Low-temperature physical properties of a new cubic compound CeMgZn₂

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Abstract. Magnetic and transport properties of a new cubic compound CeMgZn₂ have been examined by measuring the magnetic susceptibility, the magnetization, the electrical resistivity, and the specific heat. CeMgZn₂ is a Kondo-lattice compound with trivalent Ce ions. The magnetic susceptibility measured at 0.1 T exhibits a shoulder and a cusp at \( T_{N1} = 5.4 \) K and \( T_{N2} = 3.1 \) K, respectively. \( T_{N1} \) and \( T_{N2} \) correspond to the antiferromagnetic-transition temperatures since these temperatures decrease with increasing magnetic field. The large value of the paramagnetic Curie temperature divided by \( T_{N1} \) (13.5) implies that \( T_{N1} \) is suppressed by geometrical frustration on a face-centered cubic Ce sublattice. The geometrical frustration may also be responsible for the appearance of many magnetic phases in magnetic fields.

1. Introduction

Magnetic ground states of Ce- or Yb-based compounds have been understood by a concept proposed by Doniach [1], i.e., if the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction is dominant, a magnetically ordered state is realized, whereas the Kondo effect is dominant, a paramagnetic heavy fermion state is realized. In particular, a quantum critical point (QCP) where the ground state changes from a magnetically ordered state to a heavy fermion state has been a subject of intensive studies. This is because quantum fluctuation in the vicinity of a QCP leads some anomalous phenomena such as unconventional superconductivity. Recently, the other viewpoint of quantum criticality has been discussed theoretically, i.e., quantum zero-point motion of the spins induced by magnetic frustration [2]. The magnetic ground states are described by two-axis diagram with the Kondo screening axis (K) and quantum-zero point motion axis (Q) [2]. According to the QK diagram, Ce and Yb compounds possessing magnetic frustration can exhibit a variety of magnetic ground states as a result of competition between the RKKY interaction, the Kondo effect, and the quantum-zero point motion. Although the simplest way to introduce magnetic frustration is to locate Ce or Yb ions on geometrically frustrated lattices, such lattices are rather rare compared with those for transition-metal ions. Therefore, searching for new Ce or Yb compounds with geometrical frustration is important to explore a variety of magnetic ground states and concomitant anomalous phenomena.

We have focused on a new Heusler-type cubic compound CeMgZn₂ (space group \( Fm\bar{3}m \), \( O_h^5 \), No. 225) [3]. The Ce ions are located on the geometrically frustrated lattice since they are on a face-centered cubic lattice and form a network of edge-sharing tetrahedra. No physical properties of this compound...
have been reported thus far. In this study, polycrystalline samples of CeMgZn\(_2\) have been grown and their magnetic and transport properties have been examined by measuring the magnetic susceptibility, the magnetization, the electrical resistivity, and the specific heat.

2. Experimental methods
Polycrystalline samples of CeMgZn\(_2\) and a nonmagnetic reference compound LaMgZn\(_2\) were grown by melting the stoichiometric constituent elements in Mo crucibles at 1320 K for 12 hours and subsequent annealing at 1220 K for 50 hours. The X-ray diffraction experiments revealed that all of the Bragg peaks in the diffraction pattern of CeMgZn\(_2\) can be indexed on the basis of the reported structure, although small peaks of unidentified impurity phases are discernible for LaMgZn\(_2\). The lattice constants determined by X-ray diffraction experiments at room temperature are \(a = 7.023(1) \text{ Å}\) for CeMgZn\(_2\) and \(a = 7.101(1) \text{ Å}\) for LaMgZn\(_2\). The magnetization \(M(T, H)\) was measured using a superconducting quantum interference device magnetometer (Quantum Design, MPMS) as functions of the temperature \(T\) and magnetic field \(H\) between 1.8 and 300 K up to 5 T. The electrical resistivity \(\rho(T)\) was measured with a dc four-probe method between 0.4 and 300 K in a laboratory-built \(^3\)He cryostat. The specific heat \(C(T)\) was measured with a thermal relaxation method between 0.6 and 9 K in a \(^3\)He cryostat.

3. Results and Discussion
The inverse magnetic susceptibility \(1/M\) of CeMgZn\(_2\) (not shown) above 100 K can be fitted by the Curie-Weiss law. The effective magnetic moment \(\mu_{\text{eff}}\) and the paramagnetic Curie temperature \(\theta_P\) derived by Curie-Weiss fitting were 2.75 \(\mu_B/\text{Ce}\) (\(\mu_B\): Bohr magneton) and \(-73\) K, respectively. The Ce ions in this compound are trivalent because the effective magnetic moment is close to that of the free Ce\(^{3+}\) ion (2.54 \(\mu_B/\text{Ce}\)). The negative \(\theta_P\) indicates antiferromagnetic correlation between Ce moments.

