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New non-magnetically ordered heavy-fermion system CeTiGe

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Abstract

Investigations of the susceptibility, electrical resistivity, specific heat and thermopower of CeTiGe at low temperatures show that this compound is a Kondo lattice system with an enhanced Sommerfeld coefficient $\gamma \approx 0.3$ J K^{-2} mol^{-1} and where the whole $J = 5/2$ multiplet is involved in the formation of the ground state. No magnetic order was observed down to 0.4 K. In the temperature range below 10 K we observed Fermi-liquid behavior as indicated by a $\rho(T) \sim T^2$ dependence in the electrical resistivity and a linear specific heat and thermopower. Because of these results we classify CeTiGe as a moderate heavy-fermion system with a non-magnetic ground state.

1. Introduction

Systems which are located close to a ferromagnetic or antiferromagnetic quantum critical point are of great interest, because the low lying quantum fluctuations can lead to new/exotic physical phenomena like, for example, unconventional (non-Fermi liquid) metallic or unconventional superconducting states [1]. Intermetallic compounds based on Ce, Yb or U are excellent candidates for the observation of such critical points, since they can be easily tuned from a magnetic to a non-magnetic state by alloying, external pressure or magnetic field [2]. This results from the instability of their f-shell. The strong interaction between the f-electrons and the conduction electrons, which is generally discussed in the framework of Kondo theory, leads to an enhanced density of states at the Fermi level and to the formation of heavy quasiparticles, the origin of the term ‘heavy fermions’ for this class of compounds [3].

The discovery of the coexistence of ferromagnetism and superconductivity in UGe$_2$ [4], as well as the observation of comparatively high superconducting transition temperatures in the CeTIn$_5$ compound series [5], has pushed the interest towards quasi-two-dimensional (2D) heavy-fermion systems. On the one hand, it is well known that the reduction of the spatial dimensionality increases quantum fluctuations [6], but further on it was suggested that it also leads to an increase of T_c [7]. The CeFeSi structural type presents some 2D character, since it is a stacking of Ce–Si–Fe$_2$–Si–Ce layers. To the best of our knowledge, the first investigation of a Ce compound crystallizing in this structure type was reported 20 years ago on CeRuSi [8]. Only resistivity $\rho(T)$ and preliminary $C_p(T)$ data were shown, but they conclusively show a moderate heavy-fermion ground state with an enhanced Sommerfeld coefficient $\gamma \approx 200$ mJ K^{-2} mol^{-1}. In the past years, an increasing number of structural homologues have been investigated, but detailed studies on Ce-based compounds are still rather scarce, being limited to CeCoGe which presents antiferromagnetic order of a stable trivalent Ce state at $T_N = 5.5$ K [9]. CeTiGe was first observed by Welter *et al* [10] and Morozkin *et al* [11]. From preliminary susceptibility data Welter *et al* conclude the presence of a trivalent Ce^{3+} state without magnetic order in CeTiGe down to 5 K [10].

Since no further physical properties were reported and since the true ground state remained unsettled, we decided to investigate more precisely the low temperature properties of this compound. Interestingly, it is one of the rare cases of a Ce-based intermetallic compound with a transition metal from the left part of the periodic table.

2. Materials and experimental techniques

Two pellets of stoichiometric CeTiGe were prepared in two different ways. The first pellet was prepared by arc melting and homogenized by flipping and remelting four times in the arc furnace. The second pellet was also first melted in the arc furnace three times but then remelted three times in a high frequency levitation crucible to increase its homogeneity. The weight loss after melting was less than 0.1%. The ingots were then annealed for four days at 1000 °C under high vacuum conditions. Powder x-ray diffraction of these ingots confirmed the tetragonal CeFeSi-type structure (space group $P4/nmm$) with the lattice parameters $a = 4.1442$ Å and $c = 7.932$ Å for the first pellet and $a = 4.1432$ Å and $c = 7.931$ Å for the second one, close to the literature values [10, 11]. The non-magnetic reference compound LaTiGe was prepared in the same way as the first CeTiGe pellet and annealed at 1000 °C. The observed lattice parameters $a = 4.185$ Å and $c = 8.013$ Å show the expected volume increase to $V = 140.3$ Å^3 in comparison to CeTiGe with $V = 136.2$ Å^3.

