Non-Fermi-liquid behavior in $CeCu_2Si_2$ at the disappearance of the presumably magnetically ordered “A-phase”

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Abstract

We present transport and thermodynamic measurements on two high-quality single crystals of $CeCu_2Si_2$ with different ground-state properties: crystal #1 (“A/S”-type) shows a competition of superconductivity and a presumably magnetically ordered “A-phase”. The latter develops out of a low-temperature state where the resistivity varies as $\rho - \rho_0 = aT^2$ indicative of a coherent heavy Fermi liquid. By contrast, non-Fermi-liquid behavior is found for crystal #2 (“S”-type) which lacks the A-phase for $B < 5$ T.

Keywords: $CeCu_2Si_2$; Heavy-fermion superconductivity; Non-Fermi liquid system; Magnetic correlation

Recent investigations of high-quality single crystals of $CeCu_2Si_2$ showed that in this compound heavy-fermion superconductivity is almost degenerated with a state labeled “phase A” [1, 2]. Since the latter is related to distinct anomalies in magnetic properties, i.e. magnetoresistance [3], NMR [4] and μSR [5] it has been frequently associated with some magnetically ordered state. By means of a systematic study of polycrystalline slightly off-stoichiometric $CeCu_{2+x}Si_{2+y}$ samples the various ground-state properties could be related to different sections within the narrow homogeneity range of the 1–2–2 phase in the ternary Ce–Cu–Si phase diagram [6, 7]: while Cu-rich samples are superconductors (“S”-type) below $T_c \approx 0.65$ K an A-phase transition is found at $T_A \approx 0.6$–0.8 K in samples with small Ce and/or Cu deficiency (“A”-type). In between these two regions “A/S” behavior is observed, i.e. 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 [8] one might be able to tune the system through a quantum critical point (QCP) at which $T_A = 0$. Provided that T_A marks the onset of long-range magnetic order clear deviations from a Fermi-liquid (FL) behavior are expected as a consequence of the critical (long-lived, low-lying) spin fluctuations with large correlation length which strongly interact with the heavy fermions near the QCP. Both scaling [9, 10] as well as phenomenological spin-fluctuation theories [11, 12] predict the following asymptotic T dependences for the resistivity, ρ, and specific heat, C, respectively: $\Delta\rho = \rho - \rho_0 = \beta T^{3/2}$ and $C/T = \gamma_0 - \alpha T^{1/2}$. As was recently shown for “A”-type polycrystalline $CeCu_2Si_2$ the application of a pressure of 0.7 GPa, in fact, results in a replacement of the A-phase by superconductivity and in the occurrence of

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non-Fermi-liquid (NFL) behavior in the specific heat [8].

Below we communicate a comparative study of low-T transport and thermodynamic properties on high-quality $CeCu_2Si_2$ single crystals of " A/S " and " S " -type, i.e. in the presence and absence of the A-phase. While the " A/S " crystal is of the same kind as the one used by Bruls et al. [2] the " S " crystal is identical to that studied by Lang et al. [1]. The resistivity was measured using the standard four-terminal AC technique ($\nu = 17$ Hz, $I = 100$ μA) with the current, j, flowing perpendicular to the applied field. Details concerning the specific-heat measurements are described elsewhere [13]. The residual resistivities, ρ_0, of the " A/S " and " S " crystals are (5 ± 1) and (26 ± 5) μΩ cm, respectively. A somewhat smaller ρ_0 value for the " A/S " crystal is consistent with results on off-stoichiometric polycrystalline materials yielding the highest crystalline perfection for samples of the " A/S " sector [14]. Fig. 1(a) shows the temperature dependence of the resistivity, normalized to its value at 300 K, $\rho(T)/\rho(300\text{ K})$, of the " A/S " crystal. The measurements were performed in overcritical fields applied parallel to the a- and c-axis (not shown), respectively. For an extended temperature range $T_{A/B} < T < \tilde{T}(B)$ the resistivity is well described by $\rho - \rho_0 = aT^2$, with a huge and almost field-independent coefficient $a \approx (10 \pm 1)$ μΩ cm K^{-2}. The temperatures T_A, T_B below which the data points deviate from the straight lines in the representation $\rho(T)/\rho(300\text{ K})$ versus T^2 mark the transition temperatures into the phase A and the high-field phase B, respectively, in accordance with the results from thermal-expansion measurements performed in $B \leqslant 2$ T (cf. Fig. 1(b)). We note that while the transition into phase B is accompanied by an increase of the resistivity for both field orientations anisotropic scattering contributions with $\Delta\rho < 0$ for $B \| a$ ($j \| c$) and $\Delta\rho > 0$ for $B \| c$ ($j \| a$) are specific to the A-phase transition (cf. inset Fig. 1(a)). Most remarkably we find that $\tilde{T}(B)$ the upper bound of the $\rho \sim T^2$ behavior is correlated with the field dependence of T_A and T_B, respectively: a lower (higher) T_A, T_B is accompanied by a lower (higher) $\tilde{T}$ (cf. Fig. 1(b)). The same behavior was found for " A "-type polycrystalline material [14] where $\tilde{T}(B = 0) = 1.3$ K is close to the temperature below which short-range antiferromagnetic correlations have been detected by μSR measurements [15]. Apparently, short-range magnetic correlations precursive to the transitions into the phases A and

