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Entropy Evolution in the Magnetic Phases of Partially Frustrated CePdAl

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In the heavy-fermion metal CePdAl, long-range antiferromagnetic order coexists with geometric frustration of one-third of the Ce moments. At low temperatures, the Kondo effect tends to screen the frustrated moments. We use magnetic fields B to suppress the Kondo screening and study the magnetic phase diagram and the evolution of the entropy with B employing thermodynamic probes. We estimate the frustration by introducing a definition of the frustration parameter based on the enhanced entropy, a fundamental feature of frustrated systems. In the field range where the Kondo screening is suppressed, the liberated moments tend to maximize the magnetic entropy and strongly enhance the frustration. Based on our experiments, this field range may be a promising candidate to search for a quantum spin liquid.

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If competing exchange interactions prevent magnetic systems from developing long-range order, the frustrated magnetic moments can form fluidlike states of matter, so-called spin liquids (SLs) [1,2]. If the moments act as effective spin-1/2 particles, quantum fluctuations dominate and impede the moments from freezing or ordering at low temperatures T [3]. The ground states of these quantum SLs are characterized by massive many-body entanglement rendering them particularly attractive for investigations of new types of quantum matter. Ever since the first notion of SLs was advertised, there has been continual effort to search for materials that might host SLs, mainly in geometrically frustrated magnets [4–6]. Up to now, only very few candidates for metallic SLs have been discovered [6–8].

CePdAl belongs to a class of heavy-fermion (HF) metals with ZrNiAl-type crystal structure (space group $P\bar{6}2m$) that display geometric frustration owing to the fact that the Ce ions form a distorted kagome network in the hexagonal $a-b$ plane [9–11]. In HF compounds, the magnetic moments are formed by nearly localized $4f$ or $5f$ states. Magnetic correlations are enabled by the Ruderman-Kittel-Kasuya-Yoshida (RKKY) interaction which competes with the Kondo effect tending to screen the moments at low T . The presence of a Kondo effect in CePdAl is manifest through a logarithmic increase of the resistivity with decreasing T [12,13] and an extremum of the thermopower at low T [14–16].

CePdAl stands out due to the coexistence of geometric frustration with antiferromagnetic (AF) order below $T_N = 2.7$ K in a metal [9,14] and is located close to quantum criticality [17,18]. Neutron diffraction experiments [9] and ^{27}Al NMR measurements [19] reveal that one-third of the

Ce moments do not participate in the long-range order down to 30 mK. Theoretical models considering a quasi-two-dimensional magnetic structure based on the neutron experiments performed on polycrystals [9] suggest that the Ce moments of the hexagonal basal plane order in ferromagnetic chains which are antiferromagnetically coupled and separated from each other by the frustrated interjacent moments [inset of Fig. 1(b)] [20]. In the c direction, this structure is repeated with an incommensurate AF modulation. Because of the crystal-electric-field-induced large single-ion magnetic anisotropy between the easy c axis and the basal plane, CePdAl can be regarded as being effectively Ising-like [21].

Since in CePdAl the frustrated moments (1/3 of the Ce moments) are correlated but neither freeze nor order and act as effective spin-1/2 particles, this compound fulfills the basic preconditions for a fermionic quantum SL [1]. It is, however, unclear whether such a state can exist in a HF system, as at low T the Kondo interaction might screen the moments and suppress a possible SL state by quenching the correlations between the frustrated moments without destroying the magnetic order [19,20,22–24]. Even in this case, however, it has been suggested that a SL state may evolve [25]. Usually, Kondo and RKKY interactions slightly differ in their magnetic field dependence. Therefore, in an attempt to disentangle geometric frustration from Kondo interaction, we study a Czochralski-grown CePdAl single crystal [16] in magnetic fields up to $B = 14$ T between 30 mK and 10 K. Here and in the following, $B = \mu_0 H$ and H is the magnetic field strength. As the frustration enhances the degeneracy of the system, we use specific heat and magnetization measurements to

