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Metamagnetic transition of UPt_3 under pressure

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The effect of hydrostatic pressure ($P \leq 5.1$ kbar) on the metamagnetic transition of heavy-fermion UPt_3 has been investigated by means of high-field magnetoresistance measurements ($B \leq 28$ T) at a temperature of 2.0 K. The transition, which occurs at ambient pressure for a field directed in the hexagonal plane at $B^* = 21$ T, shifts upwards with pressure at a rate of 0.60 T/kbar. The deduced magnetic Grüneisen parameter, $\Gamma_B = -\partial \ln B^*/\partial \ln V$ equals 59. The value for Γ_B falls well within the range of values reported for the thermal Grüneisen parameter, $52 < \Gamma_T = -\partial \ln T^*/\partial \ln V < 71$, which verifies directly a close relation between the thermal and magnetic energy scales.

The intermetallic compound UPt_3 belongs to the select class of heavy-fermion superconductors and attracts much attention because of its unrivalled low-temperature properties. The normal state is characterized by a large coefficient of the linear term in the specific heat[1,2] ($\gamma = 420$ mJ/mol K^2), which gives rise to a description in the Fermi-liquid picture with a quasiparticle mass of ~ 200 times the free-electron mass. The strong mass renormalization is brought about by competing magnetic interactions that develop in the liquid-helium temperature range. Surprisingly, a superconducting ground state[3] ($T_c = 0.5$ K) is formed in the strongly interacting electron liquid. Therefore, it has been suggested[4] that the electron-electron interactions mediate superconductivity, instead of the normal electron-phonon mechanism. Furthermore, the unusual superconducting properties, like power-law behavior for the electronic excitation energy[5] and the occurrence of a complex superconducting phase diagram,[6] evidence unconventional (nonsinglet) superconductivity.

One of the intriguing normal-state properties of UPt_3 is the occurrence of a metamagneticlike transition at $B^* \approx 21$ T at liquid-helium temperatures for a magnetic field direction in the hexagonal plane. This anomaly was first observed in high-field magnetization measurements,[1] where it appeared as a sharp maximum in the differential susceptibility, $\Delta M/\Delta H$. In subsequent magnetoresistance data[7] the anomaly turned up as a sharp peak as well. The 21-T anomaly has furthermore been detailed by magnetostriction,[8] sound velocity,[9] Hall effect,[10] and high-field specific-heat[11] measurements. Considering the variety of thermal, magnetic, transport, and alloying studies performed for UPt_3 so far,[12] we believe that the 21-T anomaly is connected with a strong reduction of the antiferromagnetic intersite correlations, that were probed (in zero field) by inelastic neutron-scattering experiments.[13] In particular, the maximum in the magnetoresistance and the increase in the magnetization at B^* provide evidence for such an interpretation. The magnetic susceptibility,[1] $\chi(T)$, shows a maximum at a temperature of $T_{max} = 17$ K, above which the antiferromagnetic correlations become much weaker. The magnetic energy $\mu_B B^*$ is closely related to the thermal energy $k_B T_{max}$. Note that we term the anomaly at B^* as metamagneticlike, because no large static magnetic moments are found. Alloying experiments have revealed that UPt_3 is close to an antiferromagnetic instability. For instance, by alloying with Pd long-range antiferromagnetic order appears[12] with a maximum Néel temperature of $T_N = 5.8$ K and an ordered moment of $0.6\mu_B$ per U atom for $U(Pt_{0.95}Pd_{0.05})_3$. Interestingly, the magnetization increase at B^* for pure UPt_3 amounts also to $0.6\mu_B$ per U atom, which suggests a common origin of the alloying-induced and field-induced moment. Recently, antiferromagnetism with an extremely small ordered moment ($0.02\mu_B$ per U atom) was detected below $T_N = 5$ K by neutron diffraction.[13] How this tiny moment relates to the large alloying and field-induced moment is still a puzzling aspect of the magnetic properties of UPt_3.

