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LETTER TO THE EDITOR

Specific heat of heavy-fermion $CePd_2Si_2$ in high magnetic fields

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Abstract
We report specific heat measurements on the heavy-fermion compound $CePd_2Si_2$ in magnetic fields up to 16 T and in the temperature range 1.4–16 K. A sharp peak in the specific heat signals the antiferromagnetic transition at $T_N \sim 9.3$ K in zero field. The transition is found to shift to lower temperatures when a magnetic field is applied along the crystallographic a-axis, while a field applied parallel to the tetragonal c-axis does not affect the transition. The magnetic contribution to the specific heat below T_N is well described by a sum of a linear electronic term and an antiferromagnetic spin-wave contribution. Just below T_N, an additional positive curvature, especially at high fields, arises most probably due to thermal fluctuations. The field dependence of the coefficient of the low-temperature linear term, γ_0, extracted from the fits shows a maximum at about 6 T, at the point where an anomaly was detected in susceptibility measurements. The relative field dependences of both T_N and the magnetic entropy at T_N scale as $[1 - (B/B_0)^2]$ for $\boldsymbol{B} \parallel \boldsymbol{a}$, suggesting the disappearance of antiferromagnetism at $B_0 \sim 42$ T. The expected suppression of the antiferromagnetic transition temperature to zero makes the existence of a magnetic quantum critical point possible.

The heavy-fermion system $CePd_2Si_2$ has recently become a subject of considerable experimental and theoretical interest. This compound undergoes an antiferromagnetic transition at $T_N \sim 9$ K with a static moment of 0.6 μ_B at low temperature. Its spin configuration consists of ferromagnetic (110) planes with spins normal to the planes and alternating in direction along the spin axis.

The existence of a quantum critical point at a pressure of about 30 kbar, where the antiferromagnetic transition temperature is suppressed to zero, was established a long time ago [1]. It is, however, only recently that Grosche *et al* [2] discovered the appearance of superconductivity in a small pressure range in the vicinity of the quantum critical point. This pioneering work gave rise to a new wave of interest in this compound. Recently, several experimental studies of $CePd_2Si_2$ were performed both at ambient and high pressure on different samples and using different experimental techniques [3–6]. These works confirmed the emergence of superconductivity close to the critical pressure and gave insight into the nature of the superconducting ground state. It has also been shown that non-Fermi-liquid behaviour at low temperature appears close to the critical pressure. Such behaviour observed in resistivity, magnetization and specific heat measurements is a common trend in heavy-fermion compounds in the vicinity of a magnetic instability induced by application of hydrostatic pressure.

A different way to tune a heavy-fermion compound to a quantum critical point is by applying a high magnetic field. Field suppression of antiferromagnetism, like pressure, does not induce any disorder and, like pressure, opens up a new dimension in the phase diagram for study. Such a suppression of the magnetically ordered ground state by high magnetic field was found in $CeCu_{5.2}Ag_{0.8}$ [7], $YbCu_{5-x}Al_x$ [8, 9], $YbRh_2Si_2$ [10] and $CeCu_{5.2}Au_{0.8}$ [11]. In all these cases, unusual non-Fermi-liquid behaviour was also found near the magnetic quantum critical point. There are, however, other systems, e.g. $CePtSi_{0.4}Ge_{0.6}$ [7], that show Fermi-liquid behaviour upon field suppression of T_N to zero.

Two experimental investigations of $CePd_2Si_2$ at high magnetic field were recently reported. The first publication [12] reports magnetization measurements up to 28 T at 4.2 K with the magnetic field applied along both the crystallographic a- and c-axes. No anomalies have been seen for either direction of the magnetic field. The other work [7] presents specific heat measurements at 0, 13 and 28.9 T on a polycrystalline sample. At 28.9 T, the antiferromagnetic transition was found to be broadened, with both the peak in the specific heat and the transition mid-point suppressed to lower temperatures although the onset did not shift. This left the situation in this compound somewhat obscure, and no further high-field investigations have been reported.

We have, therefore, decided to re-examine the experimental situation in this material. We have performed specific heat measurements on a single crystal of $CePd_2Si_2$ in magnetic fields up to 16 T in the temperature range 1.4–16 K.

The specific heat measurements were performed on a single crystal of $CePd_2Si_2$, grown in a tri-arc furnace by the Czochralski method. Further details of the sample preparation are given elsewhere [13]. The sample, with a mass of 15 mg, was mounted on a small sapphire plate. A carbon film with four contacts made of phosphor bronze was painted on the other side of the plate. The whole set-up including two thermometers and a heater was sealed inside a vacuum sample chamber. A carbon glass resistive thermometer was used to regulate the temperature inside the sample chamber. The temperature was measured by a Cernox thermometer calibrated in field. A thermal relaxation technique was used to measure the specific heat [14]. To extract the specific heat of the sample, the contribution of the sapphire, electrical wires and grease used as an adhesive was measured and subtracted. Magnetic fields up 16 T were generated by a superconducting coil. The temperature was varied from 1.4 to 16 K using a standard variable-temperature insert (VTI).

