Non-Fermi-liquid effects in stoichiometric 4f-electron metals at ambient pressure

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Abstract

We discuss strong deviations from the signatures of a heavy Landau–Fermi liquid already at $p = 0$ that are observed for normal (n)-state $CeCu_2Si_2$, for $CeNi_2Ge_2$ and $YbRh_2Si_2$. No quantum critical point could yet be established in the case of $CeNi_2Ge_2$. For this compound, preliminary investigations of the chemical phase diagram have revealed certain similarities to $CeCu_2Si_2$. The non-Fermi-liquid (NFL) effects observed for $YbRh_2Si_2$ suggest a quasi-2D system of magnetic fluctuations. We also discuss briefly disparities between resistivity and specific-heat results in all three compounds as well as the relationship between a NFL n-state and the occurrence of heavy-fermion superconductivity.

Keywords: Heavy fermion; Non-Fermi-liquid effects; Stoichiometric compounds

1. Introduction

Quantum critical phenomena in lanthanide- and actinide-based heavy-fermion (HF) metals have recently attracted considerable interest. In such materials, HF phenomena are usually occuring in the neighborhood of some magnetically ordered state. The actual ground state of a given HF system is predominantly determined by the strength ($\mathscr{G}$) of the hybridization between the localized 4f/5f states and the itinerant conduction-electron states. $\mathscr{G}$ can be controlled experimentally by either applying hydrostatic pressure or suitable alloying. At the quantum critical point (QCP), where the magnetic ordering temperature vanishes, the abundance of strong low-lying and extended fluctuactions of the local magnetic order parameter, mediating the interactions between the heavy quasiparticles, causes strongly energy-dependent quasiparticle masses and quasiparticle–quasiparticle scattering cross sections. These lead to unusual temperature dependences in the measurable quantities $\gamma(T) = \Delta C(T)/T$ and $a(T) = \Delta\rho(T)/T^2$,[1] the 4f/5f-derived specific-heat and resistivity coefficients, respectively. In most HF metals, quantum critical behavior appears to be related to the vicinity of antiferromagnetic (AF) order. In this case the unusual T-dependences, often labeled "non-Fermi-liquid" (NFL) effects, are considered [1,2] properties of a generalized Fermi liquid, i.e. the "nearly antiferromagnetic Fermi liquid". Within this theory, the asymptotic ($T \rightarrow 0$) behavior of the specific-heat coefficient for a 3D system of spin fluctuations follows $\gamma(T) = \gamma_0 - \alpha T^{1/2}$. The important point here is that at the QCP, γ_0 remains finite, though critically enhanced compared to the heavy Landau–Fermi liquid (LFL) phase [1,2]. As far as the contribution of the quasiparticle–quasiparticle scattering to the resistivity is concerned, this should obey $\Delta\rho(T) = \beta T^{3/2}$ if the quasiparticle elastic mean-free path is not too long. Once the amount of potential scattering is reduced, $\Delta\rho(T) = \beta' T^{\varepsilon}$

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[1] Here $\Delta C(T) = C(T) - C_0(T)$ and $\Delta\rho(T) = \rho(T) - \rho_0$. $C(T)$ as well as $\rho(T)$ are determined for the HF metal and $C_0(T)$ for a suitable non-f-ion reference compound. ρ_0 denotes the residual resistivity of the HF metal.

with $1 < \varepsilon < 1.5$ is predicted [3] in a wide temperature range, although in the ideally clean case $\varepsilon = 2$ must be obtained at sufficiently low temperature [4].

Most of the investigations of the physics near an AF-QCP were done with Ce-based systems. Two strategies have so far been followed to approach the magnetic instability ($T_N \to 0$): (i) Starting from a heavy LFL and expanding the lattice by alloying with sizeable dopands, one is able to induce a long-range AF ordered state. $CeCu_{6-x}Au_x$ [5] and $Ce_{1-x}La_xRu_2Si_2$ [6] are the exemplary systems. (ii) Starting from an AF-ordered Kondo-lattice system and applying a sufficiently high hydrostatic pressure one can eventually reach a nonmagnetic state. Model systems are $CePd_2Si_2$ and $CeIn_3$ with critical pressures $p_c(T_N \to 0)$ near 2.7 GPa in either case [7]. Resistivity data only are presently available for these compounds.

