Quantum critical phenomena in undoped heavy-fermion metals

F Steglich†§, B Buschinger†, P Gegenwart†, M Lohmann†, R Helfrich†,
C Langhammer†, P Hellmann†, L Donnevert†, S Thomas†, A Link†,
C Geibel†, M Lang†, G Sparn† and W Assmus‡

† Institut für Festkörperphysik, TH Darmstadt, Hochschulstrasse 8, D-64289 Darmstadt, Germany

‡ Physikalisches Institut, Universität Frankfurt, Robert-Mayer-Strasse 2-4, D-60325 Frankfurt, Germany

Abstract. We present results of low-temperature calorimetric and resistive measurements on the isostructural heavy-fermion compounds $CeCu_2Si_2$ and $CeNi_2Ge_2$. 'Non-Fermi-liquid' effects are established which suggest the nearness of an antiferromagnetic quantum critical point (QCP) in both systems. The observed deviations from the properties of a Landau Fermi liquid (FL) may be related to anomalous energy dependences of both the quasiparticle mass and the quasiparticle–quasiparticle scattering cross section. For $CeNi_2Ge_2$, a moderately heavy FL can be recovered by application of moderate values of either magnetic field or hydrostatic pressure. For $p = 1.7$ GPa a novel, non-superconducting, phase transition has been discovered at $T_1 \simeq 1$ K.

1. Introduction

A number of intermetallics appear to be phenomenologically related to the concept of the Kondo lattice [1]. Here, the formation of a local Kondo singlet, with binding energy $k_B T^* \sim \exp(W/J_{loc})$, competes with the formation of a Néel state, with binding energy $k_B T_{\mathrm{RKKY}} \sim J_{loc}^2/W$ ($J_{loc} < 0$ being the local 4f-conduction-electron exchange integral and W the width of the conduction band). Since $|J_{loc}|$ can be varied experimentally, e.g. by application of pressure or by suitable changes in the composition, a 'quantum phase transition' at $T = 0$ from a spin-aligned, antiferromagnetic (afm) state, to a 'spin-liquid' state can be induced. The latter may show the properties of a Landau Fermi liquid (FL) with strongly renormalized quasiparticles or 'heavy fermions' (HF). Signatures of this coherent FL phase are huge coefficients, e.g. γ and a in the T-dependences of the specific heat, C, and the electrical resistivity, ρ: $C(T)/T = \gamma =$ constant and $\Delta\rho = \rho - \rho_0 = aT^2$ [2]. Whereas short-range, short-lived ('quantum') afm fluctuations mediating the interactions between quasiparticles are constituents of the coherent FL phase [3, 4], these fluctuations grow in space and time when approaching the afm phase transition.

A 'generalized' FL with critically enhanced, but finite effective quasiparticle mass m^* and anomalous energy dependences of both m^* and σ^*, the quasiparticle–quasiparticle scattering cross section, is predicted [5–7] to exist at the 'quantum critical point' (QCP), i.e. at $T_N \to 0$. Renormalization-group [5] as well as phenomenological spin-fluctuation

§ Also at: Max Planck Institute for Chemical Physics of Solids, D-01187 Dresden, Germany.

[6, 7] theory predict the following T-dependences at the QCP in the low-T limit: $\gamma = \gamma_0 - \alpha T^{1/2}$ and $\Delta\rho = \beta T^{3/2}$. At somewhat elevated temperatures, 'crossover' behaviour is expected [7] to show up within less than one decade in T: $\Delta\rho \sim T^{\epsilon}$, $\epsilon \simeq 1$, and $\gamma \sim \ln(T_0/T)$, T_0 being 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 [8] the aforementioned theoretical predictions [5–7], a substantially different criticality for $CeCu_{6-x}Au_x$, $x_c = 0.1$, has been inferred 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 $\Delta\chi = \chi - \chi_0 \sim (-T^{1/2})$ [9]. Since in such a system disorder may play a crucial role, investigations on undoped HF compounds are badly needed. Below, we present a calorimetric and resistive study of the two isostructural intermetallics $CeCu_2Si_2$ and $CeNi_2Ge_2$ (tetragonal $ThCr_2Si_2$ structure). With these systems, the asymptotic T-dependences for both $\gamma(T)$ and $\Delta\rho(T)$, as predicted by [5–7] can be verified for the first time in any undoped HF metal. Parallel attempts to 'tune' Ce_7Ni_3 [10] and $CePd_2Si_2$ [11, 12] through a QCP by application of pressure have furnished the aforementioned 'crossover' behaviour [7] in $\gamma(T)$ [10] as well as $\Delta\rho \sim T^{1.2}$ [11], but also already $\sim T^{1.5}$ [12].

The paper is organized as follows. Section 2 is devoted to $CeCu_2Si_2$. In section 2.1 we briefly discuss results of thermopower measurements on the dilute alloy $La_{0.9}Ce_{0.1}Cu_{2.2}Si_2$. In section 2.2 pressure experiments on a polycrystalline $CeCu_2Si_2$ sample are discussed which, at ambient pressure, shows an 'A-phase' transition into a (presumably) afm ordered low-T state. In section 2.3 results for an 'S-type' $CeCu_2Si_2$ single crystal are presented. Section 3 deals with $CeNi_2Ge_2$. The conclusions are given in section 4.

2. 'Non-Fermi-liquid' effects in $CeCu_2Si_2$

2.1. Thermopower of $La_{0.9}Ce_{0.1}Cu_{2.2}Si_2$

Within the last few years, single-ion multi-channel Kondo models [13] have been invoked in the interpretation of non-Fermi-liquid (NFL) phenomena observed in numerous disordered f-ion systems [14]. Recently Kim and Cox [15] discussed for Ce^{3+} impurities, with an effective $S = 1/2$ crystal-field (CF) ground state, the competition between the (local) FL fixed point of the one-channel ($M = 1$) Kondo effect and the NFL fixed points of the two- ($M = 2$) and three- ($M = 3$) channel Kondo effects. They calculated the susceptibility and the thermoelectric power (TEP) for the $M = 1, 2$ and 3 Kondo models. The sign of the TEP at low T was concluded to be a fixed-point diagnostic: a well-developed negative peak is expected for the $M = 2$ case, in contrast to the common positive peak for $M = 1$. Strict conditions concerning site symmetry and energy-level scheme have to be fulfilled for the Ce^{3+} ion to obtain the $M = 2$ model.

