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LETTER TO THE EDITOR

Double specific heat anomaly of the superconducting state of $CePt_3Si$

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Abstract
$CePt_3Si$ is the first heavy fermion superconductor without a centre of symmetry. Below an antiferromagnetic transition at 2.25 K superconductivity occurs at $T_C = 0.73$ K. The non-centrosymmetric structure is expected to have significant impact on the nature of the superconducting order parameter. This may be a reason for the unusual shape of the superconducting transition observed in the specific heat. We present specific heat data which clearly show the existence of a double anomaly in the superconducting state, signalling two consecutive phase transitions, as in the uranium based heavy fermion superconductors $U_{1-x}Th_xBe_{13}$ and UPt_3.

(Some figures in this article are in colour only in the electronic version)

The recently discovered heavy fermion antiferromagnetic (AFM) superconductor $CePt_3Si$ has attracted much attention due to the lack of a spatial inversion centre [1]. Long-range antiferromagnetic order exists below $T_N = 2.25$ K while superconductivity occurs at $T_C = 0.73$ K. Neutron scattering experiments [2] revealed an ordered Ce moment of about 0.2 μ_B with a propagation vector $\vec{k} = (0, 0, 1/2)$, doubling the magnetic unit cell with respect to the crystallographic one along the $\vec{c}$-axis. A coexistence of both superconductivity and long range magnetic order on a microscopic scale was evidenced by μSR spectroscopy [3]. The NMR relaxation rate $1/T_1$ shows unexpected features which were not found either in conventional or in heavy fermion superconductors [4]. Theoretically, the superconducting state is not yet understood in detail. A spin triplet pairing scenario is expected to require inversion symmetry while the large value of the upper critical field $H_{C2} \approx 5$ T of $CePt_3Si$ seems to be inconsistent with spin–singlet pairing. It has been shown, however, that spin–orbit coupling effects in non-centrosymmetric systems will lead to a mixed spin singlet and triplet state, able to account for the high upper critical field [1, 5, 6].

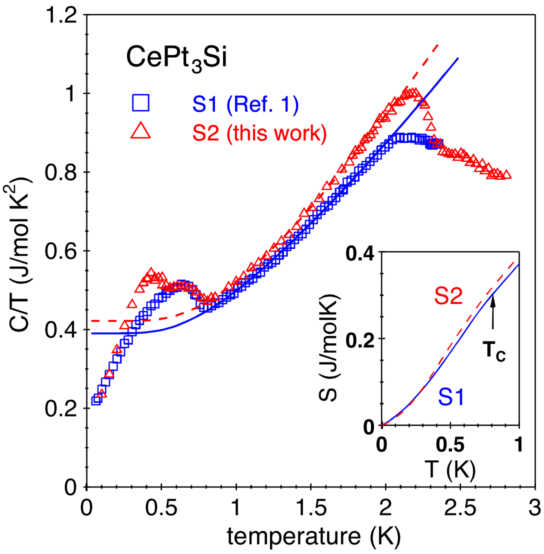

Figure 1. Temperature dependent specific heat divided by temperature of two different $CePt_3Si$ samples (S1, S2). The solid and dashed curves are least squares fits taking into account the AFM contribution, cf equation (1). The inset exhibits the temperature dependent entropy S of the nonmagnetic contribution for both samples below 1 K (see the text).

Up to now there exists no experimental evidence for a really sharp superconducting transition; even resistivity measurements provide a transition width of more than 0.1 K at $T_C^{onset} = 0.83$ K [7]. A closer inspection of the specific heat anomaly below 0.8 K [1] indicates an additional shoulder-like structure, which might signal two consecutive phase transitions. Ac-susceptibility data on polycrystalline samples reveal a small local maximum between 0.4 and 0.5 K of which the origin is still unknown [8]. In addition, pressure dependent resistivity measurements indicate two distinct superconducting phases below 0.5 K [7]. In this paper we will present specific heat measurements on polycrystalline $CePt_3Si$, which unveil a clear double anomaly in the superconducting state. This finding elucidates the unknown experimental features and will shed a new light on the theoretical discussion.

Crystalline $CePt_3Si$ was prepared in a similar manner as described in [1]. Compared to sample S1 (S1, reference [1]), the new sample S2 was annealed at slightly higher temperatures (920 °C) before quenching in cold water. X-ray diffraction measurements were carried out using a Stoe-IPDS2 diffractometer. Sample S1 reveals several single-crystal domains, whereas in the diffraction pattern of sample S2 the formation of weak powder rings beside the Bragg reflections was observed, thus indicating a very high mosaicity. This suggests a more polycrystalline character of the microstructure of S2 and thus stress and strain seem to be diminished in this sample.

Figure 1 shows the temperature dependence of the specific heat for the more polycrystalline sample S2 in comparison to that of [1]. There are two fundamental different features observed in sample S2. On the one hand the AFM transition at $T_N = 2.25$ K is more pronounced. $\Delta C/T = 0.35$ J mol^{-1} K^{-2} is larger by a factor of two than that for sample S1 and the characteristic temperature dependence below T_N ($C \propto T^3$) is valid over an expanded temperature range, corroborating antiferromagnetic ordering (AFM). A further proof of AFM is revealed from a model of Continentino [9] which can be applied to specific heat data well below T_N:

$$C_{mag} = g\Delta^{7/2}T^{1/2}\exp(-\Delta/T)\left[1 + \frac{39}{20}\left(\frac{T}{\Delta}\right) + \frac{51}{32}\left(\frac{T}{\Delta}\right)^2\right]. \quad (1)$$

