SUPERCONDUCTIVITY NEAR QUANTUM CRITICAL POINTS IN f-ELECTRON MATERIALS*

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Recent experiments on single crystals of the compounds $CeRh_{1-x}Co_xIn_5$ and $PrOs_4Sb_{12}$ are briefly reviewed. The temperature-composition $(T-x)$ phase diagram of the heavy fermion pseudoternary system $CeRh_{1-x}Co_xIn_5$, delineating the regions in which superconductivity, antiferromagnetism, and the coexistence of these two phenomena occur, has been established. Superconductivity has been observed in single crystals of the filled skutterudite compound $PrOs_4Sb_{12}$ below $T_c = 1.85$ K and appears to involve heavy fermion quasiparticles with an effective mass $m^* \sim 50m_e$, where m_e is the mass of the free electron.

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1. Introduction

Interest in Non-Fermi Liquid (NFL) behavior in strongly correlated f-electron materials has steadily increased during the past decade [1]. The NFL behavior has been identified in certain intermetallic compounds containing lanthanide and actinide ions with partially-filled f-electron shells (Ce, Yb, and U, to date). These ions carry magnetic dipole or electric quadrupole moments that interact with the spins and charges of the conduction electrons and can undergo magnetic or quadrupolar ordering at low temperatures. NFL behavior was first established in chemically substituted f-electron compounds containing nonmagnetic substituents [2–4], but has since been observed in a number of stoichiometric f-electron compounds as well [5–7]. The NFL behavior in these f-electron materials is manifested as weak power law or logarithmic divergences in temperature of the physical

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properties at low temperatures. For many of these f-electron materials, the electrical resistivity $\rho(T)$, specific heat $C(T)$, and magnetic susceptibility $\chi(T)$ have the following NFL temperature dependences for $T \ll T_0$ [2–4]:

(i) $\rho(T) \sim 1 - a(T/T_0)^n$ where $|a| \sim 1$, $a < 0$ or > 0, and $n \sim 1 - 1.5$;

(ii) $C(T)/T \sim (-1/T_0)\ln(T/T_0)$, or $\sim T^{-1+\lambda}$; and

(iii) $\chi(T) \sim 1-(T/T_0)^{1/2}$, $\sim (-1/T_0)\ln(T/T_0)$, or $\sim T^{-1+\lambda}$ ($\lambda \sim 0.7-0.8$).

In several of the f-electron systems, the characteristic temperature T_0 can be identified with the Kondo temperature T_K. These NFL characteristics are found in both chemically substituted and stoichiometric compounds, indicating that the underlying physics is not primarily driven by atomic disorder. Experiments on a variety of f-electron systems suggest that there are two routes to NFL behavior in these materials, a single ion route, involving an unconventional Kondo effect, and an inter-ionic interaction route associated with order parameter fluctuations in the vicinity of a second order magnetic (or, possibly, quadrupolar) phase transition that has been suppressed to $T = 0$ K (Quantum Critical Point — QCP) [4–7]. One of the most interesting recent developments in this field is the observation of pressure-induced superconductivity in several f-electron materials within a narrow pressure range in the vicinity of the critical pressure P_c at which the magnetic ordering temperature is suppressed to 0 K (magnetic QCP). For example, superconductivity has been observed in single crystal specimens of the AntiFerroMagnetic (AFM) compounds $CeIn_3$ and $CePd_2Si_2$ [6] at pressures in the vicinity of P_c where the Néel temperature T_N vanishes ($\sim$ 26 kbar for both compounds) and in both single crystal [8] and polycrystalline [9] specimens of the FerroMagnetic (FM) compound UGe_2 within the FM phase at pressures below P_c where the Curie temperature T_C vanishes ($\sim$ 16 kbar). These experiments suggest that magnetic interactions may play an important role in the pairing of superconducting electrons in these materials.

