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Specific heat of UPt_3 in magnetic fields up to 24.5 T

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We report the first high-precision specific-heat measurements on the heavy-fermion compound UPt_3 , over a wide temperature ($1.4 \text{ K} < T < 40 \text{ K}$) and field range ($B < 24.5 \text{ T}$), thus covering the metamagnetic transition at 20 T. The data, taken on a single-crystalline sample, show a pronounced low-temperature maximum in the enhanced γ^* value at 20 T for a field direction in the hexagonal plane. We also investigate the presence of a $T^3 \ln(T/T^*)$ contribution to the specific heat in field.

INTRODUCTION

Among the heavy-fermion compounds, UPt_3 attracts much attention due to the unusual coexistence of strong spin-fluctuation effects and superconductivity (at $T \approx 0.5 \text{ K}$).¹ In the past few years experiments have made clear that superconductivity in UPt_3 is of an unconventional type and likely mediated by electron-electron interactions rather than by an electron-phonon mechanism. It is therefore of importance to investigate the magnetic interactions to a large extent. One of the interesting magnetic properties of hexagonal UPt_3 is the metamagnetic-like transition at 20 T that occurs at low temperatures only for field directions in the basal plane. This anomaly has first been observed in magnetization experiments² as a peak in the differential susceptibility, $\Delta M/\Delta H$. In subsequent magnetoresistivity experiments³ the anomaly turned up as a pronounced peak as well. The 20 T anomaly has furthermore been studied by magnetostriction,⁴ sound velocity,⁵ and Hall-effect experiments.⁶ Taking into account the variety of thermal, magnetic, transport, and alloying experiments performed on UPt_3 until now,⁷ we believe that the high-field anomaly is connected with a strong reduction of the antiferromagnetic fluctuations that were probed in zero field by inelastic neutron scattering experiments⁸ thus explaining the increase of magnetization and the peak of the resistance at 20 T. The field of 20 T is intimately connected to the temperature of about 17 K above which temperature the antiferromagnetic correlations lose their strength as at this temperature a maximum in the low-field susceptibility $\chi(T)$ is found.²

The specific heat of UPt_3 in zero field has been studied extensively.⁹⁻¹² The γ -value amounts to $420 \text{ mJ mol}^{-1} \text{ K}^{-2}$, which, in the Fermi-liquid model, leads to an effective quasiparticle mass of $\approx 180 m_e$.⁷ Evidence for spinfluctuations has been deduced from the presence of a $T^3 \ln(T/T^*)$ term.⁹ Of particular interest is the investigation of the γ value and the presence of a $T^3 \ln(T/T^*)$ term in an external magnetic field since little is known about the evolution of the heavy-fermion state in field. Specific heat measurements in field have

been performed until now by several authors indicating an enhancement of the heavy-fermion state with field, but in none of these experiments has the metamagnetic transition been probed thus far.

The aim of this paper is to present the first study of the specific heat of UPt_3 above the metamagnetic transition over a wide temperature ($1.4 < T < 40 \text{ K}$) and field range ($B < 24.5 \text{ T}$). The data are taken on a good quality single-crystalline sample with the field perpendicular and parallel to the hexagonal axis. Comparisons with previous experiments^{9,11,12} will be made. We also investigate the presence of the $T^3 \ln(T/T^*)$ term in field.

EXPERIMENTAL

The single-crystalline sample (mass 2.356 g) used in the specific-heat experiment was of a cubic shape ($5 \times 5 \times 5 \text{ mm}^3$) with the cubic axes parallel to the main crystallographic directions. The sample has been annealed and has been used previously in high-field magnetostriction measurements.⁴