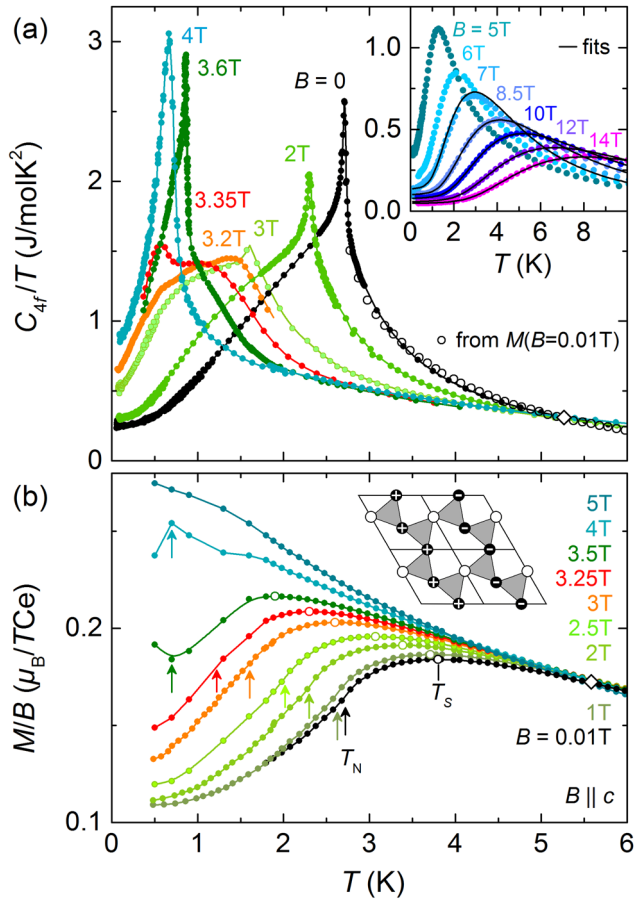


FIG. 1. (a) The $4f$ contribution to the specific heat plotted as C_{4f}/T vs temperature T for fields along the c axis. The open dots are estimated from the magnetization M at $B = 0.01$ T (see text). The inset shows the Schottky anomaly at higher magnetic fields B with the fits of the resonance-level model for $B \geq 7$ T (see text). (b) M/B vs T for different B . Open symbols indicate the entropy maximum at T_S and the arrows the magnetic transition determined from C_{4f}/T . The magnetic structure of the basal plane is sketched in the inset [9,17,20]. Crossing points $T_N^{(C)}$ and $T_N^{(x)}$ of C_{4f} and M/B , respectively, are marked by open diamonds. The lines are guides to the eyes.

determine the T and B dependence of the entropy S . In addition, we track the phase boundaries with measurements of the magnetocaloric effect, magnetostriction, and thermal expansion [26].

In Fig. 1(a), we show the $4f$ contribution C_{4f} of the Ce ions to the specific heat C as C_{4f}/T vs T at several fields. At zero field, C_{4f}/T is dominated by a sharp λ -like anomaly at $T_N = 2.7$ K. Its asymmetric shape suggests the presence of a shoulder at ≈ 2 K that shifts to lower T and becomes more pronounced with increasing B . At $B \approx 3.35$ T, this feature, now being shifted to 0.6 K, has surmounted that of the by-now broadened transition anomaly at 1.05 K. With further enhanced B , both peaks vanish, and another even larger transition anomaly appears; see the data for $B = 3.6$ and 4 T. Finally, when the

magnetic order is suppressed at $B_c(T \rightarrow 0) \approx 4.1$ T, a Schottky-like anomaly emerges and moves with B to higher T [inset of Fig. 1(a)]. This is due to the field-induced splitting of the Ce^{3+} -ground-state doublet. At $B \geq 7$ T, this anomaly can be described in terms of the single-ion resonance-level model of Zeeman-split quasi-particle levels [27]; cf. solid lines in the inset of Fig. 1(a) and fit values [26]. Recent polarized neutron experiments show that in this field range the intersite correlations are suppressed [28]. The temperature of the C_{4f}/T maximum is proportional to the Zeeman energy. A linear extrapolation of its B dependence from fields above 7 T not affected by correlations allows us to determine the crossover field $B_K \approx 2.5$ T. Here, the Kondo screening is supposedly suppressed and, consequently, the twofold degenerate ground-state levels become energetically distinct. The fact that B_K is well below B_c indicates that the Kondo screening becomes ineffective prior to the suppression of magnetic order.

