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

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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.

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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.

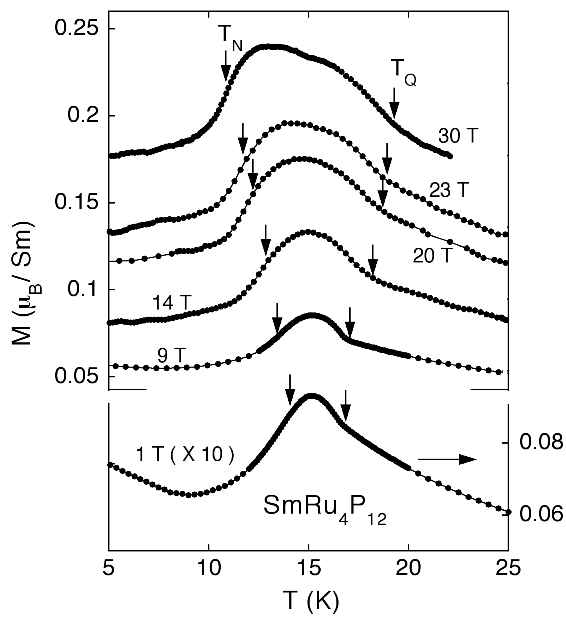

Fig. 1. Temperature dependence of magnetization at various fields.

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 $\rm CeB_6$ [4], is also expected in much higher fields for $\rm SmRu_4P_{12}$. 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.

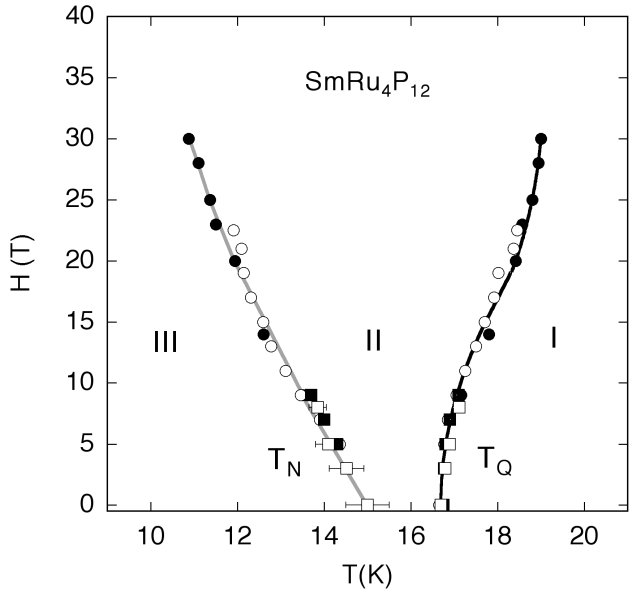

Fig. 2. *H–T* phase diagram deduced from anomalies of resistivity, magnetization, thermal expansion coefficient. Specific heat data in Ref. [2] is also plotted.

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.

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