Low-temperature physical properties of new orthorhombic compounds RE$_2$Au$_3$Sn$_6$ (RE = Ce, La)

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Abstract. We have prepared polycrystalline samples of RE$_2$Au$_3$Sn$_6$ (RE = Ce, La) and investigated their magnetic, transport and thermal properties. We found that Ce$_2$Au$_3$Sn$_6$ shows antiferromagnetic transition at $T_N = 2.54$ K, while La$_2$Au$_3$Sn$_6$ shows no phase transition above 0.4 K. The electronic specific heat coefficient of Ce$_2$Au$_3$Sn$_6$ is $\gamma = 350$ mJ Ce -mol K$^{-2}$, which indicates that Ce$_2$Au$_3$Sn$_6$ is a heavy fermion compound. The magnetic entropy of Ce$_2$Au$_3$Sn$_6$ at $T_N$ is estimated to be 70% of $R\ln2$, which can be attributed to the shielding of the magnetic moment by the Kondo effect.

1. Introduction

Compounds containing Ce show a variety of interesting phenomena, such as anomalous magnetic ordering and heavy fermion state due to the interaction between 4$f$ and conduction electrons. These phenomena are due to the interplay between the Ruderman-Kittel-Kasuya-Yosida interaction and the Kondo effect [1]. It is well known that Ce compounds with their magnetic states being at the vicinity of the quantum critical point exhibit some anomalous phenomena such as non-Fermi liquid behavior and unconventional superconductivity. One possible way to search for new compounds with quantum criticality is to search for compounds without magnetic order down to cryogenic temperatures. From this view point, we have focused on newly synthesized compounds RE$_2$Au$_3$Sn$_6$ (RE = La, Ce, Pr, Nd, Sm) [2]. RE$_2$Au$_3$Sn$_6$ crystallize with the orthorhombic La$_2$Zn$_3$Ge$_6$ type structure (space group Cmcm, $D_{2h}^{17}$, No. 63) as shown in the figure 1 [2]. This structure has two independent RE sites. The magnetic susceptibility measurement down to 2.5 K has revealed that Ce$_2$Au$_3$Sn$_6$ has trivalent Ce ions and no phase transition above 2.5 K [2]. In this study, we have prepared polycrystalline samples of RE$_2$Au$_3$Sn$_6$ (RE = Ce, La) and investigated their magnetic, transport and thermal properties down to 0.4 K.

Figure 1. Crystal structure of RE$_2$Au$_3$Sn$_6$. 

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2. Experimental Methods

The polycrystalline samples of RE$_2$Au$_3$Sn$_6$ (RE = Ce, La) have been prepared as the following procedure. First, stoichiometric amount of RE (99.9%), Au (99.99%) and Sn (99.999%) were melted by using arc furnace in an argon atmosphere. After that, the samples wrapped in Ta foils were sealed in the evacuated quartz tubes and annealed at 600°C for one week. The resulting polycrystalline samples were analyzed by powder X-ray diffraction. For both Ce$_2$Au$_3$Sn$_6$ and La$_2$Au$_3$Sn$_6$, all Bragg peaks in the diffraction patterns are indexed on the basis of the reported structure. The lattice parameters determined from X-ray diffraction agree with those reported in the Ref. [2]. We then measured the following three properties. The magnetization $M$ was measured from 1.8 K to 300 K and from 0 T to 5 T using a superconducting quantum interference device magnetometer (Quantum Design, MPMS). The electrical resistivity $\rho$ was measured from 0.4 K to 300 K using a $^3$He cryostat with the DC four-terminal method. The specific heat $C$ was measured from 0.6 K to 10 K by the thermal relaxation method using a $^3$He cryostat.

3. Results and Discussion

Figure 2 shows the temperature dependence of the magnetic susceptibility $M/H$ of Ce$_2$Au$_3$Sn$_6$. The $M/H$ above 100 K obeys the modified Curie Weiss law $M/H = C_{\text{Curie}}/(T - \theta_h) + \chi_0$ as shown by the red line in the figure 2. Here, $C_{\text{Curie}}$, $\theta_h$, and $\chi_0$ are the Curie constant, the paramagnetic Curie temperature, and the sum of the Pauli paramagnetic contribution and the core diamagnetic one, respectively. The obtained parameters are $C_{\text{Curie}} = 0.769$ emu K/Ce-mol, $\theta_h = -8.02$ K, and $\chi_0 = 0.000437$ emu/Ce-mol. The effective magnetic moment of Ce ion is calculated to be $\mu_{\text{eff}} = 2.48 \mu_B$/Ce by using $C_{\text{Curie}}$. This $\mu_{\text{eff}}$ value indicates that the Ce ions in Ce$_2$Au$_3$Sn$_6$ are trivalent since this value is close to that of the free Ce$^{3+}$ ion (2.54 $\mu_B$/Ce). The negative value of $\theta_h$ suggests that the magnetic interaction between the Ce ions is antiferromagnetic. These results are consistent with the previous report.

