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Field suppression of the heavy-fermion state in $CeRu_2Si_2$

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Specific-heat measurements have been performed on a single-crystalline sample of the heavy-fermion compound $CeRu_2Si_2$ in strong magnetic fields up to 20 T along the tetragonal axis in the temperature interval $1.5 < T < 30$ K. The linear electronic term in the specific heat (γ) passes through a pronounced maximum at a field $B^* \simeq 7.8$ T, where the metamagneticlike transition occurs. For fields above B^*, the mass enhancement decreases gradually, resulting in a reduction of the effective mass at 20 T by $\sim 80\%$ with respect to the zero-field value. In the high-field region, the data have been analyzed using a simple single-resonance-level model.

I. INTRODUCTION

The intermetallic compound $CeRu_2Si_2$ belongs to the series of rare-earth ternaries that crystallizes in the tetragonal $ThCr_2Si_2$-type structure. In the past few years, $CeRu_2Si_2$ has been studied intensively, as heavy-fermion behavior and metamagnetism dominate its properties at low temperatures. The coefficient of the term in the specific heat linear in temperaure (γ) amounts to 350 $\mathrm{mJ/mol\,K^2}$ (Refs. 1–4), allowing for a Fermi-liquid description with strongly interacting f electrons. Comparing this anomalous γ value with the value of 6.5 $\mathrm{mJ/mol\,K^2}$ obtained[1] for $LaRu_2Si_2$, one obtains an effective-mass enhancement that is 54 times larger than for the analog non-f-electron system. A description in the Fermi-liquid picture is furthermore justified by the Pauli-like susceptibility[5,6] at low temperatures, which amounts to $(150-200)\times 10^{-9}\ \mathrm{m^3/mol}$.

The ground state of $CeRu_2Si_2$ does not exhibit long-range order.[6] However, it has been shown that $CeRu_2Si_2$ is close to an antiferromagnetic instability, as follows, for instance, from alloying experiments.[1] The occurrence of electronic instabilities, either magnetic or superconducting, is a general feature among heavy-fermion systems and underlies the heavy-fermion behavior.[7]

Much attention has been focused on $CeRu_2Si_2$ because of its metamagneticlike properties. In the liquid-helium temperature region a strong increase in the magnetization occurs at $B^* \simeq 7.8$ T for a field direction along the tetragonal axis.[1,8] As the maximum of the differential susceptibility, $\Delta\sigma/\Delta H$, at B^* is accompanied by a pronounced peak in the magnetoresistance,[8] it has been proposed[8] that a strong reduction of the zero-field antiferromagnetic fluctuations is accomplished above B^*. Strong support for such an interpretation comes from inelastic-neutron-scattering experiments,[9] revealing the presence of two competing magnetic interactions: (i) a Kondo single-site contribution, appearing as a quasielastic peak with linewidth $\Gamma_{s\text{-}s} \sim 2$ meV, that is nearly field independent, and (ii) an antiferromagnetic intersite interaction, described by an energy shift $\hbar\omega_0 \sim 1.2$ meV and linewidth $\Gamma_{i\text{-}s} \sim 0.9$ meV, that is largely suppressed in fields above B^*.

The compound $CeRu_2Si_2$ is considered to be an exemplary system for the study of the competition between the Kondo effect and the Ruderman-Kittel-Kasuya-Yoshida (RKKY) interaction, generally occurring in heavy-fermion systems. Because of the availability of good-quality single-crystalline samples and the relatively low metamagnetic field, the field variation of the effective mass can be investigated in great detail. From a recent thermodynamic analysis of low-temperature susceptibility data and thermal-expansion data in field,[10–12] it follows that the field-induced mass enhancement amounts to 1.77 at B^*. It is a challenging task to account for the mass enhancement theoretically. Recently, Ohkawa[13] proposed a Kondo-collapse mechanism cooperating with a ferromagnetic exchange interaction in order to explain the metamagnetic transition in $CeRu_2Si_2$. However, the field-induced mass enhancement at B^* obtained in his model is only about 1.03. The presence of two competing magnetic interactions of a complex and incommensurate nature is undoubtedly the reason that satisfactory models are still lacking.

The purpose of this paper is to (i) measure $m_{\mathrm{eff}}(B)$ directly, and (ii) gain insight into the Kondo-type interactions in $CeRu_2Si_2$, by performing thermodynamic measurements in the field region where the intersite correlations are largely suppressed. In order to probe the high-field region, we have measured the specific-heat, $c(T)$ in the temperature interval $1.5 < T < 30$ K, up to 20 T. As it will appear, our maximum field leads to a strong suppression of the heavy-fermion state.