Figure 1 shows the temperature dependences of the magnetic susceptibilities \(M/H\) of CeMgZn\(_2\) measured in various magnetic fields. The \(M/H\) measured at 0.1 T shows a shoulder and a cusp at \(T_{N1} = 5.4\) K and \(T_{N2} = 3.1\) K, respectively. \(T_{N1}\) and \(T_{N2}\) decrease with increasing magnetic fields as indicated by solid and dashed arrows, respectively. The decrease in \(T_{N1}\) and \(T_{N2}\) by magnetic fields and the negative \(\theta_P\) indicate that these temperatures are the antiferromagnetic-transition temperatures. Above 2.5 T, only \(T_{N1}\) was observed. No anomalies were observed above 3 T.

Figure 2. Magnetic-field dependences of the magnetization \(M\) and their field derivatives \(dM/dH\) of CeMgZn\(_2\) measured at (a) 2.5 K and (b) 4 K.
The magnetic-field dependence of the magnetization $M$ at 2.5 K ($T < T_{N2}$) has two anomalies at $H_1$ and $H_2$ as shown in the figure 2(a). These anomalies are clearly observed as the peaks of the field derivative $dM/dH$. On the other hand, the $M$ at 4 K ($T_{N2} < T < T_{N1}$) has one anomaly at $H_2$ as shown in the figure 2(b).

The temperature dependence of the electrical resistivity $\rho$ is shown in the figure 3. The $\rho(T)$ decreases with decreasing temperature, followed by a minimum at 20 K and shows the $-\ln T$ dependence between 8 and 15 K. This means that CeMgZn$_2$ is a Kondo-lattice compound. A steep decrease is observed below $T_{N1}$, while no anomalies are observed below $T_{N2}$.

Figure 4 shows the temperature dependence of the specific heat $C$ of LaMgZn$_2$ and CeMgZn$_2$. The $C(T)$ of LaMgZn$_2$ shows typical nonmagnetic behavior. On the other hand, the $C(T)$ of CeMgZn$_2$ has a $\lambda$-type anomaly at $T_{N1}$. This means that the second-order antiferromagnetic transition occurs at $T_{N1}$. In contrast, the $C(T)$ of CeMgZn$_2$ has no anomaly at $T_{N2}$ as for the case of the resistivity. To examine the mass enhancement of the conduction electrons of CeMgZn$_2$, the $C(T)$ of these compounds were fitted by using a formula $C(T) = \gamma T + \beta T^3$. $\gamma$ is the electronic specific-heat coefficient. Although only the phonon contributes to $\beta$ for LaMgZn$_2$, the antiferromagnetic magnon also contributes to $\beta$ for CeMgZn$_2$. The red and light-blue lines in the figure 4 are the fitted results using this formula. The $\gamma$ and the $\beta$ values were estimated to be $\gamma = 0.0054$ J/mol K$^2$ and $\beta = 3.7 \times 10^{-4}$ J/mol K$^4$ for LaMgZn$_2$, and $\gamma = 0.018$ J/mol K$^2$ and $\beta = 0.16$ J/mol K$^4$ for CeMgZn$_2$. The rather small $\gamma$ value of CeMgZn$_2$ (only 3 times larger than that of LaMgZn$_2$) reveals that the 4$f$ electrons have localized nature even at low temperatures.

The temperature dependence of the magnetic entropy $S_{mag}$ of CeMgZn$_2$ is also depicted in the figure 4. The $S_{mag}$ was calculated by integrating $C_{mag}/T$ in $T$, where $C_{mag}$ is the magnetic specific heat that was deduced by subtracting the $C(T)$ of LaMgZn$_2$. The magnetic entropy at $T_{N1}$ is 4.2 J/mol K. Since this value is 73% of $R\ln 2$ ($R$: gas constant), it is considered that the ground state of 4$f$ energy levels split under the cubic crystalline electric field is the $\Gamma_7$ doublet.

The magnetic field versus temperature phase diagram was drawn by plotting the susceptibility anomalies and the magnetization anomalies as shown in the figure 5. Below 3 T, four magnetic phases named I, II, III, and IV appear. We now consider that competing magnetic interaction due to geometrical frustration is responsible for the appearance of many magnetic phases. The frustration effect is
evidenced by the reduction of $T_{N1}$. The value of $|\theta|$ divided by transition temperature is often used for evaluating the strength of suppression of transition temperature due to frustration and the value larger than 5 indicates a strong suppression [4]. The $|\theta|/T_{N1}$ value of CeMgZn$_2$ is 13.5. This value is comparable to $|\theta|/T_N = 12.6$ for CePdAl whose antiferromagnetically ordered state below $T_N$ is strongly affected by frustration [5].

4. Conclusions
In this study, polycrystalline samples of CeMgZn$_2$ and the reference compound LaMgZn$_2$ have been grown and their magnetic and transport properties have been examined. We found that CeMgZn$_2$ is a Kondo-lattice compound with the trivalent Ce ions. CeMgZn$_2$ shows antiferromagnetic transition at $T_{N1} = 5.4$ K and $T_{N2} = 3.1$ K. By constructing magnetic field versus temperature phase diagram, we found four magnetic phases below 3 T. Because the $|\theta|/T_{N1}$ value reaches 13.5, we consider that geometrical frustration on a face-centered cubic Ce sublattice reduces $T_{N1}$. The geometrical frustration is also responsible for the appearance of many magnetic phases.

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