$CeFeGe_3$: A concentrated Kondo compound with a stable valency and high Kondo temperature

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We report that the intermetallic compound $CeFeGe_3$ is a Kondo compound possessing a stable valency and a very high Kondo temperature of the order of 100 K. The magnetic susceptibility χ showed Curie-Weiss behavior above about 200 K and the Weiss temperature was found to be about -90 K. χ saturates at the lowest temperatures after making a faint maximum around 50 K. Below 8 K the specific heat exhibited a large temperature-independent linear term γ, amounting to 150 mJ/mol K^2 and the electrical resistivity showed T^2 dependence below about 4 K. The compound exhibited no phase transitions above 0.05 K and it is characterized as a nonmagnetic heavy-electron compound with the properties of a heavy Fermi liquid at low temperatures. The results of the magnetic susceptibility and magnetic contribution of the specific heat suggest a $j=\frac{3}{2}$ Kondo effect, which is also consistent with the result of thermoelectric power.

I. INTRODUCTION

Many heavy-electron compounds, including magnetic (mostly antiferromagnetic) and superconducting ones, are known at present.[1] They revealed a variety of properties ranging from the Kondo effect to the intermediate valence effect. Those heavy-electron compounds may be classified into two groups, the concentrated Kondo (CK) compound and the intermediate-valence (IV) compound.[2]

The CK compound has a well-defined integral or almost integral valence at higher temperatures than the characteristic Kondo temperature T_K ($T \gg T_K$), but at very low temperatures ($T \ll T_K$) enters a Fermi-liquid state with depressed magnetic moments, i.e., "Kondo compensated" state. Hitherto known compounds of this group have a low T_K of only about several degrees kelvin, which is much lower than the crystalline electric field (CEF) level splitting (Δ_{CF}). To a first approximation, the CEF level excitation can, therefore, be neglected at low enough temperatures ($T \ll T_K$), and the ground-state doublet is mainly involved in the Kondo effect with an effective total angular momentum $j=\frac{1}{2}$.

The IV compound, on the other hand, possesses a nonintegral valence at room temperature as a result of stronger hybridization of $4f$ and conduction electrons because of the anomalous proximity of the $4f$ level to the Fermi level. IV compounds such as $CePd_3$, $CeSn_3$, and CeNi have, generally, a very high T_K, of the order of 200 K. The CEF level excitation cannot be neglected, even at temperatures lower than T_K, and the full total angular momentum j ($j=\frac{5}{2}$ for Ce^{3+}) becomes effective in this case.

According to our preliminary study,[3] the compound $CeFeGe_3$ appears to possess some characteristics of these two groups, i.e., a high T_K of the ordr of 100 K and a trivalent integral Ce valence at room temperature. It may, hence, be helpful to clarify the true nature of the ground state of concentrated Kondo compounds. We herein report further details of this interesting Kondo compound.

II. EXPERIMENT

The polycrystalline samples of $CeFeGe_3$ and $LaFeGe_3$ were prepared by arc melting Ce (purity $3N$), La ($3N$), Fe ($4N$), and Ge ($5N$) in an argon atmosphere and by annealing at 800 °C ($CeFeGe_3$) and 850 °C ($LaFeGe_3$) for one week in an evacuated quartz tube. By x-ray diffraction on powdered samples with Cu-$K\alpha$ radiation and by metallography, it was confirmed that the sample was a single-phased intermetallic compound of the $BaNiSn_3$-type structure (space group $I4mm$, No. 107, $Z=2$). The tetragonal lattice parameters were $a=4.332$ Å and $c=9.955$ Å for $CeFeGe_3$, and $a=4.368$ Å and $c=9.985$ Å for $LaFeGe_3$. In order to definitely ascertain the ordered structure[4] of $CeFeGe_3$, a Rietveld analysis of x-ray powder diffraction was carried out, and the ordered structure of the $BaNiSn_3$ type was confirmed, as previously reported.[3]

The magnetic susceptibility χ was measured with a commercial superconducting quantum interference device (SQUID) magnetometer (VTS-905, BTi) from 6 to 400 K at 3 kOe. The magnetization M was measured at 2 K up to 45 kOe with the SQUID magnetometer.

