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Superconductivity and the high-field ordered phase in the heavy-fermion compound $PrOs_4Sb_{12}$

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Abstract

Superconductivity is observed in the filled skutterudite compound $PrOs_4Sb_{12}$ below a critical temperature $T_c = 1.85$ K and appears to develop out of a nonmagnetic heavy Fermi liquid with an effective mass $m^* \approx 50\, m_e$, where m_e is the free electron mass. Features associated with a cubic crystalline electric field are present in magnetic susceptibility, specific heat, electrical resistivity, and inelastic neutron scattering measurements, yielding a Pr^{3+} energy level scheme consisting of a Γ_3 nonmagnetic doublet ground state, a low-lying Γ_5 triplet excited state at $\sim$10 K, and Γ_4 triplet and Γ_1 singlet excited states at much higher temperatures. Measurements also indicate that the superconducting state is unconventional and consists of two distinct superconducting phases. At high fields and low temperatures, an ordered phase of magnetic or quadrupolar origin is observed, suggesting that the superconductivity may occur in the vicinity of a magnetic or quadrupolar quantum critical point.

1. Introduction

Since the mid-1990s, several compounds of Pr have been found to exhibit heavy-fermion behaviour. The crystalline electric field (CEF) ground state of the Pr^{3+} ions in these compounds appears to be a Γ_3 nonmagnetic doublet that carries an electric quadrupole moment. It is conceivable that the heavy-fermion state in these Pr compounds could originate from the interaction between the Pr^{3+} electric quadrupole moments and the charges of the conduction electrons. This would be the electric analogue of the exchange interaction between the magnetic dipole moments of Ce or U ions and the conduction electron spins that is widely believed to be responsible for the heavy-fermion state in most Ce and U heavy-fermion compounds. In fact, such a mechanism was proposed by Cox in 1987 [1] to account for the non-Fermi liquid temperature dependences of certain normal state physical properties of the heavy-electron superconductor UBe_{13}. The Pr compounds that display heavy-fermion behaviour include $PrInAg_2$ [2], $PrFe_4P_{12}$ [3], and, possibly, $PrFe_4Sb_{12}$ [4].

About a year ago, we reported that the compound $PrOs_4Sb_{12}$ exhibits superconductivity with a superconducting critical temperature $T_c = 1.85$ K that apparently develops out of a heavy Fermi liquid with a quasiparticle effective mass $m^* \approx 50\ m_e$, where m_e is the mass of the free electron [5, 6]. As far as we know, $PrOs_4Sb_{12}$ is the first example of a heavy-fermion superconductor based on Pr; all of the other known heavy-fermion superconductors are compounds of Ce or U. The superconducting state appears to be unconventional in nature and may consist of two distinct superconducting phases [7, 8]. An ordered phase, presumably of magnetic or quadrupolar origin, occurs at high fields >4.5 T and low temperatures <1.5 K [7, 9–13], suggesting that the superconductivity may occur in the vicinity of a magnetic or quadrupolar quantum critical point (QCP). In an effort to obtain information about the interactions that are responsible for the heavy-fermion state and superconductivity in $PrOs_4Sb_{12}$, we have performed measurements of various normal and superconducting state properties of this compound as a function of temperature, pressure, and magnetic field [5–7, 9]. Analysis of magnetic susceptibility $\chi(T)$, specific heat $C(T)$, electrical resistivity $\rho(T)$, and inelastic neutron scattering measurements within the context of a cubic CEF yields a Pr^{3+} energy level scheme that consists of a Γ_3 nonmagnetic doublet ground state that carries an electric quadrupole moment, a low-lying Γ_5 triplet excited state at $\sim$10 K, and Γ_4 triplet and Γ_1 singlet excited states at much higher temperatures ($\sim$130 and $\sim$313 K, respectively) [5–7, 9]. This scenario suggests that the underlying mechanism for the heavy-fermion behaviour in $PrOs_4Sb_{12}$ may involve the interaction of Pr^{3+} electric quadrupole moments with the charges of the conduction electrons, rather than Pr^{3+} magnetic dipole moments with the spins of the conduction electrons. It also raises the possibility that electric quadrupole fluctuations play a role in the superconductivity of $PrOs_4Sb_{12}$. In this paper, we briefly review the current experimental situation regarding the heavy-fermion state, the superconducting state, and a high-field, low-temperature phase that is apparently associated with magnetic or quadrupolar order in $PrOs_4Sb_{12}$.