The geometric frustration becomes apparent by fluctuations visible in C_{4f}/T well above T_N . The affected T and B range is marked by a crossing point at $T_N^{(C)} \approx 5.3$ K and $B < 5$ T [Fig. 1(a)] [29,30]. This point is defined by $\partial(C_{4f}/T)/\partial B = 0$, which, according to the Maxwell relation $\partial M/\partial T = \partial S/\partial B$, implies $\partial^2 M/\partial T^2 = 0$. The related sign change of $\partial^2 M/\partial T^2$ uncovers an increasing deviation from Curie-Weiss-like behavior and the tendency of M to saturate below $T_N^{(C)}$. M , indeed, reaches a maximum at a temperature T_S and drops at T_N again [Fig. 1(b)]. The absence of a Curie-Weiss-like upturn of $M(T)$ at low T demonstrates that the frustrated moments are correlated.

Figure 1(a) shows that C_{4f}/T (black closed circles) and $\partial[(M/B)T]/\partial T$ (open circles) for $B \rightarrow 0$ are proportional to each other, with a proportionality factor of $13.7 \text{ JTce}/\mu_B \text{ mol K}^2$. According to Fisher [31,32], this proportionality arises near a second-order antiferromagnetic phase transition and demonstrates that the extended tails of C_{4f}/T above T_N and the maximum of $M(T)$ are caused by magnetic fluctuations. Specifically, as C_{4f}/T neither saturates nor exhibits a peak at T_S , the Kondo effect can be ruled out as source for the maximum of $M(T)$ at T_S [33].

By virtue of the Maxwell relation above, a maximum in $M(T)$ at T_S is equivalent to a maximum of $S(B)$ at T_S . An accumulation of entropy naturally emerges when phase boundaries are crossed by employing nonthermal control parameters as the magnetic field [34]. T_S sensitively depends on critical fluctuations. In frustrated magnetic systems, entropy accumulates at much higher T than the phase-transition temperature [35]. In a mean-field description, on the other hand, no difference between T_S and T_N exists. T_S roughly reflects the temperature where the system would order without frustration. Thus, instead of the widely used frustration parameter $f_{\text{CW}} = |\Theta_{\text{CW}}|/T_N$ (with Θ_{CW} the Curie-Weiss temperature) [36], the ratio $f_S = T_S/T_N$ can serve as the measure of the frustration strength. In contrast to f_{CW} , f_S allows field-dependent

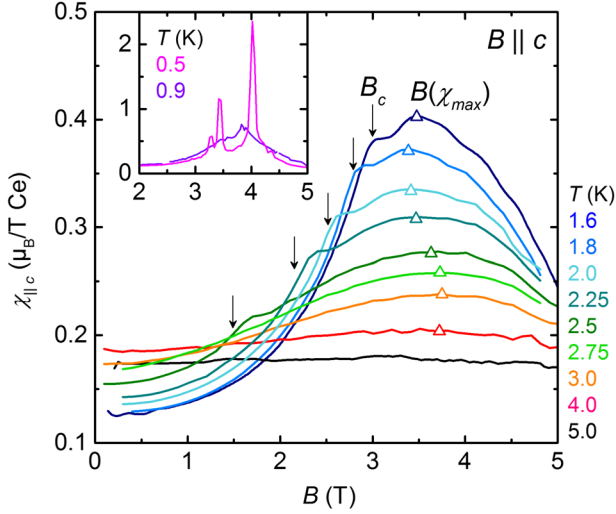


FIG. 2. Differential magnetic susceptibility $\chi_{||c} = \partial M / \partial B$ vs B . The inset presents data below $T < 1$ K. The transition fields B_c extracted from $M(T)$ and C_{4f}/T are denoted by arrows and the field of the $\chi_{||c}$ maximum $B(\chi_{\max})$ by open triangles.

studies, as long as M does not saturate at high B . Figure 1(b) confirms that indeed this is not the case in fields below 5 T and the T range of the observed M/B maximum.

