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Transport properties in the filled-skutterudite compounds $\text{RERu}_4\text{Sb}_{12}$ (RE—La, Ce, Pr and Nd); an exotic heavy fermion semimetal $\text{CeRu}_4\text{Sb}_{12}$

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Abstract

We report transport measurements on high quality $\text{RERu}_4\text{Sb}_{12}$ single crystals under magnetic fields up to 14 T and temperatures down to 0.3 K. $\text{CeRu}_4\text{Sb}_{12}$ exhibits anomalous features in all the transport properties, compared to those for RE = La, Pr and Nd. The thermoelectric power (TEP) shows a huge positive peak over $100 \mu\text{V K}^{-1}$ near 90 K. The Hall coefficient, $\sim 2.0 \times 10^{-9} \text{ m}^3 \text{ C}^{-1}$ at room temperature, exhibits a drastic increase below ~ 100 K to $R_H \approx 5.2 \times 10^{-8} \text{ m}^3 \text{ C}^{-1}$ at 0.3 K. At low temperatures below ~ 3 K, the TEP exhibits an apparent anomaly correlated with the non-Fermi-liquid behaviour reported in the resistivity and specific heat. We have succeeded in observing Shubnikov–de Haas oscillations both in the magnetoresistance and Hall effect. This result along with the Hall effect suggests small semimetallic Fermi surfaces with highly correlated electrons at low temperatures in contrast with the large FS for $\text{LaRu}_4\text{Sb}_{12}$. $\text{NdRu}_4\text{Sb}_{12}$ exhibits anomalies suggesting a ferromagnetic ground state at around 1.4 K above which the resistivity exhibits a negative temperature coefficient up to 6 K.

1. Introduction

The filled-skutterudite compounds $\text{RETr}_4\text{Pn}_{12}$ (RE—rare earth; Tr—Fe, Ru, Os; Pn—pnictogen) first synthesized by Jeitschko and Braun [1] have aroused renewed interest recently from the viewpoints of both the exotic physical properties associated with the unique body-centred cubic structure [2–6], and the potential as thermoelectric materials for the next generation [7–9]. Strong hybridization between 4f electrons and conduction electrons, associated with the unique crystal structure containing RE surrounded by 12 Pn and eight Tr ions, is expected to cause exotic phenomena. In fact, for $\text{RERu}_4\text{Pn}_{12}$ with a small lattice constant, the strong hybridization resulting from the large coordination number is thought to

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be an origin of the exotic behaviours such as the metal–insulator transition in $\text{PrRu}_4\text{P}_{12}$ [2] and the extraordinary heavy electrons in $\text{PrFe}_4\text{P}_{12}$ [3–5] which are unusual as Pr compounds. For Ce compounds such as $\text{CeRu}_4\text{P}_{12}$ and $\text{CeFe}_4\text{P}_{12}$, the stronger *c*–*f* hybridization leads to the Kondo semiconducting behaviours [10, 11]. In contrast, for $\text{RETr}_4\text{Sb}_{12}$ with larger lattice constants than $\text{RETr}_4\text{P}_{12}$, weaker *c*–*f* hybridization is expected; namely the anomalous hybridization effect is not confirmed in $\text{PrRu}_4\text{Sb}_{12}$ but is only realized in $\text{CeRu}_4\text{Sb}_{12}$. In fact, Takeda and Ishikawa [10] reported that $\text{PrRu}_4\text{Sb}_{12}$ is an ordinary superconductor exhibiting no exotic behaviours, while $\text{CeRu}_4\text{Sb}_{12}$ is a metal exhibiting non-Fermi-liquid (NFL) behaviours in the electrical resistivity and the specific heat at low temperatures. Recently, Bauer *et al* [11] reported large sample dependence of electrical resistivity with some specimens exhibiting NFL behaviour. Based on the magnetoresistance (MR) measurement, they concluded that magnetic field up to 8 T does not significantly change the NFL ground state. In contrast, Takeda and Ishikawa [12] reported the NFL behaviour to be suppressed above ~ 3 T. $\text{LaRu}_4\text{Sb}_{12}$ and $\text{NdRu}_4\text{Sb}_{12}$ are reported to exhibit a superconducting transition at 3.58 K and some magnetic order below 1.3 K, respectively [10].

For all the $\text{RERu}_4\text{Sb}_{12}$, the transport measurements have been limited mostly to the electrical resistivity until now. In this paper, we report the extended study of electrical resistivity (ρ), and the first study of thermoelectric power (TEP) (S) and Hall coefficient (R_H) on high quality single crystals to deepen the understanding of $\text{RERu}_4\text{Sb}_{12}$.

2. Experimental details

High quality single crystals of $\text{RERu}_4\text{Sb}_{12}$ (RE—La, Ce, Pr, Nd) were grown by the Sb-flux method starting from a composition of RE:Ru:Sb = 1:4:20 using raw materials 4 N (99.99%) RE (La, Ce, Pr, Nd), 4 N Ru and 6 N Sb [1, 4]. By x-ray powder diffraction, we confirmed that lattice constants agree with the reported values [6], and the absence of impurity phases within the experimental accuracy.

