Journal of Alloys and Compounds 408–412 (2006) 43–46

Journal of ALLOYS AND COMPOUNDS

www.elsevier.com/locate/jallcom

Pressure- and field-induced magnetic instabilities in a heavy-fermion antiferromagnet Ce_7Ni_3

K. Umeo [a,*], K. Motoya [b], H. Kadowaki [c], N. Aso [d], T. Tayama [e], T. Sakakibara [e],
N. Kurita [e], M. Hedo [e], Y. Uwatoko [e], T. Takeuchi [f], T. Takabatake [a]

[a] Department of Quantum Matter, ADSM, Hiroshima University, Kagamiyama 1-3-1, Higashi-Hiroshima 739-8530, Japan
[b] Faculty of Science and Technology, Tokyo University of Science, Noda 278-8510, Japan
[c] Department of Physics, Tokyo Metropolitan University Hachioji-shi, Tokyo 192-0397, Japan
[d] ISSP, University of Tokyo, Tokai, Ibaraki 319-1106, Japan
[e] ISSP, University of Tokyo, Kashiwa, Chiba 277-8581, Japan
[f] Cryogenic Center, Osaka University Toyonaka, Osaka 560-0043, Japan

---

Abstract

We investigated ground state properties of a heavy-fermion antiferromagnet Ce_7Ni_3 under hydrostatic pressures and magnetic fields. This compound undergoes two antiferromagnetic phase transitions at $T_{N1} = 1.9$ K and $T_{N2} = 0.7$ K. Below T_{N1}, a spin-density-wave (SDW) develops. Upon applying rather weak pressure 0.39 GPa = P_c, both T_{N1} and T_{N2} vanish, and non-Fermi liquid behavior appears in the specific heat and magnetic susceptibility. The enhancement of residual resistivity along the a axis near P_c is attributed to the increased spin fluctuations along the a axis. By applying fields B along the c axis, T_{N1} is suppressed and vanishes at 0.3 T. Magnetoresistance, specific-heat, and magnetization measurements revealed another field-induced magnetic (FIM) phase in the region $B \parallel c > 0.7$ T and $T < 0.5$ K. Neutron diffraction experiments indicate that the magnetic unit cell in the c-plane for the FIM phase is treble that of the chemical unit cell. Moreover, this magnetic reflection intensity remains even in the region between the FIM phase and SDW phase. This observation indicates the presence of large spin fluctuations in the c-plane associated with the magnetic frustration, which should be responsible for the magnetic instability of Ce_7Ni_3.

Keywords: Magnetically ordered materials; Neutron diffraction; Heavy fermions; High pressure; Electronic transport

---

1. Introduction

In recent years, various deviations from conventional Fermi-liquid behavior have been found for some cerium (Ce)- and uranium-based compounds near an antiferromagnetic quantum critical point. The deviations are characterized by anomalous temperature dependencies of the specific heat C, magnetic susceptibility χ, and electrical resistivity ρ; $C/T \propto -\ln T$, $\chi \propto 1 - T^{-0.5}$, and $\rho \propto T$ [1]. The breakdown of the Fermi-liquid behavior can be tuned by alloying or by applying a hydrostatic pressure or a magnetic field [1]. Most interestingly, heavy-fermion superconductivity appears in the vicinity of the pressure-induced magnetic–non-magnetic phase boundary [2].

On the other hand, the interesting physics provided by geometrically frustrated systems is also under active discussion. The geometrical frustration can occur in triangular, kagome, fcc, and pyrochlore lattices where nearest-neighbor interactions compete. The frustration in insulating materials leads to multiple phase transitions and novel field-induced magnetic phases [3]. When the frustration affects an itinerant magnet, unusual physical properties have been observed, e.g., spin-liquid behavior in $Y_{0.97}Sc_{0.03}Mn_2$ (C15 cubic Laves phase) [4]. However, there have been few studies on geometrically frustrated Ce compounds. In CePdAl with the quasikagome lattice of Ce atoms, one-third of Ce ions remain paramagnetic

---

* Corresponding author.
E-mail address: kumeo@sci.hiroshima-u.ac.jp (K. Umeo).

