Pressure Tuning of the Interplay of Magnetism and Superconductivity in $CeCu_2Si_2$

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We carried out specific-heat and ac-susceptibility experiments under hydrostatic pressure to investigate the interplay of spin-density-wave antiferromagnetism (A) and superconductivity (S) in single-crystalline AS-type $CeCu_2Si_2$. We find evidence for a line of magnetic-field- and pressure-tuned quantum critical points in the normal state in the zero-temperature magnetic field–pressure plane. Our analysis suggests an extension of this line into the superconducting state and corroborates the close connection of the underlying mechanisms leading to the formation of the antiferromagnetic and the superconducting states in AS-type $CeCu_2Si_2$.

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The discovery of superconductivity in heavy-fermion (HF) [1], organic [2], cuprate [3], and, most recently, pnictide materials [4] changed our understanding of superconductivity completely. Despite fundamental differences between these families, the proximity of a magnetic ground-state instability to the superconducting (SC) phase is a common theme in all of the SC systems. In pnictide, organic, and some HF superconductors, itinerant [spin-density-wave (SDW) type] antiferromagnetism seems to be closely related to the formation of the SC phase, suggesting a magnetically mediated SC pairing mechanism. The Néel temperature T_N can generally be tuned as a function of some external parameter, such as doping or pressure. Typically, superconductivity develops in the vicinity of the point where T_N disappears as a function of the external parameter (e.g., [5–7]). The normal state transport and thermodynamic properties close to that point often disclose a region of non-Fermi-liquid behavior hinting at the presence of a (hidden) quantum critical point (QCP) [6]. In the cuprates and iron pnictides, the SC upper-critical field (B_{c2}^0) is generally accessible only by pulsed magnetic fields, putting strong restrictions on the available experimental probes and their accuracy. In contrast to these classes of materials, in HF superconductors B_{c2}^0 is moderate and the SC state can be easily suppressed by using standard laboratory equipment. This makes the HF materials perfect model systems for an in-depth investigation of the interplay of magnetism and superconductivity and a test bed for the theoretical models developed.

$CeCu_2Si_2$, the first discovered HF superconductor [1], is ideally suited to study the interplay of SDW antiferromagnetism and superconductivity. The ground state of $CeCu_2Si_2$ depends strongly on the exact stoichiometry. It ranges from (i) antiferromagnetism (A-type), coexisting in a small parameter range with low-T_c superconductivity, via (ii) antiferromagnetism which is replaced by superconductivity on lowering temperature or recovered when superconductivity is suppressed by a sufficiently large magnetic field (AS-type), to (iii) solely superconductivity (S-type). The antiferromagnetic (AFM) order in A-type $CeCu_2Si_2$ was shown to be an incommensurate SDW, with a very small ordered moment ($\mu \approx 0.1\mu_B$) [8]. In AS-type $CeCu_2Si_2$, the AFM and SC ordering temperatures are comparable. Resistivity and specific-heat results obtained in the field-driven low-T normal state of S-type $CeCu_2Si_2$ revealed non-Fermi-liquid phenomena, highly consistent with a three-dimensional (3D) SDW QCP [9]. The SC phase in $CeCu_2Si_2$ is rather robust against external pressure covering more than 5 GPa. Introduction of disorder by Ge doping on the Si site revealed the presence of two distinct SC domes. The SC state at low pressures is supposed to be mediated by AFM spin fluctuations, while at high pressures superconductivity is suggested to be mediated by valence fluctuations [10]. An analysis of thermodynamic data evidences the existence of different SC order parameters in the two distinct SC phases [11].

In this Letter, we will substantiate the close link between superconductivity and antiferromagnetism in the low-p region in AS-type $CeCu_2Si_2$. Furthermore, we provide evidence for the existence of a line of magnetic-field- and pressure-tuned QCPs which extends into the SC region of the magnetic field–pressure phase diagram.

Heat-capacity and ac-susceptibility experiments under hydrostatic pressure have been performed on a single-crystalline sample of AS-type $CeCu_2Si_2$ (0.26 K $\leq T \leq$ 7 K, $B_{\parallel c} \leq 8$ T). The resolution of the ac-susceptibility measurements allows only for observing the SC transition; the AFM transition anomaly cannot be resolved. The measurements were carried out in a single-layer CuBe piston-cylinder type pressure cell (for details, see [11]).