The specific heat C_P was measured using about 2.5 g of sample with an adiabatic heat-pulse method in two separate calorimeters from 0.5 to 50 K and from 30 to 300 K. The accuracy of each calorimeter was checked by measuring about 4.9 and 0.84 g of high-purity copper for the low- and the high-temperature calorimeter, respectively. Deviations from the reference data[5] were less than 2% over the whole temperature range for these copper standards.

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The electrical resistivity ρ was measured by the conventional dc four-probe method from 0.5 K to room temperature with an excitation current of 2–3 mA. It was difficult to obtain precise absolute values of ρ because arc-melted samples often contained many pores and microcracks. Only specimens carefully selected under a microscope were used for the resistivity measurements. The thermoelectric power (TEP) was measured by a differential method from 2 to 300 K.

III. RESULTS AND DISCUSSION

The magnetic susceptibility at low temperatures revealed an appreciable sample dependence, particularly at low temperatures, as shown in Fig. 1(a) for several samples. χ above about 200 K was successfully fitted with a Curie-Weiss term plus a temperature-independent term χ_0. The inverse of $(\chi-\chi_0)$ is plotted as a function of temperature in Fig. 2, where χ_0 was taken as a constant value of 2.9×10^{-4} cm^3/mol. One can see that the susceptibility tends to saturate at lower temperatures below the faint maximum around 50 K, as expected for heavy-electron compounds. The upturn of χ at the lowest temperatures was, however, analyzed separately by fitting with the Curie law. The Curie constant of this portion of the χ curve gave a moment of $0.18-0.22\mu_B$ per Ce atom, if it is assumed that the paramagnetism is caused solely by the integrity of Ce atoms in the structure, for example, by uncompensated residual magnetic moments of Ce atoms. Or it can be likewise interpreted by assuming that only 0.13–0.19 at. % of the total iron atoms were magnetic with a moment of $\sim5\mu_B$. It would be, regardless, important to distinguish whether this weak paramagnetism is intrinsic or merely due to parasitic impurities.

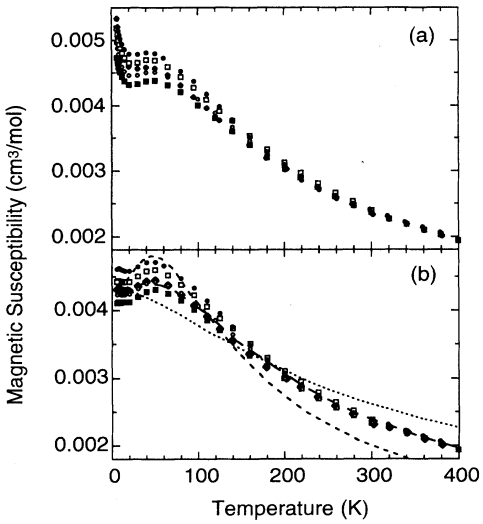

FIG. 1. (a) Raw magnetic susceptibilities of CeFeGe$_3$ (● CeFeGe$_3$ 1, ○ CeFeGe$_3$ 50 B, ■ CeFeGe$_3$ 54, □ CeFeGe$_3$ 55, and ◆ CeFeGe$_3$ 57 A). (b) Magnetic susceptibilities of CeFeGe$_3$ corrected for the low-temperature paramagnetic contribution and calculated ones of the Coqblin-Schrieffer model; observed [same symbols as for (a)] and calculated ($\cdots$ $j=\frac{1}{2}$ $T_0=180$ K, $-\,-\,-$ $j=\frac{3}{2}$ $T_0=180$ K, and $---$ $j=\frac{5}{2}$ $T_0=180$ K).

