

# Search for heavy quasiparticles in the resistivity of $\text{PrOs}_4\text{Sb}_{12}$ in magnetic fields: Comparison with $\text{Pr}_{0.7}\text{La}_{0.3}\text{Os}_4\text{Sb}_{12}$

B. Andraka,\* C. R. Rotundu,† and K. Ingersent

*Department of Physics, University of Florida, P.O. Box 118440, Gainesville, Florida 32611-8440, USA*

(Received 30 September 2009; revised manuscript received 26 January 2010; published 16 February 2010)

We study the resistivity  $\rho(T)$  of  $\text{PrOs}_4\text{Sb}_{12}$  and  $\text{Pr}_{0.7}\text{La}_{0.3}\text{Os}_4\text{Sb}_{12}$  in magnetic fields between 2.7 and 16 T and at temperatures  $20 \text{ mK} \leq T \leq 1 \text{ K}$ .  $\text{PrOs}_4\text{Sb}_{12}$  is known to exhibit field-induced antiferroquadrupolar (AFQ) order while there is no evidence for such order in the specific heat of  $\text{Pr}_{0.7}\text{La}_{0.3}\text{Os}_4\text{Sb}_{12}$ . We find the resistivity of  $\text{Pr}_{0.7}\text{La}_{0.3}\text{Os}_4\text{Sb}_{12}$  to be consistent with the accepted crystalline electric field (CEF) model. The model predicts a low-temperature plateau in  $\rho(T)$  in fields smaller or larger than the CEF level-crossing field of approximately 9 T, as is also observed in  $\text{PrOs}_4\text{Sb}_{12}$ . However, further analysis suggests that for  $\text{PrOs}_4\text{Sb}_{12}$  in fields below 5 T, this plateau is not a CEF effect but is rather due either to a crossover to a state characterized by a small electronic effective mass  $m^*$  or to the existence of two resistivity contributions, one increasing and the other decreasing with temperature. In fields between 4 and 6 T,  $\rho(T)$  shows a shallow minimum. A Kadowaki-Woods analysis of  $\rho(T)$  of  $\text{PrOs}_4\text{Sb}_{12}$  over a restricted temperature range implies that  $m^*$  depends strongly on magnetic field, increasing when the field approaches the boundary of the AFQ region. However, for all fields investigated,  $\partial^2\rho/\partial T^2$  for  $T \rightarrow 0$  remains small compared with its value in canonical heavy fermions. In the AFQ regime (only), the resistivity has different temperature dependences in fields parallel and perpendicular to the principal crystal direction of current flow, suggesting an increase in the orbital contribution to the resistivity in the ordered phase.

DOI: 10.1103/PhysRevB.81.054509

PACS number(s): 74.25.F-, 74.70.Tx

## I. INTRODUCTION

$\text{PrOs}_4\text{Sb}_{12}$  has attracted considerable interest since it is believed to be the first-discovered<sup>1</sup> heavy-fermion superconductor containing Pr. What is more, the usual picture of heavy-fermion behavior arising from hybridization between  $f$  electrons and conduction electrons does not seem to apply to this material. The experimental evidence<sup>2-7</sup> that the Pr ions have a singlet crystalline electric field (CEF) ground state and the absence of signatures of Kondo physics rule out both the standard interpretation in terms of the Kondo lattice model<sup>8</sup> and an alternative scenario based on the quadrupolar Kondo effect.<sup>9</sup> In fact, the only convincing arguments for the presence of heavy quasiparticles in  $\text{PrOs}_4\text{Sb}_{12}$  are based on properties just below the superconducting critical temperature  $T_c = 1.85 \text{ K}$ : the large specific-heat discontinuity at  $T_c$ , and the steep slope of  $H_{c2}(T)$  as the temperature  $T$  approaches  $T_c$  from below. However, just above  $T_c$  the system can be well described in terms of purely localized  $f$  electrons and light conduction electrons. For instance, the electrical resistivity is dominated at all temperatures down to 2 K by scattering of conduction electrons by excited CEF levels and by phonons.<sup>10</sup> There is also no clear evidence for heavy fermions at temperatures  $T \ll T_c$ . Intensive searches for heavy masses below 600 mK using the de Haas-van Alphen (dHvA) technique have proved unsuccessful.<sup>11,12</sup> Thus, there is a significant possibility that heavy quasiparticles exist only in a very narrow range of temperatures near  $T_c$ . Elucidation of the nature of the heavy quasiparticles that develop suddenly below 2 K and the search for heavy-fermion behavior below 1 K represent high priorities.

Electrical resistivity and magnetoresistance have played prominent roles in the identification and exploration of heavy-fermion systems. In most such systems, the high-

temperature resistivity is of a Kondo-impurity type that hints at a heavy-fermion ground state at low temperatures. In the limit  $T \rightarrow 0$ , the resistivity of canonical heavy fermions acquires a Fermi-liquid temperature variation,

$$\rho(T) = \rho_0 + AT^2. \quad (1)$$

Kadowaki and Woods<sup>13</sup> noted a correlation  $A/\gamma^2 \approx 10^{-5} \Omega \text{ cm K}^2/\text{J}^2$  between the  $T^2$  resistivity coefficient and the electronic specific-heat coefficient  $\gamma$  of many concentrated (periodic) Ce- and U-based heavy-fermion materials.

