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2003 J. Phys.: Condens. Matter 15 S2159

(http://iopscience.iop.org/0953-8984/15/28/342)

INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER

J. Phys.: Condens. Matter **15** (2003) S2159–S2162 PII: S0953-8984(03)62611-1

The field-induced magnetic phase in a heavy-fermion antiferromagnet, Ce_7Ni_3

K Umeo[1], **Y Echizen**[1], **T Takabatake**[1], **T Sakakibara**[2], **T Terashima**[3] **and C Terakura**[3]

[1] ADSM, Hiroshima University, Higashi-Hiroshima 739-8530, Japan
[2] ISSP, University of Tokyo, Kashiwa, Chiba 277-8581, Japan
[3] NIMS, Tsukuba, Ibaragi 305-0003, Japan

Received 12 November 2002
Published 4 July 2003

Abstract

In the heavy-fermion antiferromagnet Ce_7Ni_3 with three nonequivalent Ce sites (Ce_I, Ce_{II}, Ce_{III}), a field-induced magnetic (FIM) phase has been found in the field region above 0.7 T with the field applied along the hexagonal *c*-axis. Magnetoresistance, specific heat, and magnetization measurements showed that the FIM phase is separated from the spin-density-wave phase present below 0.3 T. The separation is attributed to large spin fluctuations on Ce_{III}, which originate from a geometrical frustration in the quasiregular tetrahedron made of Ce_I and Ce_{III}.

1. Introduction

The geometrical frustration in insulating magnets as well as itinerant ones leads to interesting physics such as novel field-induced magnetic (FIM) phases [1]. However, there have been few studies on geometrically frustrated cerium (Ce) compounds. In CePdAl with the quasi-Kagome lattice of Ce atoms, one third of Ce ions remain paramagnetic in the antiferromagnetically ordered state below $T_N = 2.7$ K [2].

Another possible frustrated Ce compound is Ce_7Ni_3, which crystallizes in the hexagonal Th_7Fe_3-type structure having three nonequivalent Ce sites: $1Ce_I$, $3Ce_{II}$, and $3Ce_{III}$ [3]. As shown in the inset of figure 1, the Ce_I and Ce_{III} atoms form a quasiregular tetrahedron. These tetrahedra are stacked in chains along the *c*-axis, which resemble the frustrated arrangement of Mn atoms in RMn_2 (R = rare earth) crystallizing in the hexagonal C14 Laves structure [4]. Ce_7Ni_3 undergoes two magnetic transitions at $T_{N1} = 1.9$ K and $T_{N2} = 0.7$ K [5–7]. A neutron diffraction study showed that the magnetic structure below T_{N1} is a spin-density wave (SDW). The ordered moments are estimated to be 0.46, 0.70, and 0.10 μ_B for Ce_I, Ce_{II}, and Ce_{III}, respectively [7].

The magnetically ordered state of Ce_7Ni_3 is very sensitive to pressure and magnetic field. With application of pressure, both T_{N1} and T_{N2} decrease and finally vanish at $P_c = 0.39$ GPa,

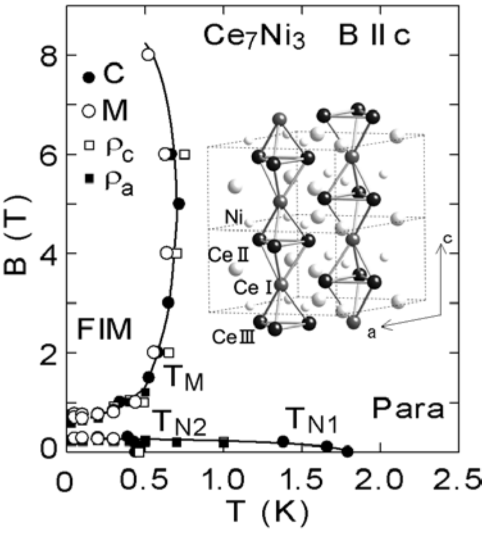

Figure 1. The field–temperature phase diagram of Ce_7Ni_3 for $B \parallel c$. The solid curves are a guide to the eye. The inset shows a schematic drawing of the crystal structure of Ce_7Ni_3. One Ce_I and three Ce_{III} atoms form a quasiregular tetrahedron, which stacks along the c-axis.

and non-Fermi liquid behaviour appears [8, 9]. Upon applying a rather weak magnetic field of 0.2 T along the c-axis, the easy magnetization axis, the SDW state collapses, and a field-induced ferromagnetic state is stabilized [7]. In order to study the ground state of Ce_7Ni_3 under magnetic fields, we have measured the field and temperature dependences of the specific heat C, magnetization M, and electrical resistivity ρ for a single-crystalline sample.

