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# Breakup of Heavy Fermions on the Brink of “Phase A” in $\text{CeCu}_2\text{Si}_2$

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We report resistivity,  $\rho(T)$ , and specific-heat,  $C(T)$ , results on near stoichiometric  $\text{CeCu}_2\text{Si}_2$  samples, in the vicinity of a quantum critical point (QCP). The latter is defined by  $T_A \rightarrow 0$ , where  $T_A \approx 0.8$  K marks the transition into a spin-density-wave-type “phase A” which competes with heavy-fermion superconductivity below  $T_c \approx 0.65$  K. Upon approaching the QCP,  $\rho(T)$  and  $C(T)$  behave very disparately, suggesting a breakup of the heavy quasiparticles. Very surprising observations are being made for samples with  $T_A > 0$  also. [S0031-9007(98)06803-3]

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The concept of the “nearly antiferromagnetic Fermi liquid” (NAFFL) has been intensively discussed in connection with the exotic normal (N) and superconducting (SC) properties of the quasi-two-dimensional (2D) high- $T_c$  cuprates [1,2]. More recently, the three-dimensional (3D) Ce-based heavy-fermion (HF) superconductors were also treated in the same frame [3]. Here, it is assumed that, in the vicinity of an AF quantum critical point (QCP), low-lying and extended spin fluctuations with wave vectors  $\mathbf{q} \approx \mathbf{Q}$ , the AF ordering wave vector, give rise to strongly  $T$ -dependent quasiparticle masses and quasiparticle-quasiparticle cross sections. These should manifest themselves in coefficients  $\gamma = C/T$  and  $a = (\rho - \rho_0)/T^2$  in the specific heat and electrical resistivity ( $\rho_0$ : residual resistivity) which are not constant as in a Landau FL, but obey the following asymptotic  $T$  dependences (in 3D) [4,5]:

$$\gamma(T) = \gamma_0 - \alpha T^{1/2} \quad (1)$$

and [4–6]

$$a(T) = \beta T^{-1/2}, \quad (2a)$$

corresponding to

$$\Delta\rho(T) = \rho(T) - \rho_0 = \beta T^{3/2}. \quad (2b)$$

Since the singular scattering expressed by Eq. (2a) is associated with the AF wave vector  $\mathbf{Q}$ , i.e., occurs only along certain “hot lines” on the Fermi surface, all other quasiparticle-quasiparticle scattering events ought to give rise to the ordinary FL term  $\Delta\rho = aT^2$  ( $a = \text{const}$ ) which, consequently, must short-circuit the anomalous  $\beta T^{3/2}$  term at sufficiently low temperature [7]. This holds even in the presence of strong impurity scattering that reduces the anisotropy of the quasiparticle lifetime and, this way, the crossover temperature between the two regimes [7].

In this Letter, we address the  $n$ -state resistivity and specific heat of the archetypical HF superconductor  $\text{CeCu}_2\text{Si}_2$

[8]. The salient results of this study are (i) a QCP exists at the disappearance of “phase A” [Fig. 1(a)], the latter being accompanied by Fermi-surface nesting (in the tetragonal plane) as expected, e.g., at a spin-density-wave

