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Muon spin relaxation in ferromagnets: I. Spin-wave fluctuations

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Abstract. Expressions for the dipolar and hyperfine contributions to the relaxation rate of muons implanted in a ferromagnet are presented, and analysed using the Heisenberg model of spin waves including dipolar and Zeeman energies. Calculations for EuO indicate that the temperature dependence of the hyperfine and dipolar contributions to the relaxation rate are similar, so the latter contribution will dominate if the ratio of the hyperfine and dipolar coupling constants is indeed very small. The hyperfine mechanism is sensitive to the dipolar energy of the atomic spins, whereas the dipolar mechanism depends essentially on the exchange energy. For both mechanisms there is an almost quadratic dependence on temperature, throughout much of the ordered magnetic phase, which reflects two-spin-wave difference events from the Raman-type relaxation processes.

1. Introduction

Recent neutron scattering experiments on ordered ferromagnets have rekindled interest in the static and dynamic properties of longitudinal spin fluctuations, i.e. components parallel to the easy axis. In the experiments, these are cleanly separated from transverse fluctuations by the use of polarization analysis. Data for insulating and metallic ferromagnets, just below the critical temperature (T_c), collected for small wavevectors ($k \sim 0.1$ Å^{-1}) reveal a quasi-elastic response and no inelastic scattering at the spin-wave energy, even though spin-wave resonances are clearly defined in the transverse fluctuations (Mitchell *et al* 1984, Mitchell *et al* unpublished, Böni *et al* 1991). These findings contrast with conclusions drawn from earlier unpolarized neutron scattering data on Fe, Ni and EuO, namely that, unlike antiferromagnets, the longitudinal response $S(\boldsymbol{k}, \omega)$ is predominantly inelastic, i.e. a minimum at $\omega = 0$.

The dependence of the longitudinal spin response on the magnitude of an external field (required in the experiments to produce a unique easy axis and to prevent severe depolarization) is of interest in view of well established results for the longitudinal susceptibility $\chi(\boldsymbol{k})$, related to the integral of $S(\boldsymbol{k}, \omega)/\omega$, to the effect that it diverges in the limit of a vanishing field or wavevector. Spin-wave theory of a Heisenberg ferromagnet, for example, predicts $\chi(0) \propto h^{-1/2}$ when the field $h \rightarrow 0$, and $\chi(k) \propto k^{-1}$ for $h = 0$ and $k \rightarrow 0$. Böni *et al* (1990) find for Ni at $T = 0.987T_c$ that the longitudinal intensity decreases moderately on increasing the field by a factor of 3. Nor have previous experiments unambiguously detected divergent behaviour in $\chi(k)$. Kötzler and Muschke (1986) report an indication of $\chi(0) \propto h^{-1/3}$ behaviour in an analysis of bulk data for EuS. Neutron diffraction experiments on EuO (Passell *et al* 1976) and a disordered alloy palladium/10% iron (Mitchell *et al* 1984) also reveal non-divergent behaviour of the susceptibility. The expression for $S(\boldsymbol{k}, \omega)$ derived from spin-wave theory (Lovesey and Trohidou 1991) possesses a field-limited enhancement at the spin-wave dispersion frequency, and appears to be at odds with the recent polarized neutron scattering data just described. Moreover, it can be shown that dipolar forces do not annul the $(1/k)$ behaviour of $\chi(\boldsymbol{k})$ for small k, but $\chi(0)$ is finite and depends on the shape of the sample.

Although Vaks *et al* (1968) argue that spin-wave results for $\chi(k)$ and $S(\boldsymbol{k}, \omega)$ evaluated for small k and ω should be useful over a wide range of temperatures, perhaps these estimates are not reliable quite so close to T_c as in the existing neutron scattering experiments. It would be valuable to have data at lower temperatures, where spin-wave theory can be applied with confidence, and to identify the temperature at which the theory becomes inadequate.

To this end, the technique of muon spin relaxation (μSR) has some attractive features (Cox 1987). The relaxation of the muon signal is directly related to $S(\boldsymbol{k}, \omega)$ and experiments can be performed with zero or a very small Larmor precession frequency, $\omega_\mu \ll 1$ μeV. Here we present results for the relaxation rate λ calculated with spin-wave theory applied to a Heisenberg ferromagnet. The model includes dipolar forces and an external magnetic field.

The properties of muons implanted in an ordered ferromagnetic are reviewed in the next section, with a view to an experiment on EuO. Ferromagnetic spin-wave theory is sketched in section 3, and the presentation largely follows Keffer (1966) and Lovesey (1987). The muon relaxation rate is calculated in section 4. It is argued that the muon Larmor frequency can be set to zero in the theory, to a good approximation. In view of this, the relaxation rate is determined by so-called Raman spin-wave scattering events, and $\lambda \propto \Sigma\, S(\boldsymbol{k}, 0)$. An expression for λ is derived using linear spin-wave theory, and its behaviour as functions of the dipolar forces, magnetic field and temperature is established using analytical and numerical techniques. Conclusions are gathered in section 5.

2. Muon spin relaxation experiments

The majority of information on spin fluctuations in magnetic materials has been obtained by neutron beam experiments, which allow spatial as well as temporal characteristics of the fluctuations to be determined. Although even very subtle effects, such as the influence if dipolar interactions between the spins in the presence of much stronger exchange interactions, can be observed in neutron scattering, the information obtained is not always unambiguous, and additional information from other spectroscopies can be helpful. Here it is argued that spin relaxation experiments using positive muons as magnetic probes can provide complementary information, in particular on the effects on the longitudinal spin fluctuations in a magnetic material.