$La_{0.9}Ce_{0.1}Cu_{2.2}Si_2$ is considered [15] a good candidate for testing these predictions for the TEP, as the above-mentioned strict conditions for the $M = 2$ model seem to be met. Further on, NFL behaviour is evident for this alloy, as both $\chi(T)$ and the specific-heat coefficient $\gamma(T)$ were found to be proportional to $\ln(T_0/T)$ at low T [16]: if $La_{0.9}Ce_{0.1}Cu_{2.2}Si_2$ were to show a low-T negative TEP peak, the $M = 2$ Kondo model might apply. In this case the latter might hold for $CeCu_2Si_2$ [15], too, since this canonical HF compound is *known* to exhibit a giant negative TEP peak at 20 K [17]. We have measured the TEP for a polycrystalline $La_{0.9}Ce_{0.1}Cu_{2.2}Si_2$ sample. As is seen in figure 1, our data are in striking contrast to the theoretical predictions [15] mentioned above. Instead of a negative peak we observe a pronounced positive one at around $T = 9$ K, consistent

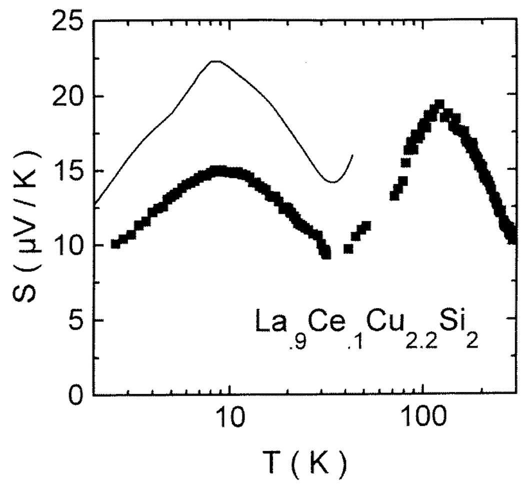

Figure 1. Measured thermoelectric power of $La_{0.9}Ce_{0.1}Cu_{2.2}Si_2$ (squares) and the 4f-derived part as calculated via Gorter–Nordheim analysis (line) for 2.5 K $< T <$ 300 K. The low-T peak is consistent with a one-channel Kondo effect of the spin-degenerate CF ground-state doublet, while the high-T peak reflects excited CF levels.

with a conventional one-channel Kondo effect. An estimate of the 4f-derived TEP via a Gorter–Nordheim analysis using TEP data for $LaCu_2Si_2$ [17] yields absolute values which are even higher and almost reach those calculated in [15] for the $M = 1$ model.

Preliminary experimental results [18] for $\gamma(T)$ and $\rho(T)$ on $La_{0.9}Ce_{0.1}Cu_{2.2}Si_2$ clearly show NFL signatures with temperature dependences compatible to those in [16], but a change to $\gamma(T) = \gamma_0 - \alpha T^{1/2}$ and $\rho(T) = \rho_0 - \beta T^{3/2}$ at still lower temperatures. It should be noted that our $\rho(T)$ results deviate strongly from $\rho(T) = \rho_0 - \beta' T^{1/2}$, the theoretical prediction for the $M = 2$ Kondo model [15]. $\Delta\gamma = \gamma - \gamma_0 \sim (-T^{1/2})$ and $\Delta\rho \sim (-T^{3/2})$ power laws have recently been calculated in the mean-field approximation for the case of a $T = 0$ spin-glass transition in a Kondo lattice with randomly quenched local magnetic moments [19, 20]. Since evidence for spin-glass transitions at low but finite temperatures in $La_{1-x}Ce_xCu_{2.2}Si_2$ with $x \geqslant 0.15$ is given in [16, 21], we propose that the NFL effects found for $La_{0.9}Ce_{0.1}Cu_{2.2}Si_2$ at low temperatures are related to the nearness to disordered magnetism, rather than a two-channel Kondo effect.

2.2. *The pressure-induced A-phase–superconducting transition*

Previous investigations on high-quality single crystals of $CeCu_2Si_2$ showed that in this compound HF superconductivity is almost degenerate with a state labelled ‘phase A’ [22, 23]. Since the latter manifests itself by distinct anomalies in magnetic properties, i.e. magnetoresistance [24], NMR [25] and μSR [26], it has been frequently associated with some afm ordered state though, so far, any attempt to observe its magnetic structure via neutron diffractometry failed. By means of a systematic study of polycrystalline off-stoichiometry $Ce_{1+x}Cu_{2+y}Si_{2+z}$ samples, the different physical ground states 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 [27, 28]: while Cu-rich samples are HF superconductors (‘S type’) below T_c = 0.65 K, an A-phase transition is found at $T_A \simeq$ 0.6–0.8 K in samples with small Ce and/or Cu deficiency (‘A type’). In between these two sectors ‘AS-type' behaviour 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 behaviour as discussed in the introductory section are expected.

