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LETTER TO THE EDITOR

The positive muon relaxation rate at low temperature in a Heisenberg ferromagnet

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Abstract. We show that the positive muon spin depolarization should be controlled at low temperature by a magnon Raman scattering for a Heisenberg ferromagnet if the degree of symmetry at the muon site is not too high. The data recently reported for the intermetallic ferromagnet GdNi₅ support our conclusion.

Recently it has been shown by Yaouanc *et al* (1990) that the positive muon spin relaxation (μ SR) method can be used to study the electronic spin excitations of rare-earth intermetallic magnets at low temperature. In particular these authors have noted that in the ordered state of the ferromagnet GdNi₅ (its Curie temperature is $T_C = 31.45$ (5) K) the longitudinal μ SR depolarization function, $P_z(t)$, can be described by an exponential function, $P_z(t) = \exp(-\lambda_z t)$. In our notation we take the z axis parallel to the mean value of the local magnetic field at the muon site in the ordered magnetic state. The longitudinal depolarization is due to the muon spin flip along this field direction (Schenck 1985). At low temperature, $13 \text{ K} < T < 25 \text{ K}$, λ_z follows a temperature power law, $\lambda_z = \eta T^2$, with $\eta = 0.191$ (4) $\times 10^{-3} \text{ MHz K}^{-2}$ (Dalmas de Réotier 1990, Dalmas de Réotier *et al* 1991a, b). Because of the restricted temperature range of the power law fit, the value of the exponent was fixed. In this letter we show that this result can be explained if we suppose that the μ SR is due to a Raman scattering of the magnons against the muon spin.

Below we first express λ_z in terms of the correlation functions of the total angular momentum of the rare-earth ions. We then compute these functions in the linearized spin-wave approximation for a Heisenberg magnet with a magnetic anisotropy represented by an effective magnetic field. Finally we use the result of the computation to discuss the data obtained on GdNi₅ at low temperature.

When the fluctuations of the total angular momentum of the rare-earth ions are sufficiently fast, it can be shown (McMullen and Zaremba 1978, Dalmas de Réotier and Yaouanc 1991) that

$$P_z(t) = \exp(-\psi_z(t)) \quad (1)$$

with

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$$\psi_z(t) = \frac{\gamma_\mu^2}{2} \int_0^t d\tau (t - \tau) (\exp(i\omega_\mu \tau) \Phi_{+-}(\tau) + \exp(-i\omega_\mu \tau) \Phi_{-+}(\tau)) \quad (2)$$

or

$$\psi_z(t) = \gamma_\mu^2 \int_0^t d\tau (t - \tau) [\cos(\omega_\mu \tau) (\Phi_{xx}(\tau) + \Phi_{yy}(\tau)) + \sin(\omega_\mu \tau) (\Phi_{xy}(\tau) - \Phi_{yx}(\tau))]. \quad (3)$$

Here

$$\Phi_{\alpha\beta}(t) = \frac{1}{2} (\langle \delta B_\alpha(\tau) \delta B_\beta \rangle + \langle \delta B_\beta \delta B_\alpha(\tau) \rangle)$$

is the symmetrized correlation function of the fluctuations of the α - and β -components of the local magnetic field at the muon site. By definition we have $\delta B_z = \beta B_x \pm \beta B_y$ and $B_\alpha = \langle B_\alpha \rangle + \delta B_\alpha$. $\langle A \rangle$ stands for the thermal average of A . The time evolution of the fluctuations is governed by the Heisenberg equation

$$\delta B_\alpha(t) = \exp(i\mathcal{H}_m t/\hbar) \delta B_\alpha \exp(-i\mathcal{H}_m t/\hbar)$$

