LOW TEMPERATURE SPECIFIC HEAT OF $PrOs_4Sb_{12}$*

R. Vollmer[a], A. Faisst[a], C. Pfleiderer[a], H. v.Löhneysen[a,b]
E.D. Bauer[c] and M.B. Maple[c]

[a]Physikalisches Institut, Unversität Karlsruhe, 76128 Karlsruhe, Germany
[b]Forschungszentrum Karlsruhe, Institut für Festkörperphysik
76021 Karlsruhe, Germany
[c]Department of Physics and Institute of Pure and Applied Physical Sciences
University of California San Diego, La Jolla, CA 92093-0319, USA

(Received July 10, 2002)

We report measurements of the magnetic field dependence of the low temperature specific heat of single crystal samples of the heavy-fermion superconductor $PrOs_4Sb_{12}$. The specific heat anomaly at T_c resides on a large Schottky anomaly and exhibits a double peak structure similar to the behaviour observed in UPt_3. For $B > 2$ T, the Schottky anomaly becomes increasingly suppressed. At low T, a maximum develops for $B > 4.5$ T which sharpens considerably and shifts to higher T with increasing B corresponding to a feature in the electrical resistivity. This suggests a field tuned $T = 0$ phase transition to a high field phase just above the suppression of superconductivity.

PACS numbers: 74.70.Tx, 65.40.–b, 71.27.+a, 75.30.Mb

The rich variety of ground states exhibited by intermetallic compounds in the filled skutterudite structure ranges from Kondo insulators to superconductivity. An especially interesting material is $PrOs_4Sb_{12}$ which appears to be the first Pr based heavy fermion superconductor [1]. Measurements of the electrical resistivity and the magnetisation show both of the two hallmarks of superconductivity, zero resistivity and the Meissner effect. The superconductivity is a bulk effect as previously inferred from a fairly broad anomaly at T_c, which resides on top of a Schottky anomaly. This Schottky anomaly and the magnetic susceptibility suggest that the crystal electric field ground state of Pr is a non-magnetic Γ_3 doublet, providing a possible opportunity to observe two channel Kondo physics.

* Presented at the International Conference on Strongly Correlated Electron Systems, (SCES 02), Cracow, Poland, July 10–13, 2002.

$PrOs_4Sb_{12}$ crystallizes in the cubic $LaFe_4P_{12}$-type structure (space group $Im3$). The single crystalline samples of $PrOs_4Sb_{12}$ have been grown from a Sb flux. The specific heat has been measured using a quasi-adiabatic heat pulse technique in a ^{4}He refrigerator in magnetic fields up to 14 T and in a dilution refrigerator in magnetic fields up to 6.65 T down to 100 mK. An aggregate of five small single crystals was studied abd compared with a large single crystalline piece. The specific heat of the single piece was 9 % lower, probably due to a small quantity of Sb inclusions, but otherwise in perfect agreement.

Fig. 1 shows the specific heat in the form C/T as a function of temperature T for low magnetic fields. With decreasing temperature C/T exhibits a maximum at $T \approx 2$ K and decreases at lower temperatures. The superconducting transition at $T_c = 1.85$ K is associated with a pronounced anomaly in the specific heat. Data for $B = 0$ and $T > T_c$ up to 10 K can be described by three contributions (solid line): *(i)* an electronic term $C/T = \gamma$ with $\gamma = 313\,\mathrm{mJ/molK^2}$, *(ii)* a phononic part with a Debye temperature of $\Theta_D = 165$ K, and *(iii)* a Schottky anomaly due to the crystal electric field splitting of the Pr^3H_4 ground state into a Γ_3 ground state doublet and a Γ_5 triplet with a separation of $\Delta = 7.0$ K. The high absolute value of the specific heat together with the coefficient of the Schottky peak (1 Pr/f.u.) shows that the samples are of high quality.

The inset of Fig. 1 shows the specific heat C/T as a function of T near T_c. Two pronounced anomalies are resolved, where the upper transition corresponds to the zero resistance and Meissner transition temperatures. The double transition has been observed on the aggregate of five crystals and the individual single crystalline piece, ruling out sample inhomogenities as a possible cause. The anomaly in C/T is suppressed for $B >\sim 0.5$ T. A double superconducting transition was previously reported in reference [2].

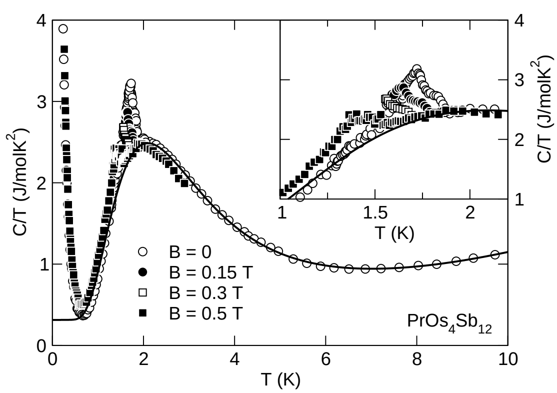

Fig. 1. Specific heat C/T *versus* temperature T in magnetic fields up to 0.5 T. The inset displays the specific heat near the superconducting transition.