Figure 1 shows the zero-field specific heat. The data are in good agreement with previous measurements [13, 15]. The antiferromagnetic transition manifests itself in a sharp, almost step-like increase of the specific heat at 9.3 K. The magnetic contribution, C_{mag}, to the specific heat is obtained by subtracting the data for the non-magnetic reference system $LaPd_2Si_2$ [16] (also shown in figure 1). At low temperature, $LaPd_2Si_2$ shows the classical behaviour $C = \gamma_r T + \beta_r T^3$ with $\gamma_r = 6.0$ mJ mol^{-1} K^{-2} and $\beta_r = 0.267$ mJ mol^{-1} K^{-4}. This is to be compared with $C/T = 160$ mJ mol^{-1} K^{-2} found for $CePd_2Si_2$ at 1.4 K, the lowest temperature of our measurements (see, however, the discussion below for fits of C/T as $T \to 0$).

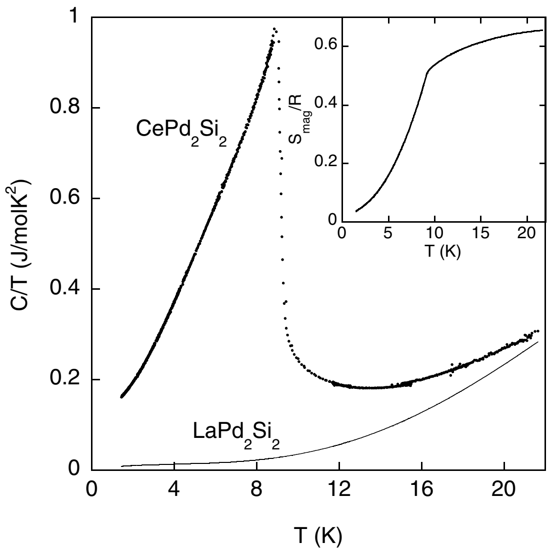

Figure 1. The specific heat C/T of $CePd_2Si_2$ in zero magnetic field. The anomaly at $T_N = 9.3$ K signals the antiferromagnetic transition. For comparison, the specific heat of the non-magnetic reference system $LaPd_2Si_2$ is shown [16]. The inset shows the magnetic entropy, S_{mag}/R, also in zero field.

Above the antiferromagnetic transition, C_{mag}/T increases with temperature decrease due to the Kondo spin fluctuations. Below T_N, $C_{mag}(T)$ can be described in terms of spin fluctuations and an additional contribution from the antiferromagnetic spin waves [13]. We have integrated C_{mag}/T to obtain the magnetic entropy. Since the lowest temperature of our measurements was 1.4 K, we extrapolated the data below 1.4 K using the empirical temperature dependence $C = \gamma T + \beta T^3$ discussed below. The calculated magnetic entropy is shown in the inset of figure 1. The entropy at the transition temperature reaches about 75% of $R \ln 2$, the value expected for a doublet ground state [13]. This is not necessarily surprising and is likely to imply that the ordered moment in the antiferromagnetic phase is somewhat compensated by the Kondo interactions. At 22 K, the highest temperature of the zero-field measurement, the magnetic entropy reaches 95% of $R \ln 2$.

When a magnetic field is applied parallel to the crystallographic a-axis, the transition gradually moves to lower temperatures, remaining, however, as sharp as in zero field, as shown in figure 2. Conversely, a magnetic field of 14 T applied along the tetragonal c-axis was not found to affect the transition temperature (see the inset of figure 2). This implies the existence of strong magnetic anisotropy between the basal plane and the tetragonal c-axis of the compound. Such anisotropy is consistent with the magnetic structure of the material with its spins aligned in the basal plane. This anisotropy explains the result of a previous report obtained on a polycrystal [7], where, at 29 T, the transition was found to be much broader with the peak in C/T suppressed to about 5 K but without any shift of the onset. Indeed, in the case of a polycrystal, the part of the sample more or less aligned with the field along the c-axis shows no change and that is why the onset of the transition does not move. On the other hand, the other field directions suppress the transition which leads both to a shift of the specific heat peak and the transition broadening.

