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Notes:
Field-dependent ordered phases and Kondo phenomena in the filled skutterudite compound PrOs$_4$As$_{12}$

M. B. Maple$^{1,5,}$, N. P. Butch$^1$, N. A. Frederick$^4$, P.-C. Ho$^4$, J. R. Jeffries$^{1,4}$, T. A. Sayles$^{1,4}$, T. Yanagisawa$^4$, W. M. Yuhasz$^{4,}$, Songxue Chi$^1$, H. J. Kang$^{1,}$, J. W. Lynn$^{1,}$, Pengcheng Dai$^{1,4}$, S. K. McCall$^5$, M. M. McElfresh$^5$, M. J. Fluss$^5$, Z. Henkie$^{1,}$, and A. Pietraszko$^{1,}$

$^1$Department of Physics, $^2$Institute for Pure and Applied Physical Sciences, and $^3$Materials Science and Engineering Program, University of California at San Diego, La Jolla, CA 92093; $^4$Department of Physics and Astronomy, University of Tennessee, Knoxville, TN 37996; $^5$Center for Neutron Research, National Institute of Standards and Technology, Gaithersburg, MD 20899-8562; $^6$Condensed Matter Sciences Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831; $^7$Lawrence Livermore National Laboratory, P.O. Box 808, Livermore, CA 94550; and $^8$Institute of Low Temperature and Structure Research, Polish Academy of Sciences, 50-950, Wroclaw, Poland

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Contributed by M. B. Maple, February 23, 2006

Electrical resistivity, specific heat, and magnetization measurements to temperatures as low as 80 mK and magnetic fields up to 16 T were made on the filled skutterudite compound PrOs$_4$As$_{12}$. The measurements reveal the presence of two ordered phases at temperatures below approximately 2.3 K and in fields below approximately 3 T. Neutron-scattering experiments in zero field establish an antiferromagnetic ground state $<2.28$ K. In the antiferromagnetically ordered state, the electronic-specific heat coefficient $\gamma \approx 1$ J/mol$\cdot$K$^2$ below 1.6 K and $0 \leq H \leq 1.25$ T. The temperature and magnetic-field dependence of the electronic resistivity and specific heat in the paramagnetic state are consistent with a single-ion Kondo behavior with a low Kondo temperature on the order of 1 K. The electronic-specific heat in the paramagnetic state can be described by the resonance-level model with a large zero-temperature electronic-specific heat coefficient that decreases with increasing magnetic field from approximately 1 J/mol$\cdot$K$^2$ at 3 T to approximately 0.2 J/mol$\cdot$K$^2$ at 16 T.

Heavy fermion | Kondo effect | Antiferromagnetism

The compound PrOs$_4$As$_{12}$ belongs to the family of filled skutterudites, which have the formula $AX_12$, where $A$ = alkali metal, alkaline earth, lanthanide, or actinide, $T$ = Fe, Os, or Ir, and $X$ = P, As, or Sb. Because the ions reside in an atomic cage with a large number of $X$ nearest neighbors, the hybridization between the lanthanide filler ion localized 4f states and the conduction–electron states is appreciable, often resulting in strongly correlated electron phenomena. In a recent article (1), we reported a study of the physical properties of single crystals of the filled skutterudite compound PrOs$_4$As$_{12}$ between approximately 0.4 and 300 K by means of magnetization $M$, specific heat $C$, and electrical resistivity $\rho$ measurements. The previous study revealed that the compound exhibits two (possibly three) ordered phases below 2.5 K in zero field and approximately 3 T at zero temperature from kinks in the magnetization, specific heat, and electrical resistivity in magnetic fields as high as 16 T and neutron diffraction measurements in zero field on PrOs$_4$As$_{12}$.

Results

A magnetic neutron-scattering pattern was obtained by subtracting the intensity of the 4-K diffraction pattern from the 0.63-K pattern. Fig. 1a shows the outcome of this subtraction, with clear magnetic intensity at positions consistent with $(h+0.5, k, l)$, where $h$, $k$, and $l$ are the Miller indices of the cubic unit cell. The observed diffraction pattern suggests a magnetic unit cell doubled along one axis of the elementary structural cubic cell. Although the cubic crystal symmetry and the apparent anisotropy of the mixed single-crystal samples prevent a conclusive determination of the spin configuration by using the diffraction intensities, it is clear that the AFM lattice consists of $(1,0,0)$ planes within which Pr ions couple ferromagnetically, while alternate planes have opposite magnetic moment orientations. The order parameter of the sample in the AFM phase was determined by measuring the $T$ dependence of the magnetic intensity at $20 = 17.9^\circ$ in the range 0.5–3.4 K. Fig. 1b indicates a Néel temperature $T_N = 2.28$ K, in excellent agreement with magnetization measurements (1).