High-field specific-heat measurements have been performed at the High Field Magnet Laboratory¹³ of the University of Nijmegen, using a 20 T Bitter-type coil and the 25 T hybrid magnet (superconducting magnet 8 T—Bitter coil 17 T). The experiments were performed adiabatically with a sapphire sample holder equipped with a ruthenium-oxide thermometer (ALPS Electric Co. Ltd., 10 k Ω nominal resistance) and a nickel-chromium film as a heater. The heat capacity of the sample holder (mass of about 120 mg) is around 5% of the heat capacity of the UPt_3 sample near 20 K and is negligible at temperatures below 5 K. The ruthenium-oxide thermometer has been calibrated below 30 K in the full field range by means of a field-insensitive capacitance thermometer. At the lowest temperatures (1.4 K), where $R^{-1}dR/dT \approx -0.5 \text{ K}^{-1}$, the field effect (for 24.5 T) on the resistance of the sensor was less than 6%, decreasing smoothly to 0.5% at 30 K where values of $R^{-1}dR/dT$ are -0.003 K^{-1} . The sample holder was mounted in a frame made out of stycast with thin copper wires embedded in it, in order to achieve short thermal equilibrium times. The sample

could be cooled by means of a mechanical heat switch that could bring a thin copper wire connected to the sapphire plate into contact with the (pumped) helium bath.

Magnetic-field ripples associated with the current-driven Bitter-type magnets disturb the specific-heat measurements in several ways:¹⁴ undesired heating of the sample by eddy currents and generation of electrical noise in the temperature-sensor circuit. To reduce the effect of field ripples, the specific-heat cell was mounted in a cylinder of the new high- T_c superconducting material $\text{YBa}_2\text{Cu}_3\text{O}_7$. A considerable reduction of the noise level could be realized up to the highest fields.¹⁵ More details of the experimental setup will be published elsewhere.¹⁶

RESULTS

The specific heat has been measured in zero field and in magnetic fields of 10, 14, 16, 18, 20, 21, 23, and 24.5 T, applied along the a axis in the hexagonal plane. Characteristic curves of the specific heat of UPt_3 are shown in Fig. 1 in a plot of c/T versus T . First of all, we like to point to the high accuracy with which the experiments could be performed even in fields of 24.5 T. For magnetic fields of 0 T and 20 T the specific heat has been measured up to 40 K. Above 25 K, the experimental data for the two field values almost coincide. Below 25 K, a negative field effect on c/T is observed down to the crossover temperature that is about 3.5 K around 20 T. Below the crossover temperature, positive field effects are observed, the largest at 20 T where at 1.8 K an increase of c/T of $100 \text{ mJ mol}^{-1} \text{ K}^{-2}$ is found. Above 20 T a depression of the heavy-fermion ground state begins. Experiments with fields up to 12 T, applied parallel to the hexagonal axis, show no significant variation in the specific heat with magnetic field. This is in agreement with the result reported by Stewart *et al.*¹⁷

The 20 T anomaly is clearly present in the magnetocaloric effect. As a demonstration of the magnetocaloric effect we show in Fig. 2 the sample temperature during a magnetic-field sweep from 8 T up to 24.5 T (sweep time

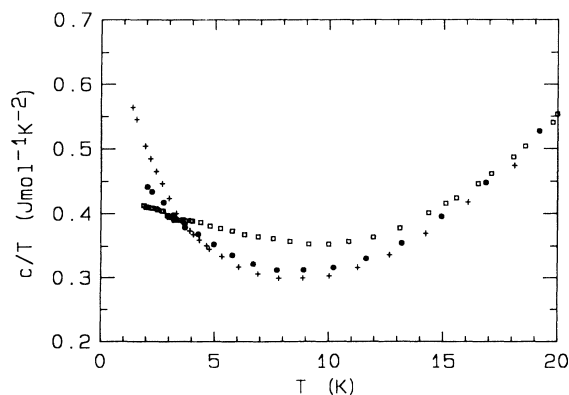


FIG. 1. The specific heat of single-crystal UPt_3 in a plot of c/T vs T at different field strengths: \square (0 T); $+$ (20 T); \bullet (24.5 T); fields have been applied along the a axis in the hexagonal plane.

