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Magnetic order in the induced magnetic moment system Pr_3In

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Abstract

Pr_3In is a single ground state compound which exhibits antiferromagnetic order below 11.4 K due to the exchange induced admixture of crystalline electric field levels. Additional information regarding the complex magnetic behavior of this compound can be gained through application of magnetic fields. We report specific heat and magnetocaloric effect measurements to 15 T and magnetization measurements to 44 T on single crystal samples of Pr_3In. A new magnetic phase is revealed above 1.9 T and below 11.4 K.

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Keywords: Induced moment behavior; Specific heat

Pr_3Tl is one of the classic examples of induced moment magnetism in singlet–triplet systems exhibiting ferromagnetic order [1,2]. Recently, we have begun to study the related system Pr_3In [3] which showed similar behavior [4] to Pr_3Tl: both compounds have identical crystal structures (Cu_3Au) and similar lattice constants, which suggest similar crystal electric field (CEF) level splittings and exchange interactions. Both materials order magnetically at similar temperatures. Pr_3Tl shows ferromagnetic order below $T_C = 11.6\,K$ whereas Pr_3In shows antiferromagnetic (AF) order below $T_N = 11.4\,K$ [3]. In these systems, the exchange interaction between Pr sites causes admixture of the crystal electric field levels, resulting in the induced moment magnetic order below T_N. Application of a magnetic field (B) can change the energies of the CEF levels in such a manner as to alter the admixture, and provide important information about the induced moment physics present in these systems.

Specific heat (C_p) was measured via a thermal relaxation method at constant B on single crystals grown by the Bridgman technique. The magnetic contribution C_{magn} was obtained after subtracting the phonon contribution, the linear coefficient γ and a low-temperature nuclear hyperfine contribution. The phonon contribution was determined from measurements of C_p of the nonmagnetic analog compound La_3In.

Fig. 1 shows C_{magn}/T vs T for values of B from 0 to 10 T. We include in Fig. 1 the Schottky specific heat calculated for a CEF scheme determined by neutron scattering [5] for temperatures above 100 K: Γ_1 singlet ($E_1 = 0\,meV$), Γ_4 triplet ($E_4 = 6.3\,meV$), Γ_3 doublet ($E_3 = 10.8\,meV$), and Γ_5 triplet ($E_5 = 30\,meV$). At fields $B < 5\,T$, C_{magn}/T increases with T faster than the CEF calculation. This is consistent with neutron scattering at low temperature [5] where the energy difference between the singlet and the lowest multiplet levels of the CEF scheme is dispersive and hence smaller at low momentum transfer Q in the induced moment phase. On the other hand, as B is raised above 5 T, C_{magn}/T increases more slowly with temperature than the CEF calculation, indicating a counterintuitive increase of the CEF splittings

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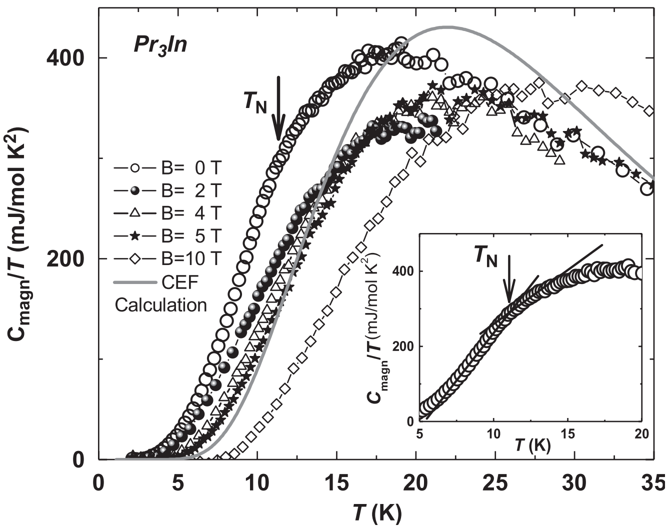

Fig. 1. Magnetic contribution to the specific heat of Pr_3In for magnetic fields $0<B<10\,T$. The line is the predicted Schottky anomaly based on the measured high temperature crystal field energies. The inset shows the anomaly at T_N.