II. EXPERIMENT

The experiments were performed on a single-crystalline sample (mass 0.461 g) for a field direction along the tetragonal (*c*) axis. A batch of $CeRu_2Si_2$ was prepared[10] from high-purity starting materials [Ce (99.99%), Si (>99.999%), both supplied by Johnson-Matthey, and Ru (99.999%), supplied by Leico Industries, Inc.] in a tri-arc Czochralski furnace. The as-grown sample was cut into the proper shape by means of a spark-erosion technique. The sample was characterized in detail by extensive thermal expansion[10–12] and susceptibility[12] measurements.

The heat-capacity experiments were performed at the High Field Magnet Laboratory of the University of Nijmegen, using a Bitter magnet ($B_{max}=20$ T), in the same way as described previously in connection with experiments on UPt_3.[14] The sample was glued with Apiezon-N grease to a sapphire plate equipped with a RuO_2 thermometer and a nickel-chromium film as heater. The adiabatic specific-heat technique was used. The sample was cooled by a mechanical heat-switch. The heat capacity of the sample holder, amounting to 36% of the heat capacity of the $CeRu_2Si_2$ sample at 20 K and zero field, has been carefully subtracted from the data.

III. RESULTS

The experimental results are presented in Figs. 1 and 2 in a plot of c/T versus T. In Fig. 1 we show mainly the data for fields $B \geq 8.1$ T in the temperature interval $1.5 < T < 20$ K. In Fig. 2 we show the data for fields $B \leq 9.0$ T in the temperature interval $1.5 < T < 5$ K. For comparison we also show in Fig. 1 the data for $LaRu_2Si_2$.

The rather complex field variation of c/T versus T at low temperatures can be summarized as follows. For $B=0$ T, c/T is almost temperature independent. For low fields, c/T at $T=1.5$ K increases and a weak maximum develops in c/T versus T ($T_{max}=2$ K at $B=6$ T). This maximum shifts towards lower temperatures (T_{max} decreases) with increasing field, falling outside our temperature range when B approaches B^*. For fields above B^*, c/T at $T=1.5$ K drops monotonously with increasing field and again a maximum in c/T versus T develops. T_{max} now rapidly increases with field—it shifts from 2.7 K at 9 T up to 20 K at 20 T.

As it is not obvious how to extrapolate the (c/T)-versus-T curves to zero temperature and thus how to determine $\gamma(B)$, we plot in Fig. 3 the field variation of c/T at $T=1.5$ K. It follows from Fig. 3 that a pronounced maximum is found at $B^*=7.8$ T. Note that the c/T values at 1.5 K only approach a true γ value in the low- and high-field limits of our data set, where c/T is nearly temperature independent at low temperatures. If we were to extend our measurements below 1.5 K, the field variation of c/T (for $T \rightarrow 0$) would become much more pronounced, eventually approaching $\gamma(B)$. We also compare in Fig. 3 the measured c/T values with the ones calculated from high-field susceptibility data[11] taken in the same temperature range. Assuming a T^2 low-temperature limit of the magnetization (M), it follows from thermodynamics that $\partial M/\partial T^2=\frac{1}{2}(\partial\gamma/\partial B)$. The resulting $\gamma(B)$ is presented by the solid line in Fig. 3. Recent low-temperature magnetization[12] measurements below 1 K have clearly illustrated the sharpening of c/T as function of B for fields close to B^* when $T \rightarrow 0$.

Our low-field results are in good agreement with the data of Fisher *et al.*[4] However, their data are limited to 7.5 T and do not probe the metamagneticlike transition. The specific heat of polycrystalline $CeRu_2Si_2$ has been measured by Kim *et al.*[15] up to 12.5 T. These authors also observe a maximum at B^* in c/T at $T=1.5$ K, but a quantitative comparison with their field data is not appropriate as the magnetic properties of $CeRu_2Si_2$ are highly anisotropic [$\chi_{\parallel}/\chi_{\perp}=15$ at 10 K (Ref. 6)].

