Thermal and magnetic properties of a low-temperature antiferromagnet Ce$_4$Pt$_{12}$Sn$_{25}$

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We report specific heat (C) and magnetization (M) of single crystalline Ce$_4$Pt$_{12}$Sn$_{25}$ at temperature down to $\sim$ 50 mK and in fields up to 3 T. $C/T$ exhibits a sharp anomaly at 180 mK, with a large $\Delta C/T \sim 30$ J/mol K$^2$.Ce, which, together with the corresponding cusp-like magnetization anomaly, indicates an antiferromagnetic (AFM) ground state with a Néel temperature $T_N = 180$ mK. Numerical calculations based on a Heisenberg model reproduce both zero-field C and M data, thus placing Ce$_4$Pt$_{12}$Sn$_{25}$ in the weak exchange coupling $J < J_c$ limit of the Doniach diagram, with a very small Kondo scale $T_K \ll T_N$. Magnetic field suppresses the AFM state at $H' \approx 0.7$ T, much more effectively than expected from the Heisenberg model, indicating additional effects possibly due to frustration or residual Kondo screening.

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I. INTRODUCTION

Compounds with elements containing 4$f$- and 5$f$-electrons, such as Ce, Yb, and U, have been subjects of intense research over the past several decades. Rich physics displayed within this class of materials can be directly attributed to the interaction of 4$f$-electrons with conduction electrons, which leads to, among other aspects, competing ground states. Heavy electron ground states are often thought to be a consequence of the Kondo interaction, which decays as a power law in $k_T$, where $k_T$ is Sommerfeld coefficient, $C/T \propto - \ln T$ behavior found in CeCu$_{5.9}$Au$_{0.1}$, YbRh$_2$Si$_2$, and CeCoIn$_5$, linear, in temperature, resistivity, and other anomalous properties, provides additional impetus to research on competition between HF physics and magnetism. The NFL behavior in d- and f-electron systems is reviewed, for example, in Refs. 11,12.

There have been a number of theoretical attempts to model such systems using the periodic Anderson model$^{13}$ and Kondo lattice model$^{14}$. A mean field model of the Kondo lattice including the nearest-neighbor magnetic interaction yields a reduction in the Kondo temperature, $T_K$, at $J > J_c$ compared to its value for a single impurity. This result emphasizes the importance of the RKKY coupling even when the system is in the Kondo (HF) ground state. On the other hand, for $J < J_c$, on the AFM side of the QCP in the Doniach analysis, a mean field renormalization group approach provided a good agreement between theory and the experimental results on the pressure-tuned AFM transition temperatures in a series of CeT$_2$Si$_2$ ($T$ = Ru, Rh, and Pd) compounds. However, the interplay between the Kondo and RKKY interactions in this regime is not yet fully understood, and additional work, both theoretical and experimental, is needed. In this paper we provide an example of a compound which orders magnetically at low temperature, in the regime where previously studied compounds display dominant Kondo screening.

An alternative to the pressure route of tuning the relative strength of the Kondo screening and AFM coupling is via synthesis, i.e. crystallographic structure of the compounds. For a specific value of the on-site f-c exchange constant $J$ one expects the RKKY interaction, which decays as a power law in $k_F r$, where $k_F$...
is the Fermi momentum and \( r \) is the distance between the magnetic ions, to become weaker the further the \( f \) - atoms are separated from one another. As a result, the Kondo screening should commonly win over magnetic ordering in such dilute systems. It would therefore be particularly interesting to explore the \( f \) - electron bearing compounds with large \( f \) - \( f \) nearest neighbor distances. Several stoichiometric dilute \( f \) - electron compounds have been useful in this regard. Ce-based filled skutterudites Ce\(_4\)X\(_{12}\) (\( T = \) Fe, Ru, Os; \( X = \) P, As, Sb, \( d_{\text{Ce}-\text{Ce}} \sim 7 \) \( \text{Å} \)) form one of these families, with most members semiconducting with the gap size that correlates with the lattice constant.\(^{19}\) Another example is a family of Yb-based HF compounds Yb\(_4\)A\(_2\)Zn\(_{20}\) (\( A = \) Fe, Co, Ru, Rh, Os, Ir, \( d_{\text{Yb}-\text{Yb}} \sim 6 \) \( \text{Å} \))\(^{20,22}\), where the Kondo physics appears to dominate the RKKY interaction, in accord with the simple argument above.

