?SR in antiferromagnetic UNiB4


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\[ \mu^+ \text{SR in antiferromagnetic UNi}_4\text{B.} \]

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Abstract

Positive-muon spin rotation (\(\mu^+\)SR) has been carried out in a single crystal of the hexagonal antiferromagnet UNi\(_4\)B, for which the magnetic structure has recently been resolved by Mentink et al. No spontaneous precession was observed in zero field with the \(\mu^+\) 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 \(\mu^+\) spin was directed along the \(c\)-axis. Transverse-field measurements indicate that the \(\mu^+\) is not stopped at a highly symmetric site. The experimental results are compared with calculations based on the magnetic structure.

UNi\(_4\)B crystallizes in the hexagonal CeCo\(_4\)B-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). UNi\(_4\)B 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. UNi\(_4\)B 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 CeCo\(_4\)B structure throughout this paper.

In this contribution, we present the results of a \(\mu^+\)SR investigation on a single crystal of UNi\(_4\)B 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 \(\mu^+\)SR experiments were carried out on the general purpose spectrometer at the Paul-Scherrer Institute, Villigen, Switzerland. For more details on \(\mu^+\)SR, see Ref. [7]. Using different mountings of the crystal, the spin of the \(\mu^+\) could be directed along the \(a\), \(b\) or \(c\)-axis.

In Fig. 2 we show the asymmetry observed in zero field.
Fig. 1. Magnetic structure of hexagonal UNi$_4$B, 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 $\mu^+$ spin directed along the c-axis. (The counter is also in the c-axis direction.) Clearly a spontaneous precession is observed with a frequency of 5 MHz (gaussian relaxation rate, $\alpha$, is 2.3 $\mu$s$^{-1}$), together with a rapidly (gaussian) decaying signal, $\sigma(T \to 0) = 38 \mu$s$^{-1}$. Above $T_N$ only an exponentially damped signal is observed, with a relaxation rate, $\lambda$, of about 0.5 $\mu$s$^{-1}$. When the $\mu^+$ spin is directed along the a or b-axis, only a rapidly decaying signal is found ($\sigma(T \to 0) = 38 \mu$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 $\lambda = 0.5 \mu$s$^{-1}$, and an asymmetry of 16% accompanied by a 3% gaussian relaxing signal with $\sigma = 2.3 \mu$s$^{-1}$, while the total asymmetry obtained from high temperature transverse field experiments is 20%. This shows that the rapid relaxation ($\sigma = 38 \mu$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 $\mu^+$ site in UNi$_4$B is not known. We can

Fig. 2. The asymmetry of the signal for UNi$_4$B at 6 K in zero field with the $\mu^+$ 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.

Fig. 3. The reduced values of the frequency of the spontaneous precession ($\Delta$) and the gaussian relaxation rate of the zero frequency signal (○), both for the $\mu^+$ 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 (○) 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 LaNi₅B.
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 in-
equivalent $\mu^+$ 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 $\mu^+$ 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 agree-
ment with the results of a calculation of the dipolar
fields in UNi₄B at the 3f site for the different direc-
tions of the external magnetic field. We therefore
assume that these sites are at least partly occupied.

The magnetic ordering in UNi₄B has clearly been
observed by $\mu$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 UNi₄B is not due
to an extra phase transition or reorientation of the
spins, but that macroscopic properties of UNi₄B
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 $\mu^+$ 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.

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