In this paper, we briefly describe recent experiments carried out in our laboratory on two f-electron systems that were motivated by the possible connection between heavy fermion superconductivity and magnetic QCP's suggested by the aforementioned experiments. The two f-electron systems investigated are the pseudoternary system $CeRh_{1-x}Co_xIn_5$, in which the Néel temperature vanishes at a critical concentration near $x_c \sim 0.8$ (AFM QCP), and the filled skutterudite compound $PrOs_4Sb_{12}$ which apparently does not exhibit magnetic or quadrupolar order down to $\sim$ 60 mK and may be located near a magnetic or quadrupolar QCP.

2. Superconductivity and magnetism in the pseudoternary system $CeRh_{1-x}Co_xIn_5$

A new class of Ce-based heavy fermion materials with the formula $Ce_nT_mIn_{3n+2m}$ (T = Co, Rh, Ir; $n = 1, 2$; $m = 0, 1$) has recently been discovered [10]. These compounds display a variety of interesting phenomena including superconductivity (SC), pressure-induced SC, and antiferromagnetism (AFM). The compound $CeRhIn_5$ displays AFM with $T_N = 3.8$ K, while $CeCoIn_5$ and $CeIrIn_5$ exhibit SC with T_c's of 2.3 K and 0.4 K, respectively [11–13]. The $CeTIn_5$ compounds can be viewed as derivatives of the $CeIn_3$ structure, in which layers of TIn_2 and $CeIn_3$ are stacked along the c-axis. The parent compound $CeIn_3$ exhibits AFM with $T_N \sim 10$ K at atmospheric pressure; upon application of pressure to $CeIn_3$, T_N decreases and SC with a maximum $T_c \sim 0.2$ K occurs in a narrow range of pressures around $P_c \sim 26$ kbar where $T_N \to 0$ K [6]. Recently, we investigated the interplay between SC and AFM in single crystal specimens of the pseudoternary system $CeRh_{1-x}Co_xIn_5$ grown in our laboratory using a molten indium flux technique [14]. One of the objectives of this study was to compare the behavior of $T_c(x)$ in the $CeRh_{1-x}Co_xIn_5$ system to $T_c(P)$ in $CeIn_3$ in the vicinity of the critical concentration x_c and pressure P_c where T_N vanishes (the AFM QCP).

Shown in Fig. 1 are plots of the specific heat C divided by temperature T, C/T, *vs* T for samples of $CeRh_{1-x}Co_xIn_5$ with Co concentrations in the range $0.2 \leq x \leq 1$. The peaks at 1–2 K for samples with $0.4 \leq x \leq 1$ are associated with the onset of SC, while the peaks at $T \sim 3$–4 K for $0.2 \leq x \leq 0.6$ mark the onset of AFM. For the samples with $0.4 \leq x \leq 0.6$, there are two peaks in the specific heat data, indicating the coexistence of SC and AFM. The sample with $x = 0.4$ shows a double peak at $T \sim 1.7$ K which could be indicative of two slightly different Co concentrations. The entropy under the antiferromagnetic peaks is much less than $R \ln 2$, suggesting that the ordered moment is being screened by the Kondo effect. The total entropy at ~ 7 K is very similar for all of the samples with the exception of the $CeCoIn_5$ end member which implies that the same heavy electrons are involved in both the SC and the AFM.

The T–x phase diagram of the $CeRh_{1-x}Co_xIn_5$ system is shown in Fig. 2. The values of T_N and T_c, determined from measurements of $C(T)$ shown in Fig. 1 and features in $\rho(T)$ and $\chi(T)$ (not shown), are plotted as a function of Co concentration x. The Néel temperature T_N remains roughly constant at ~ 3.7 K for $0 \leq x \leq 0.4$. The values of T_N determined from $\rho(T)$ and $C(T)$ are in good agreement, while the values determined from $\chi(T)$ are 0.2–0.4 K higher. For concentrations in the range $0.4 \leq x \leq 0.6$, T_N determined from $C(T)$ drops from ~ 3.7 K to ~ 2.9 K. The resistive transition

