# Magnetic behavior of the heavy-fermion system UPd<sub>2</sub>Ga<sub>3</sub>

S. Süllow, B. Ludoph, B. Becker, G. J. Nieuwenhuys, A. A. Menovsky, and J. A. Mydosh Kamerlingh Onnes Laboratory, Leiden University, 2300 RA Leiden, The Netherlands

S. A. M. Mentink and T. E. Mason

Department of Physics, University of Toronto, Toronto, Canada M5S 1A7

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We have synthesized the heavy-fermion compound UPd<sub>2</sub>Ga<sub>3</sub>, crystallizing in a structure closely related to the hexagonal PrNi<sub>2</sub>Al<sub>3</sub> lattice. The physical properties show UPd<sub>2</sub>Ga<sub>3</sub> to behave very similar to the heavyfermion superconductor UPd<sub>2</sub>Al<sub>3</sub>. It orders antiferromagnetically at  $T_N = 13$  K, while a large decrease in the electrical resistivity below  $T_N$  and electronic specific-heat coefficient  $\gamma = 230$  mJ/mol K<sup>2</sup> classify the new compound as a heavy fermion. The magnetic structure is equivalent to that of UPd<sub>2</sub>Al<sub>3</sub>, although the ordered moment of  $0.50(5)\mu_B$  is smaller. No superconductivity has been observed in UPd<sub>2</sub>Ga<sub>3</sub> above 50 mK.

## I. INTRODUCTION

Heavy-fermion materials are of general interest in present day research due to their unusual and still not fully understood low-temperature properties. Many of these systems order magnetically, and some become superconducting as well. Two of the more recent examples are UPd<sub>2</sub>Al<sub>3</sub> and UNi<sub>2</sub> Al<sub>3</sub>. Both systems crystallize in the hexagonal PrNi<sub>2</sub>Al<sub>3</sub> structure, show magnetic transitions at 14.5 and 4.6 K, and become superconducting below 2 and 1 K, respectively.<sup>1,2</sup> It has been argued that in UPd<sub>2</sub>Al<sub>3</sub> superconductivity and magnetism are carried by different electronic *f* subsystems of "light" magnetic ( $\gamma$ =25 mJ/mol K<sup>2</sup>) and "heavy" superconducting character ( $\gamma$ =115 mJ/mol K<sup>2</sup>).<sup>3,4</sup>

In this contribution we present our results on the allomorphic compound UPd<sub>2</sub>Ga<sub>3</sub>. We were able to prepare this material in nearly single-phase polycrystalline form and determine its crystal structure. Our measurements of the specific heat, electrical resistivity, magnetoresistivity, magnetic susceptibility, magnetization, and neutron diffraction show UPd<sub>2</sub>Ga<sub>3</sub> to order antiferromagnetically at  $T_N \approx 13$  K. No superconductivity was observed down to 50 mK. The zero-temperature extrapolation of the specific heat  $c_p/T$  is 230 mJ/mol K<sup>2</sup>, thus making UPd<sub>2</sub>Ga<sub>3</sub> a moderately heavy-fermion material. The resistivity exhibits a large drop below  $T_N$ , which indicates that a very large scattering contribution is removed by a continuous formation of a coherent state and the magnetic ordering.

These and the other physical properties of  $UPd_2Ga_3$  will be compared to those of  $UPd_2Al_3$  in which superconductivity does coexist with magnetism. We will discuss both systems and give arguments for the similarities of the magnetic and disparity of the superconducting properties in terms of the differences of the crystal structures affecting the Fermi surface.

