On the Relationship of Magnetism and Superconductivity in Materials Containing Partially Filled f Shells

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We discuss the low-temperature normal-state properties of the prototypical heavy-fermion (HF) superconductor $CeCu_2Si_2$ as well as their variations as a function of stoichiometry, applied pressure and magnetic field. Related results on the homologous compound $CeNi_2Ge_2$ are also presented. In contrast to $CeCu_2Si_2$ where HF superconductivity can survive in the presence of high residual resistivities ($\rho_0 \leq 40$ $\mu\Omega$cm), the newly discovered HF superconducting state in $CeNi_2Ge_2$ requires $\rho_0 < 1.5$ $\mu\Omega$cm. Our results point to striking disparities between resistivity and specific-heat results in both compounds. These observations raise doubts as to the applicability of the "nearly-antiferromagnetic-Fermi-liquid" theory to HF compounds.

§1. Some introductory remarks concerning history and recent achievements

Usually magnetism and superconductivity are considered to be mutually exclusive phenomena. While magnetism is a consequence of the strong Coulomb repulsion between electrons on localized core shells, superconductivity arizes due to an attractive interaction between the delocalized electrons in a metal such that this attraction overcompensates their screened Coulomb repulsion. In the classical superconductors the electron-electron attraction is mediated by lattice vibrations and leads to pairs of electrons with anti-aligned spins.

The apparent antagonism between magnetism and superconductivity has been intensively studied and basically resolved within the last three decades with classical superconductors containing ions of rare-earth elements. The latter show an incompletely filled $4f$ shell residing within the core of the ion. Very bizarre behaviors have been discovered for such systems; for example, superconducting states that either disappear upon cooling to sufficiently low temperatures,1) develop only at sufficiently high magnetic fields,2) or coexist with long-range antiferromagnetic (afm) order.3) Common to all these materials are two mutually interpenetrating systems of electrons, the localized $4f$ electrons responsible for the magnetic phenomena and the delocalized conduction electrons carrying superconductivity.

A novel situation emerged in 1979 when superconductivity was discovered for $CeCu_2Si_2$, an intermetalic compound of trivalent Cerium4) whose non-f-ion reference compound $LaCu_2Si_2$ does not superconduct. This is opposed to the extremely strong pair-breaking effect a tiny amount of Ce^{3+} ions (usually less than 1 at%) has in classical superconductors:1) The $4f$ electrons in $CeCu_2Si_2$ not only give rise to the local magnetic moments as observed at sufficiently high temperatures ($T > 20$ K), but they are also essential for the superconducting state that forms below $T_c \simeq 0.6$ K.4) In a "Kondo-lattice" system like $CeCu_2Si_2$ (with Kondo temperature $T_K \approx 15$ K) the local $4f$ moments disappear gradually below T_K. Simultaneously, itinerant states of an extremely large effective mass at the Fermi energy are gradually formed. Residual interactions between these novel quasiparticles ("heavy fermions", HF) can result in exotic magnetically ordered and/or superconducting ground states.

Except for $CeCu_2Si_2$ there exist five other HF superconductors (SC), i.e., UBe_{13},5) UPt_3,6) URu_2Si_2,7) $UNi_2Al_3$8) and UPd_2Al_3.9) In addition, the afm ordered Kondo-lattice systems $CeCu_2Ge_2$,10) $CePd_2Si_2$,11) $CeRh_2Si_2$12) and $CeIn_3$13) adapt a HF superconducting state once a sufficiently large hydrostatic pressure $p \approx p_c$ is applied, where p_c is the critical pressure necessary to suppress afm order. For both $CePd_2Si_2$11) and $CeIn_3$,13) strong deviations from the properties of a heavy Landau Fermi liquid (LFL) were observed in the normal-state at low temperature. These "Non-Fermi-Liquid" (NFL) effects were associated with the nearness of the magnetic instability at $p = p_c$ where the afm ordering temperature $T_N \to 0$: The abundance of very strong, extended and long-lived fluctuations of the local staggered magnetization are expected14–17) to result in unusual energy dependences of the microscopic parameters m^*, the effective quasiparticle mass, and σ^*, the effective quasiparticle-quasiparticle scattering cross section. These lead to unusual temperature dependences in the measurable quantities $\gamma(T) = \Delta C(T)/T$ and $a(T) = \Delta\rho(T)/T^2$.18) According to the "nearly antiferromagnetic Fermi-liquid" (NAFFL) theory, a generalization of LFL theory, the asymptotic low-T laws for a 3D system of afm spinfluctuations are: $\gamma = \gamma_0 - \alpha T^{1/2}$ and $\rho = \rho_0 + \beta T^{3/2}$ $(\alpha, \beta > 0)$. For very clean samples, however, $\Delta\rho = \rho - \rho_0 = \beta T^{\varepsilon}$ with $1 < \varepsilon < 1.5$ was predicted19) to describe the measured resistivity in a wide temperature range.