ρ and R_H were measured by the ordinary dc four-probe method. S was measured by the differential method using Au–0.07% Fe versus Chromel thermocouples. The voltage measurements were made by Keithley 182 nanovoltmeters. In order to reduce the heating effect, samples were directly immersed in liquid ^3He in the MR and Hall resistivity (ρ_H) measurements using an Oxford Instrument top loading ^3He cryostat, down to 0.3 K and up to 14 T. The magnetic measurements were made by a Quantum Design SQUID magnetometer up to 5.5 T.

The residual resistivity ratio (RRR) is ~ 50 , 10 and 100 for $\text{LaRu}_4\text{Sb}_{12}$, $\text{CeRu}_4\text{Sb}_{12}$ and $\text{PrRu}_4\text{Sb}_{12}$, respectively. RRR for $\text{NdRu}_4\text{Sb}_{12}$ was estimated to be more than 100 from the temperature dependence of resistivity under 1.5 T.

3. Results and discussion

3.1. Electrical resistivity

The temperature dependence of electrical resistivity $\rho(T)$ for $\text{LaRu}_4\text{Sb}_{12}$ and $\text{PrRu}_4\text{Sb}_{12}$ is shown in figure 1, which qualitatively agrees with the previous report [10]. The solid curve shows the Bloch–Grüneisen (BG) fit using the Debye temperature (θ_D) of 195 K for comparison with $\rho(T)$ of $\text{LaRu}_4\text{Sb}_{12}$. The deviation of $\rho(T)$ from the BG law is within a reasonable range, taking into account only a single value of θ_D used in the fitting. The superconducting transition temperature (T_c) estimated from the resistivity on two different samples for each RE agree as 3.42 ± 0.02 K for $\text{LaRu}_4\text{Sb}_{12}$ and 1.08 ± 0.06 K for $\text{PrRu}_4\text{Sb}_{12}$ which are slightly higher

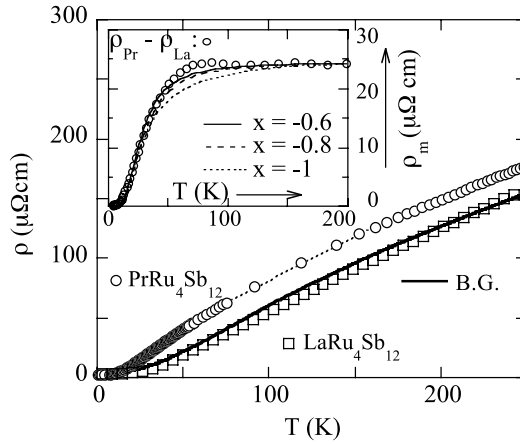


Figure 1. Temperature dependence of the electrical resistivity for LaRu₄Sb₁₂ and PrRu₄Sb₁₂. The inset shows the magnetic part of the electrical resistivity ρ_m for PrRu₄Sb₁₂ estimated by subtracting the phonon part of the resistivity using $\rho(T)$ for LaRu₄Sb₁₂. To compensate the error due to the geometrical difference between the samples, the electrical resistivity for PrRu₄Sb₁₂ was normalized so that $d\rho/dT$ agrees with that in LaRu₄Sb₁₂ above 200 K.

than those in [10]. Hence, only the characteristic features of each compound are summarized below.

The magnetic resistivity $\rho_m(T)$ of PrRu₄Sb₁₂, estimated by subtracting the phonon contribution using the temperature dependent resistivity of LaRu₄Sb₁₂, increases sharply with increasing temperature and tends to saturate near 80 K (inset in figure 1). The temperature dependence can be ascribed to the increasing magnetic conduction-electron scattering associated with crystalline electric field (CEF) excitation [13], which tends to saturate when the thermal energy $k_B T$ is comparable with the overall CEF splitting. In the following analysis, we assume that the O_6^2 – O_6^6 term, pointed out by Takegahara *et al* [14] to exist in the filled-skutterudite structure, is negligibly small, and we use the ordinary O_h -type point-group notation for CEF levels described by Lea *et al* [15]. In this notation, the two CEF parameters are represented by x and W , and the ninefold-degenerate $J = 4$ Hund multiplet of the Pr³⁺ ion on the cubic site splits into a singlet (Γ_1), a nonmagnetic doublet (Γ_3) and two triplets (Γ_4 and Γ_5). Takeda and Ishikawa [10] proposed a singlet ground state and estimated $\Delta_{14} \approx 70$ K from the magnetic susceptibility at 0 K $\{\chi(0)\}$, where Δ_{ij} refers to the energy separation between the Γ_i and Γ_j states. By taking into account the $\rho_m(T)$ data further, we can estimate Δ_{15} which is hardly determinable only from the $\chi(T)$ data because of the insensitivity of $\chi(T)$ to Δ_{15} [10]. Fixing the value of Δ_{14} to be 70 K, we fit the $\rho_m(T)$ data using the following model [13]:

