Heavy-fermion characteristics of the resistivity of $PrOs_4Sb_{12}$

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We present resistivity data at low temperatures down to 20 mK for $PrOs_4Sb_{12}$ in magnetic fields up to 18 T applied parallel to the current. Analysis of the resistivity at fixed fields in terms of the Fermi-liquid and Kadowaki-Woods models implies that the electronic effective mass m^* depends both on temperature T and magnetic field H. There is evidence for strong antiferroquadrupolar (AFQ) fluctuations in regions of the H-T phase diagram well beyond the long-range-ordered AFQ phase. We suggest that various anomalies reported at temperatures 0.4–0.7 K are signatures of a crossover that is responsible for a reduction of m^* as $T \to 0$.

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I. INTRODUCTION

Electrical resistivity and magnetoresistance have played prominent roles in the identification and exploration of heavy-fermion systems. In most such systems—primarily materials based on on Ce, Yb, and U—the high-temperature resistivity is of a Kondo-impurity type that hints at a possible heavy-fermion ground state. In the limit of temperatures $T \to 0$, the resistivity of heavy fermions acquires a Fermi-liquid temperature variation,

$$\rho(T) = \rho_0 + AT^2. \tag{1}$$

Kadowaki and Woods have noted[1] a correlation between the resistivity coefficient A and electronic specific heat coefficient γ, such that A/γ^2 is of order $10^{-5}\,\Omega\,\mathrm{cm\,K^2/J^2}$. This correlation persists in magnetic fields.[2] The sign of the magnetoresistance determines whether the heavy-fermion state is coherent or incoherent.[3]

Set against this pattern of conventional heavy-fermion behavior, the resistivity and magnetoresistance of $PrOs_4Sb_{12}$ are anomalous.[4] The reported resistivity data have not provided any evidence for a heavy-fermion ground state. At temperatures between 7.5 K and 45 K, ρ has the expected Fermi-liquid variation, but with a coefficient A of only $0.009\,\mu\Omega\,\mathrm{cm/K^2}$. This result implies, through the Kadowaki-Woods relation, that $\gamma \sim 30\,\mathrm{mJ/K^2\,mol}$—a surprisingly low value consistent with the specific heat coefficient[5–7] of $LaOs_4Sb_{12}$, a non-f-analog of $PrOs_4Sb_{12}$. When the superconductivity is suppressed with a magnetic field $H \geq H_{c2}(T = 0) = 2.2\,\mathrm{T}$, the resistivity seems to saturate for $T \lesssim 50\,\mathrm{mK}$. It can be fitted to a power law,[4]

$$\rho(T) = \rho_0 + BT^{\alpha}, \tag{2}$$

but with an exponent $\alpha > 3$. The only evidence for heavy-fermion behavior[4] (with $\gamma \sim 350\,\mathrm{mJ/K^2}$) comes from the large value of the specific-heat discontinuity at the superconducting critical temperature $T_c = 1.85\,\mathrm{K}$ and from the steep slope of $H_{c2}(T)$ as T approaches T_c from below. Intensive searches[6,8] for heavy-fermion masses below 600 mK using the de Haas-van Alphen (dHvA) technique have been unsuccessful.

Three scenarios might explain these apparent inconsistencies in the properties of $PrOs_4Sb_{12}$: (1) The heavy-fermion state might exist only at zero field. Magnetic fields could suppress the large effective mass m^*, making the results of zero-field measurements such as the specific heat appear to be inconsistent with those of dHvA and resistivity measurements performed at relatively large or overcritical magnetic fields. (2) The strong resonant electron scattering leading to an enhanced m^* might take place only in a narrow range of temperatures, somewhere above 0.4–0.6 K. (3) The heavy-fermion behavior in Pr-based metals might be of a fundamentally different character from that in canonical systems. The last scenario is rendered unlikely by the fact that $PrFe_4P_{12}$—a material closely related to $PrOs_4Sb_{12}$—exhibits low-temperature properties[9,10] consistent with conventional heavy-fermion behavior: the ratio A/γ^2 measured in magnetic fields falls in the Kadowaki-Woods range, while cyclotron electron masses determined by the dHvA technique are among the highest ever reported. There is therefore a need to re-examine the electrical resistivity of $PrOs_4Sb_{12}$, paying attention to the first two scenarios, *i.e.*, the influence of magnetic field on the effective mass, and the possibility of a reduction in m^* at very low temperatures.