The occurence of both NFL effects in the low-T normal-state and HF superconductivity in $CePd_2Si_2$ and $CeIn_3$ has led to the suggestion that the two phenomena can be traced back to the same origin.[13] At present, there is no clear correlation between them: For example, p-induced HF superconductivity in $CeCuGe_2$[10] and $CeRh_2Si_2$[12] seems to form out of a LFL state very near the critical pressure p_c which marks the afm instability. In addition, superconductivity in the U-based HF compounds like UPt_3[6] and UPd_2Al_3[9] develops out of a heavy LFL state which coexists with long-range afm order. In summary, NFL effects in the normal-state are not *necessary* for the occurence of a HF superconducting state. Such NFL effects seem not to be *sufficient* either because they have been recently established in the low-T phase of $YbRh_2Si_2$,[20] which does not superconduct down to $T \approx 10$ mK.[21]

In order to investigate the relationship between magnetic order, HF superconductivity and NFL effects by transport *and* thermodynamic measurements, we address two stoichiometric HF compounds that do show these phenomena already at ambient pressure: $CeCu_2Si_2$ (sect. 2) and $CeNi_2Ge_2$ (sect. 3). A short outlook is given in sect. 4.

§2. "Phase A", NFL effects and HF superconductivity in $CeCu_2Si_2$

Early investigations of $CeCu_2Si_2$ were plagued by severe "sample dependences" in the low-T properties. This is illustrated in Figs. 1 and 2 which display the temperature dependence of $C(T)/T$ in the range 0.05 K $< T <$ 1 K for two $CeCu_2Si_2$ samples with very similar residual resistivity ($\rho_0 \approx 20$ $\mu\Omega$cm). Although both samples are bulk superconductors according to measurements of the DC Meissner effect, their specific-heat behavior is obviously very different. The single-crystal data of Fig. 1 show[22] a sharp and huge superconducting phase-transition anomaly at $T_c = 0.62$ K. Interestingly enough, the Sommerfeld coefficient $\gamma(T) = \Delta C(T)/T \approx C(T)/T$ is not constant for T & T_c, indicating NFL properties as discussed in more detail below. No residual value, γ_{res}, can be resolved well below T_c. Also, while $C(T)$ in the superconducting state cannot be described by an exponential it does not follow a power law, T^{ϵ}, either: A T-dependent exponent ϵ would have to be assumed being ≈ 2 for T . T_c but > 3 for $T < 0.2$ K. By contrast, the polycrystal data of Fig. 2 indicate for $B = 0$ the existence of a large residual value, $\gamma_{res} \approx 0.6$ J/K^{2}mole. Further on, no superconducting phase-transition anomaly can be resolved at $T_c(B = 0)$ (0.44 K due to DC-Meissner measurements[23]). On the other hand, the large and broad anomaly in C/T *vs* T which develops below $T = 0.7$ K marks the onset of "phase A" which will be discussed below.

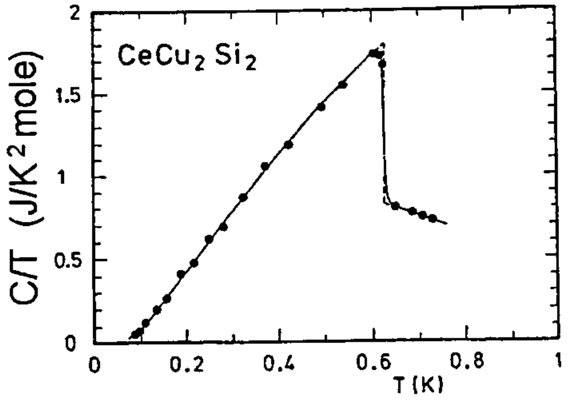

Fig. 1. Specific heat, as C/T *vs* T, of a $CeCu_2Si_2$ single crystal between 0.08 K and 0.8 K at $B = 0$.