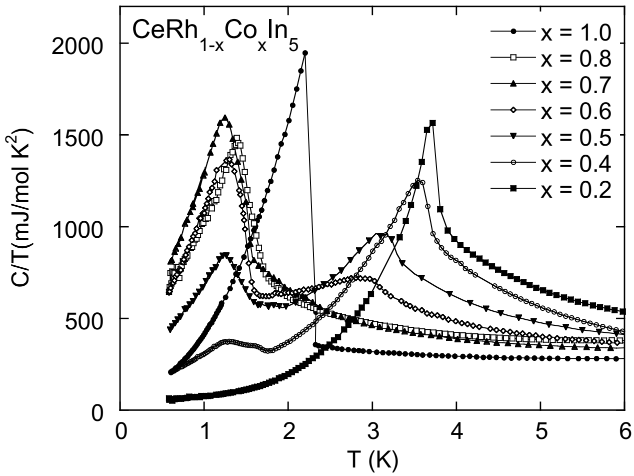

Fig. 1. Specific heat C divided by temperature T as a function of T for samples of $CeRh_{1-x}Co_xIn_5$ with $0.2 \leq x \leq 1$. From Ref. [14].

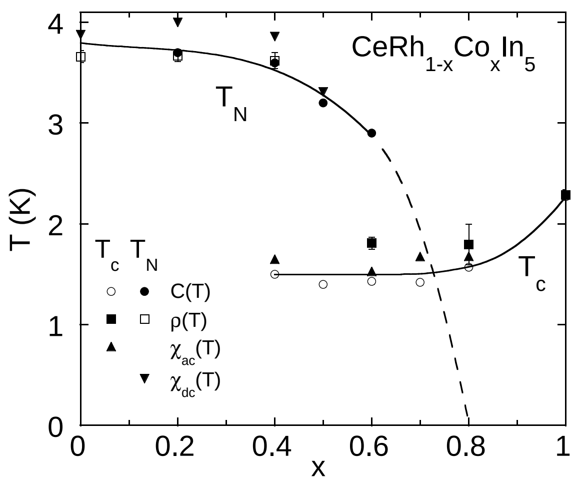

Fig. 2. Néel temperature $T_{\rm N}$ and superconducting critical temperature $T_{\rm c}$ determined from measurements of specific heat $C(T)$, electrical resistivity $\rho(T)$, and magnetic susceptibility $\chi(T)$ as a function of Co concentration x for samples of $CeRh_{1-x}Co_xIn_5$. The upper and lower bars on the $T_{\rm c}$ points determined from $\rho(T)$ correspond to the 90% and 10% values of the SC transition, and the bars on the $T_{\rm N}$ data points determined from $\chi(T)$ indicate the width of the AFM transitions. From Ref. [14].

becomes broader at $x = 0.5$ and the transitions in both $\rho(T)$ and $\chi(T)$ can no longer be distinguished for $x > 0.5$. By $x = 0.8$, there is no evidence of an antiferromagnetic transition down to $T = 0.5$ K in the specific heat data. Superconductivity is observed in $\rho(T)$, $C(T)$, and $\chi(T)$ for samples with $0.4 \leq x \leq 1$. The value of T_c remains roughly constant for concentrations in the range $0.4 \leq x \leq 0.6$, and SC appears to coexist with AFM. Between $x = 0.8$ and $x = 1$, T_c increases from 1.6 K to 2.3 K.

The SC observed in $CeRh_{1-x}Co_xIn_5$ for $0.4 \leq x \leq 1.0$ appears to be non s-wave in nature, which is typical for heavy fermion superconductors [15]. The temperature dependence of $C(T)$ below T_c varies as a power law $C/T = A + BT^n$ with $n \sim 2$ for all of the samples in which superconductivity is observed. A power law dependence of $C(T)$ below T_c has previously been observed in $CeCoIn_5$ with $n \sim 2$. For the superconductors $CeIrIn_5$ and $CeCoIn_5$ at ambient pressure, and $CeRhIn_5$ under 27 kbar of pressure, there is additional evidence for a non s-wave order parameter from ^{115}In NQR measurements [16, 17]. These measurements reveal a power law T-dependence of $1/T_1$ below T_c with a power law exponent $n \sim 3$, and the absence of a Hebel–Schlichter coherence peak below T_c.

