Vol. 34 (2003) ACTA PHYSICA POLONICA B No 2

MAGNETIC PHASE DIAGRAM OF FILLED SKUTTERUDITE COMPOUND $SmRu_4P_{12}$*

C. Sekine, I. Shirotani

Department of Electrical and Electronic Engineering, Muroran Institute of Technology, 27-1 Mizumoto-cho, Muroran 050-8585, Japan

K. Matsuhira

Department of Electronics, Faculty of Engineering, Kyushu Institute of Technology, Kitakyushu 804-8550, Japan

P. Haen, S. De Brion, G. Chouteau

CNRS, BP 166, 38042 Grenoble cedex 9, France

H. Suzuki and H. Kitazawa

NIMS, 1-2-1 Sengen, Tsukuba 305-0047, Japan

(Received July 10, 2002)

Electrical resistivity $\rho(T)$, thermal expansion and magnetization $M(T)$ measurements in high magnetic fields have been performed on filled skutterudite $SmRu_4P_{12}$ in order to investigate the field dependence of the metal–insulator (M-I) transition ($T_{\rm MI} \sim 16$ K). The M-I transition occurs in fact in two successive steps. The specific heat shows a double peak in field. The thermal expansion coefficient, the temperature derivative of the resistivity $d\rho(T)/dT$ and of the magnetization $dM(T)/dT$ also show two anomalies at the same position as the specific heat peak. The field dependence up to 30 T of the two anomalies suggests that the two successive transitions are antiferro-quadrupolar ordering and antiferro-magnetic ordering.

PACS numbers: 71.27.+a, 71.30.+h, 74.25.Ha, 75.30.Kz

* Presented at the International Conference on Strongly Correlated Electron Systems, (SCES 02), Cracow, Poland, July 10–13, 2002.

1. Introduction

$SmRu_4P_{12}$ crystallises in the cubic filled skutterudite-type structure with space group Im$\bar{3}$ (T_h^5, # 204). It has attracted much interest because it exhibits a metal-insulator (M-I) transition at $T_{MI} = 16$ K [1]. The mechanism of the M-I transition is still an enigma. Recent work has revealed that this M-I transition occurs in fact in two successive steps [2]. The specific heat exhibits a double peak in magnetic field. The temperature derivative of the electrical resistivity $d\rho(T)/dT$ and of the magnetization $dM(T)/dT$ also exhibit two anomalies at the same positions as the specific heat peaks. The magnetic entropy estimated at zero field reaches Rln4 at T_{MI} [2]. This indicates that the crystalline electric field (CEF) ground state is Γ_{67} quartet in the point group T_h [3]. The ground state Γ_{67}, which is the same as Γ_8 quartet in the point group O_h, has both magnetic and orbital degree of freedom. The existence of a double anomaly can be ascribed to two successive transitions on cooling: orbital ordering, then magnetic ordering such as in CeB_6 [4]. In order to elucidate this point, we have performed thermal expansion, electrical resistivity and magnetization measurements in high magnetic field and investigated the magnetic phase diagram of $SmRu_4P_{12}$.

2. Experimental

Single-phase polycrystalline $SmRu_4P_{12}$ was prepared at high temperature and high pressure using a wedge-type cubic-anvil high pressure apparatus. The measurement of linear thermal expansion was performed by using a three terminal capacitance method. The electrical resistivity was measured with standard DC four-prove method in magnetic field up to 23 T at Grenoble High Magnetic Field Laboratory. Temperature dependence of magnetization was measured up to 30 T by an extraction method at Tsukuba Magnet Laboratory.

3. Results

Figure 1 shows the temperature dependence of the magnetization $M(T)$ of $SmRu_4P_{12}$ at various fields. The derivative $dM(T)/dT$ exhibits a minimum and a maximum. The two anomalies at 9T appear at almost the same position as the specific heat anomalies [2]. We define the two anomalies as T_Q and T_N, respectively. These are indicated as arrows in Fig. 1. $M(T)$ exhibits a small but clear upturn at T_Q up to 30 T. $T_Q(H)$ increases with magnetic field H. The specific heat and thermal expansion data indicate that the transition at T_Q is of second order. Thus, the positive dependence of $T_Q(H)$ is consistent with the upturn in $M(T)$ considering thermodynamic relations for second order transition. Such an upturn in $M(T)$ is often observed in compounds which show antiferro-quadrupolar (AFQ) ordering [5]. On the other hand, $M(T)$ shows a steep drop below $T_{\rm N}$ for 30 T and $T_N(H)$ decreases with H. This suggests that the anomaly at $T_{\rm N}$ is due to antiferromagnetic (AFM) ordering. The upturn well below $T_{\rm N}$ at 1 T may be due to an impurity phase.

