UvA-DARE (Digital Academic Repository)

Antiferromagnetic order and spin frustration in UNiB4

Mentink, S.A.M.; Nieuwenhuys, G.J.; Menovsky, A.A.; Mydosh, J.A.; Drost, A.; Frikkee, E.

DOI

[10.1016/0921-4526\(94\)00350-5](https://doi.org/10.1016/0921-4526(94)00350-5)

Publication date

1995

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., Drost, A., & Frikkee, E. (1995). Antiferromagnetic order and spin frustration in UNiB4. *Physica B-Condensed Matter*, 206-207, 1-4. [https://doi.org/10.1016/0921-4526\(94\)00350-5](https://doi.org/10.1016/0921-4526(94)00350-5)

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: <https://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.

ELSEVIER

Physica B 206 & 207 (1995) 473–475

PHYSICA B

Antiferromagnetic order and spin frustration in UNi_4B

S.A.M. Mentink^{a,*}, G.J. Nieuwenhuys^a, A.A. Menovsky^a, J.A. Mydosh^a,
A. Drost^b, E. Frikkee^b

^aKamerlingh Onnes Laboratory, Leiden University, P.O. Box 9506, 2300 RA Leiden, The Netherlands

^bNetherlands Energy Research Foundation ECN, P.O. Box 1, 1755 ZG Petten, The Netherlands

Abstract

We present the novel magnetic ordering structure of the hexagonal antiferromagnet UNi_4B ($T_N = 20$ K) as determined by single-crystal neutron diffraction. Uranium moments of $1.2(2)\mu_B$ are oriented perpendicular to c , forming ferromagnetic chains with antiferromagnetic in-plane coupling. Surprisingly, $1/3$ of the U-spins remain paramagnetic. Large amplitude, one-dimensional ferromagnetic spin-wave excitations in these free chains are proposed to cause the highly anomalous low-temperature behavior of the ordered state.

The hexagonal, antiferromagnetic (AF), heavy-fermion superconductors UNi_2Al_3 [1] and UPd_2Al_3 [2,3] exhibit rather complicated magnetic structures and rich magnetic phase diagrams. Here we study the anomalous AF ($T_N = 20$ K) ordering and excitations in UNi_4B [4], which is another compound with CaCu_5 -type crystal structure. We determined the magnetic structure by single-crystal neutron diffraction [5], and the magnetic phase diagram is presented elsewhere [6]. Now we will show that this magnetic structure gives rise to anomalous low-temperature behavior, which we argue is caused by low-dimensional excitations of possibly non-Fermi-liquid character in the c -axis chains of free moments.

Our measurements were performed on Czochralski-grown single crystals of UNi_4B . The crystal structure of UNi_4B is related to the hexagonal CeCo_4B -type structure with lattice parameters $a = 4.953$ Å and $c = 6.964$ Å [4]. We performed neutron-diffraction measurements between 2.4 K and 365 K at the triple-axis

spectrometer HB3 in Petten, using a wavelength $\lambda = 1.4269$ Å, within the $(a^* b^* 0)$ and $(a^* 0 c^*)$ scattering planes. Although all strong nuclear reflections can be indexed on the basis of the CeCo_4B -type subcell, weak additional reflections with Miller indices $(h - k/3, h + 2k/3, 0)$ imply the existence of a superstructure of this subcell [5]. The real hexagonal unit-cell is related to the subcell by a rotation over 30° in the basal plane, with a $\sqrt{3}$ enlargement of the a -axis [5,7].

Below 20 K, we observed magnetic intensity at distances $\pm Q = (1/3 \ 1/3 \ 0)_{\text{super}}$ from the reciprocal lattice points of the superstructure, leading to a hexagonal magnetic unit-cell with lattice parameter $a_{\text{magn}} = 3a_{\text{sub}}$. The absence of magnetic $(h \ k \ 1)$ and the presence of magnetic $(h \ k \ 2)$ reflections give $c_{\text{magn}} = 1/2c_{\text{sub}}$. The magnetic structure is shown in Fig. 1. Six U-moments are ordered in two triangular sublattices of next-nearest neighbors, with moments perpendicular to the c -axis and 120° apart, while the directions for the remaining three moments are not defined. The U-layers are ferromagnetically (FM) coupled along the c -axis. We can distinguish two paramagnetic sites: the $(0 \ 0 \ 0)$ site, (1) in Fig. 1 and the $(1/3 \ 2/3 \ 0)$ and $(2/3 \ 1/3 \ 0)$ sites, (2) in Fig. 1. From the saturation value at 4 K of various integrated

* Corresponding author. Present address: Department of Physics, University of Toronto, 60 St. George Street, Toronto, Ontario, Canada M5S 1A7.

Fig. 1. (a) Crystallographic CeCo_4B -type subcell of UNi_4B . (b) Magnetic structure of hexagonal UNi_4B , projected on the hexagonal basal plane. The magnetic layers are ferromagnetically stacked along the c -axis. The thin solid lines represent the magnetic unit-cell. The free U-moments, indicated by (1) and (2), are located in the center of the magnetic vortices (thick lines). The nearest and next-nearest-neighbor magnetic coupling constants are indicated by J_1 and J_2 .

magnetic intensities we estimate the ordered moment $\mu_{\text{ord}} = 1.2(2)\mu_{\text{B}}/\text{U-atom}$ [5].

