Doniach Phase Diagram, Revisited: From Ferromagnet to Fermi Liquid in Pressurized $CeRu_2Ge_2$

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We present the resistivity and ac susceptibility of $CeRu_2Ge_2$ at pressures p up to 130 kbar. Pressure transforms the system from a ferromagnet into a nonordering Fermi liquid (FL). The suppression of magnetic order at $p_c = 67$ kbar is accompanied by non-Fermi liquid (NFL) behavior. By comparing our results to isoelectronic $CeRu_2(Ge_{1-x}Si_x)_2$ we derive a unified hybridization J phase diagram for the entire material class. The phase diagram is characterized by the FL and Kondo energy scales T_{FL} and T_K, with the NFL behavior appearing at $T_{\mathrm{FL}} = 0$ K, while T_K remains finite and a smoothly increasing function of J. [S0031-9007(99)08832-8]

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An extensive body of experimental work has been developed which describes the stability of magnetic order in concentrated transition metal, rare earth, and actinide based compounds [1]. The results are strikingly universal, both for ferromagnets and antiferromagnets. Under the action of the control parameter Γ, which can be pressure p or composition x, magnetic order is initially stabilized, reaches a maximum T_{mag}, and is ultimately suppressed, vanishing for a critical value Γ_c. Increasing Γ further induces Fermi-liquid (FL) behavior, with the specific heat $c_p \propto T$, the magnetic susceptibility $\chi = \text{const}$, and the electrical resistivity $\rho \propto T^2$ below the characteristic temperature of the FL, T_{FL}. Recently much interest focused on the region near Γ_c, where weak power law or logarithmic temperature dependences of c_p, χ, and ρ are observed below a material specific temperature T_0 [1,2]. It is believed that these unusual temperature dependences, collectively called non-Fermi-liquid (NFL) behavior, represent the critical phenomena associated with the magnetic phase transition, occurring now at $T = 0$ K.

It is generally agreed that the fundamental parameter Γ which controls this phase diagram increases with the electronic hybridization J between the local moments and conduction electrons. The Doniach model [3,4] provided the first description of this phase diagram in terms of a competition between indirect local moment exchange, represented by the RKKY interaction $T_{\mathrm{RKKY}} \propto J^2$, and the Kondo screening, characterized by $T_K \propto \exp(-1/J)$. At small J, the RKKY exchange is dominant and the system orders magnetically at $T_{\mathrm{mag}} \simeq T_{\mathrm{RKKY}}$. For intermediate J, T_{RKKY} and T_K are of comparable strength; magnetic order still occurs, but now of increasingly screened moments. Ultimately, magnetism is suppressed upon further increase of J, with $T_{\mathrm{mag}} = 0$ K at a critical value J_c. For $J \gg J_c$, a system of strongly interacting quasiparticles is found, which can be mapped onto a FL with the characteristic energy scale $T_{\mathrm{FL}} \propto T_K$.

Although the Doniach model provides a qualitative description of the main experimental features of Kondo lattices, detailed comparison to experiments is problematic, except for small J. Such a comparison is crucial for understanding the basic energy scales which control Kondo lattices. In the limit of large J, there is both theoretical and experimental evidence for single-energy scaling in Kondo lattices [5]. While FL behavior is found for $J > J_c$, the microscopic origin of the characteristic temperature T_{FL} and its relationship to T_K remains unclear. We present evidence here that close to the magnetic instability the fundamental excitations of the quasiparticle system are described by an energy scale different from the Kondo energy T_K. It is the energy scale of the collective FL, T_{FL}, which goes to zero at J_c, in contrast to T_K, which remains finite. We will show that only for $J \gg J_c$ do the two energy scales merge.

$CeRu_2Ge_2$ is particularly suited for our study as a local moment Ce ferromagnet (FM), with a Curie temperature $T_C \simeq 8$ K, a small mass enhancement $\gamma = 20\ \mathrm{mJ/mole\,K^2}$, and small Kondo temperature $T_K = 1.9$ K, implying a small J [6,7]. Crystal electric field (CEF) excitations are absent at low T because of a large level splitting of 490 K [7]. Previous pressure studies indicated a complete suppression of magnetic order below 100 kbar [8].