# **II. METALLURGY AND EXPERIMENTAL TECHNIQUES**

Polycrystalline samples of  $UPd_2Ga_3$  have been formed by arc-melting the constituents (with purity U: 3N, Pd: 5N, and

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Ga: 4N) in stoichiometric ratio under argon atmosphere in a water-cooled copper crucible. The weight loss during melting was less than 0.5%. Subsequently the samples were annealed for one week at 700 °C in high vacuum in a quartz ampule. As-cast and annealed material has been examined by electron-probe microanalysis. This proved that annealing is necessary to obtain a homogeneous sample of the proper stoichiometry. While as-cast samples consist of about 70 vol. % of a phase with the composition UPd<sub>2.4</sub>Ga<sub>3.2</sub> and 30 vol. % UPd<sub>1.2</sub>Ga<sub>2.4</sub>, the annealed samples contain 95% UPd<sub>2</sub>Ga<sub>3</sub>, 4 vol. % UPd<sub>2.6</sub>Ga<sub>3.4</sub>, and 1 vol. % UGa<sub>3</sub>.

The crystallographic structure has been determined by a full refinement of combined x-ray and neutron diffraction at room temperature, and refinement of further neutron diffractor tograms at 4.2, 20, and 150 K. Best agreement between calculated and measured diffraction patterns was achieved by assuming a hexagonal superstructure of the PrNi<sub>2</sub>Al<sub>3</sub> lattice with the unit cell doubled along the *c* axis (BaB<sub>2</sub>Pt<sub>3</sub> structure). The lattice parameters at room temperature are a=5.3015(1) Å and c=8.5112(3) Å. Details of the structure, including the free positional parameters of Pd and Ga are given in Table I. The superstructure is realized by the small displacement of the Ga and Pd atoms. In the

TABLE I. Refined structural parameters of  $UPd_2Ga_3$ , space group  $P6_3/mmc$  (BaB<sub>2</sub>Pt<sub>3</sub> type), from neutron powder diffraction at T=290, 150, and 20 K. The room-temperature refinement is the result of a refinement of combined neutron and x-ray powder diffraction patterns.

	T=290 K	<i>T</i> =150 K	T = 20  K
a (Å)	5.3015(1)	5.2904(3)	5.2840(3)
c (Å)	8.5112(3)	8.4834(8)	8.4774(9)
V (Å <sup>3</sup> )	207.17(2)	205.63(2)	204.98(2)
$x_{\rm Ga}(-)$	0.5055(4)	0.5118(7)	0.5106(7)
$y_{Ga}(-)$	0.0110(8)	0.024(1)	0.021(1)
z <sub>Pd</sub> (-)	0.0262(2)	0.0185(7)	0.0197(7)
$R_p$ (%)	16.3	17.3	18.6
Ordered mom	ent at 4.2 K: 0.50(	$5)\mu_B/U$ atom	

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FIG. 1. (a) The specific heat  $c_p/T$  vs T of UPd<sub>2</sub>Ga<sub>3</sub>. (b) The specific heat  $c_p/T$  vs  $T^2$ . The solid line indicates the influence of CEF contributions on the determination of  $\gamma$ ; the extrapolation between 15 and 20 K leads to a lower value of  $\gamma$  than the extrapolation between 1.8 and 10 K.

PrNi<sub>2</sub>Al<sub>3</sub> lattice the positions of the Ga and Pd atoms would be given by parameter values  $x_{Ga}$ ,  $y_{Ga}$ , and  $z_{Pd}$  of 0.5, 0, and 0, respectively. We have not been able to determine the crystallographic structure of the UPd<sub>2.6</sub>Ga<sub>3.4</sub> impurity phase. Since we have not excluded the reflections corresponding to this phase, the reliability factors of our refinement are rather poor.

There are obvious differences in the lattice constants between UPd<sub>2</sub>Ga<sub>3</sub> and UPd<sub>2</sub>Al<sub>3</sub>; for UPd<sub>2</sub>Al<sub>3</sub> values of 5.365 Å (*a* axis) and 4.186 Å (*c* axis) have been reported.<sup>1</sup> Thus, while half the *c* axis of UPd<sub>2</sub>Ga<sub>3</sub> is nearly 2% longer than that of UPd<sub>2</sub>Al<sub>3</sub>, the *a* axis is about 1% shorter. On the one hand, this implies a larger magnetic anisotropy between hexagonal plane and *c* axis in UPd<sub>2</sub>Ga<sub>3</sub> compared to UPd<sub>2</sub>Al<sub>3</sub>, on the other hand it gives rise to a stronger U-Pd hybridization, since the distance between U and Pd is considerably diminished here (UPd<sub>2</sub>Ga<sub>3</sub>: 3.069 Å vs UPd<sub>2</sub>Al<sub>3</sub>: 3.097 Å).

