

Neutron diffraction study of magnetic field induced behavior in the heavy Fermion $\text{Ce}_3\text{Co}_4\text{Sn}_{13}$

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Abstract

The specific heat of $\text{Ce}_3\text{Co}_4\text{Sn}_{13}$ exhibits a crossover from heavy Fermion behavior with antiferromagnetic correlations at low field to single impurity Kondo behavior above 2 T. We have performed neutron diffraction measurements in magnetic fields up to 6 T on single crystal samples. The (001) position shows a dramatic increase in intensity in field which appears to arise from static polarization of the 4f level and which at 0.14 K also exhibits an anomaly near 2 T reflecting the crossover to single impurity behavior.

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Keywords: Heavy Fermion; Neutron diffraction

$\text{Ce}_3\text{Co}_4\text{Sn}_{13}$ is a heavy Fermion system which resides close to a magnetic instability but does not appear to exhibit the normal hallmarks of a quantum critical point [1–4]. Under ambient conditions, $\text{Ce}_3\text{Co}_4\text{Sn}_{13}$ exhibits a peak in the electronic specific heat coefficient (C/T) at 0.8 K which is too narrow to represent a Kondo peak but is too broad to represent either a Schottky anomaly or long range magnetic order. Moreover, neutron powder diffraction measurements show no sign of long-range magnetic order [5]. Inelastic neutron scattering studies show two crystal field excitations at 8 and 30 meV [6] and the low temperature heat capacity gives $\approx R \ln 2$ by 10 K [3,4] indicating that the peak in C/T represents the behavior of the ground state doublet.

An analysis of the low temperature heat capacity under magnetic field shows that above 2 T, there is a crossover to

behavior characteristic of a collection of Kondo impurities [3]. This suggests that the peak in the low field heat capacity is due to short-range magnetic correlations.

To investigate this idea further, we have performed neutron diffraction on single crystal samples of $\text{Ce}_3\text{Co}_4\text{Sn}_{13}$. Neutron diffraction measurements were performed on the SPINS triple axis spectrometer at the NIST Center for Neutron Research. See Ref. [5] for further experimental and sample preparation details. Data were also collected on the PRISMA instrument at the ISIS spallation source at the Rutherford Appleton Laboratory.

Fig. 1 shows uncorrected longitudinal scans through the (001) position at 4.2 K for a number of magnetic fields applied along the (110) direction. At zero field, only a very weak nuclear intensity is observed at the (001) position. A dramatic increase in intensity is evident starting at low fields. The results of fitting these peaks to a Gaussian line shape and extracting the integrated intensity are shown in Fig. 2(a). The intensity does not saturate below 6 T. Since the (001) position is not allowed in the $\text{Ce}_3\text{Co}_4\text{Sn}_{13}$

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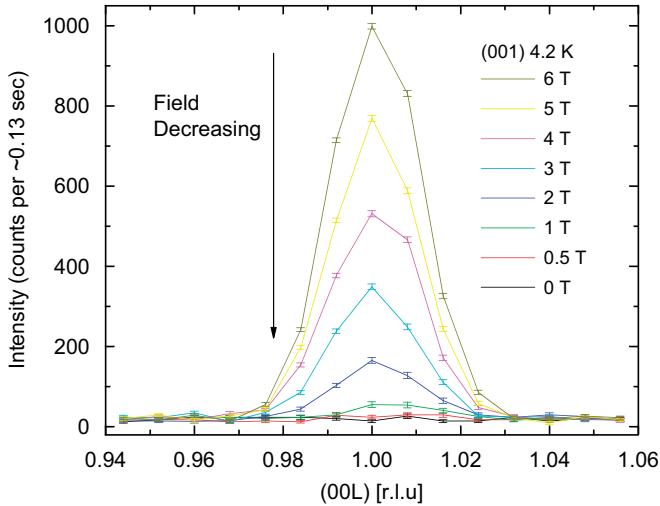


Fig. 1. Longitudinal scan through the (001) position at 4.2 K under applied magnetic fields as indicated. Error bars represent 1S.D.

