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DOI

[10.1016/0921-4526\(94\)90467-7](https://doi.org/10.1016/0921-4526(94)90467-7)

Publication date

1994

Published in

Physica B-Condensed Matter

[Link to publication](#)

Citation for published version (APA):

Mentink, S. A. M., Nieuwenhuys, G. J., Menovsky, A. A., & Mydosh, J. A. (1994). Anisotropic f-electron magnetism in Uni4B. *Physica B-Condensed Matter*, 194-196, 275-276.
[https://doi.org/10.1016/0921-4526\(94\)90467-7](https://doi.org/10.1016/0921-4526(94)90467-7)

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Anisotropic f-electron magnetism in UNi₄B

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The hexagonal uranium compound UNi₄B has been shown to exhibit strongly anisotropic hybridization of f- and d-electron states. We performed low-field (3 mT) and high-field (40 T) magnetization measurements from 1.4 K to 30 K on a single crystal of this antiferromagnetically ordering compound, $T_N=20$ K, to further investigate the magnetic anisotropy. A phase diagram, including a spin-flop transition around 9 T, is presented. The observed large values of the electronic specific heat at low temperatures are attributed to in-plane magnetic fluctuations, which persist far below T_N . No superconductivity was found down to 40 mK. The relevance of strong 5f–3d hybridization is confirmed by extensive experiments on a series of diluted compounds UCo_xNi_{4-x}B ($0 \leq x \leq 4$).

In previous papers [1,2] we demonstrated that the hexagonal intermetallic uranium compound UNi₄B, a member of the growing class of “1-5”-intermetallic compounds, exhibits highly anisotropic magnetic properties. AF ordering of U-spins lying in the basal plane occurs below $T_N=20$ K. However, both susceptibility and specific heat increase below T_N , which can be explained by the presence of strong in-plane magnetic fluctuations, persisting down to below 1 K [2]. In this contribution we show the importance of hybridization of the f and d-electron bands by alloying UNi₄B with Co on the Ni-sites. As both UCo₄B and UNi₄B crystallize in the same CeCo₄B-type structure, alloying is possible over the entire concentration range ($0 \leq x \leq 4$) in UCo_xNi_{4-x}B. In Table 1 we list the lattice parameters of this pseudo-ternary system, together with the observed AF ordering temperature. From these data, the different interatomic uranium distances can be derived: In the basal plane, $d_{U-U}=a$, while along the c-axis $d_{U-U} = \frac{1}{2}c$, yielding 4.952 Å and 3.477 Å for UNi₄B. In general, the magnetic moment will orient perpendicular to the direction of strongest f–f hybridization, i.e. in the basal plane.

In Fig.1 we present the magnetic susceptibility ($\chi \equiv M/H$) for polycrystalline material with $x=1$ and 2, together with low-field (3 mT) and high-field (0.5 T) data for single-crystal UNi₄B. While UNi₄B exhibits clear Curie-Weiss local-

Table 1: Lattice parameters and ordering temperatures of UCo_xNi_{4-x}B-compounds.

	a (Å)	c (Å)	V(Å ³)	T _N (K)
UCo ₄ B	4.895	6.933	143.88	–
UCo ₃ NiB	4.910	6.928	144.62	–
UCo ₂ Ni ₂ B	4.924	6.954	146.01	–
UCoNi ₃ B	4.931	6.964	146.64	5.0
UNi ₄ B	4.952	6.954	147.68	20.0

Figure 1: Susceptibility of UCoNi₃B (+) and UCo₂Ni₂B (●) and single-crystal UNi₄B (△) with field parallel to the *ab*-plane and along the *c*-axis (□), all measured in $\mu_0 H=0.5$ T. Note the large anisotropy for UNi₄B and the saturation of $\chi \parallel ab$ in 3 mT (○) below 10 K.

* This work is partially supported by the Dutch Foundation FOM.

moment type behavior with $p_{eff}=2.81 \mu_B$ above 100 K (not shown), its uranium $5f$ -derived moment is progressively lost upon increasing Co-concentration. This strongly suggests that the $5f$ -electrons are filling the $3d$ -band when Co is introduced in the system. As a result, the AF ordering temperature is strongly reduced, from $T_N=20.0$ K for UNi_4B to $T_N=5.0$ K for $UCoNi_3B$. The low-field magnetization of single-crystal UNi_4B , was measured with a SQUID magnetometer in fields of 3 mT and 0.5 T. See Fig.1. In the lowest fields, χ first increases below T_N , before it saturates below 10 K. A larger field suppresses these apparent basal-plane fluctuations, yielding a maximum around 7 K.

Figure 2: Specific heat of UNi_4B (o), $UCoNi_3B$ (+) and UCo_2Ni_2B (•) plotted as c/T on a logarithmic temperature scale.

The specific heat of these samples, plotted in Fig.2 on a logarithmic temperature scale, clearly shows the reduction of T_N with increasing Co-concentration. For pure UNi_4B , an increase of c/T is observed below 7 K, in accord with the susceptibility maximum. The increase in c/T , which is almost field-independent [2], follows a weak $\ln T$ -dependence, reminiscent of the formation of an unusual Fermi-liquid state [3,4]. An extension towards lower T is necessary to confirm this $\ln T$ -dependence. The extrapolated γ -values are 269 and 294 mJ/mol K² for UNi_4B and $UCoNi_3B$, respectively.

If we combine these new results with those obtained earlier [1,2], we can establish the magnetic

Figure 3: Magnetic phase diagram for UNi_4B with the magnetic field parallel to the basal plane. Lines are guides to the eye.

phase diagram for UNi_4B . This diagram, shown in Fig.3, is a combination of high-field magnetization, specific heat and resistivity, both in magnetic fields, for two field directions in the basal plane. The AF phase boundary lies at 20 T. A spin-flop transitions is found for $\mu_0 H=8$ and 11 T for the two directions, respectively. The low- T regime for in-plane fluctuations is also indicated (shaded area in Fig.3).

In conclusion, we have shown that the ordering temperature of $UCo_xNi_{4-x}B$ strongly depends on the $5f$ - $3d$ hybridization strength. We have presented a detailed magnetic phase diagram for UNi_4B , which incorporates a highly unusual low temperature spin state, thought to arise from large in-plane fluctuations in the antiferromagnetically ordered state.

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