Magnetic phase diagram and low-dimensional excitations of hexagonal UNiB4

Mentink, S.A.M.; Nieuwenhuys, G.J.; Menovsky, A.A.; Mydosh, J.A.; Sugiyama, K.; Bando, Y.; Takabatake, T.

Published in:
Journal of Magnetism and Magnetic Materials

DOI:
10.1016/0304-8853(94)01175-3

Citation for published version (APA):

General rights
It is not permitted to download or to forward/distribute the text or part of it without the consent of the author(s) and/or copyright holder(s), other than for strictly personal, individual use, unless the work is under an open content license (like Creative Commons).

Disclaimer/Complaints regulations
If you believe that digital publication of certain material infringes any of your rights or (privacy) interests, please let the Library know, stating your reasons. In case of a legitimate complaint, the Library will make the material inaccessible and/or remove it from the website. Please Ask the Library: http://uba.uva.nl/en/contact, or a letter to: Library of the University of Amsterdam, Secretariat, Singel 425, 1012 WP Amsterdam, The Netherlands. You will be contacted as soon as possible.
Magnetic phase diagram and low-dimensional excitations of hexagonal UNi$_4$B

S.A.M. Mentink $^a$, G.J. Nieuwenhuyss $^a$, A.A. Menovsky $^a$, J.A. Mydosh $^a$, K. Sugiyama $^b$, Y. Bando $^c$, T. Takabatake $^c$

$^a$ Leiden University, P.O. Box 9506, 2300 RA Leiden The Netherlands
$^b$ Department of Physics, Osaka University, Toyonaka, Osaka 560, Japan
$^c$ FIAS, Hiroshima University, Higashi-Hiroshima 724 Japan

Abstract

The hexagonal antiferromagnet UNi$_4$B ($T_N = 20$ K) exhibits highly unusual magnetic properties, such as huge anisotropy and increasing electronic specific heat and susceptibility below $T_N$. New high-field magnetization, magnetoresistance and thermoelectric power measurements are analyzed using the recently resolved zero-field magnetic structure [1], which consists of six ordered and three paramagnetic spins.

In previous papers [1–3] we have presented the anomalous low-temperature behavior of the hexagonal intermetallic compound UNi$_4$B, crystallizing in a superstructure of the CeCo$_4$B-type structure, which orders in a unique antiferromagnetic spin arrangement below $T_N = 20$ K. This material exhibits the effects of strongly anisotropic hybridization combined with those of antiferromagnetic interactions on a triangular lattice [4,5], introducing spin frustration of magnetic uranium moments. The magnetic structure is described by a magnetic unit-cell with lattice parameters $a_{mag} = 3a$ and $c_{mag} = 1/2c$. Only six out of nine U-moments, oriented in the basal plane, are ordered in a vortex-like arrangement around the three remaining, paramagnetic moments [1]. Here we report on new high-field magnetization experiments up to 50 T, magnetoresistance and thermoelectric power measurements, on well-characterized single crystals. The magnetic phase diagram is presented, and the anomalous low-temperature magnetic and transport properties are described using the predictions for low-dimensional magnetic excitations [6].

Fig. 1 displays high-field magnetization data taken at 4.2 K on a single crystal of UNi$_4$B with fields along the two basal plane axes $a$ (left panel) and $b$ (perpendicular to $a$, right panel). Depending on this field orientation, three or two hysteretic spin-flop phases are observed, respectively. Along $a$, these fields are $\mu_0 H_2 = 8.2$ T, $\mu_0 H_3 = 11.7$ T and $\mu_0 H_0 = 19.8$ T. Along $b$, the first spin-flop field is $\mu_0 H_1 = 9.3$ T at 4.2 K (the two spin-flop fields for $H // b$ at 1.4 K are 8.1 and 18.6 T). Note that the magnetization jump is only 0.1–0.2 $\mu_B$, in accord with the hexagonal magnetic structure determined in Refs. [1,7]. Interestingly, the magnetization still does not saturate in fields as large as 52 T. This implies antiferromagnetic nearest and next-nearest neighbor coupling between the U-moments, which in zero field are equal to 1.2(2)$\mu_B$ per ordered spin. This value is exactly recovered if the magnetization per formula unit is extrapolated in a plot of $M$ vs. $1/H$ towards $1/H = 0$, indicating that the paramagnetic spin also reaches 1.2$\mu_B$ in high fields. The value of the paramagnetic spin in zero field is not known. Along the $c$ axis, the magnetization is linear with field and reaches 0.12$\mu_B$/f.u. in 35 T [2].

The anisotropy of the first spin-flop field is demonstrated in a transversal magnetoresistance experiment, at

![Fig. 1. High-field magnetization of UNi$_4$B. (a) Field parallel to the $a$-axis at $T = 4.2$ K. (b) Field parallel to the $b$-axis (perpendicular to $a$ in the basal plane) at 1.4 K (—) and 4.2 K (——).](image-url)
Fig. 2. Magnetoresistance of UNi$_4$B at $T = 1.4$ K with the current along the $c$-axis and the magnetic field parallel to $a$ (O) and $b$ (O). The solid line describes the $(1/\cos \theta)$-behavior of $H_2$. The inset shows the angular dependence of the first spin-flop field, $H_2$. The angle between the $a$-axis and the magnetic field.

The huge anisotropy of UNi$_4$B in transport properties was further studied by thermoelectric power ($S$) measurements, shown in Fig. 3. Along the basal-plane direction, $S$ is small and a pronounced, reproducible minimum is found around 215 K. No anomaly is found at $T_N$, reflecting the sustained very effective scattering mechanism in the ordered state. In contrast $S$ parallel to $c$ is rather large, and displays an upward kink at $T_N$, reminiscent of the formation of new magnetic superzone boundaries due to the AF ordering.

Combination of all magnetization, resistivity and specific-heat data yields the magnetic phase diagram of UNi$_4$B, with fields parallel to the basal plane, displayed in Fig. 4. The AF phase boundary lies at 19.8 T ($\parallel a$) and 18.6 T ($\parallel b$). The low-temperature, low-field region is indicated by the dashed line. This crossover line marks the maxima as observed in the magnetoresistance measurements. Although the zero-field magnetic structure is known, the nature of the field-induced phases remains to be established. Of special interest again is the role the paramagnetic spins play in the high-field magnetization process.

The experiments on UNi$_4$B in zero and low magnetic fields were analyzed in terms of unique magnetic ordering and one-dimensional ferromagnetic (1D FM) excitations of the chains of paramagnetic U-spins [1,7]. These 1D FM excitations give rise to increasing susceptibility and electronic specific heat below $T_N$, and weak power-law behavior of specific heat and resistivity. Here we have demonstrated that the application of large magnetic fields in the basal-plane direction quenches the influence of these excitations, giving way to the regular behavior of an antiferromagnet. We propose that the crossover line in the phase diagram is related to this quenching. At the highest fields, the paramagnetic spins approach the same moment value as the ordered spins. Further neutron-diffraction studies on single crystals in magnetic fields are necessary to establish the nature of the spin-flop phases and possible unusual critical behavior, as expected for ordering on triangular lattices with antiferromagnetic interactions.

Acknowledgements: This work was partially supported by the Dutch Foundation FOM. We thank A. Drost, E. Frikkee and K.S. Bedell for fruitful discussions.

References