Our analysis of the low-temperature resistivity rules out the first scenario above (because the effective mass is enhanced, rather than suppressed, by the application of a magnetic field), but strongly supports the second scenario: $PrOs_4Sb_{12}$ is in a heavy-fermion state only in a narrow range of temperatures, roughly between 0.5 K and 3 K. Various unexplained anomalies at temperatures $T \simeq 0.5\,\mathrm{K}$ are most likely related to a crossover between heavy-fermion and non-heavy-fermion behavior as $T \to 0$.

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. The crystals used in this

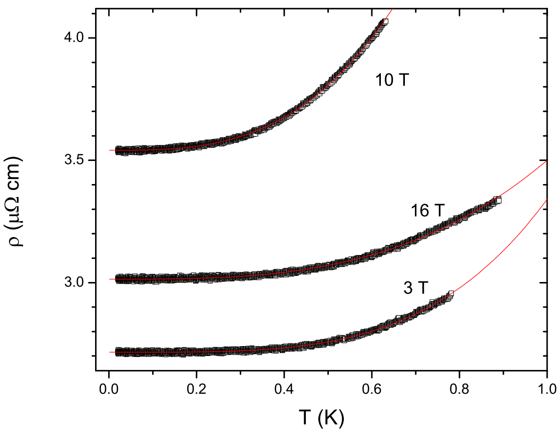

FIG. 1: (color online) Low-temperature resistivity of $\mathrm{PrOs_4Sb_{12}}$ at three representative fields applied along the (001) direction, together with fits to the form of Eq. (2). The fitted temperature exponent takes the the values $\alpha = 3.9$, 3.03, and 3.23 for $H = 3$, 10, and 16 T, respectively.

study had a residual resistivity ratio (the ratio of the zero-field resistance at room temperature to that extrapolated to absolute zero) in the range 70-80, indicative of high sample quality. All resistivity measurements were carried out with magnetic fields applied along the (001) crystallographic direction and parallel to the electric current. Measurements in fields of 3, 10, 15, 16, 17, and 18 T were made on one sample in a dilution refrigerator, down to 20 mK. Measurements for other fields between 2 and 14 T were performed on a second sample in a ^{3}He cryostat, down to about 0.35 K. Some of the experimental data have previously been reported in Ref. 11. In agreement with the results of Bauer *et al.*,[4] the temperature variation of the zero-field resistivity above 7 K—and in our case, up to at least 30 K—can be accurately described by Eq. (1) with a small coefficient $A \simeq 0.009\,\mu\Omega\,\mathrm{cm/K^2}$.

Figure 1 shows the low-temperature resistivity in fields of 3, 10, and 16 T, corresponding to three different regions of the field-temperature phase diagram. At 10 T and below 1.3 K, $\mathrm{PrOs_4Sb_{12}}$ is in a long-range-ordered (LRO) state. The order has been identified as antiferroquadrupolar, with accompanying antiferromagnetic order of small, field-induced moments.[12] The 3-T and 16-T curves correspond to paramagnetic phases. The temperature variation of the resistivity in all three cases, as well as for other studied fields, can be well described by Eq. (2), the form proposed by Bauer *et al.*[4] to describe the situation in fields transverse to the current. However, the exponent α varies appreciably, and non-monotonically, with increasing field strength. Also, the exponent $\alpha \simeq 3$ for $H = 3\,\mathrm{T}$ reported in Ref. 4 is significantly smaller than our value $\alpha \simeq 3.9$ for the same field. What is more, the temperature variation in Eq. (2) has no theoretical justification.