$$\rho = \rho_0 \text{tr}(PQ)$$

$$P_{ij} = \exp(-\beta E_i) \{ \sum_k \exp(-\beta E_k) \}^{-1} \beta (E_i - E_j) \{ 1 - \exp[-\beta (E_i - E_j)] \}^{-1} \quad (1)$$

$$Q_{ij} = |\langle i | J_z | j \rangle|^2 + 2^{-1} |\langle i | J_+ | j \rangle|^2 + 2^{-1} |\langle i | J_- | j \rangle|^2,$$

where $\beta = k_B T$, and the trace and the summation are made over $2J + 1$ CEF states. The best fitting is obtained with the CEF parameter $-0.8 \leq x \leq -0.6$, where the CEF level scheme is Γ_1 (0 K)— Γ_4 (70 K)— Γ_5 (90–140 K)— Γ_3 (120 K) as shown in the inset of figure 1. Note that if x is larger than -0.5 , there appears a maximum in $\chi(T)$, which is inconsistent with the experiment. On the other hand, if x is lower than -0.8 , the shoulder appearing around 70 K in $\rho_m(T)$ becomes too broad.

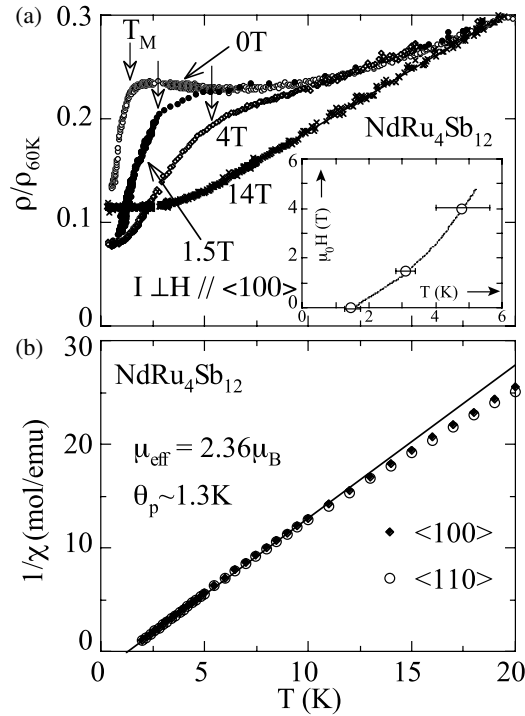


Figure 2. Temperature dependence of (a) the resistivity normalized at 60 K under selected magnetic fields, and (b) the inverse susceptibility for NdRu₄Sb₁₂. The inset shows the field versus temperature plot for the kink in resistivity.

Figure 2 shows the temperature dependence of resistivity normalized at 60 K $\{\rho(T)/\rho_{60\text{ K}}\}$ under selected values of magnetic field for NdRu₄Sb₁₂, which has not been reported before. In zero field, $\rho(T)/\rho_{60\text{ K}}$ decreases with decreasing temperature down to ~ 6 K, where it starts to increase after showing a minimum. Below a faint maximum at ~ 2 K, it decreases down to 0.3 K with no tendency to saturate. The decrease below ~ 2 K reflects the decrease of conduction electron scattering due to the magnetic order at $T_M \approx 1.4$ K inferred from the specific heat and the magnetic susceptibility measurements [10]. It should be noted that such a resistance minimum has been also observed in the ferromagnetic NdFe₄P₁₂, though the origin has not been clarified yet [3, 16].

Taking into account the magnetic field effect on resistivity in figure 2(a), i.e., the application of field

- (1) suppresses the resistivity,
- (2) shifts the shoulder in $\rho(T)$ to higher temperature and
- (3) makes the shoulder more blurred,

the magnetic ordering at 1.4 K should be ferromagnetic rather than antiferromagnetic. The Weiss temperature of ~ 1.3 K estimated by extrapolating the inverse magnetic susceptibility below 10 K is close to $T_M \approx 1.4$ K, consistent with the ferromagnetic correlation.