The approach of synthesizing compounds with large \( f \) - \( f \) nearest neighbor distance also turned out to be useful in the studies of electronic correlations in uranium compounds. Large U-U distance \( d_{\text{U}-\text{U}} \sim 6 \) \( \text{Å} \) in UM\(_2\)Zn\(_{20}\) (\( M = \) Co, Rh) family of materials reduces the overlap between the \( 5f \) - electron wave functions, allowing for observation of sharp crystal electric fields, a situation very rare in U compounds.\(^{24}\) At the same time, this U-U separation resulted in a good description of the system within a Kondo limit of the Anderson model, i.e. UM\(_2\)Zn\(_{20}\) compounds are found to be well on the Kondo side of the QCP, similar to the Yb\(_4\)T\(_2\)Zn\(_{20}\) case above, and again in accord with expectations above.

Ce\(_4\)Pt\(_{12}\)Sn\(_{25}\) presents another example of a metallic \( 4f \) - dilute Ce-based system,\(^{23}\) with Ce-Ce interatomic distance \( d_{\text{Ce}-\text{Ce}} = 6.14 \) \( \text{Å} \). It therefore appears to be a good candidate to continue exploration of correlated electron physics in \( f \) - electron systems, and in particular the competition between the RKKY interaction and Kondo screening. Ce\(_4\)Pt\(_{12}\)Sn\(_{25}\) is a cubic compound lacking 4-fold point symmetry with three inequivalent Sn sites. Recently, we succeeded in growing single crystals of Ce\(_4\)Pt\(_{12}\)Sn\(_{25}\) with relatively large physical dimensions (up to \( \sim 5 \times 5 \times 5 \) \( \text{mm}^3 \))\(^{25}\). Previous specific heat, resistivity, and ac-susceptibility measurements uncovered a phase transition at 0.18 K.\(^{25}\) However, the origin of the transition was not identified. In this paper, we report the low-temperature specific heat (\( C \)) and magnetization (\( M \)) measurements in magnetic field. From the cusp and the field dependence in \( M(T) \), which corresponds to the specific heat anomaly, we conclude that Ce\(_4\)Pt\(_{12}\)Sn\(_{25}\) is an antiferromagnet with \( T_N = 0.18 \) K in zero field. Evolution of \( C/T \) and \( M \) with magnetic field indicates disappearance of AFM ordering and subsequent splitting of an \( f \) - electron ground state doublet. We find that the signatures of the magnetic transition are reasonably close to those described by a spin-1/2 Heisenberg model, albeit with notable differences, see below. This suggests that even for the large distance between the neighboring \( 4f \) - ions the RKKY interaction plays a very significant, or even dominant, role.

## II. EXPERIMENTAL DETAILS

Single crystals of Ce\(_4\)Pt\(_{12}\)Sn\(_{25}\) were grown by Sn self-flux method. The details of the sample growth and the physical properties are described in Ref.\(^23\). Specific heat was measured in a SHE dilution refrigerator with 9 T superconducting magnet by means of a quasi-adiabatic heat pulse method with a Ru\(_2\)O\(_4\) thermometer. The low-temperature high-resolution DC magnetization measurements were performed in a commercial Oxford dilution refrigerator with 12-14 T superconducting magnet. We used a capacitive Faraday magnetometer cell with applied field gradient of 10 T/m. The principle of the magnetization measurement is described in Ref.\(^{26}\). By comparing the data from a commercial Magnetic Property Measurement System (MPMS; Quantum design) up to 7 T and down to 2 K with that obtained with capacitive method in the same field and temperature range, we are able to determine the absolute value of the magnetization. We used three samples from different batches labeled as samples #1 (7.18 mg), #2 (2.58 mg) and #3 (3.70 mg) in the figures in this paper for specific heat measurements. We used a sample weighing 4.63 mg for magnetization measurements.