TABLE I: Value of the exponent α, obtained by fitting the resistivity at fixed field H to Eq. (2) over the temperature range between 20 mK and different upper bounds T_{max}. With decreasing T_{max}, α seems to approach its Fermi-liquid value of 2.

$H = 10\,\mathrm{T}$

T_{max}(K)	0.63	0.5	0.45	0.4	0.35	0.3	0.25
α	3.03	3.09	3.03	2.88	2.66	2.45	2.31

$H = 15\,\mathrm{T}$

T_{max}(K)	0.7	0.6	0.5	0.4	0.35	0.3
α	3.01	2.97	2.86	2.63	2.41	2.32

$H = 16\,\mathrm{T}$

T_{max}(K)	0.83	0.6	0.5	0.4
α	3.23	3.19	2.91	2.35

$H = 18\,\mathrm{T}$

T_{max}(K)	0.8	0.7	0.6	0.5	0.4
α	3.84	3.77	3.32	2.95	2.86

Closer inspection reveals systematic deviations of the measured resistivity from the power-law description. These deviations become apparent when ρ vs T for a fixed field is fitted to Eq. (2) between our lowest temperature of 20 mK and some upper temperature T_{max}. The value of α obtained from such a fit depends on T_{max}. For instance, fitting the 10-T data up to $T_{\mathrm{max}} = 0.5\,\mathrm{K}$ yields $\alpha = 3.09$. However, reducing T_{max} lowers the value of α, such that $\alpha = 2.25$ for $T_{\mathrm{max}} = 0.25\,\mathrm{K}$. We observe the same trend at all investigated fields. The results of the fitting procedure for four fields[13] are summarized in Table I. These data argue against the use of a single exponent to describe the temperature variation of the resistivity below 1 K. They also strongly suggest that if the upper temperature of the fit is sufficiently small, the resistivity can be described by the usual Fermi-liquid formula, Eq. (1).

Above 2 K, the zero-field resistivity[14] of $\mathrm{PrOs_4Sb_{12}}$ seems to be dominated by scattering from excited crystalline electric field (CEF) levels, in all likelihood because of small CEF energies and the ground state being a singlet. This has motivated us to calculate the temperature variation of the CEF resistivity ρ_{CEF} for several magnetic fields at temperatures below 1 K. Details of the model can be found in Ref. 15. Theoretical ρ_{CEF} vs T curves were calculated assuming a singlet CEF ground state and equal weights $\rho_A = \rho_{\mathrm{ex}} = 0.25\,\mu\Omega\,\mathrm{cm}$ for the contributions from aspherical Coulomb scattering and from magnetic exchange scattering. These choices are consistent both with the higher-temperature[14] (above 2 K) variation of the zero-field resistivity of $\mathrm{PrOs_4Sb_{12}}$ and with the field-variation of the residual ($T \to 0$)

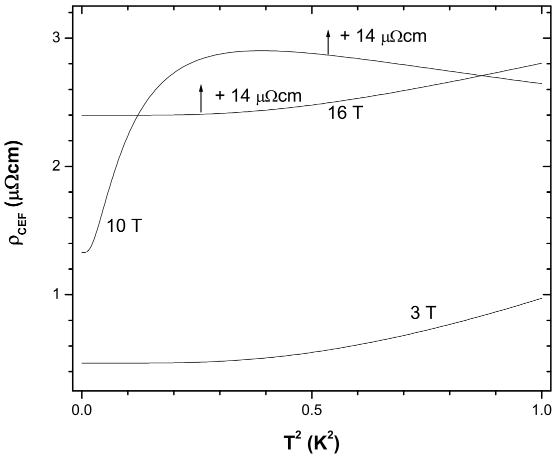

FIG. 2: Calculated crystal-field resistivity ρ_{CEF} vs T^2 for magnetic fields $H = 3$, 10, and 16 T along the (001) direction. The 10-T and 16-T curves have been shifted downward by 14 $\mu\Omega$ cm to allow them to be plotted on the same scale.