Figure 3 shows the temperature dependence of the magnetic part of the electrical resistivity in CeRu₄Sb₁₂. Compared to the other RERu₄Sb₁₂, the RRR of CeRu₄Sb₁₂ is smaller and somewhat sample dependent (8.3–12); however, the dependence is not so drastic as

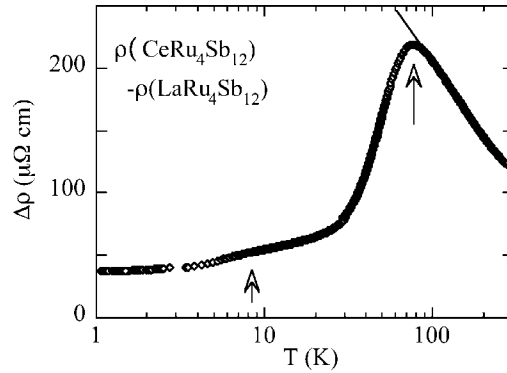


Figure 3. Temperature dependence of the magnetic part of the electrical resistivity for CeRu₄Sb₁₂.

in [11]. The electrical resistivity at RT is $\sim 300 \mu\Omega \text{ cm}$ in agreement with [11], while the overall temperature dependence is close to that in [10] and that of sample (2) in [11]. $\Delta\rho = \rho(\text{CeRu}_4\text{Sb}_{12}) - \rho(\text{LaRu}_4\text{Sb}_{12})$ exhibits a large peak at around 75 K above which it shows a clear $-\log(T)$ dependence suggesting Kondo scattering. The resistivity decrease below 75 K is ascribable to the development of coherence in the Kondo scattering. ρ for CeRu₄Sb₁₂ shows a tendency to saturate at around 10 K below which it shows NFL behaviour $\rho \propto T^{1.6}$ in agreement with [10–12]. Note that the temperature dependence below the maximum is not monotonic: i.e., a sudden change of curvature at ~ 30 K and a hump near 10 K.

3.2. Thermoelectric power

Figure 4 shows the temperature dependence of TEP S for RERu₄Sb₁₂ (RE—La, Ce, Pr and Nd). The TEP of LaRu₄Sb₁₂ is almost linear up to RT reflecting the dominance of the diffusion TEP except a small shoulder near 15 K. The relatively large temperature coefficient $dS/dT \sim 0.1 \mu\text{V K}^{-2}$ indicates the existence of high density of states $D(\varepsilon)$ near the Fermi energy, since S should be proportional to $\{\partial \ln D(\varepsilon)/\partial \varepsilon\}_{\varepsilon_F}$ in the simplest model [17]. In fact, a sharp peak is obtained near E_F in the band structure calculation [18]. The positive sign of S suggests that the peak is located below ε_F . The overall behaviour for RE = Pr and Nd is not very different from that for La, indicating basically the same electronic structure as inferred from the localized character of 4f electrons in these compounds. The complex structures below 30 K depending on RE may be due to the phonon drag or magnon drag contribution, which is not discussed in this paper. The shoulder in S for PrRu₄Sb₁₂ near 70 K, close to the saturation temperature of magnetic resistivity, might also reflect the conduction-electron scattering contribution associated with the CEF excitation.

Compared to the other RE, the TEP for CeRu₄Sb₁₂ is very different; S has a large peak $\sim 130 \mu\text{V K}^{-1}$ near 90 K below which it decreases with decreasing temperature showing a plateau near 30 K correlated with that in $\rho(T)$, and finally it shows an upturn below ~ 2 K. The overall behaviour of $S(T)$ does not depend on sample, although the absolute value is somewhat sample dependent probably due to the limited size of the samples. It is noted that the TEP and the resistivity have resemblance with those in the high T_K Kondo compound CePd₃ [19], except the low temperature upturn in TEP that may be related with the NFL behaviour found in ρ and C/T . Unfortunately, no systematic study to be compared has been reported on the

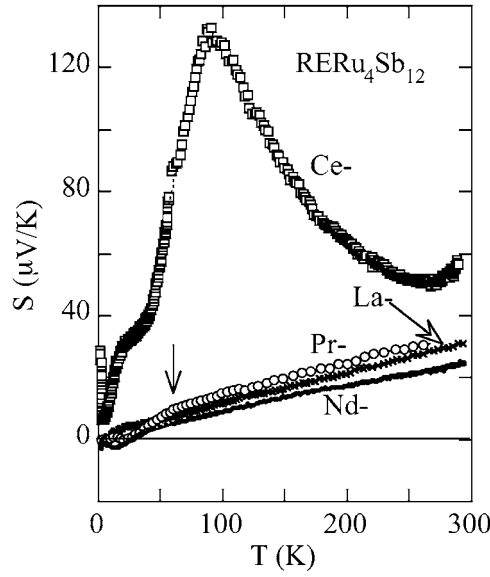


Figure 4. Temperature dependence of the TEP for $\text{RERu}_4\text{Sb}_{12}$ (RE—La, Ce, Pr and Nd).

TEP related with the NFL behaviours except one on $\text{Y}_{1-x}\text{U}_x\text{Ru}_2\text{Si}_2$. A sharp minimum in TEP reported on that compound near 8 K [20], where NFL behaviours are evident in the other physical properties, might have same origin [21]. In order to understand TEP related with NFL, more systematic studies on the compounds exhibiting NFL behaviours are necessary.

