

## Magnetic anisotropy of single-crystalline UNi<sub>2</sub>Al<sub>3</sub>

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**Abstract.** We report the magnetic field dependence of the dc susceptibility and resistivity in superconducting single crystals of the hexagonal heavy-fermion superconductor UNi<sub>2</sub>Al<sub>3</sub>. From the susceptibility we derive a crystalline-electric-field scheme, which closely resembles that of UPd<sub>2</sub>Al<sub>3</sub>. Furthermore, we examine the magnetic phase transition into the incommensurably ordered state at  $T_N = 4.1$  K and construct the magnetic phase diagram for the three crystallographic directions. While for fields  $B$  parallel to the  $a$ - or  $c$ -axis  $T_N$  is monotonically lowered, this is not the case for  $B \parallel b$ . Instead, here we find a field-induced magnetic transition into a, presumably, commensurate ordered magnetic state. We discuss our results on UNi<sub>2</sub>Al<sub>3</sub> in comparison to those for the related system UPd<sub>2</sub>Al<sub>3</sub>.

### 1. Introduction

In recent years heavy-fermion superconductors (HFS) attracted much attention due to the unusual superconducting and magnetic properties of these compounds. In particular, here the magnetic correlations could contribute to the coupling of the Cooper pairs, leading to unconventional types of superconductivity. Therefore, for HFS it is of special importance for achieving an understanding of the normal state and its magnetic properties in order to assess their influence on the superconductivity.

For one particular HFS, UPd<sub>2</sub>Al<sub>3</sub> [1], it has been argued that the long-range magnetic ordering of the U ions does not play a role in the superconductivity [2]. Instead, similarly to the case of the magnetic superconductors [3], two subsets of f-electron systems should be present, one with a small mass enhancement carrying the antiferromagnetic ordering of large U moments of  $0.85 \mu_B$  below  $T_N = 14.5$  K, and the other with heavy-fermion characteristics bearing the superconductivity below  $T_c = 2$  K [1, 2, 4].

However, with this model it is difficult to explain why the closely related system UNi<sub>2</sub>Al<sub>3</sub> exhibits a rather different behaviour with respect to its magnetic properties. This system undergoes a transition from a paramagnetic phase to an incommensurably ordered magnetic structure at  $T_N = 4$  K, with U moments of  $0.24 \mu_B$ , while heavy-fermion superconductivity is observed at  $T_c = 1$  K [5, 6]. It is not evident that the isoelectronic and isostructural systems UNi<sub>2</sub>Al<sub>3</sub> and UPd<sub>2</sub>Al<sub>3</sub> should behave so differently on a microscopic level as regards their magnetism, while the superconducting properties are qualitatively similar.

These issues are currently the focus of new experimental efforts, now that the first superconducting single crystals of UNi<sub>2</sub>Al<sub>3</sub> have become available, yielding the opportunity to study the magnetic and superconducting anisotropies. Recently Sato *et al* [7] published

the first data on the anisotropy of the superconducting properties of  $\text{UNi}_2\text{Al}_3$ . The magnetic bulk properties, however, they addressed only on a superficial level.

The magnetic anisotropies will be the subject of this report. We present susceptibility ( $\chi_{dc}$ -) and resistivity ( $\rho$ -) data on the magnetic properties of superconducting single crystals of  $\text{UNi}_2\text{Al}_3$ . The anisotropy of the magnetic susceptibility we explain as arising from single-ion crystalline-electric-field (CEF) splitting. Furthermore, from the field dependence of the dc susceptibility and the resistivity we determine the magnetic phase diagram. In particular, we find a field-induced magnetic transition for fields  $\mathbf{B}$  directed along the  $b$ -axis, and we argue that this is a transition from an incommensurably to a commensurably ordered state. We discuss our results in comparison with the magnetic properties of  $\text{UPd}_2\text{Al}_3$  and their importance regarding the appearance of heavy-fermion superconductivity in  $\text{UNi}_2\text{Al}_3$ .