Moreover, the unit-cell volume of the Ga compound is reduced compared to the Al system. While for UPd<sub>2</sub>Ga<sub>3</sub> half the volume of the unit cell is V=103.6 Å<sup>3</sup>, it is V=104.3Å<sup>3</sup> for UPd<sub>2</sub>Al<sub>3</sub>. Generally, such change of the unit-cell volume can be treated in terms of increasing the hybridization on applying pressure. Although the volume effect is generated by anisotropic lattice parameter changes (i.e., by uniaxial pressure), we can at least estimate the order of magnitude of (hydrostatic) pressure necessary to achieve a similar compression. With the isothermal compressibility  $\kappa_T=0.85$  Mbar<sup>-1</sup> of UPd<sub>2</sub>Al<sub>3</sub> this volume difference between the two systems would be equivalent to 10 kbar pressure applied to UPd<sub>2</sub>Al<sub>3</sub>.<sup>5</sup>

The physical properties of UPd<sub>2</sub>Ga<sub>3</sub> have been examined by specific heat  $c_p$ , resistivity  $\rho$ , susceptibility  $\chi$ , magnetization M, and neutron diffraction. The specific heat has been determined by a conventional adiabatic technique, at temperatures from 1.8 to 50 K. The resistivity was measured with a four-probe ac technique in zero field from 50 mK to 300 K and in fields up to 8.5 T below 1.5 and 16 K utilizing a four-probe dc technique. For the susceptibility in 0.01 T between 2 and 300 K, a commercial SQUID was employed,



FIG. 2. (a) The overall behavior of the resistivity of  $UPd_2Ga_3$ . (b) The low-temperature resistivity  $\rho$  of  $UPd_2Ga_3$  in zero field (+) and in a field of 8 T ( $\diamond$ ) with the magnetic transition. In the zero-field measurement the resistivity drops by a factor of 40 between 14 and 0.05 K. The solid lines are fits to Eq. (1) with parameters as described in the text.

while the magnetization was obtained using a FONER magnetometer between 5 and 15 K from 0 to 12 T. Powder neutron-diffraction measurements were performed with the high-resolution 800-wire multidetector on the C2 spectrometer at the NRU reactor of Chalk River Laboratories, using a neutron wave length of 1.5405 Å. The 1-cm<sup>3</sup> sample was mounted in a cylindrical vanadium container with helium exchange gas. Full diffractograms were recorded at temperatures from 4 to 290 K.

### **III. RESULTS**

The specific heat divided by temperature in zero field vs temperature T is plotted in Fig. 1(a). An anomaly (of antiferromagnetic origin, as will be shown below) is clearly visible, the transition temperature is determined by an entropy balance to  $T_N = 13.3$  K. At higher temperatures the  $c_p/T$  vs T curve starts to bend due to contributions of crystalline electric fields (CEF). The CEF contributions prohibit an evaluation of the electronic specific heat  $\gamma$  at temperatures above  $T_N$ . Extrapolating  $c_p/T$  between 15 and 20 K to T=0, as is done by the straight line in Fig. 1(b) (where  $c_p/T$  is plotted against  $T^2$ ), leads to a  $\gamma$  that is lower than that yielded by an extrapolation from below 10 K, thus illustrating the influence of CEF contributions above  $T_N$  on the determination of  $\gamma$ . From the low-temperature extrapolation we obtain  $\gamma = 230$ mJ/mol  $K^2$  indicating that UPd<sub>2</sub>Ga<sub>3</sub> belongs to the class of heavy-fermion materials [for comparison the value of UPd<sub>2</sub>Al<sub>3</sub>:  $\gamma = 140$  mJ/mol K<sup>2</sup> (Ref. 3)]. We did not correct the specific heat of UPd<sub>2</sub>Ga<sub>3</sub> for lattice contributions, since up to now no nonmagnetic allomorph having a superstructure as described above has been found and thus we cannot obtain a reliable correction for the phonons. We remark that, although there are quantitative differences between the specific heat of UPd<sub>2</sub>Ga<sub>3</sub> and UPd<sub>2</sub>Al<sub>3</sub>, the general shape of  $c_p$  is quite similar for both systems.

