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DOI

[10.1016/0921-4526\(94\)00493-F](https://doi.org/10.1016/0921-4526(94)00493-F)

Publication date

1995

Published in

Physica B-Condensed Matter

[Link to publication](#)

Citation for published version (APA):

Nieuwenhuys, G. J., Mentink, S. A. M., Menovsky, A. A., Amato, A., Feyerherm, R., Gygax, F. N., Heffner, R. H., Lee, L. P., MacLaughlin, D. E., & Schenck, A. (1995). ?SR in antiferromagnetic UNiB₄. *Physica B-Condensed Matter*, 206-207, 470-472. [https://doi.org/10.1016/0921-4526\(94\)00493-F](https://doi.org/10.1016/0921-4526(94)00493-F)

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ELSEVIER

Physica B 206 & 207 (1995) 470–472

PHYSICA B

μ^+ SR in antiferromagnetic UNi₄B.

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Abstract

Positive-muon spin rotation (μ^+ SR) has been carried out in a single crystal of the hexagonal antiferromagnet UNi₄B, for which the magnetic structure has recently been resolved by Mentink et al. No spontaneous precession was observed in zero field with the μ^+ spin directed along the rhombohedral *b*-axis; instead, rapidly and slowly relaxing components (comprising 70% and 30% volume fraction, respectively) were found. On the other hand, a frequency of 5 MHz was detected when the μ^+ spin was directed along the *c*-axis. Transverse-field measurements indicate that the μ^+ is not stopped at a highly symmetric site. The experimental results are compared with calculations based on the magnetic structure.

UNi₄B crystallizes in the hexagonal CeCo₄B-type crystal structure with $a = 4.953$ Å and $c = 6.964$ Å. Since there are two U layers in this structure, the U–U distance along the *c*-axis is much smaller (3.482 Å), than that in the basal plane (4.953 Å). UNi₄B orders antiferromagnetically for temperatures below $T_N = 20$ K [1], where it attains a unique and new magnetic structure in the ordered phase [2] (see Fig. 1). Only 2/3 of the spins order and form a hexagonal structure in the basal plane, with moment directions in the plane and ferromagnetically coupled along the *c*-axis. No magnetic difference could be found between the two crystallographically inequivalent U sites. The remaining 1/3 of the spins do not couple in the basal plane, only ferromagnetically along the *c*-axis. UNi₄B also attains a crystallographic superstructure, which is not uncommon in this type of compound [3–5]. Since the exact atomic positions are

not yet known, except that it is very unlikely that the U atoms are displaced from their original sites, we will use the ideal CeCo₄B structure throughout this paper.

In this contribution, we present the results of a μ^+ SR investigation on a single crystal of UNi₄B in its unique antiferromagnetic state.

The large single crystal was grown using the tri-arc method [6] by the FOM-ALMOS center. The polycrystalline starting material was prepared by arc-melting stoichiometric amounts of the pure elements (U:3N, Ni:5N and B:3N). The high quality of the crystal was confirmed by X-ray diffraction. Subsequently the crystal was cut by spark erosion into slabs of 0.5-mm thickness with the *a*–*c* plane parallel to the surface. The μ^+ SR experiments were carried out on the general purpose spectrometer at the Paul-Scherrer Institute, Villigen, Switzerland. For more details on μ^+ SR, see Ref. [7]. Using different mountings of the crystal, the spin of the μ^+ could be directed along the *a*, *b* or *c*-axis.

In Fig. 2 we show the asymmetry observed in zero

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Fig. 1. 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 uranium moments, indicated by (1) and (2), are located in the center of and in between the magnetic vortices (thick lines). The nearest and next-nearest-neighbor magnetic coupling constants are indicated by J_1 and J_2 . After Ref. [2].

field after subtraction of a slowly decaying signal (probably due to muons stopped in the silver sample holder) at a temperature of 6 K with the μ^+ spin directed along the c -axis. (The counter is also in the c -axis direction.) Clearly a spontaneous precession is

Fig. 2. The asymmetry of the signal for UNi_4B at 6 K in zero field with the μ^+ spin directed along the crystallographic c -axis after subtraction of the signal due to the sample holder. The line represents a fit to two gaussian relaxing signals with 0 and 5.04 MHz frequencies, respectively.