2. Results and discussion

We summarize the magnetic field (B)–temperature (T) phase diagram of Ce_7Ni_3 for $B \parallel c$ in figure 1. Both T_{N1} and T_{N2} vanish at 0.3 T. Upon increasing the field above 0.7 T, a FIM phase appears. Because this phase is separated from the SDW phase present below 0.3 T, it is unlikely to be a spin-flop phase in a conventional antiferromagnet. When the field is applied along the a-axis, T_{N1} is unchanged up to 8 T.

The panels on the left of figure 2 show the low-temperature data for C/T and M at various constant fields, $B \parallel c$. The peaks in C/T at T_{N1} vanish for $B \gtrsim 0.5$ T. Above 1 T, another peak in C/T and a small kink in $M(T)$ appear at around 0.5 K, which suggest a FIM transition. By integrating the C/T data at 2 T, we estimate the entropy released at the FIM transition temperature T_M to be 0.53 J K^{-1}/mol Ce $\cong 0.1 R \ln 2$. This small entropy may imply that the FIM transition is associated with the ordering of Ce_{III} with the small moment of 0.1 μ_B.

The magnetoresistance $\Delta\rho/\rho_0 = [\rho(B) - \rho(0)]/\rho(0)$ and M as a function of $B \parallel c$ are displayed in the panels on the right of figure 2. The sharp anomaly in $\Delta\rho/\rho_0$ at $B_M = 0.2$ T is associated with the metamagnetic transition. On cooling below 0.4 K, the FIM transition manifests itself in the decrease of $\Delta\rho/\rho_0$ and weak increase of M at around 0.7 T. The latter is more clearly seen as a maximum in $\Delta M = M(0.05\text{ K}) - M(0.45\text{ K})$ in figure 2(e). The small jump of $\Delta M = 0.004\ \mu_B$ supports the conjecture that the Ce_{III} spins having the smallest moment partially align along the c-axis.

We now discuss the origin of the separation of the FIM phase from the SDW phase in the phase diagram shown in figure 1. To our knowledge, a FIM phase appearing from