The resistivity in zero field of  $UPd_2Ga_3$  closely resembles that of  $UPd_2Al_3$  [Fig. 2(a)]. The broad maximum around 150 K with the downturn at lower temperatures is



FIG. 3. The normalized magnetoresistivity of UPd<sub>2</sub>Ga<sub>3</sub> at 15 K ( $\nabla$ ), 12 K ( $\diamond$ ), 8 K ( $\triangle$ ), 5 K ( $\bigcirc$ ), and 1.6 K (+). The solid line indicates a  $B^2$  dependence of the magnetoresistivity of the measurement at 15 K.

assigned to the Kondo effect with its reduction due to the low-temperature coherent state and to the depopulation of CEF levels. However, before the coherent state can fully form, magnetic ordering appears.  $T_N$  for B=0 T is determined to be 13.1 K, defined as the minimum in  $d^2\rho/dT^2$ , which corresponds to the kink in  $\rho$  [Fig. 2(b)]. With magnetic field only a slight decrease of  $T_N$  is observed, at 8 T we find a value of 12.3 K.

As in the case of UPd<sub>2</sub>Al<sub>3</sub>, the zero field resistivity  $\rho_{ZF}$  shows a dramatic decrease from just above  $T_N$  to lowest temperatures. For UPd<sub>2</sub>Ga<sub>3</sub> the resistivity drops by a factor of 40 between 14 and 0.05 K. Dissimilar to UPd<sub>2</sub>Al<sub>3</sub>, no sign of superconductivity is observed in UPd<sub>2</sub>Ga<sub>3</sub> down to lowest attainable temperature (50 mK). Following the approach of Caspary *et al.*<sup>3</sup> we fit  $\rho_{ZF}$  below 10 K with a power law

$$\rho(T) = \rho_0 + AT^2 + bT^5 \tag{1}$$

with  $\rho_0 = 3.37(3)$   $\mu\Omega$  cm, A = 0.663(5)  $\mu\Omega$  cm K<sup>-2</sup>, and  $b = 0.12(1) \times 10^{-3}$   $\mu\Omega$  cm K<sup>-5</sup>. Caspary *et al.*<sup>3</sup> derived fit parameters of  $\rho_0 = 9.2$   $\mu\Omega$  cm, A = 0.26  $\mu\Omega$  cm K<sup>-2</sup>, and  $b = 0.48 \times 10^{-3}$   $\mu\Omega$  cm K<sup>-5</sup>, qualitatively illustrating the similarities below  $T_N$  of UPd<sub>2</sub>Ga<sub>3</sub> and UPd<sub>2</sub>Al<sub>3</sub>. With our data we can calculate the ratio  $A/\gamma^2$  yielding a value of 12.5  $\mu\Omega$  cm (mol K/J)<sup>2</sup>. This number is in good agreement with that of other heavy-fermion systems [CeAl<sub>3</sub>:  $A/\gamma^2 = 13.3$   $\mu\Omega$  cm (mol K/J) (Ref. 6)], proving that A is really reflecting the heavy mass of the electrons. We also note that the number A we derived for UPd<sub>2</sub>Ga<sub>3</sub> is 2.5 times larger than that one given for UPd<sub>2</sub>Al<sub>3</sub>, this in good agreement with the difference between the two  $\gamma$  values.

The same fit procedure can be performed for the resistivity measured in a magnetic field of 8 T. A best fit of the resistivity below 10 K was obtained using parameters of  $\rho_0=3.37(3)$   $\mu\Omega$  cm, A=0.556(6)  $\mu\Omega$  cm K<sup>-2</sup>, and  $b=0.15(1)\times10^{-3}$   $\mu\Omega$  cm K<sup>-5</sup>. From the fit parameters it is obvious that the magnetic field has mainly influenced the  $T^2$  term.