where \mathcal{H}_m is the Hamiltonian that describes the magnetic properties of the magnet. $\omega_\mu = 2\pi\nu_\mu = \gamma_\mu B$ where γ_μ is the muon gyromagnetic ratio, $\gamma_\mu = 8.516 \times 10^8 \text{ rad s}^{-1} \text{ T}^{-1}$. We have given $\psi_z(t)$ with the $\{+, -, z\}$ and $\{x, y, z\}$ coordinates because these two forms of $\psi_z(t)$ can both be useful, depending on what one is looking for. The rare-earth magnetic moments fluctuate with a characteristic time of $\approx 10^{-12} \text{ s}$. Therefore, as the experimental time window is $10^{-8} \text{ s} \leq t \leq 10^{-5} \text{ s}$, we can neglect τ in the $t - \tau$ factor of (2) and (3) and extend the integrals to infinity. In practice we have $\nu_\mu < 500 \text{ MHz}$ in intermetallics (Schenck 1985). This means that $\omega_\mu \tau \ll 1$. Taking into account the experimental values just mentioned, the fact that $\Phi_{\alpha\beta}(\tau) = \Phi_{\beta\alpha}(-\tau)$ when $\alpha = \{x, y, z\}$, and equations (2) and (3), it is a good approximation to write $\psi_z(t) = \lambda_z t$ with

$$\lambda_z = \frac{\gamma_\mu^2}{2} \int_{-\infty}^{\infty} d\tau \text{Re}(\Phi_{+-}(\tau)) \quad (4)$$

or

$$\lambda_z = \frac{\gamma_\mu^2}{2} \int_{-\infty}^{\infty} d\tau (\Phi_{xx}(\tau) + \Phi_{yy}(\tau)). \quad (5)$$

$\text{Re}(A)$ stands for the real part of A . As expected, equation (5) indicates that the depolarization is produced by the fluctuations of the transverse components of the local magnetic field at the muon site. Within the approximations mentioned above (basically the typical rare-earth fluctuation time must be sufficiently small) we deduce that the longitudinal depolarization function is an exponential function with a damping rate λ_z . If we identify λ_z with $1/T_1$, where T_1 is the muon spin-lattice relaxation time, the above μSR expressions (equations (4) and (5)) are equivalent to the nuclear magnetic resonance (NMR) formula given by Moriya (1962). We notice that it is possible to give an expression for the transverse depolarization function in terms of correlation functions $\Phi_{\alpha\beta}(\tau)$ as we have just done for the longitudinal function. This expression is the same as that found for NMR only if the mean value of the local magnetic field at the muon site is sufficiently large (Dalmas de Réotier 1990, Dalmas de Réotier and Yaouanc 1991).

In order to compute λ_z we need to give an expression for the magnetic field at the muon site and to specify the magnetic part of the Hamiltonian of the magnet.

The muon spin interacts with the polarized (in this paper we consider the case of an intermetallic ferromagnet with $T < T_C$) conduction electrons through the Fermi contact interaction and with the total angular momentum of each rare-earth ion, J_i ($J_i = S_i + L_i$ where S_i and L_i are respectively the spin and the orbital momentum vectors of rare-earth ion i), through the classical dipolar interaction. In a good approximation the first interaction can be expressed in terms of an effective isotropic and short-range interaction between the muon spin and J_i for each ion. Therefore we write

$$\delta B_+ = \sum_i [(C_i + F_i) \delta J_{i,+} + B_i \exp(2i\varphi_i) \delta J_{i,-} + A_i \exp(i\varphi_i) \delta J_{i,z}]. \quad (6)$$

where the geometrical factors are $A_i = 3D_i \cos \theta_i \sin \theta_i$, $B_i = \frac{3}{2}D_i \sin^2 \theta_i$ and $C_i = D_i(1 - 3 \cos^2 \theta_i)/2$ with $D_i = (\mu_0/4\pi)(\mu_{B}g_i/r_i^3)$. r_i , θ_i and φ_i are the distance and polar and azimuthal angles for ion i relative to the muon. The z axis is directed, as already mentioned, along the mean value of the magnetic field at the muon site. g_i is the Landé factor of ion i . The constant F_i is the contact magnetic field at $T = 0$ K produced by the conduction electrons polarized by J_i . The sum is over the N rare-earth ions of the lattice. We have $\delta B_- = (\delta B_+)^*$.

