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Electronic and magnetic investigation of the filled skutterudite compound $CeRu_4Sb_{12}$

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Abstract

We synthesized the filled skutterudite $CeRu_4Sb_{12}$ in single-crystal form using a molten-metal-flux technique with Sb flux. The specific heat and magnetic susceptibility of $CeRu_4Sb_{12}$ are well described by a logarithmic divergence or a power law in temperature indicating non-Fermi-liquid behaviour at low temperatures. The electrical resistivity is sample dependent with some specimens exhibiting non-Fermi-liquid behaviour below $T \sim 5$ K in which $\rho \propto T^n$ with $n \sim 1.4$. The application of magnetic fields up to $H = 80$ kOe does not significantly change the non-Fermi-liquid ground state.

1. Introduction

Strong electronic correlations in lanthanide and actinide materials give rise to a wealth of novel types of phenomenon. In heavy-fermion compounds, the small energy scale (∼1–10 K) associated with interaction between the f and conduction electrons is believed to produce the large effective masses ($m^* \sim 10^2$–$10^3\ m_e$) observed in these systems (for a review see [1–4]). Their physical properties correspond to those of localized non-interacting magnetic moments at high temperatures and a strongly interacting Fermi liquid at low temperatures. There are, however, a large number of f-electron heavy-fermion compounds which do not exhibit Fermi-liquid behaviour at low temperatures [5–8]. Most of these non-Fermi-liquid (NFL) materials are Ce-, Yb-, or U-based intermetallics in which long-range magnetic order has been suppressed by the substitution of a non-magnetic species or by the application of hydrostatic pressure [9–11]. Their physical properties at low temperature are characterized by weak power-law or logarithmic divergences in temperature of quantities such as the specific heat ($C/T \propto \ln T$ or T^n), magnetic susceptibility ($\chi \propto \ln T$ or T^n), and electrical resistivity ($\rho \propto T^n$ with $n < 2$). As there appear to be many routes to this NFL behaviour, a number of theories with distinct microscopic origins have been put forward including: a multichannel

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Kondo effect [12, 13], fluctuations of an order parameter near a $T = 0$ K second-order phase transition [14–16], a disorder-induced distribution of Kondo temperatures [17–19], and the fluctuations of an inhomogeneous Griffiths' phase [20].

The filled skutterudites RT_4X_{12} (R = alkaline earth, rare earth, or actinide; T = Fe, Ru, or Os; X = P, As, or Sb), crystallize in the $LaFe_4P_{12}$ BCC structure [21, 22] and exhibit an extraordinary number of strongly correlated electronic ground states [23–31]. Recently, Takeda and Ishikawa observed NFL behaviour in the filled skutterudite $CeRu_4Sb_{12}$ [32, 33] which is particularly interesting as most non-Fermi-liquid materials are disordered alloys (e.g., $Y_{1-x}U_xPd_3$ [34], $CeRhRuSi_2$ [35]).

In this paper, we report on a study of the physical properties of the filled skutterudite $CeRu_4Sb_{12}$ by means of x-ray diffraction, electrical resistivity, magnetoresistivity, dc magnetization, and specific heat measurements.

2. Experimental details

Single crystals of the filled skutterudite compounds RRu_4Sb_{12} (R = La, Ce) were grown from a Sb flux by placing the starting materials (R: AMES Laboratory 4N; T: Colonial Metals, 3N5; Sb: Alfa Aesar, 6N) in the ratio R:T:Sb = 1:4:20 in an evacuated carbon-coated quartz tube. The tubes were heated to 950 °C for 24 hours and then cooled slowly (∼3 °C h^{-1}) to 600 °C; this was followed by a quench to room temperature. The single-crystal specimens were removed from the excess antimony flux by etching in aqua regia. The crystals were usually cubic or rectangular in shape and ranged from 0.1 to 2 mm in diameter.