FIG. 4. The susceptibility  $\chi$  and the inverse susceptibility  $\chi^{-1}$  of UPd<sub>2</sub>Ga<sub>3</sub> plotted against temperature. The solid line indicates a Curie-Weiss fit of the data with  $\mu_{\text{eff}}=2.95\mu_B$  and  $\Theta_{\text{CW}}=-33$  K.

To examine this influence in more detail we performed measurements of the magnetoresistivity in fields up to 8.5 T. In Fig. 3, we show the normalized magnetoresistivity  $\Delta \rho / \rho_{\rm ZF} = [\rho(B) - \rho_{\rm ZF}] / \rho_{\rm ZF}$  at different temperatures. Two effects can be distinguished: (i) Above  $T_N$  the resistivity decreases with  $B^2$  (the solid line in Fig. 3 indicates a  $B^2$  dependence of the magnetoresistivity at 15 K). (ii) At 1.6 K the resistivity shows, instead of a decrease, a small increase with field. In the intermediate temperature range both effects contribute to the magnetoresistivity leading to a maximum in  $\Delta \rho / \rho_{\rm ZF}$ .

The negative  $\Delta \rho / \rho_{ZF}$  at high temperatures can have two sources, either spin disorder scattering is removed by the field or the field is affecting the coherent state. Both effects should be  $\propto B^2$ , and therefore, we cannot separate the two. On the other hand, the small magnetoresistivity at 1.6 K is entirely due to the field trying to rearrange the antiferromagnetic moments. Hence, the low-temperature magnetoresistivity reveals a complicated interplay between the influence of magnetic field on the coherent state, on the one hand, and on the magnetic moments on the other. We note that the magnetoresistivity of UPd2Ga3 resembles qualitatively that one of UPd<sub>2</sub>Al<sub>3</sub> determined by de Visser *et al.*<sup>7</sup> for a single crystal with the current and the field oriented along the a axis (i.e., the low resistive axis). The small influence of a magnetic field of 8 T on  $T_N$  of UPd<sub>2</sub>Ga<sub>3</sub> suggests a phase diagram similar to that of UPd<sub>2</sub>Al<sub>3</sub>. Taken together the experiments on resistivity and magnetoresistivity indicate that there is a continuous development of the coherent state below  $T_N$ and that the very large drop in the electrical resistivity upon antiferromagnetic order is not solely due to the decrease of spin-disorder scattering.

In order to clarify the nature of the magnetic transition we measured the susceptibility  $\chi$ . In Fig. 4, we plot our data both as  $\chi$  and  $\chi^{-1}$  vs *T*. At high temperatures the susceptibility follows a Curie-Weiss law with an effective moment  $\mu_{\text{eff}}=2.95\mu_B$  and  $\Theta_{\text{CW}}=-33$  K (by extrapolating  $\chi^{-1}$  above 100 K as straight line). This effective moment is in agreement with the expected value for either a free U<sup>3+</sup> or U<sup>4+</sup> atom. The negative Curie-Weiss temperature, indicative of antiferromagnetic coupling, is in accord with the antifer-



FIG. 5. The magnetization of UPd<sub>2</sub>Ga<sub>3</sub> in fields up to 12 T. The measurements have been taken at 15 K ( $\bigcirc$ ), 12 K ( $\diamondsuit$ ), 8 K ( $\triangle$ ), and 5 K (+).

romagnetic peak at  $T_N = 12.2$  K [determined as maximum in  $d(\chi T)/dT$ ]. The transition temperature derived from the susceptibility is somewhat lower than those extracted from specific heat or resistivity: This is probably due to the relatively broad transition [see the specific heat (Fig. 1)] blurring the exact determination of the transition temperature in the various measurements.