0925-8388/$ – see front matter © 2005 Elsevier B.V. All rights reserved.
doi:10.1016/j.jallcom.2005.04.024

in the antiferromagnetically ordered state below $T_N = 2.7$ K [5,6].

A heavy-fermion antiferromagnet Ce_7Ni_3 possesses above two features. Ce_7Ni_3 crystallizes in the hexagonal Th_7Fe_3-type structure having three non-equivalent Ce sites; 1Ce I, 3Ce II, and 3Ce III. As shown in Fig. 1, the Ce I and Ce III atoms form a hexahedron. The hexahedrons are stacked in chains along the *c* axis, which resemble the arrangement of Mn atoms in geometrically frustrated RMn_2 (R, rare earth) compounds crystallizing in the hexagonal C14 Laves structure [7]. Ce_7Ni_3 undergoes two magnetic transitions at $T_{N1} = 1.9$ K and $T_{N2} = 0.7$ K [8]. A neutron-diffraction study showed that the magnetic structure below T_{N1} is a spin-density wave (SDW) with a modulation vector $k = 0.22c^*$ [9]. With application of pressure, both T_{N1} and T_{N2} decrease and vanish at $P_c = 0.39$ GPa, and non-Fermi liquid behaviors, $C/T \propto -\ln T$, and $\chi \propto (1 - T^{-0.5})$ appear [10,11]. On cooling below 0.5 K, the C/T curve near P_c deviates from $-\ln T$ dependence, and exhibits a broad maximum around 0.15 K [11]. These observations indicate that the $-\ln T$ dependence in C/T between 0.5 and 6 K is a crossover phenomenon to a Fermi-liquid ground state, and is consistent with the result predicted by the self-consistent-renormarization (SCR) theory of spin fluctuations [12]. In order to study the spin fluctuations in Ce_7Ni_3 relevant to the pressure induced non-Fermi liquid behavior, we have measured the electrical resistivity along the *a* and *c* axis, ρ_a and ρ_c, under pressure up to 0.7 GPa at temperatures down to 0.05 K.

The geometrical frustration mentioned above may lead to anomalous magnetic behavior under magnetic fields. In fact, both T_{N1} and T_{N2} vanish above 0.3 T when the magnetic field *B* is applied along the *c* axis, the magnetic easy axis. For $B \parallel c > 0.7$ T, another field-induced magnetic (FIM) phase appears below 0.5 K, which was found by magnetoresistance, specific-heat, and magnetization measurements [13]. The findings of a small entropy gain of $0.1R\ln 2$ and the very small increase of magnetization of $0.005\,\mu_B$ at the phase boundary suggested that the FIM transition is associated with the ordering of the small moment of $0.1\,\mu_B$ on Ce III. The separation of the FIM phase from the SDW phase was attributed to large spin fluctuations originated from a geometrical frustration in the hexahedron made of Ce I and Ce III atoms in Ce_7Ni_3. In aiming at determining the magnetic structure of the FIM phase, we have performed neutron diffraction study on a single crystal sample in a temperature range $0.04 \leq T \leq 1$ K and a field range $0 \leq B \leq 5$ T.