The microprobe analyses also gave quite similar results for both samples of CeTiGe. The main phase was confirmed to be CeTiGe with a stoichiometric composition within the accuracy of this technique. Additionally three minor foreign phases were detected: Ti_5Ge_3, $Ti_{0.96}Ge_{0.04}$ and Ce_5Ge_3, which, however, were not observed in the x-ray powder pattern. The presence of these foreign phases is tentatively attributed to the high melting point of Ti_5Ge_3 ($T_m = 1980$ °C), which likely leads to a peritectic formation of CeTiGe, Ti_5Ge_3 being the primary phase, while Ce_5Ge_3 belongs to the final eutecticum. Microprobe analyses for the LaTiGe pellet showed similar results, with minor amounts of the phases Ti_5Ge_3, La_3Ge_2 and $La_{0.02}Ti_{0.95}Ge_{0.03}$ in addition to the stoichiometric main phase LaTiGe.

CeTiGe polycrystalline bars were cut from the ingots for the measurements. Pieces from pellet 1 will be denoted by number #1 and pieces from pellet 2 by number #2. The magnetic susceptibility was measured in the temperature range 2–400 K and in an applied field up to 5 T with a commercial Quantum Design superconducting quantum interference device (SQUID) magnetometer (MPMS). The specific heat and the resistivity were determined in a commercial PPMS (Quantum Design) equipment using the relaxation method and a conventional four-probe ac technique, respectively. With the ^{3}He refrigerator insert of the PPMS the temperature range could be extended to 0.4–300 K. The thermoelectric power $S(T)$ was measured with the thermal transport option (TTO) of the PPMS utilizing a one-heater two-thermometer technique in the temperature range 2 K $\leqslant T \leqslant$ 400 K.

3. Results and discussion

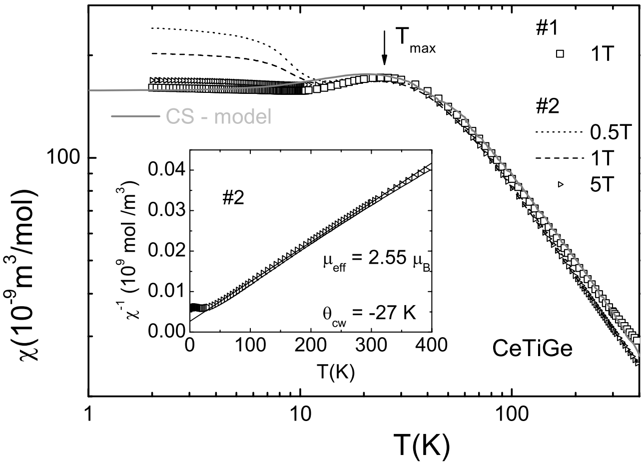

Figure 1. Temperature dependence of the magnetic susceptibility $\chi_{dc}(T)$ versus log T for two polycrystalline samples of CeTiGe. The first sample (#1) was measured in an external field $B = 1$ T, while the second sample (#2) was measured in $B = 0.5$, 1 and 5 T. The gray line shows the prediction of the Coqblin–Schrieffer (CS) model with $J = 5/2$ and $T_0 = 82$ K. Inset: for sample (#2) the inverse susceptibility $1/\chi$ in an external field of $B = 5$ T is plotted versus T. The gray line shows the Curie–Weiss fit.