Within the context of this pattern of conventional heavy-fermion behavior, the resistivity and magnetoresistance of  $\text{PrOs}_4\text{Sb}_{12}$  are anomalous.<sup>1</sup> The reported resistivity data have not provided a convincing argument for a heavy-fermion ground state. In zero field at temperatures between 7.5 and 45 K,  $\rho(T)$  has the expected Fermi-liquid variation but with a coefficient  $A$  of only  $0.009 \mu\Omega \text{ cm}/\text{K}^2$  (Ref. 14). This result would imply, through application of the Kadowaki-Woods relation, that  $\gamma \approx 30 \text{ mJ}/\text{K}^2 \text{ mol}$ —an order of magnitude smaller than the values estimated<sup>1,15</sup> from the specific heat of  $\text{PrOs}_4\text{Sb}_{12}$  above 7 K and one similar to the specific-heat coefficient measured<sup>11,16,17</sup> in the non- $f$ -electron analog  $\text{LaOs}_4\text{Sb}_{12}$ . When the superconductivity is suppressed with a magnetic field  $H > H_{c2}(T=0) \approx 2.3 \text{ T}$ , the resistivity seems to saturate<sup>1,15,18</sup> for  $T < 300 \text{ mK}$ .

For  $H = 3 \text{ T}$  and over a restricted temperature range  $0.5 \text{ K} \leq T \leq 1 \text{ K}$ ,  $\rho(T)$  can be approximated by Eq. (1) with an  $A$  value of order  $1 \mu\Omega \text{ cm}/\text{K}^2$ , consistent with the Kadowaki-Woods relation.<sup>1,15</sup> However, also at  $H = 3 \text{ T}$  and over the broader temperature window from 0.1 to 1.1 K, the resistivity can instead be fitted to<sup>1</sup>

$$\rho(T) = \rho_0 + BT^\alpha \quad (2)$$

with an exponent  $\alpha \approx 3$ , undermining the case for heavy-fermion behavior. Furthermore, fits to Eq. (1) ignore two potentially significant contributions to the resistivity: scattering from single-ion CEF excitations and collective fluctuations associated with field-induced long-range order. For fields between 4.5 and 14.5 T,  $\text{PrOs}_4\text{Sb}_{12}$  exhibits antiferro-quadrupolar (AFQ) order<sup>2-4,19</sup> up to temperatures as high as 1.3 K. This order is believed to arise from a crossing (or near crossing<sup>20</sup>) between 8 and 9 T of two CEF levels: the zero-field ground state (a  $\Gamma_1$  singlet) and one member of the zero-field first excited multiplet (a  $\Gamma_4^{(2)}$  triplet). Short-range AFQ fluctuations are expected to persist even to zero field and have been postulated to play an important role in the unconventional properties of  $\text{PrOs}_4\text{Sb}_{12}$ .

One motivation for the present work was to further investigate possible heavy-fermion behavior in  $\text{PrOs}_4\text{Sb}_{12}$  via resistivity measurements carried out over a wide range of magnetic fields. We have estimated the CEF contribution by direct numerical calculations within a theoretical model. We have compared our measurements with ones for a related alloy,  $\text{Pr}_{0.7}\text{La}_{0.3}\text{Os}_4\text{Sb}_{12}$ , which shows no evidence in its specific heat for any AFQ phase and exhibits a magnetoresistance at  $T \rightarrow 0$  that is well described by the CEF model.<sup>20</sup> Our results suggest that although CEF effects can explain certain aspects of  $\text{PrOs}_4\text{Sb}_{12}$ , such as the low-temperature plateau in  $\rho(T)$ , the resistivity is most likely dominated by fluctuations of the field-induced AFQ order parameter.

The other goal of this investigation was to explore the possibility of a significant variation in the electronic effective mass  $m^*$  with magnetic field. In particular, we have considered the possibility raised in Ref. 11 that a discrepancy between dHvA measurements performed in relatively high fields and the zero-field specific heat can be attributed to a field suppression of  $m^*$ . Our analysis indicates quite the opposite, i.e., that magnetic fields *enhance*  $m^*$ . However,  $\lim_{T \rightarrow 0} \partial^2 \rho / \partial T^2$  remains small for all applied fields.

## II. METHODS AND RESULTS

Samples were prepared, by a standard Sb-self-flux growth method, in the form of long bars with a rectangular cross section that allowed for a fairly accurate determination of the resistivity. Resistance measurements were performed directly in the mixing chamber of a dilution refrigerator in the ac four-probe configuration using a Linear Research LR-700 AC resistance bridge. In order to minimize possible Joule heating, current leads were spot welded at the far edges of the bars. Voltage leads, at least 1 mm away from current contacts, were silver epoxied. Probable errors, about 10% of measured values, were due to uncertainty in the separation of voltage contacts.