3.3. Hall effect

Figure 5 shows the temperature dependence of R_H in $\text{RERu}_4\text{Sb}_{12}$. R_H for $\text{LaRu}_4\text{Sb}_{12}$ is $2.5 \times 10^{-9} \text{ m}^3 \text{ C}^{-1}$ at 4.2 K and only weakly temperature dependent. Assuming a single-band model, the carrier concentration is about 1 hole/fu at RT. That is consistent with a band structure calculation, where two hole Fermi surfaces (a small spherical one in the 47th bands and a multiply connected one in the 48th bands) are predicted [18].

At 4.2 K, R_H for $\text{PrRu}_4\text{Sb}_{12}$ and $\text{NdRu}_4\text{Sb}_{12}$ is 2.2×10^{-9} and $2.7 \times 10^{-9} \text{ m}^3 \text{ C}^{-1}$, respectively. For both compounds, the Hall coefficient is slightly temperature dependent, which could be ascribed to the anisotropy (the wavevector dependence) in the relaxation time. These facts also indicate the similarity of the electronic structure and the well localized nature of 4f electrons in these compounds.

R_H for $\text{CeRu}_4\text{Sb}_{12}$ at RT is $\sim 2.3 \times 10^{-9} \text{ m}^3 \text{ C}^{-1}$, not very different from that in $\text{LaRu}_4\text{Sb}_{12}$, and slowly increases with decreasing temperature. Below ~ 80 K close to the peak temperature in $\rho_m(T)$, R_H increases sharply and reaches $5.2 \times 10^{-8} \text{ m}^3 \text{ C}^{-1}$ at 0.5 K, corresponding to the carrier concentration of 0.05 holes/fu in a single-band model. Such temperature dependence cannot be simply explained by the skew scattering model of conduction electrons. According to Fert and Levy [22], the Hall coefficient from the skew scattering is described by a general expression,

$$R_H(T) = R_{H0} + \gamma \chi'(T) \rho_r(T), \quad (2)$$

using the reduced magnetic susceptibility $\chi'(T)$ and the resistivity $\rho_r(T)$ due to the resonance scattering. The parameter γ takes different values above and below T_K depending on the

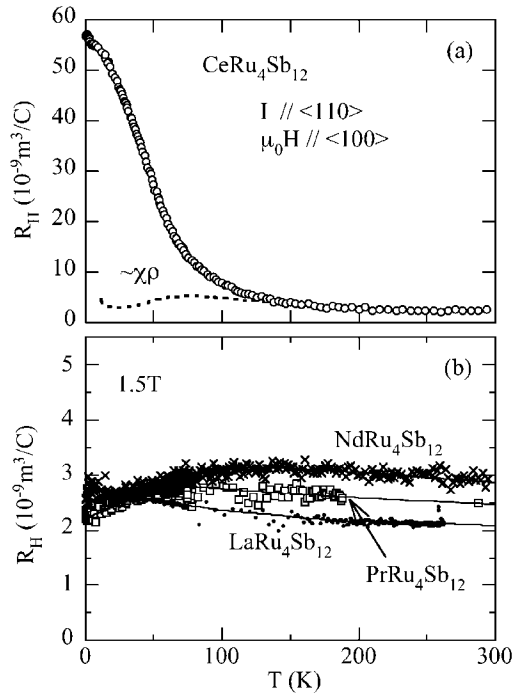


Figure 5. Temperature dependence of the Hall coefficient for (a) CeRu₄Sb₁₂ and for (b) RERu₄Sb₁₂ (RE—La, Pr and Nd). The broken curve in (a) shows the skew scattering contribution estimated by using the experimental magnetic susceptibility and electrical resistivity.

phase shift associated with the scattering channels. Even for the coherent state, they found that equation (2) accounts for the main feature of the Hall effect. The overall behaviours of $\rho(T)$ and $S(T)$ with a large peak at around 100 K in CeRu₄Sb₁₂ have similarities with those in the typical intermediate valence compound CePd₃ [19], the $R_H(T)$ of which can be roughly explained by equation (2). For CeRu₄Sb₁₂, however, equation (2) along with the experimental $\chi'(T)$ and $\rho_r(T)$ using a constant $R_{H0} = 1.7 \times 10^{-9} \text{ m}^3 \text{ C}^{-1}$ explains $R_H(T)$ only above 170 K, where the temperature dependence of R_H is minor. Especially below ~ 100 K, where both $\rho(T)$ and $\chi(T)$ shows a peak, R_H increases sharply in contradiction with the decrease expected from equation (2). From this viewpoint, some new mechanism must be taken into account to explain the $R_H(H)$ behaviours below ~ 100 K. The simplest explanation for the different temperature dependence of the Hall coefficient in CeRu₄Sb₁₂ compared to that in RERu₄Sb₁₂ (RE—La, Pr and Nd) is the temperature dependent change of Fermi surfaces in CeRu₄Sb₁₂. Based on the ultrahigh resolution photoemission spectra, Kanai *et al* [23] have recently reported a temperature dependent reduction of the electronic density of states at E_F in CeRu₄Sb₁₂, becoming noticeable below ~ 100 K. Including the temperature range, the dependence correlates well with the increase in R_H (figure 5). However, the more intense studies utilizing the techniques to investigate the FS at higher temperatures are necessary to confirm such a temperature dependent FS reconstruction.

