Muon Spin Dynamics and Sites in GdNi₅

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Abstract. In GdNi₅, the muon localizes at the $3f$ interstitial site and below ~ 80 K a second muon site becomes populated, the so-called 6_{ring} site. This ring site is metastable and the muon hops to the $3f$ site. We determine the mean time of stay of the muon in the ring site and show that it is governed by a multi-phonon quantum diffusion process. The coincidence energy is measured to be $E_a = 272(10)$ K and the tunneling matrix element $J = 0.11(2)$ meV.

Key words: muon diffusion, muon spin rotation and relaxation.

1. Introduction

The magnetic properties of GdNi₅ have been studied intensively and are well understood. The Gd $4f$ shell is spherical and therefore its magnetic moment is insensitive to crystal field effects. The magnetic interaction is strong compared to other rare earth atoms (de Gennes scaling) and governs the second order magnetic phase transition at $T_C = 30.85$ K. GdNi₅ is a ferrimagnet with a Gd moment of $7\mu_B$ and an induced Ni moment of $0.16\mu_B$. The moments are preferentially aligned along the c axis and this anisotropy is caused by the dipolar interaction [1].

In general, the muon localizes interstitially inside its host and this may lead to a distortion of its near neighbour environment. Because of its positive charge, it acts as a light proton, i.e., it can be considered as a hydrogen-like particle. At elevated temperatures the muon will diffuse through the lattice of its host and motional narrowing is commonly observed.

Two non-equivalent muon sites become populated below 80 K in GdNi₅ [2]. One stable site, the $3f$ site, which is also occupied by the muons at temperatures above 80 K, and a metastable site, the $6k$ or $6m$ site, both located in a ring surrounding the $1b$ site (Figure 2) and further referred to as 6_{ring} . The muon occupies the six 6_{ring} sites simultaneously within the time range of the μ SR experiment (10^{-9} s).

Figure 1. Unit cell of GdNi₅ (hexagonal crystal structure, space group P6/mmm). The muon localizes at the $3f$ site in the whole temperature range and also below ~ 80 K in either the $6m$ ($0.13, 0.26, \frac{1}{2}$) or $6k$ ($0.225, 0, \frac{1}{2}$) sites. The muon occupies all six sites within the time range of the experiment [2, 4]. Similar work on UNi₂Al₃ [3] suggests that the muon forms an extended localized ring-shaped orbit. We refer to this site as the 6_{ring} site.

An equivalent muon localization has been observed recently in the isostructural compound UNi₂Al₃ [3], where the authors suggest that the muon actually forms an extended state over the six ring sites. In this paper we report on the muon hopping from the metastable 6_{ring} site to the $3f$ site which occurs between 35 and 80 K. This work is a continuation of [4] where we present a first determination of the muon sites. Here we present the combined results obtained from transverse field μ SR measurements with the external field B_{ext} applied along the c and a axis.

2. Experimental

A GdNi₅ single crystal was prepared by the Czochralski method in Amsterdam from starting materials of at least 99.99% purity. The sample was shaped into a sphere (diameter of 5.5 mm) with spark erosion in order to avoid demagnetisation corrections [2]. The μ^+ SR transverse field measurements took place at the π M3 beamline of PSI and the data presented here were taken with the a or c axes parallel to B_{ext} ($B_{\text{ext}} = 0.6$ T) between 35 and 80 K. In this temperature region the muon hopping from the 6_{ring} to the $3f$ site is prominent. Below 35 K the muon remains at the 6_{ring} site during its lifetime when implanted, whereas above 80 K the muon does not populate the 6_{ring} site in the time window of the experiment (longer than ~ 1 ns).

Including the muon hopping, the total polarization function is written as [5]

Figure 2. Temperature dependence of the muon residence time at the δ_{ring} site. The solid line is a fit to Equation (2) but an Arrhenius law gives a similar result as discussed in the text. At $T = 60$ and 65 K where only an open dot is visible, τ_{ring} is equal for both directions of B_{ext} .

