Journal of Magnetism and Magnetic Materials 226–230 (2001) 145–147

Journal of magnetism and magnetic materials

www.elsevier.com/locate/jmmm

Magnetic properties of the dense Kondo system CeB_6 in high magnetic fields

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Abstract

This work reports on the phase diagram study of the dense Kondo system CeB_6 by means of magnetization (M) and transverse magnetoresistance (MR) measurements in magnetic fields (B) to 18 T, in the temperature (T) range from 1.8 to 40 K. The zero field resistivity displays Kondo behavior in the paramagnetic P-phase, while two discontinuities at low T were observed at $T_Q \approx 3.5$ K and $T_N \approx 2.3$ K, the boundaries of the antiferro-quadrupolar (AFQ), and antiferromagnetic (AF) phases, respectively. The isothermal transverse MR is large and negative at low T, and it shows a distinct feature when the P–AFQ line of the B–T phase diagram is crossed. Similarly, the M vs. B curves at low temperatures show upward discontinuities at the phase boundary. Both the features in MR and M at the P–AFQ line correlate well in T, revealing the gradual increase of the T for the onset of AFQ ordering with B. No evidence for reentrant behavior could be observed in fields to 18 T. © 2001 Published by Elsevier Science B.V.

Keywords: Kondo systems; Quadrupole ordering; Magnetic phase diagram

The dense Kondo compound CeB_6 crystallizes in a CsCl structure in which B_6 octahedra occupy the Cl site. The competition between intra-site Kondo and inter-site RKKY interactions leads this compound to display a remarkably complex magnetic phase diagram at low temperatures (T) [1,2]. In zero magnetic field (B) CeB_6 is paramagnetic (P-phase) down to 3.2 K. At $T_Q = 3.2$ K it undergoes a transformation to an antiferro-quadrupolar AFQ-phase, which is stable down to 2.3 K, undergoing then another transformation into an AF-phase at $T_N = 2.3$ K. There are two lines in the T-B phase diagram, one starting at T_Q which separates the AFQ- and P-phases, and one starting at T_N separating the AF- and AFQ-phases. The T of the AF–AFQ phase transition decreases with B, while the T of the AFQ–P transition increases with B. Raman and neutron scattering measurements suggest that the cubic symmetry crystal field splits the Ce^{3+} multiplet into a Γ_8 quartet (groundstate) and a Γ_7 doublet, and that the difference in energy between these two levels is 46 meV [3]. A calculation based on the inter-site quadrupole–quadrupole interaction and a Zeeman term suggests that the AFQ–P line should be reentrant, i.e., $dT_{P\text{-}AFQ}/dB$ should change from positive in low B to negative in higher B, until the AFQ phase is completely suppressed [4]. The magnetic structure below T_N corresponds to a double-k commensurate structure. For $B > 0$ several magnetic substructures are formed in the AF-phase [5]. The increase of $T_{AFQ\text{-}P}$ with B is quite counterintuitive as one could expect a tendency of B to align the spins in a common direction, which should cause $dT_{AFQ\text{-}P}/dB$ to be negative. The structure of the AFQ ordered phase has been studied using a number of microscopic techniques (NMR [6], neutron diffraction [5], and μSR[7]) but it has not been unequivocally determined. A number of theoretical approaches has been utilized in order to interpret both the

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0304-8853/01/$ - see front matter © 2001 Published by Elsevier Science B.V.
PII: S0304-8853(01)00117-2

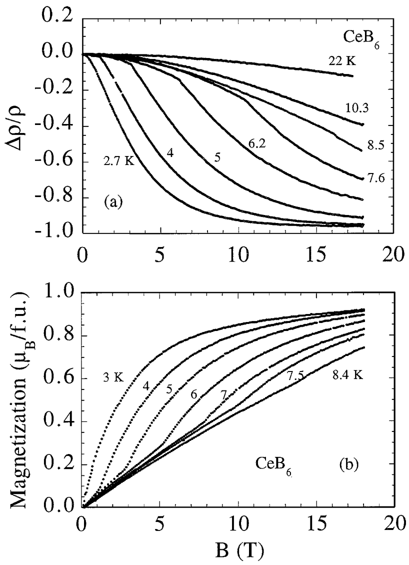

Fig. 1. (a) Isothermal transverse magnetoresistance vs. B for CeB_6 in the T-range $2.7\,K \leqslant T \leqslant 22\,K$; (b) Magnetization curves for $3.0\,K \leqslant T \leqslant 8.4\,K$ (B//001).