It is interesting that in the $CeRh_{1-x}Co_xIn_5$ compounds, which display the coexistence of SC and AFM, T_c remains roughly constant while T_N varies with Co concentration and appears to vanish at a critical concentration near $x_c \sim 0.8$ at which there is presumably an AFM QCP. This indicates that the superconducting gap is unchanged by the presence of the AFM. This relatively weak dependence of T_c on x in the vicinity of x_c where T_N appears to vanish in the $CeRh_{1-x}Co_xIn_5$ system is in marked contrast to the narrow peak in $T_c(P)$ in the vicinity of P_c where T_N is suppressed to zero in the parent compound $CeIn_3$.

The results of our investigation on the $CeRh_{1-x}Co_xIn_5$ system are very similar to those obtained by Pagliuso *et al.* [18] on the $CeRh_{1-x}Ir_xIn_5$ system in which coexistence of SC and AFM is found in the range $0.3 \leq x \leq 0.6$.

2.1. Superconductivity and heavy fermion behavior of the filled skutterudite compound $PrOs_4Sb_{12}$

The filled skutterudite compounds have the formula MT_4X_{12} (M = alkaline earth, lanthanide, actinide; T = Fe, Ru, Os; X = P, As, Sb). These compounds exhibit a variety of strongly correlated electron phenomena and ground states, including superconductivity (*e.g.*, $LaFe_4P_{12}$, $LaRu_4Sb_{12}$), ferromagnetism (*e.g.*, UFe_4P_{12}, $SmFe_4Sb_{12}$), “hybridization gap” semiconductivity (or “Kondo insulator” behavior) (*e.g.*, $CeFe_4P_{12}$, UFe_4P_{12}), valence fluctuation/heavy fermion behavior (*e.g.*, $CeFe_4Sb_{12}$, $YbFe_4Sb_{12}$), non-

Fermi liquid behavior (*e.g.*, $CeRu_4Sb_{12}$), and metal-insulator transitions (*e.g.*, $PrRu_4P_{12}$) [19]. These materials are also of interest because of their potential for thermoelectric applications [20].

In our recent investigations of filled skutterudite compounds, we have found that $PrOs_4Sb_{12}$ exhibits SC with a $T_c = 1.85$ K [21]. What is particularly interesting is that the SC appears to involve heavy fermion quasiparticles with an effective mass $m^* \sim 50m_e$, where m_e is the mass of the free electron, as inferred from the jump in the specific heat at T_c, the initial slope of the upper critical field H_{c2} near T_c, and the normal state electronic specific heat coefficient γ [21]. Thermodynamic measurements indicate that the ground state of the Pr^{3+} ions in the cubic Crystalline Electric Field (CEF) is probably a Γ_3 nonmagnetic doublet which carries an electric quadrupole moment, suggesting that the heavy fermion state in $PrOs_4Sb_{12}$ may be associated with screening of the Pr^{3+} Γ_3 quadrupole moments by the charges of the conduction electrons.

Shown in Fig. 3 is a plot of C/T *vs* T for $PrOs_4Sb_{12}$ between 0.7 K and 20 K. Between $\sim$ 8 K and 20 K, the data can be fitted by the sum of a Debye lattice contribution $C_l = \beta T^3$ and an electronic contribution $C_e = \gamma T$, where the value of β corresponds to a Debye temperature of 259 K and $\gamma = 750$ mJ/mol K^2, typical of a heavy fermion metal. At temperatures below $\sim$ 8 K, there is a pronounced Schottky anomaly with a peak