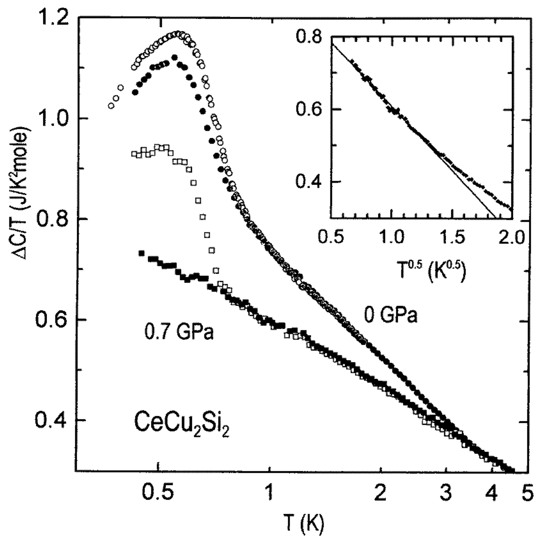

Figure 2. $\Delta C/T$ versus T (on a logarithmic scale) for a polycrystalline (A-type) $CeCu_2Si_2$ sample for $p = 0$ and 0.7 GPa, $B = 0$ (open symbols) and 2 T (closed symbols). The $p = 0$ data display the A-phase transition; the $p = 0.7$ GPa ($B = 0$ T) data display the superconducting transition.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

Figure 2 displays $\gamma = \Delta C/T$ versus T on a logarithmic scale for a polycrystalline sample which shows a transition into phase A at $T_A \simeq 0.7$ K [29]. 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 very different natures of the respective $p = 0$ and 0.7 GPa transitions are demonstrated when we apply a magnetic field $B = 2$ T, being harmless in the former but overcritical in the latter case. A comparison of the $B = 2$ T data at differing pressures is given in figure 3. We identify the following ground-state properties: 'A' ($p = 0$), 'AS' ($p = 0.1$ GPa) and the normal (n) state of 'S' ($p \geqslant 0.5$ GPa). For comparison, the n-state ($p = 0$, $B = 4$ T) data of the S-type single crystal to be discussed in the following subsection are also shown. As can be seen, above $T \simeq 2$ K the specific heats of A-, AS- and ($p = 0$) S-type $CeCu_2Si_2$ samples almost agree. In addition, if we extrapolate the zero-field data for both $p = 0$ and $p = 0.1$ GPa to $T = 0$, we estimate that the corresponding entropies, taken at $T = 2$ K, coincide within a few per cent. This seems to indicate the existence of a low-lying multi-critical point: very subtle changes in the control parameters (composition/pressure) result in very different ground-state behaviours. In fact, no measurable differences in the lattice parameters a and c of (A-, AS- and S-type) single crystals could be resolved [21, 22]. This agrees with the estimated relative changes $|\Delta a|/a$ and $|\Delta c|/c < 3 \times 10^{-4}$ (being within the resolution of the x-ray diffraction measurements), if $p = 0.1$ GPa is applied to our polycrystalline sample. Here, we have used a bulk modulus $b = 125$ GPa [30]. On the other hand, for $p = 0.7$ GPa

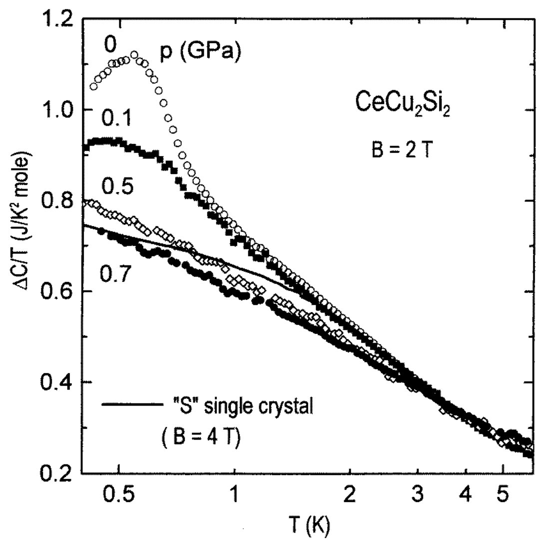

Figure 3. $\Delta C/T$ versus T (on a logarithmic scale) at $B = 2$ T and differing pressures for the same $CeCu_2Si_2$ sample as in figure 2. $B = 4$ T ($p = 0$) results for S-type single crystal (cf. figure 5) are also shown.

we estimate a reduction of both lattice parameters by 2×10^{-3}, which should be easily resolvable by x-ray diffraction. Clearly, for $p \geqslant 0.5$ GPa entropy is found to be shifted to $T > 4$ K when compared to the low-p data. The main reason for this is an increase of the Kondo temperature T^* by about 30% [31].

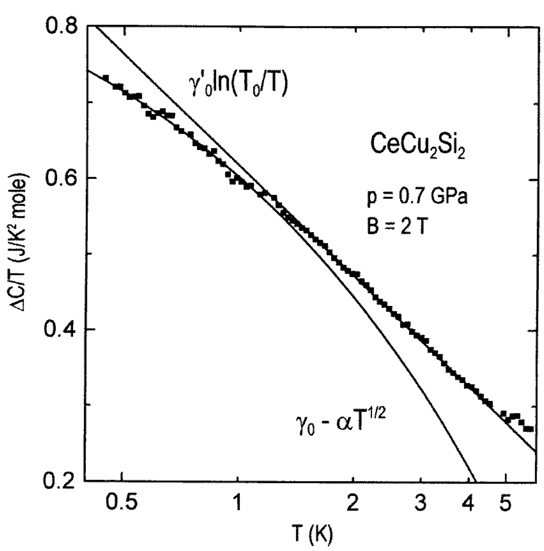

Figure 4. $\Delta C/T$ versus T (on a logarithmic scale) for the same sample as in figure 2 at $p = 0.7$ GPa and $B = 2$ T. Lines indicate 'asymptotic' and 'crossover' temperature dependences; see the text.

In the inset of figure 2 our $\Delta C(T)/T$ results taken at $p = 0.7$ GPa and $B = 2$ T 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^{-1} and $\alpha \simeq 0.38$ J K$^{-2.5}$ mol^{-1}. As inferred from figure 4, for higher T we find the expected [7] crossover law $\gamma(T) = \gamma_0' \ln(T_0/T)$, where $\gamma_0' = 0.21$ J K^{-2} mol^{-1} and $T_0 = 18.8$ K. To our knowledge this is the first observation [32], for any undoped HF compound, of the predicted [5–7] behaviour in the specific heat near a QCP of afm type.

Since we expect the A-phase transition temperature T_A to vanish somewhere within the 'S' sector of the homogeneity range, we address in the following subsection the n-state properties of an S-type $CeCu_2Si_2$ single crystal, measured at ambient pressure.