3.4. High field magnetoresistance and Hall effect in CeRu₄Sb₁₂

The NFL behaviour is one of the most attractive features in CeRu₄Sb₁₂; however, even the sensitivity of the NFL feature against magnetic field has not been settled as described in

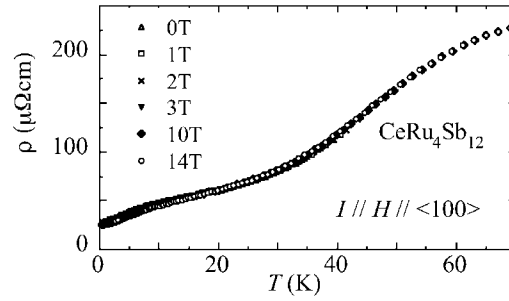


Figure 6. Temperature dependence of electrical resistivity for $\text{CeRu}_4\text{Sb}_{12}$ under selected magnetic fields in the longitudinal geometry.

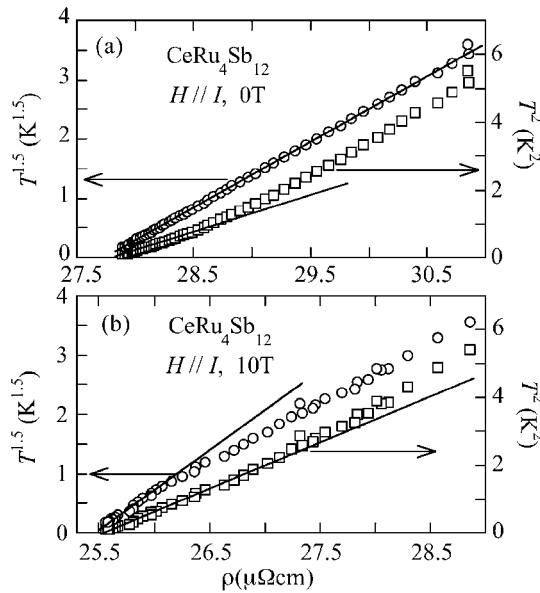


Figure 7. Electrical resistivity for $\text{CeRu}_4\text{Sb}_{12}$ as functions of $T^{1.5}$ and T^2 under (a) 0 T and (b) 10 T in the longitudinal geometry.

section 1. In order to settle the problem, we have measured the MR and Hall resistivity of $\text{CeRu}_4\text{Sb}_{12}$ up to $\mu_0 H = 14$ T and down to $T = 0.3$ K.

Firstly, we have measured the magnetic field effect on the temperature dependence of resistivity in the longitudinal geometry up to 70 K as shown in figure 6, since Bauer *et al* [11] have reported an exotic huge magnetic field effect on MR over a wide temperature range up to RT. On all the samples we prepared, we have not succeeded in reproducing such a huge magnetic field effect in the longitudinal geometry. For the transverse geometry (not shown), positive MR up to $10 \mu\Omega \text{ cm}$ at 0.3 K has been observed. However, it decreases with increasing temperature, which could be ascribed to the ordinary magnetoresistance due to the electrons' cyclotron motion. In the longitudinal geometry, the negative MR predominates over the positive MR except under a limited condition described later. We have no definitive answer for the origin of such a huge field effect as reported by Bauer *et al*, which may be ascribed to some extraordinary sample dependence.

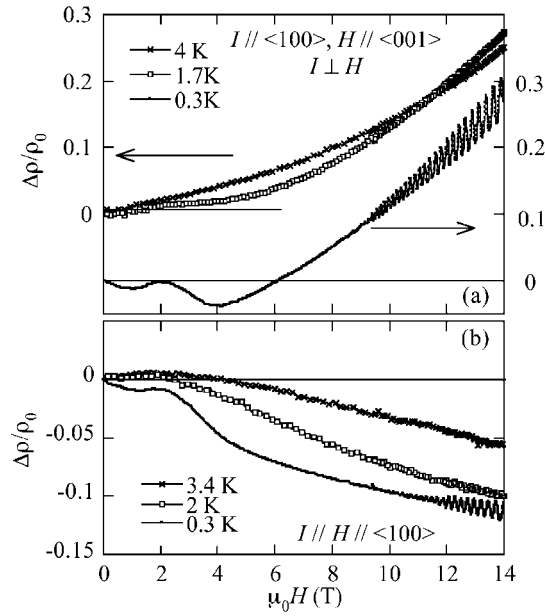


Figure 8. MR $\Delta\rho/\rho_0$ for CeRu₄Sb₁₂ at selected temperatures for both (a) transverse and (b) longitudinal geometry.