2. Evidence for a heavy-fermion state in $PrOs_4Sb_{12}$

The first evidence for a heavy-fermion state in the filled skutterudite compound $PrOs_4Sb_{12}$ emerged from specific heat $C(T)$ measurements on a $PrOs_4Sb_{12}$ pressed pellet (formed by pressing a collection of small single crystals in a cylindrical die) at low temperatures. Specific heat data in the form of a plot of C/T versus T between 0.5 and 10 K for the $PrOs_4Sb_{12}$ pressed pellet from [5] and [6] are shown in figure 1. The $C(T)$ data have been corrected for excess Sb derived from the molten Sb flux in which the crystals were grown. The line in the figure represents the expression $C(T) = \gamma T + \beta T^3 + C_{Sch}(T)$, where γT and βT^3 are electronic and phonon contributions, respectively, and $C_{Sch}(T)$ is a Schottky anomaly for a two-level system consisting of a doublet ground state and a triplet excited state at an energy Δ above the ground state. The best fit of this expression to the data yields the values $\gamma = 607$ mJ mol^{-1} K^{-2}, $\beta = 3.95$ mJ mol^{-1} K^{-4} (corresponding to a Debye temperature $\theta_D = 203$ K), and $\Delta = 7.15$ K. Superimposed on the Schottky anomaly is a feature in the specific heat due to the onset of superconductivity at $T_c = 1.85$ K which is also observed as an abrupt drop in $\rho(T)$ to zero and as a sharp onset of diamagnetism in $\chi(T)$. The feature in $C(T)/T$ due to the superconductivity is also shown in the top inset of figure 1 along with an entropy-conserving construction from which the ratio of the jump in specific heat ΔC at T_c, $\Delta C/T_c = 632$ mJ mol^{-1} K^{-2}, has been estimated. Using the BCS relation $\Delta C/\gamma T_c = 1.43$, we obtain another estimate for γ of 440 mJ mol^{-1} K^{-2}. The value of $\Delta C/T_c$ is larger than

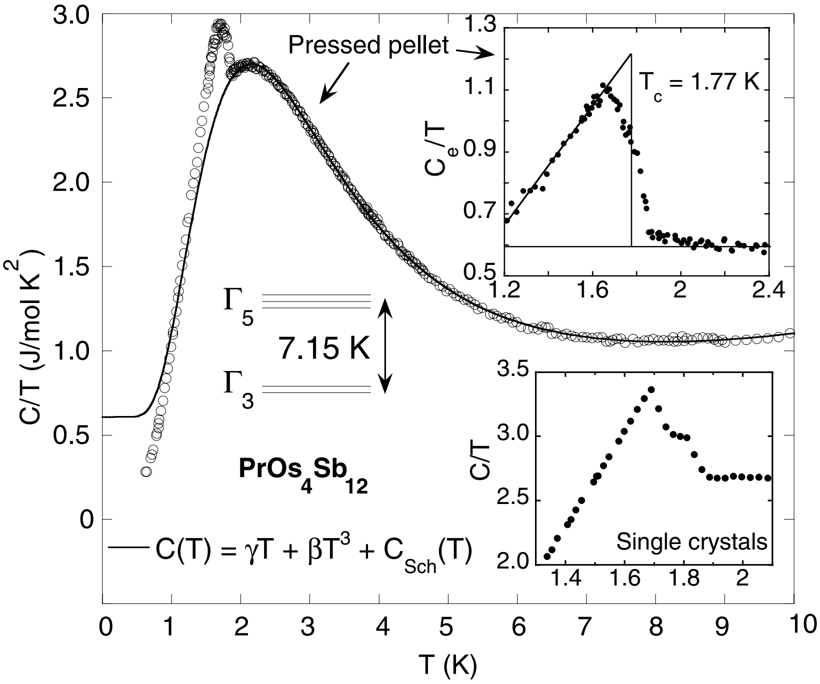

Figure 1. Specific heat C divided by temperature T, C/T, versus T for a $PrOs_4Sb_{12}$ pressed pellet. The line represents a fit of the sum of electronic, lattice, and Schottky contributions to the data. Upper inset: C_e/T versus T near T_c for a $PrOs_4Sb_{12}$ pressed pellet (C_e is the electronic contribution to C). Lower inset: C/T versus T near T_c for $PrOs_4Sb_{12}$ single crystals, showing the structure in ΔC near T_c. Data from [5, 6].