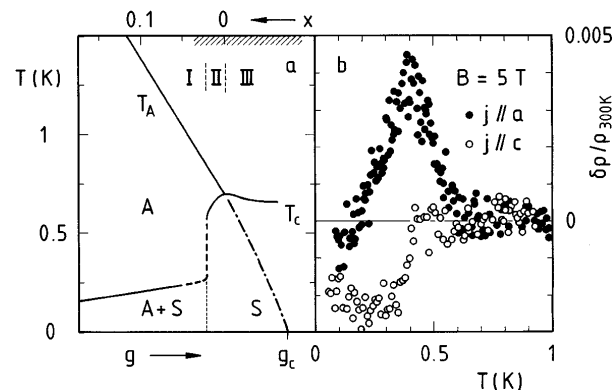


FIG. 1. (a) Schematic phase diagram for  $\text{CeCu}_2\text{Si}_2$  at zero field, indicating existence ranges for phase A, superconductivity (S), and coexistence range (A + S). For samples labeled type I, II, and III:  $T_A > T_c$ ,  $T_A \approx T_c$ , and  $T_A < T_c$ , respectively (see text). The form of the phase boundaries between S and A + S (dotted line) and between S and A (dashed line) is tentative since considerable stress dependence and homogeneity problems prevent a precise determination. We expect it to be rather steep when determined with monodomain single crystals. On the abscissa an effective coupling constant  $g$  is used which is a complicated function of the composition in homogeneous  $\text{CeCu}_2\text{Si}_2$  samples [9] (hatched regime) or is proportional to the Ge content  $x$  in  $\text{CeCu}_2(\text{Si}_{1-x}\text{Ge}_x)_2$  [10].  $g = g_c$  marks  $T_A \rightarrow 0$ . The phase boundaries  $T_A(g)$  [10,11] and  $T_c(g)$  [10] are determined from  $B = 0$  measurements (solid lines) or extrapolated to  $B = 0$  from data taken at  $B > B_{c2}$  [12] (dash-dotted line). (b): Normalized resistivity of a type II  $\text{CeCu}_2\text{Si}_2$  single crystal as  $\delta\rho/\rho_{300\text{K}}$  vs  $T$ , with  $\delta\rho = \rho - \rho_0 - aT^2$ , measured along the respective  $a$  and  $c$  axes at  $B = 5$  T (applied perpendicular to the current) [13]. The data indicate the transition into the SDW-type phase A, with a nesting wave vector lying within the basal plane.

(SDW) transition [Fig. 1(b)]. (ii)  $\text{CeCu}_2\text{Si}_2$  loses the signatures of a NAFFL upon approaching this QCP sufficiently closely. (iii) A strange behavior is also found upon approaching the  $A$ -phase transition at  $T_A > 0$ .

We discuss below the properties of two  $\text{CeCu}_2\text{Si}_2$  samples which, according to x-ray diffractometry, were found to be single phase with the proper  $\text{ThCr}_2\text{Si}_2$  structure. The  $\text{Ce}_{0.99}\text{Cu}_{2.02}\text{Si}_2$  polycrystal was prepared in an argon-arc furnace and subsequently annealed at  $700^\circ\text{C}$  for 24 h and  $1000^\circ\text{C}$  for 120 h. The single crystal was already studied in [14]. Measurements of the resistivity were done using a four-terminal, low-frequency (113 Hz) lock-in technique. The specific heat at ambient pressure was measured utilizing a thermal-relaxation technique [15] while, for measurements of the heat capacities of the pressure cell (with and without the sample), a compensated heat-pulse method [16] was used. The Ce increment to the specific heat was determined by subtracting from the measured specific heat that of  $\text{LaCu}_2\text{Si}_2$ . Hydrostatic pressure was applied by utilizing a CuBe piston-cylinder cell with a 1:1 mixture of isoamyl alcohol and  $n$ -pentane as pressure-transmitting medium. The pressure,  $p$ , was determined inductively from the  $p$ -induced shift of the SC transition of a small piece of Pb mounted together with the sample. For the investigation of the single crystal, a  $^3\text{He}$ - $^4\text{He}$  dilution refrigerator ( $T \geq 20$  mK) and a superconducting solenoid ( $B < 17$  T) were used. The experiments on the polycrystal utilizing the pressure cells were done in a  $^3\text{He}$  cryostat ( $T > 0.4$  K).

The nature of phase  $A$  is still unknown. Neutron diffractometry has so far failed to resolve magnetic Bragg reflections. Different assignments spanning the whole range from spin-glass [17] to dynamical [18] and unconventional SDW [19] order have been made. Partial Ge substitution for Si was found to stabilize phase  $A$  and to support strong evidence for an AF transition at  $T_A$  [11]. As shown in Fig. 1(a),  $\text{CeCu}_2(\text{Si}_{1-x}\text{Ge}_x)_2$  samples