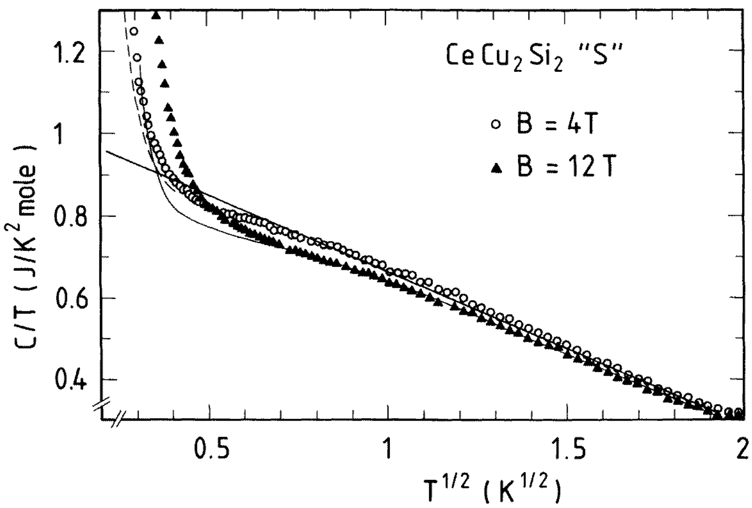

Figure 5. Normal-state specific heat of S-type $CeCu_2Si_2$ single crystal as C/T versus $T^{1/2}$ for two different magnetic fields applied parallel to the a-axis. Dashed ($B = 4$ T) and thin solid ($B = 12$ T) lines represent data after subtraction of nuclear contributions due to the external field. The thick solid line representing $C/T = \gamma_0 - \alpha T^{1/2}$ is intended as a guide to the eye.

2.3. Normal-state properties of S-type $CeCu_2Si_2$

An S-type $CeCu_2Si_2$ crystal, identical to that studied by Lang *et al* [22], was recently reexamined via specific-heat and resistivity measurements; cf. figures 5 and 6. As can be seen in figure 5, $C(T)/T$ follows a $\gamma_0 - \alpha T^{1/2}$ dependence only over a limited temperature range between 0.5 K and 3 K (the straight line). For $B = 2$ T, both $\gamma_0 \simeq 1.12$ J K^{-2} mol^{-1} and $\alpha \simeq 0.42$ J K$^{-2.5}$ mol^{-1} exceed the corresponding values for the polycrystalline sample (measured at $p = 0.7$ GPa) as discussed in the preceding subsection. With increasing field (up to $B = 5$ T) both parameters are found to gradually decrease (cf. figure 5). Upon lowering the temperature, however, $\gamma(T)$ starts to deviate from the $T^{1/2}$-behaviour, with the tendency to saturate for $T \to 0$. A detailed analysis of this seeming change from 'NFL' to FL behaviour for $T < 0.4$ K is, unfortunately, impeded by the steep upturn in $\gamma(T)$ below $T \simeq 0.25$ K. The latter cannot be explained by the effect of the external field only on the nuclear Cu/Si spins. An anomalous enhancement of the hyperfine coupling, i.e. an (average) finite 'internal magnetic field' transferred to the Cu/Si sites has to be invoked to quantitatively account for the anomalous T-dependence. One possibility is that the 'internal

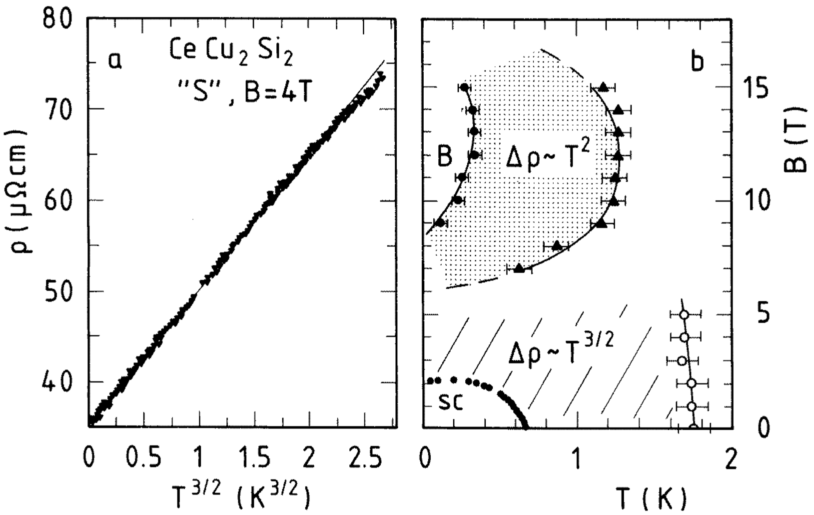

Figure 6. (a) The resistivity of the same single crystal as in figure 5 at $B = 4$ T, parallel to the a-axis, i.e. longitudinal to the current. (b) The B–T diagram displaying the superconducting (sc) phase and the B phase as well as ranges in which the resistivity shows 'FL' ($\Delta\rho \sim T^2$) and 'NFL' ($\Delta\rho \sim T^{3/2}$) behaviour, respectively.

field' marks a polarization of the Kondo-compensated 4f spins, i.e. via Ce–Ce correlations. However, in contrast to both A- and AS-type $CeCu_2Si_2$ samples no phase transition anomaly is associated with the formation of this 'internal field'.

Even more surprising, for $T \lesssim 1.7$ K the n-state resistivity *does* obey the asymptotic behaviour predicted [5–7] at the afm QCP, $\rho = \rho_0 + \beta T^{3/2}$ ($\rho_0 \simeq 36$ $\mu\Omega$ cm and $\beta = 14.9$ $\mu\Omega$ cm $K^{-1.5}$). This holds down to $T \simeq 20$ mK, the lowest temperature of the measurement (figure 6(a)) and, as indicated in figure 6(b), for magnetic fields $B \leqslant 5$ T. For $B > 6$ T, the low-temperature resistivity turns into $\rho = \rho_0 + aT^2$, characterized by a huge and almost field-independent coefficient, $a \simeq 10$ $\mu\Omega$ cm K^{-2}. This change from 'NFL' behaviour at $B \leqslant 5$ T to a seeming FL behaviour 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 $B > 5$ T.

