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N. Oeschler, Philipp Gegenwart, F. Weickert, I. Zerec, P. Thalmeier, F. Steglich, E. D. Bauer, N. A. Frederick, M. B. Maple

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B-T phase diagram of $PrOs_4Sb_{12}$ studied by low-temperature thermal expansion and magnetostriction

N. Oeschler, P. Gegenwart, F. Weickert, I. Zerec, P. Thalmeier, and F. Steglich
Max-Planck Institute for Chemical Physics of Solids, Noethnitzer Strasse 40, D-01187 Dresden, Germany

E. D. Bauer,* N. A. Frederick, and M. B. Maple
Department of Physics and Institute for Pure and Applied Physical Sciences, University of California, San Diego, La Jolla, California 92093, USA

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We report low-temperature thermal expansion $\alpha(T)$ and magnetostriction $\lambda(B)$ measurements on the filled skutterudite $PrOs_4Sb_{12}$ in magnetic fields up to 18 T. This compound appears to be the first example of a Pr-based heavy-fermion (HF) superconductor. Superconductivity manifests in a double jump at $T_{c1}=1.82$ K and $T_{c2}=1.72$ K, as also observed in specific heat measurements. The pressure dependencies of the two transitions are calculated by means of the Ehrenfest relation. For $B \leqslant 3$ T, the normal-state thermal expansion shows a pronounced minimum related to crystalline electric field (CEF) splitting of the Pr^{3+} ions. At low temperatures, the thermal expansion is dominated by a nuclear spin contribution of the Pr ions. Furthermore, anomalies in $\alpha(T)$ and $\lambda(B)$ for $4.4\ \mathrm{T} \leqslant B \leqslant 15$ T and $T \leqslant 1.3$ K have made it possible to determine the boundary of an ordered phase in the *B-T* plane that is presumably associated with quadrupolar order.

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I. INTRODUCTION

The study of superconductivity in systems with large effective masses which have a nonmagnetic ground state is of fundamental interest. The recently discovered filled skutterudite compound $PrOs_4Sb_{12}$ appears to be the first example of a Pr-based heavy-fermion (HF) superconductor.[1,2] Evidence for a HF state in this compound is provided by the large Sommerfeld coefficient γ_0 of 300–600 mJ/mol K^2, corresponding to a mass enhancement of the itinerant *f* electrons by ~ 50 with respect to the free electron mass.[1–5] The large value of γ_0 was inferred from the electronic contribution to the normal-state specific heat,[1–5] the jump in the specific heat at T_c,[1–4] and the slope of the upper critical field near T_c.[1–3] Magnetic susceptibility $\chi(T)$ measurements on $PrOs_4Sb_{12}$ revealed a nonmagnetic ground state.[1–3] The $\chi(T)$ data for $PrOs_4Sb_{12}$ were analyzed within the context of the theory of Lea, Leask, and Wolf[6] for a cubic crystalline electric field (CEF), in which the Pr^{3+} $J=4$ Hund's rule multiplet is split into a nonmagnetic Γ_1 singlet, a nonmagnetic Γ_3 doublet which carries an electric quadrupole moment, and Γ_4 and Γ_5 triplets, and could be described by two Pr^{3+} energy level schemes, one with a Γ_3 ground state and the other with a Γ_1 ground state. Both of these Pr^{3+} energy level schemes have a low lying Γ_5 triplet excited state at an energy ~ 10 K above the ground state, with the two remaining states at much higher energies ($\gtrsim 10^2$ K). The Pr^{3+} energy level scheme with the Γ_3 ground state provided a better description of the $\chi(T)$ data at low temperatures,[2] and was found to be consistent with inelastic neutron scattering measurements,[3] the pronounced Schottky anomaly in the normal-state specific heat,[4,5] and, recently, features in the *T* and *B* dependence of the electrical resistivity.[7] On the other hand, based on their specific heat measurements on $PrOs_4Sb_{12}$, Aoki *et al.* concluded that the Pr^{3+} ions have a Γ_1 singlet ground state and a Γ_5 triplet first excited state.[8] Recently neutron scattering measurements on $PrOs_4Sb_{12}$ were performed in the high-field ordered phase.[9] From the analysis of these experiments, it was concluded that the Pr^{3+} ions exhibit antiferroquadrupolar order in high fields and have a Γ_1 singlet ground state.