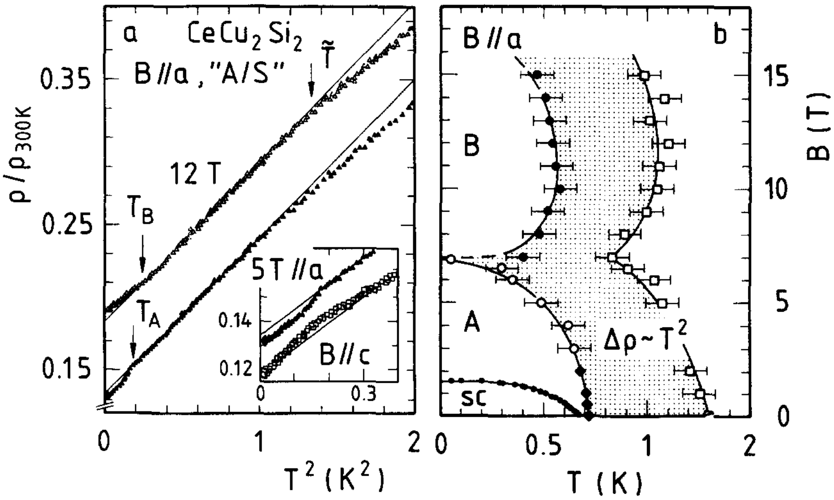

Fig. 1. (a) Normalized resistivity versus T^2 for the " A/S "-type $CeCu_2Si_2$ single crystal at fields $B = 5$ and 12 T applied parallel to the a-axis. (b) Corresponding B–T phase diagram including the superconducting (sc), A and B phases as well as the ranges where the resistivity follows FL-like ($\rho \sim T^2$) behavior. For $B \leqslant 2$ T the A-phase boundary was also determined by thermal-expansion measurements (◆).

B are at the origin of the T^2 dependence in the resistivity [16].

By contrast, deviations from an FL-like T-dependence is found for the normal-state resistivity of the "S"-type crystal (Fig. 2(a)), which lacks any indication of phase A for fields below 5 T [1]. In this field range the resistivity can be well described by $\rho - \rho_0 = \beta T^{\varepsilon}$ with $\varepsilon = 1.5 \pm 0.05$. As exemplary shown in Fig. 2(a) for $B = 4$ T this description holds for nearly two decades in temperatures, i.e. below 1.7 K down to 20 mK, the lowest temperature of our experiment. Upon increasing the field to above 6 T the low-T resistivity turns into a $\rho - \rho_0 = aT^2$ dependence (cf. Fig. 2(b)), again characterized by a huge and almost field-independent coefficient of $a \approx 10\ \mu\Omega\mathrm{cm\,K}^{-2}$. For $8\ \mathrm{T} \leqslant B \leqslant 15.5$ T, the highest field of our experiment, this T^2 behavior precedes the transition into phase B (cf. Fig. 2(b)) which manifests itself in an increment of the resistivity for $T \leqslant T_B$. Compared to the "A/S" crystal the B phase of the "S"-type crystal forms at somewhat lower temperatures indicative of a weakening of its thermodynamic stability. Moreover, these results prove that phase B may form even in the absence of phase A. The crossover from NFL behavior for $B \leqslant 5$ T to FL-like behavior for $B \geqslant 6$ T is substantiated by the results of magnetoresistance measurements: in an isothermal field sweep at $T = 20$ mK $\Delta\rho(B) = \rho(B) - \rho(0)$ changes sign from an anomalous $\Delta\rho(B) < 0$ for $B_{c2}(20\ \mathrm{mK}) < B \leqslant 5$ T to the ordinary $\Delta\rho(B) > 0$ for $B > 5$ T.