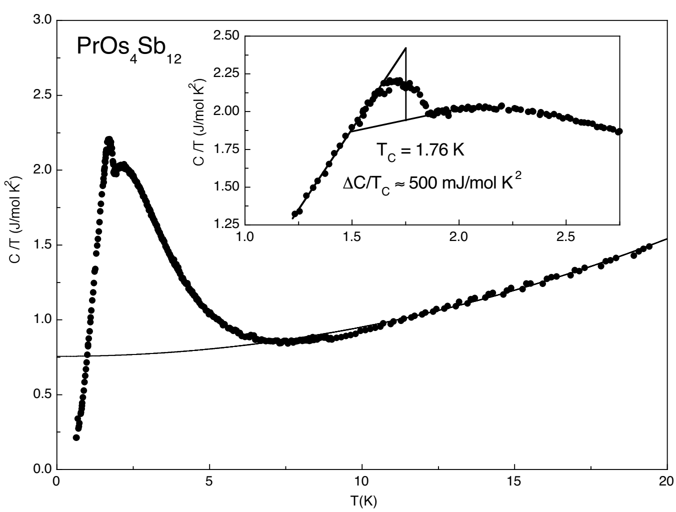

Fig. 3. Specific heat C divided by temperature T *vs* T for $PrOs_4Sb_{12}$ between 0.7 K and 20 K. Inset: Expanded view of the superconducting specific heat jump at T_c. The solid lines are an equal entropy construction that yields $\Delta C/T_c \sim 500$ mJ/mol K^2.

near ~ 3 K. Superimposed on this anomaly is a specific heat jump associated with the onset of superconductivity at $T_c \sim 1.75$ K. The specific heat jump ΔC is displayed in the upper inset of the Fig. 3 in which an entropy conserving construction has been made which yields $\Delta C/T_c \sim 500$ mJ/mol K^2. Since the microscopic theory of superconductivity predicts that $\Delta C \sim \gamma T_c$ ($\Delta C = 1.43\gamma T_c$ for the weak coupling BCS theory), this indicates that the superconductivity is a bulk phenomenon and that it involves the same heavy fermion quasiparticles that are responsible for the large value of γ inferred from the normal state specific heat.

The electronic specific heat of $PrOs_4Sb_{12}$, ΔC, which is calculated by subtracting the lattice contribution of the isostructural compound $LaOs_4Sb_{12}$, can be fit by a Schottky anomaly for a system of two levels with equal degeneracy split by 6.6 K and an electronic term $\gamma' T$ with $\gamma' = 570$ mJ/mol K^2.

Shown in Fig. 4 is a plot of χ *vs* T which reveals that $\chi(T)$ exhibits a maximum at ~ 3 K, below which it decreases and approaches a finite value as $T \to 0$ K, indicative of a nonmagnetic ground state. The upper most inset shows that χ undergoes an abrupt decrease at the superconducting transition, while the lower most inset shows that $\chi(T)$ follows a Curie–Weiss law between 50 K and room temperature with an effective moment $\mu_{\text{eff}} = 2.97\,\mu_B$ and a Curie–Weiss temperature $\theta_{\text{CW}} = -16$ K.

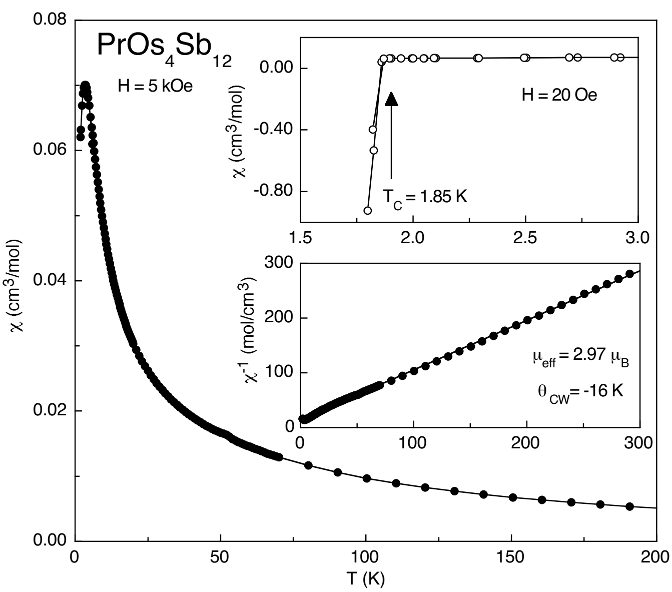

Fig. 4. Magnetic susceptibility χ *vs* T for $PrOs_4Sb_{12}$ between 1.8 K and 300 K. Upper inset: $\chi(T)$ at $H = 20$ Oe, showing the superconducting transition. Lower inset: χ^{-1} *vs* T between 1.8 K and 300 K.