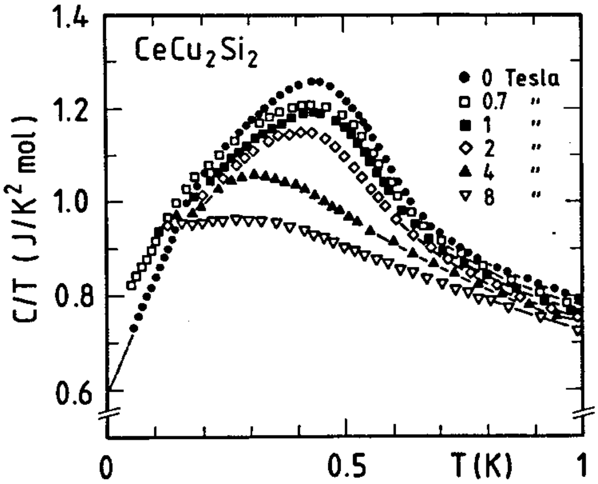

Fig. 2. Specific heat, as C/T *vs* T, of a $CeCu_2Si_2$ polycrystal between 0.05 K and 1 K at $B = 0$ and differing magnetic fields.

Owing to the size of the broadened anomaly shown in Fig. 2 as well as that of the (negative) thermal-expansion jump,[24] "phase A" was considered an ordering phenomenon in the system of HFs.[24] It manifests itself by phase-transition signatures in several magnetic properties, e.g., magnetoresistance,[25] NMR/NQR[26] and μSR.[27] Fig. 3 displays the temperature dependence of the normal-state electrical resistivity measured at $B = 5$ T for a $CeCu_2Si_2$ single crystal near the "A-phase" transition temperature, $T_A \approx 0.6$ K. A spin-density-wave (SDW) phase including a nesting wavevector lying within the basal tetragonal plane and a minor gapping of the Fermi surface would be compatible[28] with the data shown in the inset of Fig.3. No magnetic satellites could, so far, be detected in neutron-diffraction patterns, either because the staggered moment is too small to be resolved, or because "phase A" is of an unconventional SDW type.[29] Recently, a disorder-induced long-range antiferromagnetically ordered state was proposed to explain "phase A". In addition, comparison of NQR and μSR results leads to the assumption of a slowly fluctuating state (time constant $\tau \approx 10^{-7}$sec).[30]

A thorough study of the chemical Ce-Cu-Si phase diagram revealed the existence of up to four different

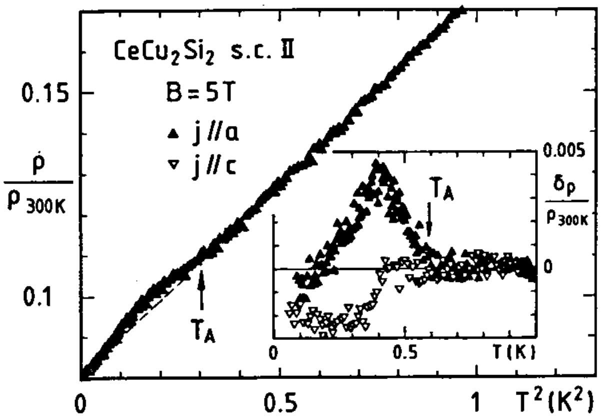

Fig. 3. Resistivity, normalized to its value at $T = 300$ K, ρ_{300K}, as a function of T^2 at $B = 5$ T for a CeCu$_2$Si$_2$ single crystal of "AS type". Current density, j, injected parallel to the tetragonal basal plane. Inset: $\delta\rho = \rho - \rho_0 - aT^2$, normalized to ρ_{300K}, as a function of T at $B = 5$ T. j injected both parallel and perpendicular to the basal plane. T_A mark onset of "phase A" and resisitivity drop, respectively.