## 2. Experimental techniques

The single crystals investigated in this work were grown by use of a vertical floating-zone technique using a mirror-image furnace. Details regarding the growth and the characterization, including the superconducting properties, will be published elsewhere [8]. The crystals were annealed at 1000 °C for one week in high vacuum in quartz ampoules. Each crystal was checked by Laue diffraction for single crystallinity and alignment. In the following we denote as the  $a$ - and  $c$ -axes the conventional unit-cell vectors, while the term  $b$ -axis refers to a vector with indices  $\langle 110 \rangle$ , spanning an angle of 30° with the  $a$ -axis. The homogeneity of the crystals was checked by electron probe microanalysis. They were found to be single phase, and the composition was determined to be  $\text{UNi}_{1.92 \pm 0.06}\text{Al}_{2.95 \pm 0.09}$ .

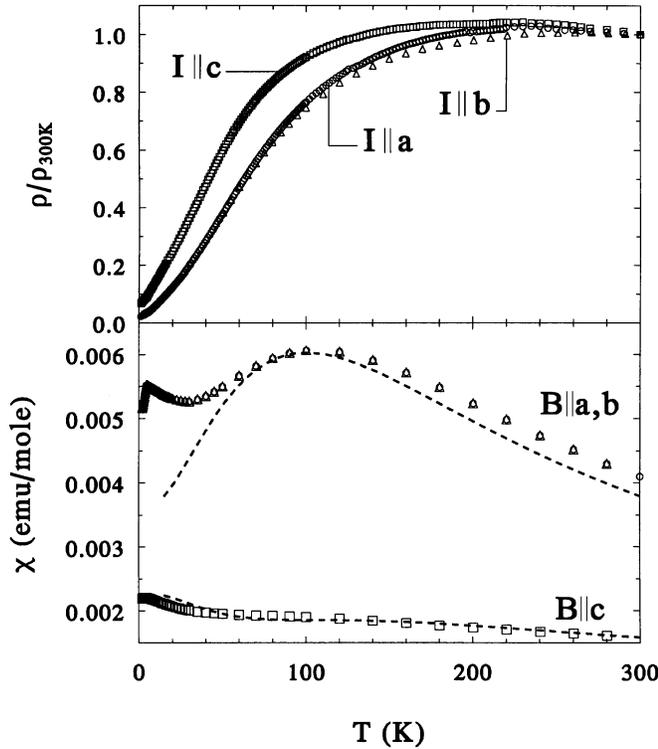
The magnetic susceptibility was measured in a commercial SQUID, between 2 and 300 K in a field of 0.1 T, and at different fields up to 5 T between 2 and 6 K. The resistivity was obtained employing a conventional ac four-point technique, in zero magnetic field between 50 mK and 300 K, and in fields up to 8 T between 1.6 and 6 K.

The superconducting properties of the crystals were checked by means of ac resistivity measurements. For the two crystals cut along the  $a$ - and  $b$ -axis, superconducting transitions were observed at  $T_c = 850$  mK with a typical transition width of 200 mK, while the transition for the crystal  $\parallel c$  was much broader with a width of about 500 mK.

## 3. Results

In figure 1 the overall magnetic susceptibility  $\chi_{dc}$  and the normalized resistivity  $\rho/\rho_{300\text{K}}$  of  $\text{UNi}_2\text{Al}_3$  are plotted for the three principal crystallographic directions. Due to unfavourable shapes of the crystals we could not determine accurately the resistivity of our crystals, though—as an estimate—the room temperature values of  $\rho$  are similar to those reported by Sato *et al* [7], i.e. about 150  $\mu\Omega$  cm.

