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Gubbens, P.C.M.; Moolenaar, A.A.; Dalmas de Réotier, P.; Yaouanc, A.; Kayzel, F.E.; Franse, J.J.M.; Prokes, K.; Snel, C.E.; Bonville, P.; Hodges, J.A.; Imbert, P.; Pari, P.

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SPIN DYNAMICS IN RENi₅ FERROMAGNETS BY μSR MEASUREMENTS

P.C.M. GUBBENS and A.A. MOOLENAAR
Interfaculty Reactor Institute, Technical University Delft, 2629 JB Delft, The Netherlands

P. DALMAS DE RÉOTIER and A. YAOUANC
CEA/Département de Recherche Fondamentale sur la Matière Condensée, 85X, F-38041 Grenoble-Cedex, France

F. KAYZEL, J.J.M. FRANSE and K. PROKES
Natuurkundig Laboratorium, Universiteit van Amsterdam, 1018 XE Amsterdam, The Netherlands

C.E. SNEL
Kamerlingh Onnes Laboratorium der Rijksuniversiteit Leiden, 2300 RA Leiden, The Netherlands

P. BONVILLE, J.A. HODGES, P. IMBERT and P. PARI
Centre d'Etudes de Saclay, DRECAM/SPEC, F-91191 Gif-sur-Yvette, France

We report a zero field μSR study of single crystals of the $RENi_5$ ferromagnets where RE=Gd, Er, Dy or Tm. Our work points out the strong influence of the crystal electric field on the rare earth total momentum dynamics: whereas the spin-lattice relaxation process below T_C is controlled by a two magnon mechanism for a weakly anisotropic magnet ($GdNi_5$), the phonons drive the relaxation for a strongly anisotropic magnet such as $ErNi_5$. For $TmNi_5$ a comparison is made between the fluctuation time measured by μSR and ^{169}Tm Mössbauer spectroscopy.

The $RENi_5$ intermetallics compounds where RE is a heavy rare earth ion are ferromagnets which crystallize with the $CaCu_5$ crystal structure. Their static magnetic properties are now well understood. They are controlled by the crystalline electric field (CEF) acting on the rare earth ions (the Ni contribution to magnetism is negligible). Their dynamic magnetic properties have practically not been investigated up to now. In this paper we show that, as expected [1], μSR relaxation measurements are a valuable tool for the identification of the mechanisms responsible for the rare earth moment dynamics. We will discuss in some details μSR data recorded in the easy axis (c axis) ferromagnets $GdNi_5$ ($T_C \sim 37$ K) and $ErNi_5$

($T_C \sim 9.2\ K$). Because of lack of space we will only mention the results obtained for $TmNi_5$. The data obtained for $DyNi_5$ are similar to those presented previously for $TbNi_5$. Preliminary work in $GdNi_5$, $TbNi_5$ and $ErNi_5$ have been presented elsewhere [2 − 4].

The measurements have been performed either at the μSR spectrometer of the ISIS facility located at the Rutherford Appleton Laboratory (RAL, UK) or at the General Purpose Spectrometer of the μSR facility of the Paul Scherrer Institute (PSI, Switzerland). The ISIS data have been recorded from 340 K to 0.2 K with a longitudinal experimental set-up and have been analysed taking into account the two-pulse structure of the muon beam [5]. A sample is prepared using the following method: a single crystal of about 1 cm^3 grown by the Czochralski technique is cut by spark erosion and the pieces are glued on a silver plate. The sample is then a mosaic of single crystals of about 8 cm^2. We have used $GdNi_5$, $DyNi_5$ and $TmNi_5$ samples with the c axis perpendicular to the sample plane and two $ErNi_5$ samples with the c axis either parallel or perpendicular to the sample plane. Thanks to the PSI spin rotator, we have been able to perform measurements with the initial muon beam polarisation either parallel or perpendicular to the c axis using only one sample. With this trick we have checked that the μSR damping rate recorded on the two $ErNi_5$ samples is the same for identical experimental geometry. The samples have been grown at the university of Amsterdam. Most of them have been also cut in Amsterdam, the others in Leiden.