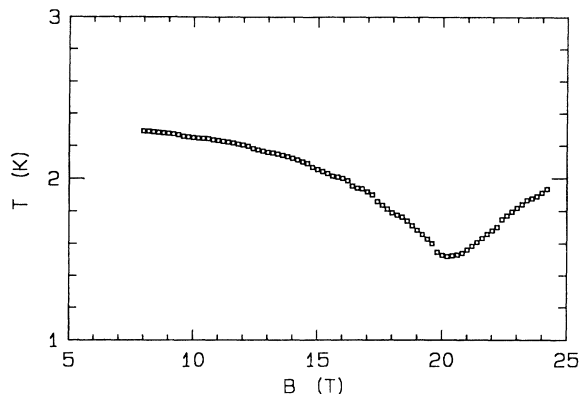


FIG. 2. The magneto-caloric effect of an adiabatically mounted single-crystalline UPt_3 sample with the field B applied parallel to the a axis; the shift in temperature as a function of the applied field can be evaluated by the thermodynamic relation: $\Delta T = -(TB/c)\partial M/\partial T$, with M the sample magnetization and c the specific heat.

5.5 min). In principle, these magnetocaloric data can be used to deduce the field effect on the specific heat, provided that the temperature dependence of the magnetization is known for all field values. In the present investigation the magnetocaloric effect has not been analyzed in more detail. In employing an adiabatic method for specific-heat measurements of UPt_3 in field, we can make use of the field-cooling effect, that originates from the positive temperature derivative of the susceptibility at the lowest temperatures. Due to this effect, the starting temperature of 1.8 K at 8 T could be reduced to 1.2 K in the 20 T experiment.

ANALYSIS

For analyzing the specific-heat data of UPt_3 in zero field the expression

$$\frac{c}{T} = \gamma^* + \beta T^2 + \delta T^2 \ln(T/T^*) = \gamma^* + \beta^* T^2 + \delta T^2 \ln T \quad (1)$$

has been used by several authors.⁹⁻¹² In this expression γ^* is an enhanced electronic coefficient, β is the usual phonon coefficient, and δ is the coefficient of the $T^3 \ln(T/T^*)$ term which will be discussed below. The coefficient β^* is defined as $\beta^* = \beta - \delta \ln T^*$, where T^* is a characteristic temperature.

In order to compare quantitatively the different sets of specific-heat data, we summarize in Table I the values for the coefficients γ^* , β^* , and δ from Refs. 9, 11, and 12, including our results. Equation (1) gives a good description of the zero-field specific heat of UPt_3 for temperatures below 5 K, that is to say, in all cases the deviations between the calculated and the as-measured data is smaller than the experimental error ($\sim 1\%$). However, as follows from Table I, there is some spread in the values for γ^* , β^* , and δ , which possibly indicates a sample dependence.

Our main concern in the present analysis is to determine the variation of γ^* with field. Since the experimen-

TABLE I. Comparison of the fit coefficients from Eq. (1) applied to the specific-heat data of UPT_3 below 5 K.

Sample	B (T)	γ^* ($\text{mJ mol}^{-1} \text{K}^{-2}$)	β^* ($\text{mJ mol}^{-1} \text{K}^{-4}$)	δ ($\text{mJ mol}^{-1} \text{K}^{-4}$)	T interval (K)
Data from de Visser ^a	0	424	-4.50	1.85	1.4-5
Brodale ^b	0	426	-6.92	3.22	0.5-4
Fisher ^c					
Sample 1	0	434	-8.92	2.89	0.5-5
Sample 2	0	430	-7.00	2.88	0.5-5
This work	0	427	-6.0	2.5	1.8-5
	20	624	-46	22	
	24.5	503	-21	9.5	

^aReference 9.

^bReference 11.

^cReference 12.

tal data are limited to 1.8 K an extrapolation procedure is needed to obtain γ^* . In a first attempt to have a handle on $\gamma^*(B)$ we have applied Eq. (1) to the data in the interval $1.8 \text{ K} < T < 5 \text{ K}$. Actually, it turns out that Eq. (1) gives also a fairly good description of our nonzero field results. Again deviations of the experimental data from the fit are below 1% for all field values. As an example for the quality of the fit we show in Fig. 3 a plot of $(c/T - \gamma^*)/T^2$ versus $\ln T$ for the experimental data in 0, 20, and 24.5 T, using the γ^* values reported in Table I. The β^* and δ values deduced from the data in Fig. 3 are listed in Table I as well. Having determined the γ^* values for each investigated magnetic field by the fitting procedure described above, we are able to construct $\gamma^*(B)$. The result is shown in Fig. 4, which gives evidence that γ^* passes through a pronounced maximum at the metamagnetic transition. Interestingly, also the coefficients β^* and δ have their extreme values at 20 T. We will come back to this later.