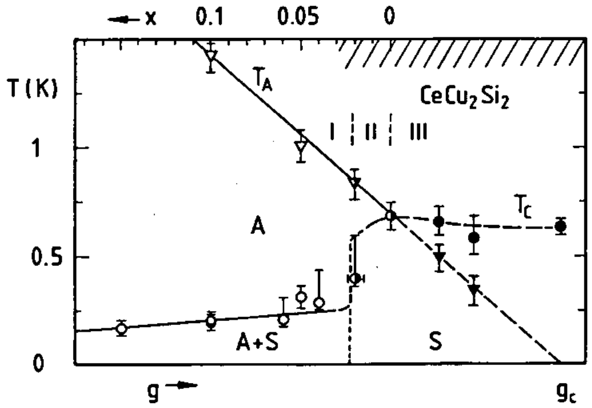

Fig. 4. Generic phase diagram of CeCu$_2$Si$_2$ (from Ref. 28, but with a linear $T_A(g)$ dependence, see text).

ground-state properties in different sectors in the homogeneity range of the primary 1:2:2 phase.[31] If the results obtained with CeCu$_2$Si$_2$ samples out of this homogeneity range are combined with those for slightly Ge-doped samples, one arrives at the magnetic/superconducting phase diagram shown in Fig. 4. Since it was found[32] that T_A depends linearly on the Ge concentration, in the dilute CeCu$_2$(Si$_{1-x}$Ge$_x$)$_2$ alloys the general coupling constant g, which should be considered a measure of the strength of the hybridization between the localized $4f$ electrons and the conduction electrons, was intentionally defined to depend linearly on the Si concentration, $1 - x$. The same linear dependence $T_A(g)$ was then *assumed* to hold for undoped CeCu$_2$Si$_2$, too. Three kinds of samples have to be distinguished: "type A" (I), "type AS" (II) and "type S" (III). "A-type" CeCu$_2$Si$_2$ samples, both Ge-doped and undoped, exhibit an "A-phase" transition and an additional superconducting one at lower T. Here, a "weak" superconducting state showing weak or no phase-transition anomalies at T_c in thermodymamic properties coexists in the whole volume with "phase A",[33] cf. Fig. 2 and Fig. 5d. "AS-type" samples exhibit first a transition into "phase A", followed by a superconducting one at T_c slightly below T_A. Pronounced phase-transition anomalies at T_c indicate a "strong" superconducting state which replaces "phase A" from most of the volume,[34,35] cf. Fig. 5c. "S-type" samples are HF superconductors. For polycrystals, application of a magnetic field apt to suppress superconductivity can result in a recovery of "phase A", cf. Figs. 5a and b. Extrapolating the corresponding "A-phase" boundary to $B = 0$, one may obtain fictitious T_A values, see Fig. 4. "S-type" single crystals (Fig. 1) are lacking "A-phase" signatures even at $B > B_{c2}$.

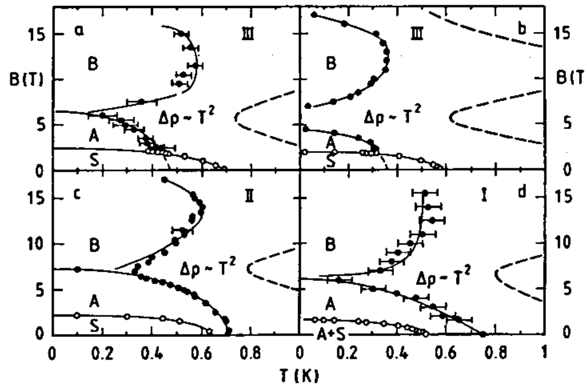

Fig. 5. $B - T$ phase diagrams for four CeCu$_2$Si$_2$ polycrystals of "S type" (a, b), "AS type" (c) and "A type" (d). Dashed lines at high temperatures mark limit of the validity range of the T^2 dependence of $\rho(T)$.