The effective magnetic moment $\mu_{\rm eff}$ and the Weiss temperature Θ_W deduced from the high-temperature data above 200 K were $2.53-2.57\mu_B$ per Ce atom and $-86--97$ K, respectively. The large negative value of Θ_W is consistent with the high Kondo temperature determined in the following. The effective moments are nearly equal to that of a free ion of Ce^{3+}, indicating that CeFeGe$_3$ is a little different from ordinary IV materials with a similarly high T_K, such as CePd$_3$ and CeSn$_3$.[2] The preliminary results of photoemission experiments also indicate that the spectra are different from those of IV compounds.[8] The fact further suggests that the irons in the structure of this compound are not magnetic. In fact, LaFeGe$_3$ was confirmed to be Pauli paramagnetic, although inhomogeneity caused very large parasitic paramagnetism, often as large as 1×10^{-2} cm^3/mol, at low temperatures. The Pauli paramagnetic susceptibility of LaFeGe$_3$ was almost T independent, and the value at 300 K was 2.10×10^{-4} cm^3/mol.

It is here remarked that the sample dependence of the low-temperature magnetic susceptibility was most striking among properties investigated in the present study, amounting to a dispersion of $\pm5\%$ in $\chi(0)$, as can be seen in Fig. 1(b). In order to understand this strong variation of $\chi(0)$, we measured $\chi(0)$ of CeFe$_{1.01}$Ge$_3$ with excess Fe and found about a three times larger $\chi(0)$, compared with that of CeFeGe$_3$. This means that only a 0.03% shift of iron concentration could explain such a distribution of $\chi(0)$, if the sample dependence were caused solely by excess iron atoms.

Figure 1(b) shows the magnetic susceptibility corrected for the paramagnetic contribution from which we obtain $\chi(0)=(4.11-4.60)\times10^{-3}$ cm^3/mol. It is very important to note here that the broad peak around 50 K has been more clearly revealed after the correction, which is certainly not an artifact of the correction and very different from the simple saturation toward $\chi(0)$, usually found for $s=\frac{1}{2}$ Kondo systems with a much lower T_K. We believe that the broad peak is a result of the orbital effect of total angular momentum j larger than $\frac{1}{2}$, as considered by Coqblin and Schrieffer.[6] In the Coqblin-Schrieffer model, spin-orbit exchange scattering is taken into account for the Kondo effect caused by Ce ions ($j=\frac{5}{2}$). Hence, in this model the multiplicity $2j+1$ for the total angular momentum plays an important role in stabilizing the Kondo state and raising T_K. Such a high T_K as in our compound may result from the multiplicity for $j=\frac{5}{2}$, rather than just from the ground-state doublet, as in many conventional Kondo compounds. Hence, for our compound with T_K of the order of CEF splitting, excited states of CEF splitted levels must be explicitly taken into account, and the simple $s=\frac{1}{2}$ Kondo model would not be applicable. The Kondo impurity problem in the Coqblin-Schrieffer model was numerically calculated for several total angular momenta by Rajan.[7] The results of his calculation are compared in Fig. 1(b) with the observed values. It is obvious that the curves for $j=\frac{1}{2}$ and $\frac{5}{2}$ do not fit the observed curves well compared with that for $j=\frac{3}{2}$. This suggests that the *effective* angular momen-