that reported in [6] due to the correction of the $C(T)$ data for the excess Sb (about 30% of the total mass). This value is comparable to that inferred from the fit to the C/T versus T data in the normal state above T_c, and indicative of heavy-fermion behaviour. A similar analysis of the $C(T)$ data taken at the University of Karlsruhe for several single crystals of $PrOs_4Sb_{12}$ prepared in our laboratory yielded $\gamma = 313$ mJ mol^{-1} K^{-2}, $\theta_D = 165$ K, $\Delta = 7$ K, and $\Delta C/\gamma T_c \approx 3$, much higher than the BCS value of 1.43 and indicative of strong-coupling effects [10]. It is interesting that we also find a large value of $\Delta C/\gamma T_c \approx 3$ in recent $C(T)$ measurements on one single crystal of $PrOs_4Sb_{12}$ at UCSD. Although the values of γ determined from these experiments vary somewhat, they are all indicative of a heavy-electron ground state and an effective mass $m^* \approx 50\, m_e$.

Further evidence of heavy-fermion superconductivity is provided by the upper critical field H_{c2} versus T curve shown in figure 2 [6, 7]. The orbital critical field $H_{c2}^*(0)$ can be derived from the slope (-19 kOe K^{-1}) of the H_{c2}-curve near T_c and used to estimate the superconducting coherence length $\xi_0 \approx 116$ Å via the relation $H_{c2}^*(0) = \Phi_0/2\pi\xi_0^2$, where Φ_0 is the flux quantum. The Fermi velocity v_F can be obtained from the BCS relation $\xi_0 = 0.18\hbar v_F/k_B T_c$ and used to determine the effective mass m^* by means of the expression $m^* = \hbar k_F/v_F$. Using a simple free electron model to estimate the Fermi wavevector k_F, an effective mass $m^* \approx 50\, m_e$ is obtained [6, 7]. Calculating γ from m^* yields $\gamma \sim 350$ mJ mol^{-1} K^{-2}, providing further evidence for a heavy-fermion state in $PrOs_4Sb_{12}$.

Recently, Sugawara *et al* [14] performed de Haas–van Alphen effect measurements on $PrOs_4Sb_{12}$. They found that the topology of the Fermi surface is close to that of the reference

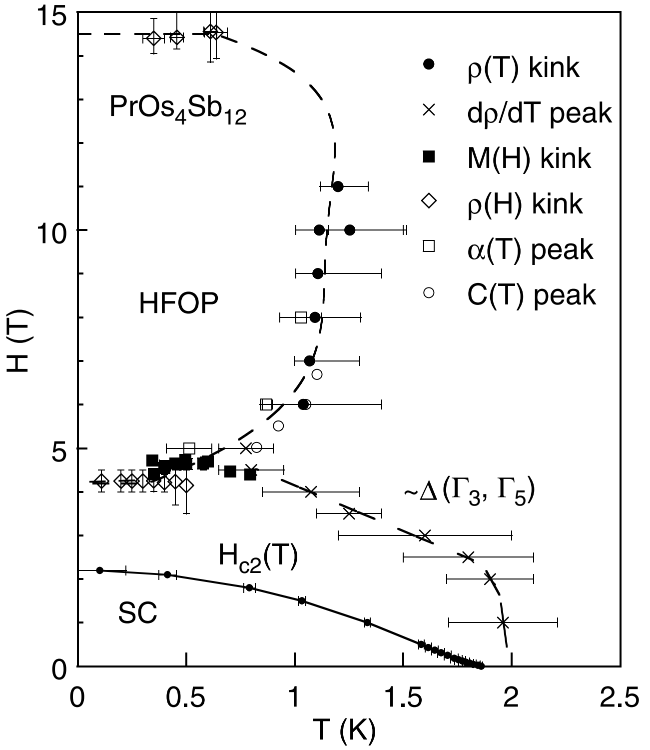

Figure 2. The magnetic field–temperature (H–T) phase diagram of a $PrOs_4Sb_{12}$ single crystal showing the regions exhibiting superconductivity (SC) and the high-field ordered phase (HFOP). The boundary delineating the SC phase is based on measurements of $\rho(H, T)$, while the HFOP phase boundary is based on $\rho(H, T)$, $C(H, T)$, $M(H, T)$, and $\alpha(H, T)$ measurements (see the text). The dashed curve, derived from a peak in $d\rho/dT$ versus T, is a measure of the splitting between the highest and lowest Zeeman levels of the Pr^{3+} Γ_3 ground and Γ_5 excited states, respectively (see the text for further details). Data from [6, 7, 9–11, 26].