The heavy-electron bands are very sensitive to volume (and shape) effects, as follows from the extremely large thermal Grüneisen parameters,[14,15] $\Gamma_T = -\partial \ln T^*/\partial \ln V$, that are roughly 2 orders of magnitude larger than for ordinary metals. Here T^* is the characteristic temperature of the heavy-fermion resonance which is of the order of several kelvin. In the case of UPt_3 Γ_T is of the order of 60.[14,15] The close connection between the thermal and magnetic energy scales suggests that the relevant free-energy term can be written as $F = F(T/T^*(V), B/B^*(V))$. This implies that the thermal and magnetic properties can be scaled by one single volume-dependent energy scale[16,17] and that the thermal and magnetic Grüneisen parameters ($\Gamma_B = -\partial \ln B^*/\partial \ln V$) are equal. Although the scaling ansatz for B^* has been investigated[14] by comparing high-field magnetization and magnetostriction data, the pressure dependence of B^* has never been measured directly so far. The purpose of the present paper is to present the first high-field ($B \leq 28$ T) high-pressure ($P \leq 5.1$ kbar) study of the metamagneticlike transition in UPt_3. The pressure dependence of B^* has been investigated by means of magnetoresistance measurements on a single-crystalline sample at 2.0 K.

The high-pressure experiments have been performed in the High Magnetic Field Facility of the University of Amsterdam.[18] Helium is used as a pressure-transmitting medium. The pressure cell is connected to the pressure generating unit via a flexible stainless-steel capillary. As in the pressure-temperature range where we performed our experiments the helium gas solidifies, special care has been taken in order to keep the pressure as large and as hydrostatic as possible. This is realized by cooling the pressure cell while maintaining a temperature gradient, in order to start solidification of the helium at the bottom of the cell. During the solidification process blocking of the capillary is prevented by a heater. In the gaseous helium regime pressures were read within 10 bars with a standard pressure transducer (Coleraine Instruments). The pressure drop upon the solidification of the helium was monotored by strain gauges that were glued on the pressure vessel. The pressure vessel is machined of a copper-beryllium alloy. It has a cylindrical shape with an outer diameter of 14 mm and a sample space with a diameter of 5 mm over a length of 15 mm. The magnetoresistance was measured with a standard four-wire dc method. The voltage and current contacts were realized by soldering copper leads onto the sample. The four leads are led through the capillary tube towards a pressure sealing at room temperature. The feedthrough is realized by embedding the wires in epoxy and glueing them onto a conically machined plug that serves as pressure sealing.

The Czochralski-grown single-crystalline UPt_3 specimen had a cylindrical shape (length 6 mm, diameter 1.4 mm). The current and magnetic field were both directed in the hexagonal plane along the a axis. The applied current amounted to 0.5 A, yielding a current density of 32 A/cm^2. The residual resistance value[19] equals 6.2 $\mu\Omega$ cm ($I \parallel a$). The sample was previously used for resistivity measurements under pressure[19] and high-field magnetoresistance measurements at zero pressure.[7]

In the high-field installation[18] pulsed magnetic fields can be generated up to 40 T. The energy is taken directly from the mains. Different pulse shapes can be chosen as the field can be regulated by a thyristor rectifier that is coupled to the mains. A serious experimental problem that is encountered by the use of a metallic pressure vessel in pulsed magnetic fields is eddy current heating of the pressure cell. As the pressure-transmitting solid helium is a good thermal conductor the temperature of the sample also will change. In order to keep the heating of the sample during the pulse within acceptable limits a low sweep rate of 20 T/sec was used. Furthermore, the pressure vessel was immersed directly in pumped helium ($T=1.3$ K) in order to improve the heat flow to the helium bath. At the start of the pulse the sample temperature increases but reaches a constant value of 2.0 ± 0.1 K, because of the heat balance, after a typical thermal relaxation time of ~ 0.1 sec ($B=2$ T). This was checked by additional measurements with various sweep rates and field pulses with constant magnetic fields during several tenths of a second. Note, furthermore, that in the temperature range of interest B^* is almost temperature independent.[7] The maximum field that can be reached is limited by the joule heating of the magnet. It amounts to 28 T with the applied sweep rate of 20 T/sec. All the data have been taken with increasing field.