Specific heat divided by temperature $C/T$ vs. $T$ data for temperatures below 4 K and in magnetic fields up to 3 T are displayed in Fig. 2. Previous zero-field measurements revealed two low-$T$ features in $C(T)$ (Fig. 2 Inset): a slight shoulder at $T_2 \approx 2.3$ K, on the high temperature side of a well defined peak at $T_1 \approx 2.2$ K (1). $T_1$ can be identified with the AFM transition observed in neutron scattering. As $H$ is applied, the two anomalies observed in zero field gradually shift toward 0 K. The second anomaly decreases with field more quickly than the first and is not observed above 0.6 K by $H = 1.5$ T, whereas the first transition persists to nearly $H = 3$ T. Additionally, the magnitude of the second transition is lowered with increasing $H$, whereas the first evolves from a rounded shoulder at the lowest fields to a clear peak by $H = 0.75$ T, although the true character of the transition is altered.

Conflict of interest statement: No conflicts declared.

Abbreviations: AFM, antiferromagnetic; PM, paramagnetic; RLM, resonance level model; UCSD, University of California at San Diego; LLNL, Lawrence Livermore National Laboratory.

*To whom correspondence should be addressed. E-mail: mbmaple@physics.ucsd.edu.

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first anomaly at low $H$ may be masked by the presence of the second. At the lowest temperatures, a slight upturn, associated with a nuclear Schottky anomaly, is visible. In fields strong enough to suppress the second transition, $H \gtrsim 2$ T, a dome-like structure develops in $C(T)$ above the first transition (see Fig. 7), and the maximum of the dome decreases in magnitude and shifts to higher $T$ with increasing $H$. These features are described in more detail in Discussion.

The features in $C(T)$ at low fields are also identified with features in $M(H, T)$ that have a similar field dependence, as described (1). Low-$T$ $M(T)$ data for various values of $H$ parallel to [111] are shown in Fig. 3. It has been reported that with increasing $H$, a peak in $M(T)$, denoted $T_1$, is suppressed toward $T = 0$ K, whereas a second peak in $M(T)$ at higher $T$, denoted $T_2$, splits off from $T_1$ and eventually shifts toward $T = 0$ K (1). Corresponding features were identified in $M(H)$ at constant $T$, as shown in Fig. 4 (1). Two kinks in $M(H)$, denoted $H_1$ and $H_2$, were found to be suppressed toward lower field with increasing $T$.

The dc magnetic susceptibility $\chi_{dc}(T)$, reported in ref. 1 and shown in Fig. 4 Inset, displays a clear peak at $T = 2.3$ K, indicative of AFM ordering, consistent with the neutron-scattering measurement reported herein.

As reported in ref. 1, $\rho(T)$ was measured for $1.9$ K $\leq T \leq 300$ K in $H$ up to $16$ T (Fig. 5). It was shown that the zero-field $\rho(T)$ decreased with decreasing temperature, displayed a minimum at approximately $16$ K, and then increased to a peak by approximately $5.5$ K followed by a sharp decrease in the vicinity of the AFM transition (Fig. 5) (1). Measurements in an applied field (displayed in Fig. 6 Inset a and Inset b) reported herein reveal a negative magnetoresistance with increasing field below approximately $25$ K. However, below $1.75$ K, $\rho$ increases with $H$, reaching a maximum at $1.5$ T, then decreases with increasing $H$. Eventually the $\rho(H)$ curves at various constant $T$ merge in the highest fields (Fig. 6). At $H = 1$ T, two features in the $d\rho/dT$ vs. $T$ curves are associated with the two phase transitions at $T_2$ and $T_1$ (Fig. 6a Inset). Isotherms of $\rho(H)$, shown in Fig. 6b, display a peak that develops at $T < 2.3$ K, indicative of a field-induced transition at $H_1$. The peak shifts to higher fields with decreasing temperature, up to $1.5$ T below $0.5$ K. A change in slope, apparently caused by a field-induced transition to the paramagnetic (PM) state at $H_2$ (Fig. 6b Inset), was observed for $H > H_1$. 

