Breakup of Heavy Fermions on the Brink of "Phase A" in $CeCu_2Si_2$

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(Received 23 October 1997)

We report resistivity, $\rho(T)$, and specific-heat, $C(T)$, results on near stoichiometric $CeCu_2Si_2$ samples, in the vicinity of a quantum critical point (QCP). The latter is defined by $T_A \to 0$, where $T_A \lesssim 0.8$ 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]

PACS numbers: 74.70.Tx, 75.30.Mb, 75.40.Cx

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 $\mathbf{q} \simeq \mathbf{Q}$, 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 (ρ_0: residual resistivity) which are not constant as in a Landau FL, but obey the following asymptotic T dependences (in 3D) [4,5]:

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

and [4–6]

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

corresponding to

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

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 $\Delta\rho = aT^2$ ($a =$ 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 $CeCu_2Si_2$ [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

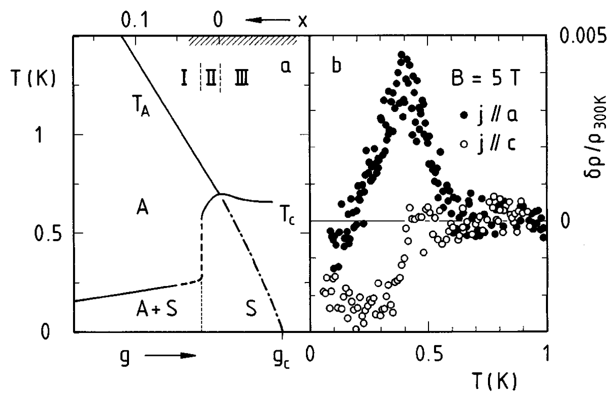

FIG. 1. (a) Schematic phase diagram for $CeCu_2Si_2$ at zero field, indicating existence ranges for phase A, superconductivity (S), and coexistence range ($A + S$). For samples labeled type I, II, and III: $T_A > T_C, T_A \gtrsim T_C$, and $T_A < T_C$, respectively (see text). The form of the phase boundaries 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 $CeCu_2Si_2$ samples [9] (hatched regime) or is proportional to the Ge content x in $CeCu_2(Si_{1-x}Ge_x)_2$ [10]. $g = g_C$ marks $T_A \to 0$. 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 $CeCu_2Si_2$ single crystal as $\delta\rho/\rho_{300\,K}$ 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.

0031-9007/98/81(7)/1501(4)$15.00

(SDW) transition [Fig. 1(b)]. (ii) $CeCu_2Si_2$ 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 $CeCu_2Si_2$ samples which, according to x-ray diffractometry, were found to be single phase with the proper $ThCr_2Si_2$ structure. The $Ce_{0.99}Cu_{2.02}Si_2$ polycrystal was prepared in an argon-arc furnace and subsequently annealed at 700 °C for 24 h and 1000 °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 $LaCu_2Si_2$. 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}He-^{4}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}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), $CeCu_2(Si_{1-x}Ge_x)_2$ samples 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" $CeCu_2Si_2$ samples of near stoichiometric composition and with $T_A \gtrsim T_C$, (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" $CeCu_2Si_2$ 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 $B = 0$ μSR 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 $CeCu_2Si_2$ 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 $CeCu_2Si_2$ 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

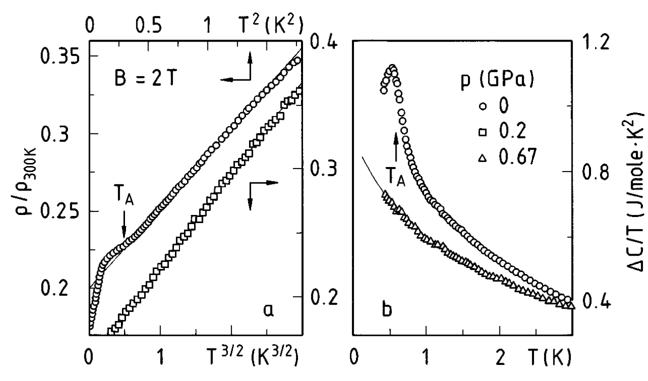

FIG. 2. Normalized resistivity as $\rho/\rho_{300\text{ K}}$ vs T^2 (upper scale) and $\rho/\rho_{300\text{ K}}$ vs $T^{3/2}$ (lower scale) (a) as well as Ce increment to the specific heat as $\gamma = \Delta C/T$ vs T (b), for $B = 2T$ and at $p = 0$ as well as two overcritical pressures for a type I $CeCu_2Si_2$ 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.