For  $\mathbf{B}\parallel\mathbf{a}$  and  $\mathbf{B}\parallel\mathbf{b}$  the susceptibilities are nearly identical. For both directions we find a broad bump in  $\chi_{dc}$  at about 100 K, while hardly any sign of a susceptibility bump is perceivable for  $\mathbf{B}\parallel\mathbf{c}$ . The resistivity exhibits common heavy-fermion behaviour with the downturn at low temperatures due to the depopulation of CEF and coherent scattering. There is some anisotropy to be seen in the resistivity between the  $a$ - $b$  plane and the  $c$ -axis, but hardly any anisotropy can be noticed in the hexagonal plane. In the susceptibility a magnetic anomaly at  $T_N = 4.1$  K (determined as the maximum in  $d(\chi_{dc}T)/dT$  [9]) is visible for  $\mathbf{B}\parallel\mathbf{a}$  and  $\mathbf{B}\parallel\mathbf{b}$ , which cannot be seen for  $\mathbf{B}\parallel\mathbf{c}$ . In contrast, for all three directions an anomaly at  $T_N$  can be observed in the resistivity. This is illustrated in figure 2, where we plot the



**Figure 1.** The dc susceptibility and the normalized resistivity of UNi<sub>2</sub>Al<sub>3</sub> for the three principal crystallographic directions:  $B\parallel a$  ( $\circ$ ),  $B\parallel b$  ( $\Delta$ ) and  $B\parallel c$  ( $\square$ ). The dashed lines indicate the result of a calculation of the susceptibility using a crystalline-electric-field scheme as described in the text.

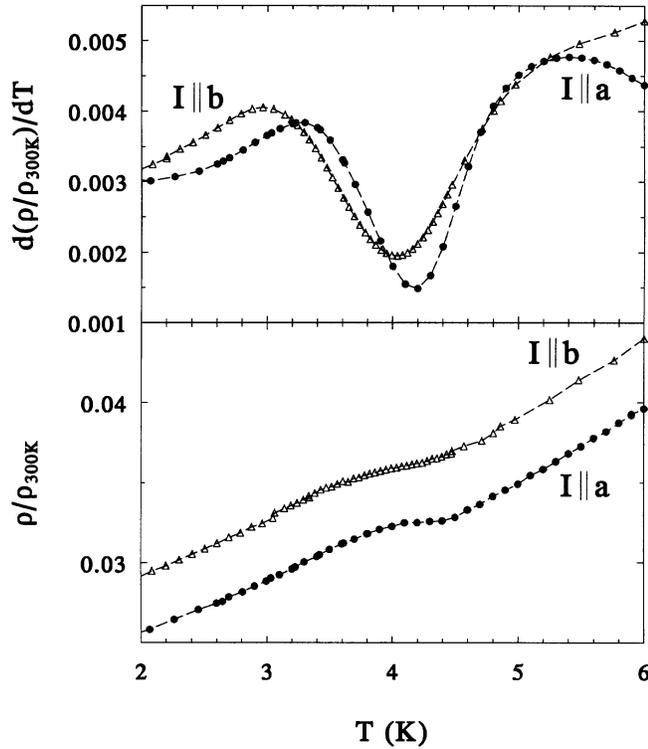
magnetic phase transition area of the resistivity for  $I\parallel a$  and  $I\parallel b$ , as examples. The magnetic transitions are reflected in the resistivity by changes of slope, and the transition temperature  $T_N$  of UNi<sub>2</sub>Al<sub>3</sub> (shown by Dalichaouch *et al* [10]) can be determined as minimum in the temperature derivative  $d\rho/dT$  of the resistivity.

These results, in so far as they overlap, are in good agreement with those of Sato *et al* [7]. The anisotropy between the  $a$ - $b$  plane and the  $c$ -axis with the maximum for  $B\parallel a, b$  qualitatively resembles the susceptibility anisotropy in UPd<sub>2</sub>Al<sub>3</sub> [11]. For the latter system the susceptibility was interpreted in terms of CEF excitations from a singlet ground state, and could quantitatively be reproduced within a mean-field approach [11].

In analogy, we describe the susceptibility of UNi<sub>2</sub>Al<sub>3</sub> in terms of CEF excitations. We follow the approach of Grauel *et al* [11], who assumed tetravalent U<sup>4+</sup> ( $5f^2$ ) as the uranium configuration in UPd<sub>2</sub>Al<sub>3</sub>. Starting from the crystalline-electric-field Hamiltonian in hexagonal symmetry

$$H_{eff} = H_{CEF} + H_{MF} = B_2^0 O_2^0 + B_4^0 O_4^0 + B_6^0 O_6^0 + B_6^6 O_6^6 + \lambda M \quad (1)$$

and including a mean-field term  $\lambda M$ , we can reproduce qualitatively and quantitatively the susceptibility of UNi<sub>2</sub>Al<sub>3</sub> with the same tetravalent U configuration and a CEF scheme similar to that for UPd<sub>2</sub>Al<sub>3</sub>. The results of our calculation are indicated in figure 1 by the dashed lines. The level scheme qualitatively resembles the one proposed for UPd<sub>2</sub>Al<sub>3</sub>; it