The residual resistivity ratio (RRR), defined as the ratio of the zero-field resistance at room temperature to that extrapolated to absolute zero, was in the range 70–80 for all our  $\text{PrOs}_4\text{Sb}_{12}$  crystals. The La-doped crystal with a nominal composition  $\text{Pr}_{0.7}\text{La}_{0.3}\text{Os}_4\text{Sb}_{12}$  had an RRR of approximately 120. In general, the RRR (as well as the residual resistivity

$\rho_0$ ) of  $\text{Pr}_{1-x}\text{La}_x\text{Os}_4\text{Sb}_{12}$  samples rises with increasing  $x$ , despite the large degree of atomic disorder induced by La doping. Flux-grown crystals of  $\text{Pr}_{1-x}\text{La}_x\text{Os}_4\text{Sb}_{12}$  show strong local variations in the Pr concentration within a single crystal, as observed in microprobe analysis.<sup>21</sup> This disorder also shows up as temperature-broadened superconducting transitions in the electrical resistivity. For instance, the investigated  $x=0.3$  crystal had an onset of superconductivity at 1.59 K and zero resistance at 1.43 K. Therefore, the standard assumption that larger RRRs are indicative of higher-quality crystals might not be valid for  $\text{Pr}_{1-x}\text{La}_x\text{Os}_4\text{Sb}_{12}$ .

All resistivity measurements were carried out with the current along the (001) crystallographic direction and with the magnetic field either parallel or perpendicular to the current. In the transverse configuration, the magnetic field was oriented along the (100) direction. The probable error in the field alignment was  $5^\circ$  for longitudinal measurements and  $10^\circ$  for transverse measurements. Previously, we have investigated a different  $\text{PrOs}_4\text{Sb}_{12}$  crystal with the magnetic field and current both approximately parallel to the (001) direction.<sup>18</sup> Some of the differences between our new results and previously reported data can be ascribed to misalignment of the crystal in the earlier study. However, there is also strong sample dependence, particularly in the ordered state. In our new measurements, special care was exercised to avoid overheating the crystal at the lowest temperatures. This possible overheating was checked by measuring at different alternating excitation currents below 300 mK.

Details of the CEF model can be found in Ref. 20. We used the singlet-ground-state CEF scheme determined from elastic<sup>3</sup> and inelastic<sup>5</sup> neutron scattering in  $\text{PrOs}_4\text{Sb}_{12}$ , and assumed equal prefactors for the contributions from aspherical Coulomb scattering and from magnetic exchange scattering. The latter choice is consistent both with the zero-field resistivity<sup>10</sup> of  $\text{PrOs}_4\text{Sb}_{12}$  above 2 K and with the field variation in the residual ( $T \rightarrow 0$ ) magnetoresistance<sup>20</sup> of  $\text{Pr}_{0.7}\text{La}_{0.3}\text{Os}_4\text{Sb}_{12}$ . This choice also best describes<sup>22</sup> the resistivity of  $\text{PrB}_6$ .

Figure 1 compares the low-temperature resistivity of  $\text{PrOs}_4\text{Sb}_{12}$  and  $\text{Pr}_{0.7}\text{La}_{0.3}\text{Os}_4\text{Sb}_{12}$  in fields of 3, 10, and 16 T along the (001) direction to the theoretical CEF resistivity  $\rho_{\text{CEF}}$  of Pr in this system. The La-doped crystal was one used previously to investigate the field variation in the residual resistivity.<sup>20</sup> For this composition, the specific heat showed no obvious anomalies that could be related to field-induced long-range order. Therefore, we expect its resistivity to be dominated by CEF excitations. As stated above, the CEF resistivity was calculated assuming equal prefactors for aspherical Coulomb scattering and exchange scattering. There is no fundamental reason to rule out different weightings of the two contributions. In fact, we find that the CEF resistivity at 16 T can be made somewhat closer to the resistivity of the  $x=0.3$  sample if magnetic exchange scattering is neglected altogether. However, since the change in the aspherical resistivity between 20 mK and 1 K is approximately the same for  $H=3$  and 16 T, omission of exchange scattering would be inconsistent with our experimental data for the same sample at 3 T. We have found no weighting that perfectly describes the electrical resistivity of the La-doped crystal for  $H=3$ , 10, and 16 T. Nonetheless, the relevance of the CEF resistivity