Assuming a doublet ground state, the heavy masses may be caused by aspherical Coulomb scattering of conduction electrons by the electric quadrupole moments of the Pr^{3+} ions, in contrast to exchange scattering of conduction electrons by the magnetic moments of the Pr^{3+} *f* electrons. The so-called quadrupolar Kondo effect is expected to lead to high effective masses and, in addition, to deviations from Fermi-liquid behavior.[10] On the other hand, in the Γ_1 singlet ground-state scenario, the mass renormalization could arise from virtual $\Gamma_1-\Gamma_5$ excitations as in Pr metal.[11]

In $PrOs_4Sb_{12}$, the nature of the true normal ground state for $T \to 0$ cannot be investigated since superconductivity evolves out of the HF state at $T_c \approx 1.85$ K with a zero temperature critical field $B_{c2}(0)$ of ~ 2.4 T. The height of the specific heat jump at the superconducting (SC) transition, $\Delta C/T_c$, is in the order of γ_0, suggesting the formation of Cooper pairs from heavy quasiparticles.[1–3] Careful specific heat measurements[3,4] on clean single crystals reveal two SC phase transitions at $T_{c1}=1.85$ K and $T_{c2}=1.75$ K, indicative of unconventional superconductivity. At higher magnetic fields $4.4\ \mathrm{T} \leqslant B \leqslant 15$ T, an ordered phase[3,4,12–16] appears below 1.3 K, which, according to recent neutron diffraction measurements, is of antiferroquadrupolar origin.[9]

This paper is organized as follows. After giving details concerning experimental techniques in Sec. II, the experimental results of thermal expansion and magnetostriction studies on $PrOs_4Sb_{12}$ are presented (Sec. III). In Sec. IV, the SC properties of $PrOs_4Sb_{12}$ are discussed and the normal-state thermal expansion is analyzed. Furthermore, the nature of the high-field phase is discussed. The conclusions are presented in Sec. V.

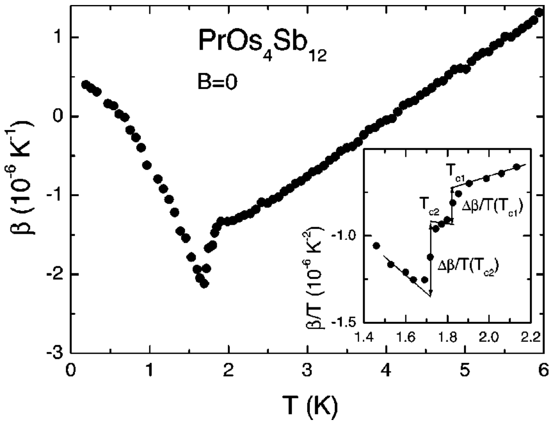

FIG. 1. Volume thermal expansion coefficient β of $PrOs_4Sb_{12}$ for $B=0$. Inset: β/T of $PrOs_4Sb_{12}$ at $B=0$ near T_{c1} and T_{c2}. Arrows in the equal-areas constructions indicate jump heights $\Delta\beta/T$ at the superconducting transitions.

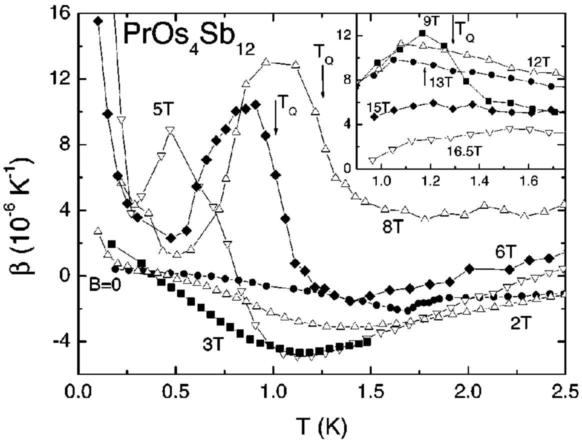

FIG. 2. Volume thermal expansion coefficient β of $PrOs_4Sb_{12}$ in several constant magnetic fields between 0 and 8 T, as well as between 9 and 16.5 T (inset). Arrows indicate the quadrupolar ordering temperature T_Q.

