"Non-Fermi-liquid" phenomena in heavy-fermion $CeCu_2Si_2$ and $CeNi_2Ge_2$

F. Steglich[a,b,*], P. Hellmann[a], S. Thomas[a], P. Gegenwart[a], A. Link[a], R. Helfrich[a], G. Sparn[a], M. Lang[a], C. Geibel[a], W. Assmus[c]

[a] *Institut für Festkörperphysik, SFB 252, TH Darmstadt, D-64289 Darmstadt, Germany*
[b] *Max-Planck-Institut für chemische Physik fester Stoffe, D-01187 Dresden, Germany*
[c] *Physikalisches Institut, SFB 252, Universität Frankfurt, D-60325 Frankfurt/M., Germany*

Abstract

We report low-temperature results of specific-heat and resistivity measurements on the heavy-fermion (HF) compounds $CeCu_2Si_2$ and $CeNi_2Ge_2$. "Non-Fermi-liquid" effects are observed which suggest the nearness of an antiferromagnetic quantum critical point (QCP) in either system. The observed deviations from the properties of a Landau Fermi liquid (FL) agree with theoretical predictions and point to an anomalous energy dependence of both the quasiparticle mass and the quasiparticle–quasiparticle scattering cross section. However, the complexity of the B–T phase diagram of $CeCu_2Si_2$ as well as the specific-heat results for $CeNi_2Ge_2$ measured at high pressure (1.7 GPa) indicate that the physics of HF metals is richer than anticipated in the theoretical models.

Keywords: Heavy fermions; $CeCu_2Si_2$; $CeNi_2Ge_2$; Non-Fermi liquid; Quantum phase transition

1. Introduction

In a Kondo lattice as approximated by certain Ce intermetallics [1], the formation of a Néel state (with binding energy $k_B T_{RKKY} \sim J_{loc}^2/W$) competes with the formation of a local Kondo singlet ($k_B T_K \sim \exp(W/J_{loc})$). Here $J_{loc} < 0$ is the local 4f-conduction electron exchange integral and W the conduction bandwidth. For a critical value of $|J_{loc}|$, tuned by a suitable control parameter (pressure or composition), antiferromagnetic (afm) order becomes suppressed, i.e. $T_N \to 0$. Beyond this quantum critical point (QCP), the properties of a coherent (Landau) Fermi liquid (FL) with strongly renormalized quasiparticles or heavy fermions (HF) is often observed; this heavy FL being characterized by huge coefficients γ and a in the T dependences of the 4f-derived specific heat, $\Delta C = \gamma T$, and the resistivity, $\rho - \rho_0 = \Delta\rho = aT^2$ [2]. Constituent to this Landau FL are short-range and short-lived ("quantum") afm fluctuations [3, 4] mediating the interactions between HF. These fluctuations grow in space and time when approaching the afm phase transition.

A "generalized" FL, with critically enhanced but finite quasiparticle mass m^* and anomalous energy dependences of both m^* and the quasiparticle-quasiparticle scattering cross section ($\sim a = \Delta\rho/T^2$), is predicted by renormalization group [5] as well as SCR spin-fluctuation [6]

* Correspondence address: Institut für Festkörperphysik, TH Darmstadt, Hochschulstr. 8, D-64289 Darmstadt, Germany.

theory: At the QCP one expects the following asymptotic low-T laws: $\gamma = \Delta C/T = \gamma_0 - \alpha T^{1/2}$ and $\Delta\rho = \beta T^{3/2}$. At elevated temperatures, "crossover" behavior is predicted [6], i.e. γ should obey $\gamma_0' \ln(T_0/T)$ and $\Delta\rho \sim T^{\varepsilon}$, $\varepsilon \simeq 1$, where T_0 is a characteristic spin-fluctuation temperature. Most published results concerning a QCP in Kondo-lattice systems were obtained with disordered systems. Whereas for $Ce_{1-x}La_xRu_2Si_2$ ($T_N \to 0$ at $x_c = 0.08$) the overall $\gamma(T)$ and $\Delta\rho(T)$ dependences follow well [7] the aforementioned theoretical predictions [5, 6], a largely different criticality has been inferred for $CeCu_{6-x}Au_x$, $x_c = 0.1$, from the observation that within about two decades in T (down to 60 mK), $\gamma \sim \ln(T_0/T)$, $\Delta\rho \sim T$ and $\Delta\chi = \chi - \chi_0 \sim (-T^{1/2})$ [8]. Since in such a system disorder may play a crucial role, we present below a study of "non-Fermi-liquid" (NFL) effects in the two isostructural intermetallics $CeCu_2Si_2$ and $CeNi_2Ge_2$. With these compounds, the asymptotic T-dependences for *both* $\gamma(T)$ and $\Delta\rho(T)$ as predicted by [5, 6] can be verified for the first time in any *undoped* HF metal. Attempts to "tune" $CePd_2Si_2$ [9, 10] and Ce_7Ni_3 [11] through a QCP by application of pressure have furnished [9, 11] the aforementioned "crossover" behaviors [6] and, in the case of $CePd_2Si_2$ ($p \geqslant 2.7$ GPa), an asymptotic $T^{3/2}$ dependence of $\Delta\rho(T)$ [10].