The experimental results are shown in Fig. 1 in a plot of $\Delta\rho = \rho(B) - \rho(0)$ vs B. For the sake of clarity the curves at various pressures are shifted vertically. The most important result of the present experiments is the shift of the maximum in the magnetoresistance towards higher fields

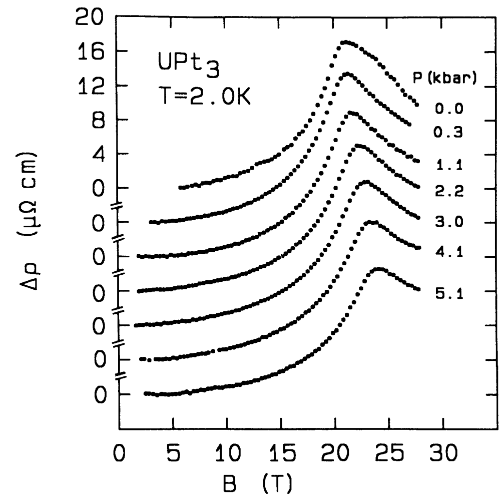

FIG. 1. High-field magnetoresistance of UPt_3 ($B \parallel I \parallel a$) at $T=2.0$ K under hydrostatic pressures up to 5.1 kbar as indicated.

with increasing pressure. The pressure variation of B^* is plotted in Fig. 2, from which it follows that dB^*/dP is constant over the pressure range 0–5 kbar and amounts to 0.60 T/kbar. The zero pressure curve ($B^*=21.0$ T, Fig. 1) is in good agreement with our previous data.[7] The somewhat different curvature of the $P=0$ curve when compared to the $P \neq 0$ curves, for fields in the vicinity of B^*, is probably due to a temperature variation caused by the magnetocaloric effect in the $P=0$ experiment, as the thermal contact was less good. An additional zero-pressure measurement without pressure vessel at a temperature of 1.3 K (not shown) gave the proper behavior and confirmed $B^*(P=0)=21.0$ T. At zero pressure an enormous magnetoresistance is observed: $\rho(0)$ equals 12 $\mu\Omega$ cm, while $\rho(B^*)$ amounts to 29.3 $\mu\Omega$ cm at $T=2.0$ K. Under pressure the absolute value of $\Delta\rho$ is slightly reduced: At 5 kbar $\rho(0)$ equals 9.4 $\mu\Omega$ cm (Ref. 19), while

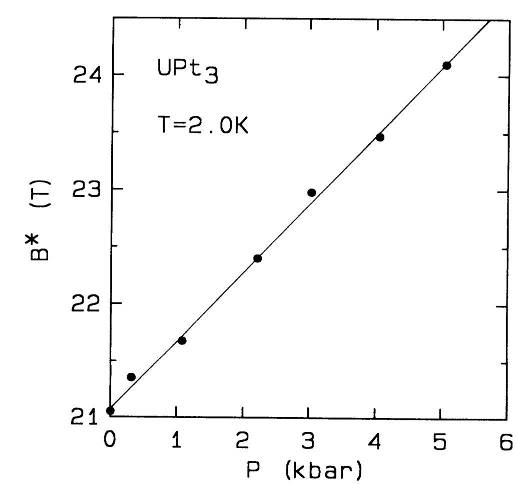

FIG. 2. Pressure variation of the metamagnetic threshold field B^* of UPt_3 at $T=2.0$ K. B^* increases at a constant rate of 0.60 T/kbar (solid line).

$\rho(B^*)$ amounts to 24.0 $\mu\Omega$ cm (at $T=2.0$ K). Apart from the increase of B^* with pressure the different curves in Fig. 1 are quite similar. After normalization with respect to B^* it appears that the width of the transition remains almost equal. Also the pressure variation of the relative magnetoresistance at B^*, $\Delta\rho(B^*)/\rho(0)$, is small. The scatter in the data at low magnetic fields does not allow for an accurate determination of the field dependence of the magnetoresistance. Plotting the data in Fig. 1 on a double logarithmic scale reveals that $\Delta\rho \propto B^n$ with $n \approx 2.0 \pm 0.3$ for $B < 10$ T for all pressures. Unfortunately, no accurate pressure variation of the power-law dependence can be deduced from the present data.