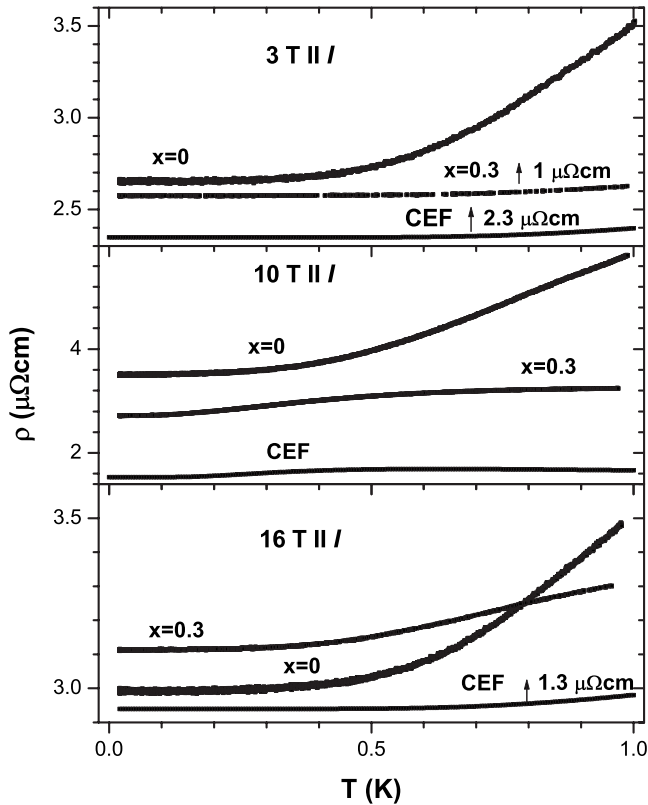


FIG. 1. Longitudinal low-temperature resistivity of  $\text{PrOs}_4\text{Sb}_{12}$  and  $\text{Pr}_{0.7}\text{La}_{0.3}\text{Os}_4\text{Sb}_{12}$ , as well as the theoretical CEF resistivity in fields of 3, 10, and 16 T applied parallel to the (001) current direction. The CEF and  $\text{Pr}_{0.7}\text{La}_{0.3}\text{Os}_4\text{Sb}_{12}$  resistivity curves are shifted upward by 2.3 and 1  $\mu\Omega\text{cm}$  for  $H=3$  T. The CEF resistivity curve for  $H=16$  T is shifted upward by 1.3  $\mu\Omega\text{cm}$ .

for this alloy is obvious. The overall temperature variation is consistent with the CEF model for all three field values. The model predicts the observed saturation of the resistivity at the lowest temperatures and also reproduces the broadened step in the 10 T data (middle panel of Fig. 1) provided that aspherical and exchange scattering are assigned roughly equal weights.

The relevance of the calculated CEF resistivity for the resistivity of  $\text{PrOs}_4\text{Sb}_{12}$  is much less obvious. The theory is clearly inconsistent with the experimental data at 10 T. This is to be expected since  $H=10$  T places the system deep in its AFQ phase and the CEF model takes into account only single-ion effects, neglecting the consequences of long-range order. However, the CEF resistivity also greatly underestimates  $\partial\rho/\partial T$  for  $T \geq 0.5$  K at both 3 and 16 T, i.e., above and below the range of fields exhibiting long-range AFQ order. One point of agreement between theory and experiment is the (near) saturation of the 3 T resistivity below 300 mK. This leveling off of the resistivity of  $\text{PrOs}_4\text{Sb}_{12}$  in fixed magnetic fields  $H > H_{c2}(0) \approx 2.3$  T has been noted previously<sup>1</sup> and remains to be understood.

The residual resistivity  $\rho_0$  of  $\text{Pr}_{0.7}\text{La}_{0.3}\text{Os}_4\text{Sb}_{12}$  (Fig. 2) exceeds the theoretical CEF resistivity at  $T=0$  by 1.5  $\mu\Omega\text{cm}$  for  $H=3$  and 16 T, and by 1.2  $\mu\Omega\text{cm}$  for  $H=10$  T, suggesting that the lowest-temperature resistivity of this alloy consists mainly of the field-dependent CEF con-

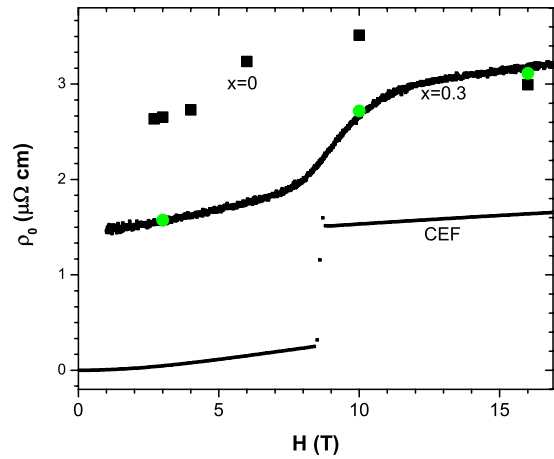


FIG. 2. (Color online) Residual resistivity vs field for  $\text{PrOs}_4\text{Sb}_{12}$  (squares) and  $\text{Pr}_{0.7}\text{La}_{0.3}\text{Os}_4\text{Sb}_{12}$  (large filled circles). Small dots correspond to the residual resistivity of  $\text{Pr}_{0.7}\text{La}_{0.3}\text{Os}_4\text{Sb}_{12}$  measured in Ref. 20 and reduced by 15%, in order to match the new measurement. Also, the theoretical CEF resistivity at 20 mK is shown.

tribution plus a field-independent<sup>23</sup> part due to crystalline defects. At 1 K, the highest temperature reached in our measurements, the difference between the measured and predicted resistivity remains in the range 1.5–1.6  $\mu\Omega\text{cm}$  for all three fields. For  $\text{PrOs}_4\text{Sb}_{12}$ , by contrast,  $\rho_0 - \rho_{\text{CEF}}(T=0) \approx 2.6, 2.0,$  and  $1.4$   $\mu\Omega\text{cm}$  in 3, 10, and 16 T, respectively. We know from our earlier investigation of the longitudinal field configuration<sup>20</sup> that this apparent monotonic decrease in  $\rho_0 - \rho_{\text{CEF}}(T=0)$  with increasing  $H$  extends beyond 16 T. These observations support our assertion that the residual resistivity measured in small fields and the zero-field RRR are poor indicators of the quality of  $\text{PrOs}_4\text{Sb}_{12}$  crystals.