Figure 1 displays the electronic contribution to the specific heat as $C_{el}(T)/T$ at selected pressures. At atmospheric pressure, AS-type $CeCu_2Si_2$ undergoes two consecutive phase transitions below $T = 1$ K upon decreasing temperature: the first one at $T_N \approx 0.69$ K to an incommensurate AFM SDW type of order [8] and the second one at a slightly lower temperature, marking the onset of superconductivity at $T_c \approx 0.46$ K. The highly enhanced value

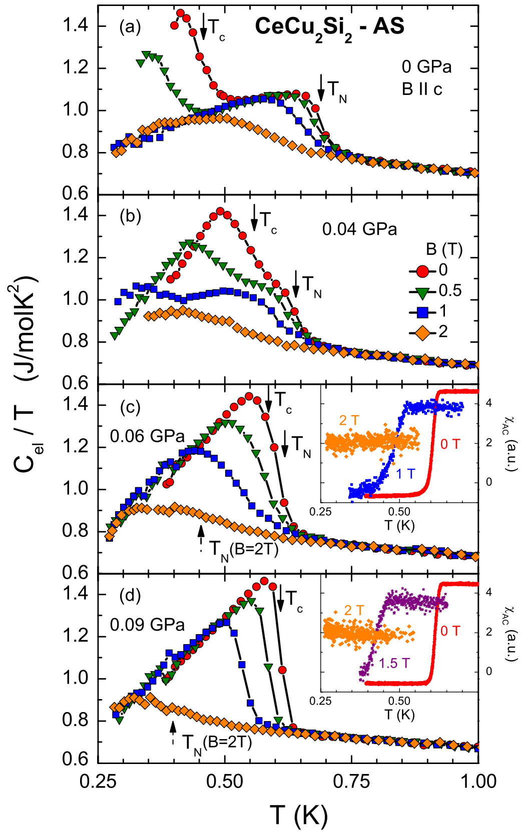

FIG. 1 (color online). $C_{\mathrm{el}}(T)/T$ as function of T of *AS*-type $CeCu_2Si_2$ for different magnetic fields $B = 0$, 0.5, 1, and 2 T ($B \parallel c$) and at four different pressures as indicated in (a)–(d). The solid arrows mark T_N and T_c at zero magnetic field. Dashed arrows denote the AFM transition at 2 T at 0.06 and 0.09 GPa. The transition temperatures were determined by an equal-entropy approximation. The insets of (c) and (d) show $\chi_{\mathrm{ac}}(T)$ for different magnetic fields as indicated.

of the electronic specific-heat coefficient at low temperatures, $C_{\mathrm{el}}/T \approx 0.73\ \mathrm{J/(mol\,K^2)}$ at $T = 0.9$ K, indicates the HF character. A Kondo temperature of $T_K \approx 13$ K can be determined by analyzing the entropy in the frame of the single-impurity Kondo model [12]. At T_N, the entropy reaches a value of only $S_{\mathrm{el}}(T_N) \approx 0.11 R \ln 2$, as anticipated from the small ordered moment of $\mu_{\mathrm{ord}} \approx 0.1\mu_B$ per Ce atom detected by neutron-diffraction experiments in the AFM state [8].