The occurrence of paramagnetic moments in an ordered matrix is very rare. No additional ordering phenomena occur down to 40 mK [4]. The consequences of this type of ordering for the low- T excitations can be analyzed by the resistivity, $\rho(T)$, and thermoelectric power. Fig. 2 displays $\rho(T)$ along the c -axis. Above T_{N} $\rho_{\parallel c}$ is nearly T -independent up to 300 K. Below the kink at T_{N} , $\rho_{\parallel c}$ decreases slowly, before falling more rapidly below 5 K. This unusual T -dependence is accurately described by a combination of two power laws:

Fig. 2. Temperature dependence of the resistivity of UNi_4B parallel to the c -axis. The solid line is a fit to Eq. (1).

$$\rho_{\parallel c} = AT^{0.11} + BT^2, \quad (1)$$

with $A = 57.995(1) \mu\Omega \text{ cm/K}^{0.11}$ and $B = 3.79977(6) \times 10^{-2} \mu\Omega \text{ cm/K}^2$. The latter term is ascribed to spin-wave excitations from the AF ordered spins, while the former arises from excitations in the chains comprising FM correlations of the paramagnetic U-moments along c . These chain correlations increase as T decreases, thereby decreasing $\rho(T)$.

In the basal plane $\rho(T)$ shows more complicated behavior (see Fig. 3). Below T_{N} , $\rho_{\perp c}$ first increases with decreasing T , caused by the formation of new magnetic superzone boundaries (see inset of Fig. 3). This increase in the critical region can be described by

$$\rho_{\perp c} = \tau_{\perp c, T=T_{\text{N}}} + A(T_{\text{N}} - T)^{1/3}, \quad (2)$$

where $\rho_{\perp c, T=T_{\text{N}}} = 256(1) \mu\Omega \text{ cm}$ and $A = 4.0(2) \mu\Omega \text{ cm K}^{-1/3}$. The decrease below 5 K is again attributed to the increasing chain correlations. Above T_{N} , $\rho_{\perp c}$ follows a logarithmic behavior, $\rho_{\perp c} = 287.5 \mu\Omega \text{ cm} - 10.0(1) \times \ln T$, suggestive of a Kondo effect.

Fig. 4 shows the low- T thermoelectric power, $S(T)$, measured at Hiroshima University [6]. In the basal plane $S(T)$ is rather small and no signature of the AF ordering is found. Below 10 K $S_{\perp c}$ is nearly linear with T . Along c , $S(T)$ is larger and exhibits a kink at T_{N} . Below 15 K, $S_{\parallel c}$ nearly follows a square-root T -dependence:

$$S_{\parallel c} = A + BT^{0.48}, \quad (3)$$

with a negligible small constant $A = 4.3(5) \times 10^{-8} \mu\text{V/K}$ and coefficient $B = 1.263(6) \mu\text{V/K}^{1.48}$. The changes of thermopower and resistivity along both directions below T_{N} are consistently related by the usual formula

Fig. 3. Resistivity of UNi_4B perpendicular to the c -axis. The solid line in the inset is given by Eq. (2). The high- T resistivity follows a logarithmic temperature dependence (see text).

Fig. 4. Thermoelectric power of UNi₄B parallel (○) and perpendicular to (●) the *c*-axis. The solid line is given by Eq. (3).

$$\Delta S = \frac{\pi^2 k_B^2 T}{3|e|} \frac{1}{\rho} \frac{\Delta \rho}{\Delta E}, \quad (4)$$

with ΔE the energy change through the transition.

Both the zero-field experiments described here and earlier magnetic and specific-heat measurements [4] indicate that the non-ordering U-atoms are also magnetic, with similar planar anisotropy as the ordered moments. The FM coupling along *c* of the free moments yields short-range correlated paramagnetic chains. Since the molecular fields arising from the ordered spins cancel at every site of a free spin, these spins do not order, and give rise to extra magnetic scattering at low *T*. From our data the excitations in these chains are effective scatterers. Only when these excitations freeze out, $\rho(T)$ drops rapidly. Note also that the specific heat exhibits an extra contribution below T_N [5], which follows the calculations for a 1D-FM chain [8]. The excitations are best described by weak power laws, which we propose relate to their 1D nature, and suggest a non-Fermi-liquid character. An appropriate theoretical model should consistently

describe and relate the various power laws observed. The multi-channel Kondo effect [9] provides small power laws of the correct size for large channel numbers, but the predicted increase of the low-*T* susceptibility is not observed [4].

We have presented the magnetic structure of hexagonal UNi₄B, which consists of U-moments of $1.2\mu_B$ which form vortices around paramagnetic moments. These free moments form isolated chains along the *c*-axis. The resulting low-*T* excitations in the ordered state are of 1D character and dominate the low-temperature properties.

Acknowledgement

This work was partially supported by the Dutch Foundation FOM. We acknowledge Y. Bando for performing the thermoelectric power measurements and M.W. Meisel and K.S. Bedell for discussions.

References

- [1] A. Schröder et al., Phys. Rev. Lett. 72 (1994) 136.
- [2] A. Krimmel et al., Z. Phys. B 86 (1992) 161; A. Krimmel et al., Solid State Commun. 87 (1993) 829.
- [3] H. Kita et al., J. Phys. Soc. Japan 63 (1994) 726.
- [4] S.A.M. Mentink et al., Physica B 186–188 (1993) 270; S.A.M. Mentink et al., Physica B 194–196 (1994) 275.
- [5] S.A.M. Mentink et al., Phys. Rev. Lett. 73 (1994) 1031.
- [6] S.A.M. Mentink et al., in: The International Conference on Magnetism, Warsaw, Poland, 1994.
- [7] A. Drost et al., unpublished.
- [8] J.C. Bonner and M.E. Fisher, Phys. Rev. A 135 (1964) 640.
- [9] A.W.W. Ludwig and I. Affleck, Phys. Rev. Lett. 67 (1991) 3160.