compound $LaOs_4Sb_{12}$ and is explained well by band structure calculations. In contrast to the similarity in the Fermi surface topologies of $PrOs_4Sb_{12}$ and $LaOs_4Sb_{12}$, the cyclotron effective masses m_c^* of $PrOs_4Sb_{12}$ are enhanced up to ~6 times relative to those of $LaOs_4Sb_{12}$. The Sommerfeld coefficient γ estimated from the Fermi surface volume and the value of m_c^*, assuming a spherical Fermi surface, is ~150 mJ mol^{-1} K^{-2}, which is two to three times smaller than the value of γ inferred from the normal and superconducting properties of $PrOs_4Sb_{12}$. Our studies of $LaOs_4Sb_{12}$ single crystals reveal superconductivity with a T_c of 1 K.

3. Normal state of $PrOs_4Sb_{12}$

The $\chi(T)$ data for $PrOs_4Sb_{12}$ exhibit a peak at ~3 K and saturate to a value of ~0.11 cm^3 mol^{-1} as $T \rightarrow 0$, indicative of a nonmagnetic ground state. At temperatures above ~5 K, $\chi(T)$ is strongly T-dependent, as expected for well defined Pr^{3+} magnetic moments. In the analysis of the $\chi(T)$ data, interactions between Pr^{3+} ions and hybridization of the Pr 4f and conduction electron states were neglected, while the degeneracy of the Hund's rule multiplet of the Pr^{3+} ions was assumed to be lifted by a cubic CEF and to have a nonmagnetic ground state. According to Lea, Leask, and Wolf (LLW) [15], in a cubic CEF, the Pr^{3+} $J = 4$ Hund's rule multiplet splits into a Γ_1 singlet, a Γ_3 nonmagnetic doublet that carries an electric quadrupole moment, and Γ_4 and Γ_5 triplets. It was assumed that the nonmagnetic ground state of the Pr^{3+} ions corresponds to either a Γ_1 singlet or a Γ_3 nonmagnetic doublet [6]. Although reasonable fits to the $\chi(T)$ data could be obtained for both Γ_1 and Γ_3 ground states, the most satisfactory fit was obtained for a Γ_3 nonmagnetic doublet ground state with a Γ_5 first excited triplet state at 11 K and Γ_4 and Γ_1 excited states at 130 and 313 K, respectively. Inelastic neutron scattering measurements on $PrOs_4Sb_{12}$ [7] reveal peaks in the INS spectrum at 0.71 meV (8.2 K) and 11.5 meV (133 K) that appear to be associated with transitions between the Γ_3 ground state and the Γ_5 first and Γ_4 second excited states, respectively, that are in good agreement with the Pr^{3+} CEF energy level scheme determined from the analysis of the $\chi(T)$ data. As noted above, the Schottky anomaly in the $C(T)$ data on $PrOs_4Sb_{12}$ taken at UCSD and at the University of Karlsruhe [10] can be described well as a two-level system consisting of a doublet ground state and a low-lying triplet excited state with a splitting of $\sim$7 K, a value that is comparable to the values deduced from the $\chi(T)$ and INS data.