2. Evidence for a quantum critical point in $CeCu_2Si_2$

A systematic study [12, 13] of polycrystalline off-stoichiometry $Ce_{1+x}Cu_{2+y}Si_{2+z}$ samples revealed that the different physical groundstates, previously established for high-quality single crystals [14, 15], could be related to different sectors within the narrow homogeneity range of the primary 1–2–2 phase in the chemical Ce–Cu–Si phase diagram: While Cu-rich samples are HF superconductors ("S type") below $T_c = 0.65$ K, in samples with small Ce and/or Cu deficiency a transition into "phase A" is found at $T_A \simeq 0.6$–0.8 K ("A type"). Though up to now not directly confirmed via neutron diffraction, phase A is associated by most researchers with some low-moment, antiferromagnetically ordered state, presumably of finite correlation length [16]. This interpretation is supported by recent investigations on the $CeCu_2(Si_{1-x}Ge_x)_2$ system [17]. In-between the S and A sectors, "AS-type" behavior is observed in that upon cooling, the incipient A-phase transition becomes replaced by bulk superconductivity. Owing to the high sensitivity of phase A to control parameters such as sample composition or external pressure, one might be able to "tune" the system through a QCP at which $T_A \to 0$. Provided that phase A is, in fact, of afm nature clear deviations from the Landau-FL behavior as discussed above are expected. Fig. 1 shows $\gamma = \Delta C/T$ versus T on a logarithmic scale for a polycrystalline sample which exhibits a transition into phase A at $T_A \simeq 0.7$ K [18]. However, if an external pressure of 0.7 GPa is applied the A-phase transition is suppressed and replaced by a bulk superconducting one at $T_c \simeq 0.65$ K. The different natures of the respective $p = 0$ and 0.7 GPa transitions become evident when we apply a magnetic field $B = 2$ T, leaving the afm transition almost unchanged but suppressing superconductivity.

In the inset of Fig. 1 our $\Delta C(T)/T$ results taken at $B = 2$ T and $p = 0.7$ GPa and are plotted versus $T^{1/2}$. For $T \leqslant 1.2$ K, the data obey well $\gamma(T) = \gamma_0 - \alpha T^{1/2}$, with $\gamma_0 \simeq 0.99$ J/K^2 mol and

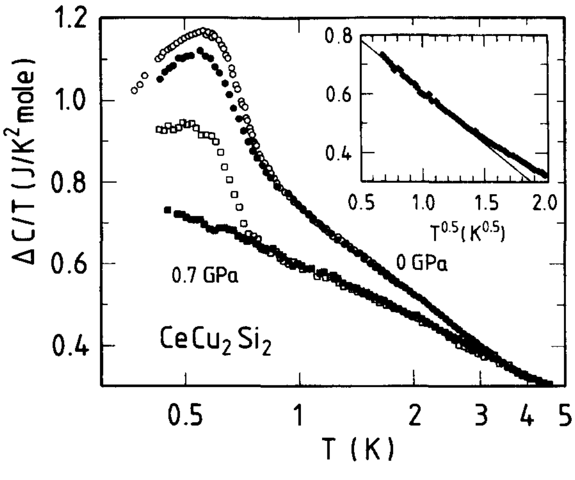

Fig. 1. $\Delta C/T$ versus T (on a logarithmic scale) for an A-type polycrystalline $CeCu_2Si_2$ sample measured at $p = 0$ and 0.7 GPa, $B = 0$ (open symbols) and 2 T (closed symbols). $p = 0$ data display A-phase, $p = 0.7$ GPa ($B = 0$ T) data superconducting transition. Inset shows $p = 0.7$ GPa ($B = 2$ T) data as $\Delta C/T$ versus $T^{1/2}$.

$\alpha \simeq 0.38$ J/K$^{2.5}$ mol. As inferred from the main part of Fig. 1, for higher T we find the expected [6] crossover law $\gamma(T) = \gamma'_0 \ln(T_0/T)$. These observations lend further support to phase A representing some (itinerant) afm ordered state.