We also note that in 3 T, the resistivity of  $\text{PrOs}_4\text{Sb}_{12}$  below 300 mK is larger than that of  $\text{Pr}_{0.7}\text{La}_{0.3}\text{Os}_4\text{Sb}_{12}$  but in 16 T, the undoped sample has the smaller low-temperature resistivity. Thus, even in the paramagnetic regime at 3 T, the resistivity of  $\text{PrOs}_4\text{Sb}_{12}$  seems to have an unknown component that is not directly related to CEF excitations and that depends on both magnetic field and temperature. At 1 K,  $\rho - \rho_{\text{CEF}} = 3.5, 4.3,$  and  $1.8$   $\mu\Omega\text{cm}$  for  $H=3, 10,$  and 16 T, respectively. The fact that the difference between the measured resistivity and the theoretical CEF resistivity is largest in the ordered state points to fluctuations of the AFQ order parameter as the source of the enhanced electron scattering in this field and temperature range.

Resistance measurements performed at 2.7 T—the lowest field of this study—using several different alternating excitation currents found the change in the resistivity of  $\text{PrOs}_4\text{Sb}_{12}$  between 20 and 200 mK to be no more than 0.001  $\mu\Omega\text{cm}$  (a bound set by the resolution of the ac bridge). Fitting to Eq. (1) places an upper limit on  $A$  of only 0.03  $\mu\Omega\text{cm K}^{-2}$ , a value that appears inconsistent with the existence of a heavy-fermion state for  $T \rightarrow 0$  in 2.7 T. We found similarly small changes in  $\rho(T)$  below 200 mK in fields of 3 and 3.5 T.

By contrast, the resistivity curves in fields between 4 and 6 T show minima at low temperatures rather than simple saturation. These minima were detected for both longitudinal

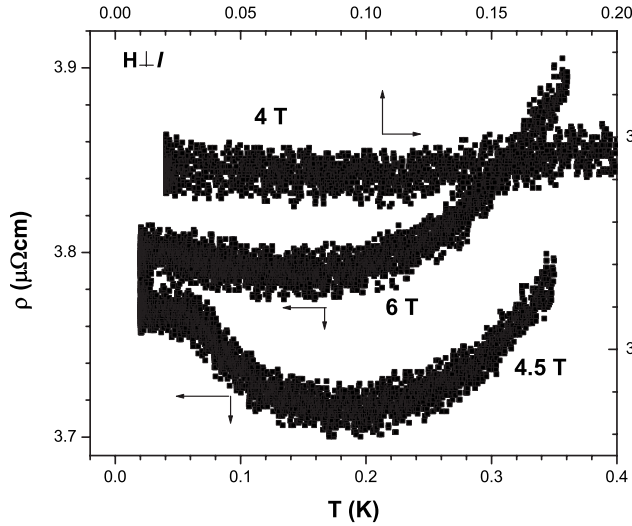


FIG. 3. Resistivity of  $\text{PrOs}_4\text{Sb}_{12}$  in fields of 4, 4.5, and 6 T, perpendicular to a current along the (001) direction. Data points for 6 T were shifted downwards by  $1.12 \mu\Omega \text{ cm}$ . Measurements performed both upon cooling and heating, and with different temperature-change rates, yielded identical results. The scatter in the data stems from small excitation currents and the absence of averaging.

and transverse configurations, independent of the rate of temperature change and of the magnetothermal history. These features are illustrated in Fig. 3 for three values of the field applied perpendicular to the current. To our knowledge, they were not reported in previous studies, which focused on somewhat higher temperatures. The minimum is particularly pronounced for  $H=4.5$  T in which field it is located at  $T=0.2$  K. The feature moves to a lower temperature when the field is reduced or increased. For 4 T, a very shallow minimum is still detectable at  $T \approx 0.1$  K but none can be resolved at 3.5 T. When the field is increased above 4.5 T, the minimum becomes shallower but its temperature decreases very slowly at an average rate of  $-0.023$  K/T. The highest field in which we are able to resolve the minimum is 6 T. The fact that this very low-temperature anomaly is most pronounced at 4.5 T suggests that it is related to the onset of field-induced long-range AFQ order. Magnetization measurements<sup>4</sup> indicate that in a 4.5 T field applied along the (001) direction, AFQ ordering sets in somewhere between 200 and 250 mK. When the system enters the low-temperature AFQ phase its resistivity increases. Although rather unusual, this behavior is consistent with magnetoresistance measurements at fixed temperatures.<sup>19,24</sup> However, the location  $T(H)$  of this resistivity minimum does not simply track the temperature of onset of AFQ order,<sup>2-4,19,24</sup> which increases rapidly between 4.5 and 5 T, and reaches approximately 1 K at 6 T. The persistence of a weak minimum down to 4 T is also at odds with the currently accepted  $H$ - $T$  phase diagram of  $\text{PrOs}_4\text{Sb}_{12}$ .