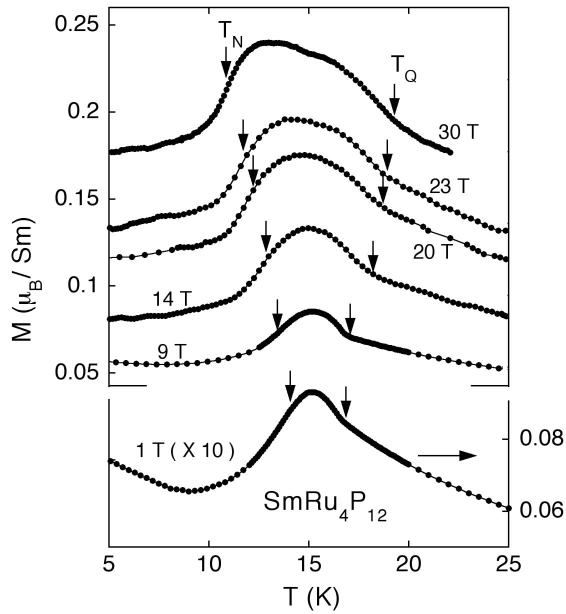

Fig. 1. Temperature dependence of magnetization at various fields.

We also determined T_Q and $T_{\rm N}$ from two anomalies of the derivative of the electrical resistivity $d\rho(T)/dT$. Further, we measured thermal expansion in order to investigate the field dependence of T_Q and $T_{\rm N}$ in low fields because it is more sensitive than specific heat measurement.

T_Q and $T_{\rm N}$ determined by our experiments are plotted in Fig. 2, where open circles, solid circles and open squares are the data points determined from $\rho(T)$, $M(T)$ and thermal expansion coefficient, respectively. The solid squares indicates the data points determined by specific heat in Ref. [2]. $T_Q(H)$ slightly increases (H^2-dependence) with increasing magnetic field H up to 20 T and shows a saturation behavior. This suggests that a 'reentrant behavior', which is observed in the H–T phase diagram of the AFQ ordering systems such as CeB_6 [4], is also expected in much higher fields for $SmRu_4P_{12}$. In Fig. 2, phase I is a paramagnetic metal. It is considered that phase III is an AFM insulator, and in phase II, AFQ and a partial gap is formed.

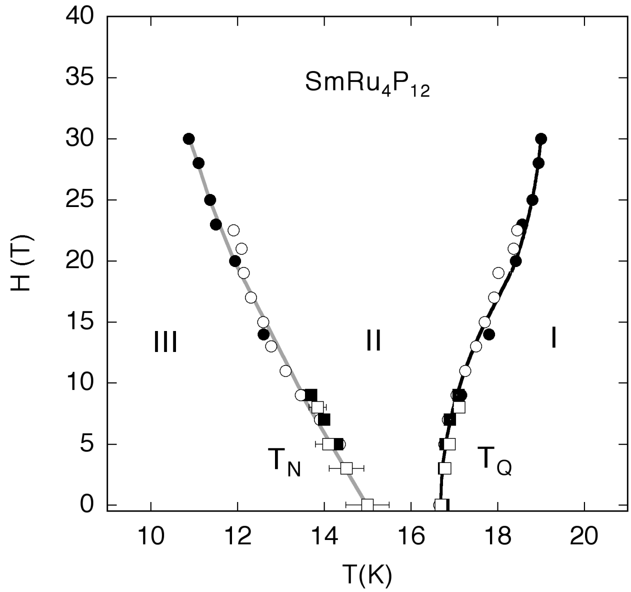

Fig. 2. *H–T* phase diagram deduced from anomalies of resistivity, magnetization, thermal expansion coefficient. Specific heat data in Ref. [2] is also plotted.

In summary, we investigated the *H–T* phase diagram of $SmRu_4P_{12}$ up to 30T. The results strongly suggests that two successive transitions are AFQ ordering and AFM ordering. At present, however, there is no microscopic confirmation of these orderings. Neutron diffraction study is desired.

The present work was partly supported by Grant-in-Aid from the Ministry of Education, Culture, Sports, Science and Technology, Japan.

REFERENCES

[1] C. Sekine, T. Uchiumi, I. Shirotani, T. Yagi, *Science and Technology of High Pressure*, Universities Press, Hyderabad, India 2000, p.826.

[2] K. Matsuhira, Y. Hinatsu, C. Sekine, T. Togashi, H. Maki, I. Shirotani, H. Kitazawa, T. Takamasu, G. Kido, *J. Phys. Soc. Jpn.* **71**, Suppl. (2002).

[3] K. Takegahara, H. Harima, A. Yanase, *J. Phys. Soc. Jpn.* **70**, 1190 (2001).

[4] J.M. Effantin, J. Rossat-Mignod, P. Burlet, H. Bartholin, S. Kunii, T. Kasuya, *J. Magn. Magn. Mater.* **47**&**48**, 145 (1985).

[5] T. Sakakibara, T. Tayama, K. Kitami, M. Yokoyama, K. Tenya, H. Amitsuka, D. Aoki, Y. Onuki, Z. Kletowski, T. Matsumura, T. Suzuki, *J. Phys. Soc. Jpn.* **69**, Suppl. A, 25 (2000).