Since we expect the A-phase transition temperature T_A to vanish within the "S" sector of the homogeneity range, we address below the ($p = 0$) n-state properties of an S-type $CeCu_2Si_2$ single crystal, identical to that studied in Ref. [14]. In Fig. 2(a), we show the T-dependence of its resistivity which, down to the lowest temperature of 20 mK, obeys well $\rho = \rho_0 + \beta T^{3/2}$ ($\rho_0 = 36$ μΩ cm, $\beta = 14.9$ μΩ cm K$^{-1.5}$ for $B = 4$ T). This T-dependence holds for $T < 1.7$ K and $B \leqslant 5$ T; see Fig. 2(b).

For $B > 6$ T, the low-temperature resistivity turns into $\Delta\rho = aT^2$, characterized by a huge and almost field-independent coefficient, $a \simeq 10$ μΩ cm K^{-2}. The apparent change from "NFL" behavior at $B \leqslant 5$ T to "FL" behavior at $B > 6$ T is substantiated by magnetoresistivity results: In an isothermal field sweep at $T = 20$ mK, $\Delta\rho(B)$ changes sign from an anomalous $\Delta\rho(B) < 0$ for $B_{c2}(T = 20\text{ mK}) < B \leqslant 5$ T to $\Delta\rho(B) > 0$ (as commonly found in a coherent FL) for $B > 6$ T. For $8\text{ T} \leqslant B \leqslant 15.5$ T, the highest field accessible, the T^2 dependence of $\Delta\rho(T)$ precedes the transition into the (as yet unidentified) high-field "phase B" (Fig. 2(b)), first discovered for an AS-type single crystal [15]. The B-phase transition manifests itself in an increase of the resistivity for $T \leqslant T_B$.

Compared to the resistivity behavior, the temperature and field dependences of the specific heat as determined for the same single crystal [19] are more complex [20]. This observation highlights a more realistic theoretical treatment of an afm QCP in $CeCu_2Si_2$, a HF compound with an anisotropic, multi-component quasiparticle system [21].

3. "Non-Fermi-liquid" effects in $CeNi_2Ge_2$

In order to investigate a "simple" reference compound isostructural to $CeCu_2Si_2$, recent activities have been devoted to non-afm ordered $CeNi_2Ge_2$, with $\gamma_0 \simeq 350$ mJ/K^2 mol [22] ($\simeq \gamma_0/2$ of $CeCu_2Si_2$). Though a peak near $T \simeq 30$ K [23] in $\chi_c(T)$, the susceptibility measured along the easy c direction, points to some short-range afm fluctuations, a low-T anomaly in $\chi_c(T)$ [23] suggests "NFL" behavior. The latter may be related to an afm QCP in $Ce(Ni_{1-x}Cu_x)_2Ge_2$ with low critical Cu concentration, $x_c < 0.2$ [13, 20].

In Fig. 3 we show new specific-heat results on a polycrystalline $CeNi_2Ge_2$ sample. While, at

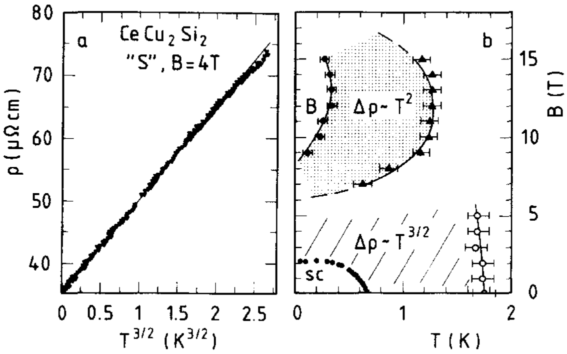

Fig. 2. (a) Resistivity of S-type $CeCu_2Si_2$ single crystal at $B = 4$ T, applied parallel to the a axis. (b): B–T diagram displaying superconducting (sc) and B phase as well as ranges in which the resistivity shows "FL" ($\Delta\rho \sim T^2$) and "NFL" ($\Delta\rho \sim T^{3/2}$) behavior, respectively.