As a check of the extrapolation procedure we next consider the involved entropy for different values of B . In zero field the electronic entropy amounts to approximate-

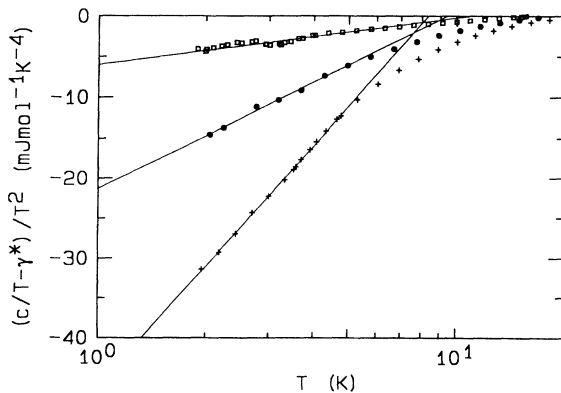


FIG. 3. The specific heat plotted as $(c/T - \gamma^*)/T^2$ vs $\ln T$; the solid curves represent fits to Eq. (1) of the data presented in Fig. 1 at different fields: \square (0 T); $+$ (20 T); \bullet (24.5 T). Values for γ^* are given in Table I.

ly $R \ln 2$ at 20 K. In Fig. 5 we present $\Delta c/T = c/T|_{(B)} - c/T|_{(B=0)}$ for B equal to 20 and 24.5 T. Below 5 K these curves have been constructed using Eq. (1) (with the parameters as listed in Table I), whereas above 5 K the experimental data are used. Similar curves can be constructed for the other investigated field values (not shown). The entropy difference between the field curves and the $B=0$ curve is given by the integral over $\Delta c/T$. The main contribution to this integral is found below $\sim 20 \text{ K}$. As a result it appears that for both curves the negative contribution is at least 30% larger than the positive one. The apparent lack of entropy in field at $\sim 20 \text{ K}$ implies that (i) the entropy is shifted towards higher temperatures and smeared out over a wide temperature range given the fact that the experimental data for $B=0$ and $B=20 \text{ T}$ nearly coincide above 20 K, and/or (ii) the extrapolation procedure leads to too small

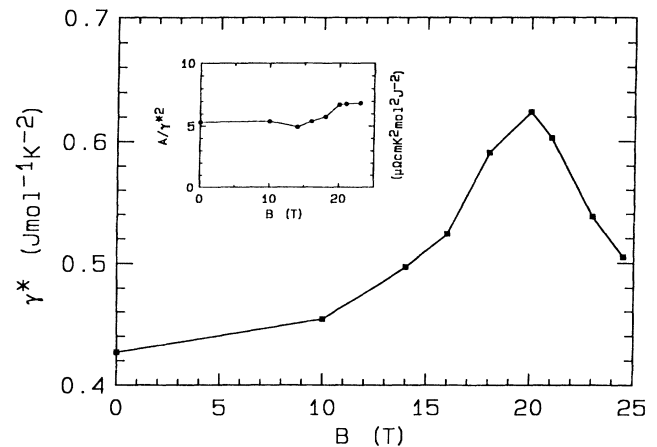


FIG. 4. Result of an analysis of the specific-heat data for UPT_3 (partly shown in Fig. 1) with the expression $c/T = \gamma^* + \beta^* T^2 + \delta T^2 \ln T$; values for the coefficient γ^* are shown as a function of the magnetic field applied along the a axis. The inset shows a plot of the field dependence of the ratio A/γ^{*2} where A is the coefficient of the T^2 term in the expression $\rho(T) = \rho_0 + AT^2$. The resistivity data are from Ref. 25.