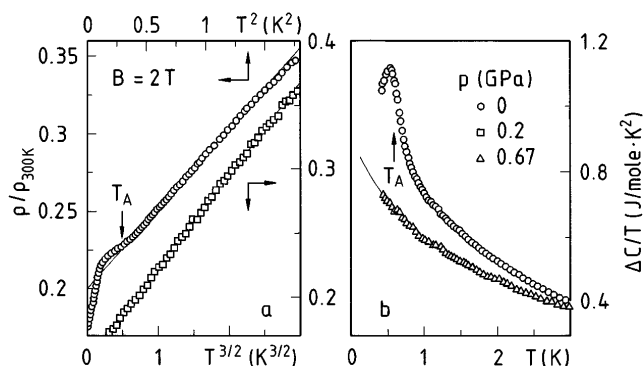


FIG. 2. Normalized resistivity as  $\rho/\rho_{300\text{K}}$  vs  $T^2$  (upper scale) and  $\rho/\rho_{300\text{K}}$  vs  $T^{3/2}$  (lower scale) (a) as well as Ce increment to the specific heat as  $\gamma = \Delta C/T$  vs  $T$  (b), for  $B = 2$  T and at  $p = 0$  as well as two overcritical pressures for a type I  $\text{CeCu}_2\text{Si}_2$  polycrystal. The low- $T$  drop in  $\rho(T)$  at  $p = 0$  is due to the onset of a SC transition. Solid lines display  $T^2$  and  $T^{3/2}$  dependences of  $\rho(T)$  (a) and a fit of the “SCR theory” [5] to the  $\gamma(T)$  data, implying  $y_0 = 0$ ,  $y_1 = 4$ , and  $T_0 = 13$  K.

with  $0.02 < x \leq 0.15$  and undoped “type I” samples with compositions out of the narrow homogeneity range exhibit an  $A$ -phase transition between  $T_A \approx 0.8$  and  $1.75$  K, followed by a bulk HF-SC transition between  $T_C \approx 0.3$  and  $0.15$  K, respectively. Phase  $A$  and (thermodynamically weak) superconductivity coexist on a microscopic scale [20]. In “type II”  $\text{CeCu}_2\text{Si}_2$  samples of near stoichiometric composition and with  $T_A \geq T_C$ , (thermodynamically strong) HF superconductivity expels phase  $A$  [21]. The phase boundary separating sectors I and II is expected to be rather steep [cf. Fig. 1(a)]. Resistivity measurements of “type III”  $\text{CeCu}_2\text{Si}_2$  polycrystals (intentionally prepared with a slight excess of Ce or Cu [9]) reveal clear  $A$ -phase signatures with reduced  $T_A$  when superconductivity is suppressed by a magnetic field [10,12], while  $B = 0$   $\mu\text{SR}$  experiments indicate the presence of a “magnetic minority domain” whose volume fraction shrinks upon cooling to well below  $T_C$  [22]. The  $B$ - $T$  phase diagrams collected for such polycrystals with varying  $T_A$ ’s clearly indicate a continuous evolution  $T_A \rightarrow 0$  [10,12]. Type III single crystals do not show any  $A$ -phase signatures. For all  $\text{CeCu}_2\text{Si}_2$  samples studied in resistivity so far, an additional “phase B” [21], phenomenologically related to but unidentified as phase  $A$ , was observed to form at fields  $B > 6$  T.

Figure 1(a) suggests the existence of a critical coupling constant (measuring the strength of the  $4f$  hybridization with conduction electrons),  $g_C$ , at which  $T_A \rightarrow 0$ . In order to investigate whether this defines a QCP and, if so, how heavy fermions behave in its vicinity, we discuss in the following a pressure-induced  $A \rightarrow S$  transition on a type I  $\text{CeCu}_2\text{Si}_2$  polycrystal (Fig. 2) as well as specific-heat (Fig. 3) and resistivity (Fig. 4) results for a type III single crystal.