X-ray powder diffraction measurements were made using a 9 kW Rigaku diffractometer to check the sample purity and crystal structure. A silicon standard was used for a more accurate determination of the lattice parameter. The compounds $LaRu_4Sb_{12}$ and $CeRu_4Sb_{12}$ were found to crystallize in the $LaFe_4P_{12}$ structure ($Im3$) and the lattice parameters were in agreement with the previously reported values [22] (the results are collected in table 1). The electrical resistivity was measured in a commercial Quantum Design PPMS cryostat in a transverse geometry in magnetic fields up to 8 T at temperatures 1.8 K $\leqslant T \leqslant$ 300 K using a standard four-wire technique with excitation currents from 1 to 10 mA parallel to the ⟨100⟩ direction. Measurements of the electrical resistivity from 0.05 K to 1.8 K were made in an Oxford ^{3}He–^{4}He dilution refrigerator with a Linear Research LR 700 low-dissipation ac resistance bridge operating at a frequency of 12 Hz and an excitation current of 500 μA. The measurements were performed by stabilizing the temperature against a germanium or RuO_2 thermometer and averaging the sample resistance for ∼60 s. Specific heat measurements were

Table 1. Physical properties of RRu_4Sb_{12} (R = La, Ce). a: cubic lattice parameter; V: unit-cell volume; μ_{eff}: effective moment; θ: Curie–Weiss temperature; γ: electronic specific heat coefficient; θ_D: Debye temperature; T_K: Kondo temperature; χ_0: Pauli susceptibility; R_W: Wilson ratio. The values of the effective moment μ_{eff} and Curie–Weiss temperature θ were obtained from fits of the high-temperature magnetic susceptibility. The electronic specific heat coefficient γ and Debye temperature θ_D were determined from fits to the low-temperature specific heat. The Kondo temperature T_K and Wilson ratio R were calculated using the parameters χ_0 and γ at $T = 1.8$ K as discussed in the text. Errors in the last digit are given in parentheses.

	a (Å)	V (Å^3)	μ_{eff} (μ_B)	θ (K)	γ (mJ mol^{-1} K^{-2})	θ_D (K)	T_K (K)	χ_0 (cm^3 mol^{-1})	γ(1.8 K) (mJ mol^{-1} K^{-2})	R_W
$CeRu_4Sb_{12}$	9.270(1)	796.5(2)	2.26	−37	73	262	101	0.008	103	2.6
$LaRu_4Sb_{12}$	9.273(1)	797.3(2)	—	—	39	262	—	—	—	—

made on a collection of single-crystal specimens in a semi-adiabatic ^{3}He calorimeter using a standard heat-pulse technique. Magnetization measurements were made on a collection of single-crystal specimens in a Quantum Design MPMS magnetometer at temperatures between 1.8 K and 300 K and in magnetic fields up to 5.5 T.

3. Results

3.1. Magnetic susceptibility

The magnetic susceptibility $\chi \equiv M/H$, measured in a field of 1 kOe, versus T for $CeRu_4Sb_{12}$ is shown in figure 1. A broad maximum appears in $\chi(T)$ at 100 K followed by an upturn at low temperatures, which is similar to the behaviour of many intermediate-valence materials [36]. A least-squares fit to a Curie–Weiss law

$$\chi(T) = C/(T - \theta) \tag{1}$$

where $C = N_A\mu_{eff}^2/3k_B$, θ is the Curie–Weiss temperature, and μ_{eff} is the effective moment in Bohr magnetons, yields $\mu_{eff} = 2.26\ \mu_B$ and $\theta = -37$ K as displayed in the lower inset of figure 1. A different temperature-dependent effective moment, $\mu'_{eff}(T)$, defined as

$$\mu'_{eff}(T) \equiv \sqrt{\frac{3k_B\chi T}{N_A}} \tag{2}$$

is displayed in the upper inset of figure 1. For high temperatures, $\mu'_{eff}(T)$ approaches a constant value of 2.1 μ_B which is reduced from the free-ion Hund's rule multiplet value of