We have tried to fit the data below T_N with the commonly used empirical fitting functions $C_{mag} = \gamma T + A \exp(-\Delta/T)$ for a spin gap [17] and $C_{mag} = \gamma T + \beta T^3$ for antiferromagnetic

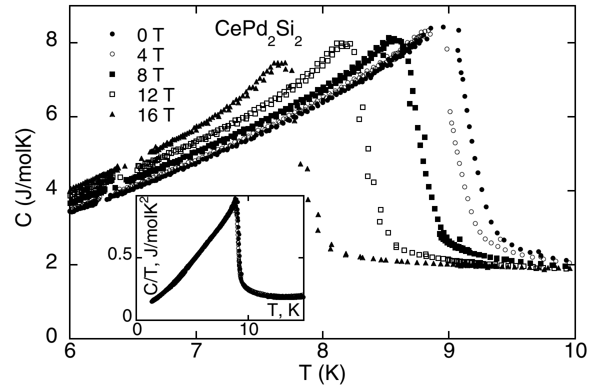

Figure 2. The specific heat C of $CePd_2Si_2$ around the antiferromagnetic phase transition in magnetic fields of 0, 4, 8, 12 and 16 T ($\boldsymbol{B} \parallel \boldsymbol{a}$). Both the transition temperature, T_N, and the jump in the specific heat decrease with magnetic field. The inset shows the transition in zero field (closed circles) and at 14 T applied along the c-axis (open circles). The two curves are practically indistinguishable, as the transition does not shift in fields parallel to the c-axis.

spin waves [13]. While both of them fit well the low-temperature part of the data, neither of them succeeded in providing a satisfactory fit over the whole temperature range. We found, however, that the magnetic contribution to the specific heat is well described by a model composed of a linear electronic contribution as above, $\gamma_0 T$, plus a term that accounts for the contribution from antiferromagnetic spin waves with the dispersion relation $\omega = \sqrt{\Delta^2 + Dk^2}$ [18]:

$$C_{\rm mag} = \gamma_0 T + \alpha \Delta^{7/2} T^{1/2} {\rm e}^{-\Delta/T} [1 + (39/20)(T/\Delta) + (51/32)(T/\Delta)^2]. \quad (1)$$

Here Δ is the spin-wave gap, and α is related to the spin-wave stiffness D by $\alpha \propto D^{-3/2}$. As shown in figure 3 for $B = 0$ and 16 T, the above equation yields a good fit to the data, except for a small temperature range just below T_N, which was ignored when fitting the data. The failure of the fit over this small temperature range is due to the existence of a positive curvature just below T_N. This curvature becomes gradually stronger with field, as can be seen in figure 3. We associate this curvature with thermal fluctuations that are expected to play a role in the vicinity of the transition, and to become stronger in magnetic field.

The field dependence of the low-temperature electronic contribution to the specific heat extracted from the fits is shown in figure 4. As one can see, the low-temperature Sommerfield coefficient, γ_0, passes through a maximum at about 6 T before starting to decrease monotonically. This maximum matches a clear anomaly observed in susceptibility measurements when the magnetic field was also applied along the crystallographic a-axis [19]. The anomaly can be associated with a spin-flop process. Indeed, the behaviour of γ_0 in a magnetic field that we find here is very similar to that of the coefficient A of the T^2-term of the resistivity found by McDonough and Julian in $CePb_3$ around a spin-flop transition [20]. Note that the coefficient A also reflects the many-body enhancement, the ratio A/γ_0 having a universal value in heavy-fermion systems, as pointed out by Kadowaki and Woods [21].

The field-dependent values of T_N have been determined by an equal-entropy construction with an ideal step transition. Figure 5 shows the field dependence of the transition temperature and the magnetic entropy at T_N. Both follow a simple scaling relation $[1 - (B/B_0)^2]$. Here B_0 corresponds to the field necessary to suppress the antiferromagnetic order, and is found to be (41.5 ± 0.6) T. This scaling relation explains quantitatively why no anomalies were observed in the magnetization measurements up to 28 T at $T = 4.2$ K [12]. According to the above formula, at $T = 4.2$ K the transition should occur at about 31 T for $\boldsymbol{B} \parallel \boldsymbol{a}$, while in [12]