Similar to what was found for a type II single crystal [cf. Fig. 1(b)], the  $A$ -phase transition in the polycrystal chosen for the present study, when measured at  $p = 0$  and  $B = 2$  T, manifests itself in broadened anomalies

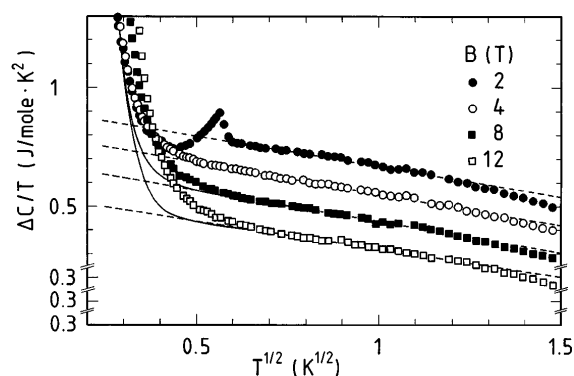


FIG. 3.  $\gamma = \Delta C/T$  vs  $T^{1/2}$  at varying fields for a type III  $\text{CeCu}_2\text{Si}_2$  single crystal. Dashed lines indicate  $(-T^{1/2})$  dependence of  $\gamma(T) - \gamma_0$ . Solid lines display  $\Delta C(T)/T$  data after subtraction of nuclear hyperfine contributions due to the applied  $B$  fields. For  $B = 2$  T, the SC transition anomaly at  $T_C \approx 0.3$  K is seen.

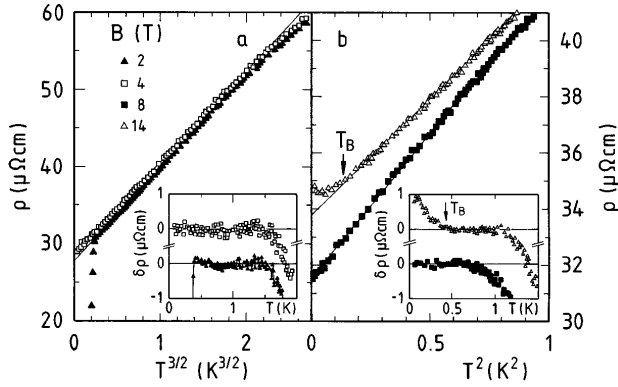


FIG. 4. Resistivity for the same crystal as in Fig. 3 as  $\rho$  vs  $T^{3/2}$  for  $B = 2$  and  $4$  T (a) as well as  $\rho$  vs  $T^2$  for  $B = 8$  and  $14$  T (b). Insets show  $\delta\rho$  vs  $T$  with  $\delta\rho = \rho - \rho_0 - \beta T^{3/2}$  (a) and  $\delta\rho = \rho - \rho_0 - aT^2$  (b), respectively. Arrows in (b) indicate  $B$ -phase transition for  $B = 14$  T.

in the  $T$  dependences of  $\rho(T)$  and  $\gamma(T) = \Delta C(T)/T$  at  $T_A \approx 0.75$  K [Figs. 2(a) and 2(b)]. At pressures exceeding a critical value  $p_C \approx 0.1$  GPa, phase  $A$  is completely suppressed and replaced, at  $B = 0$ , by a strong HF-SC state below  $T_C \approx 0.65$  K (not shown).

The ambient-pressure results of Fig. 2 display a strikingly dual behavior at  $T > T_A$ :  $\rho = \rho_0 + aT^2$  with a giant coefficient  $a \approx 10 \mu\Omega \text{ cm K}^{-2}$  suggests a heavy Landau FL state [Fig. 2(a)], with which notion the strongly  $T$ -dependent Sommerfeld coefficient  $\gamma(T)$  is, however, incompatible [Fig. 2(b)]. On the other hand, the results taken at finite pressure,  $p > p_C$ ,  $0.4 \text{ K} < T < 2 \text{ K}$  and  $B = 2 \text{ T}$  fulfill the theoretical predictions for  $T_N \rightarrow 0$  [4–6]:  $\rho = \rho_0 + \beta T^{3/2}$  and  $\gamma = \gamma_0 - \alpha T^{1/2}$ . An extension of these experiments to lower temperatures is in preparation in order to determine the true asymptotic behavior.

