?SR in antiferromagnetic UNiB4


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Abstract

Positive-muon spin rotation ($\mu^+\text{SR}$) 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 $a = 4.953 \, \text{Å}$ and $c = 6.964 \, \text{Å}$. Since there are two U layers in this structure, the U–U distance along the $c$-axis is much smaller ($3.482 \, \text{Å}$), than that in the basal plane ($4.953 \, \text{Å}$). UNi$_4$B orders antiferromagnetically for temperatures below $T_N = 20 \, \text{K}$ [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 $\mu^+\text{SR}$ 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 $\mu^+\text{SR}$ experiments were carried out on the general purpose spectrometer at the Paul-Scherrer Institute, Villigen, Switzerland. For more details on $\mu^+\text{SR}$, 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
Fig. 1. Magnetic structure of hexagonal UNi₄B, 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. [21.

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, $\sigma(T\rightarrow0)=38$ $\mu$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 ($\sigma(T\rightarrow0)=38$ $\mu$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 UNi₄B is not known. We can

Fig. 2. The asymmetry of the signal for UNi₄B 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 ($\bigcirc$), 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 ($\bigtriangleup$) 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$_5$B. 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 $\mu^+$ 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 agreement with the results of a calculation of the dipolar fields in UNi$_4$B 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$_4$B 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 UNi$_4$B is not due to an extra phase transition or reorientation of the spins, but that macroscopic properties of UNi$_4$B 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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References