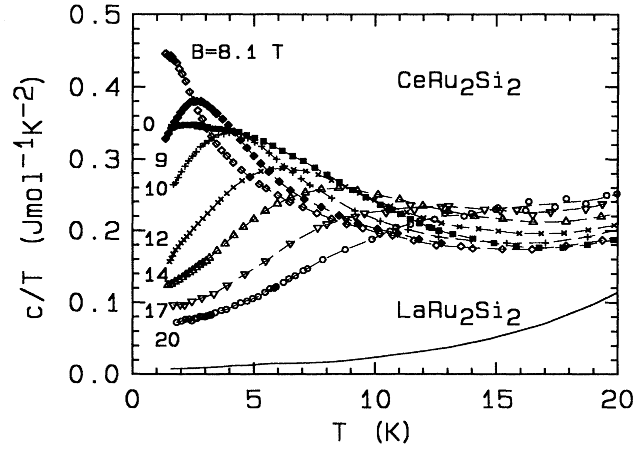

FIG. 1. Specific heat of single-crystalline $CeRu_2Si_2$, plotted as c/T vs T, in the temperature interval $1.5 < T < 20$ K, for magnetic fields along the tetragonal axis as indicated. The dashed lines serve as a guide to the eye. The solid line represents c/T vs T for $LaRu_2Si_2$ after Ref. 1.

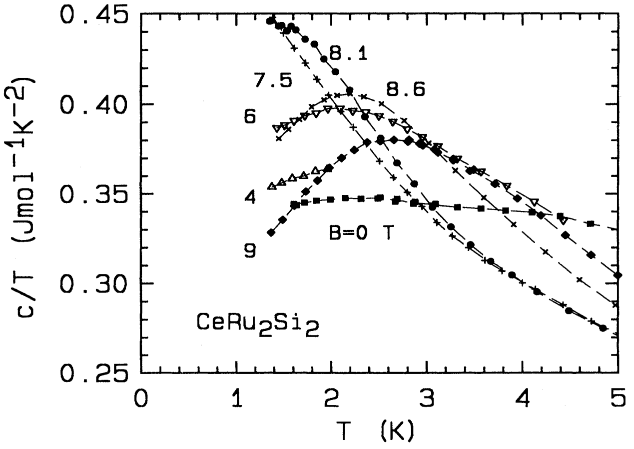

FIG. 2. Specific heat of single-crystalline $CeRu_2Si_2$, plotted as c/T vs T, in the temperature interval $1.5 < T < 5$ K, for magnetic fields along the tetragonal axis as indicated. The dashed lines serve as a guide to the eye.

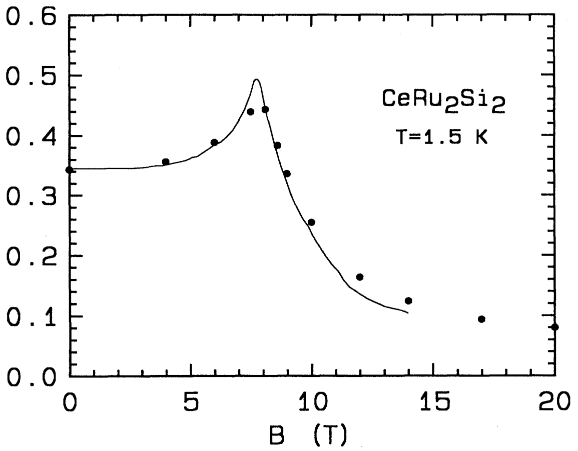

FIG. 3. Field variation of c/T at 1.5 K. The solid line has been calculated from magnetization data (Ref. 11).

IV. ANALYSIS

The f-electron specific heat of CeRu$_2$Si$_2$ in zero field consists of two distinct contributions.[1] A low-temperature anomaly centered at 11 K, caused by the competition of the magnetic interactions, and a high-temperature anomaly centered at 90 K that has been ascribed to the population of the first-excited crystal-field doublet at 220 K above the ground-state doublet (the tetragonal crystal field splits the $J=\frac{5}{2}$ multiplet into three doublets). The entropy associated with the low-temperature contribution amounts to $\sim 0.8R \ln 2$ at 20 K. In a first attempt to model this contribution, it has been compared to a single-ion Kondo $S=\frac{1}{2}$ anomaly, yielding values for the Kondo temperature, T_K, of 24.5 K (Ref. 1) and 19 K (Ref. 10). However, as CeRu$_2$Si$_2$ is a Kondo-lattice compound and as competing magnetic interactions prevail, such a comparison must be far from appropriate. Furthermore, in a simple Kondo model one cannot account for an initial increase of γ with field. Since for large magnetic fields the intersite correlations are largely suppressed, and the remaining interactions are dominantly of the Kondo type, it is, however, of interest to compare the specific-heat data at high field ($B>B^*$) with the theoretical curves obtained in a Kondo model. For this we choose the simple single-resonance-level model as worked out by Schotte and Schotte.[16]