Turning now to the type III crystal which is lacking any  $A$ -phase signature, we expect its properties at  $p = 0$  to be similar to the properties of the polycrystal at  $p > p_C$  (cf. Fig. 2). We focus first on the results taken at sufficiently low fields ( $B < 6 \text{ T}$ ) and at intermediate temperatures ( $T > 0.2 \text{ K}$ ) [cf. Figs. 3 and 4(a)]. In the normal state, both  $\gamma(T)$  and  $\Delta\rho(T)$  obey Eqs. (1) and (2b) for  $T < 1.7 \text{ K}$ . The slopes in the respective plots  $\gamma$  vs  $T^{1/2}$  and  $\Delta\rho$  vs  $T^{3/2}$  are almost independent of the field, whereas the  $T = 0$  values move slightly up ( $\rho_0$ ) or down ( $\gamma_0$ ) if the field is increased. The data at  $T > 0.2 \text{ K}$  and  $B < 6 \text{ T}$  for the single crystal measured at ambient pressure suggest the existence of an AF-QCP. This is corroborated by the polycrystal data taken at pressures  $p > p_C$ ,  $B = 2 \text{ T}$ , and  $T > 0.4 \text{ K}$ , the minimum temperature accessible in the  $^3\text{He}$  cryostat. It is straightforward to relate this QCP to the disappearance of phase  $A$  at a critical coupling constant  $g_C$  [Fig. 1(a)]. However, when approaching the QCP by cooling the single crystal to below  $0.2 \text{ K}$ , the  $n$ -state specific-heat coefficient  $\gamma(T)$  does *not* follow the  $T^{1/2}$  dependence

anymore (Fig. 3). It rather shows a steep upturn at the low- $T$  end. The latter cannot be ascribed to the Zeeman splitting of the nuclear  $^{63}\text{Cu}$ ,  $^{65}\text{Cu}$ , or  $^{29}\text{Si}$  spin states through the external  $B$  field (cf. the solid lines in Fig. 3). 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. The origin of this internal field is, however, not clear. In the same low-temperature range where  $\gamma(T)$  deviates from Eq. (1), the  $n$ -state resistivity at sufficiently low field is still obeying Eq. (2b):  $\Delta\rho \sim T^{3/2}$  [cf. Fig. 4(a)]. Most remarkably, no crossover to a Landau-FL-type  $T^2$  behavior, which must necessarily occur in a NAFFL [7], can be resolved down to  $20 \text{ mK}$ .

Finally, we wish to address the surprising effect a magnetic field  $B > 6 \text{ T}$  has on the low- $T$  properties of our  $\text{CeCu}_2\text{Si}_2$  single crystal. While the gross  $T$  dependence of  $\gamma(T)$  remains unaffected (Fig. 3), the  $\Delta\rho(T)$  dependence becomes qualitatively changed into  $\rho = \rho_0 + aT^2$  [Fig. 4(b)]. The residual resistivity keeps rising, i.e., by  $\approx 10\%$  when  $B$  is increased from  $8$  to  $14 \text{ T}$ , whereas the giant coefficient  $a$  decreases by almost the same fraction. In addition, the  $B = 14 \text{ T}$  data display the broadened transition into phase  $B$  which is not visible in  $\gamma(T)$  measured at, e.g.,  $B = 12 \text{ T}$  (Fig. 3). Our resistivity and specific-heat results on the type III single crystal at  $B > 6 \text{ T}$  are phenomenologically related to the  $p = 0$  data taken on the type I polycrystal in an overcritical field  $B = 2T$  [cf. Figs. 2(a) and 2(b)]: While  $\Delta\rho(T)$  suggest that phase  $B$  as well as phase  $A$  form out of a heavy Landau-FL phase, such an interpretation becomes obsolete in view of the pronounced  $T$  dependences of  $\gamma(T)$  precursive to both the  $B$ - and  $A$ -phase transitions. The strikingly different  $T$  dependences of the resistivity for the  $\text{CeCu}_2\text{Si}_2$  single crystal below and above  $B \approx 6 \text{ T}$  are shown as  $a(T) = \Delta\rho(T)/T^2$  vs  $T$  in Fig. 5(a), along with the resistively determined  $B$ - $T$  phase diagram in Fig. 5(b). We note that the field dependence of the limiting temperature  $T_L$  below which the  $\Delta\rho = aT^2$  dependence is obeyed tracks that of the phase transition temperature,  $T_B(B)$ . Likewise, for our type I polycrystal measured at  $p = 0$ , the field dependence of  $T_L$  ( $\approx 1.2 \text{ K}$  at  $B = 0$ ) is tracking  $T_A(B)$  (not shown). In addition, we have recently found that, at  $p < p_C$ , the pressure dependences of  $T_L(p)$  and  $T_A(p)$  are very similar [23]. One might be inclined from these observations to ascribe the  $\Delta\rho = aT^2$  dependence preceding the  $A/B$  transitions to some critical fluctuations. However, assuming the  $A/B$  phases to be of an itinerant nature,  $\Delta\rho \sim T$  is predicted [24] in the critical regime,  $T > T_{A,B}$ .