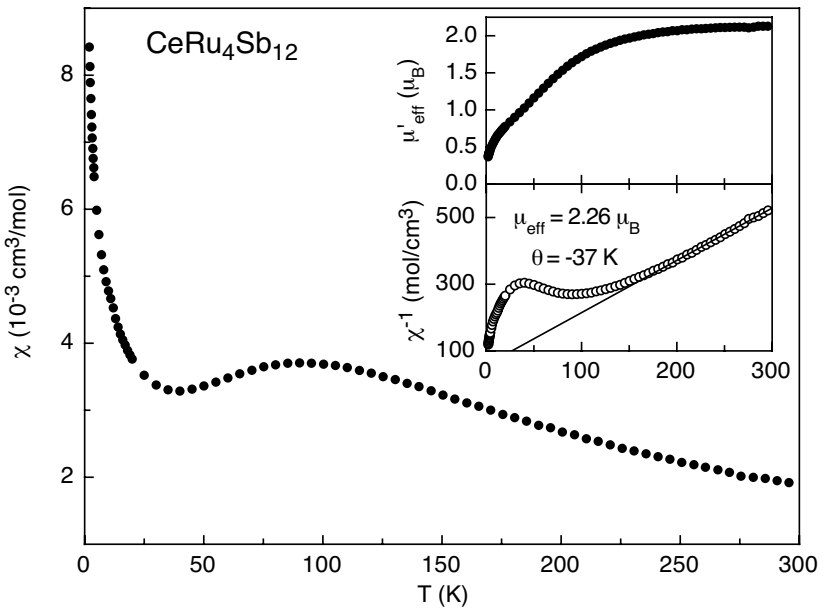

Figure 1. Magnetic susceptibility χ versus temperature T for $CeRu_4Sb_{12}$ in a magnetic field of $H = 1$ kOe. Upper inset: temperature-dependent effective moment $\mu'_{eff}(T) \equiv \sqrt{(3k_B\chi T/N_A)}$ versus T. Lower inset: inverse magnetic susceptibility χ^{-1} versus T. The line is a fit of a Curie–Weiss law $\chi(T) = C/(T - \theta)$ to the data.

2.54 μ_B. The effective moment $\mu'_{eff}(T)$ decreases for $T < 100$ K, indicating the influence of crystalline-electric-field or intermediate-valence effects.

The magnetic susceptibility of $CeRu_4Sb_{12}$ at low temperatures is shown in figure 2 on a log–log scale. $\chi(T)$ can be well described by a power law $\chi \propto aT^n$ from 1.8 K to 20 K. A least-squares fit to the data yields $a = -0.011$ cm^3 mol^{-1} K^{-n} and $n = -0.35$, as indicated by the solid line in figure 2. This type of non-Fermi-liquid behaviour is consistent with the Griffiths' phase model which predicts that

$$\chi(T) \propto T^{-1+\lambda}. \tag{3}$$

In the Griffiths' phase model, the low-temperature divergences in the physical properties (such as the magnetic susceptibility and specific heat) are universal functions of λ. The value of $\lambda_\chi = 0.65$ is comparable to the values obtained from fits of equation (3) to the $\chi(T)$ data at low temperatures for other non-Fermi-liquid materials such as $UCu_{5-x}M_x$, $Y_{1-x}U_xPd_3$, and $U_{1-x}Th_xPd_2Al_3$ [37]. The magnetic susceptibility can also be fitted by the expression

$$\chi(T) \propto -\ln(T) \tag{4}$$

but only over a limited temperature range of 1.8 K to ∼5 K. The upturn in $\chi(T)$ of $CeRu_4Sb_{12}$ at low temperatures could not be described by a Curie–Weiss law. In addition, we attempted to fit the data to the magnetic susceptibility of trivalent Ce for the case where the crystalline electric field (CEF) splits the sixfold-degenerate Ce $4f^1$ Hund's rule multiplet into a doublet and a quartet as was proposed, for example, for $Ce_{0.9}Fe_3CoSb_{12}$ [38]. However, fits to the CEF model were of poor quality, particularly in the low-temperature region at the maximum in χ^{-1} at 50 K; nor did the effective moment track either the expected moment for the doublet or quartet ground state. While it is possible that the increase in $\chi(T)$ at low temperatures is due to magnetic impurities, the results are also consistent with NFL behaviour that is observed in other measurements as described below.

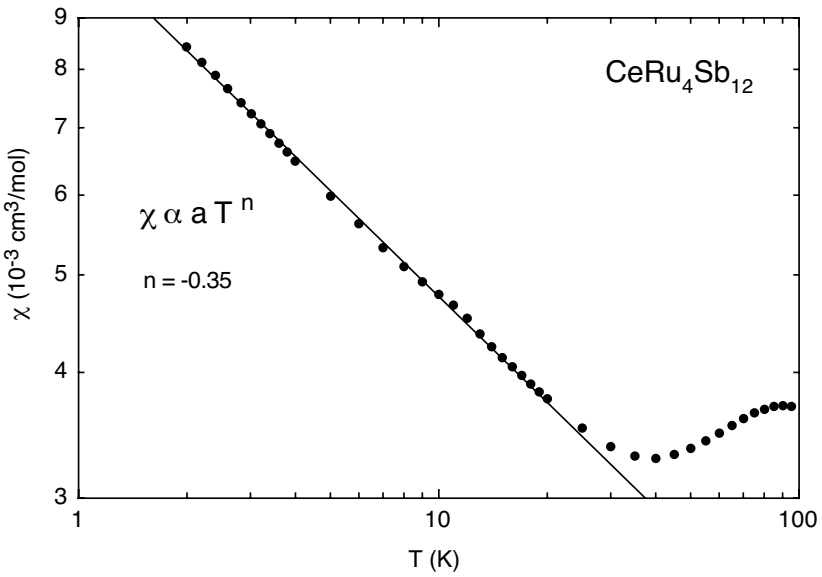

Figure 2. Magnetic susceptibility χ versus temperature T for $CeRu_4Sb_{12}$ on a log–log scale. The line is a fit of a power law $\chi \propto aT^n$ to the data from 1.8 K to 20 K with $n = -0.35$.