While a magnetic Γ_5 Pr^{3+} ground state (Γ_4 is not a possible ground state for a cubic system in the LLW formulation) could also produce a nonmagnetic heavy-fermion ground state via an antiferromagnetic exchange interaction (Kondo effect), the behaviour of $\rho(T)$ of $PrOs_4Sb_{12}$ in the normal state does not resemble the behaviour of $\rho(T)$ expected for this scenario. For a typical magnetically induced heavy-fermion compound, $\rho(T)$ often increases with decreasing temperature due to Kondo scattering, reaches a maximum, and then decreases rapidly with decreasing temperature as the highly correlated heavy-fermion state forms below the coherence temperature. At low temperatures, $\rho(T)$ typically varies as AT^2 with a prefactor $A \approx 10^{-5}(\mu\Omega\ \mathrm{cm\ K^2\ mJ^{-2}\ mol^2})\gamma^2$ that is consistent with the Kadowaki–Woods relation [16]. In contrast, $\rho(T)$ for $PrOs_4Sb_{12}$ [5, 6] exhibits typical metallic behaviour with negative curvature at higher temperatures and a pronounced ‘roll-off’ below $\sim$8 K before it vanishes abruptly when the compound becomes superconducting. The ‘roll-off’ in $\rho(T)$ is consistent with a decrease in charge- or spin-dependent scattering of conduction electrons by the Pr^{3+} ions due to the decrease in population of the low-lying first excited state (Γ_5) as the temperature is lowered. The ‘roll-off’ below $\sim$8 K and the negative curvature at higher temperatures in $\rho(T)$ can be described reasonably well by calculations based on magnetic and aspherical Coulomb scattering of conduction electrons by the Pr^{3+} ions with a low-lying Γ_5 excited state separated from the Γ_3 ground state by $\sim$6 K and excited Γ_4 triplet and Γ_1 singlet states at much higher energies comparable to those found from the analysis of the $\chi(T)$ and INS measurements [17]. The $\rho(T)$ data can be described by a temperature dependence of the form AT^2 between $\sim$8 and 45 K, but with a prefactor $A \approx 0.009\ \mu\Omega\ \mathrm{cm\ K^{-2}}$ that is nearly two orders of magnitude smaller than that expected from the Kadowaki–Woods relation ($A \approx 1.2\ \mu\Omega\ \mathrm{cm\ K^{-2}}$ for $\gamma \approx 350\ \mathrm{mJ\ mol^{-1}\ K^{-2}}$) [16]. Interestingly, $\rho(T)$ is consistent with T^2-behaviour with a value $A \approx 1\ \mu\Omega\ \mathrm{cm\ K^{-2}}$ in fields of $\sim$5 T [9] in the high-field ordered phase discussed in section 5. The zero-field temperature dependence of $\rho(T)$ is similar to that observed for the compound $PrInAg_2$, which also has a low value of the coefficient A, an enormous γ of $\sim$6.5 J $\mathrm{mol^{-1}\ K^{-2}}$, and a Γ_3 nonmagnetic doublet ground state [2]. Another possible source of the enhanced effective mass in $PrOs_4Sb_{12}$ may involve excitations from the ground state to the low-lying first excited state in the Pr^{3+} CEF energy level scheme [18].

Two studies of the nonlinear magnetic susceptibility have been performed in an attempt to determine the CEF ground state of the Pr^{3+} ion in $PrOs_4Sb_{12}$ [12, 19]. The nonlinear susceptibility χ_3 is the coefficient of the H^3-term in the expansion of the magnetization M in a series of odd powers of H; i.e., $M \approx \chi_1 H + (\chi_3/6)H^3$, where χ_1 is the ordinary linear susceptibility. In an ionic situation, χ_3 is isotropic and varies as T^{-3} for a magnetic ground state, whereas χ_3 is anisotropic and diverges at low temperatures for $\boldsymbol{H} \parallel [100]$ and approaches a constant for $\boldsymbol{H} \parallel [111]$ for a non-Kramers Γ_3 doublet ground state [20]. This type of study was previously employed in an attempt to determine the ground state of U in the compound UBe_{13} [21]. In both studies of $PrOs_4Sb_{12}$, $\chi_3(T)$ was found to behave similarly for $\boldsymbol{H} \parallel [100]$ and $\boldsymbol{H} \parallel [111]$, exhibiting a minimum near 4 K followed by a maximum near 1 K and a negative divergence with decreasing temperature. Calculations based on the quadrupolar Anderson–Hamiltonian described the $\chi_3(T)$ data reasonably well for $\boldsymbol{H} \parallel [100]$, but not very well for $\boldsymbol{H} \parallel [111]$. It was concluded that the data were qualitatively consistent with a Γ_3 ground state, given the limitations of the experiment and the complexity of the theory. The $\chi_3(T)$ studies are difficult to interpret because of the curvature of $M(H)$ and the complications that arise at lower temperatures $T \leqslant T_c$ and lower fields $H \leqslant H_{c2}$ due to the superconductivity and at temperatures $T \leqslant 2$ K and higher fields $H \geqslant 4.5$ T by the onset of the high-field ordered phase, discussed in section 5.