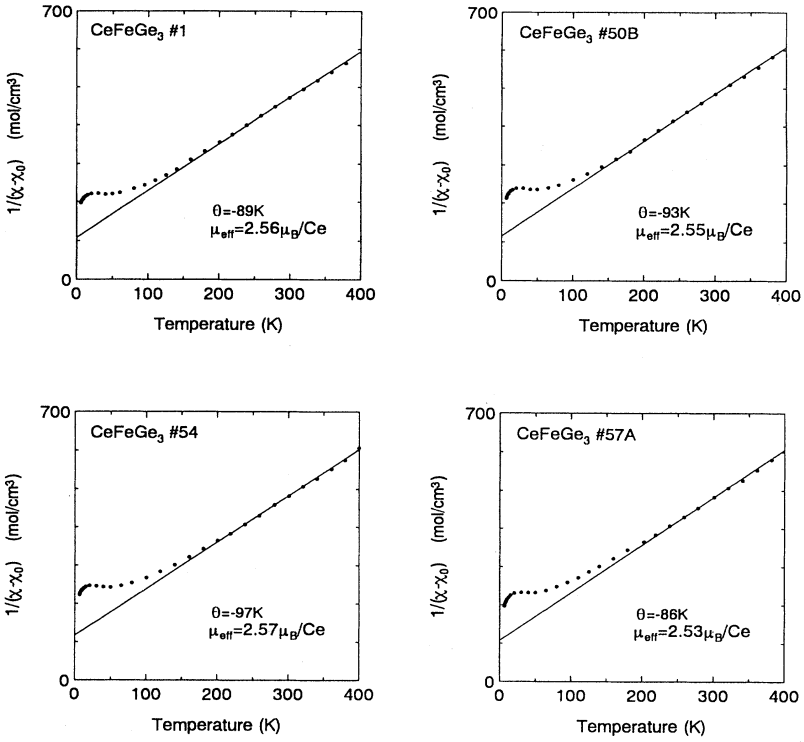

FIG. 2. Inverse magnetic susceptibilities of four different samples of $CeFeGe_3$. The straight lines designate Curie-Weiss laws.

tum of $CeFeGe_3$ for the Kondo effect at low temperatures ($T < \sim T_K$) would be $\frac{3}{2}$, which is also implied by the following analysis of specific heat. The Kondo temperature T_K calculated from the characteristic temperature T_0 of this fitting was 232 K. We can alternatively estimate T_K from the following relation using the average value 4.33×10^{-3} cm^3/mol for $\chi(0)$ and find 217 K;

$$T_K^{\chi(0)}=\frac{\nu(\nu^2-1)\mu_B^2 g^2 W}{24\pi k_B \chi(0)} ,$$

where $\nu=2j+1=6$, $g=\frac{6}{7}$, and W is the Wilson number $0.1026\times4\pi$.[7]

$CeRu_2Si_2$ has been known as a CK compound with one of the highest T_K's (15–20 K), and properties similar to those of $CeFeGe_3$ have been reported.[9,10] Among them, a similar manner of saturation of χ with a faint maximum below 10 K as just found for our compound is noteworthy.[9] Furthermore, high-field magnetization[10] and neutron-diffraction[11,12] experiments on $CeRu_2Si_2$ disclosed many new interesting features concerning the ground state of the Kondo compound. A metamagnetic-like transition around $H=8$ T at $T \ll T_K$ was interpreted as a collapsing of magnetic correlations found in inelastic neutron-diffraction experiments.[11] We believe it extremely important to likewise check for such magnetic correlations and a metamagneticlike transition in our compound in order to comprehend the true magnetic nature of the Kondo ground state. Our measurement of the magnetization up to 35 T did not reveal any evidence for such a metamagneticlike transition. If it is assumed that the critical magnetic field H_c is scaled with T_K, H_C of $CeFeGe_3$ would be about 80 T. We are planning to perform a higher-field magnetization measurement.

The specific heat of $CeFeGe_3$ between 0.5 and 300 K is shown together with that of $LaFeGe_3$ in Fig. 3. There are no anomalies implying long-range order in the temperature range of investigation. The linear term γ is as large as 150 mJ/mol K^2 and almost temperature independent up to about 8 K, as can be seen in the inset of Fig. 3, where Cp/T is plotted as a function of temperature. This *T-independent* Cp/T in such a wide temperature range is one of the striking features of this compound with a higher T_K than those of previously reported typical heavy-electron Kondo systems.[1,2]

In the Coqblin-Schrieffer model,[7,13] T_K is related to γ by the following equation:

$$T_K^{\gamma}=\frac{Wj\pi R}{3\gamma} ,$$

where R is the gas constant. We find $T_K=187$ K for $j=\frac{5}{2}$ and $\gamma=150$ mJ/mol K^2. This value of T_K is in fair agreement with those estimated above from the susceptibility.