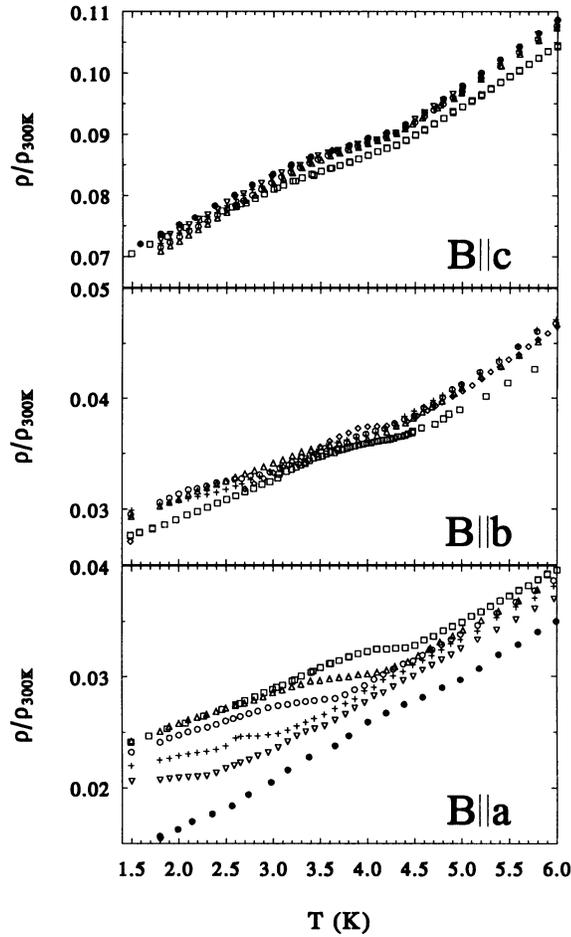


**Figure 2.** The magnetic phase transition of  $\text{UNi}_2\text{Al}_3$ , as seen in the resistivity for  $I||a$  and  $I||b$ .  $T_N$  is determined as the minimum in  $d\rho/dT$ .

is only the energy splittings of the levels which we find to be larger by a factor of about 3 for the Ni compound than for  $\text{UPd}_2\text{Al}_3$ . The CEF scheme for  $\text{UNi}_2\text{Al}_3$ , thus, consists of a singlet ground state  $|\Gamma_4\rangle$ , an excited singlet  $|\Gamma_1\rangle$  at 100 K, two excited doublets  $|\Gamma_6\rangle$  and  $|\Gamma_5\rangle$  at 340 and 450 K, another singlet  $|\Gamma_3\rangle$  at 1300 K, and a doublet  $|\Gamma_5\rangle$  at 1800 K, with a mean-field coefficient  $\lambda$  of  $-15$  K (for a detailed account of the CEF calculations see Grauel *et al* [11] and Böhm *et al* [12]).

We regard our CEF analysis as evidence for a close relationship of the magnetic states of the two systems,  $\text{UPd}_2\text{Al}_3$  and  $\text{UNi}_2\text{Al}_3$ . In the two compounds the basic magnetic anisotropies are similar, and they can be described by employing a comparable CEF picture. Only in  $\text{UNi}_2\text{Al}_3$  is the level splitting larger than in  $\text{UPd}_2\text{Al}_3$ . On the basis of the CEF scheme and taking into account that the moment in  $\text{U}(\text{Ni}, \text{Pd})_2\text{Al}_3$  is an induced moment, we expect the ordered magnetic moment  $\mu_{ord}$  and the magnetic transition temperature  $T_N$  of  $\text{UNi}_2\text{Al}_3$  to be much smaller than for  $\text{UPd}_2\text{Al}_3$ , which is in agreement with the experimental findings [4, 6]. The two parameters,  $T_N$  and  $\mu_{ord}$ , appear to be closely connected. With  $\mu_{ord} \propto T_N$  and using  $\text{UPd}_2\text{Al}_3$ ,  $T_N = 14.5$  K and  $\mu_{ord} = 0.85 \mu_B$ , we expect the ordered magnetic moment of  $\text{UNi}_2\text{Al}_3$  to be, with  $T_N = 4.1$  K,  $\mu_{ord} = 0.24 \mu_B$ , in agreement with experiment [6].