Equation (1) is based on antiferromagnetic magnons with a dispersion relation $\omega = \sqrt{\Delta^2 + D^2k^2}$, where Δ is the spin-wave gap and D is the spin-wave velocity; $g \propto 1/D^3 \propto 1/\Gamma^3$ and Γ is an effective magnetic coupling between the Ce ions. Least squares fits of equation (1) to both sets of data below T_N (solid (S1) and dashed (S2) curve, figure 1) reveal $\Delta \approx 2.7$ and 3.0 K, respectively, a reasonable gap value with respect to the ordering temperature. An extrapolation of these fits towards zero yields Sommerfeld values of 390 and 420 mJ mol^{-1} K^{-2} for S1 and S2, respectively. These values are slightly larger than those derived from the simple T^3 extrapolation, 367 and 375 mJ mol^{-1} K^{-2}, respectively. The entropy deduced for S2 from these measurements at $T = T_N$ is about 6% higher than that of S1.

On the other hand, there are two anomalies below the superconducting transition, similar to those known from the uranium based heavy fermion superconductors $U_{1-x}Th_xBe_{13}$ [10] and UPt_3 [11]. Following the same procedure as in [1], we find two idealized jumps at $T_{C1} = 0.8 \pm 0.01$ K and $T_{C2} = 0.54 \pm 0.01$ K. Surprisingly, $\Delta C/T_{C1} \simeq 0.1$ J mol^{-1} K^{-2} is of the same size as that of the S1 sample. Both transitions together exceed the size of $(\Delta C_1/T_{C1} + \Delta C_2/T_{C2}) \simeq 0.18$ J mol^{-1} K^{-2}. To estimate the balance of the nonmagnetic entropy S (inset of figure 1) between the superconducting ($S_{SC} = (\int C(T)/T\,dT - \gamma_R T_{C1})$) and normal state ($S_N = (\gamma_n - \gamma_R)T_{C1}$) one has to take into account a residual γ_R which marks a normal state contribution in the superconducting state not associated with superconductivity. An extrapolation of the linear temperature dependence of C/T below T_{C2} yields $\gamma_R = 0.17$ and 0.12 J mol^{-1} K^{-2} for samples S1 and S2, respectively. For both samples the basic requirement of entropy balance equals $S_{SC}/S_N \approx 0.92$, slightly smaller than expected for normal type II superconductors. Furthermore, for sample S2 the superconducting contribution to the entropy is larger by a factor of 1.4 than for S1.

It is interesting to note that the low temperature properties are in some respect similar to the prominent heavy fermion superconductor UPt_3 [11–13]: (i) the superconducting transition displays a double structure in the specific heat, (ii) C/T shows below T_{C2} an unusual linear temperature dependence and (iii) an AFM transition is found about 5 K. But in contrast to UPt_3, where the AFM spin state is of a short range, fluctuating nature with no detectable anomaly in the specific heat, $CePt_3Si$ develops long range magnetic order.

Further information about the two anomalies can be deduced by magnetic field dependent measurements of the specific heat (figure 2). For the upper transition we confirm the large change of $dB_{C2}/dT \simeq -9$ T K^{-1} also reported for sample S1 [1]. The lower transition is more sensitive to the magnetic field:

(i) $\Delta C_2/T_{C2}$ nearly vanishes at $B = 1$ T, while the shape of the specific heat curve looks like that of sample S1 in zero field.
(ii) The initial slope $dB_{C2}/dT \simeq -2.5$ T K^{-1} at T_{C2} is much smaller than for the upper transition (inset of figure 2).

The different T_C-dependences of $\Delta C/T_C$ for the upper and lower transitions indicate that the origins of the two anomalies are based upon two different superconducting states.

Summarizing, based on specific heat measurements, we have shown for the first time two superconducting phase transitions in the Ce-based heavy fermion superconductor $CePt_3Si$. The phase diagram looks like that of other well studied U based systems, like $U_{1-x}Th_xBe_{13}$ and UPt_3, while the mechanism of superconductivity might be very different. Experimentally our findings give a hint that the forming of the lower transition becomes more pronounced

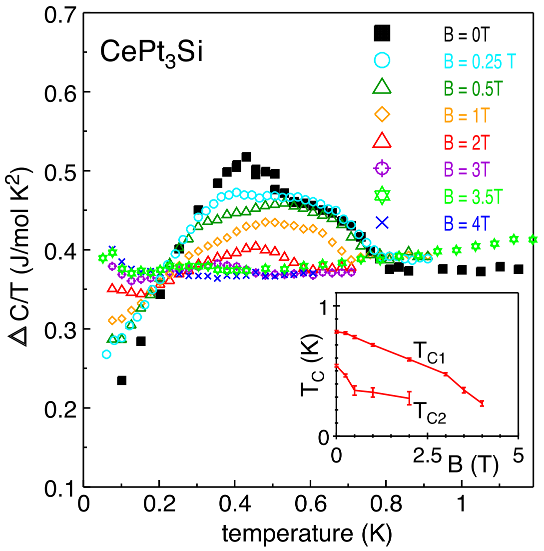

Figure 2. Temperature dependent $\Delta C/T$ of $CePt_3Si$ (S2) in various magnetic fields (the AFM contribution was subtracted; see figure 1). The inset shows the magnetic field dependence of T_{C1} and T_{C2}.

with an enhanced development of the long range AFM order at 2.25 K or the reduction of γ_R or by minimizing the internal structural stress due to the more polycrystalline character. The lack of an inversion centre establishes $CePt_3Si$ in a prominent role among the unconventional superconductors. The theoretical challenge will mainly culminate in the question of which kind of order parameter might govern these two exotic superconducting phases.

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