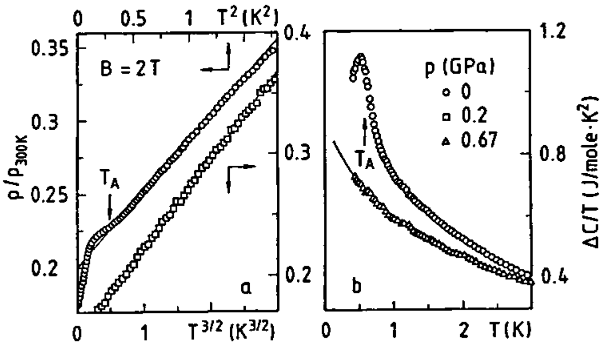

Fig. 6. Ambient and finite-pressure results for the temperature dependence of the resistivity, normalized to its value at $T = 300$ K (a), and the specific-heat coefficient, $\gamma(T) = C(T)/T$ (b) for an "A-type" CeCu$_2$Si$_2$ polycrystal. Note the different temperature scales for $p = 0$ and $p = 0.2$ GPa in (a).

As illustrated in Fig. 4, the magnetic phase boundary $T_A(g)$ extrapolates to a critical coupling constant g_c at which $T_A \to 0$. In agreement with the theoretical predictions,[14-17] for "S-type" single crystals (when measured at ambient pressure in an overcritical, but sufficiently low, magnetic field B & $B_{c2} \approx 2.5T$) $\gamma(T) =$

$\Delta C(T)/T = \gamma_0 - \alpha T^{1/2}$ and $\rho(T) = \Delta\rho(T) = \rho_0 + \beta T^{3/2}$ was observed[28] for moderate temperatures, $T > 0.4$ K. These findings are excellently corroborated by the results taken at $B = 2\ T$ for an "A-type" polycrystal and shown in Figs. 6a and b. At ambient pressure, broadened "A-phase"-transition anomalies are resolved in both $\rho(T)$ (Fig. 6a) and $\Delta C(T)/T$ (Fig. 6b). Preceding the "A-phase" transition $\Delta\rho(T) = aT^2$ is found, with a gigantic coefficient $a \approx 10\ \mu\Omega\mathrm{cmK}^{-2}$. However, this does not characterize a heavy LFL phase as the Sommerfeld coefficient γ is *not* constant but depends strongly on temperature. As is seen in Figs. 5a-d, the limiting temperature, T_L, for the validity range of the aT^2 law does follow the field dependences of both T_A and T_B, at which the high-field "phase B" forms in $CeCu_2Si_2$.[34] Application of a minute hydrostatic pressure to this "A-type" polycrystal apparently locates the sample beyond, but sufficiently close to, the critical coupling constant g_c (Fig.4). Consequently, no "A-phase" signatures but $\Delta\rho(T) = \beta T^{3/2}$ (Fig. 6a) as well as $\Delta C(T)/T = \gamma_0 - \alpha T^{1/2}$ (Fig. 6b) are observed above T = 0.4 K. We wish to note here that, so far, the experiments utilizing pressure cells had to be done in a ^{3}He cryostat with a minimum temperature of ≈ 0.4 K. An extension of these experiments to lower temperatures is in preparation to determine the true asymptotic behavior. For an "S-type" single crystal investigated at ambient pressure, striking disparities between $\Delta\rho(T)$ and $\gamma(T)$had been noticed[28] to develop below $T \approx 0.3$ K. They will be discussed in the last section.

§3. The homologue $CeNi_2Ge_2$: A "vegetable"?

In order to investigate NFL effects related to a 3D afm instability as discussed in the preceding section, it would be most favorable to find a suitable "vegetable", i.e., a HF compound lacking any superconducting and magnetic phase transitions, which is behaving as a "NFL" at $B = 0$ and low temperature. The absence of superconductivity is essential: In the presence of a finite magnetic field strong enough to suppress superconductivity, the low-lying, extended spinfluctuations causing NFL behavior should be gradually frozen out and a heavy LFL state recovered at sufficiently low T. Such a behavior can, in fact, be observed via resistivity measurements for $CeNi_2Ge_2$ (Fig.7), a homologue to $CeCu_2Si_2$: For $B =$ 0 only, the NFL-type power law, $\Delta\rho \sim T^\epsilon$, is found over a substantial T regime (0.1 K < T < 2.7 K), while the finite-field results can be well fit by $\Delta\rho(T, B = \mathrm{const})$ = $a(B)T^2$ below a crossover temperature $T_\rho(B)$ which increases with B. The slope of the T^2 fits to the low-T $\Delta\rho(T)$ data increases strongly when lowering the field (Fig. 7b). This is qualitatively expected for an afm instability to occur close to $B = 0$.[36] Therefore, it seems at first glance that $CeNi_2Ge_2$ behaves as a "vegetable" suited to study NFL properties. But, what is the origin for the latter? Can a magnetic instability be established for $CeNi_2Ge_2$, too? In order to answer to this question we have recently initiated a thorough investigation of the ternary chemical Ce-Ni-Ge phase diagram. Below, some of the most remarkable discoveries made during the course of this study on polycrystalline $CeNi_2Ge_2$ samples will be briefly addressed.