3.2. Specific heat

A plot of the specific heat divided by temperature C/T versus T^2 for $CeRu_4Sb_{12}$ and the non-magnetic analogue $LaRu_4Sb_{12}$ is shown in figure 3. The C/T versus T^2 data for $CeRu_4Sb_{12}$ and $LaRu_4Sb_{12}$ are similar between 4 K and 10 K but different below 4 K, with $LaRu_4Sb_{12}$ displaying a feature that is typical for a superconductor with a superconducting critical temperature $T_c = 3.75$ K and $CeRu_4Sb_{12}$ exhibiting an upturn in C/T below 2 K. For $LaRu_4Sb_{12}$, the electronic and phonon contributions to the specific heat were extracted from a least-squares fit to the data (shown as the solid line in figure 3) with the expression $C/T = \gamma + \beta T^2$, where γ is the electronic specific heat coefficient, $\beta = 12\pi^4 r R/(5\theta_D^3)$ is the Debye phonon coefficient, r (=17) is the number of atoms per formula unit, R is the universal gas constant, and θ_D is the Debye temperature. It was assumed that the phonon contributions to $C(T)$ of $CeRu_4Sb_{12}$ and $LaRu_4Sb_{12}$ are similar, so any changes in $C(T)$ are attributed to differences in electronic structure. The Sommerfeld coefficient $\gamma \equiv C/T$ for $CeRu_4Sb_{12}$ (with the same Debye temperature as $LaRu_4Sb_{12}$) is 73 mJ mol^{-1} K^{-2}, which classifies this compound as a moderately heavy-fermion material. The fit results are listed in table 1. An upturn in $C(T)/T$ that is linear in $-\ln T$ below 2 K is observed for $CeRu_4Sb_{12}$ which is evident in the semi-log plot of C/T versus T in the inset of figure 3. The two-channel spin-1/2 Kondo model [39–41] predicts that C/T should have the following form:

$$C/T \propto -(A'R/T_K)\ln(T/bT_K) \tag{5}$$

where A' (=0.251) is a constant and $b = 0.41$, leading to a residual entropy of $(R/2)\ln 2$. For many NFL materials the specific heat is described by equation (5) with the slope yielding an estimate of T_K that is comparable to values obtained from other measurements, even though the residual entropy often does not amount to $(R/2)\ln 2$ [5, 6]. Thus, we fit the