We first present our $GdNi_5$ data. This compound has a weak magnetic anisotropy which is probably only due to the dipolar interaction between the Gd^{3+} magnetic moments. At PSI we have recorded a spectrum in the ferromagnetic phase at 3.3 K with the initial beam polarisation perpendicular to the c axis. We see two frequencies. We denote respectively a_i, λ_i and ν_i the initial asymmetry, exponential damping rate and frequency for component i. A fit gives $a_1 = 0.098$ (3), $\lambda_1 = 5.3$ (2) MHz, $\nu_1 = 220.89$ (3) MHz and $a_2 = 0.061$ (3), $\lambda_2 = 4.2$ (3) MHz, $\nu_2 = 79.68$ (4) MHz. At ISIS we have recorded zero field spectra with the initial muon beam polarisation parallel to the c axis. As the value of the initial asymmetry does not change when crossing the Curie temperature we deduce that the local magnetic field at the muon site is parallel to the c axis. The PSI data lead to the same conclusion. This result strongly suggests that the c axis is the easy axis. This conclusion is not consistent with reported magnetisation measurements [6] but agrees with ^{155}Gd Mössbauer data [7 − 9]. This discrepancy is maybe explained by the fact that the magnetisation measurements are done in an applied field which can modify the magnetic properties of the compound.
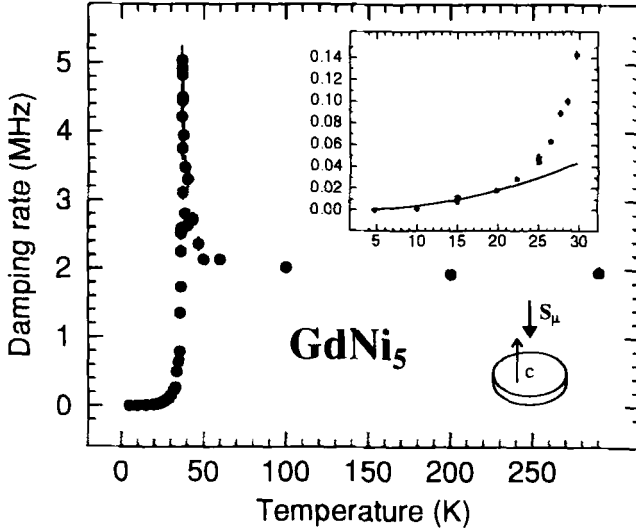
Fig. 1. $\lambda_Z(T)$ measured in zero field at ISIS on *GdNi$_5$*. In the insert is displayed the low temperature part.

Because we observe two frequencies, there are two localization sites for the muon. Symmetry arguments tell us that a crystallographic site gives only one magnetic site. In $PrNi_5$ we note that the muon seems to be localized in only one site [10]. It is surprising that the muon does not have the same localization site(s) in compounds of a given rare earth series. Definitely more work is needed on the muon localization problem but the fact that we do not know the muon localization sites will not influence the results of our data analysis. The zero field depolarisation functions recorded with the initial beam polarisation and the c axis parallel are all well described by exponential functions. In Fig. 1 we show $\lambda_Z(T)$. At high temperature λ_Z is temperature independent. This is expected for a Heisenberg magnet [11]. λ_Z exhibits a maximum near T_C due to the slowing down of the dynamics of the Gd^{3+} total moments. We will not analyse these data because the temperature regulation was not precise enough. We note that such a study would yield information on the effect of the dipolar interaction on the spin fluctuations [12]. At sufficiently low temperature, $T \lesssim T_C/2$, $\lambda_Z(T)$ varies as $T^2 \ln(T)$ which can be explained by a two magnon scattering process [12]. From the fit (see the insert of Fig. 1) we deduce that the spin wave stiffness constant is $D = 4.2 \ (2) \ meV \cdot \mathring{A}^2$. The deduced D value is independent of the muon site [12].

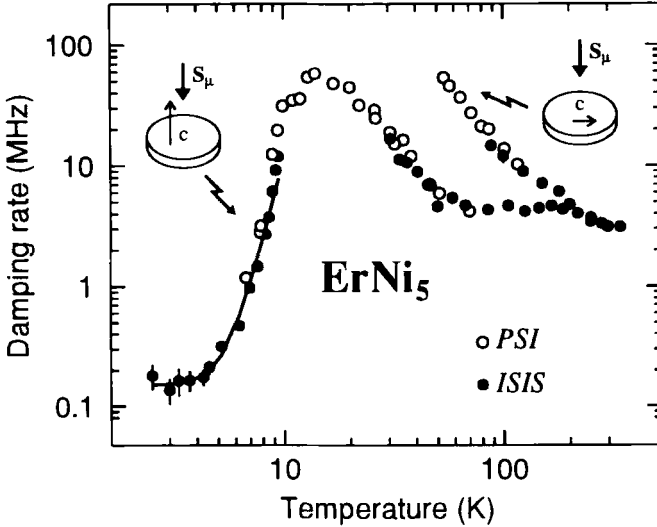We report now our $ErNi_5$ data. When the muon beam polarisation is