For 8 T $\leqslant B \leqslant$ 15.5 T, the highest field of our experiment, the T^2-dependence of $\Delta\rho(T)$ precedes the transition into the high-field 'phase B' (figure 6(b)), first discovered for an AS-type single crystal [23]. The B-phase transition manifests itself in an increase of the resistivity for $T \leqslant T_B$. Our experiments show that in the S-type single crystal, phase B forms at somewhat lower temperatures than in the AS-type crystals [33]. Moreover, the present results prove that phase B can form even in the absence of (the low-field) phase A.

In contrast to the resistivity results, the specific-heat data for $B = 12$ T of figure 5 do not reflect the onset of phase B. This may indicate a gross reduction of the ordered Ce moment, compared to that of an A-type (or AS-type) sample [33]. Note, however, that the 'internal field' inferred from our specific-heat data cannot be associated with the B-phase transition as the former seems to be present already for $B = 4$ T. The apparent change from 'NFL' ($B \leqslant 5$ T) to FL ($B > 6$ T) as inferred from the resistivity experiments also has no correspondence in the specific-heat results. In particular, the high-field $\Delta\rho \sim T^2$ behaviour contrasts with a rather complex 'NFL' behaviour in $\gamma(T)$ (figure 5). A similar discrepancy is well known already at zero field for both A- [34] and AS-type [33] samples, showing a $\gamma(T)$ of 'NFL' kind (cf. figure 3), but $\Delta\rho = aT^2$ with $a \simeq 10\ \mu\Omega$ cm K^{-2}. These complications add to the problem of the, as yet, incompletely identified nature of phase A. They pose a challenge to the theoretical treatment of a QCP in a strongly anisotropic, multi-component quasiparticle system as realized in $CeCu_2Si_2$ [35].

In order to study a comparatively simple reference compound we have recently initiated low-T measurements on $CeNi_2Ge_2$. They are the subjects of the next section.

3. $CeNi_2Ge_2$: 'non-Fermi-liquid' effects at low magnetic field and low pressure

$CeNi_2Ge_2$ exhibits neither afm order nor superconductivity. A highly anisotropic uniform susceptibility, $\chi(T)$, was reported by Fukuhara *et al* [36]. $\chi_c(T)$, as measured when the field was applied along the easy tetragonal c-axis, shows a broad maximum near $T_0 \simeq 30$ K. This was interpreted as evidence for short-range afm correlations, similar to the case for $CeRu_2Si_2$ with $T_0 \simeq 10$ K [37]. However, in the data of Fukuhara *et al* [36] an upturn in $\chi_c(T)$ at lower temperature shows up whose origin is not clear. It may be, at least partly, intrinsic and related to the negative temperature coefficient of $\gamma(T)$ that was observed for $T > 0.3$ K already some time ago [38]. Knopp *et al* [38] reported a low-T value $\gamma_0 \simeq 350$ mJ K^{-2} mol^{-1} for this compound ($\simeq 1/2$ of γ_0 for $CeCu_2Si_2$) and, therefore, labelled it 'medium heavy fermion'.

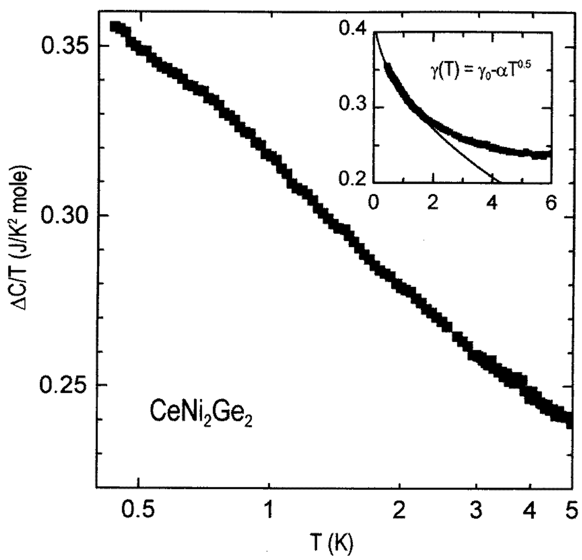

Figure 7. $\Delta C/T$ versus T (on a logarithmic scale) for a polycrystalline $CeNi_2Ge_2$ sample. Inset: the same data on a linear temperature scale. The solid line represents $\Delta C/T = \gamma_0 - \alpha T^{1/2}$; see the text.

A reinvestigation of a polycrystalline $CeNi_2Ge_2$ sample yielded the specific-heat results displayed in figure 7: for $1\ \text{K} \leqslant T \leqslant 4$ K, $\gamma = \gamma_0' \ln(T_0/T)$ with $\gamma_0' \simeq 0.05$ J K^{-2} mol^{-1} and $T_0 \simeq 400$ K [39], whereas the low-T data can be well fitted by $\gamma = \gamma_0 - \alpha T^{1/2}$, with $\gamma_0 \simeq 0.42$ J K^{-2} mol^{-1} and $\alpha \simeq 0.10$ J $K^{-2.5}$ mol^{-1}; see the inset. If Ni is partially replaced by Cu, a transition into a long-range afm ordered state can be induced [40]. For polycrystalline $Ce(Ni_{1-x}Cu_x)_2Ge_2$ samples, the critical Cu concentration, at which $T_N \to 0$, was found to be $x_c \simeq 0.2$ [28]. However, current investigations on single crystals indicate a substantially smaller value of x_c and, therefore, $CeNi_2Ge_2$ to be sufficiently close to a QCP.