A calculation of $\chi(T)$ for a single Pr^{3+} ion in a cubic CEF based on the theory of Lea, Leask, and Wolf (LLW) [22] provides a reasonable description of the χ *vs* T data shown in Fig. 4. In a cubic CEF, the $J = 4$ Hund's rule multiplet of Pr^{3+} is split into a Γ_1 singlet, a Γ_3 doublet, and Γ_4 and Γ_5 triplets. According to the LLW theory, the CEF Hamiltonian in cubic symmetry can be expressed in terms of a parameter x, the ratio of the fourth to the sixth order terms of the angular momentum operators, and an overall scale factor W. For the calculation of $\chi(T)$, the Pr^{3+} ground state was taken to be either a Γ_1 singlet ($W > 0$) or a Γ_3 doublet ($W < 0$), in accordance with the nonmagnetic behavior of Pr^{3+} in this compound. The best overall fit to the data corresponds to the Γ_3 ground state with Γ_5, Γ_4, and Γ_1 excited states at respective temperatures of 11 K, 139 K, and 313 K above the Γ_3 ground state.

Electrical resistivity ρ *vs* T data for $PrOs_4Sb_{12}$, shown in Fig. 5, reveal typical metallic behavior. Below 7 K, $\rho(T)$ decreases by nearly 50% relative to its value at 7 K and then drops abruptly to zero at T_c, as shown in the upper inset of Fig. 5. The decrease in $\rho(T)$ below 7 K is apparently due to the decrease of scattering of conduction electrons by electric quadrupole or magnetic dipole moments associated with thermal depopulation of a low lying Pr^{3+} energy level in the CEF. This is consistent with the Schottky anomaly in $C(T)/T$ and the peak in $\chi(T)$ near 3 K. Between 8.5 K and 45 K, $\rho(T)$ exhibits T^2 behavior, as shown in the lower inset of Fig. 5, consistent with Fermi liquid behavior. However, the coefficient $A = 0.009\,\mu\Omega\,\text{cm/K}^2$ of the T^2 term in $\rho(T)$ (*i.e.*, $\rho(T) = \rho_0 + AT^2$) is nearly two orders of magnitude smaller than the value expected for a heavy fermion compound with a γ of 500 mJ/mol K^2 (for heavy fermion compounds, the Kadowaki–Woods relation [23] $A/\gamma^2 = 1 \times 10^{-5}\,\mu\Omega\,\text{cm (mol K/mJ)}^2$ is usually satisfied). Moreover, the overall shape of the $\rho(T)$ curve does not resemble that of a typical heavy fermion compound, for which $\rho(T)$ is only weakly temperature dependent, often increasing with decreasing temperature, reminiscent of the Kondo effect, above a characteristic "coherence temperature", below which $\rho(T)$ decreases rapidly and then saturates as T^2 at low temperature as the heavy Fermi liquid ground state develops. It is noteworthy that the general shape of the $\rho(T)$ curve of $PrOs_4Sb_{12}$ is similar to that of $PrInAg_2$, in which quadrupolar fluctuations associated with a Γ_3 ground state are suspected to be responsible for the enormous electronic specific heat coefficient $\gamma \sim 6.5$ J/mol K^2 observed for that material [24]. Another possibility is that a T^2 term of magnitude comparable to that expected from the Kadowaki-Woods relation is obscured by the decrease in scattering below 7 K and SC below 1.8 K. Measurements of $\rho(T)$ in magnetic fields high enough to suppress SC indicate the existence of a low temperature region in which $\rho(T)$ has a strong power law dependence which can be best described as $\rho(T) = BT^3$ with $B = 0.88\,\mu\Omega$ cm/K^3 in the range 0.1 K $\leq T \leq$ 1.1 K.

