B-T Phase Diagram of $PrOs_4Sb_{12}$ Studied by Low-Temperature Thermal Expansion and Magnetostriction

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The recently discovered skutterudite superconductor (T_c = 1.8 K) $PrOs_4Sb_{12}$ is considered to be the first Pr-based heavy-fermion (HF) superconductor [1]. The analysis of the temperature dependence of the susceptibility [1,2] and specific heat [3] suggests a nonmagnetic Γ_3 ground state separated from a first excited doublet at 6 K. Thus, the quadrupolar Kondo effect, arising from the scattering of the conduction electrons of the quadrupolar moment [4], might give rise to a HF ground state, although no indications for Non-Fermi liquid behavior expected for the quadrupolar Kondo scenario, are found. Evidence for a mass enhancement of about 50 arises from i) the extrapolated Sommerfeld coefficient $\gamma_0(T\rightarrow 0)$ = 300 mJ/molK^2 of the normal-state specific heat [3] and ii) the initial slope of the upper critical magnetic field $(\partial B_{c2}/\partial T)_{T_c} = -2$ T/K [1]. Furthermore, specific heat measurements revealed a double peak structure at T_c [3] that, if intrinsic, would indicate two superconducting phase transitions, as has been observed, e.g., in UPt_3 [5].

Below, we report on thermal expansion and magnetostriction measurements on single crystals of $PrOs_4Sb_{12}$ performed along the cubic [100] direction. The thermal expansion coefficient α and the magnetostriction coefficient λ are defined as $\alpha = l^{-1}(\partial l/\partial T)$ and $\lambda = l^{-1}(\partial l/\partial B)$, respectively, where l denotes the sample length. For the thermal expansion a high-resolution capacitive dilatometer made from silver was used in fields up to 8 T, whereas the magnetostriction has been measured with the aid of the new dilatometer described in the section “experimental development”.

The low temperature thermal expansion coefficient α of $PrOs_4Sb_{12}$ is displayed in Fig. 1. In zero magnetic field, two phase transitions can be resolved at $T_{c1} = (1.82 \pm 0.02)$ K and $T_{c2} = (1.72 \pm 0.02)$ K (cf. inset of Fig. 1). Unfortunately, one cannot follow both transitions as a function of an applied magnetic field, because their signatures in thermal expansion weakens substantially in $B > 0$. At the moment it is not clear, whether the two phase transition anomalies found in thermal expansion and specific heat [3] are intrinsic or arise due to sample inhomogeneities. Assuming two intrinsic superconducting transitions, we could use the Ehrenfest-relation $(\partial T_{c1,2}/\partial p)_{p\rightarrow 0} = V_{mol}T_{c1,2}\cdot\Delta\beta/\Delta C$, where $\Delta\beta$ and ΔC denote the discontinuities in the volume thermal expansion and specific heat, respectively, to calculate the pressure, p, dependences of the two superconducting transitions at T_{c1}

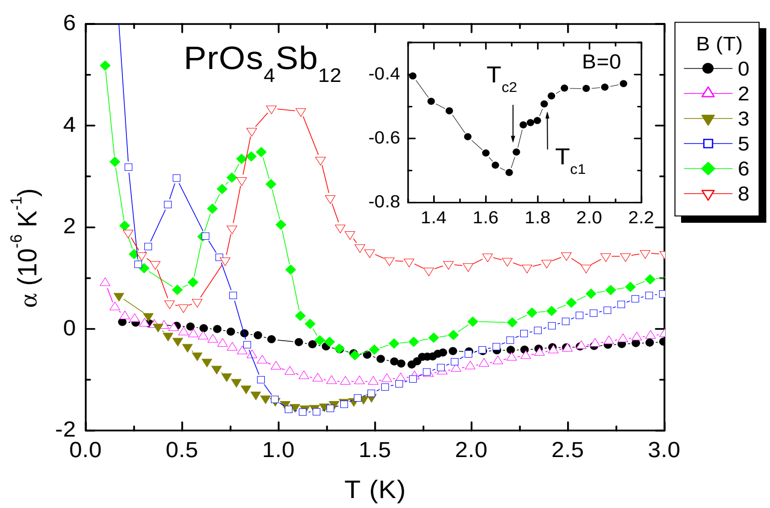

***Fig. 1:** Thermal expansion coefficient α vs T of $PrOs_4Sb_{12}$ at varying magnetic fields. Inset displays α(T) near the superconducting phase transitions indicated by the arrows.*