4. Superconducting state of $PrOs_4Sb_{12}$

A number of features in the superconducting properties of $PrOs_4Sb_{12}$ indicate that the superconductivity of this compound is unconventional in nature. One of these features is the 'double-step' structure in the jump in $C(T)$ near T_c in single crystals (lower inset of figure 1) that suggests two distinct superconducting phases with different T_cs: $T_{c1} \approx 1.85$ K and $T_{c2} \approx 1.70$ K [7, 10]. This structure is not evident in the $C(T)$ data taken for the pressed pellet of $PrOs_4Sb_{12}$ shown in the upper inset of figure 1, possibly due to strains in the single crystals out of which the pressed pellet is comprised that broaden the transitions at T_{c1} and T_{c2} with the result that they overlap and become indistinguishable. However, at this point, we are unable to eliminate the possibility that the two apparent jumps in $C(T)$ are due to sample inhomogeneity. It is noteworthy that all of the single-crystal specimens prepared in our laboratory and investigated by our group and our collaborators exhibit this 'double-step' structure. Multiple superconducting transitions, apparently associated with distinct superconducting phases, have previously been observed in two other heavy-fermion superconductors, UPt_3 [22] and $U_{1-x}Th_xBe_{13}$ $(0.1 \leqslant x \leqslant 0.35)$ [23]. Measurements of the specific heat in magnetic fields reveal that the two superconducting features shift downward in temperature at nearly the same rate with increasing field, consistent with the smooth temperature dependence of the $H_{c2}(T)$ curve [10]. These two transitions have also been observed in thermal expansion measurements [11], which, from the Ehrenfest relation, reveal that T_{c1} and T_{c2} have different pressure dependences, suggesting that they are associated with two distinct superconducting phases. Another feature is the power law T-dependence of $C_s(T)$, $C_s(T) \sim T^{2.5}$, after the Schottky anomaly and βT^3 lattice contributions have been subtracted from the $C(T)$ data. (As reported in [7], $C_s(T)$ follows a power law with $C_s(T) \sim T^{3.9}$ when the Schottky anomaly is not subtracted.) However, this dependence can only be established from T_c down to $\sim 0.4\ T_c$, since it is not possible to reliably correct the $C(T)$ data at lower temperatures for an enormous nuclear Schottky anomaly. Power law T-dependences of the superconducting properties are generally attributed to nodes in the superconducting energy gap at points or lines on the Fermi surface. Among three recent experiments on $PrOs_4Sb_{12}$, described below, two yield evidence for an isotropic energy gap, while another provides evidence for two distinct superconducting phases in the H–T plane with different numbers of point nodes in the energy gap.

Recent transverse field μSR [24] and Sb NQR measurements [25] on $PrOs_4Sb_{12}$ are consistent with an isotropic energy gap. Along with the specific heat, these measurements indicate strong-coupling superconductivity. These findings suggest an s-wave, or, perhaps, a Balian–Werthamer p-wave order parameter. On the other hand, the superconducting gap

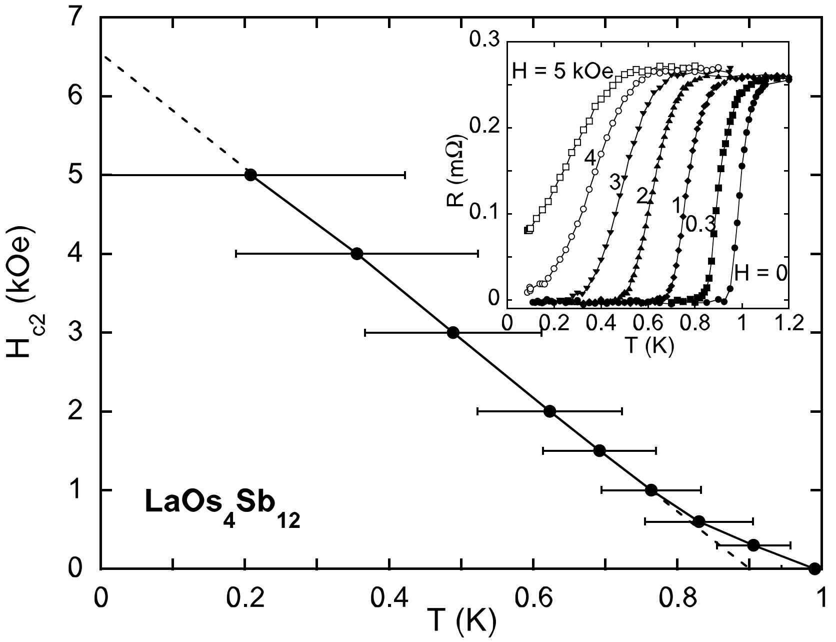

Figure 3. Upper critical field H_{c2} versus T for $LaOs_4Sb_{12}$, based on resistance R versus temperature T data shown in the inset.