In order to keep the degree of disorder as low as possible [8] and to perform transport as well as thermodynamic experiments we will address *undoped* HF metals which do show NFL effects already at *ambient* pressure. Only a few of such materials have been known to date, e.g. normal (n)-state UBe_{13} [9], $CeCu_2Si_2$ [10] and $CeNi_2Ge_2$ [11]. As another Ce-based compound we mention $CeRu_4Sb_{12}$ [12]. In the following, we will review some of the key properties of $CeCu_2Si_2$, discuss recent results on $CeNi_2Ge_2$ and introduce as a new exciting member of this class of materials $YbRh_2Si_2$ [13]. Finally, we also wish to briefly address two topics of more general interest: (i) disparities between resistivity and specific-heat data and (ii) the relationship between NFL n-state properties and the occurrence of HF superconductivity.

2. Materials: $CeCu_2Si_2$, $CeNi_2Ge_2$ and $YbRh_2Si_2$

Among the Ce- and U-based stoichiometric HF compounds showing NFL behavior at ambient pressure, a QCP could so far be established for the HF superconductor $CeCu_2Si_2$ [14] only: The phase transition temperature T_A was found to vanish [12] inside the so-called "S sector" of the homogeneity range of $CeCu_2Si_2$ in the chemical ternary Ce–Cu–Si phase diagram [15]. T_A signals the transition from the paramagnetic state into "phase A", a (perhaps unconventional [16]) spin-density-wave state. In Fig. 1a the B–T phase diagram is shown for an "S-type" single crystal which has to be located slightly beyond the QCP: There are no signatures for the "A-phase" transition, and the n-state resistivity follows $\Delta\rho \sim T^{3/2}$ in a wide temperature range, $0.005\ \mathrm{K} < T < 1.8\ \mathrm{K}$. HF superconductivity forms below $T_c \simeq 0.6$ K and at moderate magnetic fields ($\leqslant 2$ T). For fields $B > 8$ T a, so far, unidentified "phase B" [17] exists at temperatures below 0.2 K. For the "AS-type" single crystal of Fig. 1b, the existence range of

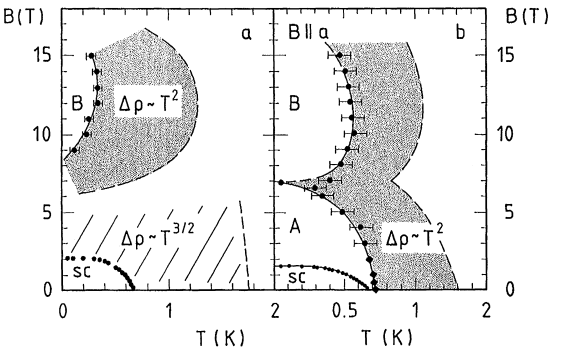

Fig. 1. B–T phase diagrams, based upon resistivity results, for an "S-type" $CeCu_2Si_2$ single crystal (a) and an "AS-type" single crystal. (b) Dashed lines indicate limits of the validity ranges for the $T^{3/2}$ and T^2 laws, respectively.

"phase B" is considerably larger. HF superconductivity sets in at a somewhat higher T_c, but shows a reduced upper critical field, $B_{c2}(T)$. The latter reflects a strong competition between superconductivity and the embedding "phase A" which becomes expelled from the (major fraction of the) sample volume below B_{c2} [17,18]. The transitions from the paramagnetic state into both "phase A" and "phase B" are preceded by a temperature interval in which the resistivity follows $\Delta\rho = aT^2$, with a gigantic coefficient $a \simeq 10\ \mu\Omega\ \mathrm{cm\ K^{-2}}$. However, this does *not* signal a heavy LFL state as, in the same T interval, the Sommerfeld coefficient $\gamma(T)$ is found to depend strongly on temperature [10].

For the $CeNi_2Ge_2$ compound, the chemical phase diagram is currently under investigation by systematically varying the stoichiometry of polycrystalline samples [19]. With two almost stoichiometric samples we could establish "phase B" (at magnetic fields in excess of 6 T) for this compound as well (Fig. 2). For several slightly Ni-rich samples, resistivity measurements done at $B = 0$ have revealed an anomaly that strongly resembles the "A-phase" transition in $CeCu_2Si_2$ [10] (cf. Fig. 3). Application of $B = 0.5$ T shifts the transition temperature to somewhat lower values. Unlike $CeCu_2Si_2$, however, $\Delta\rho(T)$ follows a $T^{3/2}$ rather than a T^2 law for $T > T_A$. The assignment of this anomaly to the transition into "phase A" has yet to be confirmed by thermodynamic and microscopic experiments. For two samples with larger Ni excess, incipient superconductivity had been previously observed [11]. New results for $Ce_{1.005}Ni_{2+x}Ge_{2-x}$ samples with $x = 0.02$ and 0.025 are illustrated in Figs. 4a and b. Their onset T_c (≈ 100 mK) is about a factor of two lower than that of a single crystal discussed in Ref. [20] which must, therefore, be located at a somewhat different place in the chemical phase diagram. The $x = 0.025$ sample exhibits a full superconducting transition with a mid-point $T_c \simeq 50$ mK and an initial