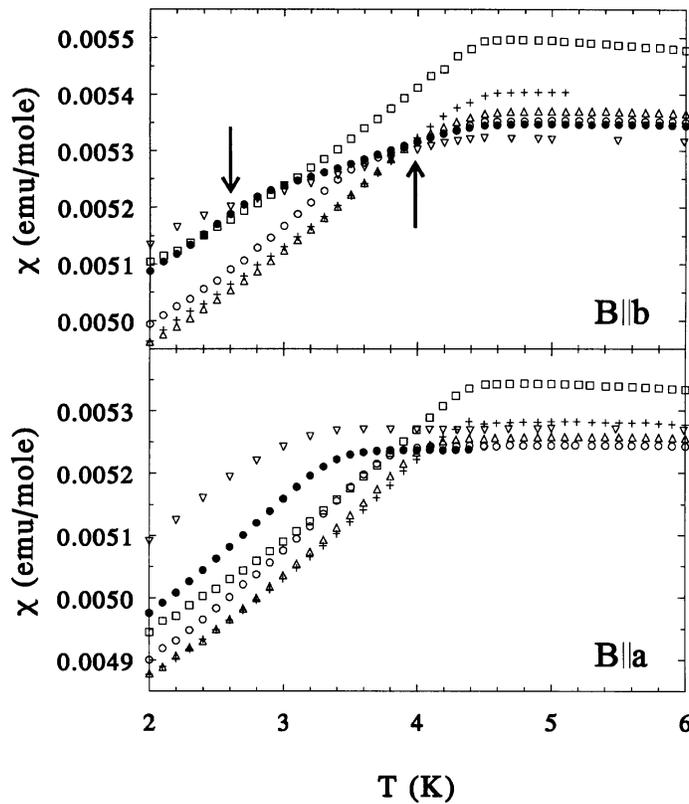
Furthermore, we studied the field dependence of the magnetic transition temperature by resistivity and dc susceptibility measurements. In figure 3 we plot the field dependence of the normalized resistivity for the three principal directions. In figure 4 the field dependence



**Figure 3.** The magnetic anomaly in the resistivity of  $\text{UNi}_2\text{Al}_3$  in different magnetic fields for the three principal crystallographic directions. The applied fields are 0 T ( $\square$ ), 1 T ( $\diamond$ ), 3 T ( $\triangle$ ), 4 T ( $\circ$ ), 5 T ( $+$ ), 6 T ( $\nabla$ ) and 8 T ( $\bullet$ ).

of the susceptibility for  $B\|a$  and  $B\|b$  is shown. From these plots it is obvious that the field dependences of the magnetic transition temperature—as of the measured properties in general—differ for the three principal directions.

The difference in behaviour for fields directed along the  $a$ - and the  $c$ -axis is similar to what is found for  $\text{UPd}_2\text{Al}_3$  [11, 13]. While for  $B\|c$  the decrease of  $T_N$  is weak, a much stronger decrease of  $T_N$  is found for  $B\|a$ . In contrast, the anomalous evolution of  $T_N$  for fields  $B$  directed along the  $b$ -axis is quite remarkable. For fields  $>3$  T two kinks appear, and the resistivity susceptibility has two changes of slope. To illustrate this the two transitions are marked by the arrows in figure 4 for  $\chi_{dc}$  at 4.5 T. And in figure 5 we plot the temperature derivatives of  $\chi_{dc}T$  and  $\rho/\rho_{300\text{K}}$ , both measured in 4.5 T for  $B\|b$ . The two anomalies observed in both derivatives suggest a field-induced magnetic transition in an intermediate-temperature regime. Since at both kinks in  $\chi_{dc}$  the susceptibility drops as the temperature is lowered, such behaviour indicates antiferromagnetic coupling in both magnetic phases. Therefore, both transition temperatures can be determined as the maximum