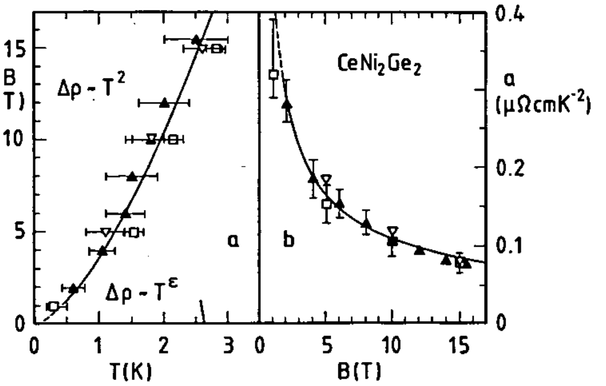

Fig. 7. a: $B - T$ phase diagram demonstrating, at given fields, the limiting temperatures of the low-T $\rho = \rho_0 + aT^2$ behavior for three $CeNi_2Ge_2$ polycrystals with $\rho_0 = 2.7\mu\Omega cm$ (Λ), 0.43 $\mu\Omega cm$ (N) and 0.34 $\mu\Omega cm$ (O). Line through the data points is a guide to the eye. Other line hitting the abscissa marks the temperature limit of the NFL regime at very low fields, see text.

b: Slope a of straight lines in the $\Delta\rho$ *vs* T^2 plots of the low-T data, taken in fixed fields, as a function of magnetic field. Symbols as in (a).

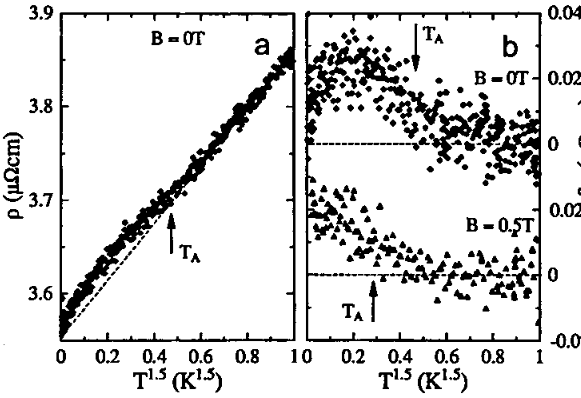

Fig. 8. Low-T resistivity as a function of temperature for a $Ce_{1.005}Ni_{2.005}Ge_{1.995}$ polycrystal as ρ *vs* $T^{1.5}$ at $B = 0$ (a) and as $\delta\rho = \rho - \rho_0 - \beta T^{1.5}$ *vs* $T^{1.5}$ at $B = 0$ and 0.5 T (b). Onset of "A-phase anomaly" is indicated by the arrows.

For slightly Ni-rich $CeNi_2Ge_2$ samples the "B-phase" transition, already known for $CeCu_2Si_2$ (Figs. 5a-d), could be established once a magnetic field B > 6 T was applied.[37] Further on, for nearly stoichiometric and slightly Ge-rich samples, an anomaly in the $\rho(T)$ measurements at $B = 0$ was discovered similar to the "A-phase" transition in the copper-silicide compound (Fig. 4). This anomaly is illustrated in Figs. 8a and b. Note, however, that in contrast to $CeCu_2Si_2$, a $T^{3/2}$ rather than T^2 power-law dependence of $\Delta\rho(T)$ was found precursive to the transition. In future work, we will attempt (i) to establish the "A-phase" transition in $CeNi_2Ge_2$ by performing thermodynamic and microscopic measurements as well as (ii) to determine that point in the chemical phase diagram at which $T_A \rightarrow 0$. Like in $CeCu_2Si_2$, the NFL effects in $CeNi_2Ge_2$ could then be traced back to the nearness of a very similar kind of magnetic instability.