$$P_X(t) = f_{3f} e^{-\lambda_X^{(3f)} t} \cos(\omega_\mu^{(3f)} t) + f_{\text{ring}} \left\{ e^{-\check{\lambda}_X^{(\text{ring})} t} \cos(\omega_\mu^{(\text{ring})} t) + \frac{e^{-\lambda_X^{(3f)} t} \cos(\omega_\mu^{(3f)} t + \theta_m) - e^{-\check{\lambda}_X^{(\text{ring})} t} \cos(\omega_\mu^{(\text{ring})} t + \theta_m)}{\tau_{\text{ring}} \sqrt{(\check{\lambda}_X^{(\text{ring})} - \lambda_X^{(3f)})^2 + 4(\delta\omega_\mu)^2}} \right\} \quad (1)$$

with

$$\check{\lambda}_X^{(\text{ring})} = \lambda_X^{(\text{ring})} + \frac{1}{\tau_{\text{ring}}}, \quad \delta\omega_\mu = \frac{\omega_\mu^{(\text{ring})} - \omega_\mu^{(3f)}}{2} \quad \text{and} \\ \tan \theta_m = \frac{2 \delta\omega_\mu}{\check{\lambda}_X^{(\text{ring})} - \lambda_X^{(3f)}}.$$

$f_{\text{ring}}, f_{3f}, \lambda_X^{(\text{ring})}, \lambda_X^{(3f)}, \omega_\mu^{(\text{ring})}, \omega_\mu^{(3f)}$ are the relative weight factors ($f_{\text{ring}} + f_{3f} = 1$), damping rates and precession frequencies of the muon at the specific sites. We analyzed our spectra with Equation (1) and a fixed ratio $f_{3f}/f_{\text{ring}} = 1/2$. It is difficult to separate $\lambda_X^{(\text{ring})}$ and τ_{ring} since the fit is mostly sensitive to the sum of both, $\check{\lambda}_X^{(\text{ring})} = \lambda_X^{(\text{ring})} + 1/\tau_{\text{ring}}$. Therefore we have determined $\lambda_X^{(\text{ring})}$ by other means. This is new compared to our previous analysis [4] and briefly described below.

An analysis of the zero-field data at elevated temperature presented in [1] shows that the spin correlations of the Gd moments are isotropic. The increase of $\lambda_X^{(3f)}(T)$ observed when approaching the magnetic ordering temperature from above reflects

the critical slowing down of the Gd moments. Since $\lambda_X^{(\text{ring})}$ is fully due to the Gd spin dynamics, it can be calculated assuming the Gd spin fluctuations are still isotropic below 80 K. We used this in our fits to extract τ_{ring} . The details on the calculation of the spin dynamics will be given in a paper which is in preparation.

The result for $\tau_{\text{ring}}(T)$ is plotted in Figure 1. The consistency between $\tau_{\text{ring}}(T)$ for $B_{\text{ext}} \parallel c$ and $B_{\text{ext}} \perp c$ supports the validity of our whole method of analysis.

3. Muon hopping analysis

The temperature dependence of τ_{ring} (Figure 1) suggests either a multi-phonon assisted tunneling process is at play or simply a muon jumping over the energy barrier between two interstitial sites. In the first process, multi-phonon processes equalize for an extremely short time (of the order of 10^{-13} s) the local energy levels of the potential well where the muon is localized and where it likes to jump. When this occurs the muon tunnels through the energy barrier between the two sites. Within the framework of the small polaron theory [6, 7],

$$\tau_{\text{ring}} = \frac{\hbar}{J^2} \sqrt{\frac{4E_a k_B T}{\pi}} \exp\left(\frac{E_a}{k_B T}\right), \quad (2)$$

where E_a is an energy necessary to get the coincidence configuration of both sites and J the tunneling matrix element. From Figure 1 we obtain $E_a = 272(10)$ K and $J = 0.11(2)$ meV. The jumping mechanism over the energy barrier is described by an Arrhenius law. The data of Figure 1 are equally well described by this law with $E_a = 244(10)$ K and $\tau_0 = 1.2(2)$ ns. $1/\tau_0$ is the attempt frequency. But, referring to the discussion of Karlsson [8], the anomalous low value of τ_0 indicates that the muon mobility is governed by the multi-phonon assisted tunneling process.

Multi-phonon assisted muon tunneling has been reported before [9, 10] in Al and Cu and the spread in J is large: ~ 1 meV for Al and 36 μeV for Cu. Our value of 0.12 meV for GdNi₅ seems consistent with these results. Note that the uncertainty for Al is relatively large [8], in contrast to GdNi₅. The value of E_a for Al (32 meV [8]) and GdNi₅ (23 meV) are in the same range.

4. Conclusions

We have determined the residence time of the muon at the 6_{ring} site in GdNi₅ as function of temperature. The muon hopping process from this site to the $3f$ site is characterized by a multi-phonon-assisted tunneling process with coincidence energy $E_a = 272(10)$ K and tunneling matrix element $J = 0.11(2)$ meV.

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