In this Letter, we address the entire magnetic and nonmagnetic phase diagram of $CeRu_2Ge_2$, and the energy scales which control it. We present the complete p-T phase diagram of $CeRu_2Ge_2$, obtained from transport and ac-susceptibility measurements at pressures up to 130 kbar between 1.3 to 300 K. Pressure tunes $CeRu_2Ge_2$ from a local moment ferromagnet via two antiferromagnetic states into a nonordering FL. The suppression of magnetic order at the critical pressure $p_c = 67$ kbar is accompanied by NFL behavior and an enhanced electronic mass. Combining our results with those on isoelectronic

$CeRu_2(Ge_{1-x}Si_x)_2$ [9] enables us for the first time to quantitatively describe the evolution of the fundamental energy scales in a large family of isostructural materials over a very broad range of the hybridization J, both above and below J_c. Signatures of NFL behavior such as a resistivity $\propto T$ appear for the broadest range of temperatures at J_c, while above J_c FL behavior is recovered. We determine the J dependence of the effective Kondo temperature, and demonstrate that it is not the dominant energy scale near J_c.

Our study has been carried out on single crystals of $CeRu_2Ge_2$ taken from the surfaces of arc-melted polycrystalline buttons [7]. Neutron diffraction experiments prove the crystal structure to be tetragonal $ThCr_2Si_2$ ($a = 4.268$ Å; $c = 10.03$ Å), in good agreement with previous works [6]. At ambient pressure magnetic susceptibility establishes $CeRu_2Ge_2$ as a FM below $T_C = 8$ K, while no second magnetic phase above T_C was detected in our single crystals, unlike observations in polycrystalline material [6,8]. The pressure dependences of the resistivity ρ and the ac susceptibility $\chi_{\rm ac}$ of the crystals were determined employing a Bridgman anvil technique [10].

In Figs. 1(a) and 1(b), we plot the normalized resistivity $\rho/\rho_{300\text{-K}}$ of $CeRu_2Ge_2$ up to 130 kbar below 300 and 20 K, respectively. Our data below 20 kbar resemble previously obtained results [8]. In this pressure range, ρ increases with increasing T between 20 and 300 K, as expected for a metallic f electron system with a large CEF splitting [7]. A kink in the low-T resistivity, together with the saturation of the ac susceptibility signal [inset of Fig. 1(b) [11]] marks the onset of FM order at ambient pressure, while additional changes of slope, as well as features in the ac susceptibility, mark antiferromagnetic (AFM) transitions at higher pressures.

We have compiled the p dependences of the magnetic transition temperatures of $CeRu_2Ge_2$ in the phase diagram in Fig. 2 [12]. The main effect of pressures $p < 20$ kbar is to split the ambient FM phase into a low-T FM phase I below T_C and high-T phase II below T_N, with T_C decreasing and T_N increasing with p. Between 20 and 67 kbar T_N is reduced with p. A second change of slope is seen at $T_M < T_N$ (phase III), implying at least a change in the staggered magnetization below T_M. Our high pressure $\chi_{\rm ac}$ measurements prove that phase I is FM, while both phases II and III are AFM.

We see no evidence for magnetic order above 1.3 K for pressures above ~60 kbar. Figure 2 indicates that magnetic order is completely suppressed at the critical pressure $p_c = 67$ kbar, and ρ becomes a smooth function of T. For a narrow pressure range $64 \le p \le 71$ kbar and below temperatures as high as $T_0 = 60$ K the resistivity is well described by $\rho = \rho_0 + bT$. FL behavior is recovered at 76 kbar with $\rho = \rho_0 + AT^2$ below 4.5 K. A further increase of p stabilizes FL behavior, with $\rho - \rho_0 = AT^2$ over an increasingly broad temperature range $T_{\rm FL}$, while A decreases, with $T_{\rm FL} \propto A^{-1/2}$.

The p-T phase diagram of $CeRu_2Ge_2$, as well as the overall evolution of the resistivity under pressure, bears a striking similarity to that of the alloying series $CeRu_2(Ge_{1-x}Si_x)_2$, which also has three magnetic phases for $0 \le x \le 1$ [9]. The three phase lines of the alloying study can be scaled onto those of our pressure experiment assuming a bulk modulus $B_0 = 120$ GPa for $CeRu_2Ge_2$ (Fig. 2). This value compares favorably to those of related isostructural compounds [13] in which the pressure-volume relations were found to be almost linear over pressure ranges similar to those used here. In the alloying study, the phases have been identified by neutron diffraction: I—FM; II and III—AFM [ordering vector $\vec{k} = (0.31, 0, 0)$ in both cases]. This comparison suggests an exact and extensive equivalence between applied pressure and chemical pressure of Si alloying in $CeRu_2Ge_2$. Figure 2 implies that $CeRu_2Ge_2$ at 67.5 kbar is the electronic equivalent of $CeRu_2Si_2$ at ambient pressure, assuming $B_0 = 120$ GPa for $CeRu_2Ge_2$. Finally, since no difference is found between alloying experiments, in which disorder is profound, and our pressure study, in which disorder is minimal, we conclude that disorder is not a dominant factor in the physics of $CeRu_2(Ge_{1-x}Si_x)_2$.