The corresponding results of the low-T specific heat for fields 4 and 12 T applied parallel to the a-axis are displayed in Fig. 3 as C/T versus $T^{1/2}$. To demonstrate the contribution of the nuclear specific heat, C_{nuc}, caused by field-induced Zeeman splitting of Cu and Si nuclear spin states, we included the data sets corrected for C_{nuc}. The latter was calculated by assuming that the hyperfine field acting on the nuclear spins is identical to the external field. In contrast to the crossover behavior observed for $\rho(T, B)$ the specific heat does not show a significant change on going from the low-field ($B < 5$ T) to the high-field ($B > 6$ T) regime. In a limited temperature range $1\ \mathrm{K} \leqslant T \leqslant 3$ K the data sets reveal NFL behavior of the form $C/T = \gamma - \alpha T^{1/2}$ (straight line in Fig. 3). Upon lowering the temperature, however, C/T starts to deviate from the $T^{1/2}$ behavior with the tendency to saturate for $T \to 0$. This suggests a crossover from an NFL to a seeming FL behavior for $T < 1$ K. A detailed analysis of the low-T specific heat data is, however, impeded by the steep upturn in C/T below about 0.25 K. As evident from Fig. 3 this

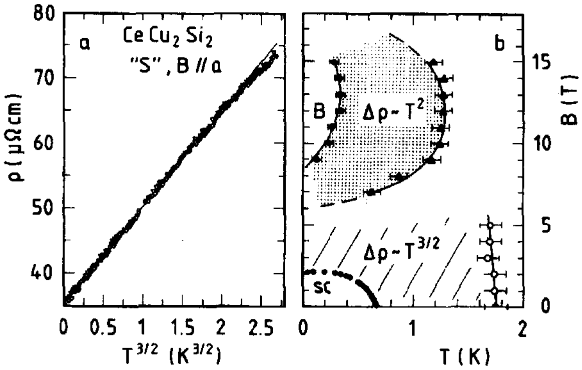

Fig. 2. (a) Resistivity versus $T^{3/2}$ for the "S-type" $\mathrm{CeCu_2Si_2}$ single crystal at $B = 4$ T applied parallel to the a-axis. (b) Corresponding B–T phase diagram including the superconducting (sc) and B phase as well as the ranges where the resistivity follows FL-like ($\rho \sim T^2$) and NFL ($\rho \sim T^{3/2}$) behavior.

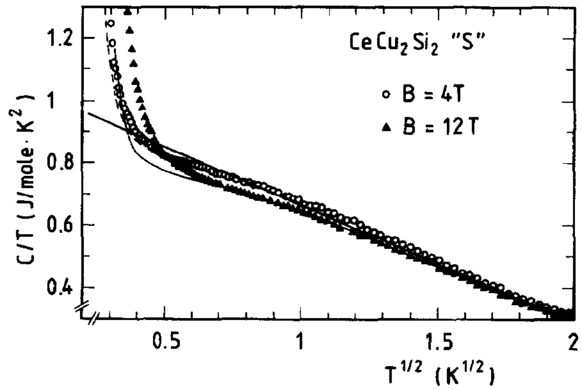

Fig. 3. Specific heat of the "S"-type $\mathrm{CeCu_2Si_2}$ single crystal as C/T versus $T^{1/2}$ for various fields applied parallel to the a-axis. Dotted ($B = 4$ T) and thin solid ($B = 12$ T) lines represent the data after subtraction of a nuclear contribution (see text). The thick solid line represents the dependence $C/T = \gamma - \alpha T^{1/2}$.

behavior cannot be explained by the effect of only the external field on the nuclear spins. An anomalous enhancement of the hyperfine coupling, i.e. an (average) finite "internal magnetic field" at the Cu and/or Si sites has to be invoked to quantitatively account for the anomalous T dependence. We also stress that in contrast to the resistivity results no indication of phase B is visible in the raw data of $C(T, B)$ reflecting a substantial reduction of the ordered moment compared to that of an "A" or "A/S"-type sample [17].

In conclusion, by means of low-T resistivity measurements on two slightly different high-quality single crystals of $CeCu_2Si_2$ we could track the change from a behavior consistent with a heavy FL for the "A/S-type" crystal to NFL behavior in the "S-type" crystal. While the resistivity shows the characteristic behavior expected above a QCP, i.e. $\rho \sim T^{3/2}$ the corresponding $C/T \sim T^{1/2}$ law is observed only for a limited T range down to about 1 K.

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