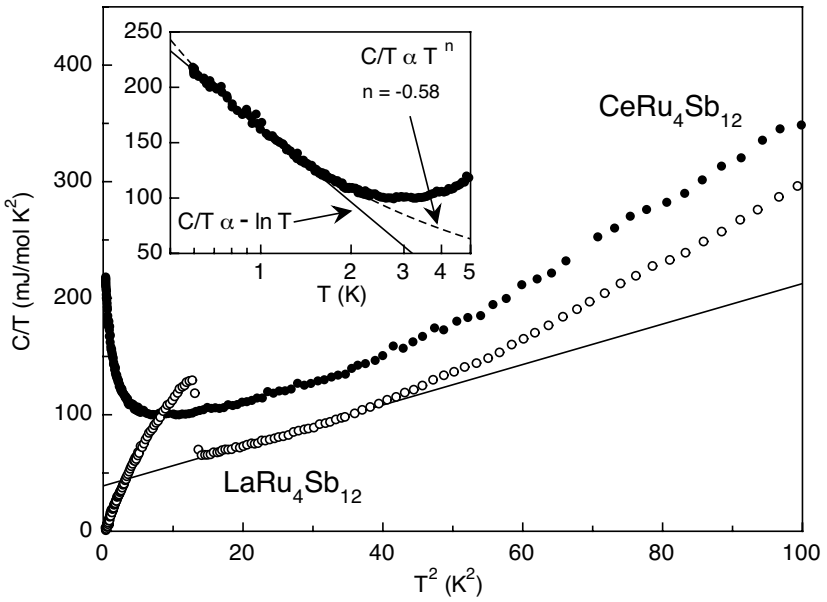

Figure 3. Specific heat divided by temperature C/T versus T^2 for $LaRu_4Sb_{12}$ (open circles) and $CeRu_4Sb_{12}$ (filled circles). The line is a fit of $C/T = \gamma + \beta T^2$ to the data. Inset: specific heat divided by temperature, C/T, versus T for $CeRu_4Sb_{12}$ on a semi-log scale. The solid line is a fit of $C/T \propto -\ln(T)$ to the data and the dashed line is a fit of $C/T \propto T^n$ to the data with $n = -0.58$.

C/T data of $CeRu_4Sb_{12}$ to the two-channel spin-1/2 Kondo model in a phenomenological way (without placing emphasis on the residual entropy) to equation (5) and obtain a Kondo temperature of $T_K = 21$ K. The specific heat of $CeRu_4Sb_{12}$ can also be described well over a similar temperature range by a power law $C/T \propto T^n$ with an exponent $n = -0.58$ as shown in the inset of figure 3. In the Griffiths' phase model, $n = -1 + \lambda$, for both the specific heat and magnetic susceptibility [20]. The value of λ obtained from specific heat, $\lambda_C = 0.42$, is not equal to the value obtained from the magnetic susceptibility, $\lambda_\chi = 0.65$. It is possible that better agreement between λ_C and λ_χ might be obtained if the measurements were performed on the same (large) single crystal rather than a collection of small crystals.

3.3. Electrical resistivity

The normalized electrical resistivity $\rho/\rho(300\ \mathrm{K})$ versus T is shown in figure 4 for five different samples of $CeRu_4Sb_{12}$. We estimate $\rho(300\ \mathrm{K})$ to be of the order of $\sim 300\ \mu\Omega$ cm. The resistivities of all the samples have a somewhat similar weak temperature dependence above 100 K, but exhibit markedly different behaviour below 100 K. Samples (1) and (2) show a knee at ~ 75 K, presumably due to the coherent scattering in the Ce-ion sublattice. This type of behaviour is also observed in another heavy-fermion skutterudite, $CeFe_4Sb_{12}$ [28], and also in the intermediate-valence skutterudite $YbFe_4Sb_{12}$ [29–31]. For temperatures below ~ 10 K, the resistivities of $CeRu_4Sb_{12}$ samples denoted as (1) and (2) decrease with decreasing temperature. The inset of figure 4 shows the normalized resistivity minus a constant term ρ_0 versus T on a log–log plot for both of these samples. A least-squares fit to the normalized resistivity

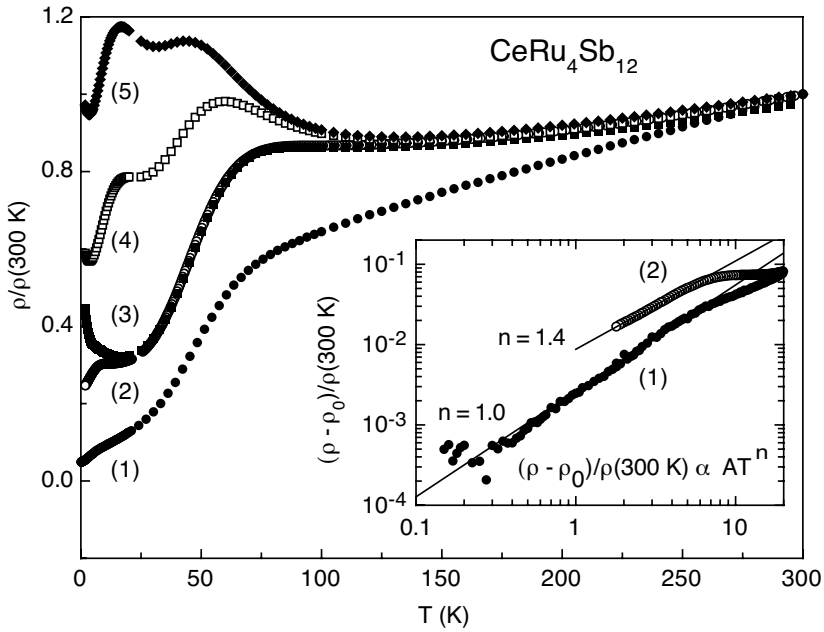

Figure 4. Normalized resistivity $\rho/\rho(300\ \mathrm{K})$ versus temperature T for five different samples of $CeRu_4Sb_{12}$. See the text for details. Inset: $(\rho - \rho_0)/\rho(300\ \mathrm{K})$ versus T on a log–log scale at low temperatures for samples (1) and (2). The lines are fits of $(\rho - \rho_0)/\rho(300\ \mathrm{K}) \propto AT^n$ to the data.