In order to clarify the effect of magnetic field on the NFL behaviour in the electrical resistivity, $\rho(T)$ is plotted against both T^2 and $T^{1.5}$ in figure 7. In zero field, $\rho(T)$ follows a $T^{1.5}$ dependence in a wider range in agreement with the reported data [10, 11]. The effect of magnetic field is not so drastic, although the T^2 dependence better fits the experiment data below ~ 1.5 K under 10 T.

Figure 8 shows the field dependence of MR for both (a) the transverse and (b) longitudinal geometries. At 4 K, the MR ratio ($=[\rho(H) - \rho(0)]/\rho(0)$) of $\sim 25\%$ under 14 T for transverse geometry is extraordinarily large compared to ordinary metals, taking into account the large residual resistivity ($>35 \mu\Omega$ cm). This fact indicates CeRu₄Sb₁₂ to be a semimetal with only small Fermi surfaces. In fact, it is further manifested in the clear Shubnikov–de Haas (SdH) oscillation described below. The transverse MR at 4 K increases almost monotonically up to 14 T, which is explainable as an ordinary MR. At lower temperatures, however, an anomalous field dependence appears below ~ 4 T; a faint peak near 2 T and a succeeding minimum in ρ are clear especially at 0.3 K in figure 8(a). The recovery to the FL behaviour, T^2 -dependence of the electrical resistivity from the NFL occurs across the field range 2–4 T. In the transverse MR of the f-electron systems, two contributions generally coexist: namely ordinary positive MR and the negative MR resulting from the field induced suppression of magnetic fluctuations. Usually, the latter dominates at higher temperatures due to larger thermal fluctuations of the magnetic moments, while the former increases with decreasing temperature reflecting the growing cyclotron motion of electrons. In contrast, CeRu₄Sb₁₂ behaves in the other way; the negative MR at low fields grows with decreasing temperature as clearly seen for 0.3 K in figure 8. We inferred that the negative MR is closely correlated with the magnetic instability related with the NFL behaviour. The competing contributions from the positive and negative MR components make the simple temperature exponent analysis impossible, which may be the reason for the disagreement between [11] and [12]. MR geometry in [12] is inferred to

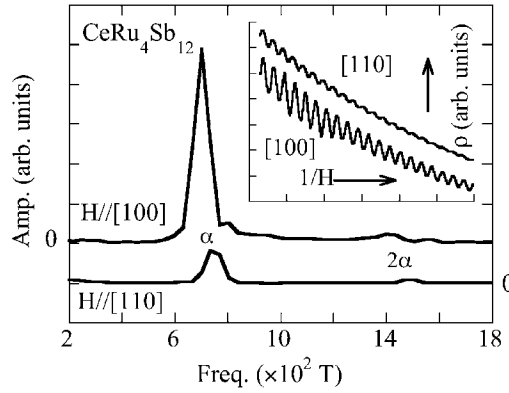


Figure 9. Fast frequency transform (FFT) spectra of SdH oscillation for $\text{CeRu}_4\text{Sb}_{12}$ for $H \parallel [100]$ and $H \parallel [110]$. The inset shows the resistivity plotted against $1/H$.

be longitudinal though it was not clearly mentioned in the text, while it is transverse in [11]. That is the main reason for the different experimental results between the two works. The longitudinal MR is simpler to extract the temperature dependence of the conduction electron scattering under a constant field, since the ordinary Lorentz MR is smaller. The negative longitudinal MR in figure 8(b) indicates that the conduction electron scattering resulting from some magnetic origin still exists even at 14 T and 0.3 K. The larger ordinary positive MR masks such a contribution in the transverse geometry. At high fields, the positive transverse MR contribution $\rho_{\perp} - \rho_{\parallel}$ (subtracting the magnetic contribution using the resistivity in the longitudinal geometry) varies approximately as H^n ($n \sim 1.5$) with a little angular dependence. These facts indicate $\text{CeRu}_4\text{Sb}_{12}$ to be a compensated metal with no open orbit.

Figure 9 shows the SdH oscillations as a function of $1/H$ for $H \parallel [100]$ and $[110]$, along with their FFT (fast Fourier transform) analysis [24]. For both field directions, only a single frequency branch is observed except their second harmonic branch; $F = 7.1 \times 10^2$ T for $[100]$ and 7.5×10^2 T for $[110]$, respectively. From the frequency, the extremum cross-sectional area of the FS perpendicular to H is estimated from $A_{ext} = \{2\pi e/\hbar\}F$ as 2% of the Brillouin zone size (~ 0.04 holes/fu) assuming a spherical FS. The carrier number is close to that estimated from the Hall coefficient. The corresponding branch has not been detected in dHvA experiment on $\text{LaRu}_4\text{Sb}_{12}$ [25], while any branches observed in $\text{LaRu}_4\text{Sb}_{12}$ have not been detected in the present experiment. This apparent difference in FS between the two compounds indicates that conduction electrons in $\text{CeRu}_4\text{Sb}_{12}$ are highly hybridized with 4f electrons in accord with the band structure calculation [18].