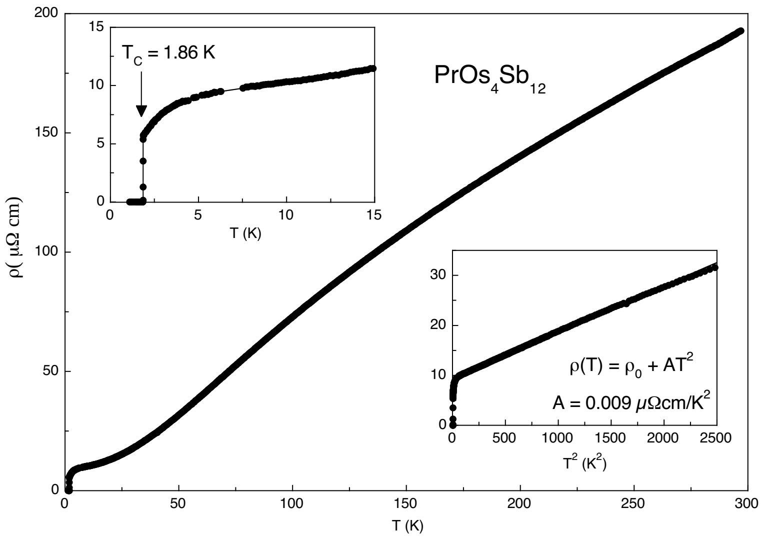

Fig. 5. Electrical resistivity ρ *vs* T for $PrOs_4Sb_{12}$ between 1.8 K and 300 K. Upper inset: $\rho(T)$ below 20 K. Lower inset: ρ *vs* T^2 below 50 K.

Measurements of the magnetoresistance in magnetic fields up to 80 kOe down to 60 mK with the current transverse to the magnetic field revealed a relatively high upper critical field $H_{c2}(T)$ with $H_{c2}(0) \sim 22$ kOe, consistent with heavy fermion behavior. Analysis of the slope of H_{c2} near T_c, $(-dH_{c2}/dT)T_c$, yields a value of ~ 19 kOe/K, from which a value for the orbital critical field at $T = 0$ K, $H^*_{c2}(0) \sim 24.5$ kOe, can be estimated. This, in turn, can be used to extract a superconducting coherence length of $\xi_0 \sim 116$ Å. Using an analysis similar to that employed for the heavy fermion superconductor UBe_{13} [25], values of the effective mass $m^* \sim 50m_e$ and $\gamma \sim 350$ mJ/mol K^2 are obtained. These values of m^* and γ are comparable to the estimates based on the specific heat jump at T_c and the normal state specific heat. Thus, it appears that $PrOs_4Sb_{12}$ may be the first example of a Pr-based heavy fermion superconductor. It is possible that the heavy fermion state is generated by the screening of the Pr^{3+} Γ_3 quadrupole moments by the charges of the conduction electrons. Such quadrupolar interactions could play some role in the pairing of superconducting electrons in this fascinating compound. The apparent absence of magnetic or quadrupolar order at temperatures down to ~ 60 mK suggests that $PrOs_4Sb_{12}$ may be near a magnetic or quadrupolar QCP.

2.2. Summary

The temperature-composition ($T-x$) phase diagram of the heavy fermion pseudoternary system $CeRh_{1-x}Co_xIn_5$ has been determined from experiments on $CeRh_{1-x}Co_xIn_5$ single crystals. The experiments reveal that superconductivity and antiferromagnetic order coexist over a range of Co concentrations $0.4 \leq x \leq 0.6$. The relatively weak variation of T_c with x in the vicinity of $x_c \sim 0.8$ where T_N appears to vanish in the $CeRh_{1-x}Co_xIn_5$ system is in marked contrast to the narrow peak in $T_c(P)$ in the vicinity of P_c, where T_N is suppressed to zero in the parent compound $CeIn_3$. Superconductivity has been observed in single crystals of the filled skutterudite compound $PrOs_4Sb_{12}$ below $T_c = 1.85$ K and appears to involve heavy fermion quasiparticles with an effective mass $m^* \sim 50 m_e$, where m_e is the mass of the free electron. It is possible that the heavy fermion state is generated by the screening of the Pr^{3+} Γ_3 quadrupole moments by the charges of the conduction electrons. The apparent absence of magnetic or quadrupolar order in $PrOs_4Sb_{12}$ down to ~ 60 mK suggests that this compound may be in the proximity of a QCP.

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