AFQ–P and the AF–AFQ lines of the B–T phase diagram (for a discussion see, for example, Ref. [8]). The goal of this work was to perform measurements of magnetoresistance MR and magnetization M in order to determine the shape of the P–AFQ line in fields up to 18 T, to determine the effect that crossing the P–AFQ line has on the transport and magnetic properties, and to test for the possible reentrant behavior.

The CeB_6 single crystals for this study were grown in Al-flux. The measurements of magnetization above 2 K in B to 18 T (B//001) were performed with a vibrating sample magnetometer operating in the B provided by a superconducting magnet at the NHMFL. The measurements of transverse MR for $T > 2\,K$ (B//001) were performed in the same magnet. Both apparatuses were equipped with a gas flow cryostat. The zero field ρ vs. T data below 300 K (not shown) show a typical Kondo-like behavior, in which the rise in ρ vs. T below 50 K follows a logarithmic function. There is a sharp drop in ρ vs. T below $T_{AFQ} \approx 3.5\,K$, followed by another sharp drop below $T_N \approx 2.3\,K$. Isothermal curves of $\Delta\rho/\rho$ vs. B in the 2.7–22 K interval are displayed in Fig. 1a. The MR in this T–range is very large and negative, reflecting the suppression of spin disorder both in the P- and AFQ-phases. The $\Delta\rho/\rho$ vs. B curves for $2.7\,K \leqslant T \leqslant 8.5\,K$ show sharp downward discontinuities as B crosses the AFQ–P line into the AFQ-phase. These drops can be due both to suppression of spin-disorder scattering, as well as to spin realignment of the AFQ structure. The M vs. B magnetization curves for $3\,K \leqslant T \leqslant 8.4\,K$ are displayed in Fig. 1b. These curves show upward discontinuities as B crosses the AFQ–P line, suggesting the occurrence of spin realignment in the AFQ-phase, which is consistent with the neutron diffraction observation that B induces an AF component in the AFQ-phase [5]. The discontinuities in $\Delta\rho/\rho$ vs. B and M vs. B correlate well in temperature, and they can be taken as belonging to the AFQ–P line, as shown in Fig. 2. The value of $B(T)$ on the AFQ–P line follows closely a T^3 law, but it does not show any signs of re-entrant behavior up to 18 T.

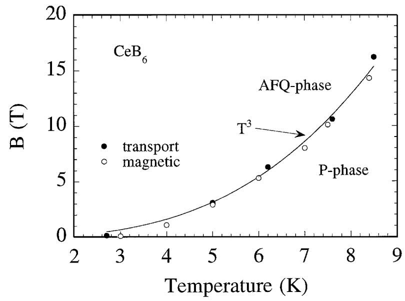

Fig. 2. The boundary between the paramagnetic and antiferroquadrupolar phases of CeB_6, as established by magnetotransport and magnetization measurements. The solid line is a heuristic T^3 function.

In summary, the negative values of MR in $B \leqslant 18\,T$ suggest that B suppresses spin-flip scattering in the P-phase, and possibly induces spin realignment in the AFQ-phase. The S-shaped features of the M vs. B curves suggest that the AFQ–P transition is consistent with the occurrence some spin realignment in the AFQ-phase. The features in $\Delta\rho/\rho$ vs. B and M vs. B at the AFQ–P line become more washed out at higher T, possibly due to thermal disorder. No reentrant behavior for the AFQ–P line could be observed for $B \leqslant 18\,T$.

The support of NSF Grant No. INT-9725929 (MST, AHL) is gratefully acknowledged. Work at the NHMFL was performed under the auspices of the NSF, the state of Florida, and the USDOE.

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