II. EXPERIMENTAL DETAILS

Low-temperature thermal expansion and magnetostriction measurements were made on single crystals of the cubic filled skutterudite compound $PrOs_4Sb_{12}$, grown in a molten Sb-flux.[2] The thermal expansion was measured between 50 mK and 6 K in fields up to 8 T utilizing a high-resolution capacitive dilatometer manufactured of pure silver. For better resoultion, two samples of 0.9 and 0.6 mm length oriented along the [100] axis were mounted on top of each other. Thermal expansion and magnetostriction measurements on the larger sample were performed in a capacitive CuBe dilatometer in fields up to 18 T and at temperatures down to 15 mK.

The thermal expansion coefficient α is defined as $l^{-1}(\partial l/\partial T)$ where l is the total sample length. Similarly, the magnetostriction coefficient is defined as $\lambda = l^{-1}(\partial l/\partial B)$. For a cubic system such as $PrOs_4Sb_{12}$, the volume expansion coefficient is given by $\beta = 3\alpha$.

III. RESULTS

The thermal expansion coefficient β of $PrOs_4Sb_{12}$ for $B=0$ is shown in Fig. 1. Superconductivity manifests itself in a double transition at $T_{c1} = (1.82 \pm 0.01)$ K and $T_{c2} = (1.72 \pm 0.01)$K. The inset of Fig. 1 shows the thermal expansion coefficient around the SC transitions in more detail. The values obtained for T_{c1} and T_{c2} are in very good agreement with those reported from specific heat measurements.[3,4] This fact excludes the possibility that the double transition is caused by slightly different T_c values of the two samples measured simultaneously. At $B=0.5$ T, only one transition is observed. No discontinuity can be resolved at 2 T, although the upper critical field derived from the resistive transition equals 2.4 T.[3] Figure 2 shows the normal state thermal expansion for fields $B \leqslant 8$ T. Three striking features are observed: (i) a negative contribution to $\alpha(T)$, resulting in a minimum for $B \leqslant 5$ T, related to the CEF splitting of the Pr^{3+} state, (ii) a strong increase at lowest temperatures caused by a hyperfine contribution of the Pr nuclear spins, and (iii) at $B \geqslant 4.4$ T, a phase transition anomaly at T_Q which, as discussed in the next section, marks the onset of quadrupolar ordering. The phase transition temperature T_Q is derived from an equal-areas construction. For magnetic fields up to 9 T, T_Q increases with increasing B. At larger fields, T_Q becomes reduced and, correspondingly, the anomaly weakens and disappears for $B \gtrsim 13$ T (see inset Fig. 2).

Our magnetostriction measurements plotted in Fig. 3 reveal phase transition anomalies at the lower B_Q as well as the upper B'_Q boundary of the high-field phase. The pronounced minima in $\lambda(B)$ observed at the lower transition B_Q correspond to the maxima in $\alpha(T)$ near T_Q and shift to higher fields with increasing temperature. At $B'_Q \approx 15$ T, the slope of $\lambda(B)$ changes sign. The B-T phase diagram (Fig. 4), derived from the thermal expansion and magnetostriction measurements, suggests that this B'_Q anomaly represents the continuation of $T_Q(B)$ (see dotted line), terminating to $T \rightarrow 0$ at around 15 T. Minor anomalies in $\lambda(B)$ between 12 and 15 T and at elevated temperatures above 0.4 K should be related

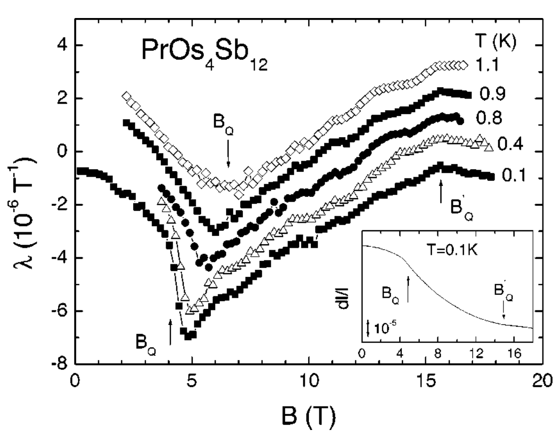

FIG. 3. Linear magnetostriction coefficient λ of $PrOs_4Sb_{12}$ at various temperatures. For clarity the curves for $T > 0.1$ K were shifted subsequently by 1×10^{-6} T^{-1}. Arrows indicate lower (B_Q) and upper (B'_Q) boundary of quadrupolar ordered phase. Inset: length change dl/l of $PrOs_4Sb_{12}$ vs magnetic field at $T=0.1$ K.