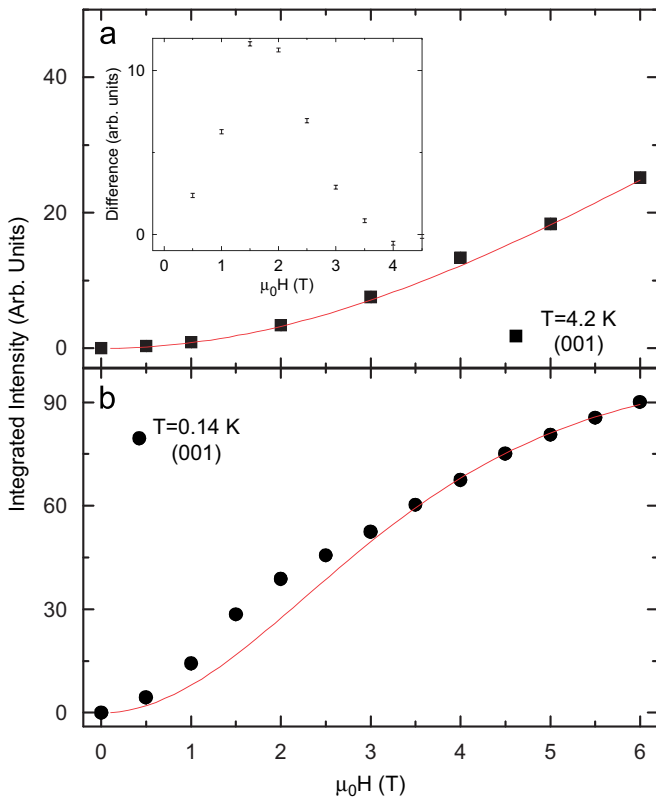


Fig. 2. Integrated intensity (symbols) as a function of magnetic field. The lines are fits as described in the text. The inset in part (a) is described in the text.

structure, the intensity at this \mathbf{Q} means that the applied field induces an antiferromagnetic alignment in the unit cell, with the peak area proportional to the square of the staggered magnetization.

We have also studied the intensity as a function of magnetic field at lower temperature (Fig. 2(b)). Several features are worth noting: (1) The increase in intensity has become larger in magnitude; at 6 T the increase is about 3.5 times larger than observed at 4.2 K. (2) Initially, there is a sharper rise in intensity after which a more gradual rise occurs which shows a tendency to saturate. The data above 4 T at 0.14 K and the entire field range at 4.2 K have been fit (lines in Fig. 2) with the square of a modified $J = 1/2$ Brillouin function with $g_{\text{eff}} = 1.59$ and the temperature, T , replaced by $T + T_K$ with $T_K = 1.7$ K, where T_K is the Kondo temperature. The value of T_K obtained here is similar to the values obtained from specific heat [3] and inelastic neutron scattering measurements [5]. In addition, these parameters are able to reproduce the magnetization data of Ref. [4] at 2 K with small deviations appearing above 4 T (not shown). This indicates that the antiferromagnetic alignment responsible for the (001) intensity has the same field dependence as the magnitude of the moment induced by the field and that the polarization of the 4f electrons occurs under the influence of Kondo spin fluctuations as is observed in the high field specific heat [3]. Subtracting the fit extrapolated to low fields from the data at 0.14 K yields the inset in Fig. 2(a). The excess intensity peaks at a similar field to the 2 T field scale observed in the specific heat and hence has a similar origin, reflecting the crossover from short-range order to single impurity Kondo behavior. Further experiments are planned to elucidate the nature of the correlations as well as to determine why under ambient conditions, the correlations are not strong enough to promote long-range order.

Work at Oak Ridge, UC Irvine and Los Alamos was supported by the Department of Energy. This work utilized facilities supported in part by the NSF agreement no. DMR-0454672.

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