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A μSR magnetic study of UNiGe

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Abstract

We have carried out μSR spectroscopy on polycrystalline UNiGe between 2 K and 100 K. The existence of two magnetic transitions at $T_N = 51$ K and $T_1 = 41.5$ K is confirmed. The μSR spectra clearly reveal that the magnetic state between 51 K and 41.5 K is an incommensurate spin structure ruling out a spin–slip structure which had been considered an alternative. Below 41.5 K the spectra are compatible with simple antiferromagnetic order. The local field for $T \to 0$ is $B_\mu = 170$ mT, a comparatively low value, indicating a rather small uranium ordered moment. When going from the commensurate to the incommensurate structures at T_1 a sudden reduction in local field by 23% occurs reflecting an equal change in ordered moment. The transition at T_1 is sharp, but T_N extends over roughly 5 K. The antiferromagnetic spin structure exhibits persistent spin fluctuations in the limit $T \to 0$, implying the presence of some additional spin interactions which tries to suppress long-range magnetic order.

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1. Introduction

Compounds of the type MTX (M = 4f- or 5f-element, T = transition metal, X = p-electron element), also referred to as 1:1:1 compounds, display a multitude of highly interesting, often controversial features, especially with respect to their magnetic properties. In particular, many materials with M = Ce are well known heavy-fermion compounds, but also some M = U alloys belong to this class. Thus, MTX compounds have been a subject of extensive studies over many years [1].

The MTX alloys crystallize in a number of different structures. UNiGe forms the orthorhombic TiNiSi structure (space group *Pnma*), which is the ordered variant of the $CeCu_2$ structure that in turn is a distortion of the hexagonal AlB_2 structure. The nearest U–U distance has a slight zig-zag along the *a*-axis, which is also the magnetically hard axis. Originally, only one magnetic transition at 41.5 K from paramagnetism to collinear antiferromagnetism with moments either along the *b*- [2] or *c*-axis [3] was proposed. Subsequent bulk magnetic measurements [4–6] indicated the existence of a second magnetic transition at 51 K, and this prompted a new thorough investigation of the magnetic structure by neutron diffraction [7,8] on a single crystal of UNiGe. Below 41.5 K, a single-*q* antiferromagnetic structure with $q = (0, \frac{1}{2}, \frac{1}{2})$ and moments of 0.96 μ_B (at 20 K) mainly in the *b*–*c* plane was found. Polarized neutron diffraction proved the structure not be precisely collinear, the moments having a canting angle around 20° with respect to the *b*–*c* plane. In the range $41.5\text{ K} < T < 51$ K, UNiGe was initially proposed to exhibit an incommensurate spin structure with $q = (0, \delta, \delta)$, where δ varies between 0.37 at 44 K and 0.35 at 50 K. A more recent study [9], however, indicated that $q = (0, \delta_1, \delta_2)$ with δ_1 to be very close to $\frac{1}{3}$ and only δ_2 varying between 0.35 and 0.37. For the incommensurate phase, the moments are reduced to 0.36 μ_B (at 46 K), but the tilting out of the *b*–*c* plane is probably still in existence. Even though the *x*-components to the magnetic moment of UNiGe are small, theoretical consider-

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ations [10] predicted that these out-of-plane components will persist even in magnetic fields beyond the metamagnetic transition fields applied in the *b*–*c* plane. Recently, this was confirmed experimentally for fields applied along the *c*-axis [11].

The interesting magnetic behavior of UNiGe prompted us to look for the local magnetic properties employing μSR spectroscopy. In particular, we were interested to test whether the moment configuration of UNiGe between 41.5 K and 51 K is truly incommensurate or whether it exhibits a so-called 'spin–slip' structure similar to the case of another MTX compound, namely CePtSn. For this compound, neutron diffraction data [12] were initially interpreted in terms of an incommensurate magnetic structure, but subsequent μSR studies [13] excluded incommensurability and induced a reanalysis of the magnetic structure data [14] which revealed that the magnetic spin arrangement in this compound consists of islands with commensurate moment configuration separated by coherent spin–slips every 20th layer or so. It was then verified in turn that this spin–slip structure is fully compatible with the μSR results [15].

So far, μSR experiments have naturally concentrated on the established heavy-fermion members (see, for example [16]). However, besides heavy-fermion properties, the MTX compounds can be used to study the transition from local moment behavior to more itinerant moment character and finally to Pauli paramagnetism, particularly in U compounds [1]. Although this subject would be well suited for μSR studies, only a rather limited number of experiments on non-heavy-fermion MTX alloys has appeared in the literature. The few examples include CeCuSn [17], UNiGa [18] and UPdSn [19]. However, these compounds all crystallize in different structures than UNiGe.