To estimate the field range governed by AF correlations, we determined the differential magnetic susceptibility $\chi_{||c} = \partial M / \partial B$ from M measurements for $B || c$ at fixed T (Fig. 2). In accordance with C_{4f}/T and $M(T)$, $\chi_{||c}$ reveals AF correlations above T_N as witnessed by the maximum of $\chi_{||c}(B)$ at $B(\chi_{\max})$. This maximum is produced by the suppression of the correlations with the magnetic field. In the temperature range from T_N down to 1 K, a single phase transition appears at the critical field B_c as additional shoulder of $\chi_{||c}$. In contrast to usual antiferromagnets, $B(\chi_{\max})$ is well separated from B_c and stays roughly constant at ≈ 3.6 T (Fig. 2). Its upper temperature limit is given by the crossing region of M/B at $T_N^{(x)} = 5.5 \pm 1$ K and $B < 5$ T [Fig. 1(b)]. As here $\partial(M/B)/\partial B = 0$, $\partial M / \partial B = \chi_{||c}$ is constant. At further decreased $T < 1$ K, three sharp peaks arise (cf. inset of Fig. 2), which indicate metamagnetic transitions in agreement with previous measurements [37].

To establish the phase boundaries, we measured the magnetostriction $\lambda_a = (1/L_a)(\partial L_a / \partial B)$ (with L_a as the crystal length in the a direction) by varying B at fixed T to obtain horizontal cuts through the (B, T) phase diagram. A three-dimensional plot of λ_a vs T and B is shown in Fig. 3. Just as $\chi_{||c}$, λ_a clearly reveals three sharp peaks at $T \leq 1$ K indicative of first-order transitions. At higher T , only one transition remains present, visualized by the dotted line in the (B, T) plane of Fig. 3. The change of the height and sharpness of the peaks with increasing T suggests crossovers from first- to second-order transitions.

By extracting the transition temperatures from all data and from additional measurements of the thermal

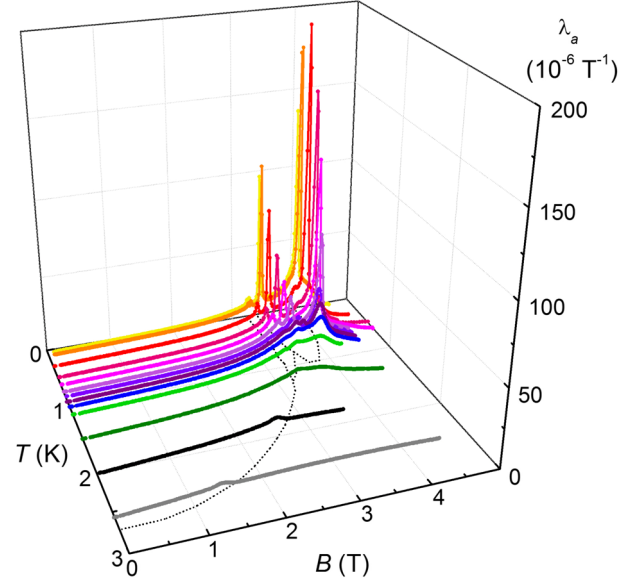


FIG. 3. Magnetostriction λ_a of the a axis for fields along c . The phase diagram is shown beneath under the data. The discontinuous length changes indicative of first-order transitions give rise to sharp peaks at low temperatures.

expansion and magnetocaloric effect (not shown), we construct the magnetic phase diagram depicted in Fig. 4(a), which comprises an extended phase (AF₁) between zero field and $B_{c1} = 3.25$ T, an intermediate phase (AF₂) at $B_{c1} < B < B_{c2} = 3.4$ T, and a smaller pocket (AF₃) which ends at $B_c = B_{c3}$. The phase boundaries between AF₁ and AF₂ and those surrounding the AF₃ phase are first-order transitions. The large magnetic anisotropy below T_N (with $10\chi_{\perp c} \approx \chi_{||c}$) prohibits a canted alignment of the moments [21,38] and renders the low- T metamagnetic transitions discontinuous.