Unlike in NMR, for which the contact field is usually much larger than the dipolar field due to the localized electronic dipole moments, in μ SR spectroscopy these two magnetic fields are of about the same order of magnitude. The dipolar field even seems to be a little stronger than the contact field (Hartmann *et al* 1986). In any case, it is not negligible. This has an important consequence for the muon spin relaxation mechanism (first mentioned by Dalmas de Réotier 1990). This can be understood qualitatively as follows. The muon spin can be depolarized if the magnet in which the muon is implanted is able to flip its spin. The fluctuating part of the coupling Hamiltonian between the muon spin and the electronic spins can be written

$$\delta \mathcal{H}_1 = -\frac{1}{2}\gamma_\mu \hbar [\sigma_z \delta B_z + \frac{1}{2}(\sigma_+ \delta B_- + \sigma_- \delta B_+)] \quad (7)$$

where σ_α is the α -projection of the Pauli operator of the muon. Only the last two terms of this Hamiltonian can induce a muon spin flip. δB_+ (δB_-) is a linear combination of $\delta J_{i,+}$, $\delta J_{i,-}$ and $\delta J_{i,z}$ for each rare-earth ion i (see (6)). We now consider the first two types of terms of this combination. Since a relaxing muon spin flips an electronic spin (a magnon is created or annihilated), the z component of spin angular momentum of the ensemble muon system is conserved. However, as already observed for NMR (Mitchell 1957, Beeman and Pincus 1968), fulfilment of the energy-conservation requirement is more difficult to ensure. The energy of the created (or annihilated) magnon must be equal to $\hbar\omega_\mu$. The minimum energy of a magnon is equal to the gap energy of the magnon dispersion curve which is usually much larger than $\hbar\omega_\mu$ ($2 \mu\text{eV}$ if $\nu_\mu = 500$ MHz). Thus the first two types of terms of the linear combination, involving a single magnon, cannot in general induce a muon spin flip. On the other hand, a two-magnon, or Raman, process can be quite effective as regards inducing the muon spin to flip. As noted previously for NMR (Mitchell 1957, Beeman and Pincus 1968), this two-step process does not present problems as regards energy conservation because the only requirement is that the energies of the annihilated and created magnons must be equal (we neglect $\hbar\omega_\mu$). In addition there is no problem with the conservation of the z component of the angular momentum because this conservation is not required. In summary, for the computation

of λ_z (in lowest order) we have only to use the following part of the fluctuating magnetic field at the muon site:

$$\delta B'_+ = 3\mu_B \left(\frac{\mu_0}{4\pi}\right) \sum_i \frac{g_i \cos \theta_i \sin \theta_i \exp(i\varphi_i)}{r_i^3} \delta J_{i,z}. \quad (8)$$

Notice that $\delta B'_+$ involves only the classical dipolar interaction. The contact term, which we suppose to be isotropic, does not participate. This explains why in NMR the nuclear relaxation due to a magnon Raman process is usually rarely of significance (Beeman and Pincus (1968) and references therein). In order to compute λ_z we now specify the Hamiltonian of the magnet.

