Electrical Resistivity Measurements of Antiferromagnetic Compound Ce₃TiSb₅ under Pressure

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Abstract. We performed electrical resistivity measurements of Ce₃TiSb₅ under pressure. From the pressure dependences of antiferromagnetic ordering temperature $T_N$ and lower anomalous temperature $T^*$, temperature–pressure phase diagram of Ce₃TiSb₅ up to 2.3 GPa has been constructed. $T^*$ rapidly decreases with increasing pressure and disappears around 1 GPa. $T_N$ increases with increasing pressure, however a hump structure of electrical resistivity below $T_N$ becomes small. Hence, some change for the magnetic ordered state is expected to occur in higher pressure region.

1. Introduction

Odd-parity multipole ordering has attracted great attention owing to various unique properties such as cross-correlation phenomena [1]. An antiferromagnetic (AFM) compound with broken local inversion symmetry at magnetic ion site is an ideal system for studying odd-parity multipole ordering. On the UNi₅B, which has a toroidal ordered state with a vortex-like magnetic structure, some cross-correlation responses and an unconventional multipole ordered state predicted by theoretical study have been experimentally verified [2-4]. Recently, we reported that Ce₃TiBi₅ also shows magnetoelectric (ME) effect which is considered to be originated from magnetic ferrotoroidal order [5, 6]. There are many family ternary compounds expressed as $R_2TX_3$ ($R$: rare-earth element, $T$: transition metal, $X$: p-block element). Hence, it is expected to be an existence of the odd-parity multipole ordering other than Ce₃TiBi₅.

On the other hand, CeCoSi, in which there is no local inversion symmetry at the Ce site, has the complex magnetic temperature–pressure ($T$–$P$) phase diagram and the possibility of the antiferroquadrupolar ordering at $T_0$ has been extensively studied [7-9]. The phase transition temperature of $T_0$ shows strong pressure dependence. Note that the broad kink anomaly at $T_0$ appeared at low pressure ranges changes a broad hump anomaly at high pressure ranges [7].

Ce₃TiSb₅ is a hexagonal system with space group $P6_3/mcm$ (No. 193) and has identical crystal structure as Ce₃TiBi₅ [10]. Ce₃TiSb₅ also shows AFM transition, although the transition temperature of $T_N$ is 5.2 K and the direction of ordered Ce magnetic moments is presumed to differ from Ce₃TiBi₅ from the anisotropy of the magnetic susceptibility [11]. There is a possibility of odd-parity multipole...
ordering in Ce₃TiSb₅ due to the expected magnetic structure. Meanwhile, Ce₃TiBi₅ has not been reported about physical properties under pressure. Pressure dependence of $T_N$ is one of the keys to investigate the magnetic phase realizing in this system. To discuss about cross-correlation phenomena, it is necessary to elucidate the detail of the ordered state. In the present work, we show the results of electrical resistivity measurements of Ce₃TiSb₅ under pressure and $T$–$P$ phase diagram up to 2.3 GPa in this system.

2. Experimental

Single-crystalline samples were grown by a Sn-flux method, as described in the literature by M. Matin et al. [11]. Electrical resistivity was measured by a standard four-terminal method using indenter-type pressure cell. Daphne oil 7373 was used as the pressure transmitting media.

3. Results

Figure 1 shows temperature dependence of electrical resistivity ($\rho$) of Ce₃TiBi₅ and Ce₃TiSb₅ at ambient pressure. An electric current was applied parallel to the c-axis. Both $\rho(T)$ curves of Ce₃TiBi₅ and Ce₃TiSb₅ exhibit a double hump structure: one is around 100 K and the other is at lower temperature (Ce₃TiBi₅: $\sim$7 K, Ce₃TiSb₅: $\sim$9 K). The double hump structure is considered to originate in Kondo effect in the excited CEF states. $\rho(T)$ of Ce₃TiBi₅ exhibits a clear kink at AFM transition temperature $T_N$ = 5.0 K, then $\rho(T)$ only decreases with decreasing temperature. Note, however, that the derivative resistivity with temperature ($d\rho/dT$) shows a broad peak about 3 K. In contrast, $\rho(T)$ of Ce₃TiSb₅ exhibits a minimum at $T_N$ = 5.2 K, then a hump structure appears at just below $T_N$. This anomaly reminds us the charge-density-wave (CDW) or spin-density-wave (SDW) transition with Fermi surface reconstruction. This is consistent with the result of neutron scattering experiment [12]. The difference of the $\rho(T)$ between Ce₃TiBi₅ and Ce₃TiSb₅ is considered to be due to the magnetic anisotropy, but microscopic studies as NMR and neutron diffraction measurements are needed in the future. We can also notice there is another feature of $\rho(T)$ of Ce₃TiSb₅ which is a small anomaly at $T'$ = 2.5 K. Ce₃TiSb₅ often contains Sn as an impurity phase at the crystal growth by a Sn-flux method, then the sudden decrease at about 3.5 K is often observed [11]. However, the anomaly at $T'$ is also observed with applying higher magnetic field than the critical field of Sn. Therefore, this anomaly is considered to be an intrinsic behavior for Ce₃TiSb₅. $T'$ is close to the temperature at which one of the two kinds of the magnetic scattering intensities on the neutron powder diffraction measurements increases [12].