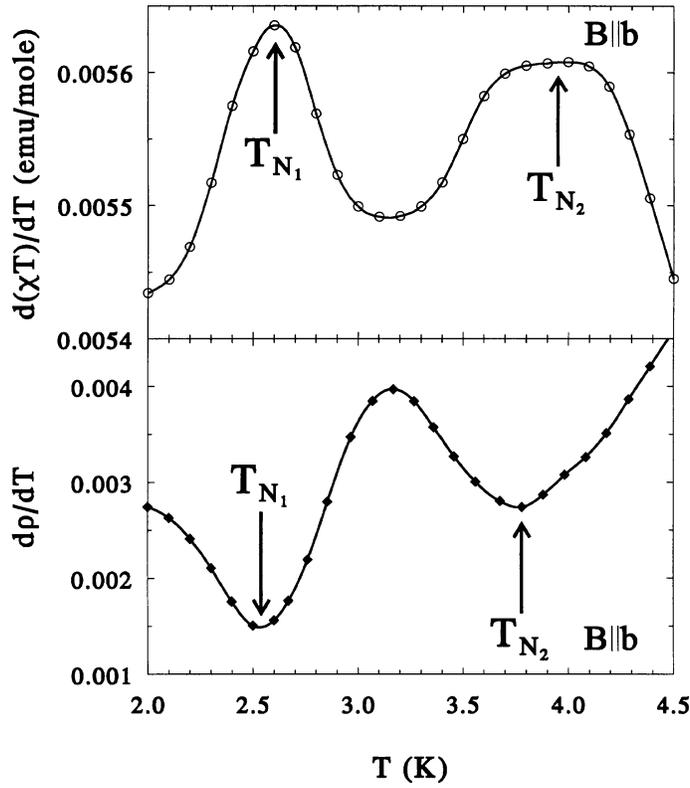
**Figure 4.** The magnetic anomaly in the susceptibility of  $\text{UNi}_2\text{Al}_3$  for different magnetic fields directed along the  $a$ - and the  $b$ -axis. Measurements are taken at 0.5 T ( $\square$ ), 1.5 T ( $+$ ), 2.5 T ( $\Delta$ ), 3.5 T ( $\circ$ ), 4.5 T ( $\bullet$ ) and 5 T ( $\nabla$ ). The arrows indicate the two transitions in the susceptibility for  $B\parallel b$  at 4.5 T.

in  $d(\chi_{dc}T)/dT$  and minimum in  $d\rho/dT$ , respectively.

From the anomalies in  $\rho/\rho_{300\text{K}}$  and  $\chi_{dc}$  we can construct the magnetic phase diagrams for the three principal crystallographic directions. The three phase diagrams are displayed in figure 6. As mentioned above, we find pronounced differences for the three crystallographic directions. Although for  $B\parallel a$  a smooth decrease of the phase borderline is seen, with  $T_N$  leaving our temperature window above 6 T, we only observe a decrease of  $T_N$  by 0.2 K for  $B\parallel c$ . On the other hand, for  $B\parallel b$  we find three magnetic phases: (i) the incommensurably ordered phase I with a boundary similar to that determined for  $B\parallel a$ , but with smaller critical fields; (ii) a magnetic phase II, which is much more stable in high magnetic fields than phase I; and (iii) at higher temperatures the paramagnetic phase III.