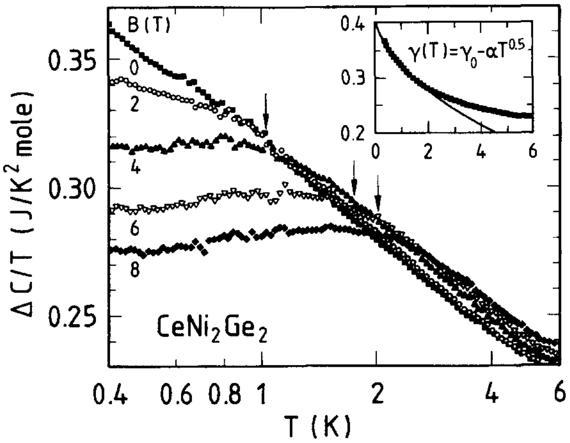

Fig. 3. Field dependence of $\Delta C/T$ versus T (on a logarithmic scale) for polycrystalline $CeNi_2Ge_2$ sample. Arrows mark onset of low-T "FL" regime for $B = 4$, 6 and 8 T, respectively (from Fig. 4(a)). Inset: $B = 0$ data on a linear temperature scale. Solid line represents $\Delta C/T = \gamma_0 - \alpha T^{1/2}$, see text.

$B = 0$, $\gamma(T) = \Delta C(T)/T \sim \ln(T_0/T)$ for $1\text{ K} \leqslant T \leqslant 4\text{ K}$, at lower T it follows $\gamma(T) = \gamma_0 - \alpha T^{1/2}$ ($\gamma_0 \simeq 0.42\text{ J/K}^2\text{ mol}$; $\alpha \simeq 0.1\text{ J/K}^{2.5}\text{ mol}$); see inset of Fig. 3. These observations are corroborated by our resistivity results on several (poly- and single crystalline) $CeNi_2Ge_2$ samples, showing residual resistivities between 1.5 and 2.5 μΩ cm: For $2.5\text{ K} < T < 10\text{ K}$ ($B = 0$) we find $\Delta\rho \sim T^{\varepsilon}$, $\varepsilon \simeq 1.2$, but for $T < 2.5\text{ K}$ ($B = 1\text{ T}$), again $\Delta\rho = \beta T^{3/2}$ ($\beta = 0.22\ \mu\Omega\text{ cm K}^{-1.5}$) holds down to 20 mK.

The isothermal magnetoresistivity indicates a change, at $B = B_\ell$, from a negative to a positive field coefficient, i.e. from anomalous scattering off long-range, long-lived afm fluctuations at low fields to scattering off some short-range, short-lived fluctuations, typical for a coherent FL, at higher fields (see Fig. 4(a)). In fact, as can be seen in Fig. 4(b), for $B > 2\text{ T}$, $\Delta\rho(T)$ is found to be $\sim T^2$ below some limiting temperature T_ℓ. As inferred from Fig. 4(a), a nearly quadratic relationship exists between B_ℓ and T_ℓ. The arrows in Fig. 3 indicate that the regime, in which $\Delta\rho \sim T^2$ holds, satisfactorily coincides with the one in which γ becomes almost independent.

However, this field-induced "FL" state shows scaling properties strongly different from that of a canonical heavy FL [2]: In the latter, $\gamma \simeq 0.3\text{ J/K}^2\text{ mol}$ would relate to a T_K of the order of several tens of K, while in Fig. 3 broad humps near $T = 1\text{ K}$ occur in $\Delta C/T$ versus T (for $B = 6$ and 8 T). In addition, for $B = 8\text{ T}$ the coefficient a of the low-T $\Delta\rho = aT^2$ law is found to be too small by one order of magnitude when compared to the a value expected from the "universal" $a \sim \gamma^2$ relation for HF compounds [2].

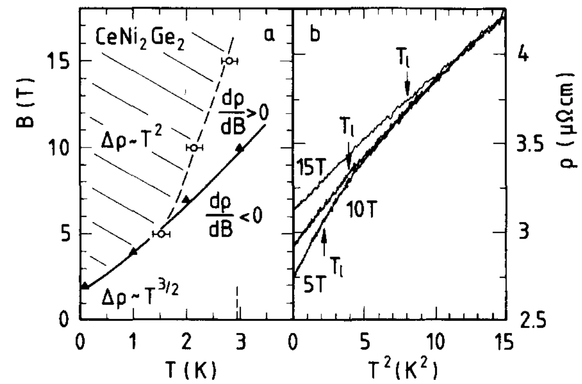

Fig. 4. B–T phase diagram for $CeNi_2Ge_2$ as derived from resistivity measurements (a), including change from negative (low B) to positive (high B) magnetoresistivity as well as ranges in which the resistivity shows "NFL" ($\Delta\rho \simeq T^{3/2}$) and "FL" ($\Delta\rho \sim T^2$) behavior. Their limits are indicated by dashed lines, the latter being described by $B_\ell = AT_\ell^2$, see text. T_ℓ values for fixed values of B_ℓ are read off ρ versus T^2 plots in (b) (arrows).

