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Observation of field-induced single impurity behavior in the heavy fermion compound $Ce_3Co_4Sn_{13}$

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Abstract

We have performed heat capacity measurements in magnetic fields to 90 kOe on single crystals of the cubic heavy fermion compound $Ce_3Co_4Sn_{13}$. In zero field, there are no signs of long-range magnetic order down to 0.35 K. However, C/T increases rapidly below 2 K, reaching a very large maximum value of ~4 J/mol Ce–K around 0.8 K in zero field, and the high-field magnetic entropy approaches $R\ln 2$ at 20 K. Above 25 kOe, the data are consistent with a Kondo impurity with $T_K = 1.2$ K. Short-range magnetic correlations are suppressed by magnetic fields giving way to single impurity behavior above 25 kOe.

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Keywords: Heavy fermion; Heat capacity; Quantum critical point

The ternary Ce stannides $Ce_3M_4Sn_{13}$ (M = Ir, Co, or Rh) are known to crystallize in the cubic $Yb_3M_4Sn_{13}$ structure, space group Pm-3n with 40 atoms per unit cell [1–3]. The Ir compound displays heavy fermion behavior and two magnetic transitions at 2.1 and 0.6 K [1,2]. Previous measurements on the Co compound show heavy fermion behavior with no magnetic ordering down to 1.8 K [2]. To further investigate the heavy fermion state of $Ce_3Co_4Sn_{13}$, we have performed powder neutron diffraction at 0.8 K and specific heat measurements down to 0.35 K and in magnetic fields upto 90 kOe. The neutron diffraction results show no evidence of long-range magnetic order. The zero field heat capacity shows no obvious sharp features typical of long-range order but cannot rule out the possibility of long-range order. However, for low fields ($H < 10$ kOe), heavy fermion behavior with very large values of C/T (~4 J/mol K^2 in zero field) is observed. Above 25 kOe, the heat capacity data are fit quite well to what is expected for the $S = \frac{1}{2}$ single impurity model [4]. The data are consistent with short-range order that is suppressed by applied magnetic field, and a crossover above 25 kOe to single impurity behavior.

Single crystals of $Ce_3Co_4Sn_{13}$ and $La_3Co_4Sn_{13}$ were grown in a Sn-flux as described elsewhere [2]. X-ray diffraction measurements confirm the cubic crystal structure with a lattice constant of 9.590 Å for $Ce_3Co_4Sn_{13}$. The non-magnetic La compound was used to measure the lattice contribution to the heat capacity. This data were subtracted from the measured heat capacity of the Ce compound to give the magnetic heat capacity C_{mag}. Quasiadiabatic heat capacity measurements in applied magnetic fields up to 90 kOe were performed in a Quantum Design PPMS.

The results are shown in Fig. 1. In the top panel of Fig. 1, a large increase in C_{mag}/T is seen at low temperatures. In fact the maximum value of 4 J/mol K^2 is very large, being close to the largest report value of 5.5 J/mol K^2 in tetragonal $CeNi_9Ge_4$ [5]. This would seem to indicate that $Ce_3Co_4Sn_{13}$ is likely very near a magnetic