The magnetic susceptibility $\chi_{dc}(T)$ in the temperature range 2–400 K is presented in (figure 1) for one sample of each pellet. Sample #2 was measured in three different fields $B = 0.5$, 1 and 5 T, while sample #1 was only measured for $B = 1$ T. The results at higher temperatures are very similar for both samples, while at low T some differences appear, which, however, as discussed below, can be attributed to different amounts of foreign phases. Above 60 K, $\chi(T)$ follows a Curie–Weiss law as demonstrated in the inset of (figure 1). From a fit $1/\chi(T) = (T - \theta)/C$ we obtained an effective moment $\mu_{eff} = 2.55\mu_B(2.68)\mu_B$ and a Weiss temperature $\theta = -27$ K (-30)K for the different samples. This evidences a trivalent Ce state and suggests a dominant antiferromagnetic exchange. Below 50 K the T dependence of the susceptibility deviates from the Curie–Weiss behavior and presents a well-defined broad maximum at $T_{max} = 24$ K. While $\chi(T)$ in sample #1 (and in sample #2 at 5 T) levels off below 20 K at a value $\chi_0 = (164 \pm 4)10^{-9}$ m^3 mol^{-1}, an upturn is observed at lower field in sample #2 below 10 K. However, the T and B dependence of this additional contribution is very similar to that reported for Ce_5Ge_3, which exhibits ferromagnetic behavior below 10 K [12]. Thus, taking into account the comparison between both samples and the results of the microprobe analyses, this additional contribution can safely be attributed to the Ce_5Ge_3 foreign phase. The magnetization at 2 K associated with this additional contribution in sample #2 corresponds to a Ce_5Ge_3 content of the order of 0.1 mol%. Thus the maximum in $\chi(T)$ at T_{max} and the leveling off at lower temperatures corresponds to the intrinsic behavior of CeTiGe. Such a behavior is typical for a heavy-fermion system located on the non-magnetic side of the critical point associated with the suppression of magnetic order. It is quite similar to the behavior observed in the archetypical heavy-fermion system $CeRu_2Si_2$ [13]. The value of T_{max} gives a first estimate of the characteristic 4f energy of CeTiGe. On the other hand, such a maximum in $\chi(T)$ is typical for a Kondo lattice system with a larger characteristic energy, i.e. at the border to the intermediate-valent state. Then not only the crystal electric field (CEF) ground state doublet, but some of the excited CEF states or even the whole multiplet is involved in the physics at low T, leading to a maximum in $\chi(T)$ and $C(T)/T$ [14]. For a more precise analysis of our data, we compared our data with the theoretical results for the Coqblin–Schrieffer (CS) model of Rajan [14] which were scaled with the experimental χ_0 and a scaling energy T_0. A quite good agreement was obtained assuming the whole multiplet $J = 5/2$ of Ce^{3+} to be involved and a scaling energy

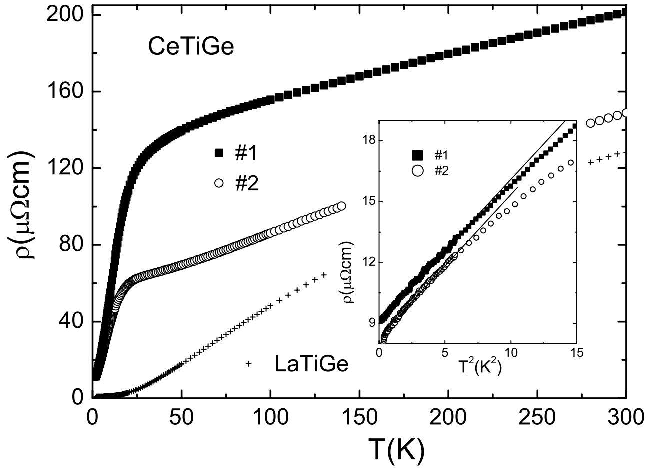

Figure 2. Temperature dependence of the electrical resistivity plotted for the two CeTiGe ingots. Inset: below 3 K, $\rho(T) \sim T^2$ is observed for both samples indicating a Fermi-liquid ground state. Data of sample (#2) are shifted vertically by $\rho'(T) = \rho(T) - 1\ \mu\Omega$ cm.