If an external pressure of $p = 1.3\text{ GPa}$ is applied to $CeNi_2Ge_2$, the "NFL" effects are found to be completely suppressed. Rather, one observes a constant $\gamma \simeq 150\text{ mJ/K}^2\text{ mol}$ for $T \leqslant 5\text{ K}$, typical of a moderately heavy Landau FL: At $p = 1.7\text{ GPa}$, $\gamma \simeq 130\text{ mJ/K}^2\text{ mol}$ is even somewhat smaller. Fully unexpectedly, a non-superconducting second-order phase transition at $T_1 = 1\text{ K}$ was recently discovered [20] at this pressure.

4. Outlook

For both $CeCu_2Si_2$ and isostructural $CeNi_2Ge_2$ distinct low-temperature anomalies have been found in the T dependences of $\gamma = \Delta C/T$ and $\Delta\rho$: $\gamma(T) = \gamma_0 - \alpha T^{1/2}$ and $\Delta\rho = \beta T^{3/2}$ suggest the nearness of an afm QCP in either system, i.e. (in space and time) slowly varying fluctuations of an afm order parameter [5, 6]. In the case of $CeNi_2Ge_2$, this holds for relatively low values of external magnetic field and hydrostatic pressure. The near degeneracy of two collective states in $CeCu_2Si_2$, i.e. HF superconductivity and phase A, as well as the occurrence of the additional (high-field) phase B display a more complex behavior of this canonical HF compound. Inconsistencies found between the $\gamma(T, B)$ [19] and $\Delta\rho(T, B)$ results for an S-type single crystal highlight a theoretical treatment of the QCP in $CeCu_2Si_2$, by taking into account its strongly anisotropic, multi-component quasiparticle system [21].

Acknowledgements

We gratefully acknowledge numerous conversations with G.G. Lonzarich and G. Zwicknagl and the expert preparation of this manuscript by C. Lang.

References

[1] S. Doniach, Physica 91 B (1977) 231.
[2] N. Grewe and F. Steglich, in: Handbook on the Physics and Chemistry of Rare Earths, Vol. 14, eds. K.A. Gschneidner Jr. and L. Eyring (Elsevier, Amsterdam, 1991) p. 343.
[3] G. Aeppli et al., Phys. Rev. Lett. 57 (1986) 122.
[4] J. Rossat-Mignod et al., J. Magn. Magn. Mater. 76 and 77 (1988) 376.
[5] A.J. Millis, Phys. Rev. B 48 (1993) 7183; M.A. Continentino, Phys. Rep. 239 (1994) 179; Z. Phys. B 101 (1996) 197.
[6] G.G. Lonzarich, College on Quantum Phases, ICTP Trieste (1994), unpublished; T. Moriya and T. Takimoto, J. Phys. Soc. Japan 64 (1995) 960.
[7] S. Kambe et al., Physica B 223 and 224 (1996) 135.
[8] H. von Löhneysen, J. Phys.: Condens. Matter 8 (1996) 9689.
[9] F.M. Grosche et al., Physica B 223 and 224 (1996) 50; S.R. Julian et al., J. Phys.: Condens. Matter 8 (1996) 9675.
[10] P. Link et al., Physica B 223 and 224 (1996) 303.
[11] K. Umeo et al., J. Phys.: Condens. Matter 8 (1996) 9743.
[12] R. Modler et al., Physica B 206 and 207 (1995) 586.
[13] F. Steglich et al., Physica B 223 and 224 (1996) 1.
[14] M. Lang et al., Phys. Scripta T 39 (1991) 135.
[15] G. Bruls et al., Phys. Rev. Lett. 72 (1994) 1754.
[16] F. Steglich et al., in: Physical Phenomena at High Magnetic Fields-II, eds. Z. Fisk, L. Gor'kov, D. Meltzer and R. Schrieffer, (World Scientific, Singapore, 1996) p. 125.
[17] O. Trovarelli et al., Physica B 223 and 224 (1996) 295.
[18] Throughout this paper, $\Delta C = C - C_{\mathrm{La}} - C_{\mathrm{hf}}$, where C_{La} denotes the specific heat of the La homologue and C_{hf} the nuclear contribution due to the hyperfine splitting of nuclear spin states by the externally applied B field.
[19] R. Helfrich, Dissertation, TH Darmstadt (1996), unpublished.
[20] F. Steglich et al., J. Phys.: Condens. Matter 8 (1996) 9909.
[21] G. Zwicknagl, Adv. Phys. 41 (1992) 203.
[22] G. Knopp et al., J. Magn. Magn. Mater. 74 (1988) 341.
[23] T. Fukuhara et al., J. Magn. Magn. Mater. 140–144 (1995) 889.