#### 4. Discussion

The wave vector of the incommensurate structure of  $\text{UNi}_2\text{Al}_3$  was determined by Schröder *et al* [6] to be  $q = (\frac{1}{2} \pm \delta, 0, \frac{1}{2})$ ,  $\delta = 0.110(3)$ , with ordered moments of  $\mu_{ord} = 0.24 \mu_B$ . This structure can be viewed as a longitudinal spin-density wave (LSDW) in the hexagonal plane, with the moments directed along the  $b$ -axis, and antiferromagnetic stacking of the

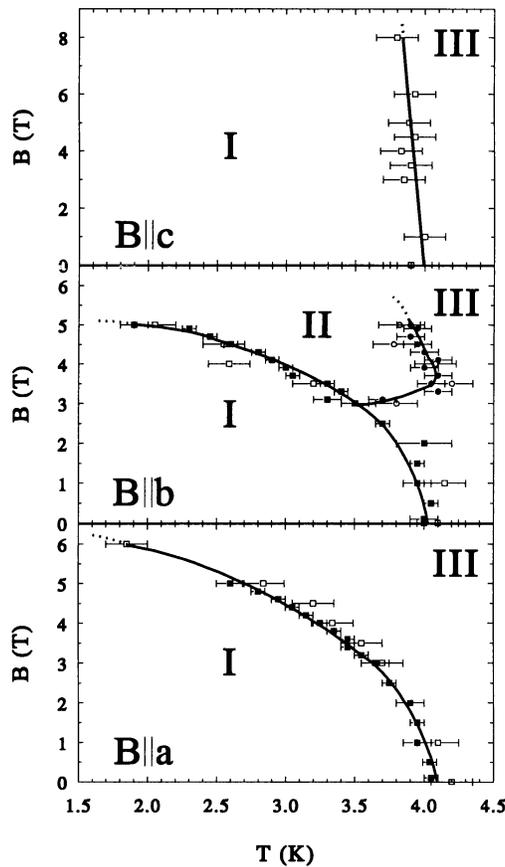


**Figure 5.** The temperature derivatives  $d(\chi_{dc}T)/dT$  and  $d(\rho/\rho_{300\text{K}})/dT$  for  $B\parallel b$  in a field of 4.5 T. The transition temperatures are determined as the maximum of  $d(\chi_{dc}T)/dT$  and the minimum of  $d(\rho/\rho_{300\text{K}})/dT$ .

moments along the  $c$ -axis. In UPd<sub>2</sub>Al<sub>3</sub> the wave vector in the magnetically ordered state is commensurate,  $\mathbf{q} = (0, 0, \frac{1}{2})$ , implying that U moments of  $\mu_{ord} = 0.85 \mu_B$  are coupled ferromagnetically in the hexagonal plane and antiferromagnetically along the  $c$ -axis. Here, the easy magnetic axis is the  $a$ -axis [4].

We argue that the magnetic transition from phase I to phase II for  $B\parallel b$  is one from the incommensurate state in UNi<sub>2</sub>Al<sub>3</sub> with wave vector  $\mathbf{q} = (\frac{1}{2} \pm \delta, 0, \frac{1}{2})$  to the commensurate one of UPd<sub>2</sub>Al<sub>3</sub> with  $\mathbf{q} = (0, 0, \frac{1}{2})$ . Then, the ferromagnetic alignment of the moments in the hexagonal plane is stabilized. With the antiferromagnetic coupling along the  $c$ -axis two antiferromagnetically coupled sheets are rotated such that the spins lie perpendicularly with respect to the magnetic field, i.e. they point along the  $a$ -axis [14]. In contrast, this would be impossible for  $B\parallel a$ , and thus the magnetic field destroys the ordered state. In addition, fields directed along the  $c$ -axis can hardly affect the magnetically ordered state, which can be understood as arising from the single-ion anisotropy.

The different behaviours of the magnetically ordered state of UNi<sub>2</sub>Al<sub>3</sub> for fields  $B$  oriented along the  $a$ - or  $b$ -axis implies that the basal-plane anisotropy energy is of a similar order of magnitude to the energy of the magnetic field at which the magnetic transition takes place. For  $B\parallel b$ , the contribution of the basal-plane anisotropy lowers the energy of the intermediate (commensurate) phase II both with respect to the paramagnetic phase III



**Figure 6.** The magnetic phase diagrams of  $\text{UNi}_2\text{Al}_3$  for the three principal crystallographic directions. Phase I is incommensurably ordered, phase II presumably commensurate and phase III paramagnetic. Filled data points have been derived from the susceptibility; open symbols indicate those determined via resistivity.

and the incommensurate phase I, while for  $B \parallel a$  it increases the energy of phase II, thereby prohibiting a field-induced magnetic transition into II below 6 T. Now, since the magnetic field energy at the field-induced magnetic transition is rather small, it indicates that the basal-plane anisotropy energy is also comparably small. This again resembles the situation for  $\text{UPd}_2\text{Al}_3$ , where in torque measurements the basal-plane anisotropy energy was found to be small [14].