observed with a frequency of 5 MHz (gaussian relaxation rate, σ , is $2.3 \mu\text{s}^{-1}$), together with a rapidly (gaussian) decaying signal, $\sigma(T \rightarrow 0) = 38 \mu\text{s}^{-1}$. Above T_N only an exponentially damped signal is observed, with a relaxation rate, λ , of about $0.5 \mu\text{s}^{-1}$. When the μ^+ spin is directed along the a or b -axis, only a rapidly decaying signal is found ($\sigma(T \rightarrow 0) = 38 \mu\text{s}^{-1}$). We have plotted the normalized results for the spontaneous frequencies and relaxation rates together with the ordered moment as obtained from neutron diffraction [2] as a function of reduced temperature in Fig. 3. The similarity between the temperature dependences is clear.

An experiment at 5.5 K in a longitudinal field of 0.25 T revealed an exponentially damped signal with $\lambda = 0.5 \mu\text{s}^{-1}$, and an asymmetry of 16% accompanied by a 3% gaussian relaxing signal with $\sigma = 2.3 \mu\text{s}^{-1}$, while the total asymmetry obtained from high temperature transverse field experiments is 20%. This shows that the rapid relaxation ($\sigma = 38 \mu\text{s}^{-1}$) observed in zero field is not due to dynamic behavior.

Transverse-field experiments have been carried out in 0.025 and in 0.5 T. The low field experiments showed an exponentially damped signal which disappeared rapidly below T_N , giving way to a rapidly (gaussian) damped signal as observed in zero field. The experiments in 0.5 T showed a (negative) frequency shift roughly proportional to the measured magnetic susceptibility at temperatures above 40 K.

The μ^+ site in UNi_4B is not known. We can

Fig. 3. The reduced values of the frequency of the spontaneous precession (Δ) and the gaussian relaxation rate of the zero frequency signal ($\circ$), both for the μ^+ spin parallel to the c -axis; the gaussian relaxation rate ($+$) for the spin parallel to the b -axis; and the ordered moment as observed by neutron diffraction ($\diamond$) as a function of the reduced temperature, T/T_N .

compare with the observations of Spada et al. [5] for the sites occupied by the hydrogen ion in LaNi_4B . They concluded that the 3f and/or the 12n sites (Wyckoff notation) are most probable. Both sites are not highly symmetrical with respect to the U magnetic structure. At 30 K a number of frequencies could be observed, indicating more than one magnetically inequivalent μ^+ site. Rotating the sample along the *c* axis revealed an anisotropy. Unfortunately, a unique fit to a limited number of frequencies appeared to be impossible, making a determination of the anisotropy of the Knight shift, and thus of the μ^+ site, impossible. At best we could distinguish a 10% signal, which was almost isotropic and two signals of between 2 and 6%, respectively, which showed an anisotropy in agreement with the results of a calculation of the dipolar fields in UNi_4B at the 3f site for the different directions of the external magnetic field. We therefore assume that these sites are at least partly occupied.

The magnetic ordering in UNi_4B has clearly been observed by μ^+ SR. Also no changes have been found to occur around a temperature of 10 K, where the magnetic susceptibility and the electronic specific heat attain a second maximum, in agreement with the results from neutron diffraction [2], where also no effects were found around 10 K. This supports the conclusion [2] that the second maximum in the specific heat and magnetic susceptibility in UNi_4B is not due to an extra phase transition or reorientation of the spins, but that macroscopic properties of UNi_4B should be considered as the summation of those of a three-dimensionally ordering antiferromagnet (two-thirds) and those of a ferromagnetic linear chain (one-third).

Based on the deduced magnetic structure [2], the possible spontaneous frequencies for the 3f sites have been calculated. The 3f sites surrounding the 'free' U ion in the hexagon depicted in the lower left of Fig. 1, have a frequency of 4.98 MHz, close to that observed when the μ^+ spin is directed along the *c*-axis. Due to

the magnetic structure most of the other possible sites become inequivalent, leading to frequencies between 3 and 50 MHz.

Acknowledgements

We acknowledge the expert help preparation of the single crystal by C.E. Snel and valuable discussions with J.A. Mydosh. This work was supported in part by the Dutch Foundation FOM and by the U.S. National Science Foundation, Grant no. DMR-9114911.

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