![Fig. 1. Neutron diffraction measurements on PrOs$_4$As$_{12}$ at various temperatures between 0.63 and 4 K. (a) Intensity spectrum at 0.63 K after subtraction of the 4-K spectrum. The positive and negative intensities around $2\theta = 32^\circ$ represent imperfect subtraction around the nuclear Bragg peak (1,1,1) reflection. The positions of magnetic scattering are clearly marked, and the solid curve is a guide to the eye. (b) The $T$ dependence of the (0.5,1,0) magnetic Bragg peak. The solid curve is a fit of mean field theory, yielding an estimated Néel temperature of $2.28$ K.](image)

![Fig. 2. Specific heat divided by temperature $C/T$ vs. $T$ in various magnetic fields $H$, showing the decrease of $T_2$ and $T_1$ with increasing $H$. (Inset) $C/T$ vs. $T$ in the zero field in the vicinity of the transition.](image)

![Fig. 3. Magnetization $M$ vs. $T$ for several values of $H$. Features in $M(T)$ at constant $H$, with $T_2$ defined by a small peak, and $T_1$ defined by the prominent peak, are illustrated for $H = 1.5$ T.](image)
Fig. 4. Magnetization $M$ vs. $H$ isotherms for several values of $T$. Features in $M(H)$ at constant $T$, with $H_1$ and $H_2$ defined by a kink in the slope, are illustrated for PrOs$_4$As$_{12}$. (Inset) DC magnetic susceptibility $\chi$ vs. $T$ for PrOs$_4$As$_{12}$.

Discussion

Contributions to Specific Heat. In ref. 1, the specific heat was analyzed in zero field. The low-$T$ zero-field-specific-heat was assumed to consist of four terms, $C(T) = C_m(T) + C_e(T) + C_g(T) + C_{pm}(T)$, which are the nuclear Schottky, electronic, lattice, and magnetic contributions, respectively. At higher temperatures, where $C_m(T)$ and $C_e(T) \approx 0$, the specific heat was dominated by the electronic and lattice terms. A fit of $C/T = \gamma + \beta T^2$ to the data in the range $10 \, K \leq T \leq 18 \, K$ yielded an enhanced electronic specific heat coefficient $\gamma = 211 \, mJ/molK^2$ and, from $\beta$, a Debye temperature $\theta_D = 260 \, K$ (1). The calculated magnetic entropy $S_m(T)$ leveled off to a value of $90% Rln3$ by 8 K (1).

Analysis of the zero-field specific heat $\Delta C$, after subtraction of the lattice contribution, was based on an expression of the following form, $\Delta C(T) = C_m(T) + C_e(T) + C_{pm}(T)$ (1), where $C_m(T) = A/T^2$, $C_e(T) = \gamma T$, and $C_{pm}(T) = B T^\nu$. In zero field, it was found that $A = 128 \, mJ/molK$, $n = 3.2 \pm 0.1$, and $\gamma = 1 \, J/molK^2$ (1). As stated in ref. 1, the discrepancy between the value of $\gamma$ estimated from these fits to $\Delta C(T)$ in the ordered phase and the $\gamma$ value taken from the earlier fit to $C(T)$ in the PM state is likely caused by a $T$ dependence of the electronic contribution to the specific heat, as has been observed in a number of heavy fermion systems (11). Fits of this form have now been performed in the ordered state ($T < 0.6 \, T_1$) in field. Analysis of $\Delta C(T)$ in an applied field reveals that $A$ increases monotonically to $A = 215 \, mJ/molK$ by 1.25 T, but does not change substantially with field, and $\gamma$ remains close to 1 $J/molK^2$.