Application of a small hydrostatic pressure leads to a rapid shift of $T_N(p)$ to lower temperatures with an initial slope of $dT_N/dp|_{p=0} \approx -1.17$ K/GPa, while $T_c(p)$ at first strongly increases ($dT_c/dp|_{p=0} \approx 2.33$ K/GPa). In contrast to hydrostatic pressure, a magnetic field suppresses both T_N and T_c, T_c being much more sensitive to the magnetic field than T_N. As a consequence, at 0 and 0.04 GPa the two transition anomalies in specific heat become more separated in an external magnetic field. At ambient pressure no indication for a SC transition is present in the data at $B = 1$ T anymore in our accessible temperature range ($T > 0.27$ K), while at 0.04 GPa the anomaly at T_c is visible in $B = 1$ T but absent in 2 T, indicating an enhanced SC upper-critical field B_{c2}^0, compared with zero pressure. A further pressure increase to only $p = 0.06$ GPa is sufficient to shift the two phase transitions very close to each other, resulting in a single broadened anomaly in $C_{\mathrm{el}}(T)/T$. In a closer analysis the transition temperatures $T_N \approx 0.62$ K and $T_c \approx 0.60$ K can be extracted. Upon increasing the magnetic field, the anomaly in $C_{\mathrm{el}}(T)$ further broadens, reflecting the different dependencies of T_N and T_c on the magnetic field. While for $B < 2$ T the $\chi_{\mathrm{ac}}(T)$ data confirm the presence of the SC transition, at 2 T no diamagnetic signal is observed anymore, proving that the anomaly observed in specific heat corresponds only to the AFM transition and superconductivity is already suppressed. At a slightly larger pressure $p = 0.09$ GPa, a single sharp anomaly in specific heat at $B = 0$ signals the transition to the SC state. $\chi_{\mathrm{ac}}(T)$ experiments prove the presence of superconductivity up to $B = 1.5$ T. We find no hint at a magnetic transition below T_c. At 2 T, $\chi_{\mathrm{ac}}(T)$ does not show any diamagnetic signal anymore, but $C_{\mathrm{el}}(T)/T$ exhibits a broad anomaly. Therefore, we identify this anomaly with the transition into the AFM state. At higher pressures (not shown), no indication for a magnetic transition is observed in the field-induced normal state anymore. Especially, no signature of an AFM phase transition inside the SC state is found at any magnetic field and pressure. Our results clearly indicate that once $T_c(p)$ for a fixed B becomes larger than $T_N(p)$, the presence of the AFM phase transition cannot be detected anymore. This strongly suggests the absence of any long-range magnetic ordering inside the SC phase. We therefore conclude that the antiferromagnetically ordered state in *AS*-type $CeCu_2Si_2$ is expelled once superconductivity has been established.

The deduced low-pressure T-p phase diagram of *AS*-type $CeCu_2Si_2$ is presented in Fig. 2. At zero magnetic field, $T_c(p)$ exhibits a weak maximum around $p_{T_{c,\max}} \approx 0.4$ GPa. Although we cannot follow $T_N(p)$ inside the SC state, we can extrapolate $T_N(p)$ from the normal into the SC state. We follow the predictions of the spin-fluctuation theory for a 3D SDW QCP and use $T_N(p) \propto (p - p_c)^{2/3}$ to extrapolate $T_N(p)$ [13]. By our analysis, we obtain a critical pressure $p_c \approx 0.39$ GPa, which nearly coincides with the position of the maximum in $T_c(p)$. We have utilized the same approach to extrapolate the $T_N(p, B = \mathrm{const})$ data taken in magnetic fields ($B = 0.5$, 1, and 2 T). With increasing magnetic field, the critical pressure $p_c(B)$ and the position of the maximum in $T_c(p, B = \mathrm{const})$, $p_{T_{c,\max}}(B)$, also coincide and shift to lower pressures. The tight correlation of $p_c(B)$ and $p_{T_{c,\max}}(B)$ thus suggests a strong link between the underlying mechanisms leading to the formation of the two ordered phases in $CeCu_2Si_2$.

As pointed out before, we do not detect any magnetic phase-transition anomaly inside the SC state. Clear

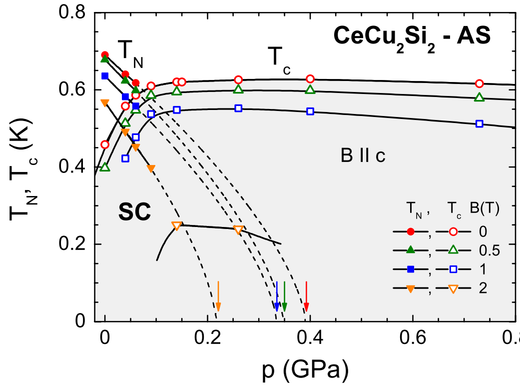

FIG. 2 (color online). Effect of a magnetic field ($B \parallel c$) on the low-pressure T-p phase diagram of AS-type $CeCu_2Si_2$. Full symbols correspond to T_N, while open symbols mark T_c. The dashed lines extrapolate $T_N(p)$ to zero temperature (see the text for details). The arrows mark the critical pressures p_c for the corresponding magnetic field.