The p dependences of ρ and A of $CeRu_2Ge_2$ between p_c and 100 kbar resemble those of $CeRu_2Si_2$. Therefore, to obtain absolute A values we assume the same room temperature resistivity for $CeRu_2Ge_2$ at 76 kbar as was

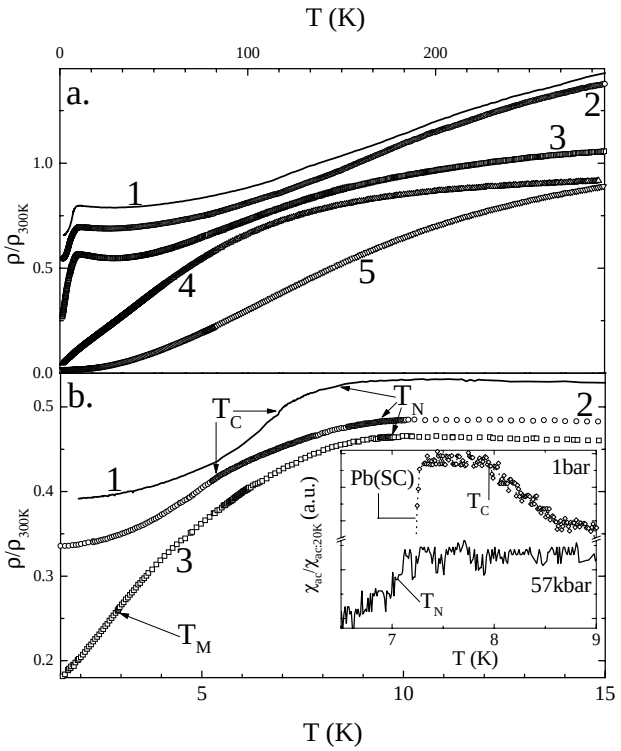

FIG. 1. The normalized resistivity $\rho/\rho_{300\text{-K}}$ of $CeRu_2Ge_2$ at 1.6 kbar (1), 10 kbar (2), 42 kbar (3), 67 kbar (4), and 130 kbar (5) below room temperature (a) and 20 K (b), offset for clarity. In the inset, we plot the normalized ac susceptibility signal at ambient pressure (◇) and 57 kbar (–).

found for $CeRu_2Si_2$ at 9 kbar, 91 $\mu\Omega$ cm. We quantify the emergence of the FL state above p_c by including the pressure dependence of the quasiparticle masses, taken from the resistivity coefficients of $CeRu_2Ge_2$ and $CeRu_2Si_2$ as $A^{-1/2}(p)$ in Fig. 2. The combined data of $A^{-1/2}(p)$ increase by almost a factor of 20 between 67.5 kbar (= ambient pressure $CeRu_2Si_2$) and 130 kbar, but saturate at highest pressures. Further, with $A/\gamma^2 = 3.3\ \mu\Omega\,\mathrm{cm\,mol^2\,K^2/J^2}$ for $CeRu_2Si_2$, we find $\gamma \approx 80$ mJ/mole K for $CeRu_2Ge_2$ at 76 kbar. These findings suggest that the suppression of magnetism in $CeRu_2Ge_2$ is accompanied by an enhanced electronic mass m^*, compared to its ambient pressure value, and a rapid decrease of m^* with p in the nonmagnetic part of the phase diagram.

We relate the parameter p to the underlying hybridization J using the similarity of $CeRu_2(Ge_{1-x}Si_x)_2$ and $CeRu_2Ge_2$. Since in Ce-based Kondo lattices the energy difference between Fermi and f electron level $E_F - E(f)$ is nearly constant [14], the hybridization J is a simple function of the hybridization matrix element V: $J \propto V^2$. The matrix elements V for each Ce bond can be calculated by tight binding methods [15]. Based on observations in related compounds [13], we assume the lattice parameters of $CeRu_2Ge_2$ to decrease linearly with p and extrapolate from their ambient pressure values to those at 67.5 kbar of $CeRu_2Si_2$ to establish the relationship between J and p [16]. In Fig. 3, we replot the phase diagram of $CeRu_2(Ge, Si)_2$ as a function of the normalized hybridization J/J_c, which thus is established for a broad range of J above and below J_c.

