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LETTER TO THE EDITOR

Electronic and magnetic properties of a new heavy-fermion compound, $CeRuSn_3$

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Abstract. Measurements of electrical resistivity, Hall coefficient, thermoelectric power, magnetic susceptibility and specific heat have been made on $CeRuSn_3$ with cubic $Pm\bar{3}n$ structure. The results indicate that $CeRuSn_3$ is a new heavy-fermion compound with a specific heat coefficient of $1.4\,J\,mol^{-1}\,K^{-2}$ at 0.6 K. Above 0.6 K, it shows no magnetic ordering.

The properties of heavy-fermion Ce compounds with non-magnetic ground states have been studied extensively in recent years. Their common features are: an enhanced electronic specific heat coefficient, a large Pauli paramagnetic susceptibility at low temperature, a resistivity drop below the Kondo temperature and a large positive thermoelectric power at high temperature.

Heavy-fermion compounds have relatively large Ce spacing: 4.43 Å and 4.83 Å in $CeAl_3$ and $CeCu_6$, respectively [1, 2]. The spacing of f-electron atoms in heavy-fermion compounds is an important parameter, presumably describing the effective width of the hybridised f band [3, 4]. Almost all the cubic Kondo compounds, such as CeB_6 and $CeAl_2$ [5, 6], order magnetically at low temperatures, except $CeInCu_2$ which has recently been reported to be a heavy-fermion compound [7]. $CeInCu_2$ also has a large Ce spacing of 4.80 Å, which is close to that of $CeCu_6$.

The existence of compounds with the formula $RRuSn_3$ (R ≡ La, Ce, Pr, Nd) was reported by Eisenmann and Schäfer [8]. $CeRuSn_3$ crystallises in a cubic structure (with $Pr_3Rh_4Sn_{13}$ structure $Pm\bar{3}n$) and with a Ce spacing of 4.86 Å. It is of interest to know physical properties of $CeRuSn_3$. In this Letter we report the first experimental studies of resistivity, magnetic susceptibility, Hall coefficient, thermoelectric power and specific heat on this compound.

Polycrystals of $CeRuSn_3$ and $LaRuSn_3$ were obtained under argon atmosphere by arc melting stoichiometric amounts of 99.9% purity Ce and La, 99.98% purity Ru and 99.999% purity Sn. To ensure homogeneity, the ingots were turned over and remelted several times. The samples obtained were analysed by x-ray diffraction. No parasitic phase was detected even in as-cast samples. The lattice constants obtained by x-ray analysis agree with literature values, namely 9.73 Å and 9.77 Å for $CeRuSn_3$ and $LaRuSn_3$, respectively. Single-crystal samples were grown by the Czochralski pulling

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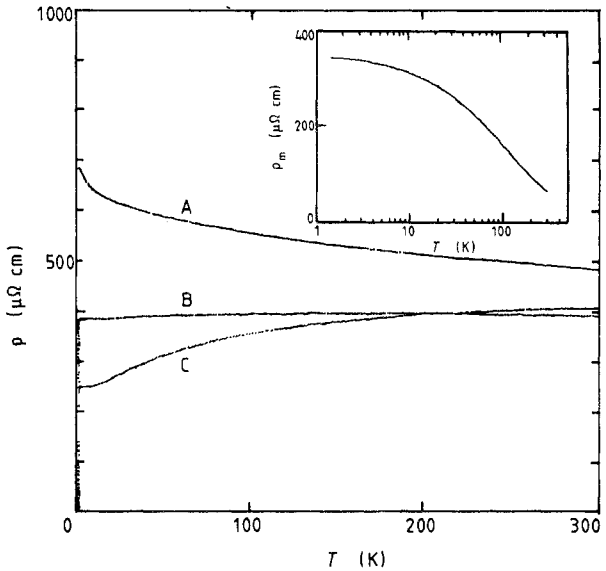

Figure 1. Temperature dependence of electrical resistivity $\rho(T)$ of $CeRuSn_3$ (curve A) and $LaRuSn_3$: as-grown (curve B), annealed (curve C). Inset shows temperature dependence of magnetic resistivity $\rho_m = \rho(CeRuSn_3) - \rho(LaRuSn_3$ (curve C)).

method using a tri-arc furnace. Samples were annealed at 950 °C for three days. For transport measurements, samples were cut into rectangular rods by a spark cutter.