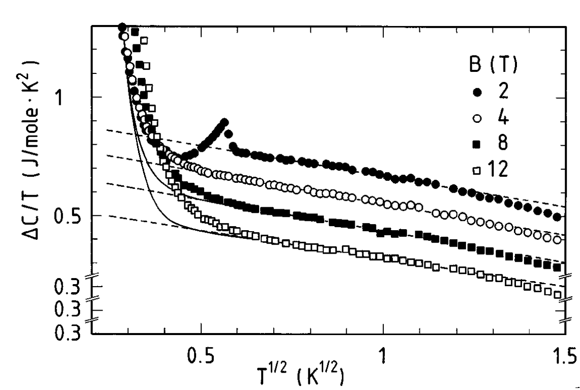

FIG. 3. $\gamma = \Delta C/T$ vs $T^{1/2}$ at varying fields for a type III $CeCu_2Si_2$ single crystal. Dashed lines indicate $(-T^{1/2})$ dependence of $\gamma(T) - \gamma_0$. Solid lines display $\Delta C(T)/T$ data after subtraction of nuclear hyperfine contributions due to the applied B fields. For $B = 2\ T$, the SC transition anomaly at $T_C \approx 0.3$ K is seen.

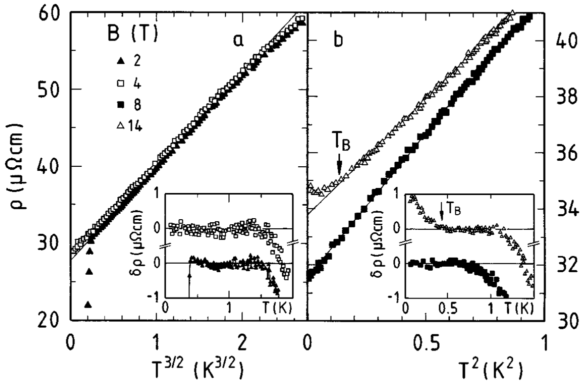

FIG. 4. Resistivity for the same crystal as in Fig. 3 as ρ vs $T^{3/2}$ for $B = 2$ and 4 T (a) as well as ρ vs T^2 for $B = 8$ and 14 T (b). Insets show $\delta\rho$ vs T with $\delta\rho = \rho - \rho_0 - \beta T^{3/2}$ (a) and $\delta\rho = \rho - \rho_0 - aT^2$ (b), respectively. Arrows in (b) indicate 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 $a \approx 10\ \mu\Omega\,\mathrm{cm\,K^{-2}}$ 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 K $< T <$ 2 K and $B = 2$ 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 ($B < 6$ T) and at intermediate temperatures ($T > 0.2$ K) [cf. Figs. 3 and 4(a)]. In the normal state, both $\gamma(T)$ and $\Delta\rho(T)$ obey Eqs. (1) and (2b) for $T < 1.7$ K. The slopes in the respective plots γ 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 (ρ_0) or down (γ_0) if the field is increased. The data at $T > 0.2$ K and $B < 6$ 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 $p > p_C$, $B = 2$ T, and $T > 0.4$ K, the minimum temperature accessible in the ^{3}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 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}Cu, ^{65}Cu, or ^{29}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 mK.

Finally, we wish to address the surprising effect a magnetic field $B > 6$ T has on the low-T properties of our $CeCu_2Si_2$ 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 T, whereas the giant coefficient a decreases by almost the same fraction. In addition, the $B = 14$ T data display the broadened transition into phase B which is not visible in $\gamma(T)$ measured at, e.g., $B = 12$ T (Fig. 3). Our resistivity and specific-heat results on the type III single crystal at $B > 6$ 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 $CeCu_2Si_2$ single crystal below and above $B \approx 6$ 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 ($\simeq 1.2$ 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 $CeCu_2Si_2$ 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

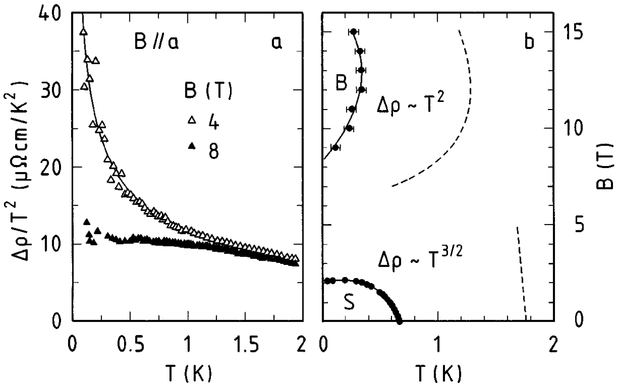

FIG. 5. (a) $\Delta\rho/T^2$ vs T ($\Delta\rho = \rho - \rho_0$) at $B = 4$ and 8 T for the same crystal as in Figs. 3 and 4. Solid line marks $T^{-1/2}$ dependence of $a(T) = \Delta\rho(T)/T^2$. (b) B-T phase diagram for the same crystal derived from $\rho(T)$ measurements. Existence ranges for superconductivity 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 $CeCu_2Si_2$, 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].

We thank Piers Coleman for a most fruitful correspondence and for sending us his new results [26] prior to publication. We are grateful to Gil Lonzarich, Julian Sereni, Peter Thalmeier, and Octavio Trovarelli for several valuable conversations.

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