In summary, the low-temperature properties of homogeneous  $\text{CeCu}_2\text{Si}_2$  samples are governed, depending on composition, by a complicated interplay between phase  $A$  and HF superconductivity: One can get rid of the  $A$ -phase signatures by applying a minute hydrostatic pressure or by

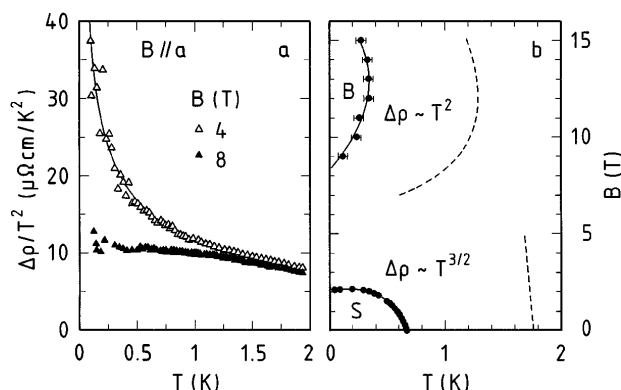


FIG. 5. (a)  $\Delta\rho/T^2$  vs  $T$  ( $\Delta\rho = \rho - \rho_0$ ) at  $B = 4$  and  $8$  T for the same crystal as in Figs. 3 and 4. Solid line marks  $T^{-1/2}$  dependence of  $a(T) = \Delta\rho(T)/T^2$ . (b)  $B$ - $T$  phase diagram for the same crystal derived from  $\rho(T)$  measurements. Existence ranges for superconductivity and phase  $B$  are indicated along with limiting temperatures for  $T^{3/2}$  and  $T^2$  dependences of  $\Delta\rho(T)$  (dashed lines).

choosing a suitably prepared (type III) single crystal. In either case [25], measurements of the  $n$ -state resistivity and specific heat performed in sufficiently low fields and at intermediate temperatures suggest that an AF-QCP exists where  $T_A \rightarrow 0$ . This is concluded from the agreement of the experimental data with theoretical predictions for a NAFFL in a one-band system of itinerant fermions [4–6]. However, at low temperatures, there are two striking observations that strongly violate this NAFFL scenario: (1) The cross section of quasiparticle-quasiparticle scattering measured by  $a(T) = \Delta\rho(T)/T^2$  keeps diverging  $\sim T^{-1/2}$  down to mK temperatures rather than becoming constant [7]. This suggests that singular scattering occurs on the whole Fermi surface rather than along some hot lines only. (2)  $\Delta\rho(T)$  and  $\gamma(T)$  behave very disparately. This indicates a decoupling of the itinerant and the local (4f) parts out of which the heavy fermions are composed [26]. In addition, extremely disparate behavior of  $\Delta\rho(T)$  ( $= aT^2$ ) and a strangely  $T$ -dependent  $\gamma(T)$  is found with the polycrystal (of type I) precursive to the  $A$ -phase transition at  $T_A > 0$ . The same is observed, precursive to the  $B$ -phase transition at  $B > 8$  T, for the single crystal (of type III). For the latter a dramatic change in the  $T$  dependence of  $\Delta\rho(T)$  occurs as a function of magnetic field near  $B = 6$  T.

We conclude by speculating that, in near stoichiometric  $\text{CeCu}_2\text{Si}_2$ , it is the formation of Cooper pairs below  $T_C \approx 0.65$  K which preserves the heavy quasiparticles. In the absence of superconductivity, the latter appear to break up in the vicinity of the competing phase  $A$ . More insight into the nature of phase  $A$  is expected from experiments on slightly Ge-doped samples which are in progress [27].

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