data of the form $(\rho - \rho_0)/\rho(300\ \mathrm{K}) \propto AT^n$ yields an exponent $n \simeq 1.4$ for sample (1) and $n \simeq 1.0$ for sample (2), which differs from Fermi-liquid theory which predicts $n = 2$ [42]. Thus, we observe non-Fermi-liquid behaviour in $\rho(T)$ for these two samples, similarly to Takeda and Ishikawa who found $\rho(T) \propto T^n$ with $n \sim 1.6$–1.7 [32, 33]. The behaviour of $\rho(T)$ for $CeRu_4Sb_{12}$, denoted as (3), which was previously studied by means of infrared reflectivity measurements in reference [43], is similar to that of samples (1) and (2), except that it exhibits an upturn below ∼10 K. This type of behaviour could be due to the presence of Kondo holes arising from uncompensated Ce moments in grain boundaries as proposed for $CeFe_4Sb_{12}$ which showed somewhat similar characteristics [44], or other impurities. Another possibility for the origin of the upturn at low temperatures is the presence of a small energy gap. Samples (4) and (5) exhibit more complicated behaviour below 100 K suggesting a delicate balance between several scattering mechanisms.

Measurements of the magnetoresistivity (MR) of $CeRu_4Sb_{12}$ sample (1) in magnetic fields up to 80 kOe are shown in figure 5. The sign of the MR is positive for all magnetic fields and shows the largest increase between 0 kOe and 20 kOe as displayed in the inset of figure 5. With increasing field, the knee in the resistivity at 75 K sharpens considerably and the $\rho(T)$ curves resemble those of sample (2) and reference [32] in zero field.

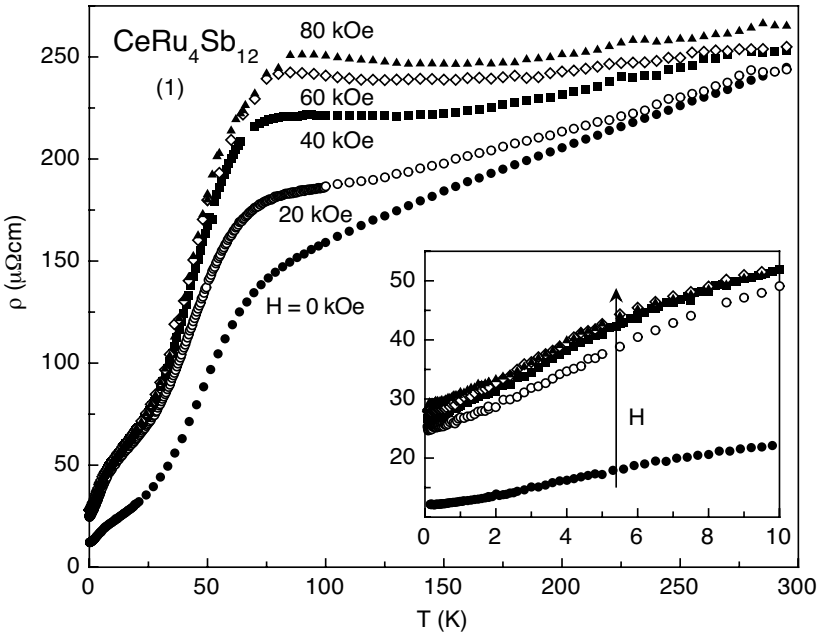

Figure 5. Electrical resistivity ρ versus temperature T for $CeRu_4Sb_{12}$ sample (1) in magnetic fields H up to 80 kOe. The current was applied parallel to the ⟨100⟩ direction and the magnetic field applied parallel to the ⟨001⟩ direction. Inset: an enlarged view of the magnetoresistivity of $CeRu_4Sb_{12}$ at temperatures below 10 K.

The MR of $CeRu_4Sb_{12}$ (1) at low temperatures below 20 K is shown in log–log plots of $\rho - \rho_0$ versus T in figure 6. The value of ρ_0 was chosen such that the curves are as linear as possible at the lowest temperatures. Least-squares fits to the expression $\rho - \rho_0 \propto AT^n$ in the range $0.2\ \mathrm{K} \lesssim T \lesssim 5\ \mathrm{K}$ yield exponents $n \sim 1.0$–1.5. Magnetic fields up to $H = 80$ kOe do not significantly alter the temperature dependence of ρ at low temperatures.