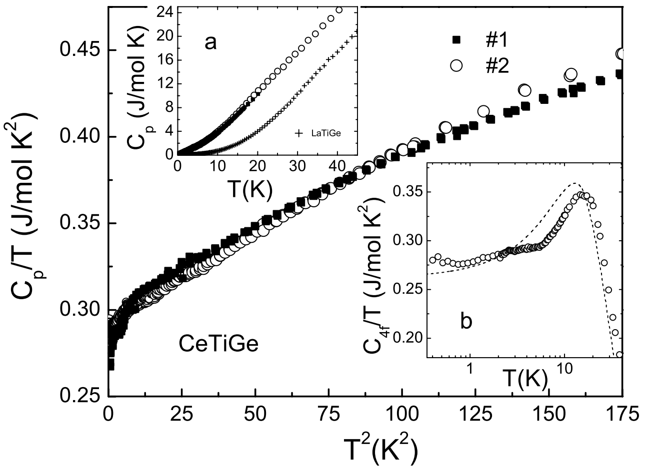

Figure 3. Temperature dependence of the specific heat of samples #1 and #2 plotted as C_p/T versus T^2 at low temperature. Below 10 K we observe a $C_p/T = \gamma + \beta T^2$ behavior. Inset (a) shows C_p up to 50 K for samples #1, #2 and LaTiGe. In (b) the $4f$ contribution to the specific heat is plotted as C_{4f}/T versus T. The prediction for the Coqblin–Schrieffer model with $J = 5/2$ and $T_0 = 82$ K is represented by the dotted line.

$T_0 = 82$ K (solid line in figure 1). This scaling energy can be converted into a Kondo temperature (in its high T definition) according to $T_K = T_0 \cdot 2 \cdot W_j \cdot \pi/(2J+1)$, which for $J = 5/2$, $W_{5/2} = 0.6468$ [15] gives $T_K = 55$ K. A close inspection of the comparison between theoretical and experimental data reveals a sharper increase of $\chi(T)$ from low T towards the maximum in the experimental data. This shall be discussed later.

Magnetization measurements performed at 2 and 10 K exhibit no saturation in M versus B curves and a linear field dependence up to $B = 5$ T (not shown). The magnetization value for CeTiGe reaches a value of $\mu_{\mathrm{eff}} = 0.12\mu_B$/Ce in 5 T. In order to look for the existence of a metamagnetic transition, often seen in Kondo systems, investigations towards much higher fields are necessary.

The heavy-fermion character is confirmed by the T dependence of the resistivity (figure 2). Below room temperature, $\rho(T)$ presents a weak, linear in T decrease down to 50 K, where a pronounced drop sets in. $\rho(T)$ decreases by one order of magnitude down to the lowest temperature. Such a pronounced decrease is typical for the formation of a coherent Kondo lattice. The comparison with the homologue LaTiGe (figure 2, symbol (+)) shows clearly that the resistivity is dominated by magnetic scattering of the $4f$-electrons. The magnetic part of the resistivity, which can be guessed qualitatively by looking at the difference between the CeTiGe and LaTiGe data, shows a maximum at around 35 K followed by a decrease towards higher temperatures, as expected for a Kondo lattice. The two samples display slight differences in $\rho(T)$ between 20 and 50 K, which might be due to an anisotropy induced either by crystal electric field (CEF) effects, as seen in $CeCu_2Si_2$ [16], or by a quasi-two-dimensional Fermi surface, like, for example, in the structurally related $CeCuSb_2$ and $CeAuSb_2$ [17]. Such anisotropies become visible even in polycrystalline samples because of the texture commonly induced by the strong temperature gradient upon solidification of arc-melted samples. Similar differences in the absolute values of $\rho(T)$ and its temperature dependence between different samples are quite common in Kondo lattice systems, see, for example, the results on the archetypical heavy-fermion system[1] $CeCu_2Si_2$.