These results suggest that the nature of the low-temperature state of  $\text{PrOs}_4\text{Sb}_{12}$  may be more complex than hitherto assumed. The low-temperature plateau in  $\rho(T)$  may result from the addition of two contributions, one increasing with increasing temperature and the other decreasing. At temperatures sufficiently far above 200 mK, the resistivity

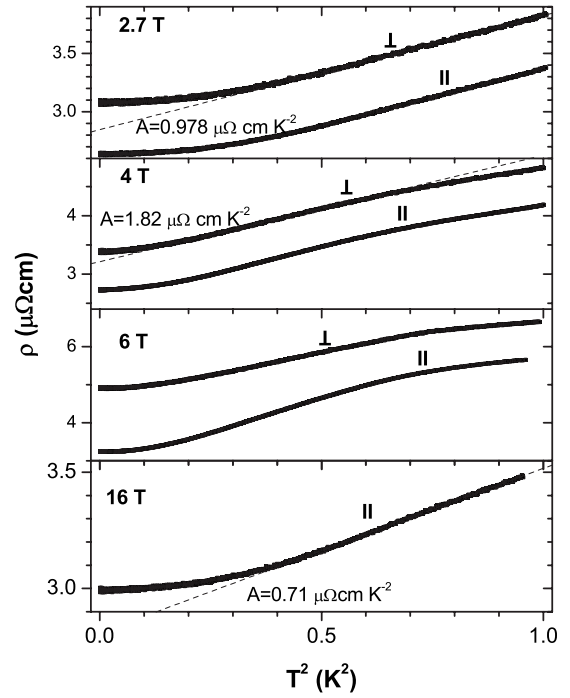


FIG. 4. Resistivity vs square of temperature for  $\text{PrOs}_4\text{Sb}_{12}$  in magnetic fields of 2.7, 4, 6, and 16 T applied parallel (||) or perpendicular ( $\perp$ ) to the (001) current direction. The broken lines represent least-squares fits over finite temperature intervals. See text for the discussion.

seems to exhibit Fermi-liquid character. Figure 4, upper panel, shows  $\rho$  vs  $T^2$  for a 2.7 T field in both the parallel and perpendicular configurations. For  $T^2 \geq 0.5$  K<sup>2</sup>, the data fall on parallel straight lines, yielding identical  $A$  coefficients. Very similar behavior is observed at other fields  $H < 4.5$  T. However, with increasing  $H$  there is a continuous decrease in the upper limit of the temperature window over which this quadratic variation holds. (For example, see the  $H=4$  T data in Fig. 4.) The departure from a  $T^2$  variation cannot be accounted for by CEF effects. Instead, it suggests the importance of fluctuations of the AFQ order parameter in fields significantly smaller than that ( $H=4.5$  T) at which long-range order first sets in. At the same time, the  $A$  coefficient increases monotonically between 2.7 and 4 T. It should be noted that the value  $A=1.1 \mu\Omega \text{ cm K}^{-2}$  extracted from the 3 T data is in agreement with other reported results,<sup>1,15</sup> which fall in the range  $1.0$ – $1.4 \mu\Omega \text{ cm K}^{-2}$ .

We do not believe that the  $A$  coefficients of  $\text{PrOs}_4\text{Sb}_{12}$  in fields between 2.7 and 4 T are significantly affected by possible CEF contributions to the resistivity. We have avoided a direct subtraction of the theoretical  $\rho_{\text{CEF}}$  since this quantity is known only approximately (due to the aforementioned uncertainty in the prefactors of aspherical and exchange scattering). Subtraction of the theoretical estimate of  $\rho_{\text{CEF}}$  with equal exchange and aspherical scattering prefactors results in a greater deviation from Fermi-liquid  $T^2$  behavior than that seen in the raw resistivity but the effect is quite small. At 2.7 T, for instance, the least-squares fit of  $\rho_{\text{CEF}}$  vs  $T^2$  between  $T^2=0.4$  and 1 K<sup>2</sup> (the same temperature interval over which  $\rho$  vs  $T^2$  was fitted) yields  $A_{\text{CEF}} < 0.08 \mu\Omega \text{ cm K}^{-2}$ , which

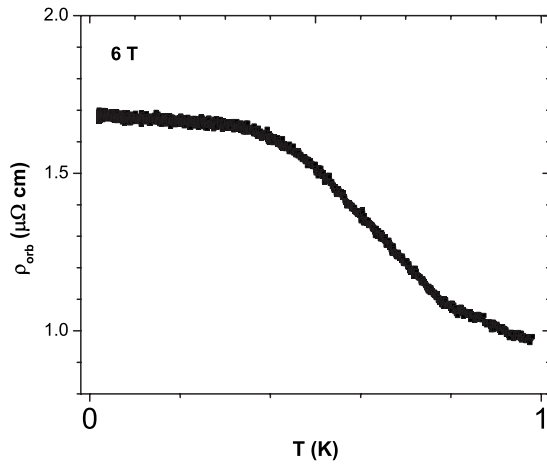


FIG. 5. Difference between transverse and longitudinal resistivity for  $\text{PrOs}_4\text{Sb}_{12}$  in a 6 T magnetic field.

is negligible compared with the full  $A$  coefficient of  $1.1 \mu\Omega \text{ cm K}^{-2}$ .