A similar analysis for isostructural $CeRh_2Si_2$ [17], in which it is claimed that NFL behavior is absent, shows a much greater electronic compressibility. If we calculate the J dependence of T_N for $CeRh_2Si_2$ with the same bulk modulus as for $CeRu_2Ge_2$, we find a much stronger J dependence of T_N in the Rh system than in the Ru compound (Fig. 3). In $CeRu_2Ge_2$, NFL behavior is found above 1 K for a range $J_c \pm 0.1\ [J_c - J(T_{\max})]$. In $CeRh_2Si_2$ this corresponds to a pressure range of only 0.5 kbar around the magnetic instability, possibly explaining the absence of NFL characteristics in Ref. [17].

The J dependence of the single ion Kondo temperature T_K is in good agreement with the predictions of the revisited Doniach model [4], as shown in Fig. 3. Here, $T_K(J)$ up to J_c is taken from inelastic neutron scattering studies (INS) on $CeRu_2(Ge_{1-x}Si_x)_2$, $0 \le x \le 1$ [7,18]. For larger J values $T_K(J)$ is taken from the maximum of $d\rho(p)/dT$ of $CeRu_2Ge_2$; the absolute scale of T_K is set by the values found in the INS measurements [7,18]. While the single impurity expression $T_K \propto \exp(-1/J)$ describes the experimental T_K up to $J \simeq J_c$, T_K saturates for $J > J_c$, perhaps due to intersite magnetic correlations, as proposed in the revisited Doniach model [4].

For a 3D antiferromagnet close to a magnetic instability, the phase line for the AFM state, $T_N(J)$, is predicted to vary like $(J_c - J)^{2/3}$ [2]. Our data are consistent with this prediction in a range $\Delta J/J_c = 0.07$, as is indicated by the solid line 2 in Fig. 3. In the same range ΔJ above J_c the FL ground state should exist below $T_{\mathrm{FL}} \propto J_c - J$. We utilize the identity between $CeRu_2Ge_2$ at 67.5 kbar and $CeRu_2Si_2$ at ambient pressure to test this prediction. By combining our data T_{FL} for $CeRu_2Ge_2$ ($J/J_c \ge 1.04$) with

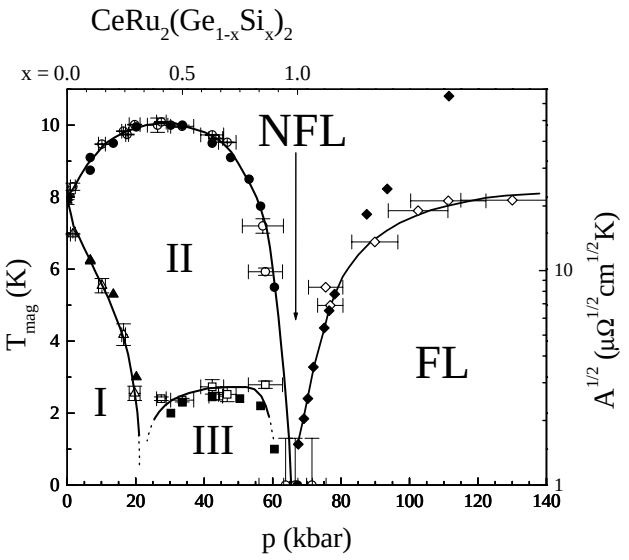

FIG. 2. The p-T phase diagram of $CeRu_2Ge_2$. The magnetic transition temperatures are T_N (∘), T_C (△), and T_M (□). The FL ground state above p_c is characterized by the T^2 resistivity coefficient, $A^{-1/2}$ (∘). We include the magnetic transition temperatures of $CeRu_2(Ge_{1-x}Si_x)_2$ (filled symbols, top axis) and the p dependence of $A^{-1/2}$ (◆) for $CeRu_2Si_2$ [9]. Solid and dashed lines are guides for the eye.