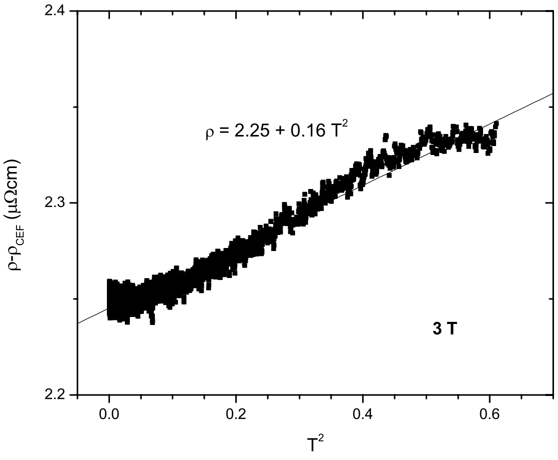

FIG. 3: Difference $\rho - \rho_{\mathrm{CEF}}$ vs T^2 for $\mathrm{PrOs_4Sb_{12}}$ in a 3-T field. The straight line is a least-squares fit.

magnetoresistance[15] of $\mathrm{Pr_{0.7}La_{0.3}Os_4Sb_{12}}$.

Figure 2 shows ρ_{CEF} vs T for $H = 3$, 10, and 16 T. The 3-T curve resembles the measured resistivity ρ at the same field. The difference $\rho - \rho_{\mathrm{CEF}}$, plotted in Fig. 3, can be reasonably well approximated between 0.4 K and 0.7 K by a quadratic function of temperature, although systematic deviations are seen outside this temperature window. Interestingly, the least-squares fit forced through all the points shown in Fig. 3 yields T^2 coefficient $A = 0.16 \mu\Omega\mathrm{cm\,K^{-2}}$, more than an order of magnitude greater than the $A \simeq 0.009\,\mu\Omega\,\mathrm{cm/K^2}$ extracted from the zero-field resistivity between 7.5 and 45 K. This larger A corresponds in the Kadowaki-Woods picture to a specific heat coefficient $\gamma \sim 400$–$500\,\mathrm{mJ/K^2\,mol}$.

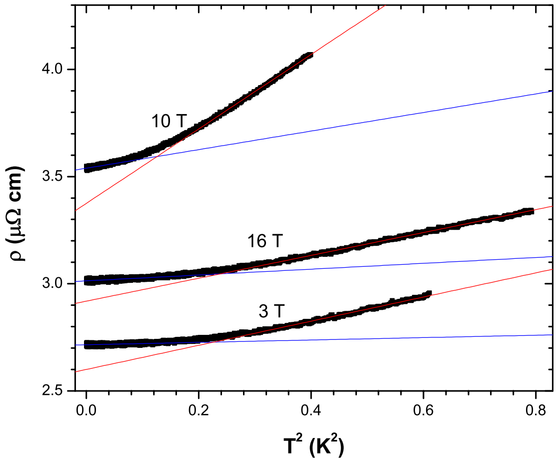

FIG. 4: (color online) Resistivity ρ versus T^2 for $\mathrm{PrOs_4Sb_{12}}$ in fields of 3, 10, and 16 T. Straight lines are least square fits for very low ($T < 0.2$ K) and intermediate ($0.5\,\mathrm{K} < T < 1\,\mathrm{K}$) temperatures, with slopes A_1 and A_2, respectively. The fitted values are (in order of increasing field) $A_1 = 0.06$, 0.51, and 0.14 $\mu\Omega\,\mathrm{cm/K^2}$, and $A_2 = 0.56$, 1.9, and $0.53\,\mu\Omega\,\mathrm{cm/K^2}$.