Figure 1 shows the temperature dependence of the electrical resistivity for $CeRuSn_3$ and $LaRuSn_3$. For $CeRuSn_3$, the resistivity increases at least down to 1.3 K. All $CeRuSn_3$ samples measured have almost the same resistivity of about $470 \pm 40\ \mu\Omega$ cm at 300 K and about $670 \pm 50\ \mu\Omega$ cm at 1.3 K. We have plotted the magnetic resistivity ρ_m ($\rho_m = \rho_{CeRuSn_3} - \rho_{LaRuSn_3}$) in the inset of figure 1. The ρ_m is proportional to $-\log T$ between 300 K and 70 K. At the present stage, it is not clear why we do not see a decrease in resistivity due to the coherence effect at low temperatures.

In order to study the dependence of the resistivity on heat treatments, we repeated the measurement after annealing $CeRuSn_3$ under different conditions: 1000 °C for three days; 950 °C for three days; 600 °C for two weeks. There is no detectable change of the resistivity. In the case of $LaRuSn_3$, the effect of annealing on the temperature dependence of the resistivity is larger than that on $CeRuSn_3$. The resistivity of an as-grown sample increases with decreasing temperature, reaches a broad maximum and then decreases at lower temperatures. For a sample annealed at 950 °C for three days, the resistivity shows normal metallic behaviour. $LaRuSn_3$ shows a transition to a superconducting state at $T_c \simeq 1.5$ K.

Figure 2 shows the temperature dependence of the inverse magnetic susceptibility χ^{-1} of $CeRuSn_3$ between 1.7 and 300 K. The susceptibility follows the Curie–Weiss law above 100 K. The paramagnetic Curie temperature and the effective Bohr magneton μ_{eff} are about −38 K and $2.5\mu_B$ mol^{-1} respectively. μ_{eff} is close to the value for Ce^{3+} ($2.56\mu_B$). The susceptibility shows a large value of about 1.5×10^{-1} emu mol^{-1} at 1.7 K, which is about five times as large as the values that have been observed for $CeCu_6$, $CeAl_3$ and $CeInCu_2$. The preliminary AC susceptibility measurement below 1.7 K reveals a sharp maximum around 0.5 K suggesting some magnetic ordering. The susceptibility of

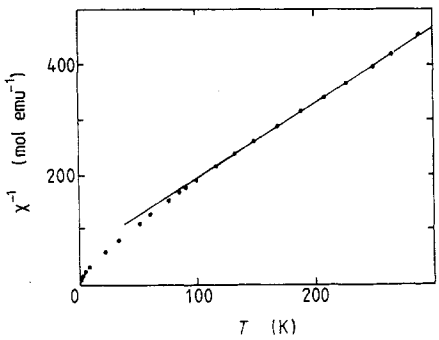

Figure 2. Temperature dependence of inverse magnetic susceptibility $\chi^{-1}(T)$ of $CeRuSn_3$.

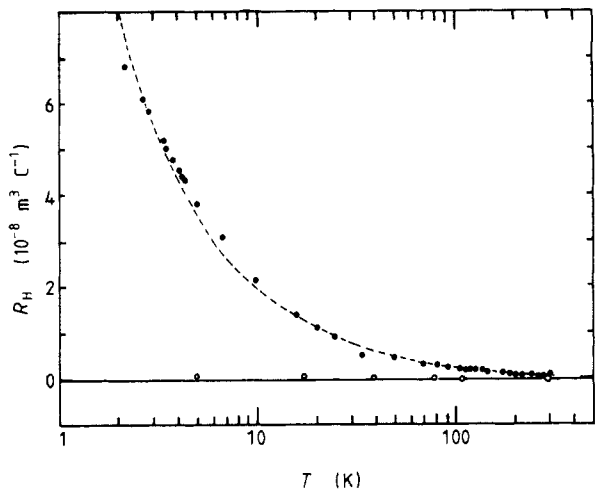

Figure 3. Temperature dependence of Hall coefficient $R_H(T)$ of $CeRuSn_3$ (●) and $LaRuSn_3$ (○) at 1.0 T. The broken curve is calculated from equation (1).

$LaRuSn_3$ is 1.6×10^{-4} emu mol^{-1} at 300 K and is almost independent of temperature down to 1.5 K.