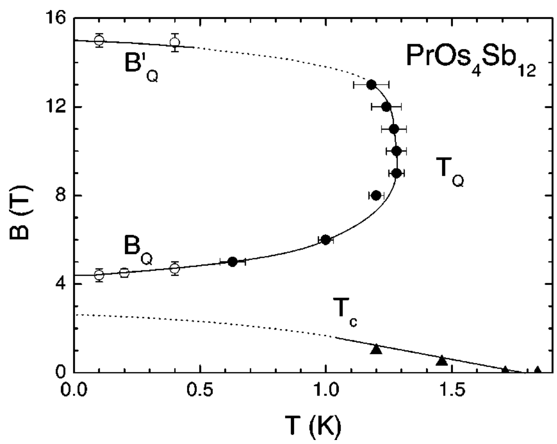

FIG. 4. B-T phase diagram of $PrOs_4Sb_{12}$ as derived from thermal expansion (closed symbols) and magnetostriction (open symbols) measurements. Triangles and circles mark transitions into SC state and high-field quadrupolar-ordered phase, respectively. Lines are guides to the eyes.

to the upper boundary of the high-field phase. The precise determination of the $T_Q(B)$ boundary in this region of the phase diagram is, however, hampered by the disappearance of its signature in thermal expansion above 13 T.

IV. DISCUSSION

Both specific heat and thermal expansion measurements have revealed the existence of two SC phase transitions in $PrOs_4Sb_{12}$.[3,4,8,13] However, the origin of the two SC phases has not yet been clarified. Further analysis of the discontinuities in the thermal expansion coefficient has provided information concerning the SC transitions. By using the Ehrenfest relation, the pressure dependence of both SC phase transitions can be calculated from the jump heights in thermal expansion and specific heat which can then be compared to the resistively determined pressure dependence of T_c. The jump height in β/T is obtained via an equal-areas construction, analogous to the construction of the jump in specific heat,[3,4] see inset of Fig. 1. Using the values given in Table I and $V_{\text{mol}}=2.42\times10^{-4}\ \text{m}^3/\text{mol}$, we obtain $\partial T_{c1}/\partial p = V_{\text{mol}}T_{c1}\Delta\beta_{c1}/\Delta C_{c1} = -(180\pm60)$ mK/GPa and $\partial T_{c2}/\partial p = -(340\pm80)$ mK/GPa.[17] Assuming only one broadened jump, the pressure dependence of T_c derived from the Ehrenfest relation would equal $-(240\pm60)$ mK/GPa. On the other hand, resistivity measurements under hydrostatic pressure[3] revealed -150 mK/GPa. This value is close to the calculated value of $\partial T_{c1}/\partial p$. We therefore exclude a broadened transition at T_c and conclude that two different phase transitions occur at slightly different temperatures.

TABLE I. Values for the jump height of the thermal expansion $\Delta\beta/T$ and specific heat (Ref. 4) $\Delta C/T$ at the two superconducting phase transtitions, as well as calculated pressure dependencies $\partial T_{c,i}/\partial p$.

	first SC transition	second SC transition
T_c [K]	$T_{c1}=(1.82\pm0.01)$	$T_{c2}=(1.72\pm0.01)$
$\Delta\beta/T[10^{-6}\ \text{K}^{-2}]$	-0.22 ± 0.06	-0.38 ± 0.06
$\Delta C/T$ [mJ/mol K^2]	540 ± 40	460 ± 40
$\partial T_{c,i}/\partial p$ [mK/GPa]	-180 ± 60	-340 ± 80

The temperature dependence of the magnetic penetration depth λ as determined from μSR experiments is characteristic for a superconducting state with an isotropic energy gap.[18] This is supported by NQR studies.[19] More recent μSR experiments[20] find evidence for static moments in the SC phase pointing to a nonunitary triplet state. Angular-dependent measurements of the thermal conductivity have revealed a low-field "B phase" with twofold and a high-field "A phase" with fourfold symmetry.[21] Various preliminary order parameter models for the A and B phase, including extended s-wave, d-wave, or nonunitary triplet state have been proposed.[22–24] However, the boundary between these two phases, observed only in these angular-dependent thermal conductivity measurements so far, is in disagreement with the field dependence of the lower SC transition $T_{c2}(B)$ derived from specific heat[4,8] and magnetization[16] measurements at $B\leqslant 0.5$ T which reveals a suppression of both $T_{c1}(B)$ and $T_{c2}(B)$ with similar rates. For $B>0.5$ T, the lower transition can no longer be resolved by thermodynamic measurements.