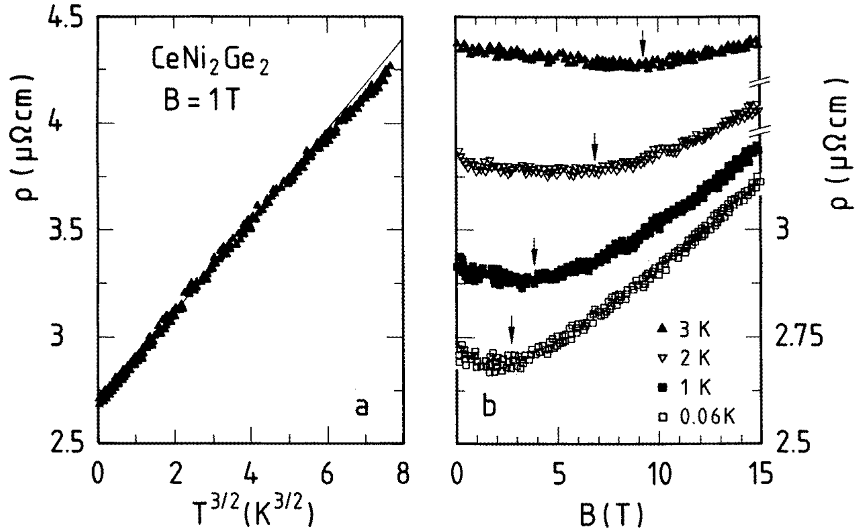

Figure 8. The resistivity of a polycrystalline $CeNi_2Ge_2$ sample. (a) ρ versus $T^{3/2}$ at $B = 1$ T. The line represents $\rho = \rho_0 + \beta T^{3/2}$. (b) ρ versus B at differing temperatures. The arrows mark changes from negative to positive magnetoresistivity; see the text.

Recently we commenced resistivity experiments on both polycrystalline and single-crystalline $CeNi_2Ge_2$ samples whose residual resistivities varied between 1.5 and 2.5 $\mu\Omega$ cm. At low fields ($B \leqslant 2$ T), $\Delta\rho(T)$ follows surprisingly well a $\beta T^{3/2}$-power law ($\beta = 0.22$ $\mu\Omega$ cm $K^{-1.5}$) between the lowest temperature of 20 mK and 2.5 K; see figure 8(a). This unique T-dependence, independently reported by the Cambridge group [11], is indicative of anomalous scattering, presumably due to soft long-range afm fluctuations near a QCP which can be suppressed by an external magnetic field. The arrows in figure 8(b) mark the transition between the anomalous negative and the common (for a coherent FL) positive magnetoresistance; cf. figure 9(a). In the B–T plane, the existence range of a coherent (Landau) FL is found above the upper dashed line. The latter marks the highest temperature up to which both $\Delta\rho \sim T^2$ (figure 9(b)) and $\gamma \simeq$ constant (figure 10; cf. also [9]) is observed at a fixed field $B > 2$ T. A nearly quadratic relationship is found between the B_ℓ- and T_ℓ-values limiting the Landau-FL regime. In summary, the magnetic field appears to suppress the low-lying, extended afm fluctuations near a QCP and, thus, to induce a coherent FL where the quasiparticle–quasiparticle interactions are mediated by short-range quantum fluctuations.

Like application of an external field, application of an external hydrostatic pressure can provide important information on the magnetic phase diagram of $CeNi_2Ge_2$: as is shown in the inset of figure 11, $p = 1.3$ GPa is sufficient to remove completely the NFL effects in the specific heat below $T \simeq 5$ K and to establish a 'moderately heavy' Landau FL ($\gamma \simeq 0.15$ J K^{-2} mol^{-1}) on the threshold to 'intermediate Ce valence' [2]. This effect of pressure is in qualitative accord with the expectation for the schematic Kondo-

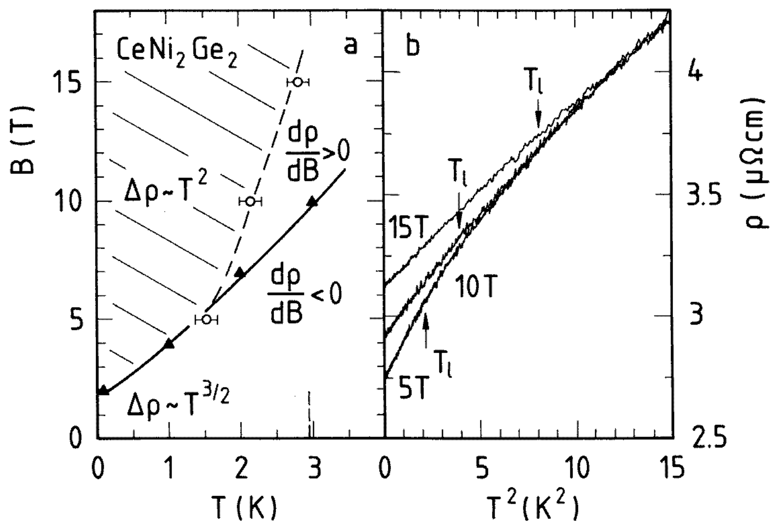

Figure 9. The B–T phase diagram for $CeNi_2Ge_2$ as derived from resistivity measurements (a), including the change from negative (low-B) to positive (high-B) magnetoresistance as well as ranges in which the resistivity shows 'NFL' ($\Delta\rho \sim T^{3/2}$) and 'FL' ($\Delta\rho \sim T^2$) behaviour. Their limits are indicated by dashed lines, the latter being described by $B_\ell = AT_\ell^2$; see the text. T_ℓ-values for fixed values of B_ℓ are read off ρ versus T^2 plots in (b) (arrows).

