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LOW TEMPERATURE MAGNETIZATION OF THE SKUTTERUDITE SUPERCONDUCTOR $PrOs_4Sb_{12}$*

K. Tenya†, N. Oeschler, P. Gegenwart, F. Steglich

Max Planck Institute for the Chemical Physics of Solids
Nöthnitzer Str. 40, 01187 Dresden, Germany

N.A. Frederick, E.D. Bauer and M.B. Maple

Department of Physics and Institute for Pure and Applied Physical Sciences
University of California, San Diego, La Jolla, CA 92093-0319, USA

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Magnetic and superconducting properties have been investigated on a single crystal of the filled skutterudite superconductor $PrOs_4Sb_{12}$ by means of static magnetization measurements at very low temperatures. There is no trace of the paramagnetic suppression of the superconductivity, indicating that the normal state paramagnetism is due to the Van Vleck contributions. An anomalous field-induced phase transition whose onset field gradually increases with temperature is found above 4.5 T and below 1.1 K. The nonlinear susceptibility increases from a negative value with decreasing temperature and attains a maximum at around 1 K. The origin of these anomalous magnetization behaviors is discussed with respect to the crystal-electric field excitations in $PrOs_4Sb_{12}$.

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The skutterudite compound $PrOs_4Sb_{12}$ presents the first example of a Pr-based heavy fermion superconductor [1]. In the normal state, a pronounced Schottky-like anomaly and a peak in the magnetic susceptibility are observed around 3 K. The latter behavior can be roughly explained by the crystalline-electric field (CEF) model based on the $4f^2$ electrons. It is likely that the CEF ground state is a Γ_3 nonmagnetic doublet with a Γ_5 triplet first excited state. On the other hand, a Γ_1 singlet ground state with a Γ_5 excited state

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† Present address: Graduate School of Science, Hokkaido University, Sapporo 060-0810, Japan.

is also possible. We have performed static magnetization measurements on $PrOs_4Sb_{12}$ down to 0.1 K in order to check the ground state as well as to investigate the superconducting properties.

A single crystal of $PrOs_4Sb_{12}$ was grown using an Sb-flux method [1]. The residual resistivity ratio exceeded 30, indicating the sample is a clean superconductor. We do not expect that the impurity-driven spin–orbit scattering is relevant in this sample. The resistive superconducting transition temperature T_c was 1.86 K. Magnetization measurements were performed at temperatures from 0.1 K to 4 K and in fields up to 11.5 T, using a capacitive magnetometer installed in a ^{3}He–^{4}He dilution refrigerator [3].

Fig. 1 shows isothermal magnetization $M(B)$ and differential susceptibility $dM(B)/dB$ curves of $PrOs_4Sb_{12}$ at 0.23 K in magnetic fields applied parallel to the $\langle 100\rangle$ and $\langle 110\rangle$ directions. Besides the isotropic superconducting-normal transition B_{c2} at 2.2 T, we observe an anomalous magnetization behavior at 4.7 T (5.3 T) for $B\|\langle 100\rangle$ ($B\|\langle 110\rangle$). The onset field of this anomaly increases gradually with increasing temperature and disappears completely for $T \geq 1.1$ K. The stepwise structure of $dM(B^*)/dB$ becomes a broad peak above 1.1 K (0.9 K) for $B\|\langle 100\rangle$ ($B\|\langle 110\rangle$). We note that corresponding anomalies have been found in the thermodynamic properties as well [2]. There is no anisotropy in the magnetization curves in the fields ranging from B_{c2} to B^*, while sizable anisotropy is observed above B^*. Below B^*, the susceptibility at 0.23 K is about 60 % of that at 3 K where the susceptibility has a maximum [1]. At high fields de Haas–van Alphen oscillations are observed for $B\|\langle 100\rangle$, reflecting the high purity of the sample. The frequency is 70 T, leading to a small Fermi surface contribution.

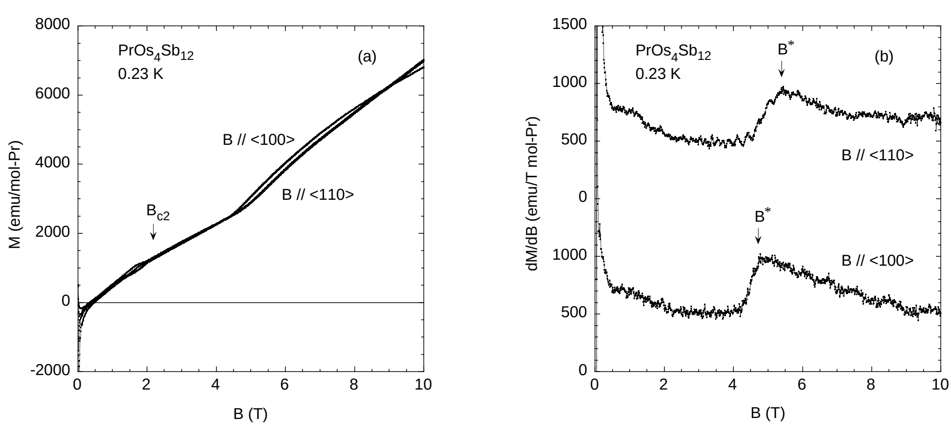

Fig. 1. Magnetization and differential susceptibility curves for $PrOs_4Sb_{12}$ at 0.23 K with $B\|\langle 100\rangle$ and $B\|\langle 110\rangle$.

