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Thermal and magnetic properties of a low-temperature antiferromagnet $Ce_4Pt_{12}Sn_{25}$.

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Abstract. We report specific heat (C) and magnetization (M) of single crystalline $Ce_4Pt_{12}Sn_{25}$ at temperature down to $\approx$ 50 mK and in fields up to 3 T. C/T exhibits a sharp anomaly at 180 mK, with a large jump in a Sommerfeld coefficient $\gamma = C/T$ of $\Delta\gamma$ = 30 J/molK^2-Ce, which, together with corresponding cusp-like magnetization anomaly, indicate antiferromagnetic (AFM) ground state with Nel temperature $T_N = 0.18$ K. Numerical calculations based on Heisenberg model reproduce well zero field specific heat data, and point to a very small Kondo scale T_K, clearly placing $Ce_4Pt_{12}Sn_{25}$ in the weak exchange coupling $J < J_c$ limit of the Doniac diagram. Magnetic field suppresses AFM state at $H^* \approx 0.7$ T, much more rapidly than indicated by theoretical calculations.

1. Introduction

Competition between magnetically ordered and heavy fermion states is ubiquitous in compounds with *f*-electrons. Formation of the heavy electron states is widely believed to be due to Kondo screening of the local *f*-moments [1], while the magnetism is driven by the Ruderman-Kittel-Kasuya-Yoshida (RKKY) [2, 3, 4] interaction between the *f*-moments, mediated by the conduction (c) electrons. This competition was originally addressed by Doniach in the 1D-chain model [5, 6, 7]. At low f-c exchange coupling J the long-range AFM ground state of f-ions is stabilized, while beyond some critical coupling J_c Kondo screened state is stabilized.

$Ce_4Pt_{12}Sn_{25}$ presents an example of a metallic $4f$-dilute Ce-based system [8], with Ce - Ce interatomic distance d_{Ce-Ce}=6.14Å. It therefore appears to be a good system to study the Kondo-limit of the Doniach model. $Ce_4Pt_{12}Sn_{25}$ is a cubic compound with a rather complicated unit cell with a lack of 4 - fold point symmetry, three inequivalent Sn sites, and a lattice constant a=12.38Å. Previous specific heat, resistivity, and ac-susceptibility measurements uncovered a phase transition at 0.18 K. [9] In this paper, we report the low-temperature specific heat

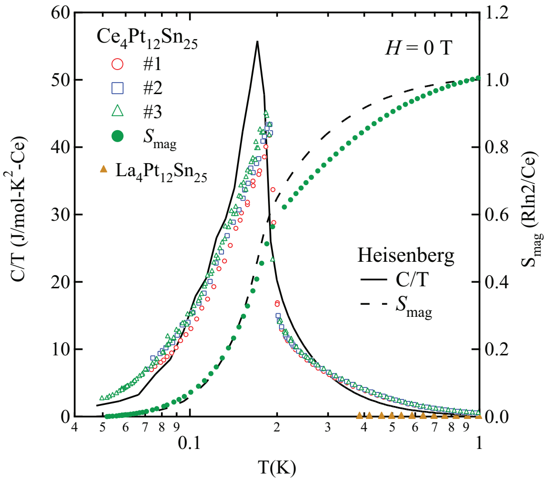

Figure 1. (Color online) (a) Temperature dependence of C/T and entropy S of $Ce_4Pt_{12}Sn_{25}$ for samples #1, #2 and #3, and of the La-analog $La_4Pt_{12}Sn_{25}$ in zero field. Solid dots represent the temperature dependence of magnetic entropy $S_{\rm mag}$ in units of $R\ln 2$. Solid and dashed lines represent specific heat and entropy, respectively, from the Heisenberg model calculations described in the text.

measurements in magnetic field, and compare them to the results of numerical calculation based on Heisenberg model. We find that the zero-field signatures of the magnetic transition are reasonably close to those described by a spin-1/2 Heisenberg model. Notable differences, however, arise with increasing magnetic field.