The residual resistivity ratio in sample #1 amounts to $\mathrm{RRR}_0 = 23$, indicating a rather good quality of this batch, while RRR_0 of sample #2 is slightly smaller $\mathrm{RRR}_0 = 16$. In a plot of $\rho(T)$ versus T^2 (inset of figure 2) the data of sample #1 follow a straight line below $T = 3$ K, while those of sample #2 show a weak curvature down to low T. Thus a higher sample quality results in a better defined Fermi-liquid behavior. A fit of the data with the simple power law $\rho(T) = \rho_0 + AT^2$ leads to $\rho_0 = 9.1\ \mu\Omega$ cm with $A = 0.71\ \mu\Omega$ cm K^{-2} for sample #1 and $\rho_0 = 9.12\ \mu\Omega$ cm with $A = 0.77\ \mu\Omega$ cm K^{-2} for sample #2, respectively. No anomaly could be detected in $\rho(T)$, showing the absence of intrinsic magnetic order above 0.4 K.

The specific heat $C_p(T)$ was measured in the temperature range from 0.4 to 10 K with the ^{3}He insert of the PPMS. The measurements were extended to 50 K using the regular ^{4}He set-up. The two samples showed the same results within the accuracy of the measurements. $C_p(T)$ increases continuously with T in the whole investigated T range, without any evidence for an anomaly related to a phase transition (figure 3(a)). A plot of C_p/T versus T^2 at low temperatures shows a straight line below 10 K (figure 3), indicating a power law $C = \gamma T + \beta T^3$ in this region. The small deviation from linear behavior below $T = 2.5$ K is within the inaccuracy of the experimental technique. The T linear term can be attributed to the electronic contribution, while the βT^3 term corresponds to the contribution expected for the phonons. The large Sommerfeld coefficient $\gamma_0 = 290$ mJ K^{-2} mol^{-1}, only slightly smaller than in the archetypical heavy-fermion system $CeRu_2Si_2$ [13], indicates the formation of a heavy Fermi liquid at low temperatures. In order to get more precise information on the $4f$ contribution to the specific heat C_{4f}, $C_p(T)$ of

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[1] See figure 2 table 1 in [18].

LaTiGe was measured too (figure 3(a)) and subtracted from the data of CeTiGe (figure 3(b)). LaTiGe also presents a power law $C = \gamma T + \beta T^3$ at low T with $\gamma_0 = 23$ mJ/(K^2 mol) and $\beta = 5.2 \times 10^{-4}$ J K^{-4} mol^{-1}, the latter one corresponding to a Debye temperature $\theta_D = 224$ K. In confirmation of our preliminary analysis, C_{4f}/T is almost constant below 10 K (C_{4f}/T is lower than $\gamma_0 = 290$ mJ K^{-2} mol^{-1} because the contribution of non-f conduction electrons is subtracted), but increases above 10 K towards a clear maximum at 16 K and then decreases as expected for a Kondo lattice system. The maximum in C_{4f}/T at low T is a further indication that the local moment involved has a larger degeneracy. Therefore we again compare our experimental data with the prediction of Rajan [14] for the Coqblin–Schrieffer model for $J = 5/2$ and $T_0 = 82$ K (as determined from the analysis of $\chi(T)$). The theoretical curve fits quite nicely to the experimental data (figure 3(b)), supporting that the whole multiplet is involved in the formation of the Kondo state. As for the susceptibility, the maximum in C_{4f}/T is sharper in the experimental data than in the theoretical data. The reason why these features are sharper than theoretically predicted is not yet clear, but we suspect that it is related to some intersite interactions. A further estimate for the characteristic energy can be gained from the entropy, by taking twice the temperature $T = 23$ K at which half of the full entropy $R \ln 6$ is recovered [19], which leads to $T^* = 46$ K, slightly smaller than $T_K = 55$ K estimated from the analysis of $\chi(T)$ and $C_{4f}(T)$ using the Coqblin–Schrieffer model. The entropy reaches $R \ln 4$ at 40 K, above which the experimental $C_{4f}(T)$ data deviates upwards from the Coqblin–Schrieffer model and seems to extrapolate to a larger value than $R \ln 6$ at high temperatures, indicating that the subtraction of the phonons is not anymore reliable. The large entropy collected until 40 K is direct evidence that the CEF splitting is small and thus that the whole 5/2 multiplet is involved in the formation of the Kondo ground state. Therefore the specific heat data confirm that CeTiGe is a magnetically non-ordered heavy-fermion system, with a larger degeneracy of the involved local moment and a characteristic energy of the order of 50 K. Thus CeTiGe is at the border between the pure Kondo and the intermediate valence regime. Accordingly, no change in C/T is observed upon applying an external field $B = 7.0$ T.

