

## FIELD EFFECT ON THE SPECIFIC HEAT OF $UPt_3$

H.P. VAN DER MEULEN<sup>a</sup>, Z. TARNAWSKI<sup>a,1</sup>, J.J.M. FRANSE<sup>a</sup>, J.A.A.J. PERENBOOM<sup>b</sup>,  
D. ALTHOF<sup>b</sup> and H. VAN KEMPEN<sup>b</sup>

<sup>a</sup>*Natuurkundig Laboratorium, Universiteit van Amsterdam, Valckenierstraat 65, 1018 XE Amsterdam, The Netherlands*

<sup>b</sup>*High Field Magnet Laboratory, University of Nijmegen, Toernooiveld, 6525 ED Nijmegen, The Netherlands*

Specific heat measurements have been performed on a single-crystalline sample of the heavy-fermion compound  $UPt_3$  in magnetic fields up to 24.5 T. The specific heat has been measured in the temperature range 1.8–40 K with an adiabatic method. The  $c/T$  values extrapolated to zero temperature for fields applied perpendicular to the hexagonal axis show a pronounced maximum near 20 T. This maximum was previously observed in measurements of differential susceptibility and magnetoresistivity. The field effect in the specific heat is compared with the thermodynamically related temperature dependence of the susceptibility.

### 1. Introduction

In the heavy fermion compound  $UPt_3$  high field studies of the magnetization at low temperatures have revealed a metamagnetic-like transition at 20 T [1]. This 20 T anomaly has also been observed in measurements of the magnetoresistance, volume magnetostriction [2], the Hall effect [3], and the sound velocity and the acoustic damping [4]. The transition is most probably related to the breaking of short-range anti-ferromagnetic correlations that have been observed in neutron scattering experiments [5, 6]. In addition, the neutron experiments have revealed magnetic ordering below 5 K, in which uranium moments of the order of  $0.02\mu_B$  are involved [7]. In the specific heat of  $UPt_3$  hardly any sign of magnetic ordering can be observed [1]. Substitution of a few percent of U by Th or Pt by Pd, easily introduces long-range antiferromagnetic order with maximum values for the ordering temperature of 6 K [8, 9], and values for the moment per uranium atom of about  $0.5\mu_B$ . This order is clearly observed in the specific heat of these compounds in the form of a well-pronounced peak [8, 10].

In most of the cerium-based heavy fermion compounds it has been observed that the heavy fermion state is suppressed relatively rapidly with increasing field [11–13]. The compound  $CeRu_2Si_2$ , however, shows a different behaviour since an enhancement of  $\gamma$  with field is reported [14]. The increase of  $\gamma$  with field is about 20% near the metamagnetic-like transition

that occurs at 8 T for a field direction along the tetragonal axis [15]. Inelastic neutron-scattering experiments on this compound 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 anti-ferromagnetic correlations. Neutron-scattering experiments in field 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  $CeRu_2Si_2$  [15] strongly suggest that the metamagnetic-like transition has a very similar origin in both compounds. 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 a recent paper [16] we have reported on specific-heat experiments on  $UPt_3$  in magnetic fields up to 24.5 T in the temperature range 1.8–40 K. In this contribution we present some details of this investigation and concentrate on the thermodynamic relation between the field effect of the specific heat and the temperature dependence of the susceptibility.

For analysing the data of the zero-field specific heat measurements, the following expression, proposed by several authors [17, 18], is used:

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

where  $\gamma^*$  is an enhanced electronic coefficient and where  $\beta$  is the usual phonon coefficient. The presence of a  $T^3 \ln(T/T^*)$  contribution to  $c$  seems to be a

<sup>1</sup> On leave from Solid State Physics Dept. Academy of Mining and Metallurgy, al Mickiewicza, 30, 30-059 Krakow, Poland.

logical consequence of many-body interactions as it results within different theoretical frameworks: i) the paramagnon approach for ferromagnetic spinfluctuations [19], ii) quasiparticle interactions with small momentum transfer processes in the Fermi-liquid approach [20], and iii) a Fermi liquid theory of the Kondo lattice with the  $1/N$ -Kondo-boson expansion [21]. An extended paramagnon model including Fermi surface geometry has recently been used by Ihle and Fehske [22] to interpret the specific heat data of  $UPt_3$ , including pressure effects. The Fermi liquid model has been explored in detail by Coffey and Pethick [23]. 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  $\gamma^*$  corresponds to an enhancement of the effective mass, whereas  $\delta$  yields information about the interaction between the quasiparticles via the Landau parameter  $A_0^a$ . In the analysis we introduce the parameter  $\beta^*$  which is composed of two terms  $\beta^* = \beta - \delta \ln T^*$ , where  $\beta$  is the coefficient of the cubic phonon term in  $c$  and  $T^*$  a characteristic temperature for the quasiparticle interaction.