2. Experimental

Our μSR data were taken at TRIUMF using the M20 surface muon beam line. Since surface muons penetrate only small thicknesses of material, the sample container must be open on the beam entrance side. We used a powder sample of UNiGe made of depleted U^{238}, and clearly the powder had to be fixed rigidly to ensure radiation safety rules (including cooling to Kelvin temperatures). For that, the powder was set in an aluminum can by STYCAST epoxy, which is known to give reliable bonding even at cryogenic temperatures. The sample was then mounted inside a He-gas flow cryostat, which allows continuous setting of temperatures between 300 K and 2 K with a stability better than 0.1 K. Since the sample is inside the He-gas flow, good temperature exchange to the sample and negligible temperature gradients in the sample are ensured. The longitudinal geometry with positron telescopes in backward (B) and forward (F) direction was used. A longitudinal field (LF) could be applied via a pair of Helmholtz coils. Most of our data were taken in zero field (ZF). With the help of small correction coils the residual field from the cyclotron and other magnets in the surrounding was fully zeroed. A weak (<10 mT) transverse field (TF) could be applied using a pair of cross coils.

3. μSR magnetic response

Positive muons are implanted in the material to be studied and come to rest at one or more interstitial lattice site(s). The temporal behavior of the muon spin due to the interaction with the local field B_μ at the muon stopping site (Larmor precession, static and/or dynamic relaxation of the initial spin polarization) is reflected in the time dependence of the asymmetry $A(t)$ of count rates $N(t)$ in the backward (B) and forward (F) positron telescopes:

$$A(t) = \frac{N_{\rm B}(t) - N_{\rm F}(t)}{N_{\rm B}(t) + N_{\rm F}(t)} = A(0)\,G(t) \qquad (1)$$

The time t starts when the muon is deposited in the sample and extends usually to about four muon mean lifetimes ($\tau_\mu = 2.2\,\mu$s). $A(0)$ is the initial asymmetry (or signal strength) which is given by geometrical properties of the μSR spectrometer and commonly around 0.2. $G(t)$ is the muon response function containing the physical relevant parameters like precession frequency and relaxation rates. In practice, one has to apply some corrections in forming the asymmetry $A(t)$ such as taking into account the different sensitivities of the two telescopes and the random background counts (e.g. from cosmic radiation) leading to the so-called 'corrected asymmetry'. This is routinely done within the least square algorithm which fits a properly selected theory function $G(t)$ to the measured $A(t)$. A plot of the corrected asymmetry versus time is generally referred to as the μSR spectrum. In most experimental setups, it is unavoidable that some muons, which are stopped outside the sample (e.g. in the sample holder), are recorded as well. This produces a 'background signal' $A_{\rm bkg}(t)$ that has to be subtracted from the measured signal $A(t)$ to obtain the true sample signal $A_{\rm spl}(t)$. The strength of the background signal $A_{\rm bkg}(0)$ can usually be estimated from TF data. For the physical interpretation of μSR spectra, ZF and sometimes LF measurements are preferred over TF data.

In a powder sample with commensurate long-range magnetic order (LRO), the μSR response function takes the form:

$$G_{\rm LRO}(t) = \tfrac{2}{3}\,\exp[-\lambda_{\rm tr}t]\,\cos[\omega_\mu t + \phi] + \tfrac{1}{3}\,\exp[-\lambda_{\rm lg}t] \qquad (2)$$

with the Larmor precession frequency:

$$\omega_\mu = \gamma_\mu \langle B_\mu \rangle. \qquad (3)$$

Here, $\gamma_\mu = 850\,\mu{\rm s}^{-1}\,{\rm T}^{-1}$ is the muon gyromagnetic ratio and $\langle B_\mu \rangle$ is the average field at the muon site generated by surrounding magnetic dipoles (atomic and/or nuclear). A phase shift $\phi \neq 0$ may arise from slight misalignment of the B and F telescopes. Since the oscillatory term in $G_{\rm LRO}(t)$ is present without an external field being applied one often speaks of 'spontaneous spin precession'. Its appearance is the tell-tale μSR signature of a commensurate long-range ordered magnetic state. μSR cannot determine the spin structure directly. However, if the muon stopping site is known (which is rarely the case), the field B_μ can be calculated by dipolar lattice sums for the proposed spin structure and compared with the measured spectrum. A pertinent example is CePtSn discussed in Section 1.

The transverse relaxation rate $\lambda_{\rm tr}$ in $G_{\rm LRO}(t)$ (corresponding to $1/T_2$ in NMR) is predominantly given by the static distribution width of B_μ. In an ideal ordered spin structure this distribution should be zero. In practice, however, there are always local distortions of the spin lattice and $\lambda_{\rm tr}$ is by no means negligible. The longitudinal relaxation rate $\lambda_{\rm lg}$ is solely determined by mag-