## III. RESULTS AND DISCUSSIONS

### A. Specific heat

Figure 1 shows the temperature dependence of \( C/T \) of Ce\(_4\)Pt\(_{12}\)Sn\(_{25}\) (50 mK - 3 K) for samples #1, #2 and #3, and that of a non-magnetic analogue La\(_4\)Pt\(_{12}\)Sn\(_{25}\) (0.4 K - 3 K) in zero field. \( C/T \) of La\(_4\)Pt\(_{12}\)Sn\(_{25}\) is negligible compared to that of Ce\(_4\)Pt\(_{12}\)Sn\(_{25}\) in the investigated temperature range. Therefore, the obtained specific heat of Ce\(_4\)Pt\(_{12}\)Sn\(_{25}\) can be regarded as purely of magnetic origin \( C_{\text{mag}} \). With temperature decreasing from 3 K, \( C/T \) monotonically increases down to 0.18 K, at which point it exhibits a sharp anomaly with the magnitude of the jump \( \Delta C/T \sim 30 \text{J/mol K}^2 \cdot \text{Ce} \). From magnetization measurements we identify this anomaly as due to antiferromagnetic (AFM) ordering, as discussed below. In addition, low-temperature \( T^2 \)-variation of \( C/T \), shown in the inset, is consistent with the spin-wave contribution in the AFM state. Solid circles represents the temperature dependence of the magnetic entropy \( S_{\text{mag}}(T) \) of Ce\(_4\)Pt\(_{12}\)Sn\(_{25}\):

\[
S_{\text{mag}}(T) = \int_0^T \frac{C_{\text{mag}}(T)}{T} \, dT
\]  

(1)

Entropy gain is \( \approx 0.5 \text{Rln} 2 \) at \( T_N \) and reaches the value of 1.0 \text{Rln} 2 at 3 K. This indicates that the crystal electric field (CEF) ground state of Ce\(^{3+} \) in Ce\(_4\)Pt\(_{12}\)Sn\(_{25}\) is a \( \Gamma_7 \) doublet. One remarkable feature of the specific heat of Ce\(_4\)Pt\(_{12}\)Sn\(_{25}\) is the long tail above \( T_N \). As we show below, a substantial part of this tail is due to quantum fluctuations of \( f \) - electron spins above \( T_N \). However, from our
FIG. 1: (Color online) (a) Temperature dependence of $C/T$ of Ce$_4$Pt$_{12}$Sn$_{25}$ for samples #1, #2 and #3, and of the La-analog La$_3$Pt$_{12}$Sn$_{25}$ in zero field. Solid dots represent the temperature dependence of magnetic entropy $S_{\text{mag}}$ in units of R ln 2. Inset: $C/T$ vs $T^2$ at low temperature. The solid line is a linear least-squared fit to the data. (b) $\Delta - C/T$ and $\bullet$ - $S$ of Ce$_4$Pt$_{12}$Sn$_{25}$ (sample #3). Solid and dashed lines represent specific heat and entropy, respectively, from the Heisenberg model calculations described in the text.

FIG. 2: (Color online) Temperature dependence of $C/T$ of Ce$_4$Pt$_{12}$Sn$_{25}$ in the magnetic fields, (a) 0-0.65 T, (b) 0.6-3 T. The arrows indicate the temperature where $C/T$ behavior was reported in the experimental data is rather good. In particular, the behavior of the entropy below $T_N$ is very similar, and the entropy at $T_N$ for model calculations is about 0.55 R ln 2, about 10% larger than the experimental value. The most notable difference is that experimental entropy is lower than the calculated values above the transition, indicating that some degrees of freedom remain "locked". This discrepancy is also seen from the somewhat longer tail of the measured specific heat in Fig. 2(b) relative to the Heisenberg model. Such contribution may still be due to remnant Kondo physics. Recall that the distance between Ce ions is large, and therefore RKKY exchange, and correspondingly the critical $J_c$, is reduced. In the framework of Doniach phase diagram, the only situation when RKKY interaction can still dominate the Kondo coupling is when the bare exchange $J$ is very small. Ce$_4$Pt$_{12}$Sn$_{25}$ is therefore in the low $J < J_c$ limit of the

analysis it is likely that other processes, such as residual Kondo screening, or frustration of spin-spin interactions play some role. Similar $C/T$ behavior was reported in the structurally frustrated system Yb$_2$Pt$_2$PdSn, for example.