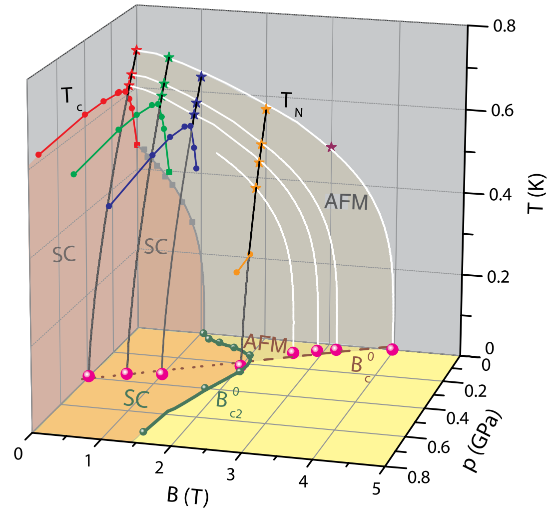

FIG. 3 (color online). Magnetic and superconducting phase diagram of AS-type $CeCu_2Si_2$ as a function of p and B for $B \parallel c$. Stars and circles correspond to $T_N(p, B)$ and $T_c(p, B)$, respectively. Black lines represent fits to the $T_N(p, B = \text{const})$ data and white lines to the $T_N(B, p = \text{const})$ data. A detailed description can be found in the text. The critical points $B_c^0(p)$ obtained by the extrapolation of the fitting curves to zero temperature are marked with bullets in the p-B plane. "SC" and "AFM" mark the superconducting and the antiferromagnetic phase, respectively. $B_{c2}(T)$ data at ambient pressure are shown. The extrapolated upper-critical field $B_{c2}^0(p)$ is indicated by the solid line.

evidence for microscopic coexistence of magnetism and (low-T_c) superconductivity has been found only at negative chemical pressures [14]. Thus, we speculate that, despite the fact that we cannot identify a magnetic phase transition or a QCP inside the SC state, their *halo* is present. For AFM spin-fluctuation-mediated superconductivity, the maximum of the SC dome is expected at the critical point where magnetism is completely suppressed [15,16]. This is in agreement with our findings for AS-type $CeCu_2Si_2$. However, in zero magnetic field, $T_c(p)$ exhibits only a weak maximum at the critical pressure $p_c \approx 0.39$ GPa, in contrast to the pronounced SC dome typically observed in HF superconductors, e.g., [6]. The almost pressure-independent behavior of $T_c(p)$ in AS-type $CeCu_2Si_2$ might be related to the influence of the additional pairing mechanism provided by the valence fluctuations present at higher pressures [10,11,17], thus indicating that already in this relatively low-pressure region of the phase diagram the two SC pairing mechanisms have to be considered.

Application of both magnetic field ($B \parallel c$) and pressure leads to a gradual suppression of the AFM order in AS-type $CeCu_2Si_2$. While, as already discussed above, for a fixed magnetic field the $T_N(p)$ curve cannot be followed inside the SC phase anymore, at a constant pressure T_N can be continuously suppressed to zero temperature by increasing the magnetic field, suggesting the presence of field-induced QCPs. The experimental data $T_N(B, p = \text{const})$ are well described by the empirical formula $B_c(T) = B_c^0[1 - (T/T_N^0)^n]$, where T_N^0 is the Néel temperature at $B = 0$, $B_c^0 = B(T_N = 0)$, and n is a fitting parameter. n was determined once at ambient pressure ($n = 3.9$) and then kept constant for all other pressures. The results of the fits are indicated by the white lines in Fig. 3. The critical points in the p-B plane at $T = 0$, $B_c^0(p)$, follow a straight line. This is indicated in Fig. 3 by the dashed line. At ambient pressure $B_c^0(p = 0) \approx 4$ T. An increase of pressure leads to a gradual decrease of $B_c^0(p)$, $B \parallel c$ [for $B \parallel a$, $B_c^0(p)$ shows initially only a weak pressure dependence [18]]. At about $p = 0.18$ GPa and $B = 2.1$ T, the line of critical points hits the SC phase boundary (solid line in Fig. 3), right at the maximum of the upper-critical field, $B_{c2}^0(p)$, suggesting again a close connection of antiferromagnetism and superconductivity in AS-type $CeCu_2Si_2$. The estimated critical points inside the SC phase [$T_N(p, B = \text{const}) \rightarrow 0$; see above], which coincide rather well with the position of the maximum in $T_c(p)$, lie on the same straight line (dotted line in Fig. 3) as the critical points in the normal state. We note that the HF compound $CeRhIn_5$ exhibits a similar T-p phase diagram at $B = 0$ [7]. However, in $CeRhIn_5$ the AFM state is robust against the application of a magnetic field; even field-induced magnetism extending into the SC state is observed [19,20]. This behavior in a magnetic field is in strong contrast to the observations in $CeCu_2Si_2$.