In order to evaluate the magnetic specific heat C_m, the lattice and conduction-electron contribution were subtracted from the measured specific heat by assuming that the specific heat of $LaFeGe_3$ represents the nonmagnetic part of the specific heat of $CeFeGe_3$. The linear term γ and the Debye temperature Θ_D of $LaFeGe_3$ were 9.4 mJ/mol K^2 and 298 K, respectively. The subtraction of

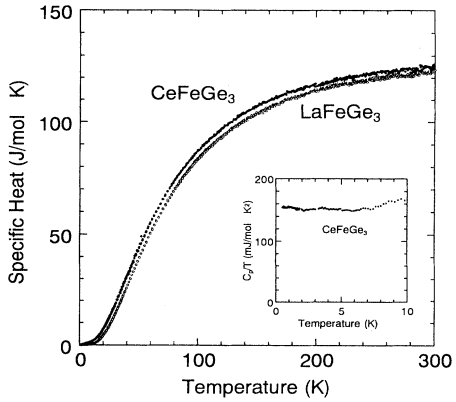

FIG. 3. Specific heat of $CeFeGe_3$ and $LaFeGe_3$. The inset shows temperature-independent Cp/T of $CeFeGe_3$ below about 8 K.

nonmagnetic specific heat was done after each specific heat curve of $CeFeGe_3$ and $LaFeGe_3$ had been fitted with Chebyshev's polynomials. Thus, C_m obtained from $CeFeGe_3$ is plotted in Fig. 4 with calculated C_m in the Coqblin-Schrieffer model for ($j=\frac{1}{2}$, $T_0=80$ K), ($j=\frac{3}{2}$, $T_0=110$ K), and ($j=\frac{5}{2}$, $T_0=150$ K).[7] The calculated values for $j=\frac{1}{2}$ are obviously too far off the observed curve, compared with those for $\frac{3}{2}$ and $\frac{5}{2}$. If we take further account of the Schottky specific heat to be added to the calculated values and the best fit of $\chi(T)$ obtained for $j=\frac{3}{2}$, as shown in Fig. 1(b), we come to the conclusion that the Coqblin-Schrieffer model with $j=\frac{3}{2}$ reasonably explains the thermal variation of both the magnetic susceptibility and the specific heat of $CeFeGe_3$. The Kondo temperature T_K obtained in this analysis was 142 K, again in fair agreement with those obtained above from γ and $\chi(0)$.

The magnetic entropy S_m was obtained by integrating C_m/T with respect to T. The inset of Fig. 4 shows that S_m monotonously increases until it reaches 12.5 J/mol K at 300 K, which is close to R ln6=14.9 J/mol K of the full entropy for the Ce^{3+} ion ($j=\frac{5}{2}$). This fact guarantees that our low-temperature state, say below 10 K, is the ground state of the system; in other words, there would be no more magnetic phase transition below the limit of the temperature range of the present specific-heat measurement. In fact, we observed no phase transitions down to 0.05 K in electrical resistivity and ac susceptibility measurements with a dilution refrigerator. It is also noteworthy that the initial T-linear increase of S_m is a result of the T-independent γ, and the full entropy R ln6 seems to be reached only around 600 K or more by a simple extrapolation. This suggests that the total CEF splitting Δ_{CF} is much higher than T_K. Any distinct CEF excitations were not, however, observed up to 40 meV of energy transfer in our preliminary inelastic neutron-scattering experiments. This fact suggests that CEF levels in $CeFeGe_3$ may be severely broadened by the Kondo effect, as found in $CeRu_2Si_2$.[12]

The electrical resistivity ρ of $CeFeGe_3$ is plotted with that of $LaFeGe_3$ in Fig. 5. The resistivity ratio, $\rho_{300}/\rho_{4.2}$ for $CeFeGe_3$ and $LaFeGe_3$, was found to be ~ 20 and ~ 50, respectively. These large values for the polycrystalline samples are consistent with the result of our x-ray analysis,[3] claiming that Fe and Ge atoms in these compounds occupy their specific sites in an ordered fashion. Another point to be noted in Fig. 5 is that the resistivity of $CeFeGe_3$ remains large at high temperatures and marks a sudden decrease below about 100 K, while $LaFeGe_3$ demonstrates ordinary metallic behavior. This difference in the thermal variation of ρ between these two compounds should be attributed to a magnetic origin. In