We next discuss the normal-state thermal expansion which, similar to specific heat,[2,4,5,8] is strongly influenced by the CEF splitting. Two different CEF energy level schemes for the localized Pr^{3+} ions have been proposed to describe the magnetic susceptibility and specific heat data. In one case, the ground state is a Γ_3 doublet,[2,4,5] whereas in the other, it is a Γ_1 singlet.[2,8] In both scenarios the first excited state is the Γ_5 triplet state and both scenarios are based on the cubic O_h symmetry, although the tetrahedral T_h CEF could be realized in $PrOs_4Sb_{12}$.[25] However, this does not changes the number of sublevels nor their degeneracies, but introduces a nonzero sixth order term of strength y that affects the eigenfunctions and eigenvalues.

The application of magnetic fields leads to a Zeeman splitting of the CEF levels. The magnetic field dependence of the two lowest levels, i.e., Γ_3 and Γ_5 in the first scenario and Γ_1 and Γ_5 in the second, has been calculated in Refs. 4 and 16. However, a quantitative analysis of the Schottky anomaly in specific heat at $B>0$ with the two different CEF models has not been performed yet. Below 4 K, where the phonon contributon is negligible, the thermal expansion, shown in Fig. 5, consists of an electronic, a CEF, and, as discussed below, a nuclear contributon. Since the effective Grüneisen ratio $\Gamma_{\text{eff}}\propto\alpha/C$ is not constant but is instead strongly temperature dependent,[13] a separation of the different contributions similar to the specific heat analysis in zero magnetic field,[2,4,5] is impossible. We relate the minimum in $\alpha(T)$ to the CEF Schottky contribution; however, its position differs from that of the corresponding maximum in specific heat. For example, at $B=3$ T, a specific heat maximum[4,8] occurs at 3 K, whereas the minimum in thermal expansion is located around 1.5 K. It is thus not a simple task to determine an energy splitting from the position of the minimum in $\alpha(T)$. As each CEF sublevel possesses a contribution proportional to its respective Grüneisen parameter and these different

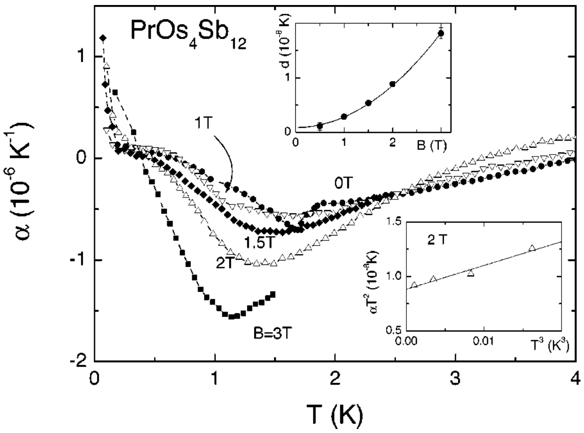

FIG. 5. Linear thermal expansion coefficient α of $PrOs_4Sb_{12}$ in fields $B \leqslant 3$ T. Upper inset: fit parameter d for nuclear spin contribution vs magnetic field. Solid line indicates $d(B)=d_0+d_1B^2$ with $d_0=8.5\times10^{-10}$ K^{-1} and $d_1=1.84\times10^{-9}$ K. Lower inset: thermal expansion data at 2 T for $T \leqslant 0.25$ K as αT^2 vs T^3.

Grüneisen parameters are unknown because the elastic constants have not yet been measured, a fitting of $\alpha(T)$ with too many unknown parameters is not sensible.