2. Experimental details

Single crystals of $Ce_4Pt_{12}Sn_{25}$ were grown by Sn self-flux method. The details of the sample growth and the physical properties are described in Ref. [9]. 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 RuO_2 thermometer. We used three samples from different batches labeled in the figures as samples #1 (7.18 mg), #2 (2.58 mg) and #3 (3.70 mg).

3. Results and Discussion

Figure 1 shows the temperature dependence of C/T of $Ce_4Pt_{12}Sn_{25}$ (50 mK - 3 K) for samples #1, #2 and #3, and that of a non-magnetic analogue $La_4Pt_{12}Sn_{25}$ (0.4 K - 3 K) in zero field. C/T of $La_4Pt_{12}Sn_{25}$ is negligible compared to that of $Ce_4Pt_{12}Sn_{25}$, and the specific heat of $Ce_4Pt_{12}Sn_{25}$ is of purely magnetic origin $C_{\rm mag}$. 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 $\Delta C/T \sim 30$ J/mol K^2-Ce. We identify the anomaly at 0.18 K as due to antiferromagnetic (AFM) ordering by means of magnetization measurements, not shown. Solid circles represents the temperature dependence of the magnetic entropy $S_{\rm mag}(T)$ of $Ce_4Pt_{12}Sn_{25}$. Entropy gain is $\approx 0.5\,R\ln 2$ at $T_{\rm N}$ and reaches the value of $1.0\,R\ln 2$ at 3 K. This indicates that the crystal electric field (CEF) ground state of Ce^{3+} in $Ce_4Pt_{12}Sn_{25}$ is a Γ_7 doublet. One remarkable feature of the specific heat of $Ce_4Pt_{12}Sn_{25}$ is the long tail above $T_{\rm N}$. There is a number of possible origins of such tail feature, including the onset of the Kondo screening, the development of short-range magnetic correlations in the lattice of Ce moments above the bulk transition temperature $T_{\rm N}$, or frustration of spin-spin interactions. Similar C/T behavior was reported in the structurally frustrated system Yb_2Pt_2Pb [10], for example.

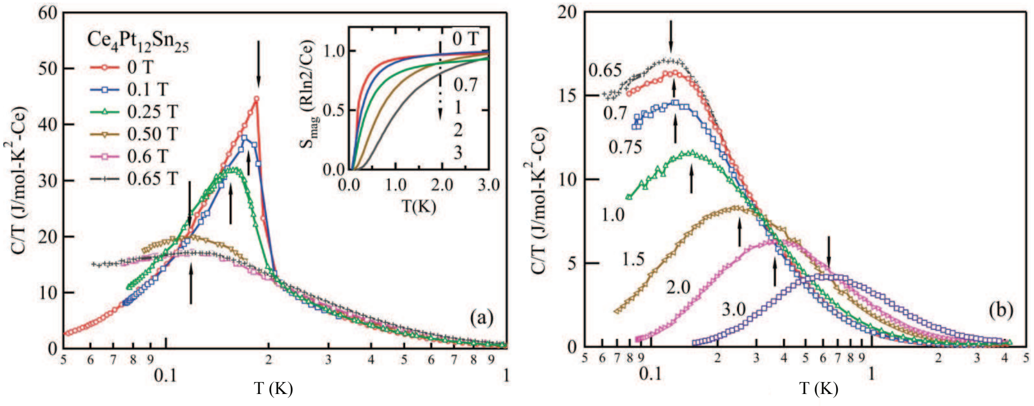

Figure 2. (Color online) Temperature dependence of C/T of $Ce_4Pt_{12}Sn_{25}$ in the magnetic fields, (a) 0-0.65 T, (b) 0.65-3 T. The arrows indicate the temperature where C/T has maximum. The inset (a): $S_{\mathrm{mag}}(T)$ in several fields up to 3 T.

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. The results for zero field specific heat and entropy are displayed in Fig. 1 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 Rln 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 the lower panel of Fig. 1 relative to the Heisenberg model.