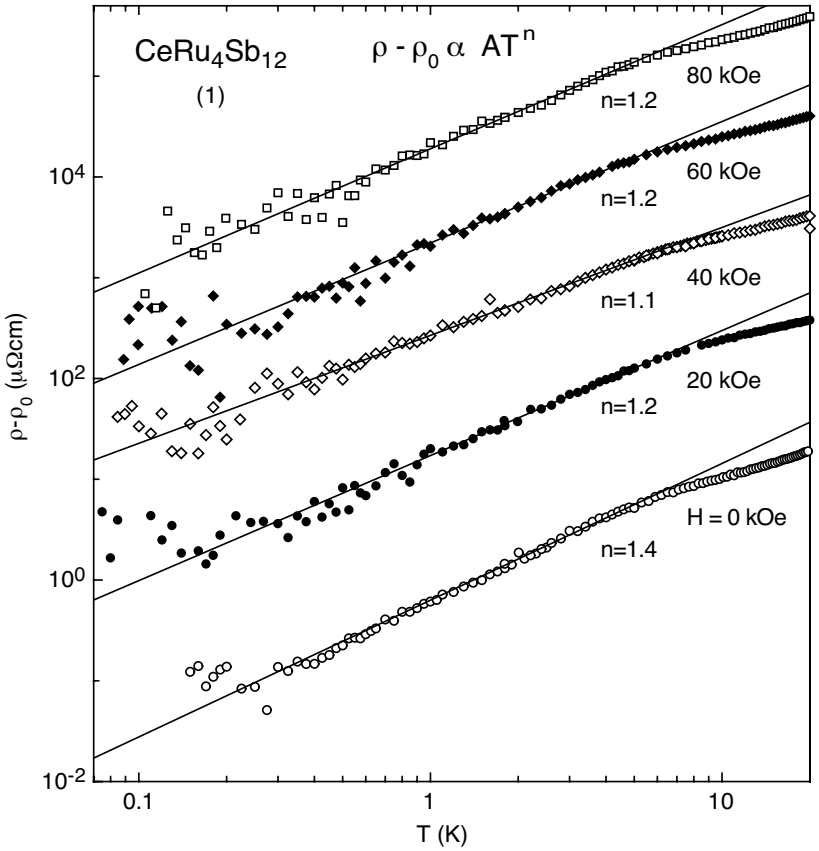

Figure 6. $\rho - \rho_0$ versus T for $CeRu_4Sb_{12}$ (1) in magnetic fields up to 80 kOe on a log–log scale. The curves have been shifted up by one decade from the one at the next-lowest field for clarity. The lines are fits to the data of the form $\rho - \rho_0 \propto AT^n$.

4. Discussion

In order to further describe the physical properties of $CeRu_4Sb_{12}$, we estimated the Kondo temperature T_K and the Wilson ratio R_W. The Kondo temperature was determined from the following formula:

$$T_K = \frac{N_A \mu_{eff}^2}{3 k_B \chi_0}. \tag{6}$$

Using the value of the Pauli susceptibility $\chi_0 = 0.008$ cm^3 mol^{-1} and the free-ion value of $\mu_{eff} = 2.54\ \mu_B$, we obtained the value $T_K \sim 100$ K. The value of T_K for the isostructural compound $CeFe_4Sb_{12}$, estimated the same way, was also ∼100 K [28]. The estimates of T_K for these materials seem reasonable in view of the maximum in $\chi(T)$ and the shoulder in $\rho(T)$ signalling the onset of coherence, both of which occur at ∼100 K. The Kondo temperature estimated from specific heat measurements, $T_K \sim 21$ K, is comparable to the value $T_K \sim 100$ K obtained from magnetic susceptibility given the fact that estimates of T_K often vary by about half an order of magnitude between different types of measurement. In order to determine whether the enhanced magnetic susceptibility and specific heat at low temperatures are due to

itinerant electrons, we calculated the Wilson–Sommerfeld ratio, $R_W = (\pi^2 k_B^2/3\mu_{eff}^2)(\chi_0/\gamma)$, which is generally close to unity for heavy-fermion systems. Since an unambiguous choice of γ for $CeRu_4Sb_{12}$ was not possible, we selected the value of $\gamma \equiv C/T$ at 1.8 K, which was the lowest temperature of the susceptibility measurements (the results are listed in table 1). We used the value of $\mu_{eff} = 2.54\ \mu_B$ appropriate for the Hund's rule $J = 5/2$ Ce multiplet. The Wilson–Sommerfeld ratio is of the order of unity ($R_W \sim 2.6$) indicating that the low-temperature physical properties of $CeRu_4Sb_{12}$ are associated with the moderately heavy itinerant electrons.