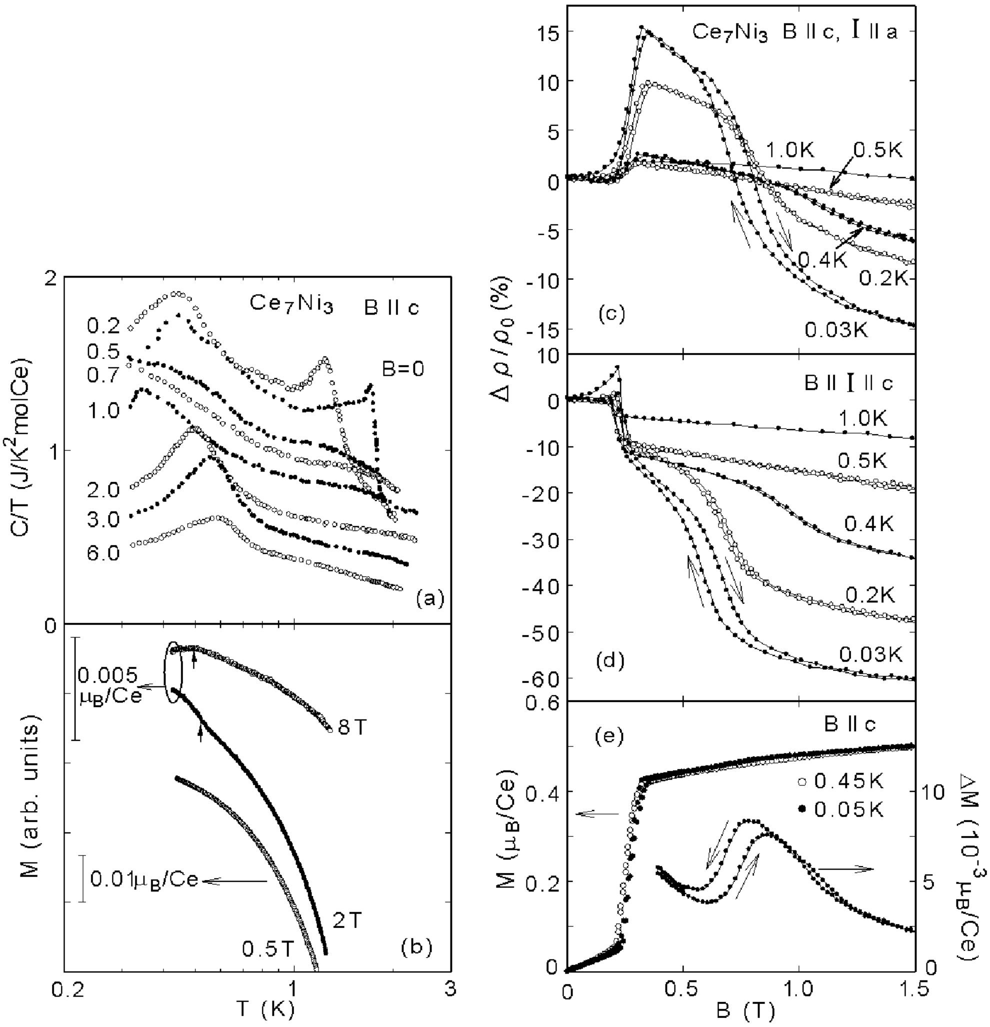

Figure 2. Left panels: the temperature dependence of the specific heat divided by the temperature C/T and the magnetization M of Ce$_7$Ni$_3$ under magnetic fields $B \parallel c$. Right panels: the field ($B \parallel c$) dependence of $\Delta\rho/\rho_0 = [\rho(B) - \rho(0)]/\rho(0)$ and the magnetization M of Ce$_7$Ni$_3$. The inset shows the difference $\Delta M = M(0.05\ \mathrm{K}) - M(0.45\ \mathrm{K})$.

a paramagnetic phase has been observed in four distinct systems: (a) spin-singlet ground state systems, e.g., CsFeCl$_3$ [10]; (b) the Ce-based superconductor CeCu$_2$Si$_2$ located in the vicinity of a quantum critical point [11]; (c) quasi-two-dimensional organic conductors such as (TMTSF)$_2$ClO$_4$ [12]; and (d) geometrically frustrated magnets such as Gd$_3$Ga$_5$O$_{12}$ [13]. We consider the applicability of each scenario to Ce$_7$Ni$_3$. First, the singlet ground state system, which possesses a low-lying excited multiplet, can order magnetically at the level-crossing field if the exchange interaction is larger than the Zeeman energy. This scenario cannot be applied to Ce$_7$Ni$_3$ because the crystal-field ground state is a doublet according to the specific heat data [6]. Second, the origin of the FIM phase in CeCu$_2$Si$_2$ is still vague to the best of our knowledge, and therefore we do not consider it further. Third, for the quasi-two-dimensional system, the field-induced SDW (FISDW) occurs when the Fermi surface nesting is incomplete. If FISDW occurs, the magnetoresistance should become positive due to gap formation at the

Fermi level. For Ce_7Ni_3, however, the magnetoresistance for both $I \parallel a$ and $I \parallel c$ is negative at the FIM transition, as is shown in figures 2(c) and (d).

Fourth, for the geometrically frustrated system, a lack of magnetic order at low fields is caused by the strong spin fluctuations due to a geometrical frustration. Application of a magnetic field quenches the frustration, and leads to a transition to a magnetically ordered state. This scenario is possible for the FIM transition in Ce_7Ni_3. On using the atomic positions of Ce_7Ni_3 determined at 7 K by powder neutron diffraction [7], the average distances between one Ce atom and neighbouring atoms in the coordination polyhedra defined by the Brunner–Schwarzenbach method [14] are 3.58, 3.58, and 3.54 Å, respectively, for Ce_{I}, Ce_{II}, and Ce_{III}. These nearly equal distances for the three sites suggest similar hybridization strengths and thus similar sizes of magnetic moments for the three sites. Therefore, the strongest reduction of the ordered moment for Ce_{III} may result from not only the Kondo effect but also a sort of frustration effect. For Ce_7Ni_3, the geometrical frustration is expected in the quasiregular tetrahedron formed by Ce_{I} and Ce_{III} as shown in the inset of figure 1. The frustration effect on Ce_{III} should be larger than that on Ce_{I}, because the three Ce_{III} atoms in the tetrahedron form a regular triangle. The spin fluctuation due to the frustration prevents the magnetic moment of Ce_{III} from ordering in the field range $0.3 < B < 0.7$ T. Beyond 0.7 T, however, Ce_{III} moments may be partially aligned along the c-axis by the exchange fields produced by the field-induced ferromagnetic moments of Ce_{I} and Ce_{II}.

3. Summary

We have found a FIM phase of the heavy-fermion antiferromagnet Ce_7Ni_3 for $B \parallel c > 0.7$ T and $T < 0.5$ K, which is separated from the SDW phase in the region $B \parallel c < 0.3$ T. Combination of the results on specific heat and magnetization suggests that the rearrangement of the Ce_{III} moments is responsible for the FIM order. We attribute the separation from the low-field SDW phase to large spin fluctuations on Ce_{III} originating from a geometrical frustration in the quasiregular tetrahedron made of Ce_{I} and Ce_{III} atoms in Ce_7Ni_3.

Acknowledgments

We acknowledge H Kadowaki for helpful discussion. Specific heat measurements were carried out at the Cryogenic Centre, Hiroshima University. This work was supported by COE Research (13CE2002) in a Grant-in-Aid from the MEXT, Japan.

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