Such contribution may still be due to remnant Kondo physics, although it would require a rather low temperature scale $T_K < T_N$. The primary factor responsible for the low ordering temperature is the large distance between Ce ions of $Ce_4Pt_{12}Sn_{25}$. In spite of that, RKKY interaction still dominates the Kondo interactions, and the system orders magnetically below $T_N = 0.18$ K. $Ce_4Pt_{12}Sn_{25}$ is therefore in the low $J < J_c$ limit of the Doniach phase diagram. Note that Ce-based skutterudite compounds and YbT_2Zn_{20}, whose Ce-Ce and Yb-Yb distances are similar to that in $Ce_4Pt_{12}Sn_{25}$, do not show magnetic ordering, and are in the $J > J_c$ 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_{max}, where C/T has its maximum value, gradually shifts to lower temperature, as indicated by arrows, reflecting suppression of the AFM ordering with field. The feature associated with the AFM order is suppressed entirely around 0.7 T. With further increase of magnetic fields, T_{max} shifts to higher temperature, while the peak hight is continuously suppressed, as shown in Fig. 2(b). This is due the to increase of magnetic field Zeeman splitting of the crystal electric field (CEF) doublet ground state. As seen in the inset of Fig. 2(a), S_{mag} reaches 1.0 Rln2 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

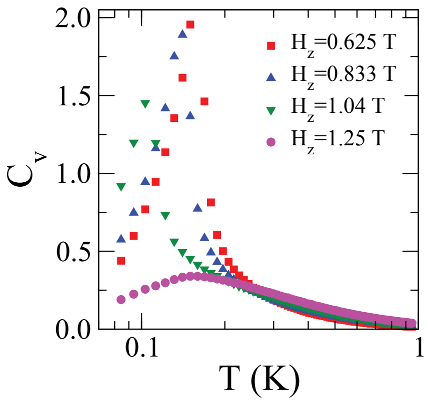

Figure 3. (Color online) Numerical results for C/T (in arbitrary units) for the spin-1/2 Heisenberg model as described in the text in the high field region.

doublet.

4. Theoretical modeling in field

Some of the results of our numerical calculations with magnetic field included are shown in Fig 3. At low fields (not shown) the anomaly is very robust, and does not change up to $H = 0.4$ T. At higher field the discrepancy between the experimental data and numerical modeling is even greater, since experimentally the AFM anomaly is suppressed completely for $H \approx 0.5T$, whereas theoretical calculations reveal a strong anomaly even at $H = 1.04$ T. This is an important difference, and can be due to a variety of factors absent in the model, including next nearest neighbors interactions, frustration, remaining Kondo physics, or RKKY coupling strength changing in magnetic field, and is an important issue to address in the future investigations of RKKY-Kondo competition.

5. Conclusion

In conclusion, we have performed low-temperature field-dependent specific heat to elucidate the ground state properties of $Ce_4Pt_{12}Sn_{25}$. T_N is suppressed with an initial increase of field up to 0.6 T, whereas T_{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 Γ_7 doublet. Therefore, the ground state of $Ce_4Pt_{12}Sn_{25}$ changes above 0.6 T from AFM to paramagnetic.

The model calculations, based on a CEF scheme with a Γ_7 ground state, reproduce rather well experimental high field specific heat and magnetization data. Lower field data is described very well by the numerical calculation based on the Heisenberg model. Small deviation between experimental data and numerical results can be ascribed to 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 $Ce_4Pt_{12}Sn_{25}$ in the $J \ll J_c$ of the Doniach phase diagram. The Kondo temperature T_K is 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. $Ce_4Pt_{12}Sn_{25}$ therefore presents us with a *counter example* to an expectation that dilute f-electron compounds will likely fall into the Kondo screened regime, and therefore should be good candidates to study Kondo physics. Weak $c - f$ hybridization in $Ce_4Pt_{12}Sn_{25}$ may be due to large distances between Ce and surrounding Sn and Pt atoms [9].

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