_2$ near its $T = 0$ magnetic-nonmagnetic boundary when this boundary is accessed by application of pressure. Comparisons to CeCu$_6$$_{-1.2}$Au$_{1.2}$ and Ce$_2$Ni$_3$ indicate that NFL behavior should have been observed in our data if, in those cases, NFL signatures were due to quantum or local spin fluctuations. This raises the possibility that disorder in CeCu$_6$$_{-1.2}$Au$_{1.2}$ and Ce$_2$Ni$_3$ plays a role in producing $C_m/T \propto -\ln T$ behavior near their magnetic-nonmagnetic boundaries. It is noteworthy that the residual resistivity of CeCu$_5$$_{0.9}$Au$_{0.1}$, which is just at the magnetic-nonmagnetic boundary in this system at $P = 0$, and Ce$_2$Ni$_3$ near $P_c$ is comparable to that found in CeRhRuSi$_2$. Although by some measure CeRhRuSi$_2$ is closer to a $T = 0$ critical point than CeRh$_2$Si$_2$ under pressure, which might contribute to the different low temperature behaviors [26]. Kondo disorder in CeRhRuSi$_2$ is clearly a significant factor in producing logarthmic divergences of $C_m/T$ and $\chi$ over a large low-temperature interval.

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[26] NFL behavior is less pronounced in CeRh$_{0.8}$Ru$_{1.2}$Si$_2$, i.e., $\Delta = 0.2$, but $\chi$ and $C_m/T$ still exhibit $-\ln T$ dependences over a limited low temperature interval and $C_m/T$ approaches 500 mJ/mole K$^2$ as $T \rightarrow 0$. 