Figure 3 shows the temperature dependence of the Hall coefficient R_H. The Hall coefficient of $LaRuSn_3$ is 2×10^{-10} m^3 C^{-1} at 300 K and is weakly temperature dependent. The estimated carrier density based on a single carrier model is normal metallic (3×10^{28} electrons m^{-3} at 300 K). The Hall coefficient of $CeRuSn_3$ increases with decreasing temperature and has a large value of 6.8×10^{-8} m^3 C^{-1} at 2 K. The Hall coefficient of this f-electron system is still increasing at 2 K. According to Coleman and co-workers [9], the Hall coefficient due to skew scattering by independent Ce impurities is given by

$$R_H \sim g\mu_B \rho_m \tilde{\chi}(1 - \tilde{\chi}T) \tag{1}$$

where g is the gyromagnetic ratio of the f electron and $\tilde{\chi}$ is the normalised susceptibility

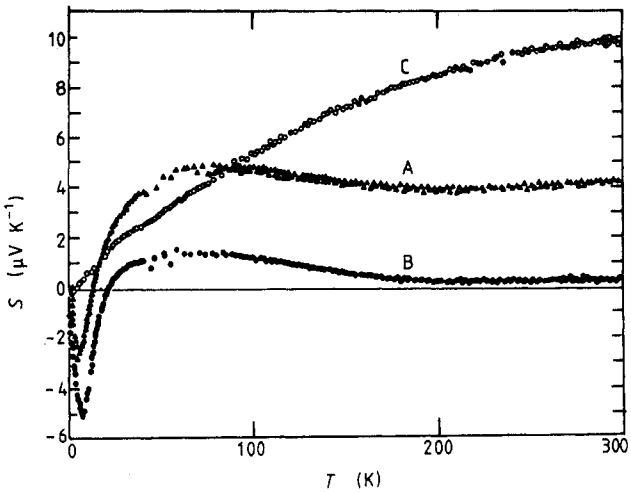

Figure 4. Temperature dependence of the thermoelectric power $S(T)$ of $CeRuSn_3$ (curves A and B) and $LaRuSn_3$ (curve C).

$\tilde{\chi} = \chi/C$ (C being the Curie constant). In figure 3 the broken curve shows R_H calculated by equation (1). In order to obtain a good fit, we multiplied ρ_m by a factor of 3.3. All these experimental features are characteristic of Ce dense Kondo compounds.

Figure 4 shows the temperature dependence of the thermoelectric power. Compared to the other properties, the dependence of thermoelectric power on the sample is larger. The temperature dependencies measured on different samples, however, have common characteristics. In figure 4, two examples of the temperature dependence of $CeRuSn_3$ are shown. We observe a negative sharp minimum around 6 K, and a positive broad maximum around 80 K. The sign of thermoelectric power is positive at 300 K, though the positive value is somewhat smaller compared to general Ce Kondo compounds. Maekawa and co-workers analysed the thermoelectric power of the Ce Kondo system theoretically [10]. They obtained positive thermoelectric power in all the temperature range, unless they take into account the potential scattering and/or coherence effect of electron scattering by Ce ions. Since in $CeRuSn_3$ the coherence effect is not clearly seen in resistivity or Hall effect in this temperature range, the effect of potential scattering is the possible origin of the negative peak near 6 K. For $LaRuSn_3$, thermoelectric power at 300 K is positive and decreases monotonically with lowering temperature without change of sign down to T_c.

Figure 5 shows the temperature dependence of the specific heat coefficient divided by temperature, C/T, of $CeRuSn_3$ between 0.6 and 20 K. The C/T has a minimum around 7 K then increases at lower temperatures without any sign of phase transition down to 0.6 K. At 0.6 K, it reaches 1.40 J mol^{-1} K^{-2}, which is comparable with those of $CeCu_6$, $CeAl_3$ and $CeInCu_2$. The C/T versus T curve bears a close resemblance to that of $CeInCu_2$, although C/T starts increasing at a somewhat higher temperature and shows saturation below 1 K in $CeInCu_2$.

To summarise, the present experimental results suggest that $CeRuSn_3$ is a new heavy-fermion compound. It shows no magnetic ordering down to 0.6 K. There still remain several questions to be answered in order to clarify the low-temperature behaviour. To this end, measurements on samples with controlled stoichiometry and at lower temperature are now in progress.

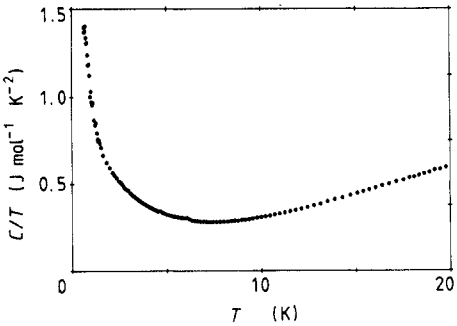

Figure 5. Temperature dependence of specific heat coefficient of $CeRuSn_3$ $C(T)$ divided by temperature.

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