In this model a resonance of Lorentzian shape is formed at the Fermi energy. The width (Δ) of the resonance is of the order of the Kondo temperature: $\Delta \simeq k_B T_K$. The molar specific heat for an external field H and spin $S=\frac{1}{2}$ is given by

$$C=\frac{R\Delta}{\pi T}-2R\,\mathrm{Re}\left\{\frac{(\Delta+ig\mu_B H)^2}{(2\pi k_B T)^2}\times\left[4\Psi'\left(1+2\frac{\Delta+ig\mu_B H}{2\pi k_B T}\right)-\Psi'\left(1+\frac{\Delta+ig\mu_B H}{2\pi k_B T}\right)\right]\right\}, \quad (1)$$

where R is the gas constant and Ψ' is the derivative of the digamma function. In Fig. 4 we compare our experimental results, after subtracting the lattice contribution obtained for LaRu$_2$Si$_2$, with Eq. (1), where the effective g factor $g=g_{\mathrm{eff}}=3.8$ was determined from the saturation magnetization along the c axis.[17] The fit parameter $T_\Delta=\Delta/k_B$ determines the position of the maximum in c versus T for $H=0$. Fitting only T_Δ, while taking for H the applied field, leads to an overly large peak height. The best results were obtained by introducing a second fit parameter, defining an effective field $H\equiv H_{\mathrm{eff}}=B_{\mathrm{eff}}/\mu_0$. The field variation of T_Δ and B_{eff} is shown in Fig. 5. For comparison we also show in Fig. 4 the zero-field specific heat as measured and as calculated using $T_\Delta=19$ K and $B_{\mathrm{eff}}=0$.[10]

V. DISCUSSION

The overall picture that emerges from Figs. 1 and 2 is that the composite contribution to the specific heat, (i.e., from the Kondo and RKKY interactions) shifts towards lower temperatures with increasing fields for $B<B^*$, while for $B>B^*$ the remaining contribution (mainly of the Kondo type) shifts upwards with increasing field.

From Fig. 3 it follows that the field-induced mass enhancement at B^* amounts to $m_{\mathrm{eff}}(B^*)/m_{\mathrm{eff}}(0)=1.28$ at 1.5 K. In order to determine the mass enhancement in the vicinity of B^* at $T=0$ K, the measurements obviously need to be extended to lower temperatures. The exact determination of B^* is furthermore complicated by the weak temperature dependence of B^* itself [$B^*=8.323$ T at 4.2 K, while $B=7.665$ T at 0.12 K (Ref. 18)]. From the low-temperature magnetization measurements[12] a field-induced mass enhancement $m_{\mathrm{eff}}(B^*)/m_{\mathrm{eff}}(0)=1.77$ for $T \rightarrow 0$ has been deduced. Hence $\gamma(B^*)$ amounts to 630 mJ/mol K^2, which is approximately equal to the γ

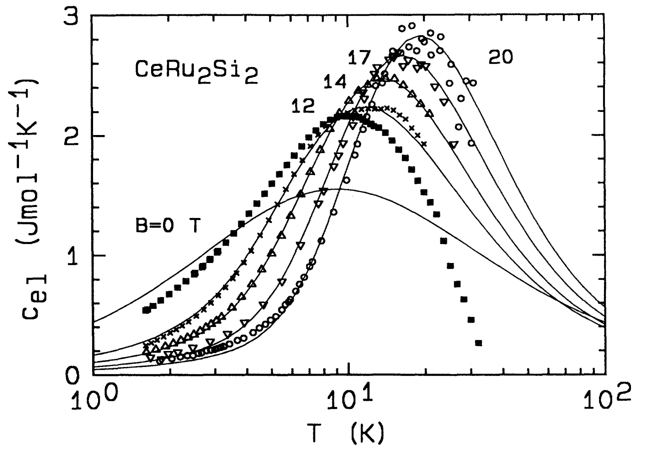

FIG. 4. The electronic specific heat of CeRu$_2$Si$_2$ for fields ($B>B^*$) as indicated along the tetragonal axis on a logarithmic temperature scale. The solid lines are calculated using Eq. (1) (see text). For comparison we also show the experimental and calculated data (using $\Delta/k_B=19$ K) at $B=0$ T.