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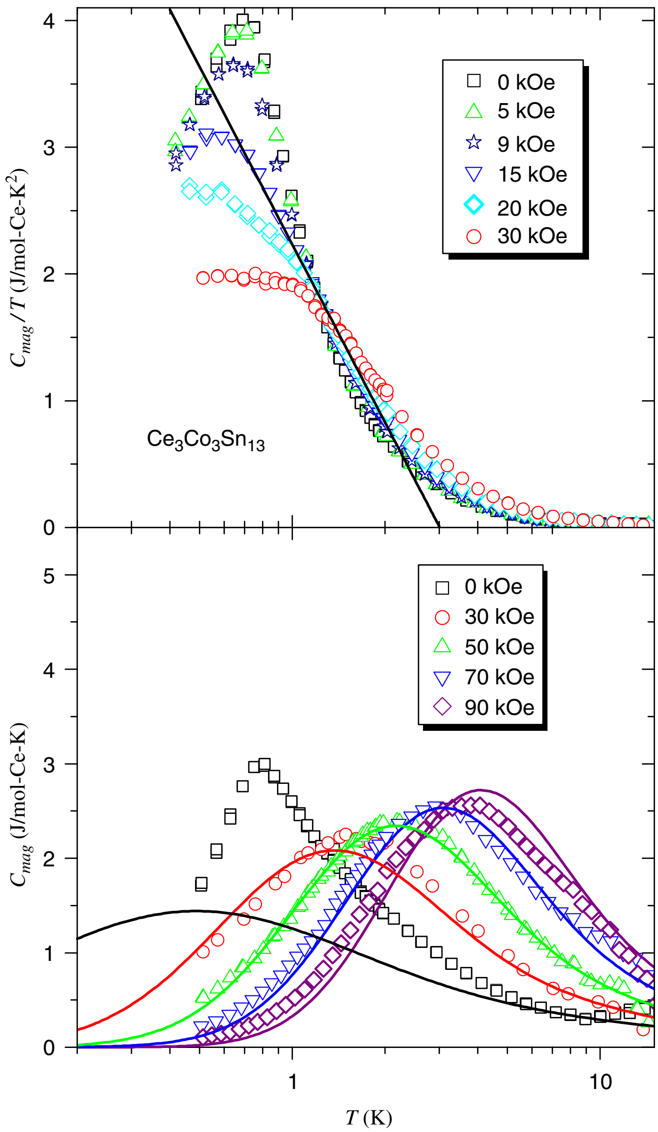

Fig. 1. Magnetic heat capacity data C_{mag} in various applied DC fields. (Top) C_{mag}/T versus T in low fields. The straight line is a logarithmic divergence of the form $\log(T_0/T)$. (Bottom) C_{mag} versus T in various fields. The solid lines are calculated values using the single impurity model with $T_K = 1.2\,K$ as described in the text.

quantum critical point (QCP). For the measurements above 30 kOe, nearly all of $R\ln 2$ of entropy is recovered by 20 K. For lower fields, slightly less than $R\ln 2$ is measured. However, we believe that if data were taken to lower temperatures, all of the $R\ln 2$ entropy would be recovered. In zero field, there are no signs of long-range magnetic order, however, there is likely short-range magnetic order as discussed below. As field is increased, the peak in C_{mag}/T moves to lower temperatures, with no clear maximum observed for fields above approximately 25 kOe.

In applied fields, no distinctive region of non-Fermi liquid (NFL) behavior with $\log T$ behavior is observed. The solid line in the top panel of Fig. 1 shows typical NFL logarithmic behavior, and the largest temperature region (less than half a decade) of logarithmic behavior occurs at 15 kOe. Again, this behavior seems reminiscent of $CeNi_9Ge_4$ where La substitution is required to see large regions of NFL behavior [5]. As field is increased further, single impurity behavior is observed as discussed below.

The bottom panel in Fig. 1 shows the magnetic field-dependent heat capacity for $H \geqslant 30$ kOe along with the zero field data. The solid lines are calculations using the $S = \frac{1}{2}$ single channel single impurity model with $T_K = 1.2\,K$ being the only adjustable parameter [4]. For all of the high field data shown, the fits are quite good. For zero field, the fit is quite poor as short-range magnetic correlations lead to a large increase in C_{mag}/T relative to the single impurity calculations. However, a satisfactory fit (not shown) to the zero field data is achieved assuming half of the entropy is due to single impurity effects and half is due to a Schottky like contribution that has been seen in other Ce compounds displaying short-range magnetic order [6].

In summary, we find a cross-over from a magnetically correlated state with an extremely large value of $C_{mag}/T = 4\,J/mol\,K^2$ to a single impurity state in applied magnetic fields of 20–25 kOe in the heavy fermion system $Ce_3Co_4Sn_{13}$. In between these two regions, no clear NFL behavior is observed. To further study the interesting properties, including the magnetic state in zero field, low-temperature magnetic and transport measurements along with lower temperature neutron scattering experiments are planned. Also, La dilution studies are in progress to study the closeness of the system to a magnetic QCP.

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