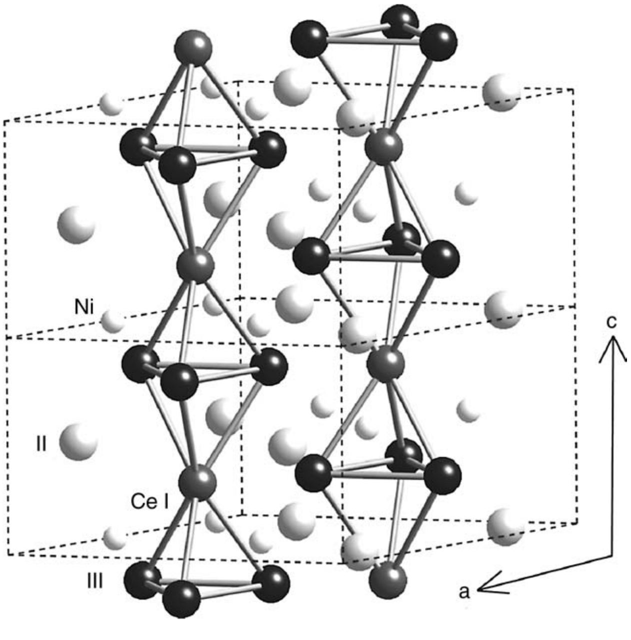

Fig. 1. Crystal structure of Ce_7Ni_3. Two Ce I atoms and three Ce III atoms form a hexahedron stacking along the *c* axis.

2. Experimental procedures

A single crystal of Ce_7Ni_3 was grown by a Czochralsky pulling method using a radio-frequency induction furnace with a hot tungsten crucible. In order to decrease defects, strains, and impurity ions, the as-grown crystal was heat-treated by the technique of the solid-state electrotransport in a high vacuum. The electrical resistivity under pressure at low temperatures ($0 \leq P \leq 0.7$ GPa and $0.05 < T < 300$ K) was measured by a four terminal ac method by using a clamp-type piston–cylinder pressure cell. The pressure was determined by the measurement of the superconducting transition of Pb. Daffne oil was used as a pressure-transmitting medium. The magnetization measurements in the temperature range $0.05 \leq T \leq 1.3$ K were performed by a Faraday method using a high-resolution capacitive magnetometer [14]. Neutron diffraction experiments were performed using triple-axis spectrometer 4G-GPTAS installed at JRR-3 M in JAERI (Tokai). Neutron energy was fixed at 30.5 meV. The sample was cooled down to 0.04 K by a ^{3}He–^{4}He dilution refrigerator, and magnetic fields up to 5 T were applied by a superconducting solenoid.

3. Pressure effect on the resistivity

Fig. 2 displays the temperature dependence of the electrical resistivity along the *a* and *c* axis, ρ_a and ρ_c, under pressures. For $P = 0$, there is very weak anomaly at T_{N1} and a maxima at T_{N2}. At $P = 0.27$ GPa, T_{N1} is recognized as an upturn in ρ_a and a drop in ρ_c, but T_{N2} goes below 0.05 K, the lowest measured temperature. This observation at T_{N1} suggests the formation of an energy gap along the *a* axis due to the SDW with a modulation vector along the *a* axis. It is noteworthy that at 0.27 GPa, the value of $\rho_a(T)$ for $T < 0.1$ K is slightly larger than that at $P = 0$, whereas $\rho_c(T)$ is reduced to the half of the value at $P = 0$. This finding suggests that the spin fluctuation along the *a* axis remains at $0.27\ \text{GPa} < P_c = 0.39$ GPa. The spin fluctuations along the *a* axis may lead to the non-Fermi liquid behavior in *C* and χ as mentioned in Section 1.