To elucidate the origin of this behavior we performed Quantum Monte Carlo simulations of the spin - 1/2 three-dimensional Heisenberg model in zero and applied magnetic field. The exchange interaction was fixed to have the AFM ordering temperature of $T_N = 0.18$ K for $H = 0$, while the $g$ - factor of the spins was determined from the saturation magnetization at high fields (see below). The results for zero field specific heat and entropy are displayed in Fig. 3 as solid and dashed curves, respectively. The agreement between the model calculations and experimental data is rather good. In particular, the behavior of the entropy below $T_N$ is very similar, and the entropy at $T_N$ for model calculations is about 0.55 R ln 2, about 10% larger than the experimental value. The most notable difference is that experimental entropy is lower than the calculated values above the transition, indicating that some degrees of freedom remain "locked". This discrepancy is also seen from the somewhat longer tail of the measured specific heat in Fig. 3(b) relative to the Heisenberg model. Such contribution may still be due to remnant Kondo physics. Recall that the distance between Ce ions is large, and therefore RKKY exchange, and correspondingly the critical $J_c$, is reduced. In the framework of Doniach phase diagram, the only situation when RKKY interaction can still dominate the Kondo coupling is when the bare exchange $J$ is very small. Ce$_4$Pt$_{12}$Sn$_{25}$ is therefore in the low $J < J_c$ limit of the
Doniach phase diagram, and the system orders magnetically below $T_N = 0.18$ K. Note that Ce-based skutterudite compounds and Yb$_2$Al$_2$Zn$_{20}$, whose Ce- Ce and Yb-Yb distances are similar to that in Ce$_4$Pt$_{12}$Sn$_{25}$, do not show magnetic ordering, and are in the $J > J_\text{K}$ Kondo limit. It should also be noted that there is no significant sample dependence for different samples #1, #2, and #3, with respect to the ordering temperature, the magnitude of the anomaly in $C/T$, and its width.

Figure 2 shows $C/T$ for magnetic field (a) $0 \leq B \leq 0.65$ T and (b) $0.65 \leq B \leq 3.0$ T. As shown in Fig. 2(a), the steep jump in $C/T$ associated with AFM ordering is quickly suppressed, and $T_{\text{max}}$, where $C/T$ has its maximum value, gradually shifts to lower temperature, as indicated by arrows. On the other hand, Fig. 2(b) shows that with further increase of magnetic fields, $T_{\text{max}}$ shifts to higher temperature, while the peak height is continuously suppressed. This behavior can be understood as follows: In the low field region the reduction of $T_{\text{max}}$ is ascribed to the suppression of the AFM ordering with field ($T_{\text{max}} \approx T_N$). Around 0.7 T, the feature associated with the AFM order is suppressed entirely. Further increase of magnetic field increases Zeeman splitting of the crystal electric field (CEF) doublet ground state (see below), leading to a Schottky anomaly in specific heat with rising $T_{\text{max}}$. Similar behavior has been observed in a number of other Ce compounds, e.g. CeCu$_{5.9}$Au$_{0.1}$
, CeCu$_2$Si$_2$
, and (La, Ce)Al$_2$
. As seen in the inset of Fig. 2(a), $S_{\text{mag}}$ reaches 1.0$R\ln2$ independently of applied fields up to 3 T, although the ground state and the shape of the specific heat anomaly both vary strongly with applied field, reflecting a two-fold degeneracy of the CEF doublet.

At high magnetic fields, far exceeding the Heisenberg exchange coupling, we can obtain the approximate behavior of Ce$_4$Pt$_{12}$Sn$_{25}$ based on the CEF level scheme. To avoid complication in parameterization, we assume that a cubic point symmetry of Ce ions in Ce$_4$Pt$_{12}$Sn$_{25}$ is $O_h$ and the CEF Hamiltonian $H_{\text{CEF}}$ can be reduced to the following formula:

$$H_{\text{CEF}} = \sum_{m,n} B^m_n O^m_n = B^4_0 (O^4_0 + 5O^4_4),$$

(2)

where $B^m_n$ and $O^m_n$ are the CEF parameters and the Stevens operators.