In the superconducting state, the irreversible magnetization decreases rapidly with field. The magnetization irreversibility, however, increases slightly again around 1.3 T. This is a so-called "peak effect", observed in other f-electron superconductors [3]. The peak rapidly decreases with increasing temperature and, above 1 K, is too small to be detected.

From the magnetization curves with small hystereses, the equilibrium magnetization $M_{\rm eq}(B)$ can be obtained by averaging the increasing- and decreasing-field magnetizations. The thermodynamic critical field $B_{\rm c}$ can be estimated from the $M_{\rm eq}(B)$ integration with respect to B [3]. The extrapolated value of $B_{\rm c}(0)$ is ~ 0.06 T.

The temperature dependence of the superconducting upper critical field $B_{\rm c2}$ and onset field of the anomaly B^* are displayed in Fig. 2(a). These results are similar to those reported in [1,4,5]. There is no anisotropy in $B_{\rm c2}$. It should be noted that $B_{\rm c2}(0)$ is almost the same as the orbital critical field estimated from $dB_{\rm c2}/dT$ just below $T_{\rm c}$, suggesting no paramagnetic suppression of the superconductivity. The Ginzburg–Landau parameter $\kappa_2(T)$, which is obtained from the average slope of the equilibrium magnetization just below $B_{\rm c2}$, increases with decreasing temperature, typical for superconductors without paramagnetic suppression [3]. The spin–orbit scattering mechanism which could recover spin paramagnetism is not relevant since the sample is clean. These results indicate that the origin of the normal state paramagnetism is the Van Vleck contribution, consistent with the Pr^{3+} energy level scheme whose ground state is Γ_1 or Γ_3 in the CEF [1].

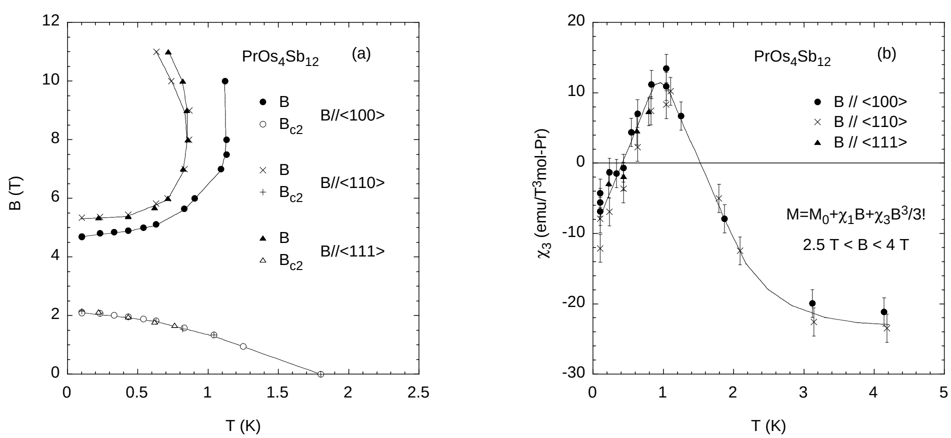

Fig. 2. (a) $B-T$ phase diagram in $PrOs_4Sb_{12}$. (b) Temperature dependence of nonlinear susceptibility in $PrOs_4Sb_{12}$.

As mentioned above, B^* increases with temperature up to $\sim$ 8 T and gradually shifts to the lower temperature side above this field. This anomalous field-induced transition should reflect the nature of the ground state in the CEF. The nonlinear contributions to the isothermal magnetization curves are analyzed to check the ground state properties in detail. The magnetization curves are almost linear with field below B^*, leading to the small contribution of the nonlinear term. The nonlinear susceptibility χ_3 is estimated in the field range between 2.5 T ($> B_{c2}$) and 4 T ($< B^*$), using the expression $M = M_0 + \chi_1 B + \chi_3 B^3/3!$ in order to avoid impurity effects.

Fig. 2(b) displays the temperature dependence of $\chi_3(T)$ for B along the principal axes. While the T-dependence of $\chi_3(T)$ is qualitatively similar to that reported in [5], the magnitude of $\chi_3(T)$ is considerably different from that in [5]. This discrepancy may originate from the difference of the field range where the $\chi_3(T)$ estimation is performed. The temperature dependence is quite strange; $\chi_3(T)$ gradually increases from negative values with decreasing temperature, exhibits a maximum at about 1 K and becomes negative again at lower temperatures. No anisotropy in $\chi_3(T)$ could be detected within experimental error in spite of the small but sizable anisotropy in B^*. Since $\chi_3(T)$ is expected to be anisotropic for both a Γ_1 and a Γ_3 ground state [6], this result calls into question the nature of the ground state in this material. However, recent $C(B,T)$ measurements suggest a crossover to a Γ_5 ground state in a magnetic field $B \sim 5$ T due to the Zeeman splitting of the Γ_5 triplet state, in agreement with these $M(B)$ measurements.

In summary, magnetic and superconducting properties have been investigated on the filled skutterudite superconductor $PrOs_4Sb_{12}$ by means of static magnetization measurements at very low temperatures. Besides the superconducting-normal transition, an anomalous field-induced phase transition whose onset field increases with increasing-T is found below 1.1 K. The paramagnetism below B^* originates from the Van Vleck contribution which does not suppress the superconductivity. The small anisotropy in the linear and nonlinear susceptibilities suggests that the ground state of Pr^{3+} in the CEF may be more complicated than expected for ionic Γ_1 or Γ_3 states due to hybridization with conduction electron states. The research at UCSD was supported by the US NSF and DOE.

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