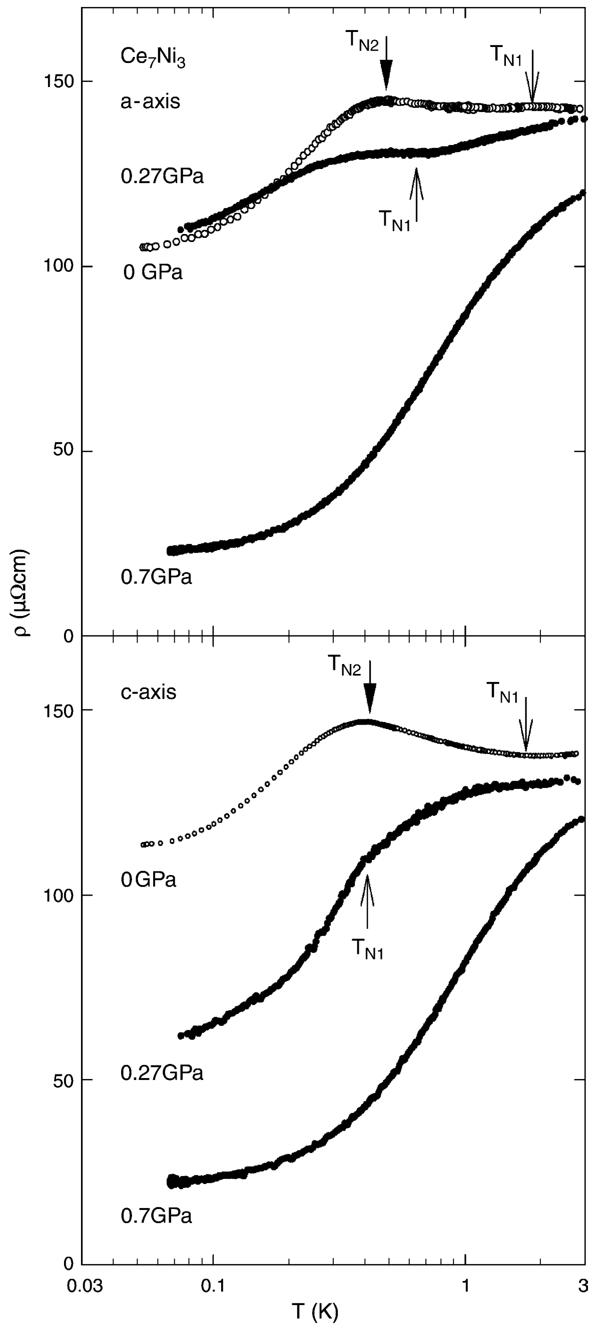

Fig. 2. Temperature dependence of the electrical resistivity of Ce_7Ni_3 along the *a* and *c* axis under various pressures.

With increasing pressure up to 0.7 GPa, both ρ_a and ρ_c are suppressed strongly and the anisotropy between $\rho_a(T)$ and $\rho_c(T)$ becomes quite small. This implies that the spin fluctuations along both the *a* and *c* axis are totally quenched by applying pressure above 0.7 GPa.

4. Field-induced magnetic phase

We summarize the magnetic field (B)–temperature (T) phase diagrams of Ce_7Ni_3 for $B \,||\, c$ in Fig. 3. Both T_{N1} and

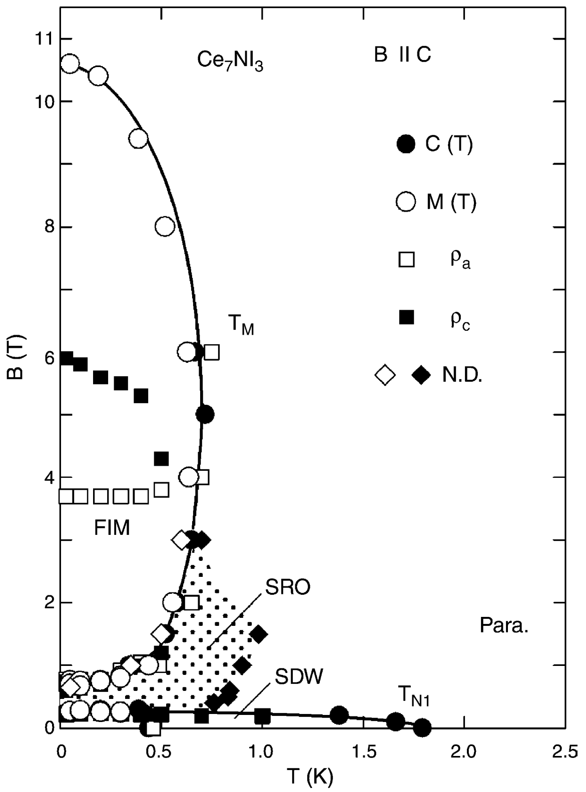

Fig. 3. Magnetic field vs. temperature phase diagram of Ce_7Ni_3 for $B \,||\, c$. The short-range order (SRO) is found by the present neutron diffraction study.