The agreement between CEF theory and experiment becomes much worse at higher magnetic fields. First of all, the measured ρ below 1 K is significantly smaller than the calculated ρ_{CEF}. (We note that in Fig. 2 the ρ_{CEF} curves for $H = 10$ and 16 T have been shifted down by $14\,\mu\Omega$ cm.) Also, the temperature variation of ρ_{CEF} is in strong disagreement with the measured 10-T resistivity. At 10 T and below 1.3 K this system is in the LRO state, so we expect that the ordering accounts for the marked decrease of ρ relative to ρ_{CEF}. It seems likely that for this field the temperature variation of ρ below 1 K is determined mainly by scattering of conduction electrons from fluctuations of the long-range order parameter, rather than from excited CEF levels.

As we demonstrate below, the resistivity has similar temperature variation below 1 K at all investigated magnetic fields, irrespective of whether the system is paramagnetic or in the LRO state. In particular, it is possible to identify two different regimes of Fermi-liquid behavior.

First, in all cases Eq. (1) is obeyed at the lowest temperatures, from 20 mK to approximately 200 mK. We denote the T^2 coefficient fitted over this range by A_1. Figure 4 shows ρ versus shows this behavior at 3, 10, and 16 T, for the crystal investigated in a dilution refrigerator.

Second, Bauer *et al.*,[4] have reported that the resistivity at 3 T (in a transverse configuration) satisfies Eq. (1) between 0.5 and 1.0 K, and that the corresponding coefficient A is consistent with the magnitude of the specific-heat jump at the superconducting critical temperature. We too find that the resistivity at intermediate temper-

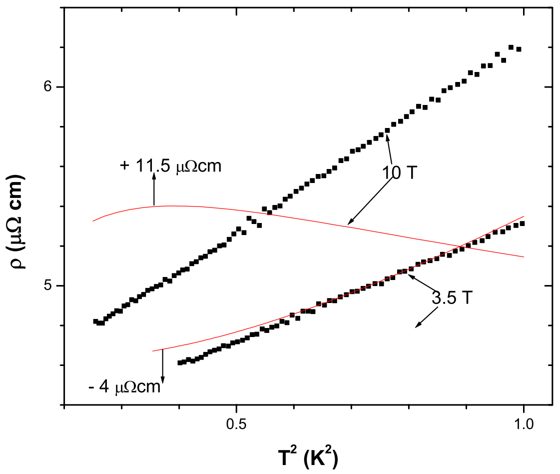

FIG. 5: (color on line) Measured resistivity ρ versus T^2 for $\mathrm{PrOs_4Sb_{12}}$ at fields of 3.5 T and 10 T. These data, obtained for the crystal investigated in a ^{3}He cryostat down to 0.3 K, demonstrate the existence of a regime of intermediate temperatures within which Eq. (1) holds. Solid lines correspond to the theoretical ρ_{CEF} offset by 4 and -11.5 $\mu\Omega$ cm for 3.5 and 10 T, respectively.

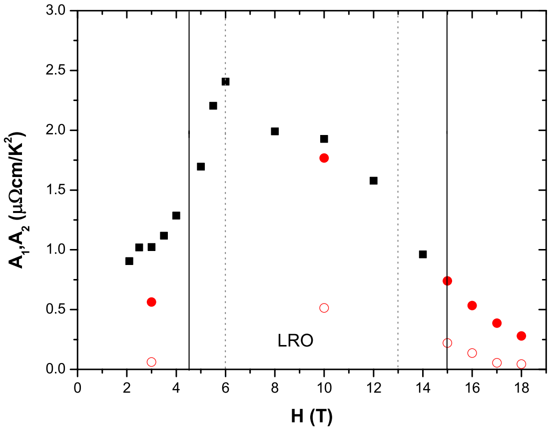

FIG. 6: (color online) Resistivity coefficients A_1 (open symbols) and A_2 (closed symbols) vs magnetic field H for $\mathrm{PrOs_4Sb_{12}}$. Circles correspond to the crystal investigated in a dilution refrigerator, squares to the crystal measured in a ^{3}He cryostat (A_2 only). Solid vertical lines show the antiferroquadrupolar (AFQ) to paramagnetic phase boundaries inferred from the magnetization. Specific heat measurements indicate a somewhat smaller AFQ region. Pronounced peaks due to the AFQ transition are clearly observed between the magnetic fields represented by dotted vertical lines. See text for explanation and discussion.

