Evidence for an antiferromagnetic quantum critical point near stoichiometric $CeCu_2Si_2$

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Abstract

We report on a comparative study of the electrical resistivity, ρ, and specific heat, C, on the presumably magnetically ordered phase A in near stoichiometric $CeCu_2Si_2$ at very low-T, in magnetic fields and as a function of pressure. Our data indicate the formation of a spin-density-wave (SDW) phase below T_A. Phase A depends sensitively on the strength g of hybridisation between 4f- and conduction electrons and g can be varied by the application of pressure or suitable changes in composition. Our results suggest a continuous evolution $T_A \to 0$ for $g \to g_c$. Hydrostatic-pressure experiments on a near stoichiometric polycrystal prove that g_c marks a quantum critical point of antiferromagnetic type consistent with the SDW-nature of phase A.

Keywords: Heavy-fermion superconductivity; Non-Fermi liquid; Magnetic correlations

Up to now the tetragonal compound $CeCu_2Si_2$ [1] is the only Ce-based heavy-fermion system (HFS) that shows superconductivity (SC) already at ambient pressure whereas for several antiferromagnetically (AFM) ordered isostructual HFS SC occurs near the critical pressure that suppresses magnetism [2–4]. The aim of this contribution is to provide evidence that $CeCu_2Si_2$ is located close to a quantum critical point (QCP) of AFM type.

Measurements of the elastic constants and thermal expansion [5] on a high-quality $CeCu_2Si_2$ single crystal revealed that SC competes with a so-called “phase A” inside which slowly fluctuating internal magnetic fields were observed in μSR [6] and NMR [7] experiments. By means of a detailed investigation of slightly off-stoichiometric $Ce_{1+x}Cu_{2+y}Si_2$ polycrystals Geibel et al. [8,9] found that phase A can be suppressed in $B = 0$ in favour of SC by a minor Ce- or Cu-excess. The observed changes in the physical behaviour were ascribed to an increase of the strength g of hybridisation between 4f- and conduction-electrons although no change in the lattice parameters can be resolved by standard X-ray diffraction.

Below we report on a detailed investigation of the electrical resistivity and the specific heat of a high-quality single crystal and near stoichiometric polycrystals in magnetic fields up to 17 T. Measurements at zero pressure were performed using a dilution refrigerator while in pressure experiments up to 0.7 GPa CuBe piston–cylinder cells were adopted to ^{3}He-cryostats. The resistivity was measured utilising a standard four-terminal AC-technique ($\nu = 17$ Hz, $I = 100$ μA) with the current, j, flowing perpendicular to the applied field. For measurements of the heat capacity a compensated heat-pulse method was used [10].

The high-quality single crystal #1 is the same as studied in [11] and of the same kind as the one used by Bruls et al. [5]. The different slightly off-stoichiometric polycrystals were prepared in an argon-arc furnace and

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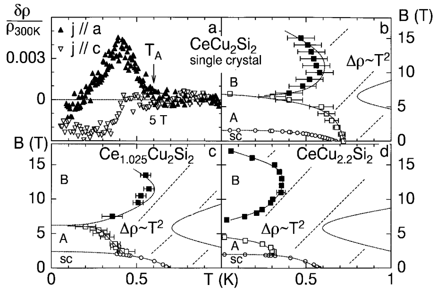

Fig. 1. Normalized resistivity of a $CeCu_2Si_2$ single crystal in $B = 5$ T measured along the *a*- and *c*-axes as $\delta\rho = \rho(T) - (\rho_0 + aT^2)$ versus *T* (a). *B–T* phase diagrams for the same single crystal (b) and polycrystals $Ce_{1.025}Cu_2Si_2$ (c) and $CeCu_{2.2}Si_2$ (d) as derived from resistivity measurements. Lines are separating SC, phase A, phase B and range in which $\Delta\rho = (\rho - \rho_0) \sim T^2$ holds.

subsequently annealed at 700°C for 24 h and 1000°C for 120 h. According to X-ray diffractometry, they are single phase with the proper $ThCr_2Si_2$ structure [9].

Fig. 1b shows the *B–T* phase diagram of single crystal #1 as derived from resistivity measurements which is quite similar to that published in Ref. [5]. Phases A and B develop out of a state, where the resistivity shows a T^2-behaviour with a giant coefficient $a \approx 10\ \mu\Omega\,\mathrm{cm\,K^{-2}}$. This would suggest a heavy Landau–Fermi-liquid state with which notion the strongly *T*-dependent Sommerfeld coefficient $\gamma(T)$ in specific-heat measurements in the same *T*-range (see, e.g. Fig. 2b, $p = 0$) is, however, incompatible.