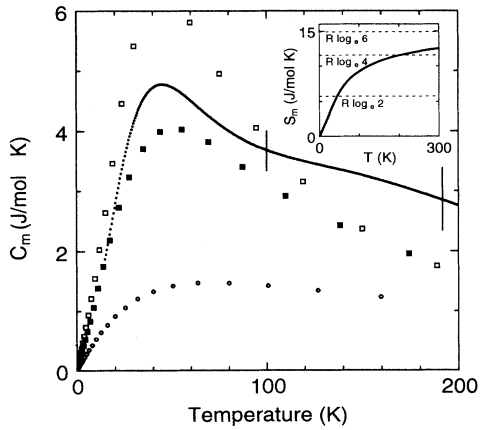

FIG. 4. Magnetic specific heat of $CeFeGe_3$; observed (denoted by ● with error bar) and calculated with Coqblin-Schrieffer model (○ $j=\frac{1}{2}$ $T_0=80$ K, ■ $j=\frac{3}{2}$ $T_0=110$ K, and □ $j=\frac{5}{2}$ $T_0=150$ K). The inset shows the temperature variation of magnetic entropy of $CeFeGe_3$.

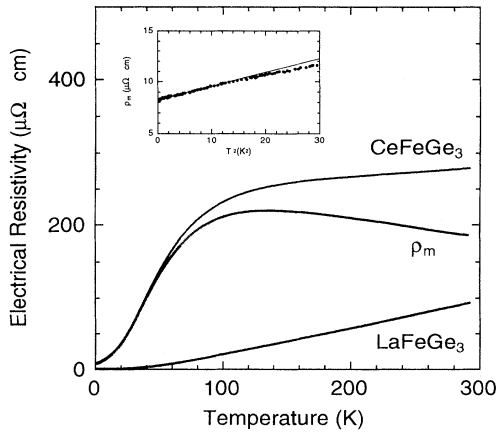

FIG. 5. Electrical resistivity of $CeFeGe_3$ and $LaFeGe_3$. The difference of the two is also included as the magnetic resistivity for $CeFeGe_3$, ρ_m. The inset shows the T^2 dependence of ρ_m below 5 K.

order to estimate the magnetic contribution to the electrical resistivity ρ_m, the resistivity of $LaFeGe_3$ is subtracted from the resistivity of $CeFeGe_3$. As can be seen in Fig. 5, ρ_m has a very broad peak around 140 K and decreases logarithmically above 220 K as expected for a typical Kondo scattering. The relatively sharp decline on the low-temperature side of the peak must be due to the coherency characteristic of a Kondo lattice. In any case, the broad peak of ρ_m is qualitatively understood as a Kondo effect strongly influenced by the CEF splitting, as was first calculated by Cornut and Coqblin.[14] It should be further noted that ρ_m exhibits a strong T dependence even at low temperatures of a usual residual-resistivity range. ρ_m below 5.5 K is plotted as a function of T^2 in the inset of Fig. 5, where it can be seen that ρ_m varies as AT^2 with a relatively small coefficient A of 0.13 $\mu\Omega$ cm/K^2. This T^2 dependence is another characteristic feature predicted by Fermi-liquid theory for heavy-electron Kondo systems.[1,2] This means that $CeFeGe_3$ enters a coherent Fermi-liquid state already at such a high temperature, whereas most Kondo compounds are reported to demonstrate the T^2 dependence of ρ_m well below 1 K.[15]

The thermoelectric power of $CeFeGe_3$ is shown as a function of temperature between 2 and 300 K in Fig. 6. The Seebeck coefficient S, which is related to the logarithmic derivative of the electronic density of states, increases with decreasing temperature and manifests a broad peak around 100 K. Below 100 K, it almost linearly decreases with temperature toward 0 K remaining positive, while ordinary Kondo compounds of Ce such as $CeCu_2Si_2$ and $CeAl_3$ show a negative minimum at low temperatures. This difference seems to result from the effective angular momentum j. When $j=\frac{1}{2}$, the Kondo resonance energy E_R is situated just at the Fermi level E_F and the sign inversion takes place at a low temperature, while $E_R > E_F$ for $j > \frac{1}{2}$; S then remains positive.[2] This result is consistent with the $j=\frac{3}{2}$ Kondo effect inferred above from the analyses of χ and C_P.