Unlike UPd<sub>2</sub>Al<sub>3</sub> there is no pronounced maximum in the susceptibility for UPd<sub>2</sub>Ga<sub>3</sub> above  $T_N$ , which for the Al compound was attributed to CEF contributions. In UPd<sub>2</sub>Ga<sub>3</sub> this maximum is hidden behind the antiferromagnetic transition and is visible here only as small anomaly in  $\chi^{-1}$ . Qualitatively the susceptibility of UPd<sub>2</sub>Ga<sub>3</sub> can be explained within a CEF scheme as proposed by Grauel *et al.*<sup>8</sup> for UPd<sub>2</sub>Al<sub>3</sub> but now with a singlet ground state, an excited singlet at about 15 K and a further doublet around 60 K. Especially, we note that the susceptibility for  $T \rightarrow 0$ ,  $\chi_0$ , being about two times larger for the Ga system than for the Al compound, arises from those low-lying CEF levels. However, without data on the anisotropy of the physical properties, we are unable to obtain more quantitative information about the crystalline electric fields in UPd<sub>2</sub>Ga<sub>3</sub>.

Further, we measured the magnetization of UPd<sub>2</sub>Ga<sub>3</sub> between 5 and 15 K in fields up to 12 T (Fig. 5). The magnetization at 12 T is about  $0.8-0.9\mu_B/U$  atom and shows no sign of saturation. The absolute values of M for UPd<sub>2</sub>Ga<sub>3</sub> are larger than those of UPd<sub>2</sub>Al<sub>3</sub> [~0.3 $\mu_B$  at 10 T, determined for a single crystal with a  $B \parallel a$  axis (Ref. 9)]. This is a consequence of the smaller CEF splitting in UPd<sub>2</sub>Ga<sub>3</sub> compared to UPd<sub>2</sub>Al<sub>3</sub>. M seems initially linear in B for all temperatures. However, a slight upward curvature begins at lower and lower field as the temperature is reduced. This behavior might arise from domain reorientation processes, as they have been found for UPd<sub>2</sub>Al<sub>3</sub> in the same temperature and field range.<sup>8</sup>

In order to determine the magnetic structure of UPd<sub>2</sub>Ga<sub>3</sub>, we performed a neutron-diffraction study.<sup>10</sup> Full diffractograms were recorded from  $2\theta = 5^{\circ} - 85^{\circ}$  at temperatures from 4.2–20 K. In the difference spectrum of I(4.2 K) - I(20 K) four extra reflections are observed, which are indexed as the (001), (101), (111), and (103) reflections of a



FIG. 6. Integrated intensity vs temperature of the magnetic (001) reflection of UPd<sub>2</sub>Ga<sub>3</sub>. The solid line denotes a fit to Eq. (2), yielding  $\beta = 0.28(2)$ .

magnetic unit cell equal to the nuclear unit cell,  $a_{\text{magn}} = a_{\text{nucl}} = 5.286 \text{ Å and } c_{\text{magn}} = c_{\text{nucl}} = 8.474 \text{ Å}.$  The ordering wave vector is (001), implying that ferromagnetic planes are stacked antiferromagnetically along the c axis. The integrated intensities of these magnetic reflections closely agree with those calculated for an ordered structure in which uranium moments are restricted to the basal plane. The orientation of the moment in the basal plane cannot be determined from our powder experiment. This magnetic structure is the same as that of  $UPd_2Al_3$ .<sup>11</sup> The ordered moment at 4.2 K is evaluated from a comparison of the integrated intensities of the (001) and (101) magnetic reflections with the nuclear (200) and (202) reflections, using the reported uranium form factor.<sup>12</sup> This procedure yields  $\mu_{ord}(4.2 \text{ K}) = 0.50(5)\mu_B/U$ atom, well above the detection limit of our powderdiffraction experiment, which is about  $0.15\mu_B$ . The small U moment explains the absence in the difference spectrum of the other reflections of this magnetic structure. Figure 6 displays the temperature evolution of the integrated intensity of the (001) magnetic reflection. The temperature dependence can be nicely described by the expression

$$I = A \left( \frac{T_N - T}{T_N} \right)^{2\beta}, \tag{2}$$

where  $\beta$  is the critical exponent for the ordering and A a scaling factor. We find  $T_N = 13.4(2)$  K and  $\beta = 0.28(2)$ . This value is suggestive of conventional 3D XY ordering for UPd<sub>2</sub>Ga<sub>3</sub>.