and $\boldsymbol{T}_{c2}$ in the limit $\boldsymbol{p} \rightarrow 0$. We find $\partial \boldsymbol{T}_{c1}/\partial \boldsymbol{p}$ = (180 ± 60) mK/GPa. This value is consistent with that obtained from resistivity measurements under hydrostatic pressure (150 mK/GPa) [2]. Thus, although the origin for the splitting of the superconducting transition is yet unclear, our measurements strongly suggest it to be intrinsic.

The low-$\boldsymbol{T}$ normal-state thermal expansion behavior is dominated by a negative contribution below 4 K for $\boldsymbol{B} = 0$. Upon increasing the field this minimum structure becomes more pronounced and the minimum temperature $\boldsymbol{T}_{min}$ shifts to lower $\boldsymbol{T}$. This feature corresponds to the Schottky anomaly observed in the specific heat [3,5] which is caused by the crystal electric field (CEF) splitting of the Pr^{3+} states. Indeed, in the field range $0 < \boldsymbol{B} < 4$ T our thermal expansion data can be fit by a Schottky anomaly with a characteristic energy splitting $\Delta(\boldsymbol{B})$ as well, if we assume a linear-in-$\boldsymbol{T}$ variation of the Grüneisen ratio between thermal expansion and specific heat [6]. The magnetic field dependence of the so-derived $\Delta(\boldsymbol{B})$ is well explained by the Zeeman-effect on the proposed CEF scheme with the Γ_3 ground state and Γ_5 first excited state [2].

At higher magnetic fields a low-$\boldsymbol{T}$ upturn becomes visible in $\alpha(\boldsymbol{T})$. Since nuclear contributions cannot lead to such large effects in the thermal expansion, this anomaly is most likely caused by the Zeeman splitting of the Γ_3 ground state doublet. Thus, our results strongly support the proposed CEF scheme with a Γ_3 doublet ground state.

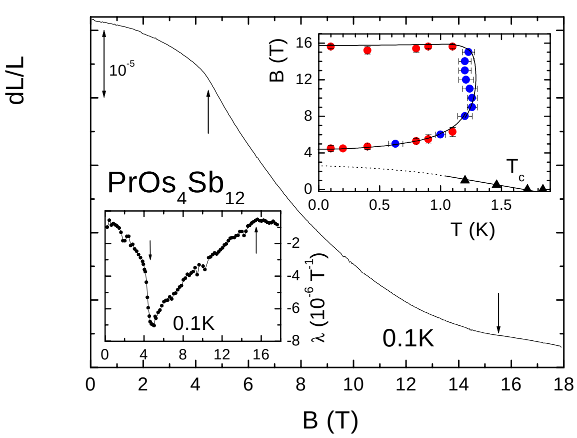

Fig. 2: Relative change dL/L vs B of $PrOs_4Sb_{12}$ at T = 0.1 K. Lower inset shows magnetostriction coefficient λ at 0.1 K. Arrows mark position of phase transitions. Upper inset displays B-T phase diagram as determined from thermal expansion (blue symbols) and magnetostriction (red symbols). Dotted line displays upper critical field as derived from electrical resistivity measurements [2].