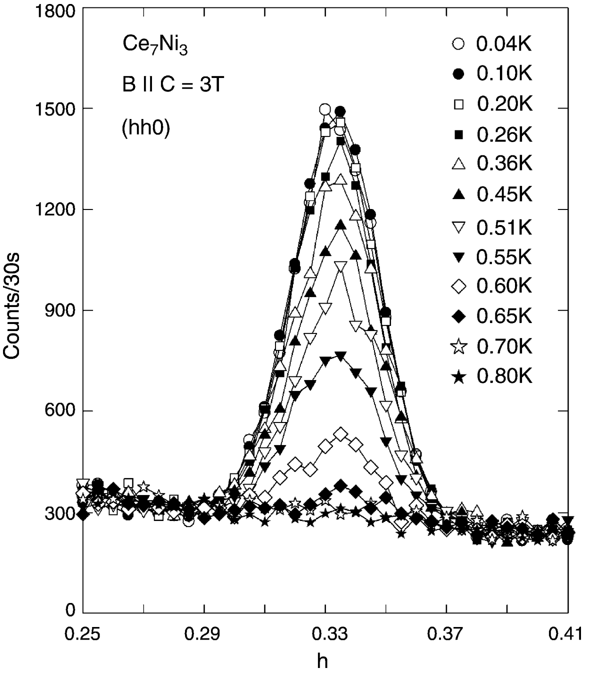

Fig. 4. Temperature dependence of the scan along ($h\,h\,0$) line around $h = 1/3$ in Ce_7Ni_3 at the magnetic field of 3 T applied along the *c* axis.

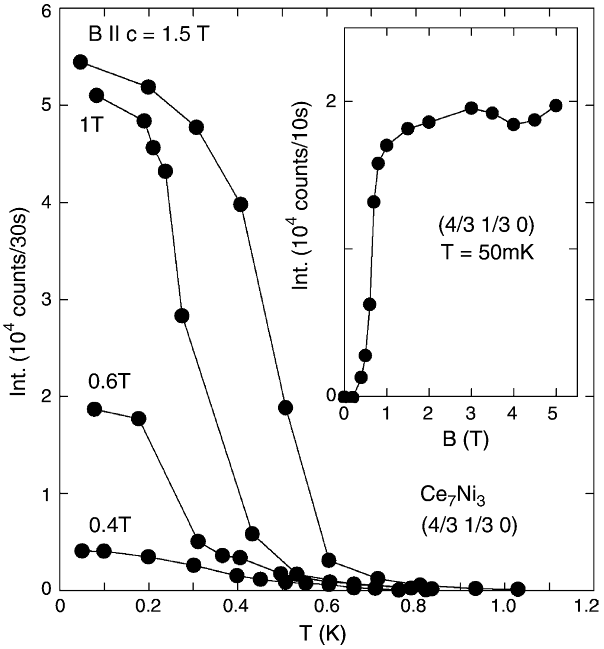

Fig. 5. Temperature dependence of the intensity of magnetic reflection at (4/3 1/3 0) of Ce_7Ni_3 in various fields applied along the c axis. The inset shows the field dependence of the intensity at $T = 50$ mK.

T_{N2} vanish at 0.3 T. Upon increasing $B \,||\, c$ above 0.7 T, a field-induced magnetic (FIM) phase appears and its boundary closes at $B \,||\, c = 10.5$ T. Furthermore, a short-range order (SRO) is found in the range $0.3 \leq B \leq 0.7$ T by the present neutron diffraction study as shown below.

At first, we scanned along a $(0\,0\,l)$ line. No magnetic reflection at $(0\,0\,l)$ for $0 < l < 1$ was detected at $T = 50$ mK when the field $B \,||\, c$ was increased to 5 T. By the scan along the $(h\,h\,0)$ lines, magnetic reflections were observed at (1/3 1/3 0) in the FIM phase. Thus, the magnetic unit cell in the c-plane is treble that of the chemical unit cell. Fig. 4 shows the temperature dependence of the scan around $h = 1/3$ at $B \,||\, c = 3$ T. The peak vanishes at 0.7 K, which corresponds with the boundary of the FIM phase in Fig. 3.