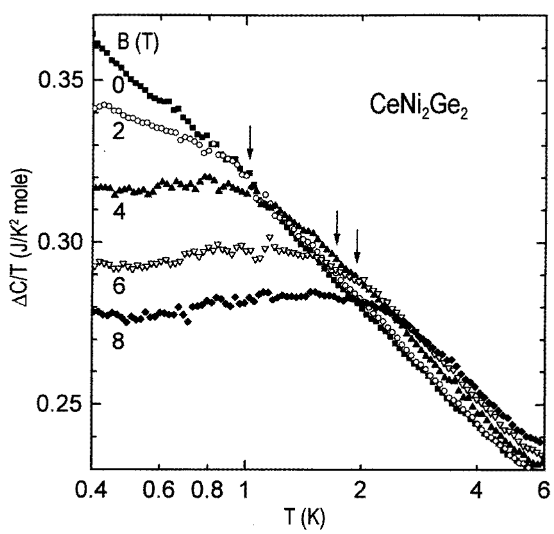

Figure 10. The field dependence of $\Delta C/T$ versus T (on a logarithmic scale) for the same sample as in figure 7. The arrows mark the onset of the low-T 'FL' regime for $B = 4, 6$ and 8 T, respectively (from figure 9(a)).

lattice phase diagram [5, 41]. In agreement with the common observation that the Kondo temperature in Ce systems is increased by pressure, we find a further reduction of γ to about 0.13 J K^{-2} mol^{-1} if $p = 1.7$ GPa is applied.

Fully unexpected, however, is the discovery that this Landau FL undergoes a mean-

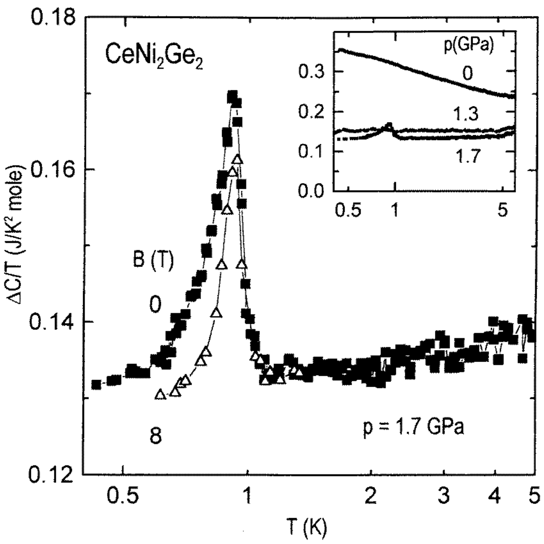

Figure 11. The pressure dependence of $\Delta C/T$ versus T (on a logarithmic scale) for the same sample as in figure 7. $B = 0$ data for differing pressure values are shown in the inset. The $p = 1.7$ GPa data at $B = 0$ and $B = 8$ T indicate a non-superconducting second-order phase transition at $T_1 = 1$ K; see the text.

field-type phase transition at $T_1 \simeq 1$ K whose position is unaffected by a magnetic field up to 8 T. AC-susceptibility measurements indicate a very small signal to be associated with this transition. Therefore, we can safely discard the possibility that it is superconducting in nature. In view of the medium size of the specific-heat coefficient, indicating a large $T^* \simeq 50$ K, an antiferromagnetic transition would also be very surprising. It is interesting to note that the unit-cell volume of $CeNi_2Ge_2$ at $p = 1.7$ GPa corresponds to $V_c \simeq 0.166$ nm^3 (using $b = 87$ GPa [42]). In three isostructural Ce compounds, low-T afm transitions occur which can be replaced by superconducting transitions (with $T_c < 0.7$ K), once a similar value of the cell volume is dictated by a suitable pressure: $CeCu_2Ge_2$ ($p_c = 7.1$ GPa, $V_c \simeq 0.168$ nm^3 [43]), $CeRh_2Si_2$ ($p_c \simeq 1$ GPa, $V_c \simeq 0.168$ nm^3 [44]) and, of course, $CeCu_2Si_2$ ($p_c \simeq 0$, $V_c \simeq 0.167$ nm^3). Future activities should explore the crucial role of this particular cell volume [45].

4. Conclusion

Through measurements of both the specific heat and the resistivity the nearness of an afm QCP could be demonstrated for two undoped HF compounds; the indication for this is asymptotic power laws predicted by theory [5–7], i.e. $\gamma = \gamma_0 - \alpha T^{1/2}$ and $\rho = \rho_0 + \beta T^{3/2}$. These temperature dependences indicate a strong violation of Landau-FL behaviour, the latter essentially relying on quasiparticle–quasiparticle interactions that are of short range. While afm nearest-neighbour quantum fluctuations are, in fact, constituents of the Landau-FL phase of canonical HF metals, these fluctuations become soft and long range when approaching the afm ordered state at very low T. As a result the quasiparticle–quasiparticle cross section (being proportional to the coefficient of the aT^2-term in the resistivity) appears to diverge $\sim T^{-1/2}$ at the QCP as $T \to 0$. In addition, the effective carrier mass m^* appears to be critically enhanced and to show also an unusual energy dependence [5–7]: thus, HF compounds behave as 'generalized' (i.e. non-Landau) FL at $T_N = 0$.

For $CeNi_2Ge_2$ a nearby long-range afm ordered state can be made visible via substituting at most 20 at.% Cu for Ni. In the case of $CeCu_2Si_2$, a (presumably) afm ordered phase A becomes suppressed for very tiny Cu/Ce excess which, on the other hand, stabilizes HF superconductivity. Application of a relatively low pressure, $p \geqslant 0.5$ GPa, is sufficient to turn an A-type into an S-type sample. The n-state specific heat of the latter exhibits the asymptotic temperature dependence mentioned before. On the other hand, if the n-state specific heat of an S-type single crystal is measured *at zero pressure*, some 'internal magnetic field' is suggested by both a flattening of $\gamma(T)$ below $T \simeq 0.4$ K and a substantially enhanced hyperfine splitting of nuclear spin states. Interestingly enough, no cooperative phase transition anomalies can be resolved in our measurements. Moreover, resistivity experiments on the same single crystal do show, for $B \leqslant 5$ T and over about two decades in temperature, the $T^{3/2}$-power law as expected at an afm QCP [5–7]. At higher magnetic fields, a seemingly coherent FL is inferred from $\Delta\rho = aT^2$ and is found to precede a low-T transition into the unidentified phase B. These apparent differences in the $\gamma(T, B)$ and $\Delta\rho(T, B)$ results obtained at $p = 0$ for an S-type $CeCu_2Si_2$ single crystal highlight a theoretical treatment of the QCP in this HF compound, taking into account its strongly anisotropic, multi-component quasiparticle system [35].