The effective mass (m^*) is estimated to be $\sim 5 m_0$, several times larger than those found in $\text{LaRu}_4\text{Sb}_{12}$ [25] despite the smaller FS cross-section, reflecting the highly correlated electrons in $\text{CeRu}_4\text{Sb}_{12}$. Using thus determined m^* and FS volume, based on a single spherical hole FS, the specific heat coefficient is estimated to be $\gamma \approx 10 \text{ mJ mol}^{-1} \text{ K}^{-2}$ that is still an order of magnitude smaller than the experimental value [10] of $\gamma \approx 100 \text{ mJ mol}^{-1} \text{ K}^{-2}$. These facts suggest the existence of an order of magnitude heavier electron FS that compensates the observed hole FS. SdH oscillations for such a heavy FS may be hardly detectable within the present experimental conditions; however, some fingerprint for such an electron FS appears in the field dependence of R_H in figure 10. The SdH oscillation is also clearly seen in R_H , shown in the inset. The gradual decrease of R_H up to 14 T could be evidence for the heavier electron FS whose dominance increases with increasing field. In the simplest two-band model

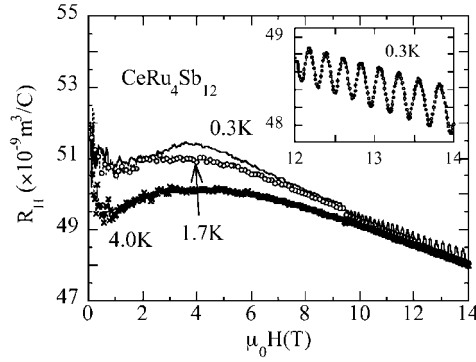


Figure 10. Field dependence of Hall coefficient for CeRu₄Sb₁₂ at selected temperatures. The inset shows the enlarged view in high fields at 0.3 K.

for a compensated semimetal, irrespective of the high field or low field condition, R_H can be presented as

$$R_H = (1/nec)\{(\mu_e^2 - \mu_h^2)/(\mu_e + \mu_h)^2\}, \quad (3)$$

where $\mu_h = \tau_h e/m_h$ and $\mu_e = \tau_e e/m_e$ are the mobilities of holes and electrons, respectively. In real compensated metals and semimetals, R_H is field dependent, which results from the deviation from a spherical FS along with the wavenumber (k) dependence of the effective mass and relaxation time. At low fields, R_H is determined as the Fermi surface integration of the local FS curvature weighted with the k -dependent relaxation time [26]. Thus, the contribution from the lighter effective mass region dominates R_H even within a single Fermi sheet. With increasing field ($\omega\tau$ —cyclotron frequency times conduction electron's relaxation time), the increased averaging path along the cyclotron orbits leads to the change in the effective mobility, resulting in the field dependence of R_H [26, 27]. In the present case, the large difference in the effective mass between the lighter holes and the heavier electrons gives R_H dominated by the hole FS. Taking into account the SdH oscillation visible above ~ 4 T, the hole FS is in the high field condition. Thus, the field dependence of R_H in figure 10 at higher fields cannot be ascribed to the low to high field transition within the hole FS, but is indirect evidence for the presence of (an)other FS(s). The FS(s) may be (an) electron-like one(s) with a heavier effective mass which compensates the hole FS, since the MR experiment suggests CeRu₄Sb₁₂ to be a compensated semimetal with no open orbit. This scenario well explains the discrepancy between the specific heat coefficient in the experiment and the value estimated from only the hole FS determined by SdH experiment.

Recently, Goll *et al* [28] reported the temperature dependence of the FS in CeBiPt based on the SdH measurement, i.e., the change of frequency from ~ 35 T at 10 K to ~ 60 T at 0.4 K, which they ascribed to the temperature dependent c - f hybridization. In the present experiment, SdH oscillation is observable only below ~ 4 K due to the high mass, where no temperature dependence of FS was observed within the present experimental accuracy.

In summary, for all the RERu₄Sb₁₂ except RE = Ce, the transport properties are not very different, suggesting the well localized nature of 4f electrons. At high temperatures, the electrical resistivity and TEP for CeRu₄Sb₁₂ exhibit similarity with those in the typical Kondo compounds with high $T_K \approx 100$ K. The highly enhanced Hall coefficient at low temperatures indicates the reduced carrier density of 0.05 holes (electrons)/fu at 0.3 K. The SdH oscillation further evidences CeRu₄Sb₁₂ to be a heavy fermion semimetal at 0.3 K, in agreement with the Hall effect at low temperatures. Combining all these facts, we infer the FS of this compound to

be temperature dependent below ~ 100 K, which is consistent with the recent high resolution photoemission spectrum measurement.

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