The temperature dependence of the intensity of the magnetic reflection at (4/3 1/3 0) in various constant fields $B \leq 1.5$ T is shown in Fig. 5. The temperature at which the slope has the maximum coincides with the boundary of the FIM phase in Fig. 3. However, there is a tail of the intensity up to 1 K, which is beyond the phase boundary. Furthermore, as shown in the inset of Fig. 5, the intensity at the same Q-vector exists even in the field range $0.3 < B < 0.7$ T, where no anomaly was found in the measurements of C, M and μSR [15]. Thus, the weak magnetic reflection is a manifestation of a sort of SRO. In this state, large spin fluctuations associated with the magnetic frustration should play the central role.

5. Summary

We have reported the results of the electrical resistivity under hydrostatic pressure and neutron diffraction study under magnetic field for a heavy-fermion antiferromagnet Ce_7Ni_3. The enhancement of the residual resistivity along the a axis at 0.27 GPa near the critical pressure $P_c = 0.39$ GPa is attributed to the spin fluctuations along the a axis. For $B \,||\, c > 0.3$ T, the magnetic unit cell in the c-plane is treble that of the chemical unit cell. The (1/3 1/3 0) magnetic reflection remains even in the field range $0.3 < B \,||\, c < 0.7$ T and temperature range $T < 1$ K. These results suggest that large spin fluctuations in the c-plane are responsible for both the non-Fermi liquid behavior under pressure and the anomalous magnetic phase diagram for $B \,||\, c$. The pressure- and field-induced magnetic instability of Ce_7Ni_3 should be explained by taking account of the dependence of the spin fluctuations on pressure and magnetic field. To make clear this point, further microscopic study, neutron diffraction measurements under pressure are highly desirable.

Acknowledgements

Resistivity and neutron diffraction measurements were preformed by the joint researches in ISSP of University of Tokyo. This work was supported by the COE Research (13CE2002) in a Grant-in-Aid from MEXT, Japan.

References

[1] G.R. Stewart, Rev. Mod. Phys. 73 (2001) 797.
[2] I.R. Walker, et al., Phys. C 282–287 (1997) 303;
H. Hegger, et al., Phys. Rev. Lett. 84 (2000) 4986.
[3] A.P. Ramirez, in: K.H.J. Buschow (Ed.), Handbook of Magnetic Materials, vol. 13, Elsevier Science B.V., Amsterdam, 2001, p. 426.
[4] M. Shiga, K. Fujisawa, H. Wada, J. Phys. Soc. Jpn. 62 (1993) 1329.
[5] A. Donni, et al., J. Phys.: Condens. Matter 7 (1995) 1663.
[6] Y. Isikawa, et al., J. Phys. Soc. Jpn. 65 (Suppl. B) (1996) 117.
[7] D. Gignoux, D. Schmitt, in: K.A. Gschneidner Jr., L. Eyring (Eds.), Handbook on the Physics and Chemistry of Rare Earths, vol. 20, Elsevier Science B.V., Amsterdam, 1995, p. 293.
[8] K. Umeo, et al., J. Phys. Soc. Jpn. 66 (1997) 2133.
[9] H. Kadowaki, et al., J. Phys. Soc. Jpn. 69 (2000) 2269.
[10] K. Umeo, H. Kadomatsu, T. Takabatake, J. Phys.: Condens. Matter 8 (1996) 9743.
[11] K. Umeo, et al., Phys. Rev. B 58 (1998) 12095.
[12] T. Moriya, T. Takimoto, J. Phys. Soc. Jpn. 64 (1995) 960.
[13] K. Umeo, et al., Phys. Rev. B 67 (2003) 144408.
[14] T. Sakakibara, et al., Jpn. J. Appl. Physica 33 (1994) 5067.
[15] A. Schenck, et al., Phys. B 326 (2002) 394.