Single-Ion Kondo Behavior. For $H \approx 3 \, T$, a broad maximum was observed in $C_e(T)$, reminiscent of that observed in $C_e(T)$ for PrFe$_4$P$_{12}$ (6). Therefore, the $C_e(T)$ data for PrOs$_4$As$_{12}$ were analyzed in terms of the resonance-level model (RLM) of Schotte and Schotte (12) used in ref. 6 for PrFe$_4$P$_{12}$. This model has three parameters: the width of a Lorentzian peak in the density of states $\Delta = k_B T_K$ (where $T_K$ is the Kondo temperature), the spin $S$, and a Zeeman term $gH$ associated with the applied field, although it was also necessary to scale the curves by a factor $f$. The parameters $S$, $\Delta$, and $f$ were held fixed for all fields, while the term $gH$ was varied. Three spin values, $S = 1/2$, 1, and 3/2, give reasonable fits; however, $S = 1$ results in the best fits and was consistent with the entropy released, as well as the proposed crystalline electric field split Pr$^{3+}$ $T_2$ triplet ground state (1). The best results are found for $T_K \approx 3.5 \, K$ and $f \approx 1.3$ for the University of California at San Diego (UCSD) data and $T_K \approx 5 \, K$ and $f \approx 1.6$ for the Lawrence Livermore National Laboratory (LLNL) data. Fig. 7 shows the high field data and the corresponding fits to the RLM with $S = 1$. In zero field, the Zeeman parameter $gH = 0$ and it monotonically increases with $H$, as expected, because the fits were applied only to data in the PM state. However, $gH$ is a linear function of $H$ only below 5 T, with $g = 3.4$, whereas in $H > 5 \, T$, $g$ decreases to a value of $\approx 1.7$. This nonlinearity may be caused by the inaccuracy of the single-ion description of what is actually a cooperative phenomenon in PrOs$_4$As$_{12}$. That the RLM roughly describes the $C(T)$ data likely reflects generic behavior associated with a sharply peaked electronic density of states at the Fermi level, whereas a Lorentzian distribution is a coarse approximation of the actual density of

Fig. 5. Electrical resistivity $\rho$ vs. $T$ from 2 to 300 K at various fields $H$. (Inset a) $\rho(T)$ from 2 to 25 K. (Inset b) $\rho(H)$ at various temperatures around $T_2$.

Fig. 6. Electrical resistivity $\rho$ measurements as a function of temperature $T$ and magnetic field $H$ for PrOs$_4$P$_{12}$. (a) Low-temperature electrical resistivity $\rho$ vs. $T$ for $0 \, T < H < 3 \, T$. (Inset) $T_1$ and $T_2$, as determined from features in $\rho$ vs. $T$ at 1 T. (b) Isotherms of $\rho$ vs. $H$. (Inset) An expanded view of the 0.1-K $\rho(H)$ isotherm with two linear fits indicating a break in slope at 2.56 T.
states in PrOs$_4$As$_{12}$ that renders the RLM inaccurate even at moderate $H$. For this reason, the values of $\gamma$ estimated from the RLM fits cannot be regarded as accurate.

Despite the aforementioned caveats, the RLM was used to estimate the value that $\gamma(0)$ would have if PrOs$_4$As$_{12}$ were to remain in the PM phase at $T = 0$ K in $H < 3$ T rather than undergoing transitions into ordered phases. In this analysis, the parameters $S$, $\Delta$, and $f$ were assumed to have the same values as they have in $H > 3$ T. The RLM fits describe the $C_e(T)$ data in the PM phase, but obviously deviate from the $C_e(T)$ data in the ordered phases. The resulting $C_{RLM}/T$ values at $T \approx 0$ for all fields are plotted in Fig. 8 along with $\gamma$ values estimated from the fits to $\Delta C(T)$ data in the ordered state. In Fig. 8 the open circles represent values of $\gamma(0)$ derived from fits of the RLM to the $C_e(T)$ data only in the PM phase, $T > T_2$ in $H < 3$ T. As calculated by the RLM fits, $\gamma(0)$ peaks in zero field and decreases sharply with increasing $H$, leveling off once the PM state is entered. In contrast, the fits to $\Delta C(T)$ in the ordered state yield values of $\gamma \approx 1$ J/mol$^2$K$^2$ (1), similar in magnitude to $\gamma(0)$ values from the RLM fits to the 2-T and 3-T $C_e(T)$ data. These results suggest that upon entering the AFM phase at low $T$ the triply degenerate Kondo-like resonance splits because of the development of an effective internal field, which reduces the electronic density of states at the Fermi level, thus decreasing the electronic contribution to $C(T)$. Because the RLM predicts $\gamma(T = H = 0) = 5$ J/mol$^2$K$^2$, it may be argued that the AFM ground state “shields” the system from the massive itinerant electronic degeneracy that would exist at low $T$ in the absence of resonance splitting. It should be noted that attempts were made to fit the broad feature in $C_e(T)$ with Schottky anomalies caused by crystalline electric field split Pr$^{3+}$ states, but those fits were unable to reproduce the feature.