The total Hamiltonian $H$ and specific heat $C$ of CEF levels in external magnetic fields is then given by the following expression:

$$H = H_{\text{CEF}} - g_J \mu_B \mathbf{J} \cdot \mathbf{H}$$

(3)

$$C = \frac{\partial}{\partial T} \frac{1}{Z} \sum_n E_n e^{-\beta E_n}$$

(4)

where, $g_J$ is the Lande $g$-factor, $E_n$ and $|n\rangle$ are the $n$th eigenvalue and eigenfunction, respectively, $Z = \sum_n e^{-\beta E_n}$, $\beta = 1/k_B T$ and $k_B$ is the Boltzmann constant. We assume that the ground state is a $\Gamma_7$ doublet.

$B^0_0 = 0.5$ T then corresponds to the CEF splitting between $\Gamma_7$ and the first excited state $\Delta \approx 200$ K, which can be inferred from $S_{\text{mag}}(T)$ and resistivity data, respectively. As shown in the inset of Fig. 2(b), the calculated specific heat anomaly at 3 T (dotted curve) closely reproduces experimental results, supporting the CEF scheme suggested above.

Quick suppression of the AFM specific heat anomaly in the low field regime, displayed in Fig. 2(a), and particularly the broadening of the anomaly with magnetic field, makes precise identification of $T_N(H)$ difficult. Encouraged by the success of our Heisenberg model calculations in zero field, we performed calculations for this model in finite field, with the goal to aid in identification of $T_N$ in experimental data. Figure 3 displays the results of these calculations. There are striking differences between the model and experimental data. In the Heisenberg model the specific heat anomaly at the AFM phase transition remains sharp, and persists to high fields over 1 Tesla, while the experimentally observed anomaly is washed out already by 0.5 T. There is a number of possible reasons for this discrepancy. A Heisenberg model is often used to describe insulating compounds, with coupling $J$ independent of the magnetic field. In our case the magnetic coupling is mediated by the conduction electrons, with a potential for a field-dependent magnetic coupling. The effect of the next nearest neighbor interaction, with accompanying effects of possible frustration, is also neglected in the present calculations. Among other complications is a possible proximity to a HF (Kondo) ground state. Our results point to a number of fruitful future theoretical inquiries, such as field dependent RKKY interaction, or including frustration within the Heisenberg model due to next nearest neighbor interactions, and its response to magnetic field.
the magnetization is monotonic, suggesting that AFM order is suppressed in the vicinity of this field. Above 2.0 T, the magnetization develops a plateau at low temperature, as seen in Fig. 4(b). The plateau is related to the field splitting of the CEF ground state doublet. In fact, calculations based on the CEF scheme assumed above also show similar behavior, as well as good agreement in absolute value, as indicated by a dashed curve in the inset of Fig. 4(b) obtained for 3 T using the following formula:

\[
\chi = \frac{N(g_J\mu_B)^2}{Z} \left( \sum_{m \neq n} |\langle m | J_z | n \rangle|^2 \frac{1 - e^{E_m - E_n}}{E_m - E_n} e^{-\beta E_n} \right) + \sum_n |\langle n | J_z | n \rangle|^2 \beta e^{-\beta E_n} \tag{5}
\]

The results of the Heisenberg model calculations of magnetization are displayed in Fig. 5. Similar to specific heat, the AFM transition remains very sharp and persists to higher fields within the model calculations compared to the experimental data.

Figure 6 shows isothermal magnetization curves \( M(H) \) of Ce\(_4\)Pt\(_{12}\)Sn\(_{25}\) at temperature of 0.1 K (< \( T_N \)), 0.2 K (≈ \( T_N \)) and 2 K (≫ \( T_N \)). In contrast to \( M(H) \) data at 2.0 K, which increases slowly with field and does not show complete saturation up to 7 T, \( M(H) \) at 0.2 K rapidly increases with field and exhibits a plateau above 2 T. The saturated value of the magnetic moment \( \mu_s \) corresponds to about 0.71 \( \mu_B/\text{Ce} \), which is much smaller than the value expected for a free Ce\(^{3+} \) ion of \( g_J J \mu_B = 2.14 \mu_B \) (\( g_J = 6/7, J = 5/2 \)). This reduction, as well as deviation of \( \chi \) at low temperature, reflects CEF effects in Ce\(_4\)Pt\(_{12}\)Sn\(_{25}\). Using formulas (2), (3), and (6) below, we can calculate the magnetization by using the value of \( B_d^0 = 0.5 \text{ K} \), the same as the one employed in calculations of the specific heat above.