Next, we show temperature dependence of $\rho$ of Ce₃TiSb₅ at various pressure in Fig. 2. An electric current was applied parallel to the c-axis because of the very thin need-shape single crystal. The double hump structure caused by Kondo effect is enhanced with increasing pressure. Similar behaviors are seen in $\rho(T)$ of Ce₃TiBi₅ [13]. On the $\rho(T)$ of the Ce₃TiBi₅, the higher hump of the double hump structure clearly changes to a peak anomaly above 6 GPa. On the other hand, $\rho(T)$ of the Ce₃TiSb₅ at
1.81 GPa clearly exhibits a peak anomaly at the higher hump of the double hump structure. These differences are considered to be due to the chemical pressure effect replacing Bi with Sb. Down triangle symbols indicate the anomaly at $T^*$. $T^*$ rapidly decreases with increasing pressure and disappears around 1 GPa. From a perspective of the anomaly appearing temperature at ambient pressure, the anomaly at $T^*$ is consistent with the ordered phase observed by neutron scattering measurement below 3 K. However, the decrease of the lattice constant along the $a$-axis in the ordered phase is observed at ambient pressure [12]. It seems to contradict the decrease of $T^*$ under pressure. In the previous report of magnetic properties of Ce$_3$TiBi$_5$, there is no clear anomaly at $T^*$ in the temperature dependence of the magnetic susceptibility and only small broad anomaly below 2 K on the temperature dependence of the specific heat was observed.

Upward triangle symbols indicate the AFM ordering at $T_N$. $T_N$ increases with increasing pressure and reaches 6.7 K at 2.31 GPa. The hump anomaly just below $T_N$ becomes smaller with pressure application despite the increase in $T_N$. This hump structure is expected to disappear around 3 GPa by further pressurization. This behaviour implies the possibility that some change occurs in AFM ordered state. In the case of that the hump anomaly just below $T_N$ changes to an anomaly with only decreasing, the order parameter of AFM ordered state might be changed by pressure.

Finally, we show the $T$–$P$ phase diagram of Ce$_3$TiBi$_5$ in Fig. 3. The $T$–$P$ phase diagram has been constructed from the results of Fig. 2. $T_N$ monotonically increases with increasing pressure at least up to 2.31 GPa. The increase of $T_N$ under pressure is more gradual than that of Ce$_3$TiBi$_5$. The double hump structure on Ce$_3$TiSb$_5$ caused by Kondo effect is enhanced by the application of lower pressure than Ce$_3$TiBi$_5$. Due to the substitution of Bi with Sb, it is expected that quantum critical point of Ce$_3$TiSb$_5$ emerges lower pressure region than Ce$_3$TiBi$_5$. The existence of the high-pressure phase and the details of the low-temperature phase are still unknown, but the existence of these phases is

Figure 2. Temperature dependences of electrical resistivity of Ce$_3$TiSb$_5$ under several pressure.

Figure 3. Temperature–pressure phase diagram of Ce$_3$TiSb$_5$. 
interesting because the changes in the order parameter affect the multipole ordered state and the ME effect derived from it.

4. Summary
We performed electrical resistivity measurements of Ce₃TiSb₅ under pressure. T–P phase diagram of Ce₃TiSb₅ up to 2.3 GPa has been constructed. The lower anomaly at T* is found to rapidly decrease with increasing pressure and disappears around 1 GPa. T_N is still increasing in this pressure range. In contrast, the hump structure of ρ below T_N becomes small. Thus, we expect some change of AFM ordered state with further pressurization.

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