Further experimental work on both compounds is necessary in order to understand why the 'correct' asymptotic behaviour [5–7] can be observed *off* the QCP, e.g. for S-type $CeCu_2Si_2$ under a finite pressure, $p \geqslant 0.5$ GPa. Other topics for future research are (i) the nature of the novel phase transition in $CeNi_2Ge_2$ under $p = 1.7$ GPa and (ii) the low-temperature properties near the QCP of spin-glass type in the dilute (**La**, Ce)$Cu_{2.2}Si_2$ system.

Acknowledgments

We have greatly benefited from enlightening discussions with Gil Lonzarich and Gertrud Zwicknagl. One of us (FS) would also like to acknowledge numerous conversations with Piers Coleman, Mucio Continentino, Dan Cox, Andy Millis, Subir Sachdev and Alexei Tsvelik as well as support by the National Science Foundation under Grant No PHY94-07194 through which participation in the ITP Workshop on Non-Fermi-Liquid Behaviour in Solids became possible. This work was performed within the research programme of the Sonderforschungsbereich 252 Darmstadt/Frankfurt/Mainz.

References

[1] Doniach S 1977 *Physica* B **91** 231
[2] Grewe N and Steglich F 1991 *Handbook on the Physics and Chemistry of Rare Earths* vol 14, ed K A Gschneidner Jr and L Eyring (Amsterdam: Elsevier) p 343
[3] Aeppli G *et al* 1986 *Phys. Rev. Lett.* **57** 122
[4] Rossat-Mignod J *et al* 1988 *J. Magn. Magn. Mater.* **76+77** 376
[5] Millis A J 1993 *Phys. Rev.* B **48** 7183
[6] Lonzarich G G 1994 *College on Quantum Phases (Trieste)* (unpublished)
[7] Moriya T and Takimoto T 1995 *J. Phys. Soc. Japan* **64** 960
[8] Kambe S *et al* 1996 *Physica* B **223+224** 135
[9] von Löhneysen H *et al* 1996 *J. Phys.: Condens. Matter* **8** 9689
[10] Umeo K, Kadomatsu H and Takabatake S 1996 *J. Phys.: Condens. Matter* **8** 9743
[11] Julian S R, Pfleiderer C, Grosche F M, Mathur N D, McMullan G J, Diver A J, Walker I R and Lonzarich G G 1996 *J. Phys.: Condens. Matter* **8** 9675
[12] Link P *et al* 1996 *Physica* B **223+224** 303

[13] Cox D L *et al* 1996 *J. Phys.: Condens. Matter* **8**
[14] Maple M B *et al* 1996 *J. Phys.: Condens. Matter* **8**
[15] Kim T S and Cox D L 1995 *Phys. Rev. Lett.* **75** 1622
[16] Andraka B 1994 *Phys. Rev.* B **49** 3589
[17] Franz W *et al* 1978 *Z. Phys.* B **31** 7
[18] Hellmann P *et al* 1996 to be published
[19] Sachdev S *et al* 1995 *Phys. Rev.* B **52** 10 286
[20] Sengupta A M and Georges A 1995 *Phys. Rev.* B **52** 10 295
[21] Aliev F G *et al* 1984 *J. Low Temp. Phys.* **57** 61
[22] Lang M *et al* 1991 *Phys. Scr.* T **39** 135
[23] Bruls G *et al* 1994 *Phys. Rev. Lett.* **72** 1754
[24] Rauchschwalbe U *et al* 1987 *J. Magn. Magn. Mater.* **63+64** 447
[25] Nakamura H *et al* 1988 *J. Magn. Magn. Mater.* **76+77** 517
[26] Uemura Y J *et al* 1989 *Phys. Rev.* B **39** 4726
[27] Modler R *et al* 1995 *Physica* B **206+207** 586
[28] Steglich F *et al* 1996 *Physica* B **223+224** 1
[29] Throughout this paper, $\Delta C = C - C_{La} - C_{hf}$, 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 field $\boldsymbol{B}$.
[30] Spain I L *et al* 1986 *Physica* B **139+140** 449
[31] Bleckwedel A and Eichler A 1985 *Solid State Commun.* **56** 693
[32] Hellmann P *et al* 1996 *Czech. J. Phys. Suppl.* 5 **46** 2591
[33] Gegenwart P *et al* 1996 *Proc. Int. Conf. on Strongly Correlated Electron Systems (SCES '96) (Zürich); Physica* B at press
[34] Steglich F *et al* 1996 *Physical Phenomena at High Magnetic Fields II* ed Z Fisk, L Gor'kov, D Meltzer and R Schrieffer (Singapore: World Scientific) p 125
[35] Zwicknagl G 1992 *Adv. Phys.* **41** 203
[36] Fukuhara T *et al* 1995 *J. Magn. Magn. Mater.* **140–144** 889
[37] Flouquet J *et al* 1985 *J. Magn. Magn. Mater.* **52** 85
[38] Knopp G *et al* 1988 *J. Magn. Magn. Mater.* **74** 341
[39] For a discussion of such extreme parameters, see
Sereni J G *et al* 1996 *Proc. Int. Conf. on Strongly Correlated Electron Systems (SCES '96) (Zürich); Physica* B at press
[40] Sparn G *et al* 1988 *J. Magn. Magn. Mater.* **76+77** 153
[41] Continentino M A 1994 *Phys. Rep.* **239** 179; 1996 *Z. Phys.* B **101** 197
[42] Fukuhara T *et al* 1996 *Proc. Int. Conf. on Strongly Correlated Electron Systems (SCES '96) (Zürich); Physica* B at press
[43] Jaccard D *et al* 1992 *Phys. Lett.* **163A** 475
[44] Movshovich R *et al* 1996 *Phys. Rev.* B **53** 8241
[45] Wassilew-Reul C *et al* 1996 *Proc. Int. Conf. on Strongly Correlated Electron Systems (SCES '96) (Zürich); Physica* B at press