## 2. 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.

High-field specific heat measurements have been performed at the High Field Magnet Laboratory [24] of the University of Nijmegen, using the 20 T Bitter-type of coil and the 25 T hybrid magnet (superconducting magnet 8 T, Bitter coil 17 T). The experiments were performed in an adiabatic way with a sapphire sample holder equipped with a ruthenium-oxide thermometer and a nickel-chromium film as heater. Details of the experimental set up and of the  $YBa_2Cu_3O_7$ -shield used to shield the magnetic field ripples of the Bitter magnet are to be found in ref. [16].

## 3. Results and discussion

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. For magnetic fields of 0 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. The experimental results of the specific heat of  $UPt_3$  are shown in fig. 1 in a plot of  $c/T$  versus  $T$  for temperatures below 10 K. Starting at 10 K the  $c/T$  values start to decrease with applied magnetic fields down to the cross-over temperature that is about 3.5 K around 20 T. Below the cross-over 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-firmion ground state starts. This is in agreement with the results recently obtained by Müller et al. [25]. 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. [12].

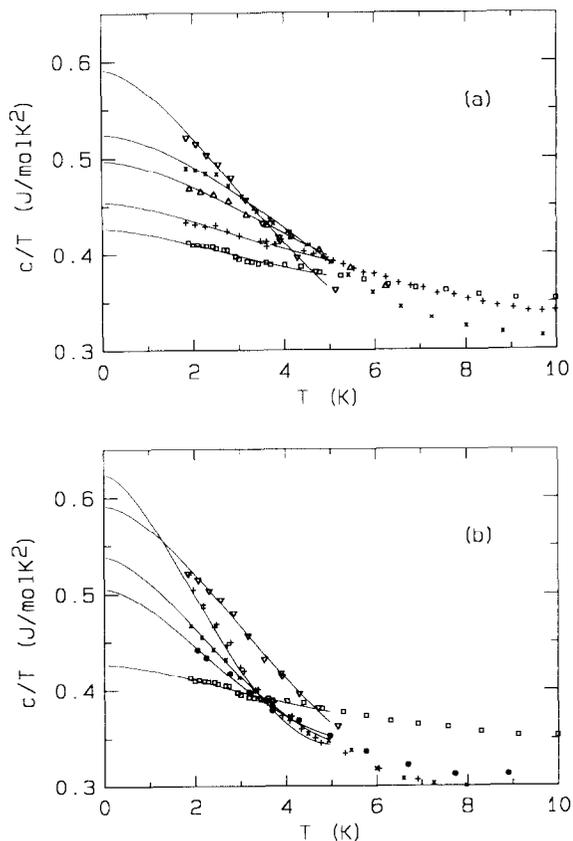


Fig. 1. The specific heat of single-crystalline  $UPt_3$  in a plot of  $c/T$  vs.  $T$  at different field strengths. (a):  $\square$  (0 T),  $+$  (10 T),  $\triangle$  (14 T),  $\times$  (16 T),  $\nabla$  (18 T). (b):  $\square$  (0 T),  $\nabla$  (18 T),  $+$  (20 T),  $\times$  (23 T),  $\bullet$  (24.5 T). Fields have been applied along the  $a$ -axis in the hexagonal plane. The full curves represent fits to eq. (1) at different fields, extrapolated to  $T=0$ .

Equation (1) has been applied to the specific heat data in zero field by different authors (see the discussion in ref. [16]). We can conclude from these references that a fit must be limited to the data below 5 K. Still sample dependencies in the values of  $\beta^*$  and  $\delta$  can be observed as well as quite different results between different groups.

In our case eq. (1) is also applied to specific heat data measured in magnetic field. Deviations of the experimental data from the fit with eq. (1) up to 5 K are below 1% for all field values. The values of  $\beta^*$  and  $\delta$  both have their extreme values at 20 T but still have an error of 10% over the whole field range. According to our experience with the fitting procedures in the limited temperature interval 1.8–5 K we estimate the accuracy of  $\gamma^*$  to be of the order of 1–2%. In fig. 2 we present the field dependence of the coefficient  $\gamma^*$ . A pronounced maximum is found at 20 T. The present zero-field value for the coefficient  $\gamma^*$  is very much the same as that reported by other authors.