Our measurements on $CeRu_4Sb_{12}$ suggest that the transport properties are influenced greatly by a delicate balance between competing effects leading to a large variation in behaviour between samples at low temperatures. However, magnetic susceptibility and specific heat measurements made on a collection of single crystals are less sensitive to sample variations. It is worth mentioning that a $CeRu_4Sb_{12}$ single crystal (sample (3)), which has an upturn in $\rho(T)$ below 10 K and, hence, does not exhibit NFL behaviour in its transport properties, does, in fact, exhibit an increase below 2 K in $C(T)$ as seen in other collections of crystals (this work and reference [32]) and is consistent with NFL behaviour, but the mass of the sample ($\sim$10 mg) precludes a more quantitative analysis. This type of sample variation has been reported in other filled skutterudite compounds such as $CeFe_4P_{12}$ [45]. It seems plausible to classify $CeRu_4Sb_{12}$ as a NFL material given that the magnetic susceptibility, specific heat, and the electrical resistivity of at least some samples display NFL characteristics.

In most non-Fermi-liquid compounds, a magnetic–non-magnetic transition is induced by chemical substitution or by some other external parameter such as pressure or magnetic field. A number of NFL theories have different microscopic origins and can describe the behaviour of many of these systems with reasonable success. For instance, models involving a quantum phase transition [14, 15], in which the quantum fluctuations take the place of thermal fluctuations as the magnetic transition is tuned to $T = 0$ K, are thought to be most applicable to systems such as $CeCu_{6-x}Au_x$ [46]. Since many NFL materials are disordered alloys, various theories such as the Kondo disorder model [17–19] or Griffiths' phase model [20] have been proposed to describe the low-temperature behaviour of systems such as $UCu_{5-x}Pd_x$ [47, 48]. Other NFL theories, for instance that of the quadrupolar Kondo effect [13], are based upon the single-ion multichannel Kondo model [12]. These models (or variations of them) would, in principle, be applicable to systems in which single-ion scaling is observed as in the cases of $U_{1-x}Th_xPd_2Al_3$ [49, 50] and $Y_{1-x}U_xPd_3$ [34]. However, it is not clear which model would best describe the physical behaviour of $CeRu_4Sb_{12}$. For instance, there is no evidence as of yet to suggest that sufficient disorder exists in $CeRu_4Sb_{12}$ to produce the observed NFL behaviour via a Kondo disorder-type model. It is conceivable that $CeRu_4Sb_{12}$ lies close to a magnetic transition, thereby being most easily described by a quantum critical point model; further doping or pressure studies may be helpful in deciding whether or not this is the case. Takeda and Ishikawa substituted La into $CeRu_4Sb_{12}$ and found that the NFL properties disappeared by 10% La substitution and suggested that the coherence of the Ce sublattice was critical to the NFL state [33]. Thus, it is unclear which type of non-Fermi-liquid theory (if any) is most appropriate for $CeRu_4Sb_{12}$. Recently, there have been a few reports of NFL behaviour in stoichiometric compounds such as $CeNi_2Ge_2$ [51], UBe_{13} [13, 52], and $YbRh_2Si_2$ [53] in addition to $CeRu_4Sb_{12}$, so it is possible that a new class of ordered NFL materials are emerging.

5. Conclusions

The magnetic susceptibility and specific heat of $CeRu_4Sb_{12}$ can be described by a power-law or logarithmic divergence at low temperatures indicating that $CeRu_4Sb_{12}$ exhibits

non-Fermi-liquid behaviour. The resistivity of $CeRu_4Sb_{12}$ appears to be sample dependent, with some specimens displaying NFL characteristics and others having more complicated behaviour at low temperatures. The application of magnetic fields up to 8 T does not significantly influence the NFL behaviour. $CeRu_4Sb_{12}$ shows an enhancement of the magnetic susceptibility and Sommerfeld coefficient at low temperatures yielding a Wilson ratio of order unity and therefore could be classified as a moderately heavy-fermion compound.

Acknowledgments

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