A further hallmark of heavy-fermion systems is the presence of a very large thermopower, resulting from the enhanced density of states at the Fermi level due to the hybridization between the f and the conduction electrons [20]. The thermopower of CeTiGe is shown in figure 4 for 2 K $\leqslant T \leqslant$ 400 K on a logarithmic T scale. The $S(T)$ curve is dominated by one single broad peak with its maximum at an astonishingly low temperature of 17 K, exhibiting a large positive value of $S \approx 60\ \mu$V K^{-1}. With increasing temperature the thermopower decreases and a sign change from positive to negative can be observed at around 230 K. The S values at high temperatures are relatively small with a negative diffusion contribution. This indicates light electron-like charge carriers. At temperatures below 6 K the thermopower of CeTiGe vanishes linearly with T (inset of figure 4), as expected for a Fermi-liquid system. A universal ratio of S/T and γ has been found for a number of metals disregarding their effective mass [21], $q = S/T \cdot N_{Av} \cdot e/\gamma \approx 1$ with N_{Av} as Avogadro's constant and e as electronic charge. For CeTiGe we found $q = 1.7$, in good agreement with this empirical law.

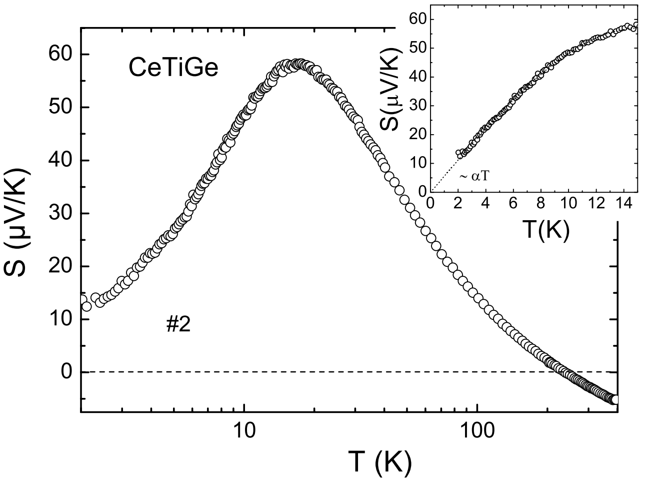

Figure 4. Temperature dependence of the thermopower $S(T)$ of CeTiGe below 400 K on a semilogarithmic scale. Inset: linear extrapolation of $S(T)$ to zero at low temperatures for CeTiGe.