As for the nature of the transition at T_A the resistivity contribution $\delta\rho(T) = \rho(T) - (\rho_0 + aT^2)$ (ρ_0 is the residual resistivity) is increasing below T_A for $j \| a$ while no anomaly can be resolved for $j \| c$ (Fig. 1a) at this temperature. This observation could be ascribed to the effect of Fermi-surface nesting in the tetragonal plane due to the formation of a SDW that reduces the effective number of charge carriers. At somewhat lower temperature a nearly isotropic reduction of $\delta\rho$ may indicate the freezing-out of incoherent scattering as expected for an SDW.

Next we turn to the near stoichiometric polycrystals $Ce_{1.025}Cu_2Si_2$ and $CeCu_{2.2}Si_2$ in which due to the increased strength of hybridisation *g* [8,9] only a minor magnetic volume fraction was observed in zero-field μSR experiments [6]. The resistively determined *B–T* phase diagrams are shown in Fig. 1c and Fig. 1d. Most remarkably, in magnetic fields $B > B_{c2}$, signatures of both the A- and B-phase though slightly reduced in temperature can still be found. The extrapolation to $B = 0$ yields fictitious transition temperatures of $T_A(B = 0) \approx 0.5$ K and $T_A(B = 0) \approx 0.35$ K for $Ce_{1.025}Cu_2Si_2$ and $CeCu_{2.2}Si_2$, respectively, that are below T_c. Apparently, in $B = 0$ SC is more stable than phase A in these polycrystals with enhanced *g*.

These results indicating a continuous evolution of $T_A \to 0$ for $g \to g_c$ strongly suggest the existence of a QCP at $T_A = 0$. To prove this, the pressure-induced A–S transition is studied on a $Ce_{0.99}Cu_{2.02}Si_2$ polycrystal with non-reduced T_A [12]. The A-phase transition, when measured at $p = 0$ and $B = 2$ T, manifests itself in broadened anomalies in the *T*- dependences of $\rho(T)$ and $\gamma(T) = \Delta C(T)/T$ at $T_A \approx 0.75$ K (Fig. 2). At pressures exceeding a critical value $p_C \approx 0.1$ GPa, phase A is completely suppressed. For $p > p_c$ and in a magnetic field of 2 T that destroys SC the characteristic *T* dependences $\rho - \rho_0 \sim T^{3/2}$ and $\gamma - \gamma_0 \sim (-T^{1/2})$ indicate a 3d QCP [13]. At temperatures lower than 0.4 K, the minimum temperature of our ^{3}He-cryostats, however, measurements on a single crystal near g_c, that are discussed elsewhere [14], revealed that $\rho(T)$ and $C(T)$ behave very disparately, suggesting a decoupling of the itinerant from the local parts out of which the HF are composed.

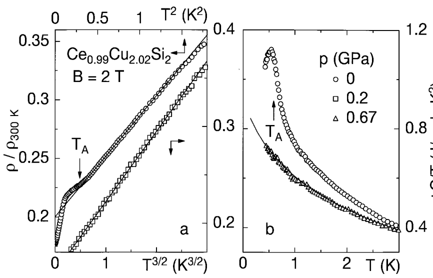

Fig. 2. $\rho/\rho_{300\,K}$ versus T^2 (upper scale) and $\rho/\rho_{300\,K}$ versus $T^{3/2}$ (lower scale) (a) as well as Ce-increment to the specific heat as $\gamma = \Delta C/T$ versus *T* (b), for $B = 2$ T and at $p = 0$ and two overcritical pressures for a $Ce_{0.99}Cu_{2.02}Si_2$ polycrystal. Solid lines display T^2 and $T^{3/2}$ dependences of $\rho(T)$ (a) and a fit of the "SCR theory" [13] to the $\gamma(T)$ data, implying parameters $y_0 = 0, y_1 = 4$ and $T_0 = 13$ K that correspond to a $\gamma - \gamma_0 \sim (-T^{1/2})$ dependence below 1.2 K.

To summarize, phase A in $CeCu_2Si_2$ shows signatures of an SDW and is suppressed by the slight changes in composition or via the application of weak hydrostatic pressure. Our results provide strong evidence for the existence of a nearby AFM QCP at $T_A \to 0$.

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