atures, say 0.5 K $< T <$ 1 K, can be satisfactorily described by Eq. (1) at all investigated magnetic fields between 2 T and 18 T. We denote the corresponding T^2 coefficient by A_2. The temperature window within which Eq. (1) holds is only weakly field-dependent. Figure 4 shows this behavior for the sample studied in a dilution refrigerator. Figure 5 presents ρ versus T^2 at 3.5 and 10 T for the second sample, measured in a ^{3}He cryostat, together with the corresponding ρ_{CEF}. (Note that each ρ_{CEF} curve has been shifted vertically to fit on the scale of the experimental data.) This limited T^2 variation of ρ seems to be characteristic of all $\mathrm{PrOs_4Sb_{12}}$ samples. In addition to Bauer *et al.*[4] and ourselves, Aoki *et al.*[16] have found such a temperature variation of the resistivity for magnetic fields of 2 and 3 T transverse to the current direction.

In a fixed magnetic field, then, there are two distinct temperature regimes below 1 K within which ρ can be described as a quadratic function of T. Slope A_1 describes the regime 20 mK $< T <$ 200 mK (and presumably characterizes the $T \to 0$ behavior), while slope A_2 governs the window $0.5 < T < 1$ K. The ratio A_2/A_1 changes with H, such that, when the resistivity is instead approximated by Eq. (2), the value of the exponent α depends not only on the range of the fit but also on the field. The fitting parameter α is particularly large for small fields (say, 3 T), since there A_2/A_1 is large (of order 10).

The data shown in Fig. 4 can be used to extract a crossover temperature corresponding to the intersection of the straight lines representing the two T^2 regimes. This temperature varies weakly with field between 3 and 16 T, remaining between 0.35 and 0.5 K. Interestingly, there have been a number of unexplained anomalies reported in the range 0.4–0.5 K. Sb nuclear quadrupole resonance measurements[17] have found a maximum in T_1 versus T at $T = 0.45$ K. Electronic specific heat measurements[18] show a shoulder, or a weak maximum, near 0.4 K. There have also been anomalies reported at somewhat higher temperatures, but not much above 0.5 K. The London penetration depth exhibits[19] a sudden decrease below 0.5 K. A pronounced enhancement of the lower critical field H_{c1} has been observed[20] below 0.7 K. These anomalies may all be plausibly explained by a reduction of the electronic effective mass at low temperatures.

This analysis of the resistivity in terms of a Fermi-liquid model provides a method for probing the possible variation of the electronic effective mass m^* as a function of magnetic field strength and temperature. A_1 and A_2 for the two investigated crystals are shown in Fig. 6. A_1 takes values of 0.06, 0.51, 0.22, 0.14, and 0.04 $\mu\Omega\,\mathrm{cm/K^2}$ in fields of 3, 10, 15, 16, and 18 T, respectively. Probable errors are 10% of the quoted values, except for $H = 3$ T, where the error may be as much as 50% due to the relatively small changes in the sample resistance below 200 mK and the high electrical noise at 3 T of the superconducting magnet employed in the experiment. The Kadowaki-Woods relationship implies that γ is less than 100 mJ/K^2 mol at 3 T. Considering that A_1 increases between 3 and 10 T, we expect the low-temperature, zero-field γ to be even smaller. A_1, and probably γ, is a non-monotonic function of field, going through a maximum somewhere in the ordered state, and decreasing above 15 T. A_2, which was determined for a larger number of magnetic fields, initially increases with H. It exhibits a peak at 6 T, superimposed on a wide maximum centered near 8 T.