At the lowest temperatures, $\rho(T)$ in PrOs$_4$As$_{12}$ is not dominated by electron–electron scattering ($\rho$ goes as $T^2$) or simple AFM magnon scattering ($\rho$ goes as $T^3$), but is described well by an expression for an antiferromagnet with an energy gap $\Delta$ in the magnon spectrum (13):

$$\rho(T) = \rho_0 + b \left( \frac{T}{\Delta} \right) \left( 1 + \frac{T}{2\Delta} \right)^{\Delta T^2}.$$  

The $\rho(T)$ data are fit well by Eq. 1 for $H < 2$ T (Fig. 9a), and a plot of the energy gap $\Delta$ vs. $H$ is shown in Fig. 10a. Notably, the value of $\Delta$ drops sharply near 1.5 T in the vicinity of the transition at $H_1$. To check for possible non-Fermi-liquid behavior associated with a field-tuned phase transition ($H \approx 3$ T), the $\rho(T)$ data in $H \approx 8$ T were also fit with a power-law $\rho(T) = \rho_0 + b T^n$ (Fig. 9b). The residual resistivity $\rho_0$ has a pronounced maximum at $H = 1.5$ T, corresponding to $H_1$ (Fig. 10c). The power-law exponent $n$ decreases from approximately 2.9 to approximately 2.3 as $H$ increases from 0 to 2 T, reaches a minimum between 2
and 3 T, then gradually increases to ~2.8 when H reaches 8 T (Fig. 10b). In no range of H does PrOs$_4$As$_{12}$ display the $T^{\frac{2}{3}}$ non-Fermi-liquid behavior predicted to occur near a 3D AFM quantum critical point (14).

For $H < 4$ T in the PM state, the $\rho(T)$ data exhibit a local minimum at approximately 16 K (Fig. 5), while the $\rho(H)$ curves decrease with increasing $H$ and $\rho(H = 0) - \rho(H)$ decreases rapidly as $T$ increases; this behavior is consistent with the Kondo effect (15–17). To determine whether PrOs$_4$As$_{12}$, which shows Kondo lattice behavior, can be described in the PM regime by the single-ion Kondo impurity model, a scaling analysis developed by Schlottmann (18) was applied to the $\rho(H, T)$ data. A lattice contribution $\rho_l$ (estimated from LaOs$_4$Sb$_{12}$ (24)) was subtracted before the data were scaled, although it was subsequently found that it makes little difference whether this correction is made. Fig. 11 displays the dependence of the normalized magnetoresistivity $\rho_{\text{norm}}(T) = [\rho(H, T) - \rho_l(T)]/[\rho(H = 0, T) - \rho_l(T)]$ on the scaled magnetic field $H^*/H^*(T)$ in the PM state, where $H^*(T)$ is the characteristic $T$-dependent Kondo field. The scaling of the magnetoresistivity of PrOs$_4$As$_{12}$ agrees well with Schlottmann’s $J = 1$ scaling curve (18), in accordance with a $T$'s triplet ground state inferred from magnetization and specific heat data. The values of $H^*$ deduced from the scaling analysis are plotted vs. $T$ in Fig. 11 Inset; $H^*$ is approximately linear in the temperature regime from 6 to 22 K, whereas below 6 K and above 22 K, $H^*(T)$ has shallower slopes.

In the single impurity model, $H^*(T)$ is given by the relation

$$H^*(T) = H^*(0) + \frac{k_B T}{g_i \mu_{\text{ion}}} = \frac{k_B}{g_i \mu_{\text{ion}}} (T_K + T),$$

where $H^*(0)$ is the zero-$T$ characteristic Kondo field, $T_K$ is the Kondo temperature, $g_i$ is the Landé g factor ($g_i = 0.8$ for the Pr$^{3+}$ ion), and $\mu_{\text{ion}}$ is the ionic local magnetic moment (16, 17).