A similar Fermi-liquid-type analysis cannot be performed consistently within the ordered state of  $\text{PrOs}_4\text{Sb}_{12}$ . For fields between 4.5 and 15 T, measurements on three different  $\text{PrOs}_4\text{Sb}_{12}$  crystals yielded significantly different results. Some of these differences might be related to sample misalignment with respect to the direction of the applied magnetic field. The importance of the field orientation in the ordered phase is illustrated in Fig. 4 for  $H=6$  T, where the longitudinal and transverse resistivities have different temperature variations. In both cases, the current was along the principal (001) direction. In the transverse measurement, the field was approximately along the (100) principal direction. Figure 5 plots the difference between the transverse and longitudinal resistivities as a function of temperature. We have obtained very similar results for  $H=10$  T. At each field, the longitudinal and transverse resistivities move further apart below the AFQ ordering temperature. This suggests that orbital effects, which affect the transverse transport much more than the longitudinal transport, become markedly stronger on entry to the ordered state. Such behavior in the orbital resistivity is consistent with the generally accepted picture of long-range AFQ order.

The highest field used in this investigation was 16 T. Only the longitudinal resistivity was measured at this field. The plot of  $\rho$  vs  $T^2$  (lowest panel of Fig. 4) is similar in appearance to that for 2.7 T (the top panel of Fig. 4). The  $T^2$  coefficient extracted over approximately the same temperature range as that for 2.7 T is  $A \approx 0.7 \mu\Omega \text{ cm K}^{-2}$ . Our previous measurements<sup>18</sup> on a different crystal of  $\text{PrOs}_4\text{Sb}_{12}$  indicated that the resistivities for fields between 15 and 18 T have similar temperature variations to that shown in the lowest panel of Fig. 4 for  $H=16$  T, with  $A$  (and also  $\rho_0$ ) monotonically decreasing with  $H$ . However, even in these strong fields, the resistivity shows a characteristic plateau at the lowest temperatures. This high-field plateau is most likely related to CEF effects. However, straightforward subtraction of  $\rho_{\text{CEF}}$  does not entirely eliminate the plateau.

Figure 6 summarizes the results of our analysis for four different measurements of  $\text{PrOs}_4\text{Sb}_{12}$ . No data for the or-

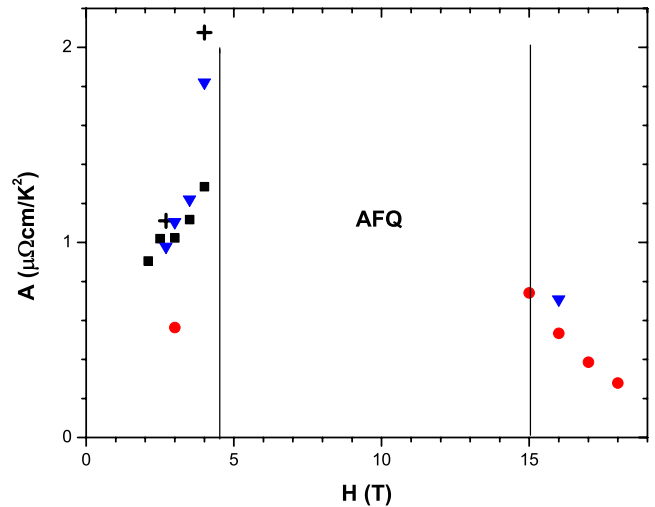


FIG. 6. (Color online) Resistivity coefficient  $A$  entering Eq. (1) vs magnetic field  $H$  along the (001) direction. Data from four different measurements are plotted with different symbols. Triangles and crosses represent two different crystals studied in the present work. A third crystal was investigated previously (Ref. 18), first in a dilution refrigerator down to 20 mK (dots) and then, after reattachment of leads, in a  $^3\text{He}$  refrigerator between 0.35 and 1 K (squares). Solid vertical lines show the boundaries of the AFQ phase as inferred from the magnetization (Ref. 4). See text for a discussion of the outlying  $A$  value at 3 T.

dered phase are included because, as mentioned above, the resistivity is sample dependent, depends on the orientation of the sample with respect to the field, and  $\rho(T)$  can be approximated by Eq. (1) only over narrow temperature intervals. A significantly lower value at 3 T for one of the measurements (plotted with a dot in Fig. 6) is most likely due to the very limited temperature range of the fit<sup>18</sup> (0.55–0.7 K) and an error in a geometrical factor. The average of  $A$  over the remaining three measurements at 3 T is in good agreement with other studies.<sup>1,15</sup>  $A$  is largest near the AFQ boundaries and decreases when the field is increased above 15 T or reduced below 4 T. A straight-line extrapolation yields a zero-field value of approximately  $0.5 \mu\Omega \text{ cm K}^{-2}$ .