Fig. 2. $\lambda_Z(T)$ measured in zero field on $ErNi_5$. The small discrepancy between the data at low T can be due to the fact that they have been recorded at two different laboratories.

perpendicular to the c axis, the spectra are well described by a single exponential component, the asymmetry of which is temperature independent. Below $\simeq 55$ K the damping rate is too large to be measured. We could not find any signal below $T_C = 9.2$ K. When the muon beam is parallel to the c axis a single exponential function describes the data recorded above about 7 K. Below that temperature the signal is a sum of two components: an exponential and a time independent function. Below 12 K we observe a drop of the total initial asymmetry by $\simeq 25$ % compared to its high temperature value. Note that at this temperature $\lambda_Z(T)$ is maximum (see Fig. 2). This complicated temperature behaviour of the asymmetry indicates that the spin dynamics is quasi-static [12] and/or the possibility of more than one muon site. Fig. 2 shows that the damping is strongly anisotropic even at $T \sim 20$ T_C. At these high temperatures this behaviour can be understood as an effect of the CEF on the fluctuations induced by the spin-spin interaction [3]. At lower temperatures (below ~ 50 K) the fluctuations are quasi-static as indicated by the ^{166}Er Mössbauer study [13]. Again we attribute this slow dynamics to the CEF. But in this case the relaxation is probably induced by an indirect process between the two levels of the Kramers ground state doublet. We now consider the data for $T < T_C$ which are, at present, more amenable to a quantitative understanding. The
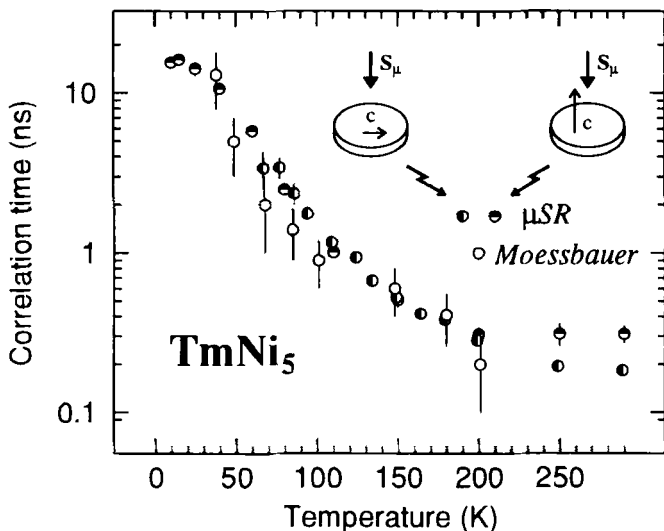
Fig. 3. Comparison between τ deduced from μSR and ^{169}Tm Mössbauer data for $TmNi_5$.

full line is a fit to the following formula

$$\lambda_Z(T) = a \coth(\Delta/k_BT) + bT^7, \tag{1}$$

where $a = 0.15\ MHz$, $\Delta/k_B \geq 5\ K$ and $b = 1.1\ Hz \cdot K^{-7}$. This strong temperature dependence is a clear signature that the muon depolarisation is induced by the phonons [14]. The physical picture is as follows. For $T <$ T_C, λ_Z is proportional to the fluctuation rate of the Er^{3+} total moments. The interaction between these moments is described in the molecular field approximation. In addition we take into account the CEF acting on the ions. Therefore, in this approximation, *the ions are treated independently.* Thus we can use well known Electron Paramagnetic Resonance results. The muon depolarisation is induced by the relaxation of the ions between their energy levels. This relaxation is produced by the magnetoelastic coupling described by an Hamiltonian which is the sum of terms consisting of products of strain operators by Stevens operators [14]. This coupling induces transitions between energy levels with $\Delta m = 1$ and $\Delta m = 2$. In our model the first and second terms of Eq. (1) describe respectively the relaxation induced by one and two phonon processes. This interpretation is consistent with the known energy level diagram [15].

In Fig. 3 we compare the correlation time τ extracted from ^{169}Tm Mössbauer [16] and μSR data for $T > T_C$. For the μSR data we have used

the "motional narrowing" limit formula. Because of the uncertainty concerning the muon site(s) and the coupling of its spin to the rare earth total momenta, the comparison is made for the two directions of the initial muon beam polarisation relative to the c axis. It is noteworthy that the Mössbauer and μSR correlation times have the same temperature dependence in their common temperature range. Note that we have performed the same type of comparison with the same success in the case of $YbNi_5$ [17].

Acknowledgements

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