By using thermodynamic relations we can connect the specific heat in field with the temperature dependence of the susceptibility. From the appropriate Maxwell relation we derive:

$$\frac{\partial c/T}{\partial B} = \frac{\partial^2 M}{\partial T^2} = H \frac{\partial^2 M/H}{\partial T^2} . \quad (2)$$

Measurements of  $M/H$  at low field show a maximum at 19 K and a quadratic increase with temperature at the low temperature side [1], see fig. 3. According to eq. (2) this implies an enhancement of the specific

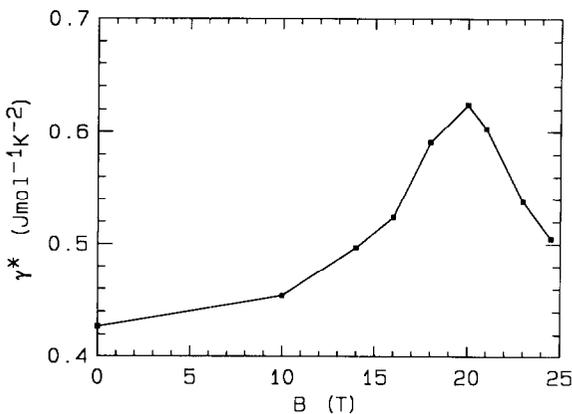


Fig. 2. The coefficient  $\gamma^*$  as a function of the magnetic field applied along the  $a$ -axis, resulting from an analysis of the specific heat data for  $UPt_3$  (shown in fig. 1) with the expression  $c/T = \gamma^* + \beta^*T^2 + \delta T^2 \ln T$ .

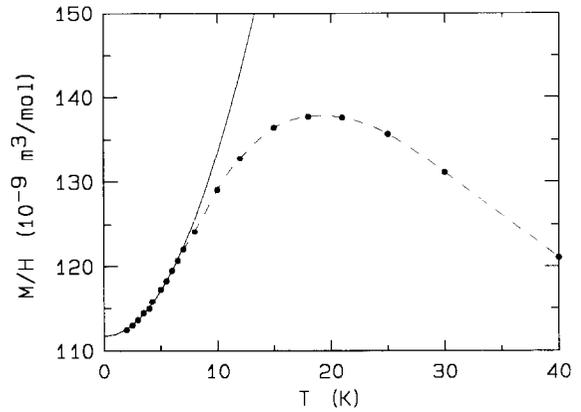


Fig. 3. The temperature dependence of the susceptibility  $M/H$  of  $UPt_3$  measured at a field of 1.3 T parallel to the  $a$ -axis. The full curve represents a fit to  $M(T)/H = M(0)/H + aT^2$ , the broken curve is a smooth line connecting the datapoints.

heat at low temperatures with field, as is confirmed by the measurements shown in fig. 1. Again according to eq. (2), we expect the field derivative of  $c/T$  to change sign at the inflection point of the  $M/H-T$  curve. From fig. 3 we deduce a temperature of about 8 K for this inflection point. Previous specific heat experiments by Frings et al. [1] indicate a crossing of specific heat curves in zero field and in 5 T near 7 K. In our case we observe a crossing of the  $c(10 T)/T$  and  $c(0 T)/T$  curves again near 7 K. Apparently below 10 T, there is hardly any field effect on the temperature where  $\partial(c/T)/\partial B$  changes sign. The temperature of the inflection point in the  $M/H-T$  curve and the temperature where the field effect on the specific heat is zero, almost coincide. At further increasing the field above 10 T, the crossover of the  $c(B)/T$  and  $c(0 T)/T$  curves occurs at lower temperatures, indicating a shift to lower temperatures of the inflection point in the  $M(H)/H-T$  curve.

In a quantitative analysis we deduce from the specific heat data the change  $\Delta c(T=0)/T$  to be  $27 \times 10^{-3} J/mol K^2$ , where  $\Delta c = c(10 T) - c(0 T)$ . Using a low temperature quadratic fit to the data of fig. 3 (obtained for a low field of 1.3 T) we calculate with eq. (2) a value for  $\Delta c(T=0)/T$  of  $35 \times 10^{-3} J/mol K^2$ , in satisfactory agreement with the analysis of the specific heat data. In summary we conclude that a thermodynamic analysis of the field effect of the specific heat and the temperature dependence of the susceptibility leads to a consistent picture of the thermomagnetic measurements on  $UPt_3$ .

### Acknowledgements

Part of this work was supported by the "Stichting voor Fundamenteel Onderzoek der Materie" (FOM) with financial support from the "Nederlandse Organisatie voor Wetenschappelijk Onderzoek" (NWO).