The low-temperature part of the $M/H$ is shown in the inset of the figure 2. The $M/H$ shows a cusp at $T_N = 2.54$ K, implying that the antiferromagnetic transition occurs at $T_N$.

The temperature dependence of the electrical resistivity $\rho$ is also shown in the figure 2 and the inset. The $\rho$ decreases with decreasing temperature and shows a broad shoulder around 70 K. The steep decrease at $T_N$ is ascribable to the decrease in scattering of conduction electrons by Ce magnetic moments.

The magnetic-field dependence of the magnetization $M$ of Ce$_2$Au$_3$Sn$_6$ is shown in the figure 3. At 5 K (> $T_N$), the $M$ increases with increasing magnetic field and shows no anomalies. On the other hand, the $M$ at 1.8 K (< $T_N$) shows the metamagnetic transition due to a spin flop at 2 T. The transition field
was determined as the peak field of the field derivative $dM/dH$ as shown in the figure 3.

Figure 4 shows the temperature dependence of the specific heat $C$ of Ce$_2$Au$_3$Sn$_6$ and La$_2$Au$_3$Sn$_6$. The $C$ of Ce$_2$Au$_3$Sn$_6$ shows a $\lambda$-type anomaly at $T_N$, which indicates that the cusp of $M/H$ and the decrease in $\rho$ are due to the intrinsic second-order antiferromagnetic transition. On the other hand, La$_2$Au$_3$Sn$_6$ does not show any anomaly down to 0.4 K. The electronic specific heat coefficient of Ce$_2$Au$_3$Sn$_6$ is estimated to be $\gamma = 350 \text{ mJ/Ce-mol K}^2$, which is about 600 times larger than that of La$_2$Au$_3$Sn$_6$, $\gamma = 0.59$ mJ/La-mol K$^2$. This indicates that Ce$_2$Au$_3$Sn$_6$ is a heavy fermion compound. Here, $\gamma$ and the phonon specific heat coefficient $\beta$ were determined by fitting the formula $C/T = \gamma + \beta T^2$ on the experimental $C/T$ from 6 K to 10 K for Ce$_2$Au$_3$Sn$_6$ and from 0.4 K to 4 K for La$_2$Au$_3$Sn$_6$.

Figure 5 shows the temperature dependence of the magnetic entropy $S_{mag}$ calculated by integrating $C_{mag}/T$ in temperature. $C_{mag}$ is the magnetic specific heat obtained by subtracting the $C$ of La$_2$Au$_3$Sn$_6$ from that of Ce$_2$Au$_3$Sn$_6$ under the assumption that the nonmagnetic contribution to the $C$ of these compounds is identical. The $S_{mag}$ at $T_N$ is estimated to be 4.0 J/Ce-mol K which is 70% of Rh2 ($R$: gas constant). The reduction of $S_{mag}$ value from Rh2 can be attributed to the shielding of the magnetic moment by the Kondo effect.

![Figure 4](image_url)  
**Figure 4.** Temperature dependence of the specific heat $C$ of Ce$_2$Au$_3$Sn$_6$ and La$_2$Au$_3$Sn$_6$.

![Figure 5](image_url)  
**Figure 5.** Temperature dependence of magnetic entropy $S_{mag}$ of Ce$_2$Au$_3$Sn$_6$.

4. Conclusion
We have prepared polycrystalline samples of RE$_2$Au$_3$Sn$_6$ (RE = Ce, La) and investigated their magnetic, transport, and thermal properties by measuring the magnetization, the electrical resistivity, and the specific heat. Ce$_2$Au$_3$Sn$_6$ is an antiferromagnet with the transition temperature at $T_N = 2.54$ K. The large electronic specific heat coefficient $\gamma = 350 \text{ mJ/ Ce-mol K}^2$ indicates that Ce$_2$Au$_3$Sn$_6$ is a heavy fermion compound. The reduced magnetic entropy of Ce$_2$Au$_3$Sn$_6$ (70% of Rh2) at $T_N$ can be attributed to the Kondo effect. The nonmagnetic reference compound La$_2$Au$_3$Sn$_6$ shows a nonmagnetic metallic behavior and does not show any phase transition down to 0.4 K.

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References
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