The AF₁ and AF₂ phases show the additional, previously mentioned, shoulder of C_{4f}/T below T_N [Fig. 1(a)]. As displayed in Fig. 4(a) and its inset, the shoulder (open circles) becomes more pronounced with increasing B within the AF₁ order, shifts to lower T until it reaches B_{c1} , and remains constant at $T \approx 0.5$ K in the AF₂ phase. This feature finally terminates at B_{c2} , the border to the AF₃ phase. Because of its broadness, it cannot be attributed to the onset of long-range order but rather points to a crossover. Although the origin of this shoulder is unknown, its field dependence implies a change in the correlations of the frustrated magnetic moments. This conjecture is supported by neutron-scattering experiments which reveal a lock-in of the magnetic propagation vector in a similar temperature range at zero field [39,40].

We note that our data display no signs of additional transitions below the AF phase boundaries. Classical SLs can, however, leave the thermodynamic equilibrium by freezing into disordered spin configurations which are manifested in a finite zero-point entropy [2,41]. We, therefore, determined S as a function of T and B by

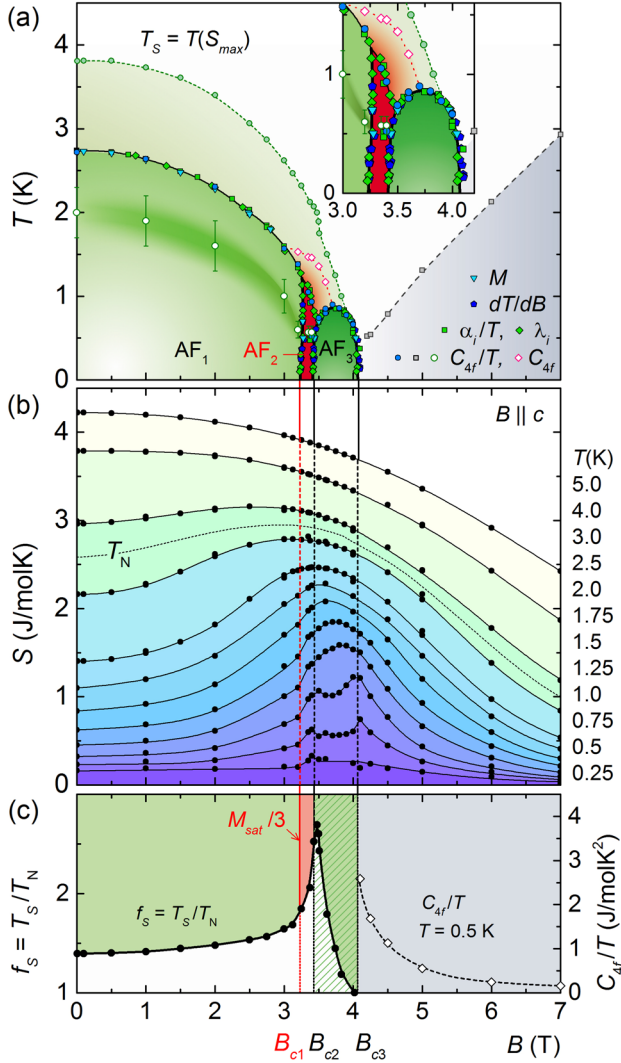


FIG. 4. (a) Magnetic phase diagram of CePdAl for $B \parallel c$ obtained from measurements of different properties as indicated in the figure. The subscript i for α_i and λ_i denotes the different axes a or c . Open circles indicate the approximate temperature of the broad shoulders of C_{4f}/T below T_N , gray squares the position of the maxima of the Schottky anomalies [Fig. 1(a)], and red open diamonds the temperature of the C_{4f} maximum. The inset shows an enlarged view of the AF_2 and AF_3 phases. (b) Entropy $S(B)$ at different fixed temperatures. (c) Frustration parameter $f_s = T_S/T_N$ vs B . For $B > B_{c3}$, C_{4f}/T at $T = 0.5$ K is plotted against B . All lines are guides to the eye.

combining C_{4f}/T and M measurements. The integration of $C_{4f}/T = \partial S/\partial T$ allows us to calculate $S(T)$ apart from a field-dependent constant $S_0(B)$. S_0 was estimated from $\int \partial M/\partial T|_T dB$ using the above Maxwell relation. The remaining integration constant was set to zero at $B > 9$ T where, as mentioned before, the geometric frustration is lifted. The derived S values are plotted in Fig. 4(b). In the entire investigated field range, S approaches zero with decreasing T , ruling out a strongly degenerate ground state. This opens the possibility that, while below $B_K = 2.5$ T the

intersite correlations between the frustrated moments might be removed by Kondo screening, at higher fields a SL dominated by quantum fluctuations develops [1].