In previous work,[38] "incipient" superconductivity had already been discovered for slightly Ni-rich $CeNi_2Ge_2$ polycrystals. Subsequent investigations on $CeNi_2Ge_2$ single crystals confirmed[39,40] our discovery. In one case, even a full superconducting transition below an onset-T_c of $\approx$ 200 mK could be registered resistively.[39] When scrutinizing the chemical phase diagram we have recently found several Ni-rich polycrystals with incomplete superconducting transitions (Fig. 9). As shown in Fig. 9b, the $\rho(T)$ data for the 2.5 at% Ni-excess sample even indicate a complete transition. Its onset-T_c of $\approx$ 100 mK is almost a factor of two lower than that of the single crystal discussed in Ref. 39. Consequently, these two samples must be located at somewhat different points in the chemical phase diagram. The initial slope at T_c of the upper-critical field curve, B_{c2}', was found to amount to $\approx$ 2 T/K for the sample with $T_c^{onset} \approx$ 200 mK. This strongly emphasizes that superconductivity in $CeNi_2Ge_2$ is of the HF variety.[41] Since in the clean limit, almost perfectly met in our samples (cf. Fig. 9a), $B_{c2}' \sim T_c^2$, we expect $B_{c2}' \approx$ 0.5 T/K for the $Ce_{1.005}Ni_{2.025}Ge_2$ sample with $T_c^{onset} \approx$ 100 mK, in agreement with preliminary results.[42] The data contained in Fig.9a indicate that superconductivity occurs in such $CeNi_2Ge_2$ samples that exhibit (i) a reduced c-lattice parameter or unit-cell volume (causing a stronger $4f$, conduction-electron hybridization), (ii) low intrinsic residual resistivities and, therefore,[19] (iii) exponents ε of the normal-state resistivity being slightly smaller than 1.5.

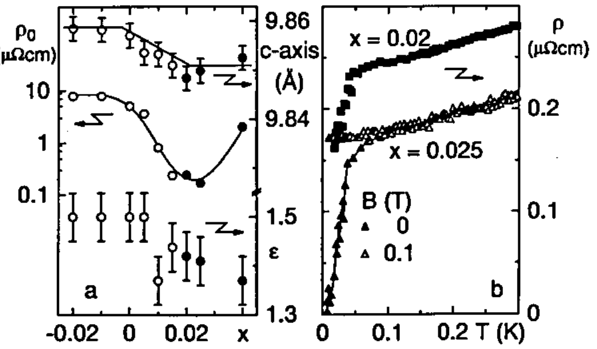

Fig. 9. a: Residual resistivity, ρ_0, c-axis lattice parameter and resistivity exponent, ε, for polycrystals of compositions $Ce_{1.005}Ni_{2+x}Ge_{2-x}$. Full circles indicate samples which exhibit a superconducting transition. b: Low-T resistivity at $B = 0$ (full symbols) and $B = 0.1$ T (open triangles) for two of the latter samples.

§4. Outlook

The two Ce-based homologues discussed in this paper should behave quite differently, owing to their different electronic configurations: While the Ni-$3d$ states in $CeNi_2Ge_2$ are crossing the Fermi level E_F, the Cu-$3d$ states in $CeCu_2Si_2$ are located way below E_F.[43] No surprize, the characteristic Kondo temperature of $CeNi_2Ge_2$ was estimated[44] to be $T_K \approx$ 30 K, i.e., about twice as high as that of $CeCu_2Si_2$. It remains to be shown whether and, if so, how this difference in the electronic structure determines the striking difference found in the sensitivity of superconductivity against ordinary potential scattering: While superconductivity with $T_c > 0.6$ K was observed[41] in $CeCu_2Si_2$ samples with $\rho_0 \leq 40$ $\mu\Omega$cm, $CeNi_2Ge_2$, samples with $\rho_0 \geq 1.5$ $\mu\Omega$cm do not superconduct.