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**B. Magnetization**

Figure 4 shows temperature dependence of the magnetization \( M(T) \) for \( T \ll 2 \text{ K} \) in fields (a) up to 0.8 T and (b) between 0.7 and 3.0 T. The low-temperature data connect well to the higher temperature data obtained using a MPMS (not shown). The cusp in \( M \) vs. \( T \) in the low field region identified by arrows in Fig. 4(a) reflects the AFM ordering phase transition. As an example, the inset of Fig. 4(a), shows \( M(T) \) at 0.15 T, on expanded scale, which exhibits a clear cusp around \( T_N = 0.18 \text{ K} \). It shifts to lower temperatures with field, in accord with the field evolution of the specific heat anomaly. Together, specific heat and magnetization measurements prove that Ce\(_4\)Pt\(_{12}\)Sn\(_{25}\) undergoes an AFM phase transition at \( T_N = 0.18 \text{ K} \) in zero field. With magnetic field increasing above 0.6 T, the cusp feature disappears, and
also displayed in Fig. 6, for the CEF model and the data is good for $T = 0.2$ K and $2$ K, respectively, based on a CEF scheme. Solid curve is a result of the Kondo screening model described in the text. Dotted curve and crosses are numerical results based on the Heisenberg model at $0.2$ K and $0.1$ K, respectively. The inset shows the magnetization curve at $0.1$ K. The arrow indicates the field where the data show anomaly due to antiferromagnetic ordering.

Equation (6)

$$M = \frac{g_1 \mu_B}{Z} \sum_n \langle n | J_z | n \rangle \beta e^{-\beta E_n}$$

The magnetization calculated from this CEF model is shown in Fig. 6 for $T = 0.2$ K (dashed line) and for $T = 2$ K (dash-dotted line). Agreement between the CEF model and the data is good for $T = 2$ K $\gg$ $J$ (similar to that in Fig. 3(b)). As expected, there is a large discrepancy at $T = 0.2$ K between experimental results and the calculated field dependence based on a single ion CEF model, so that only the saturated magnetic moment $\mu_s = 0.74 \mu_B$/Ce is comparable to the experimental value ($\mu_s = 0.71 \mu_B$/Ce). The best fit to the data at 0.2 K below 0.4 T using a Kondo screening model gives $T_K = 1.2$ K. However, this value of $T_K$ is incompatible with specific heat results and the analysis performed above, which indicate $T_K \leq 0.2$ K. At intermediate fields the dominant physics is in the $f - f$ interactions inherent in the Heisenberg model that we used to describe specific heat results above. The magnetization calculated for $T = 0.1$ K and 0.2 K is also displayed in Fig. 6 and indeed reproduces the experimental data well. Small discrepancies at higher fields (calculations overestimate magnetization) that are more pronounced at low temperature, are most likely again due to a weak Kondo screening. It should be noted that at 0.1 K, below $T_N$ in zero field, $M(H)$ shows a kink anomaly around 0.6 T, indicated by an arrow in the inset of Fig. 6. This kink corresponds to the critical field $H^*$ where AFM order disappears, and is consistent with $C(T)/T$ and $M(T)$ in constant magnetic fields described above.

FIG. 6: (Color online) $M$ vs. $H$ curves of Ce$_4$Pt$_{12}$Sn$_{25}$ at several temperatures of 0.1, 0.2 and 2 K. The dashed and dash-dotted curves are obtained from the calculation at 0.2 K and 2 K, respectively, based on a CEF scheme. Solid curve is a result of the Kondo screening model described in the text. Dotted curve and crosses are numerical results based on the Heisenberg model at 0.2 K and 0.1 K, respectively. The inset shows the magnetization curve at 0.1 K. The arrow indicates the field where the data show anomaly due to antiferromagnetic ordering.