The ordered moment  $\mu_{ord}$  being considerably smaller than the moment of UPd<sub>2</sub>Al<sub>3</sub> (0.85 $\mu_B$ ) is in agreement with the observation of the higher effective masses in UPd<sub>2</sub>Ga<sub>3</sub> (higher density of states at the Fermi level), since stronger screening is to be expected. Also, a smaller ordered moment is expected from the differences of the crystal structures between the two systems, because, as remarked, in UPd<sub>2</sub>Ga<sub>3</sub> the U-Pd distance is smaller than in the Al allomorph leading to a stronger hybridization of the moments. However, one would expect that with a substantially smaller moment also the transition temperature  $T_N$  should be correspondingly reduced. This, obviously, is not the case. We remark that this rule of the dependence of  $T_N$  on the ordered magnetic moment is broken for several heavy-fermion compounds. For instance, URu<sub>2</sub>Si<sub>2</sub> has an even higher  $T_N$  than UPd<sub>2</sub>Al<sub>3</sub>, but the ordered magnetic moment is only  $0.03\mu_B$ .<sup>13</sup>

#### **IV. CONCLUSIONS**

We have presented a study of the metallurgical and physical properties of the new heavy-fermion compound  $UPd_2Ga_3$ . This compound crystallizes in the  $BaB_2Pt_3$  structure, a superstructure of the  $PrNi_2Al_3$  type. With respect to the heavy-fermion superconductor  $UPd_2Al_3$  the major differences are that (*a*) the volume of the unit cell is decreased, (*b*) the lattice constants change anisotropically, and (*c*) the superstructure causes a doubling of the unit cell along the *c* axis and slight modifications of the local site symmetry of the U atom.

The bulk properties show that UPd<sub>2</sub>Ga<sub>3</sub> is a heavyfermion system with an antiferromagnetic phase transition at  $T_N \approx 13$  K, while it is not superconducting above 50 mK. The appearance of the magnetic transition in the bulk properties of UPd<sub>2</sub>Ga<sub>3</sub> resembles qualitatively that in UPd<sub>2</sub>Al<sub>3</sub>. The quantitative differences can be accounted for by the larger effective mass of the electrons and a smaller CEF splitting in case of UPd<sub>2</sub>Ga<sub>3</sub>. Further, the magnetic structures of UPd<sub>2</sub>Ga<sub>3</sub> and UPd<sub>2</sub>Al<sub>3</sub> are identical.

(a) The decrease of the unit-cell volume of UPd<sub>2</sub>Ga<sub>3</sub>. To a first approximation this can be viewed as hydrostatic pressure applied to UPd<sub>2</sub>Al<sub>3</sub>. The decrease of the unit-cell volume of nearly 1% corresponds to a pressure of about 10 kbar. Pressure experiments on UPd<sub>2</sub>Al<sub>3</sub> revealed only negligible pressure dependences of the transition temperatures  $T_N$  and  $T_c$  in this pressure regime.<sup>3,5</sup> Therefore, something more is needed to explain the loss of superconductivity in UPd<sub>2</sub>Ga<sub>3</sub> than simple chemical pressure as, for example, Fermi-surface effects. Here, band-structure calculations of UPd<sub>2</sub>Ga<sub>3</sub> would give valuable information especially if compared to those for UPd<sub>2</sub>Al<sub>3</sub>.<sup>14</sup>