To search for further evidence for the presence of a line of field-induced QCPs in the zero-temperature p-B plane, we analyzed $C_{el}(T)$ in the normal state. In the proximity of a QCP, strong deviations from Landau-Fermi-liquid (LFL) behavior are expected. Figure 4 shows the pressure evolution of C_{el}/T at $B = 6$ T on a $\sqrt{T}$ temperature scale. At ambient pressure, $C_{el}(T)/T$ increases as $\gamma_0 - a\sqrt{T}$ on lowering the temperature ($T \lesssim 1$ K), showing a clear deviation from $C_{el}(T)/T = \text{const}$ behavior expected for a

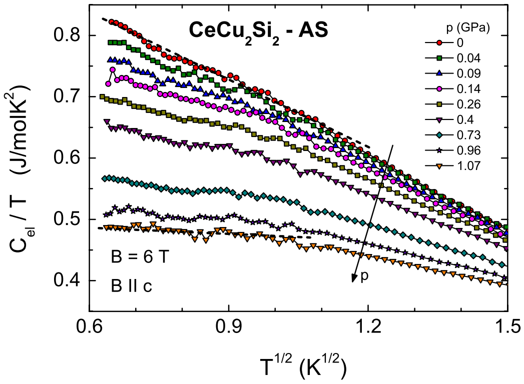

FIG. 4 (color online). Specific heat of AS-type $CeCu_2Si_2$ at $B = 6$ T ($B \parallel c$) for different pressures. C_{el}/T is plotted as a function of $\sqrt{T}$. The straight lines correspond to fits of $C_{el}(T)/T = \gamma_0 - a\sqrt{T}$ to the data. For clarity, only two lines for $p = 0$ and 1.07 GPa are indicated.

LFL. This temperature dependence of $C_{el}(T)$ has been predicted at a 3D AFM QCP [13,21]. Upon increasing pressure, the quantum critical component to the specific heat becomes smaller and smaller. At sufficiently high pressures ($p \gtrsim 1$ GPa), LFL behavior is observed. The strong deviations from LFL behavior at low pressures substantiate the proximity to a QCP. As illustrated in Fig. 3, upon increasing pressure at a constant B (e.g., $B = 6$ T), the distance to the line of QCPs in the p-B plane continuously increases, and, correspondingly, the quantum critical fluctuations become weaker until finally LFL behavior is recovered at $p \gtrsim 1$ GPa. We want to emphasize the fact that $T_c(p)$ in zero magnetic field is almost constant ($p > 0.1$ GPa), while the quantum critical component to the specific heat is strongly reduced upon increasing pressure.

In summary, we studied the magnetic and SC phase diagram of AS-type $CeCu_2Si_2$ as a function of external pressure, magnetic field, and temperature. The obtained phase diagram suggests a close connection of the underlying mechanisms leading to the formation of antiferromagnetism and superconductivity. Our results indicate the absence of long-range magnetic order in the SC phase and lead us to the conclusion that antiferromagnetism is expelled once superconductivity is established in AS-type $CeCu_2Si_2$. In the normal state, $T_N(B, p = \mathrm{const})$ can be continuously suppressed to zero temperature by increasing the magnetic field. Hence, we propose the existence of a line of field-tuned AFM QCPs in the zero-temperature p-B plane. The critical points obtained by extrapolating $T_N(p, B = \mathrm{const})$ from the normal into the SC state naturally extend this line. Furthermore, the positions of the maxima in the pressure dependence of $T_c(p, B = \mathrm{const})$, $p_{T_{c,\max}}(B)$, coincide with the critical line in accordance with the predictions for AFM spin-fluctuation-mediated superconductivity [15,16].

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