We suppose that the magnetic properties of a rare-earth intermetallic compound can be described by the sum of a Heisenberg interaction between the total angular momentum of the rare-earth ions and a Zeeman term that roughly simulates the magnetic anisotropy. For simplicity we suppose we are dealing with a Bravais lattice. Thus the Landé factor is the same for all the rare-earth ions, $g_i = g$. Therefore we write

$$\mathcal{H}_m = - \sum_{i,j,i \neq j} E_{ij} J_i \cdot J_j - g\mu_B B_a \sum_i J_{i,z} \quad (9)$$

where E_{ij} is the exchange integral between ions i and j , and B_a is the magnetic field that simulates the magnetic anisotropy. Following Holstein and Primakoff (1940) we express $\delta J_{i,z}$ in terms of boson operators a_k^+ and a_q^- (they obey the Bose commutation rule $[a_k^-, a_q^+] = N \delta_{k,q}$):

$$\delta J_{i,z} = - \frac{1}{N^2} \sum_{k,q} \exp(i(k-q) \cdot (r_i - r_0)) a_k^+ a_q^-. \quad (10)$$

r_i (r_0) is the vector that links the muon localization site to point i (the origin) of the Bravais lattice. The sum over k (q) is over the first Brillouin zone. With the Holstein-Primakoff transformation, \mathcal{H}_m can be directly diagonalized if we only retain the term bilinear in the boson operators. The result is

$$\mathcal{H}_m = \frac{1}{N} \sum_k \hbar \omega_k a_k^+ a_k^- \quad \hbar \omega_k = 2JN(E_{k=0} - E_k) + g\mu_B B_a. \quad (11)$$

We have made the definition

$$E_k = \frac{1}{N^2} \sum_{i,j} E_{ij} \exp(-ik \cdot (r_i - r_j)).$$

In (11) we have dropped constant terms irrelevant for our purposes. Notice that in the small- k approximation (long-wave approximation) we can write for cubic lattices $\omega_k = Dk^2 + g\mu_B B_a/\hbar$ where D is the spin-wave stiffness constant. For a simple cubic lattice and with only nearest-neighbour exchange interactions we have $D = 2JEa^2/\hbar$ where a is the lattice constant. The stiffness constant can still be roughly estimated from this formula for any crystal structure if a is taken as the distance between the rare-earth ions. As expected $g\mu_B B_a$ is the value of the gap of the magnon dispersion curve. Using (8), (10) and (11) we derive

$$\begin{aligned} \delta B'_+(\tau) = & -3g\mu_B \left(\frac{\mu_0}{4\pi}\right) \frac{1}{N^2} \sum_{k,q} \sum_i \frac{\cos \theta_i \sin \theta_i \exp(i\varphi_i)}{r_i^3} \exp(i(k-q) \cdot r_i) \\ & \times \exp(i\tau(\omega_k - \omega_q)) a_k^+ a_q^-. \end{aligned} \quad (12)$$

Having the $\delta B'_+(\tau)$ and $\delta B'_-$ expressions ($\delta B'_- = (\delta B'_+(\tau = 0))^*$), it is now possible to compute the longitudinal damping rate (in lowest order) using (4). We deduce that

$$\lambda_z = 9\pi(\gamma_\mu g\mu_B)^2 (\mu_0/4\pi)^2 P$$

with

$$P = \sum_{i,j} \frac{\cos \theta_i \cos \theta_j \sin \theta_i \sin \theta_j \cos(\varphi_i - \varphi_j)}{r_i^3 r_j^3} \frac{1}{N^2} \sum_{k,q} \delta(\omega_k - \omega_q) n_k (n_q + 1) \times \cos(k - q) \cdot (r_i - r_j). \tag{13}$$

To proceed further we neglect the (geometrical) cosine term in the sum over k and q . This approximation should not be unreasonable because, as mentioned below, the sum is dominated by the centre of the Brillouin zone. As the sums over the direct lattice and Brillouin zone are no longer related we obtain the simplified result

$$\lambda_z = 9\pi(\gamma_\mu g\mu_B)^2 \left(\frac{\mu_0}{4\pi}\right)^2 \frac{G}{v^2} \frac{1}{N^2} \sum_{k,q} \delta(\omega_k - \omega_q) n_k (n_q + 1). \tag{14}$$

v is the volume of the unit cell. The geometrical factor G which depends on the lattice structure and the muon localization site is given by

$$G = \sum_{i,j} \frac{v^2}{r_i^3 r_j^3} (\cos \theta_i \cos \theta_j \sin \theta_i \sin \theta_j \cos(\varphi_i - \varphi_j)). \tag{15}$$