A_1 is always smaller than A_2. Within the Kadowaki-Woods approach, this suggests a crossover from a lower effective mass below 0.2 K to a higher m^* at temperatures of order 1 K. Recent dHvA measurements seem to provide support for this scenario.[8] First, the low-temperature effective mass for the β Fermi-surface sheet[21] exhibits a broad maximum at 8 T, with about a 40% increase of m^* between 3 and 8 T. This field variation is in agreement with the trends observed both for A_1 and A_2. Second, for a field of 6.6 T applied in the (110) direction, there is an unusually rapid decrease above 0.6 K in the magnitude of quantum oscillations corresponding to the β sheet. One possible interpretation of this observation is that the cyclotron mass increases above 0.6 K.

A similar analysis of the temperature and field variation of the resistivity has previously been applied to the heavy-fermion superconductor[22] $CeCoIn_5$ and to the high-T_c superconductor[23] $Tl_2Ba_2CuO_{6+x}$. In both cases, at fields over-critical for superconductivity, the low-temperature resistivity acquires a Fermi-liquid temperature variation [Eq. (1)] with both A and the upper limit of the temperature range strongly dependent on the field. In these two cases, however, the resistivity just outside the Fermi-liquid regime is of non-Fermi liquid type. By contrast, the material $PrOs_4Sb_{12}$ studied here seems to exhibit a crossover between two Fermi-liquid-like regimes having different effective masses.

Finally, the peak in A_2 (and possibly in A_1) in Fig. 6 may be related to the field-induced LRO. For the (001) field direction, LRO is observed through magnetization measurements[24] to exist between 4.5 and 15 T. However, there are only weak signatures of LRO below 6 T, particularly in bulk measurements. For instance, 6 T is the smallest field for which the specific heat shows a pronounced lambda-like anomaly[25] near 1 K. Similarly, on the high-field side, no peak due to LRO is observed above 13 T.[26] Note that points shown in Fig. 6 are the results of fits performed either entirely within the same phase (paramagnetic or LRO). This is because the LRO boundary[12,26,27] below 1 K satisfies $dH/dT \simeq 0$. Thus, 6, 8, 10, and 12 T points correspond to the LRO phase, while the remaining points belong to the paramagnetic phases.

III. CONCLUSIONS

Although the high-temperature (above 2 K), zero-field resistivity can be adequately described by electron scattering on excited CEF levels, the low-temperature (below 1 K) resistivity in magnetic fields seems to be dominated by fluctuations of the LRO parameter. This study supports our previous analysis[15] of the field variation of the residual resistivity, $\rho_0{=}\rho(T = 0)$. We recall that changes of ρ_0 cannot be explained by CEF effects, instead they reflect strength and fluctuations of the LRO parameter. Longitudinal magnetoresistance as $T \to 0$ is strongly enhanced in the ordered state, but it still decreases with H above 15 T, *i.e.*, outside the LRO phase. LRO fluctuations persist beyond the LRO phase and are responsible for both $\rho_0(H)$ and $\rho(T)$ variations below 1 K.

The inconsistency, mentioned in the Introduction, between determinations of m^* by the dHvA technique and by the specific heat discontinuity at T_c can be explained by a reduction of m^* at temperatures below 0.4–0.7 K. Our analysis of the resistivity in fixed magnetic fields suggests that m^* is also an increasing function of the field (at least up to 6 T).

Our results suggest that heavy-fermion behavior in $PrOs_4Sb_{12}$ may be of a novel character, in the sense that this state exists only in a narrow range of temperatures around 1 K. A number of unexplained anomalies found between 0.4 and 0.7 K may be related to the crossover between heavy-fermion behavior and non-heavy fermion behavior as $T \to 0$.

Acknowledgments

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