As shown in Fig. 5, the thermal expansion coefficient α exhibits a steep increase below 0.5 K that is due to nuclear contribution of the Pr ions. A nuclear hyperfine contribution to thermal expansion has previously been observed in a single crystal of hexagonal Pr metal.[26] There, anisotropic behavior was found with strongly enhanced nuclear Grüneisen parameters of −75 and +64 along and perpendicular to the hexagonal axis, respectively. Very large nuclear specific heat contributions have also been reported for $PrOs_4Sb_{12}$.[4,8] The low-temperature thermal expansion can be described by $\alpha(T)=aT+d(B)/T^2$, resulting in a straight line in a plot of αT^2 vs T^3 (lower inset to Fig. 5). The dominating T^{-2} term in α results from the high-temperature tail of the nuclear Schottky anomaly. The magnetic field dependence $d(B)=d_0+d_1B^2$ (upper inset to Fig. 5) indicates the sum of a small field-independent nuclear quadrupolar and a field-dependent nuclear spin contribution. Corresponding behavior has been observed for the nuclear contribution to the specific heat as well.[8] Using the values of the nuclear contribution to the specific heat[8] and $\kappa_T=4.7\times10^{-12}$ Pa^{-1} taken for $PrFe_4P_{12}$,[27] one derives a nuclear Grüneisen parameter $\Gamma_n = V_{mol}/\kappa_T 3\alpha_n/C_n$ 156 (146) at $B=0(B=2$ T), which is of the same order of magnitude as observed for Pr metal.[26]

Although it is not possible at this time to accurately explain the thermal expansion data with CEF fits, it is possible to make some qualitative observations. Notably, if the ground state of $PrOs_4Sb_{12}$ is a Γ_3 doublet, it is expected to split in magnetic fields and produce a large signal at low temperatures. This feature is not observed in our data, although the presence of the nuclear contribution may also disguise it.

A high-field phase transition is observed in thermal expansion and magnetostriction measurements for fields $4.4\ T \leqslant B \leqslant 15$ T and temperatures $0.1\ K \leqslant T \leqslant 1.2$ K. The phase diagram shown in Fig. 4 agrees well with similar diagrams constructed via measurements of electrical resistivity, magnetization, and specific heat.[4,5,8,14,15] The discontinuities in specific heat and thermal expansion are reminiscent of the shape of the specific heat anomaly in $PrPb_3$ which is known to be associated with quadrupolar order.[28] Also, the boundary of the high-field phase resembles that of the ordered phase in the B-T diagram of $PrPb_3$. In contrast to $PrPb_3$, the ordered phase in $PrOs_4Sb_{12}$ only appears in magnetic fields above $B_Q=4.4$ T. Near this field, the lower branch of the first excited state Γ_5 crosses the ground state in both CEF scenarios for $PrOs_4Sb_{12}$. Thus, the quadrupolar moment carried by the Γ_5 state seems to order in the high-field phase. The appearance of a second phase transition at $B=15$ T identifies the order as antiferroquadrupolar. This is in agreement with neutron diffraction measurements which indicate that antiferroquadrupolar order occurs in the high field ordered phase.[9] In the case of ferroquadrupolar ordering, there would be no upper phase boundary at higher fields. Above the upper phase boundary, the staggered quadrupolar order vanishes and only uniform quadrupole moments persist.

V. CONCLUSION

Thermal expansion measurements on the heavy-fermion superconductor $PrOs_4Sb_{12}$ support the existence of two SC phases with only slightly different transition temperatures but clearly different hydrostatic pressure dependencies. The normal state minimum in $\alpha(T)$ is related to the CEF splitting. However, the position of the minimum disagrees with that of the corresponding specific heat maximum. Therefore, the data cannot be used to discriminate between the two different proposed CEF scenarios. At low temperatures a steep increase occurs which is caused by a nuclear contribution with a very large Grüneisen parameter. The large nuclear contribution might also disguise a Schottky anomaly expected from the Zeeman splitting of the ground-state doublet in the Γ_3 scenario.

The high-field phase was intensively studied by thermal expansion and magnetostriction measurements and a B-T phase diagram was established. The boundary of the high-field ordered phase lies within the region $4.4\ T \leqslant B \leqslant 15$ T and $T \leqslant 1.2$ K and resembles the B-T phase diagram of the quadrupolar-ordered system $PrPb_3$. We thus conclude that the high-field phase is of antiferroquadrupolar origin in accordance with recent neutron diffraction studies.[9]

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*Present address: Los Alamos National Laboratory, Los Alamos, NM 87544, USA.

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