structure of $PrOs_4Sb_{12}$ was investigated by means of thermal conductivity measurements in magnetic fields rotated relative to the crystallographic axes by Izawa *et al* [8]. These measurements reveal two regions in the H–T plane, a low-field region in which $\Delta(\boldsymbol{k})$ has two point nodes, and a high-field region where $\Delta(\boldsymbol{k})$ has six point nodes. The line lying between the low- and high-field superconducting phases may be associated with the transition at T_{c2}, whereas the line between the high-field superconducting phase and the normal phase, $H_{c2}(T)$, converges with T_{c1} as $H \rightarrow 0$. Clearly, more research will be required to further elucidate the nature of the superconductivity in $PrOs_4Sb_{12}$.

It is noteworthy that the reference compound without 4f electrons, $LaOs_4Sb_{12}$, is also superconducting, but with $T_c \approx 1$ K, considerably smaller than that of $PrOs_4Sb_{12}$. This suggests that the pairing interaction is enhanced by the presence of the Pr 4f electrons, possibly through the interaction of the Pr^{3+} electric quadrupole moments with the conduction electrons. In contrast, the values of T_c for the other PrT_4X_{12} filled skutterudites are smaller than those for their La-based counterparts. The values of T_c in K for MT_4X_{12} compounds with M = La or Pr are listed in table 1. A plot of the upper critical field H_{c2} versus T for $LaOs_4Sb_{12}$ and the resistive $R(T)$ transition curves upon which it is based are shown in figure 3.

5. High-field ordered phase in $PrOs_4Sb_{12}$

Evidence for a high-field ordered phase was first derived from magnetoresistance measurements in the temperature range 80 mK $\leqslant T \leqslant$ 2 K and magnetic fields up to 10 T [7, 9]. Recently, the magnetoresistance measurements have been extended up to 18 T for 0.35 K $\leqslant T \leqslant$ 2 K [26]. The H–T phase diagram, depicting the superconducting region and the high-field ordered phase, is shown in figure 2. The line that intersects the high-field ordered phase represents the inflection point of the 'roll-off' in $\rho(T)$ for $H < 4.5$ T at low