It is known that for heavy-electron systems there exist

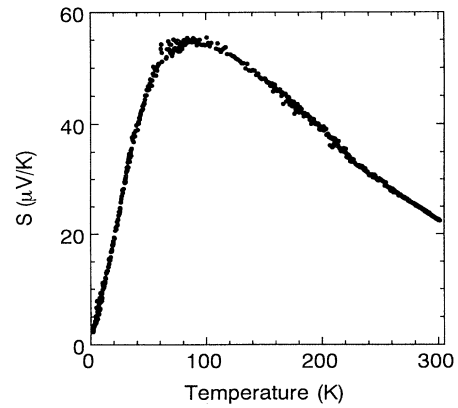

FIG. 6. Thermoelectric power of $CeFeGe_3$ between 2 and 300 K.

certain correlations among some parameters such as the enhanced $Cp/T(\gamma)$, χ at 0 K [$\chi(0)$], and the coefficient A of the T^2 term of ρ_m.[1] Most commonly discussed ratios are the Wilson ratio R_W and A/γ^2, the former of which is defined by

$$R_W = \frac{\pi^2 k_B^2 \chi(0)}{g^2 \mu_B^2 j(j+1)\gamma} .$$

Admitting that much physical significance cannot be ascribable to the magnitude of R_W, particularly in the case of partially lifted degeneracy, we estimated it merely for the purpose of comparison from $\gamma = 150$ mJ/mol K^2 and $\chi(0) = 4.33 \times 10^{-3}$ cm^3/mol for $j=\frac{5}{2}$ and $g=\frac{6}{7}$, as is usually done for the $j=\frac{1}{2}$ Kondo compounds of cerium. We thus obtained 1.03 for R_W of $CeFeGe_3$, which fits fairly well among those of other heavy-electron compounds.[16] The A/γ^2 of $CeFeGe_3$ turned out to be 5.8×10^{-6} $\mu\Omega$ cm mol^2 K^2/mJ^2, and this value is also consistent with those of other heavy-electron compounds.[17]

IV. CONCLUSION

We found a nonmagnetic Kondo compound, $CeFeGe_3$, of the $BaNiSn_3$ type of structure. The compound revealed typical heavy-electron behavior, which is consistent with Fermi-liquid theory. The magnetic susceptibility saturates at 4.33×10^{-3} cm^3/mol below the weak maximum around 50 K. The specific heat shows T-linear behavior below 8 K, i.e., temperature-independent Cp/T of 150 mJ/mol K^2, as a result of the high Kondo temperature, and the magnetic electrical resistivity exhibits a very broad peak around 140 K and T^2 dependence below about 4 K.

The results of the present study on $CeFeGe_3$ are consistent with a high Kondo temperature of about 100 K and a stable valence. The high-temperature magnetic susceptibility above about 200 K is consistent with an effective moment of $2.55\mu_B$ per Ce atom, nearly equal to a Ce^{3+} free ion.

The compound did not reveal any sort of phase transitions down to 0.05 K, and our analysis of the magnetic entropy suggests that the compound has already attained the ground state below a few tens of degrees kelvin. The presented results of magnetic susceptibility and magnetic specific heat strongly imply a $j=\frac{3}{2}$ Kondo effect rather than the more common $\frac{1}{2}$ Kondo effect. The thermal variation of thermoelectric power also supports this conclusion. Therefore, the present system would provide a unique opportunity for studying the ground-state properties of a heavy-electron Kondo system, particularly the ground state with $j > \frac{1}{2}$.

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