The specific heat for $B = 0$ near T_c after subtraction of the phonon and the Schottky contribution is shown as $\Delta C/T$ *versus* T in Fig. 2. The solid line is an equal entropy construction showing that the two transitions are of equal height. The two superconducting transition temperatures are estimated to be $T_{c1} = 1.75$ K and $T_{c2} = 1.85$ K. The ratio $\Delta C_{sc}/\gamma T_c \approx 3$ exceeds the BCS weak coupling value $\Delta C_{sc}/\gamma T_c = 1.43$ by over a factor of two, where ΔC_{sc} is the total height of both superconducting jumps taken together.

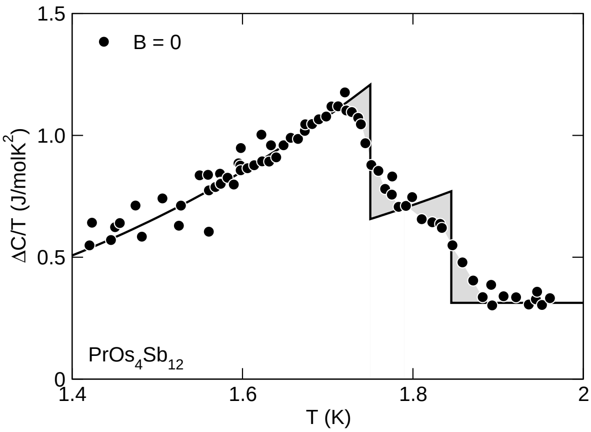

Fig. 2. Electronic contribution to the specific heat $\Delta C/T$ *versus* temperature T. The solid curve shows an entropy conserving construction as explained in the text.

The specific heat C as a function of T in magnetic fields up to 14 T is shown in Fig. 3. The Schottky anomaly is strongly suppressed by the magnetic field as expected for a Zeeman splitting of the f electron states. However, a quantitative account of the Zeeman splitting of the f electron levels in a magnetic field requires knowledge of the interactions between the f electrons. For $T < 1\,\mathrm{K}$ a pronounced maximum for $B > 4.5\,\mathrm{T}$ is resolved that shifts from $T = 0.7$ K for $B = 5$ T to $T = 1$ K for $B = 6.65$ T and sharpens considerably. This maximum corresponds to a kink in the resistivity [3] and may be related to a field tuned $T = 0$ phase transition near $B \sim 4.5\,\mathrm{T}$. At the lowest T, a strong increase of C is observed with decreasing T. This increase may be due to the splitting of the doublet ground state in a magnetic field and additional contributions from hyperfine enhanced Zeeman splitting of the Pr nuclear levels.

In summary, we have examined the superconducting transition in $PrOs_4Sb_{12}$ and observed a double transition at the onset of superconuc-tivity, possibly indicating unconventionel superconductivity. The specific heat in the normal state is influenced by interactions between the localised

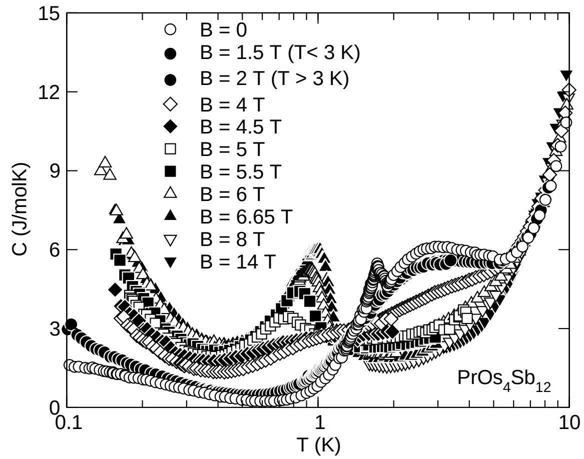

Fig. 3. Specific heat C *versus* temperature T in magnetic fields up to 14 T. The high T Schottky anomaly is rapidly suppressed up to 14 T. A pronounced anomaly, indicating an ordered state stabilized in high field, is resolved at low T.

f electrons of Pr, possibly related to a field induced phase transition for $B > 4.5\,\mathrm{T}$, just above $H_{c2} = 2.2\,\mathrm{T}$, but may be unambiguously explained by a Γ_3 doublet ground state and Γ_5 low lying triplet state.

REFERENCES

[1] E.D. Bauer, N.A. Frederick, P.-C. Ho, V.S. Zapf, M.B. Maple, *Phys. Rev.* **B65**, 100506(R) (2002).

[2] M.B. Maple *et al.*, *J. Phys. Soc. Jpn.* **71**, 23 (2002).

[3] P.-C. Ho *et al.*, Proceedings of the International Conference on Physical Properties in High Magnetic Fields, Santa Fe NM, USA, 2002.