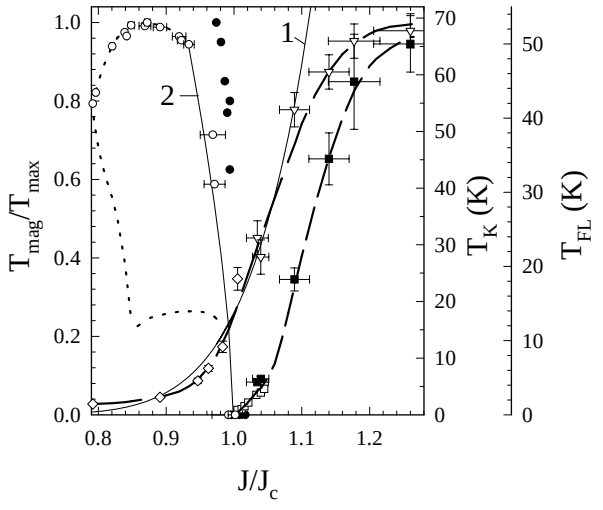

FIG. 3. The J dependence of $T_N/T_{\max}$ (∘), T_K (◇: from inelastic neutron scattering [7,18]; ▽: from resistivity) and T_{FL} in $CeRu_2Ge_2$ (■) and $CeRu_2Si_2$ (□) [9]. We include the J dependence of $T_N/T_{\max}$ in $CeRh_2Si_2$ (●) [17]. Line 1 indicates $T_K \propto \exp(-1/J)$, and line 2 denotes $T_N \propto (J_c - J)^{2/3}$ of $CeRu_2Ge_2$. Dotted lines indicate magnetic phase boundaries. Dashed lines for T_K, T_{FL} are guides to the eye.

those of pressurized $CeRu_2Si_2$ [9] ($1 \leq J/J_c \leq 1.04$), we can plot $T_{\rm FL}$ for a very wide range of J above J_c (Fig. 3). For the combined data a linear relationship is observed between $T_{\rm FL}$ and $J - J_c$ for $1 \leq J/J_c \leq 1.05$.

We have experimentally established the hybridization dependence of a Kondo lattice from the localized f-moment regime, through the magnetic instability, and far into the range of spin fluctuating materials. Our results suggest a resolution to a long standing and central question for Kondo lattices: What is the dominant energy scale for the quasiparticle interactions? The traditional view, based on studies of spin fluctuating systems with large J [5], is that the single ion Kondo temperature T_K controls both the conduction electron compensation of the single f moments at high T and the coherent FL at low T, implying a fixed relationship between $T_{\rm FL}$ and T_K. We find that this relationship is restricted to systems with $J \gg J_c$, and, in particular, is not valid near J_c. For compounds such as $CeRu_2Ge_2$ at p near p_c and ambient pressure $CeRu_2Si_2$, which both have $J \simeq J_c$, $T_{\rm FL}$ and T_K are found to have very different J dependences, which are not simply related. Specifically, the FL energy scale $T_{\rm FL}$ goes to zero, while T_K is finite and a smoothly increasing function of J. Further, Fig. 3 graphically demonstrates that in such systems T_K is no longer a single ion energy scale, but likely has substantial interion character.

Our phase diagram highlights the differing roles of the fundamental energy scales in this family of compounds. The RKKY exchange dominates for small J, leading to local moment magnetism with negligible Kondo screening. With increased J, the concept of separable intraion and interion energy scales becomes increasingly invalid. Kondo-like moment compensation ultimately leads to the suppresssion of magnetic order at J_c, and, consequently, T_N is no longer given near J_c by the familiar RKKY expression. Even so, the fact that T_N and $T_{\rm FL}$ both go to zero at J_c suggests that, at least for $J \sim J_c$, residual magnetic correlations are represented in the quasiparticle effective mass, which exhibits a corresponding divergence $m^* \propto 1/T_{\rm FL}$. Only far above J_c does $T_{\rm FL}$ become proportional to T_K, leading to the single-energy scaling behavior of relatively low-mass Kondo lattices. These conclusions are in qualitative agreement with those of the revisited Doniach model [4]. We stress that the single impurity Kondo temperature $T_K \propto \exp(-1/NJ)$ is never the dominant energy scale of this phase diagram: At small and intermediate J the physical behavior is governed by T_N and subsequently $T_{\rm FL}$, while at large J, comparison to the revisited Kondo model indicates that T_K is decidedly not single impuritylike.

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[11] $\chi_{\rm ac}$ is measured with a single pick-up coil, prohibiting a determination of its absolute value. The drop of $\chi_{\rm ac}$ at 7.2 K at ambient pressure represents the superconducting transition of the lead manometer.

[12] T_C and T_M have been computed from the maximum of $d\rho/dT$; T_N is determined from the minimum of $d^2\rho/dT^2$.

[13] B_0 for $CeRu_2Si_2$: 117 GPa; $CeCu_2Si_2$: 125 GPa; $CeRu_2Ge_2$: 98 GPa; see P. Haen *et al.*, in *Transport and Thermal Properties of f-Electron Systems*, edited by G. Oomi, H. Fuji, and T. Fujita (Plenum, New York, 1993), p. 145; C. Wassilev–Reul *et al.*, Physica (Amsterdam) **230B–232B**, 310 (1997).

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