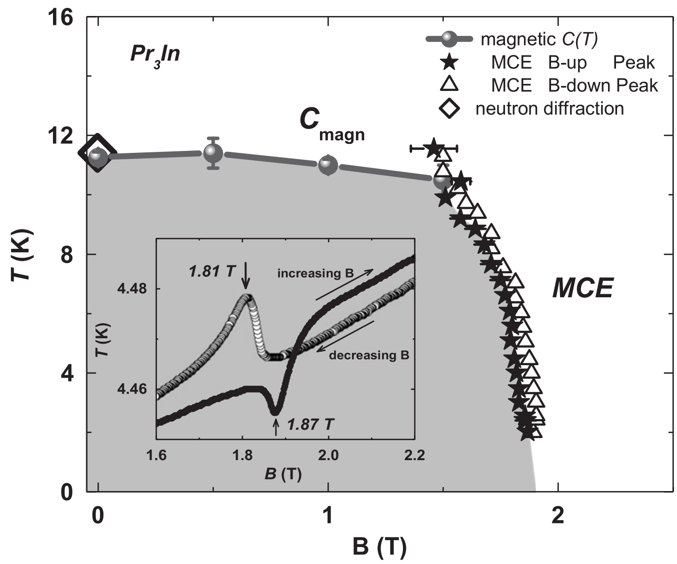

Fig. 3. The (B,T) phase diagram for Pr_3In determined from the specific heat and the magnetocaloric effect (MCE). The inset shows how the phase transition temperature is determined from a typical MCE scan.

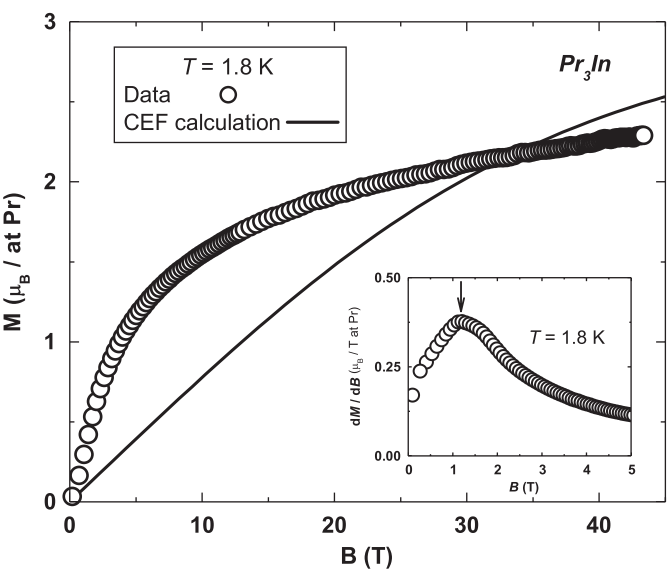

Fig. 2. The magnetization of Pr_3In compared to the value expected for the high temperature crystal field scheme. The inset shows the anomaly in $\mathrm{d}M/\mathrm{d}B$ at 1.5 T.

with B, contrary to the expected behavior of induced moment systems.

The low temperature magnetization (M) data are shown in Fig. 2 as a function of the applied field. Saturation is observed for fields between 20 and 40 T. A calculation of M based on the high temperature CEF scheme is also included in the plot. The magnetization rises more rapidly with field than the CEF calculation, again due to the reduction of the excitation energy in the induced moment phase.

There is a very small change in slope in C_{magn}/T at $T=T_N$, as shown in the inset Fig. 1. The corresponding change in entropy ΔS is also very small. As B is increased, the temperature of the anomaly decreases; this feature is no longer noticeable above 2 T in the specific heat measurement. In Fig. 3 we trace the phase diagram in the (B,T) plane with the points marked "C_{magn}" taken from the specific heat anomalies. These values are consistent with the zero field value of T_N coming from neutron diffraction [3].

To complete the phase boundary, we measured the magneto-caloric effect (MCE), which is convenient for studying B-dependent phase boundaries [6]. A phase boundary near 1.9 T is observed with MCE. The inset of Fig. 2 shows that there is also an anomaly in the first derivative $\mathrm{d}M/\mathrm{d}B$ of the magnetization between 1 and 1.5 T. We did not observe any other phase transition in fields up to 15 T with MCE. The nature of the phase above 2 T is an open question: Is it a fully polarized state or some other spin rearrangement e.g. a spin flop? Given that $T_N = 11\,K$ it would be unusual for the transition to be to a spin polarized state to occur at a field as small as 1.8 T. Neutron diffraction experiments under applied fields are planned to resolve this issue.

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