Apart from this remarkable difference, the two compounds seem to behave surprisingly similarly in various ways. This includes the rather complex (though not identical) pressure dependences of T_c.[39,45] Another similarity is noticed in the striking disparities between low-T ($T < 0.3$ K) normal-state transport and thermodynamic properties: For both compounds, the low-T, normal-state resistivity follows over more than a decade a NFL-type of power law, $\Delta\rho \sim T^{\varepsilon}, \varepsilon \leq 1.5$. On the other hand, for several $CeCu_2Si_2$[28] and $CeNi_2Ge_2$[46] samples a pronounced deviation from the theoretically predicted and, for T & 0.4 K, indeed observed $\Delta C(T)/T = \gamma_0 - \alpha T^{1/2}$ dependence was detected at low T. This is illustrated in Fig.10 by the $B = 0$ results for a high-purity $CeNi_2Ge_2$ polycrystal ($\rho_0 \approx 0.4$ $\mu\Omega$cm). For both compounds, we have found big variations in the low-T specific-heat behavior (compare for example, Fig. 10 with Fig. 1 in Ref. 44). These variations hint at an extreme sensitivity of $C(T)/T$ against stoichiometry, more precisely against the actual occupation of the different lattice sites. One can, therefore, safely discard nuclear quadrupole effects as a potential cause for the giant upturn in $\gamma(T)$ displayed in Fig. 10. A Zeeman splitting of nuclear spin states is as unlikely: Since no "A-phase" type transition was observed in $\rho(T)$ measurements, the sample exploited in Fig. 10 has to be located in the *paramagnetic* existence range, i.e., beyond the magnetic instability. In addition, because of the low residual resistivity of this very sample, we expect that the possible concentration of disorder-induced "magnetic clusters"[47] be rather low. Only the magnetic moments of such clusters could locally couple to the nuclear spins of the available isotopes. If we try to describe the data of Fig. 10 by

$$C/T = \delta/T^3 + \gamma_0 - \alpha T^{1/2} \qquad (4.1)$$

where the last two terms denote the HF ($\gamma_0 = 0.46$ J/K^2 mol) and NFL ($\alpha = 0.12$ J/K$^{5/2}$mol) contributions, respectively, we obtain $\delta = 62.8$ μJK/mol. Taking into account *all* isotopes with finite nuclear spin contained in the sample, we estimate that an unplausibly high internal field $B_{\text{int}} = 34$ T would have to act on the nuclear spins of Ge, while the Zeeman splitting of the Ni nuclei would require even $B_{\text{int}} = 91$ T. Since an only small fraction of these isotopes could be involved, the internal-field values would have to be even much larger. Having, thus, shown that these $\gamma(T)$ upturns are *not* of nuclear origin they *must* be of electronic nature. This raises the question whether the NAFFL theory can be applied to HF compounds at all.[28] More theoretical and experimental work devoted to this question is badly needed.

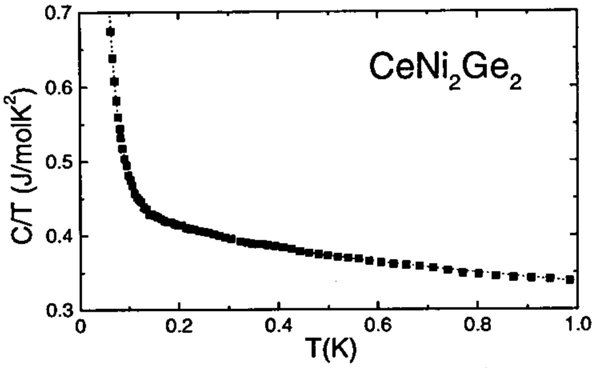

Fig. 10. Low-T specific heat, as C/T *vs* T, for a $Ce_{1.005}Ni_{2+x}Ge_{2-x}$ (x = 0) polycrystal at $B = 0$. Dotted line is a fit of eq. 4.1 to the data.

Acknowledgement

We are grateful to numerous conversations with Malte Grosche, Peter Thalmeier and Octavio Trovarelli.

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