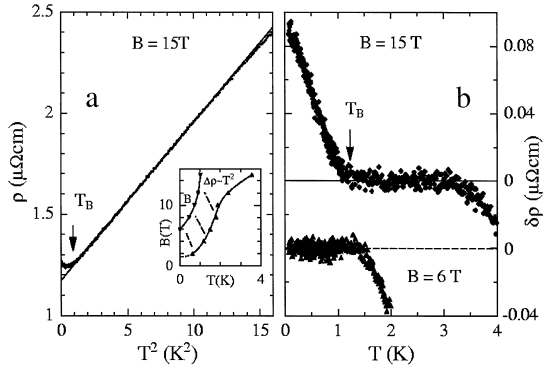

Fig. 2. Temperature dependence of the low-T resistivity as ρ versus T^2 (a) and as $\delta\rho = \rho - (\rho_0 + a(B)T^2)$ versus T (b) at $B = 15$ T and $B = 6$ T for a $Ce_{1.005}Ni_{2.025}Ge_{1.975}$ polycrystal. At T_B, the "B-phase" transition sets in. Inset: B–T diagram indicating existence range of "phase B" and validity range of the region in which $\rho = \rho_0 + a(B)T^2$ holds.

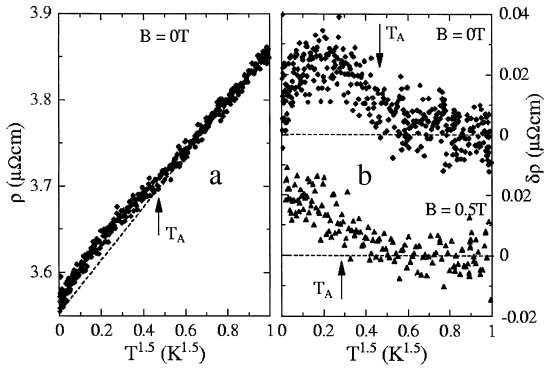

Fig. 3. Low-T resistivity as a function of temperature for a $Ce_{1.005}Ni_{2.005}Ge_{1.995}$ polycrystal as ρ versus $T^{1.5}$ at $B = 0$ (a) and as $\delta\rho = \rho - (\rho_0 + \beta T^{1.5})$ versus $T^{1.5}$ at $B = 0$ and 0.5 T. (b) Onset of "phase A" is indicated by the arrows.

slope of $B_{c2}(T)$ suggesting HF superconductivity. In Fig. 4a we show that the superconducting samples are characterized by somewhat reduced c-lattice parameters, low (intrinsic) residual-resistivity values and resistivity exponents ε of the n-state power-law dependence, $\Delta\rho \sim T^{\varepsilon}$, being lower than 1.5.[2] The investigations ascribed above are aimed at pinning down the QCP in $CeNi_2Ge_2$. Here, the main question is whether the

[2] According to X-ray diffraction measurements, the $x = 0.04$ sample contains a considerable amount of secondary phases which contribute to the measured value of both the c-lattice constant and the residual resistivity ρ_0, but *not* to that of the resistivity exponent ε: the T-dependence of the measured $\rho(T)$ appears to be predominated by the primary "122" phase.

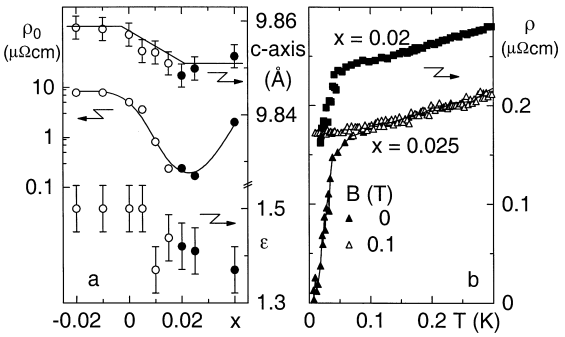

Fig. 4. (a): Residual resistivity, ρ_0, c-axis lattice parameter and resistivity exponent, ε, for polycrystals of compositions $Ce_{1.005}Ni_{2+x}Ge_{2-x}$. Full circles indicate samples which exhibit a superconducting transition. (b) Low-T resistivity at $B = 0$ (full symbols) and at $B = 0.1$ T (open triangles) for two of the latter samples.