Notice that this factor can be zero due to symmetry. For example, this is the case if the rare-earth ions are on a simple cubic lattice, the z axis being along the $[1, 0, 0]$ direction and the muon located at the site $(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$ (the sum is limited to nearest neighbours). A study of the G -factor indicates that the double sum may not be convergent when r_i and r_j are very different. This may be related to the neglect of the cosine term in the (k, q) sum. A detailed mathematical study is under way. An approximate analytical formula can be derived for the sum over k and q of (14). Replacing the sums by integrals we obtain

$$\lambda_z = 9\pi(\gamma_\mu g\mu_B)^2 \left(\frac{\mu_0}{4\pi}\right)^2 \frac{G}{v^2} \frac{1}{N^2} \frac{V^2}{(2\pi)^6} (4\pi)^2 \int_0^{k_{\max}} \int_0^{k_{\max}} k^2 dk q^2 dq \times \frac{\exp(\hbar\omega_k/k_B T)}{(\exp(\hbar\omega_k/k_B T) - 1)^2} \delta(\omega_k - \omega_q). \tag{16}$$

V , the volume of the sample, is equal to Nv . In (16) we have neglected any anisotropy in the Brillouin zone. The integrand is small for large k . Therefore we may use the small- k approximation for the magnon dispersion relation and replace the upper limit of the integrals by infinity. Then (16) becomes

$$\lambda_z = \frac{9}{16\pi^3} (\gamma_\mu g\mu_B)^2 \left(\frac{\mu_0}{4\pi}\right)^2 G \frac{(k_B T)^2}{\hbar^2 D^3} \int_{x_0}^\infty dx (x - x_0) \frac{\exp(x)}{(\exp(x) - 1)^2} \tag{17}$$

with $x_0 = g\mu_B B_a/k_B T$.

In the limit $g\mu_B B_a \ll k_B T$ the integration can be performed analytically. We then obtain

$$\lambda_z = \frac{9}{16\pi^3} (\gamma_\mu g\mu_B)^2 \left(\frac{\mu_0}{4\pi}\right)^2 G \frac{(k_B T)^2}{\hbar^2 D^3} \ln\left(\frac{k_B T}{g\mu_B B_a}\right). \quad (18)$$

The $T^2 \ln(T)$ behaviour has already been predicted for an anisotropic contact interaction, which can happen in NMR (Mitchell 1957, Beeman and Pincus 1968). The pre-factor is new. The derivation of (18) supposes that the contact interaction is isotropic. Following the method proposed here, it is easy to obtain a general expression for λ_z that includes the effect of an anisotropic contact interaction.

We are going to discuss the possibility of understanding the GdNi₅ data in terms of a Raman scattering. Equation (18) has been derived on the hypothesis that k -space is isotropic. Although this is not the case of GdNi₅, the conclusion of the present analysis should not be influenced by this simplification. Equation (18) depends on three parameters: the magnon stiffness constant, the anisotropy energy $g\mu_B B_a$ and the geometrical factor G . GdNi₅ has a hexagonal crystal structure isotypical with that of CaCu₅. The Gd³⁺ ions are located on a hexagonal Bravais lattice. Each Gd³⁺ ion has two nearest-neighbour Gd³⁺ ions at 3.97 Å and six second-nearest-neighbour Gd³⁺ ions at 4.90 Å. Thus, for simplicity, we suppose that each ion has eight nearest-neighbour ions at the average distance $a = 4.67$ Å. We estimate the exchange integral from the molecular field formula