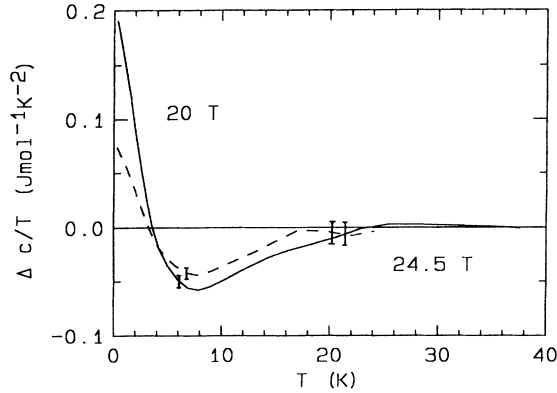


FIG. 5. The difference between c/T values in an applied field of 20 T (solid curve) and 24.5 T (dashed curve) and those in zero field; below 5 K the curves follow Eq. (1) with values of the coefficients according to Table I; above 5 K, the curves represent smooth curves through the data points; error bars for different temperature regions are indicated.

values of γ^* in field. The latter point strongly suggests that $\gamma^*(B)$ attains an even larger value at 20 T than shown in Fig. 4. Clearly very low temperature measurements are needed to settle this problem definitely.¹⁸

DISCUSSION

The most remarkable results that can be deduced from the present experiments are (i) the initial increase of $\gamma^*(B)$, (ii) the pronounced maximum of γ^* at the metamagnetic transition, and (iii) the still large value of γ^* above the transition. These features clearly underline the particular heavy-fermion properties of UPt_3 , and contrast with the relatively rapid suppression of the heavy-fermion state in most of the cerium-based Kondo-lattice systems. In polycrystalline CeCu_6 , for instance, the γ value is reduced from $1500 \text{ mJ mol}^{-1} \text{ K}^{-2}$ in zero field to $350 \text{ mJ mol}^{-1} \text{ K}^{-2}$ in 24 T.¹⁹ For CeAl_3 and CeCu_2Si_2 , a similar (but smaller) reduction of γ has been reported for fields up to 23 T.²⁰

Elaborate theoretical models that account for field effects on the density of states of heavy-fermion systems are not available at present. In general, one might expect positive as well as negative field effects on γ^* depending on the relative position of the Fermi level in the density of states curve and the detailed structure of this curve. In the case of UPt_3 , the initial positive variation of γ^* with B is likely related with an enhancement of the antiferromagnetic fluctuations, i.e., a full excitation of the spinfluctuation spectrum at a lower temperature. The subsequent quenching of the antiferromagnetic correlation above B^* leads then to a reduction of γ^* . In this respect, it is interesting to compare UPt_3 with CeRu_2Si_2 . This tetragonal heavy-fermion compound ($\gamma = 350 \text{ mJ mol}^{-1} \text{ K}^{-2}$) exhibits a metamagnetic-like transition at 8 T for a field direction along the c axis.²¹ Also here, an increase of γ with field is observed²² (about 20% near the metamagnetic transition). Inelastic neutron scattering experiments²³ on CeRu_2Si_2 have revealed that two types of

fluctuations are present at low temperatures: (i) local on-site fluctuations of the Kondo-type and (ii) intersite fluctuations giving rise to antiferromagnetic correlations. Neutron-scattering experiments in fields²³ yield a strong reduction of the intersite fluctuations at about 8 T at which field the metamagnetic-like transition occurs.

The striking similarities between the magnetic properties of UPt_3 and CeRu_2Si_2 (Ref. 21) strongly suggest that the metamagnetic-like transition has a very similar origin in both compounds. This would imply a scenario with at least two mechanisms that contribute to the specific heat. Firstly, the on-site Kondo-type fluctuations, which give rise to the main part of the specific heat in zero field, and that persist up to the upper field limit of the present experiments, and secondly, the intersite fluctuations, which give rise to a steadily growing contribution in lower fields, and rapidly lose their strength above B^* . Unfortunately, neutron-scattering data in strong magnetic fields on UPt_3 are not available, so that the precise microscopic mechanism for the 20 T transition remains to be confirmed.