"A-phase" transition can indeed be established and followed to that point in the chemical phase diagram at which $T_A \to 0$.

For UBe_{13} it appears even more difficult to find a QCP because, to our knowledge, no ordering phenomenon exists in this compound at ambient and low pressure, except for HF superconductivity [21]. One may speculate that the recently discovered weak low-T anomalies in thermodynamic properties of the superconducting state [22], tentatively ascribed to some AF correlations, are related to the occurrence of NFL behavior in the n-state.

It is generally quite a difficult task to identify a QCP for nonmagnetic Ce- or U-based compounds: If the QCP exists beyond the homogeneity range, one has to apply "negative pressure", i.e. to expand the lattice. This, however, is not possible without introducing some disorder as pointed out before. On the other hand, if one wants to avoid disorder, it is wise to search for a suitable nonmagnetic Yb-based compound showing NFL effects already at ambient pressure: Owing to the hole/electron analogy of the Yb^{3+}-$4f^{13}$ and Ce^{3+}-$4f^{1}$ configurations one can then hope to reach the magnetic instability by application of moderate hydrostatic pressure, i.e. without introducing additional disorder.

As was shown very recently by Trovarelli et al. [13] $YbRh_2Si_2$, isostructural to the two Ce compounds discussed above, meets all these requirements. In Fig. 5 we present the $\rho(T)$ behavior of a $YbRh_2Si_2$ single crystal. In a power-law representation $\Delta\rho \sim T^{\varepsilon}$, the weakly T-dependent exponent ε is found to remain within (1 ± 0.05) below 10 K and to saturate at $\varepsilon = 1$ as T decreases down to 10 mK, without showing any evidence of a magnetic or superconducting phase transition at $p = 0$ (inset a of Fig. 5). The specific-heat coefficient $\gamma(T) = \Delta C(T)/T$ diverges logarithmically in the range

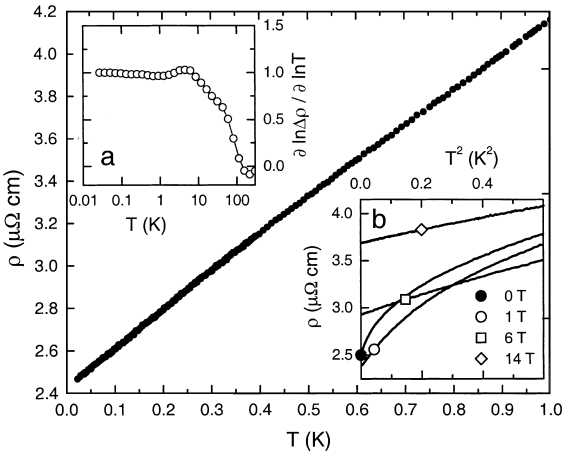

Fig. 5. Low-T resistivity of an $YbRh_2Si_2$ single crystal at $p = 0$ measured along the a-axis as a function of temperature. Inset a: T-dependence of the resistivity exponent ε, defined as the logarithmic derivative of $\Delta\rho(T) = \rho(T) - \rho_0$. Inset b: ρ versus T^2 for $B \leqslant 14$ T, applied along the c-axis. Positions of the symbols indicate the crossover temperatures below which $\Delta\rho = a(B)T^2$ holds.

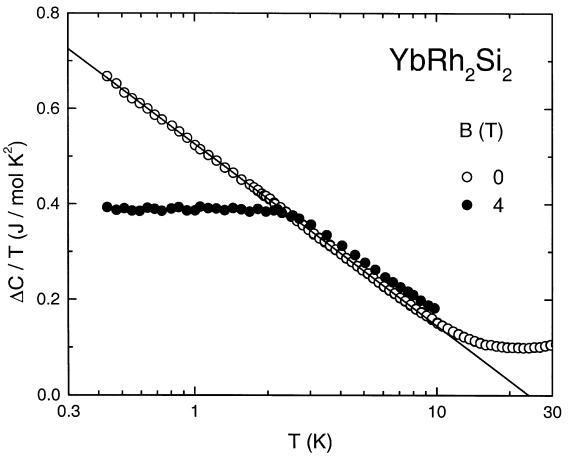

Fig. 6. Yb increment of single crystalline $YbRh_2Si_2$ to the specific heat as $\Delta C/T$ versus T (on a logarithmic scale) at $B = 0$ and 4 T, applied along the a axis. $\Delta C = C_{YbRh_2Si_2} - C_{LuRh_2Si_2}$. The solid line, representing $\Delta C/T = \gamma'_0 \ln(T_0/T)$ is a guide to the eye. $\gamma'_0 = 0.17$ J/K^2 mole and $T_0 = 24$ K.