The $S(B, T = \text{const})$ data clearly reveal a pronounced maximum at $T < 3.7$ K whose position depends on T . That temperature is nothing but T_S defined above. With decreasing T , it roughly follows the outer AF phase boundaries [Figs. 4(a) and 4(b)]. When the AF_3 phase is entered below 1 K, the maximum collapses, and two smaller peaks appear that merge into the discontinuous phase boundaries of the AF_3 pocket.

We are now able to specify the level of frustration by determining $f_s = T_S/T_N$ as a function of B [Fig. 4(c)], where T_N is defined by the outer phase boundaries. In the AF_1 phase, f_s stays almost constant over a wide field range. Upon approaching AF_2 , however, it starts to grow and shoots up when the phase boundary is crossed at B_{c1} . f_s reaches its highest value at the border B_{c2} to the AF_3 phase. Beyond B_{c2} , the sudden drop of f_s indicates that the frustration is continually removed due to the incipient order of the frustrated moments in agreement with the collapse of S in the AF_3 phase. At fields beyond $B(\chi_{\text{max}})$, the fluctuations fade out and neither a maximum of $\chi_{\parallel c}$ nor of S can be found.

According to previous measurements, M exhibits at $T = 0.5$ K three distinct steps as a function of B [12], corresponding to the sharp peaks in $\chi_{\parallel c}$ and the even stronger singularities in λ_a reported here. At B_{c1} , M reaches $1/3$ of the saturated moment $M_{\text{sat}} \approx 1.6\mu_B/\text{Ce}$ [9,12]. This suggests that with increasing B , the Kondo screening is suppressed at $B_K < B_{c1}$ and that the liberated Ce moments align along the c axis. In a simplified view, the magnetic structure of the basal plane changes at B_{c1} from $\uparrow\downarrow$ to $\uparrow\uparrow\downarrow$. The added, field-polarized moments interfere with the next-nearest-neighbor AF interaction and destabilize the magnetic order at B_{c1} . This leads to a significant strengthening of the frustration and an increase of S . Here, compared to the other phase boundaries, the transition anomalies are strongly broadened and diminished [Fig. 1(a)]. The broadening leads to a significant difference between the positions of the anomalies found in C_{4f}/T and C_{4f} . The peaks observed in C_{4f} occur at distinctively higher T [red open diamonds in Fig. 4(a)] and do not coincide with the transition temperatures extracted from $\chi_{\parallel c}$, λ_a , and thermal expansion measurements. With further enhanced field, the frustration increases even more, until the unstable frustrated magnetic structure gives way to the formation of the AF_3 phase. This leads to the collapse of S and ultimately lifts the frustration at B_{c3} .

At still higher fields where M approaches M_{sat} , the specific heat still reveals fluctuations indicated by enhanced C_{4f}/T values at low T , which slowly fade out [Fig. 4(c)]. These fluctuations, however, originate from the competition between the intersite AF correlations and the ferromagnetic alignment along the applied field as evidenced by the maximum of $\chi_{\parallel c}$.

In conclusion, our comprehensive measurements show that in CePdAl the geometric frustration persists in a wide field range and is reflected in a rich structure of the entropy $S(B, T)$. Moderate fields gradually suppress the Kondo screening of the magnetic moments. The resulting increase of the frustration and the entropy indicate that the most promising field range to search for a spin liquid is given by the AF₂ phase. This phase is characterized by rounded, ill-defined transition anomalies and a prominent shoulder of the specific heat at low temperatures. To clarify the nature of this phase and the possible existence of a corresponding new type of spin liquid intertwined with a magnetically ordered solid with competing interactions, further experimental and theoretical efforts are mandatory.

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Note added in proof.—Recently an article on the temperature-field phase diagram of geometrically frustrated CePdAl appeared [42].

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