A linear fit between 6 and 22 K resulted in a $\mu_{\text{ion}} = 0.91 \mu_B$ and $H^*(0) = -3.86$ T, corresponding to $T_K = -1.88$ K, although typically one would expect a positive value for $H^*(0)$ and $T_K$.

Possible explanations for this discrepancy are: (i) The single-ion model is too simplified for this Kondo-lattice system. Similar results were obtained previously in the case of UBe$_{13}$ (15). (ii) The Kondo temperature is not constant <40 K, which is evident in the curvature of $H^*(T)$. (iii) If the lowest three values of $H^*$ at 2.5, 3, and 3.5 K are used to estimate $T_K$, a positive $T_K \approx 0.7$ K is obtained [$\mu_{\text{ion}} = 1.68 \mu_B$ and $H^*(0) = 0.77$ T]. (iv) Magnetic correlations persist into the PM state. In any case, this scaling analysis indicates that the Kondo temperature is small, and that the Kondo effect in PrOs$_4$As$_{12}$ is sensitive to magnetic correlations.

**Phase Diagram.** The features in $C(H,T)$ (Fig. 2), $M(H, T)$ (Figs. 3 and 4), and $\rho(H, T)$ (Fig. 6) associated with $T_2$, $T_1$, $H_1$, and $H_2$ have been used to generate an $H$-$T$ phase diagram for PrOs$_4$As$_{12}$ (Fig. 12). The bars in Fig. 12 are rough estimates of the transition widths, based on the widths of those features. Comparing the magnetic, resistive, and thermodynamic phase diagrams, it is likely that the higher-field $M$ features, which correspond to peaks in $M(T)$ and $T_1$ and kinks in $M(H)$ ($H_1$ and $H_2$) are associated with the peaks in $C(T)$ ($T_2$ and $T_1$) and features in $\rho(H)$ ($H_1$ and $H_2$). Although the values of $H$ at which the phase boundaries are defined for different measurements differ by as much as 0.5 T, the field dependence of the features is very similar. These discrepancies are caused partly by measurement-dependent differences in the definitions of the various transitions and variations between samples, in addition to the fact that $M(H,T)$ and $C(H,T)$ measurements were performed on a collection of crystals.

Nonetheless, it may be inferred from the strong peaks in $C(T)$ that there are two bulk-phase transitions: PrOs$_4$As$_{12}$ has an AFM ground state, a different ordered state OP2 at intermediate $T$ and $H$, and a PM state at high $T$ and $H$. Previously, features in $M(H,T)$ and crystalline electric field considerations were argued to support an AFM ground state (1). Neutron scattering has now confirmed AFM order in this phase. Analysis of $\rho(H,T)$ data shows evidence for an energy gap that closes at $H_1$, whereas $\rho_0$ goes through a maximum at $H_1$ (Fig. 10), which is consistent with enhanced scattering off...
of critical fluctuations at a magnetic phase transition. A triplet ground state is supported by compelling evidence from $M(H, T)$ data and the Kondo-resonance-based analysis of $C(H, T)$ and $\rho(H, T)$. The recent measurements in magnetic field were unable to determine the ordering that occurs in OP2; however, as stated in ref. 1, quadrupolar or magnetic ordering remain likely candidates for OP2.

**Comparison to PrFe$_4$P$_{12}$.** Numerous compounds exhibit features similar to those observed in PrOs$_4$As$_{12}$. The most noteworthy example is PrFe$_4$P$_{12}$, which, like PrOs$_4$As$_{12}$, is a Pr-based filled skutterudite compound that displays heavy fermion behavior and AFM-like features (19, 20). The electrical resistivities of these two compounds display a Kondo-like minimum and a sharp downturn below their respective ordering temperatures. A negative magnetoresistance is also observed in these compounds in the vicinity of their respective ordering temperatures (21). At 5 K, the magnitude of the magnetization of both PrFe$_4$P$_{12}$ and PrOs$_4$As$_{12}$ below their transition temperatures is much less than the Pr$^{3+}$ free-ion value of $3.2 \mu_B$/Pr ion; however, there is a clear step-like feature in the $M(H)$ data for PrFe$_4$P$_{12}$ indicating a metamagnetic transition (19), whereas features in the $M(H)$ data for PrOs$_4$As$_{12}$ are much more subtle. In high fields, the $C(T)/T$ data for both systems display a broad dome-shaped anomaly at low temperatures, but in zero field there is no indication of a second-phase transition in the $C(T)/T$ data for PrFe$_4$P$_{12}$ (6). Because neutron diffraction measurements of PrFe$_4$P$_{12}$ in zero field did not find any indication of long-range magnetic order and measurements in an applied field have revealed the presence of an induced AFM moment, antiferroquadrupolar ordering is speculated as being the source of the observed behavior of PrFe$_4$P$_{12}$ below 6.5 K (7, 22). The transition in PrFe$_4$P$_{12}$ at 6.5 K is also accompanied by a structural change that occurs at the same temperature (23).