0.3 K $< T <$ 10 K (see Fig. 6). In addition, a field-induced LFL phase is observed, both in the resistivity and specific-heat results, below a crossover temperature which increases with the applied B field (inset b of Fig. 5). The critical pressure at which long-range (presumably) AF order develops, was found [13] to be as low as $p_c \approx 0.5$ GPa. This is by far the lowest value known for any Yb compound to date. Though the NFL effects discovered for $YbRh_2Si_2$ do *not* agree with the theoretical predictions [1,2] for the asymptotic ($T \to 0$) behavior near an AF-QCP (in 3D), they are very similar to what was found earlier for $CeCu_{6-x}Au_x$ [5] and more recently for $U_{1-x}Y_xPd_2Al_3$ [23]. For the former alloy, a system of quasi-2D spinfluctuations coupled to a 3D carrier system has been proposed [24] to account for the experimental findings. Such a scenario may also yield an adequate description of the data obtained for $YbRh_2Si_2$ [13]. This compound will certainly serve as a model system to study NFL phenomena, by a large number of experimental techniques, both in the absence of doping and at ambient pressure. Hopefully, such studies will also allow one to clarify the role disorder plays in the NFL behavior of those alloys mentioned before.

3. Final remarks

Preliminary specific-heat measurements on $YbRh_2Si_2$ at low temperatures ($T \geqslant 50$ mK) reveal a pronounced upturn in $\Delta C(T)/T$ below $T \approx 300$ mK [25], i.e. a strong deviation from the quantum critical behavior at higher T. Similar low-T features have been observed earlier, e.g. for n-state UBe_{13} [9], n-state $CeCu_2Si_2$ [10] and $CeRu_4Sb_{12}$ [12]. Recent studies on $CeNi_2Ge_2$ [25] show that for this compound, such upturns also occur and depend sensitively on stoichiometry. Interestingly enough, the low-T n-state resistivity measured at sufficiently low magnetic field *does* show quantum critical behavior, i.e. obeys a power-law dependence, $\Delta\rho \sim T^{\varepsilon}$ with $1 \leqslant \varepsilon \leqslant 1.6$, in all those compounds mentioned above. One might tend to relate these striking disparities between $\rho(T)$ and $\Delta C(T)/T$ to some disorder effects [8]. However, such a correlation could, so far, not be established.

We conclude the paper by noting that there is *no* clear correlation between the existence of NFL effects in the n-state and the occurrence of HF superconductivity: For example, in U-based compounds like UPt_3 or UPd_2Al_3, superconductivity appears to form out of a medium-heavy LFL state that coexists with long-range AF order well below T_N. Also, p-induced HF superconductivity both in $CeCu_2Ge_2$ [26] and $CeRh_2Si_2$ [27] seems to develop out of a LFL state in the very vicinity of the critical pressure, i.e. $p_c \simeq 9.4$ GPa [26] and 0.9 GPa [27], respectively. This means, NFL effects in the n-state are *not necessary* for the formation of a HF superconducting state. They are *not sufficient* either, since $YbRh_2Si_2$ is not a superconductor, at least above $T \simeq 10$ mK and in single crystals with $\rho_0 \approx 2$ μΩcm.

Finally, even those HF superconductors that *do* show NFL effects in their n-states behave phenomenologically surprisingly different: For example, while UBe_{13} exhibits a ratio of the quasiparticle mean free path to the superconducting coherence length, ℓ/ξ_0, considerably smaller

than one [28], $\ell/\xi_0 < 1$ or $\ell/\xi_0 \geqslant 1$ for $CeCu_2Si_2$ [28], but $\ell/\xi_0 \gg 1$ for $CeNi_2Ge_2$. In addition, while T_c of UBe_{13} appears to be reduced linearly by the application of pressure [29], $T_c(p)$ depends in a very complex way on p, for both $CeCu_2Si_2$ [30] and $CeNi_2Ge_2$ [20]. This non-universal behavior of the HF superconductors had been discussed [31] in the context of more than one microscopic mechanism involved in the formation of massive Cooper pairs.

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