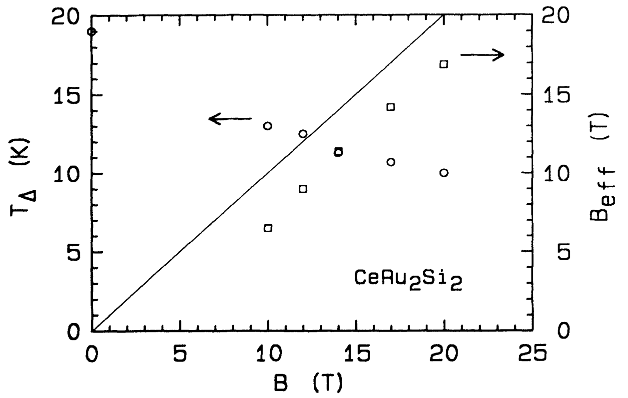

FIG. 5. The fit parameters T_Δ (open circles) and B_{eff} (open squares) plotted as a function of the applied magnetic field. The solid line indicates $B = B_{eff}$.

value for which the system $Ce_{1-x}La_xRu_2Si_2$ shows a transition to the long-range antiferromagnetic ordered state at the critical concentration of 8% La.[1,19]

At 20 T the γ value is substantially reduced to 80 mJ/mol K^2. The corresponding mass enhancement amounts to $m_{eff}(B = 20\ T)/m_{eff}(0) = 0.23$. Apparently, the heavy-fermion state is largely suppressed in the high-field limit.

The field dependence of the γ value of $CeRu_2Si_2$ is rather similar to that observed for heavy-fermion UPt_3 ($\gamma = 430$ mJ/mol K^2), which has been investigated for fields up to 24.5 T.[14] In UPt_3 a metamagneticlike transition occurs at $B^* = 20$ T for a field direction in the hexagonal plane. Using an extrapolation procedure employing a $T^3 \ln(T/T^*)$ contribution to the specific heat, the field-induced mass enhancement at B^* has been estimated at 1.44 for $T \rightarrow 0$, which is of the same order as observed for $CeRu_2Si_2$ at 7.8 T. As the maximum field in these experiments was only 22% above B^*, a suppression of the heavy-fermion state was not yet observed ($\gamma = 503$ mJ/mol K^2 at 24.5 T).

From Fig. 4 it follows that the experimental results for fields above B^* can be described fairly well by Eq. (1). For sufficiently large applied magnetic fields, the Zeeman effect will reduce Eq. (1) to a Schottky expression for a two-level system. In the case of $CeRu_2Si_2$, one would need an applied field of the order of 100 T in order to establish the pure Zeeman effect as can be shown by Eq. (1). The weak decrease of T_Δ for large fields (Fig. 5) might be connected to this. The absolute values of B_{eff} depend on the choice of g_{eff}. Using the value for g_{eff} of 3.8,[17] it appears that B_{eff} is somewhat smaller than the applied field (Fig. 5). This might find its explanation in the presence of an internal compensating field caused by remaining antiferromagnetic interactions. By further increasing the magnetic field, B_{eff} is eventually expected to become equal to the applied magnetic field. Note that the difference between B_{eff} and the applied field as follows from Fig. 5 can be reduced by taking a somewhat lower value for g_{eff}. A value for g_{eff} of 3.2 results in a difference between B_{eff} and the applied field that is reduced to zero near 20 T, while this difference remains of the order of 2 T near 10 T.

One should bear in mind that the analysis of the specific-heat data above the metamagnetic transition, using a two-parameter fit to a simple single-resonance model, should be taken with some caution. From the analysis we infer that the resonance sharpens in high fields and effectively shifts away from the Fermi level. The combination of both effects leads to a reduction of the γ value in the high-field region. It is tempting to extrapolate the curves for T_Δ and B_{eff} versus B to $B = 0$ (see Fig. 5), in order to describe the zero-field Kondo properties of $CeRu_2Si_2$. However, we believe that such an extrapolation is not justified because in the low-field region the magnetic excitation spectrum[9] is rather complex, due to the presence of intersite and on-site contributions and their interplay. Furthermore, the neutron-scattering data[9] show that the intersite interactions are gradually suppressed near B^*, and thus it is likely that some antiferromagnetic interactions will remain present for fields somewhat above B^*. Consequently, the remaining antiferromagnetic interactions will contribute to the fit parameters T_Δ and B_{eff}. This contribution, which is difficult to estimate, will become weaker when the applied field is increased ($B > B^*$).

In summary, we have measured the specific heat of heavy-fermion $CeRu_2Si_2$ in strong magnetic fields along the tetragonal axis up to 20 T. The c/T values at the lowest temperature ($T = 1.5$ K) exhibit a pronounced peak at the metamagnetic transition at $B^* = 7.8$ T. Although at the maximum field (20 T) the heavy-fermion behavior is largely suppressed, a comparison with a simple single-resonance-level model indicates that substantially larger fields (of the order of 100 T) are needed to suppress the heavy-fermion behavior completely.

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