$$E = [3/J(J + 1)] k_B T_C / 2z. \quad (19)$$

z is the number of nearest-neighbour ions ($z = 8$). From this formula we deduce $E = 0.374$ K. This leads to a stiffness constant $D = 7.48 \times 10^{-8} \text{ s}^{-1} \text{ m}^2$. As Gd³⁺ has an isotropic 4f shell, the magnetic anisotropy is small. This can be due to the dipolar interaction between the electronic magnetic moments. In any case the effective anisotropic magnetic field should be small. If we take $B_a = 0.1$ T we have $\ln(k_B T / g\mu_B B_a) = 4.57$ and 5.22 at 13 K and 25 K respectively. Therefore the temperature dependence introduced by the logarithmic factor is weak. As the GdNi₅ data are not precise enough to allow us to detect such a small effect, we will take for this factor the average of its values at 13 K and 25 K. Thus we set $\ln(k_B T / g\mu_B B_a) = 4.89$. Notice that B_a can be slightly changed without having a strong effect on the value of the logarithmic factor. The G -factor can be computed from a lattice sum using (15) if the muon localization site and the direction of the mean value of the magnetic field at that site are known. As the localization site is unknown for GdNi₅ (Dalmas de Réotier *et al* 1990) we have computed G for the two most probable sites (the hydrogen sites; site 1 and 4 of table 1 of Dalmas de Réotier *et al* 1990). We have taken the z direction parallel to the mean magnetic field produced by the Gd³⁺ dipole moments. Thus, for simplicity, we have neglected the contribution of the contact interaction to the mean value of the magnetic field at the muon site. For site 1 we find $G = 12.4$, whereas for site 4 the G -value is about 1 (the lattice sum does not seem to converge smoothly). These values should be considered as preliminary because, for example, we do not even know the direction of the magnetic field at the muon site. In addition, the lattice sum of (15) seems to have convergence problems. For an exact evaluation, equation (13) has to be used. To proceed further we take for G the value computed for site 1, $G = 12.4$. Using the parameters just estimated we can compute λ_z . We obtain $\lambda_z = \eta T^2$ with $\eta = 1.13 \times 10^{-4} \text{ MHz K}^{-2}$ whereas the experiment gives $\eta = 0.191(4) \times 10^{-3} \text{ MHz K}^{-2}$. The theoretical estimate is too small by a factor of about 1.69. Considering the facts that the exchange integral has been estimated from the simple molecular field formula and that the value given for the

geometrical factor is preliminary, we estimate that the present theory describes the experimental result. Although more work is needed, it is clear, at any rate, that Raman scattering can explain the GdNi_5 data at low temperature.

Before attempting a more detailed comparison between experiment and theory, more work is needed. The muon localization site in GdNi_5 should be determined as well as the direction of the mean value of the magnetic field. Experiments should be performed on other intermetallic ferromagnets. The theory should be completed by introducing the effect of the dipolar interaction between the rare-earth ions and taking the virtual magnon processes into account (Beeman and Pincus 1968). The sums in (13) should be studied in detail. In addition, as the compounds of interest are metals, the effect of the conduction electrons on the magnon stiffness constant should be investigated.

In this letter we have considered a ferromagnet and shown that the muon spin relaxation by a Raman process is of importance except when the degree of symmetry at the muon site is too high. The case of antiferromagnetic compounds should be studied. Preliminary data on the antiferromagnets GdCu_2 (Gubbens *et al* 1991) and NdRh_2Si_2 (Dalmas de Réotier 1990) indicate that it is possible in general to measure at low temperature the longitudinal muon relaxation rate. Notice that when the intermetallic contains ions such as uranium, which can present an itinerant magnetic character, the temperature dependence of λ_z cannot be explained within the framework given here (Dalmas de Réotier *et al* 1991a).

After this letter was written we received a preprint from Lovesey *et al* (1991) where Raman-type relaxation processes are considered for EuO . These authors take the dipolar interaction between the rare-earth ions explicitly into account. As expected, λ_z is predicted to have an almost quadratic temperature dependence. To the best of our knowledge, no μSR data exist for EuO .

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