### References

- [1] P.H. Frings, J.J.M. Franse, F.R. de Boer and A. Menovsky, *J. Magn. Magn. Mat.* 31-34 (1983) 240.  
J.J.M. Franse, *J. Magn. Magn. Mat.* 31-34 (1983) 819.  
P.H. Frings and J.J.M. Franse, *Phys. Rev. B* 31 (1985) 4355.
- [2] A. de Visser, L. Puech, W. Joss, A. Menovsky and J.J.M. Franse, *Jpn. J. Appl. Phys.* 26 S26-3 (1987) 513.
- [3] M. van Sprang, R.A. Boer, A.J. Riemersma, L.W. Roeland, A. Menovsky, J.J.M. Franse and J. Schoenes, *J. Magn. Magn. Mat.* 76 & 77 (1988) 229.
- [4] I. Kouroudis, D. Weber, M. Yoshizawa, B. Lüthi, L. Pucch, G. Bruls, U. Welp, J.J.M. Franse, A. Menovsky, E. Bucher and J. Hufnagel, *Phys. Rev. Lett.* 58 (1987) 820.
- [5] G. Aeppli, A.I. Goldman, G. Shirane, E. Bucher and M.Ch. Lux-Steiner, *Phys. Rev. Lett.* 58 (1987) 808.
- [6] P.H. Frings, B. Renker and C. Vettier, *Physica B* 151 (1988) 499.
- [7] G. Aeppli, E. Bucher, G. Broholm, J.K. Kjems, J. Bauman and J. Hufnagel, *Phys. Rev. Lett.* 60 (1988) 615.
- [8] A. de Visser, J.C.P. Klaasse, M. van Sprang, A. Menovsky, J.J.M. Franse and T.T.M. Palstra, *J. Magn. Magn. Mat.* 54-57 (1986) 375.
- [9] A.P. Ramirez, B. Batlogg, A.S. Cooper and E. Bucher, *Phys. Rev. Lett.* 57 (1986) 1072.
- [10] K. Kadowaki, M. van Sprang, J.C.P. Klaasse, A. Menovsky, J.J.M. Franse and S.B. Woods, *Physica B* 148 (1987) 22.
- [11] G.R. Stewart, B. Andraka, C. Quitmann, B. Treadway, Y. Shapira and E.J. McNiff, Jr., *Phys. Rev. B* 37 (1988) 3344.
- [12] G.R. Stewart, Z. Fisk, J.L. Smith, J.J.M. Franse, A. Menovsky and B.L. Brandt, *J. Magn. Magn. Mat.* 76 & 77 (1988) 484.
- [13] B. Andraka, G. Fraunberger, J.S. Kim, C. Quitman and G.R. Stewart, *Phys. Rev. B* 39 (1989) 6420.
- [14] R.A. Fisher, N.E. Phillips, C. Marcenat, J. Flouquet, P. Haen, P. Lejay and J.-M. Mignot, *Proc. Inter. Conf. Magn., J. de Phys.* 49 (1988) C8-759.
- [15] P. Haen, J. Flouquet, F. Lapiere, P. Lejay and G. Remenyi, *J. Low Temp. Phys.* 67 (1987) 391.
- [16] H.P. van der Meulen, Z. Tarnawski, J.J.M. Franse, J.A.A.J. Perenboom, D. Althof and H. van Kempen, to be published.
- [17] A. de Visser, J.J.M. Franse, A. Menovsky and T.T.M. Palstra, *Physica B* 127 (1984) 442.
- [18] G.R. Stewart, *Rev. Mod. Phys.* 56 (1984) 755.
- [19] W.F. Brinkman and S. Engelsberg, *Phys. Rev.* 169 (1968) 417.
- [20] C.J. Pethick and G.M. Carneiro, *Phys. Rev. A* 7 (1977) 304.
- [21] A. Auerbach and K. Levin, *Phys. Rev. B* 34 (1986) 3524.
- [22] D. Ihle and H. Fehske, *Phys. Rev. B* 39 (1989) 2106.
- [23] D. Coffey and C.J. Pethick, *Phys. Rev. B* 33 (1986) 7508.
- [24] K. van Hulst and J.A.A.J. Perenboom, *IEEE Trans. Magn. MAG-24* (1988) 1397.  
K. van Hulst, C.J.M. Aarts, A.R. de Vroomen and P. Wyder, *J. Magn. Magn. Mat.* 11 (1979) 317.
- [25] T. Müller, W. Joss and L. Taillefer, *Phys. Rev. B* 40 (1989) 2614.