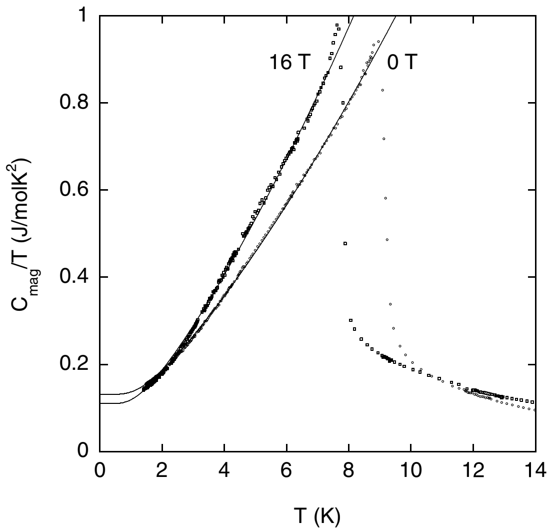

Figure 3. The magnetic specific heat C_{mag}/T of $CePd_2Si_2$ in zero field and in a magnetic field of 16 T. The solid curves correspond to a fit taking into account both the electronic contribution and antiferromagnetic spin waves (equation (1)). Parameters obtained from the fit are: $\gamma_0 = (0.131 \pm 0.003)$ J mol^{-1} K^{-2}, $\alpha = (1.7 \pm 0.1) \times 10^{-3}$ J mol^{-1} K^{-4}, $\Delta = (4.6 \pm 0.2)$ K for $B = 0$ and $\gamma_0 = (0.110 \pm 0.006)$ J mol^{-1} K^{-2}, $\alpha = (2.5 \pm 0.3) \times 10^{-3}$ J mol^{-1} K^{-4}, $\Delta = (4.2 \pm 0.3)$ K for $B = 16$ T.

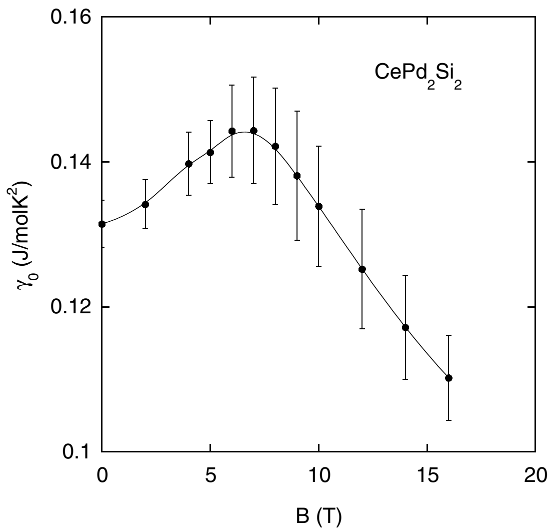

Figure 4. The magnetic field dependence of the coefficient of the electronic linear term of the specific heat extracted from the fits by equation (1). The weak maximum suggests the possibility of a magnetic phase transition, presumably a spin flop. The line is a guide for the eyes. Error bars correspond to the standard errors obtained from the fit.

the highest applied field was only 28 T. The same scaling behaviour was found for another heavy-fermion system, URu_2Si_2 [22], where the value of B_0 was found to be in relatively good agreement with the results obtained from resistivity, thermal expansion and magnetization measurements.

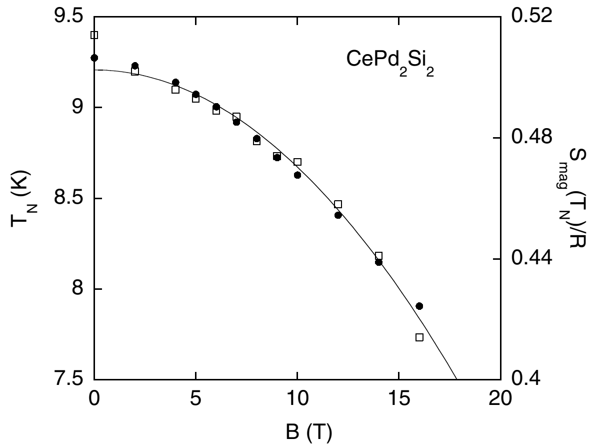

Figure 5. The field dependence of the transition temperature T_N (solid circles, left axis) and the magnetic entropy at T_N (open squares, right axis). The curve represents a scaling function of the form $T_{N0}[1 - (B/B_0)^2]$ with $B_0 = 41.5$ T.

In conclusion, we have shown that the antiferromagnetic transition in $CePd_2Si_2$ shifts to lower temperatures in magnetic fields applied in the basal plane. The absence of influence of the magnetic field parallel to the tetragonal c-axis on the transition temperature points to the existence of a strong magnetic anisotropy, which is a common trend for heavy-fermion compounds. Analysis of the field dependence of T_N and the magnetic entropy suggests the suppression of the antiferromagnetic ground state at about 42 T, raising the possibility of the existence of a quantum critical point at this field. This might make this compound a new member of a growing family of heavy-fermion antiferromagnets which can be tuned by a magnetic field through the magnetic quantum critical point, where the Néel temperature vanishes. This prediction calls for further experiments at higher, probably pulsed fields. The magnetic specific heat below T_N is best described by the sum of an electronic contribution and antiferromagnetic spin waves. The fit, however, breaks down just below T_N where thermal fluctuations give rise to a positive curvature. The low-temperature coefficient of the electronic term of the specific heat extracted from the fits is found to have a maximum at about 6 T. This suggests the existence of a magnetic transition, presumably a spin flop, the idea being also supported by the results of the magnetization measurements [19].

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