The magnetic Grüneisen parameter Γ_B deduced from the data in Figs. 1 and 2 amounts to 59, where we used a value for the isothermal compressibility[12] (κ) of 0.48 Mbar^{-1}. The thermal Grüneisen parameter of UPt_3 has been determined in several ways. From a combination of thermal-expansion and specific-heat measurements a value for Γ_T of 71 results.[14] Pressure experiments yield values of 52 [from the depression of the coefficient of the T^2 term in the resistivity,[20] where $A \propto 1/(T^*)^2$], 58 (from the pressure induced shift of $T_{\max} \propto T^*$ in the susceptibility[12]), and 55 (from specific-heat measurements under pressure,[21] where $\gamma \propto 1/T^*$). We thus conclude that $\Gamma_T \simeq \Gamma_B$.

Our high-field high-pressure experiments underline the similarity between UPt_3 and $CeRu_2Si_2$. In the latter compound a metamagneticlike transition is observed at $B^* = 8.2$ T ($T=4.2$ K) for a field direction along the tetragonal axis.[16] Magnetoresistance measurements under pressure[22] yield an increase of B^* and the anomalously large magnetic Grüneisen parameter $\Gamma_B = 179$. Interestingly, it was pointed out[22] that the metamagneticlike transition only occurs when a certain magnetic energy is attained, since $M(B^*)$ and $\Delta\rho(B^*)$ are almost pressure independent at fixed temperature. Also in the case of UPt_3 $\Delta\rho(B^*)$ is almost pressure independent, indicating a similar nature of the transition. In the past years extensive pressure[22] and magnetovolume studies[23] on $CeRu_2Si_2$ have been performed in order to demonstrate the existence of one single energy scale and in order to determine $\Gamma_B \approx \Gamma_T$.

The presence of one single energy scale that governs the low-temperature thermal, magnetic, and transport properties is now well established experimentally for both $CeRu_2Si_2$ (Ref. 22) and UPt_3. The scaling via B^* suggest that the characteristic energy scale is mainly determined by the intersite interactions. However, the notion that competing magnetic interactions, namely, the Kondo screening and the Ruderman-Kittel-Kasuya-Yosida exchange, are at the basis of the quasiparticle formation brings two energy scales into the problem. High-field specific-heat measurements[11,24] indicate that the energy scale for the on-site interactions corresponds to fields of the order of several times B^*, i.e., only for field $B \gg B^*$ the heavy-fermion state is suppressed completely. On the other hand, the complex processes of f-electron screening and f-electron exchange in heavy-fermion compounds likely imply that the on-site and intersite interactions are intimately connected. Recently, the metamagneticlike behavior of heavy-fermion compounds was investigated theoretically in a number of papers,[25–30] thereby mostly focusing on $CeRu_2Si_2$. Unfortunately, these models have not yet addressed satisfactorily the important issues of the large Grüneisen parameter and the scaling behavior as reported here.

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[1] P. H. Frings, J. J. M. Franse, F. R. de Boer, and A. Menovsky, J. Magn. Magn. Mater. **31–34**, 240 (1983).

[2] A. de Visser, J. J. M. Franse, A. Menovsky, and T. T. M. Palstra, Physica B **127**, 442 (1984).

[3] G. R. Stewart, Z. Fisk, J. O. Willis, and J. L. Smith, Phys. Rev. Lett. **52**, 679 (1984).

[4] C. M. Varma, Bull. Am. Phys. Soc. **29** (3), 404 (1984).

[5] B. S. Shivaram, Y. H. Jeong, T. F. Rosenbaum, D. G. Hinks, and S. Schmitt-Rink, Phys. Rev. B **35**, 5372 (1987).

[6] R. A. Fisher, S. Kim, B. F. Woodfield, N. E. Phillips, L. Taillefer, K. Hasselbach, J. Flouquet, A. L. Giorgi, and J. L. Smith, Phys. Rev. Lett. **62**, 1411 (1989).