According to this scenario, the commensurate and incommensurate magnetic states in  $\text{UNi}_2\text{Al}_3$  are energetically nearly degenerate. For near degeneracy there is no *a priori* reason for one of the two magnetic states to be the actual ground state. In other words, in this picture  $\text{UNi}_2\text{Al}_3$  and  $\text{UPd}_2\text{Al}_3$  appear as closely related as regards the magnetic properties. Only because of small differences in the band structures between the Pd and the Ni compounds is the magnetic ground state incommensurably ordered in  $\text{UNi}_2\text{Al}_3$ , while it is commensurate in  $\text{UPd}_2\text{Al}_3$ .

In conclusion, we have determined the anisotropy of the normal-state properties of  $\text{UNi}_2\text{Al}_3$  for all three principal directions. We found that the bulk properties of  $\text{UNi}_2\text{Al}_3$  can

consistently be described within a CEF picture, similarly to the case for previously studied  $\text{UPd}_2\text{Al}_3$ . From this scenario the quantitative differences of magnetic ordering temperatures and moments between  $\text{UNi}_2\text{Al}_3$  and  $\text{UPd}_2\text{Al}_3$  can be attributed to the larger CEF splitting in the Ni system, compared to the Pd compound. In particular,  $T_N$  and  $\mu_{ord}$  seem to scale with each other. Furthermore, we have constructed the magnetic phase diagrams for the three crystallographic directions. A magnetic phase transition for  $\mathbf{B}\parallel\mathbf{b}$  is found, and we argue that it indicates a transition from an incommensurable to a commensurable structure. Our results suggest a close relationship between the magnetic states of  $\text{UNi}_2\text{Al}_3$  and  $\text{UPd}_2\text{Al}_3$ , removing the inconsistency regarding the magnetic properties of these chemically similar compounds. However, we are aware that, in order to definitely prove our interpretation, a neutron scattering experiment on single-crystalline  $\text{UNi}_2\text{Al}_3$  in high magnetic fields should be performed.

In view of our results it is most interesting to briefly address the question of superconductivity in  $\text{U}(\text{Ni}, \text{Pd})_2\text{Al}_3$ . Since in our model the magnetic and superconducting properties of  $\text{U}(\text{Ni}, \text{Pd})_2\text{Al}_3$  are basically similar, it implies that if the 2f-subsystem picture is valid for  $\text{UPd}_2\text{Al}_3$ , it should also apply to  $\text{UNi}_2\text{Al}_3$ . Here, it would be important to perform the investigations that have been interpreted as evidence for the 2f-subsystem picture in  $\text{UPd}_2\text{Al}_3$  also on  $\text{UNi}_2\text{Al}_3$ . For instance, band-structure calculations for  $\text{UNi}_2\text{Al}_3$  should lead to Fermi surfaces similar to those for  $\text{UPd}_2\text{Al}_3$  [15–17]. Also, the experiments which have been explained within the 2f-subsystem model, like the pressure dependence of the specific heat [2] or spectroscopic experiments [18], should give similar results for  $\text{UNi}_2\text{Al}_3$ .

Another interesting possible study is that of the basal-plane anisotropy of the superconducting phase diagram. Our measurements prove the presence of basal-plane anisotropy of the magnetically ordered phase. If there is a coupling between superconductivity and the long-range magnetically ordered state, then this should be reflected in the anisotropic properties of the superconducting phase. Unfortunately, for the crystals used in this work, the superconducting transitions were too broad for us to obtain a reliable phase diagram of  $B_{c2}$  in the hexagonal plane.

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