In order to compare the magnetic interactions and their contribution to the density of states above and below the metamagnetic transition it is of interest to calculate the Wilson ratio $\chi/\gamma(B)$. However, also detailed measurements of the temperature variation of the magnetization at fixed fields are lacking. Another important parameter to investigate is the ratio A/γ^{*2} ,²⁴ where A is the coefficient of the term quadratic in temperature, that is found in the resistivity of UPt_3 below 2 K.⁷ The field variation of the coefficient A has been measured by Remenyi *et al.*²⁵ Combining our values of $\gamma^*(B)$ with their data we have calculated A/γ^{*2} up to 23 T (see insert in Fig. 4). The result shows that A/γ^{*2} remains nearly constant up to 20 T, whereas above 20 T a 25% increase is observed, apparently indicating the entrance to a different interaction regime. However, care should be taken when interpreting this result since A has been determined in the temperature regime below 2 K, whereas γ^* has been obtained by extrapolations from the data above 1.8 K.

Next we discuss the use of Eq. (1) in order to determine γ^* . The presence of a $T^3 \ln(T/T^*)$ contribution to the specific heat is a general consequence of many-body interactions as it results within different theoretical frameworks: (i) the paramagnon approach for ferromagnetic spinfluctuations,²⁶ (ii) quasiparticle interactions with small momentum transfer processes in the Fermi-liquid approach,²⁷ and (iii) a Fermi-liquid theory of the Kondo lattice with the $1/N$ -Kondo-boson expansion.²⁸ Recently the specific heat of UPt_3 has been interpreted by an extended paramagnon model including Fermi-surface geometry by Ihle and Fehske.²⁹ The Fermi-liquid model has been explored in detail by Coffey and Pethick.³⁰ Both types of analysis justify the use of a $T^3 \ln(T/T^*)$ term in the specific heat. In the Fermi-liquid picture, the enhancement of γ^* corresponds to an enhancement of the effective mass, whereas δ yields information about the interaction between the quasiparticles via the Landau parameter A_0^a . The $T^3 \ln(T/T^*)$ term can thus not be used to discriminate between the different theories. On the

other hand the aforementioned mechanisms for the low-energy fluctuations in UPt_3 are both consistent with the observations of a $T^3 \ln(T/T^*)$ term.

From a fundamental point of view it would be of interest to investigate the field dependence of the parameters in Eq. (1)— γ^* , β^* , and δ —all parameters having their extreme values at ~ 20 T. Using literature data for β we can deduce values for the characteristic temperature T^* of ~ 12 K in zero field down to ~ 9 K above B^* . We realize that the auxiliary parameter T^* is only a factor of two larger than the maximum fit temperature (~ 5 K). This problem has been encountered before and is discussed elsewhere.^{7,30} We emphasize again that very-low temperature measurements are needed to investigate the presence of the $T^3 \ln(T/T^*)$ term in field in more detail. We hope, however, that the present experimental results trigger further theoretical investigations of the variation of γ^* , β^* , and δ with field.

CONCLUSIONS

The high-field anomaly near 20 T, previously observed in magnetization, magnetostriction, magneto-transport and magneto-acoustic measurements, clearly shows up in the specific-heat measurements on UPt_3 in magnetic fields

up to 24.5 T. As a main result we find that the γ^* value steadily increases with field and passes through a pronounced maximum at the metamagneticlike transition. In order to derive $\gamma^*(B)$ we used an extrapolation procedure including a $T^3 \ln(T/T^*)$ term that represents the experimental data over the full field range sufficiently well below 5 K. Nevertheless, from entropy considerations some uncertainties remain about the appropriateness of this expression at high fields for temperatures below 1.5 K. This strongly urges the need for measurements at very low temperatures. The positive field effect on the specific heat of UPt_3 contrasts with the negative field effects for other heavy-fermion compounds, with an exception for CeRu_2Si_2 . This underlines the special features of the heavy-fermion states of UPt_3 and CeRu_2Si_2 . Finally, we mention the large magnetocaloric effect that permits considerable field cooling of UPt_3 samples in the low-temperature range.

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