[7] A. de Visser, R. Gersdorf, J. J. M. Franse, and A. Menovsky, J. Magn. Magn. Mater. **54–57**, 383 (1986).

[8] A. de Visser, L. Puech, W. Joss, A. A. Menovsky, and J. J. M. Franse, Jpn. J. Appl. Phys. **26**, Suppl. 26-3, 513 (1987).

[9] I. Kouroudis, D. Weber, M. Yoshizawa, B. Lüthi, L. Puech, P. Haen, J. Flouquet, G. Bruls, U. Welp, J. J. M. Franse, A. Menovsky, E. Bucher, and J. Hufnagl, Phys. Rev. Lett. **58**, 820 (1987).

[10] M. van Sprang, R. A. Boer, A. J. Riemersma, L. W. Roeland, A. Menovsky, J. J. M. Franse, and J. Schoenes, J. Magn. Magn. Mater. **76&77**, 229 (1988).

[11] H. P. van der Meulen, Z. Tarnawski, A. de Visser, J. J. M. Franse, J. A. A. J. Perenboom, D. Althof, and H. van Kempen, Phys. Rev. B **41**, 9352 (1990).

[12] A. de Visser, A. Menovsky, and J. J. M. Franse, Physica B **147**, 81 (1987).

[13] G. Aeppli, E. Bucher, A. I. Goldman, G. Shirane, C. Broholm, and J. K. Kjems, J. Magn. Magn. Mater. **76&77**, 385 (1988).

[14] A. de Visser, J. J. M. Franse, and J. Flouquet, Physica B **161**, 324 (1989).

[15] B. Lüthi and M. Yoshizawa, J. Magn. Magn. Mater. **63&64**, 274 (1987).

[16] L. Puech, J.-M. Mignot, J. Voiron, P. Lejay, P. Haen, and J. Flouquet, J. Low Temp. Phys. **70**, 237 (1988).

[17] P. Thalmeier and P. Fulde, Europhys. Lett. **1**, 367 (1986).

[18] R. Gersdorf, F. R. de Boer, J. C. Wolfrat, F. A. Muller, and L.

W. Roeland, in *High Field Magnetism,* edited by M. Date (North-Holland, Amsterdam, 1983), p. 277.

[19]A. de Visser, J. J. M. Franse, and A. Menovsky, J. Magn. Magn. Mater. **43**, 43 (1984).

[20]J. O. Willis, J. D. Thompson, Z. Fisk, A. de Visser, J. J. M. Franse, and A. Menovsky, Phys. Rev. B **31**, 1654 (1985).

[21]G. E. Brodale, R. A. Fisher, N. E. Phillips, G. R. Stewart, and A. L. Giorgi, Phys. Rev. Lett. **57**, 234 (1986).

[22]J.-M. Mignot, J. Flouquet, P. Haen, F. Lapierre, L. Puech, and J. Voiron, J. Magn. Magn. Mater. **76 & 77**, 97 (1988).

[23]A. Lacerda, A. de Visser, L. Puech, P. Lejay, P. Haen, J. Flouquet, J. Voiron, and F. J. Ohkawa, Phys. Rev. B **40**, 11429 (1989).

[24]H. P. van der Meulen, A. de Visser, J. J. M. Franse, T. T. J. M. Berendschot, J. A. A. J. Perenboom, H. van Kempen, A. Lacerda, P. Lejay, and J. Flouquet, Phys. Rev. B **44**, 814 (1991).

[25]W. Brenig, Solid State Commun. **64**, 203 (1987).

[26]F. J. Ohkawa, Solid State Commun. **71**, 907 (1989).

[27]R. Konno, J. Phys. Condens. Matter **3**, 9915 (1991).

[28]K. Miyake and Y. Kuramoto, Physica B **171**, 20 (1991).

[29]S. M. M. Evans, J. Magn. Magn. Mater. **108**, 135 (1992).

[30]T. M. Hong and G. A. Gehring (unpublished).