(b) The anisotropic change of lattice parameters or the increasing U-Pd hybridization. This feature can nicely account for the differences of the magnetic behavior of the two systems. A stronger U-Pd hybridization explains the larger  $\gamma$  and the smaller ordered magnetic moment  $\mu_{\mathrm{ord}}$ . The comparatively small decrease of  $T_N$  (with respect to the larger decrease of  $\mu_{ord}$ ) can qualitatively be interpreted within the model put forth by Mentink *et al.*<sup>15</sup> for CePd<sub>2</sub>Al<sub>3</sub>. These authors proposed that in the hexagonal 123-compounds magnetism is governed by an interplay of the in-plane ferromagnetic coupling and the interplane antiferromagnetic interaction. Within their model the antiferromagnetic transition temperature  $T_N$  is then determined by the interplane exchange parameter  $J_z$  and the spin value of the basal plane magnetic moment, S(S+1). Comparing now the Al system with the Ga homologue, the spin value S of  $UPd_2Ga_3$  is lowered, while  $J_{\tau}$  is increased due to the larger Ga atom creating a larger f - s, p overlap. Accordingly,  $T_N$  depends on two competing effects, and, in our case, is only slightly lowered. We remark that a similar dependence of  $J_{7}$  on the f-s,p overlap has been observed in CePd<sub>2</sub>Al<sub>3</sub> and CePd<sub>2</sub>Ga<sub>3</sub>.<sup>15,16</sup>

However, although the magnetic behavior of  $UPd_2Ga_3$ can be fairly well explained within such models, the complete absence of superconductivity is far from evident within these models. At present we can only speculate about a possible connection between the anisotropy-hybridization changes in  $UPd_2Ga_3$  and the 2-f subsystem model.<sup>3,4</sup> It is striking that in  $UPd_2Ga_3$  as in  $UPd_2$  Al<sub>3</sub> the magnetic behavior seems essentially independent of the absence or presence of superconductivity. For now, at best, we can only propose an experimental test for the role of the U-Pd hybridization. The test would be an uniaxial pressure experiment on  $UPd_2Al_3$  with the pressure applied along the *c* axis.

In our model, described above,  $T_N$  is dependent on  $J_z$  and S. Uniaxial pressure along the c axis should increase both  $J_z$  (since the f-s,p overlap gets stronger) and S (since the U-Pd hybridization becomes weaker). Hence,  $T_N$  would increase with uniaxial pressure along the c axis for UPd<sub>2</sub>Al<sub>3</sub>. The role of the hybridization change on the superconductivity should also be reflected in the uniaxial pressure dependence of  $T_c$ . If the U-Pd distance is critical for the suppression of superconductivity in UPd<sub>2</sub>Ga<sub>3</sub>, then  $T_c$  of UPd<sub>2</sub>Al<sub>3</sub> should be independent of or even increase with uniaxial pressure applied to the c axis.

(c) The crystallographic superstructure and the increase of atomic weight by the substitution of Ga for Al. The crystallographic superstructure might affect the physical properties of UPd<sub>2</sub>Ga<sub>3</sub> by two different ways. First, it will slightly change the local site symmetry of the U atoms, and accordingly modify the Fermi surface. Unfortunately, from our data we cannot assess the importance of the local site symmetry for the physical properties of UPd<sub>2</sub>Ga<sub>3</sub>. Second, the phonon spectrum might possibly be changed by the superstructure, and there will surely be an influence on the phonons by the substitution of Ga for Al, since Ga is about 2.5 times heavier than Al. This, of course, implies a simple BCS picture for superconductivity in UPd<sub>2</sub>Al<sub>3</sub> and superconductivity in UPd<sub>2</sub>Ga<sub>3</sub> would be destroyed by modifying the phonon spectrum.

In conclusion, based upon our experimental data, we believe a picture using the U-Pd hybridization and the interplane exchange mediated by the Ga or Al atoms can give a reasonable description of the magnetic behavior in these two materials. However, as far as the superconductivity is concerned we have been unable to determine the key parameter for its suppression in UPd<sub>2</sub>Ga<sub>3</sub>. It is quite possible that all effects mentioned above play a collective role. At any rate, we propose a uniaxial pressure experiment in UPd<sub>2</sub>Al<sub>3</sub> which would shed more light on the anisotropy-hybridization situation. Finally the band structure of UPd<sub>2</sub>Ga<sub>3</sub> should be calculated and compared with UPd<sub>2</sub>Al<sub>3</sub>.

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