### III. CONCLUSIONS

We find that the low-temperature resistivity of the nonordering alloy  $\text{Pr}_{0.7}\text{La}_{0.3}\text{Os}_4\text{Sb}_{12}$  is consistent with the accepted CEF model over a wide range of magnetic fields. The CEF model predicts the leveling off of the resistivity at low temperatures in fields smaller or larger than the theoretical crossing field. These plateaus are observed in  $\text{Pr}_{0.7}\text{La}_{0.3}\text{Os}_4\text{Sb}_{12}$  below 0.3 K for fields smaller than 5 T or larger than 13 T.

Low-temperature resistivity plateaus have also been observed for  $\text{PrOs}_4\text{Sb}_{12}$ . However, it is argued that for fields smaller than 5 T, these features are not entirely due to CEF effects. The residual resistivity values are too large and exhibit anomalous field variation.<sup>20</sup> Furthermore, we observe low-temperature minima in the resistivity of  $\text{PrOs}_4\text{Sb}_{12}$  in fields between 4 and 6 T. The existence of such minima suggests that there is a contribution to the resistivity that

increases with a decrease in temperature. The presence of contributions both increasing and decreasing with temperature might lead to approximately constant resistivity at low temperatures in these relatively small fields. The other possible explanation of these low-temperature plateaus is that the system crosses over at the lowest temperatures to a state with a very small effective mass, which would be consistent with the small cyclotron masses observed in dHvA measurements.

The resistivity in fields smaller than 4 T and at temperatures above 0.5 K provides evidence for the heavy-fermion character of  $\text{PrOs}_4\text{Sb}_{12}$ . However, the behavior as  $T \rightarrow 0$  remains a puzzle. The large number of anomalies<sup>25–28</sup> reported in the temperature range from 0.4 to 0.7 K may signal a transition or a crossover to a new state. Our observation of resistivity minima over a significant range of magnetic fields points to still greater complexity of the ground state of  $\text{PrOs}_4\text{Sb}_{12}$ . The presence of a weak minimum in  $\rho(T)$  in 4 T, and presumably also in lower fields, suggests that AFQ fluctuations persist well outside the AFQ domain. Strong AFQ fluctuations in the paramagnetic state have recently been postulated<sup>29</sup> based on the observation of anisotropy in the specific heat.<sup>30</sup>

Our analysis also suggests a strong-field variation in the effective mass  $m^*$ , as summarized in Fig. 6. This variation poses some challenge to the interpretation of dHvA measurements since it rules out the analysis of quantum oscillations over extended field ranges. Recent unpublished dHvA

results,<sup>12</sup> when analyzed<sup>31</sup> over narrow field intervals, indeed confirm changes in  $m^*$  with  $H$ , consistent with our analysis. However, the  $m^*$  values are still an order of magnitude too small to be compatible with the zero-field specific-heat data near 2 K. Our results essentially rule out the scenario of magnetic fields suppressing a large zero field  $m^*$ .

A similar analysis of the temperature variation in the resistivity in fixed magnetic fields was previously applied to the heavy-fermion superconductor  $\text{CeCoIn}_5$  (Ref. 32) and to the high  $T_c$  superconductor  $\text{Tl}_2\text{Ba}_2\text{CuO}_{6+x}$  (Ref. 33). In both cases, the temperature range over which the resistivity has Fermi-liquid character and the corresponding  $A$  coefficient both depend on magnetic field. The  $T^2$  range becomes very narrow while  $A$  grows large near a quantum phase transition. A similar, although more complicated, behavior is observed in  $\text{PrOs}_4\text{Sb}_{12}$  when the field is increased from 0 to 4.5 T suggesting the importance of quantum criticality.

#### ACKNOWLEDGMENTS

This work has been supported by the U.S. Department of Energy under Grant No. DE-FG02-99ER45748 (C.R.R. and B.A.), by the National Science Foundation under Grant No. DMR-0710540 (K.I.), and by the National High Magnetic Field Laboratory, supported jointly by the National Science Foundation, the U.S. Department of Energy, and the State of Florida.

\*andraka@phys.ufl.edu

<sup>†</sup>Present address: Materials Science Division, Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA.

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<sup>23</sup>The theoretical residual resistivity  $\rho_{\text{CEF}}(T=0)$  undergoes a jump between 8 and 9 T due to the crossing of the lowest CEF levels. In  $\text{Pr}_{0.7}\text{La}_{0.3}\text{Os}_4\text{Sb}_{12}$ , this step is broadened (and possibly shifted to slightly larger fields) so that the experimental  $\rho_0$  vs  $H$  is still rising at  $H=10$  T. This difference in behavior (illustrated in Fig. 3 of Ref. 20) explains why  $\rho_0 - \rho_{\text{CEF}}(T=0)$  is somewhat smaller at 10 T than at 3 and 16 T.

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