C. $H$-$T$ phase diagram

Figure 7 displays the temperature-field phase diagram of Ce$_4$Pt$_{12}$Sn$_{25}$ obtained from the data described in previous sections, with circles and squares representing $T_{\text{max}}$ obtained from specific heat and magnetization measurements, respectively. $T_{\text{max}}$ ($\approx T_N$) decreases as field increases up to about 0.6 T, and $T_{\text{max}}$ increases with further increase of magnetic field. $T_N$ appears to vanish at a critical field $H^* \approx 0.7$ T indicated by the dashed line, where the ground state of Ce$_4$Pt$_{12}$Sn$_{25}$ changes from AFM to paramagnetic. It is impossible to tell the exact manner in which $T_N$ goes to zero from the data presented here. As illustrated in the inset, the suppression of $T_N$ with fields is much more effective than expected from the Heisenberg model ($H^* > 1$ T). There is no divergence of Sommerfeld coefficient $\gamma$ with $T \rightarrow 0$, in contrast to common behav-
behavior in materials at a QCP. Perhaps this can be explained by a small amount of entropy associated with magnetic fluctuations at high fields, as most of it is released at a higher temperature Schottky anomaly. Low temperature spectroscopic investigations, such as NMR, μSR, or neutron scattering will be able to provide a definitive picture of how AFM order is suppressed in Ce$_4$Pt$_{12}$Sn$_{25}$.

IV. CONCLUSION

In conclusion, we have performed low-temperature field-dependent specific heat and magnetization measurements to elucidate the ground state properties of Ce$_4$Pt$_{12}$Sn$_{25}$. Magnetization measurements established that Ce$_4$Pt$_{12}$Sn$_{25}$ orders antiferromagnetically with a small ordering temperature $T_N = 0.18$ K in zero field, where $C(T)/T$ exhibits a huge jump of $\sim 30$ J/mol K$^2$. The small value of the saturated magnetic moment of 0.71 $\mu_B$/Ce at 0.2 K exhibited by magnetization as a function of magnetic field can be ascribed to CEF effects, with a $\Gamma_7$ ground state, as inferred from magnetic entropy. $T_N$ is suppressed with an initial increase of field up to 0.6 T, whereas $T_{\text{max}}$ of the maximum in $C(T)$ begins to move to higher temperature above 0.7 T. This latter evolution is ascribed to an electronic Schottky contribution from the Zeeman-split ground state $\Gamma_7$ doublet. Therefore, it is likely that the ground state of Ce$_4$Pt$_{12}$Sn$_{25}$ changes above $\sim 0.6$ T from AFM to paramagnetic. This picture is further supported by the magnetization data for both temperature and magnetic field sweeps.

The model calculations, based on a CEF scheme with a $\Gamma_7$ ground state, reproduce rather well experimental specific heat and magnetization data in high field. Zero field data are described very well by the numerical calculation based on the Heisenberg model. Small deviation between experimental data and numerical results may be due to frustration or Kondo screening with a low characteristic temperature $T_K \leq T_N = 0.18$ K. This indicates that electronic spins on Ce$^{3+}$ are not screened substantially by the conduction electrons, and places Ce$_4$Pt$_{12}$Sn$_{25}$ in the $J \ll J_c$ of the Doniach phase diagram. The Kondo temperature $T_K$ is expected to be exponentially small in this regime, and a $J^2$ dependence of the RKKY interaction stabilizes the AFM ground state in spite of a large distance between the Ce ions. Ce$_4$Pt$_{12}$Sn$_{25}$ therefore presents us with a counter example to an expectation that dilute f - electron compounds will likely fall into the Kondo screened regime.

Within the Heisenberg model calculations, the AFM transition is more robust to the magnetic field, compared to experimental data in low field regime. We raised a number of questions with regard to the possible origins of this discrepancy, such as magnetic field dependence of the RKKY interaction, and frustrating effects of the next nearest neighbor interactions, and point to a number of fruitful future theoretical inquiries.

Further experiments will be needed to explore the details of the disappearance of the AFM state in Ce$_4$Pt$_{12}$Sn$_{25}$, as well as the origin of the long temperature tail of the specific heat above $T_N$, and potential role of magnetic frustration. Experiments under pressure in particular can help answer why $T_N$ is so low in Ce$_4$Pt$_{12}$Sn$_{25}$ compared with other Ce compounds, as well as search for pressure induced superconductivity, occasionally found in the vicinity of pressure induced QCP at $P_c$ ($T_N \rightarrow 0$).

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