As shown in Fig. 1, a field-induced phase transition is observed for $\boldsymbol{B} \geq 5$ T, whose transition temperature increases with increasing magnetic field. Corresponding phase transition anomalies are also observed in specific heat [3] and electrical resistivity measurements [2]. In order to determine the $\boldsymbol{B}$-$\boldsymbol{T}$ phase diagram of $PrOs_4Sb_{12}$, we performed magnetostriction measurements at different temperatures. The main part of Fig. 2 shows the relative length change as a function of $\boldsymbol{B}$-field obtained at 0.1 K. Two distinct anomalies are resolved at 4.5 T and 15.5 T. Whereas the lower one manifests itself in a broadened jump in the magnetostriction coefficient λ(B), indicative for a second order phase transition, a much weaker change is observed at the high-field anomaly (cf. lower inset of Fig. 2). The phase diagram derived from thermal expansion and magnetostriction measurements is shown in the upper inset of Fig. 2. The upper boundary of the new phase is rather field independent for temperatures below 1 K.

So far the nature of the high-field phase has not been determined. From the proposed CEF scheme a crossing of the lowest excited Γ_5 triplet with the upper doublet of the Γ_3 ground state occurs at 4.5 T, the same field at which the new phase develops [3]. This suggests that the formation of the high field phase is related to the Γ_5 quadrupolar moment. We note that the signature of the phase transition in specific heat resembles that observed at the antiferroquadrupolar (AFQ) ordering transition in $PrPb_3$ [8]. However, in $PrPb_3$ AFQ order exists already at $\boldsymbol{B} = 0$, whereas for $PrOs_4Sb_{12}$ a field larger than 4.5 T is required to induce the ordered state.

To summarize we have studied $PrOs_4Sb_{12}$ by thermal expansion and magnetostriction measurements in magnetic fields up to 18 T. The thermodynamic analysis provides evidence for two intrinsic superconducting phase transitions at zero magnetic field. By applying magnetic fields we find that the Zeeman splitting of the CEF scheme removes the degeneracy of the Γ_3 ground state and fluctuations related to this degeneracy. The latter might be important for the formation of superconductivity [3]. At $\boldsymbol{B} \geq 4.5$ T, where CEF level crossing occurs, a high-field ordered phase develops which is most likely of AFQ origin.

References

[1] ***E.D. Bauer, N.A. Frederick, P.-C. Ho, V.S. Zapf, M.B. Maple,*** Phys. Rev. B **65**, 100605(R) (2002).
[2] ***M.B. Maple, P.-C. Ho, V.S. Zapf, N.A. Frederick, E.D. Bauer, W.M. Yuhasz, F.M. Woodward, J.W. Lynn,*** Acta Phys. Pol. B **32**, 3291 (2002).
[3] ***R. Vollmer, A. Faißt, C. Pfleiderer, H. v. Löhneysen, E.D. Bauer, P.-C. Ho, M.B. Maple,*** cond-mat/0207225 (2002).
[4] ***D.L. Cox,*** Phys. Rev. Lett. **59**, 1240 (1987).
[5] ***R.A. Fisher, S. Kim, B.F. Woodfield, N.E. Phillips, L. Taillefer, K. Hasselbach, J. Flouquet, A.L. Giorgi, J.L. Smith,*** Phys. Rev. Lett. **62**, 1411 (1989).
[6] ***Y. Aoki, T. Namiki, S. Ohsaki, S.R. Saha, H. Sugawara, H. Sato,*** J. Phys. Soc. Jpn. **71**, 2098 (2002).
[7] ***N. Oeschler, P. Gegenwart, F. Steglich, N.A. Frederick, E.D. Bauer, M.B. Maple,*** Proc. SCES 2002 (Krakow), to be published in Acta Phys. Pol. B (2003).
[8] ***T. Tayama, T. Sakakibara, K. Kitami, M. Yokoyama, K. Tenya, H. Amitsuka, D. Aoki, Y. Onuki, Z. Kletowski,*** J. Phys. Soc. Jpn. **70**, 248 (2001).

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