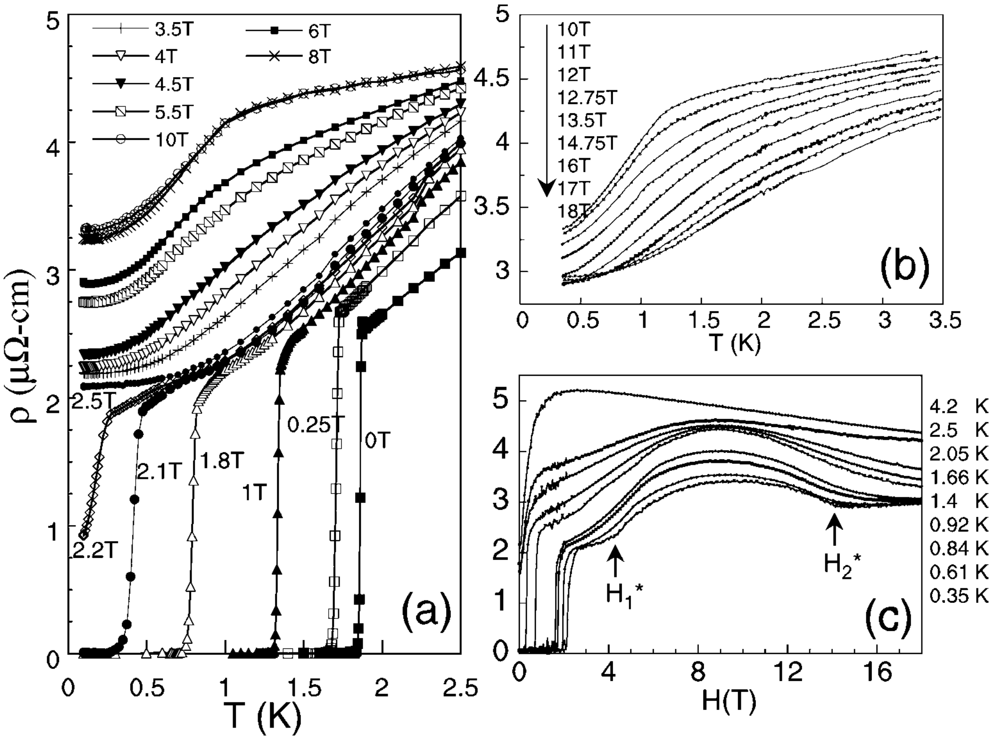

Figure 4. ((a), (b)) Electrical resistivity ρ versus temperature T at various magnetic fields H up to 18 T for a $\mathrm{PrOs_4Sb_{12}}$ single crystal. (c) ρ versus H at various temperatures between 0.35 and 4.2 K. The rapid drop in ρ to zero for $H < 2.5$ T is due to the superconducting transition, while the shoulder in $\rho(T)$ at ~1K above 4.5 T and the kinks in $\rho(H)$ (indicated at H_1^* and H_2^*) below 1.7 K are due to a field-induced phase (high-field ordered phase—HFOP). After [9, 26].

Table 1. The superconducting critical temperature T_c for $\mathrm{LnT_4X_{12}}$ compounds for Ln = La, Pr; T = Fe, Ru, Os; and X = P, As, Sb. The T_c-values listed are in K and have been derived from [4, 25, 28–35]. The symbols † and ‡ indicate that superconductivity has not been observed in those compounds above 2 and 0.35 K, respectively.

	$\mathrm{LaT_4X_{12}}$			$\mathrm{PrT_4X_{12}}$		
Fe	4.1	†	†	‡	?	†
Ru	7.2	10.3	2.8	‡	2.4	1.0
Os	1.8	3.2	1.0	†	?	1.8
	P	As	Sb	P	As	Sb

temperatures and is a measure of the splitting between the $\mathrm{Pr^{3+}}$ ground state and the first excited state, which decreases with field (see figure 4). The high-field ordered phase has also been observed by means of large peaks in the specific heat [10, 13] and thermal expansion [11] and kinks in magnetization versus magnetic field curves [12, 26] in magnetic fields >4.5 T and at temperatures <1.5 K.

Shown in figures 4(a) and (b) are $\rho(T)$ data for various magnetic fields up to 18 T for $\mathrm{PrOs_4Sb_{12}}$, which reveal drops in $\rho(T)$ due to the superconductivity for $H \leqslant 2.2$ T and features in $\rho(T)$ associated with the onset of the high-field ordered phase for $H \geqslant 4.5$ T. Isotherms of electrical resistivity ρ versus H at various temperatures $0.35\ \mathrm{K} \leqslant T \leqslant 4.2$ K and fields

$0\,T \leqslant H \leqslant 18\,T$ are shown in figure 4(c). The fields denoting the boundaries of the high-field ordered phase, H_1^* and H_2^*, are indicated in the figure.

6. Summary

Experiments on the filled skutterudite compound $PrOs_4Sb_{12}$ have revealed a number of extraordinary phenomena: a heavy-fermion state characterized by an effective mass $m^* \approx 50\,m_e$, unconventional superconductivity below $T_c = 1.85$ K with two distinct superconducting phases, and a high-field ordered phase, presumably associated with magnetic or quadrupolar order. Analysis of $\chi(T)$, $C(T)$, $\rho(T)$, and INS data indicate that Pr^{3+} has a nonmagnetic Γ_3 doublet ground state that carries an electric quadrupole moment, a low-lying Γ_5 triplet excited state at $\sim$10 K, and Γ_4 triplet and Γ_1 singlet excited states at much higher energies. This suggests that the interaction between the quadrupole moments of the Pr^{3+} ions and the charges of the conduction electrons, as well as the excitations between the Γ_3 ground state and Γ_5 low-lying excited state, may play an important role in generating the heavy-fermion state and superconductivity in this compound. The heavy-fermion state, unconventional superconductivity, and high-field ordered phase observed in $PrOs_4Sb_{12}$ and reviewed herein present a significant challenge for theoretical description [27, 36–39].

Acknowledgments

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