**Summary**

Neutron diffraction measurements of PrOs$_4$As$_{12}$ in zero field confirm that the ground state is AFM, with $T_N = 2.28$ K. Features in the $C(H, T)$ and $\rho(H, T)$ data can be identified with transitions between two ordered phases and yield an $H-T$ phase diagram similar to that inferred from the $M(H, T)$ data previously reported (1). The $T$ dependence of $\rho$ in the AFM ground state is consistent with the scattering of electrons by magnons with a gapped spectrum. Features reminiscent of single-ion Kondo behavior are observed in the PM state. The specific heat can be described in terms of the RLM for $S = 1$, which would be appropriate for a Pr$^{3+}$ triplet ground state, yielding a Kondo temperature of approximately 3 K. Moreover, $\rho(H, T)$ scales with $H/H^*$ and is consistent with a small $T_K < 1$ K and a triplet ground state.

**Experimental Details**

Single crystals of PrOs$_4$As$_{12}$ were grown by means of a molten metal flux method at high temperatures and pressures as described (1). Neutron diffraction measurements were performed on the triple-axis spectrometer BT2 at the National Institute of Standards and Technology Center for Neutron Research. The small single-crystal samples (approximately 1 mm in size) with a total mass of approximately 1.2 g were wrapped in thin aluminum foil and sealed inside a thin-wall aluminum can mounted in a $^3$He cryostat.

The incident and final neutron energies were set to 14.7 meV with pyrolytic graphite as monochromator, analyzer, and filters. The collimations were 60 minutes of arc-40 minutes of arc-sample -40 minutes of arc-open from the reactor to the detector. Scans were carried out at 0.63 and 4 K with the angle ranging from 3° to 65° with a step size of 0.1°.

Specific heat $C(T)$ measurements on PrOs$_4$As$_{12}$ were performed for 0.5 K $\leq T \leq 18$ K in $H$ up to 5 T at UCSD in a semidiabatic $^3$He calorimeter. Initial measurements in zero field were performed on a collection of single crystals with a total mass of 49 mg, while additional measurements in zero and higher fields were performed on a collection of single crystals with a mass of 29 mg. The calculated molar specific heats of the two zero-field measurements were found to be identical within experimental error. Additional $C(T)$ measurements for 0.4 K $\leq T \leq 20$ K in $H$ up to 16 T were performed at LLNL in a Quantum Design (San Diego) $^3$He Physical Property Measurement System (PPMS). Bulk magnetization $M$ for temperatures 0.4 K $\leq T \leq 10$ K and applied magnetic fields $-5.5 \leq H \leq 5.5$ T was measured in a $^3$He Faraday magnetometer. A mosaic of 27 crystals, with a total mass of 27 mg, was measured with the [111] axis of the crystals nominally aligned parallel to $H$. Measurements of $M(T, H)$ were also performed by using a Quantum Design Magnetic Property Measurement System in $H$ up to 5.5 T for 1.7 K $\leq T \leq 300$ K. Electrical resistivity $\rho(H, T)$ measurements were made by using a four-probe ac technique in fields up to 9 T for 2 K $\leq T \leq 300$ K in a Quantum Design PPMS by using a constant current of 1–10 mA and in $H$ up to 8 T for 0.06 K $\leq T \leq 2.6$ K by using a $^3$He–$^4$He dilution refrigerator and a constant current of 300 $\mu$A, with $H$ perpendicular to the electrical current and parallel to the [100] axis. Additional $\rho(H, T)$ measurements for 0.5 K $\leq T \leq 300$ K in $H$ up to 16 T were performed at LLNL in a Quantum Design $^3$He PPMS by using a constant current of 0.25–1 mA.

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