The temperature dependence of the thermopower in Kondo lattice systems arises from two contributions: Kondo scattering at the ground state and incoherent scattering at excited crystal electric field levels. The former mechanism gives rise to a first maximum at low temperature T, being roughly equal to T_K [22], while the latter one appears as a broad high temperature anomaly at $T \approx 0.3 \cdots 0.5 \Delta_{CEF}/k_B$, with Δ_{CEF} being the overall CEF splitting [23]. However, it was confirmed theoretically and experimentally that the energy scales of Kondo interaction and CEF splitting have to be separated by a factor of 10–20 for these two distinct features in $S(T)$ to be resolved [20]. So often only one feature is observed, which has to be attributed to a superposition of both the Kondo and CEF effects. As only one single feature is resolved in $S(T)$ in CeTiGe, it can be assumed that it originates from the combined Kondo scattering at the ground state doublet and at the excited doublets of the Ce^{3+} ion. This is a further confirmation that the whole $J = 5/2$ is involved in the formation of the Kondo state. The maximum in $S(T)$ is, however, located at a much lower temperature $T_{S\,\max} \approx 17$ K than expected from theoretical calculations, which for $N = 6$ predicts $T_{S\,\max} \approx 0.6\ T_0$ [22]. For $T_0 = 82$ K as obtained from $\chi(T)$ and $C_{4f}(T)$ one would expect $T_{S\,\max} \approx 49$ K. The reason for this difference is not yet clear. One can notice that the maximum in $S(T)$ occurs at the same temperature as the maximum in C_{4f}/T, a further example of the close relation between specific heat and thermopower in heavy-fermion systems [21, 24].

4. Conclusions

The observed temperature dependences of the susceptibility, resistivity, specific heat and thermopower of CeTiGe indicate that this system is a Kondo lattice located on the non-magnetic side of the (quantum) critical point within the Doniach phase diagram. At low temperatures ($T < 3$ K) CeTiGe presents all the properties expected for a Fermi liquid: a resistivity increase AT^2, a constant Pauli-like susceptibility χ_0, and a specific heat and a thermopower proportional to the temperature, $C \sim \gamma T$ and $S \sim \alpha T$, respectively. From the experimentally determined coefficient A, χ_0 and γ, we deduce a Kadowaki–Woods ratio of $A/\gamma^2 = 8\ \mu\Omega$ cm mol^2 K^2 J^{-2} and a Wilson ratio $R_{\mathrm{w}} = \chi_0/\gamma_0(\pi k_{\mathrm{B}})^2/(\mu_0\mu_{\mathrm{eff}}^2) = 1.5$ (using $\mu_{\mathrm{eff}} = 2.54\mu_B$) both being well in the range expected for heavy-fermion systems [25, 26]. The maximum observed in $\chi(T)$ at 24 K and in $C_{4\mathrm{f}}/T$ at 16 K indicates a larger degeneracy of the involved local moment. Both $\chi(T)$ and $C_{4\mathrm{f}}(T)$ can be well fitted with the prediction for the Coqblin–Schrieffer model for $J = 5/2$ with $T_0 = 82$ K, corresponding to a Kondo scale $T_{\mathrm{K}} = 55$ K, while the analysis of the entropy leads to a slightly smaller $T_{\mathrm{K}} = 46$ K. These conclusions are supported by the T dependence of the thermopower, which presents only one maximum at a comparatively small temperature $T_{S\,\max} = 17$ K, indicating that the CEF splitting is not much larger than the Kondo scale, and by the absence of a hump in $\rho(T)$ at higher temperatures related to Kondo scattering by well-separated excited crystal field levels [27].

The sample dependence and the field dependence of the anomaly observed in $\chi(T)$ around 8 K indicate that this anomaly is due to a small amount of Ce_5Ge_3 foreign phase also visible in the microprobe analysis. The absence of an anomaly in $C_p(T)$ and $\rho(T)$ suggests that CeTiGe itself does not present any phase transitions. Thus our results demonstrate that CeTiGe is a new magnetically non-ordered heavy-fermion system with a Kondo temperature $T_{\mathrm{K}} \approx 50$ K and a Sommerfeld coefficient $\gamma \approx 0.3$ J/(K^2 mol).

Acknowledgments

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