Muon spin relaxation (μSR) experiments have so far been carried out on a number of magnetic materials, notably metallic magnets (for a review see, for instance, Karlsson (1990)) but also for a limited number of insulating materials (Holzschuh *et al* 1981, Brewer *et al* 1981). The fluctuations of the atomic spins in the material are often obtained indirectly through their magnetic coupling with a probe spin, in the present case that of the μ^+ whereas in nuclear magnetic resonance (NMR) a ligand nucleus might be monitored. In the analysis of μSR data it is assumed that the implanted muon is a passive probe, and the signal is truly representative of bulk properties in the absence of the probe, i.e. μSR is regarded as a delicate and unobtrusive technique.

The relaxation of the muon spin is described by the same rate equations as in the theory of NMR (Slichter 1990). The longitudinal relaxation time T_1 is defined through the exponential decay on an initial magnetization of the probe spin $M_z(0)$ directed along the x axis,

$$M_z(t) = M_z(0)\exp(-t/T_1). \tag{2.1}$$

The microscopic mechanism behind this polarization decay is that fluctuation fields $B_x(t)$ and $B_y(t)$, perpendicular to the z direction, are acting on the probe spin and induce spin-flip transitions. The longitudinal relaxation rate λ for a muon has the following generic form:

$$\lambda \equiv (1/T_1) = \Gamma \int \mathrm{d}t\,\langle B_x(t)B_x + B_y(t)B_y\rangle \tag{2.2}$$

where the $\langle B_\alpha(t)B_\alpha(0)\rangle$ are the correlation functions for the local fields acting on the muon spins. The prefactor Γ depends on the actual form of the interaction between the probe and the atomic spins, and it will include hyperfine and dipolar interactions, both of which are discussed in the appendix. In the proposed μSR experiment the dipolar interaction dominates, and the appropriate Γ is derived. For an ordered magnet, relaxation is achieved with Raman processes that are generated by fluctuations in the atomic spins along the axis of quantization (easy axis). The final comment about (2.2) is that it is assumed that there is no net magnetic field acting on the muon, and hence there is no function in the integrand that oscillates at the Larmor frequency.

Similarly, one defines the transverse relaxation rate $\lambda_\perp$, which refers to an experiment in an applied magnetic field B_0 (along z) where the muon spins precess with a Larmor frequency ω_μ,

$$\lambda_\perp \equiv (1/T_2) = (\mu_N g_\mu/\hbar)^2 \int \mathrm{d}t\,[\langle B_z(t)B_z\rangle + \tfrac{1}{2}\cos(t\omega_\mu)\langle B_x(t)B_x + B_y(t)B_y\rangle]. \tag{2.3}$$

Here, T_2 is the transverse relaxation time, which is defined by the macroscopic equations $\mathrm{d}M_\alpha/\mathrm{d}t = M_\alpha/T_2$ ($\alpha = x, y$). A magnetization $M_x(0)$ initially oriented along the x axis is then precessing around the z axis with angular frequency ω_μ, losing its magnitude at the rate $\exp(-t/T_2)$ because of spin flips induced by the fluctuating fields $B_\alpha(t)$.

In μSR, an initial polarization of the muon spins is obtained automatically by the mechanism with which the muons are created. This polarization is along the direction of the beam coming from the accelerator. Thus, there is no need for a magnetic field to produce the initial polarization, as in NMR.

In a *longitudinal* set-up for μSR, detectors for measuring the depolarization are placed in backward and forward directions with respect to the sample where the muons are stopped, and the reference direction z is the direction of the muon beam. The relaxation rate λ can be measured in zero field or in a longitudinal field applied along the z axis.

In a *transverse* field μSR experiment, the magnetic field is applied perpendicular to the initial polarization direction (beam axis). The quantization axis is now along the applied field $\boldsymbol{B}_0$ and detectors placed in the xy plane measure the precession (and decay) of the initial polarization. Equation (2.3) refers to this geometry, which is the same as in an NMR experiment measuring the so-called free induction decay (Slichter 1990).

2.1. Longitudinal and transverse spin fluctuations

In the following we will concentrate on the interpretation of μSR experiments made in the longitudinal geometry, mainly because they allow observations in zero applied field. As observed by inspection of equation (2.2) the measurement of λ actually provides information on the local field fluctuations perpendicular to the chosen symmetry axis of this measuring geometry.

Spin dynamics of an ordered magnetic system is naturally described with an easy magnetization axis as reference direction. *Longitudinal fluctuations* of the atomic spins refer to this axis, whereas transverse fluctuations are perpendicular to it. In the following we will illustrate, for a particular magnetic crystal, how the longitudinal spin fluctuations are seen in a longitudinal μSR experiment for various orientations of a single-crystal sample with respect to the initial muon spin polarization axis (=beam axis).

It should be noted that conventional experiments in NMR and polarized neutron scattering need an external field for the observation of the relaxation phenomena (this is not true for spin-echo NMR, but such observations are less interesting in this context since the response of an echo experiment mainly comes from domain walls, which are magnetically perturbed regions). The muon spin relaxation technique, on the other hand, can be applied even in zero external field. This is of particular importance when studying dipolar effects on the spin dynamics since even a very weak external field may interfere with and overshadow the dipolar effects. This is, of course, even more important if the sample is not a single crystal so that there is a distribution of the ordering directions with respect to the easy axes over the different domains, or if the sample is not shaped so that local demagnetization effects are avoided when the field is applied.

As an illustrative example we will choose the magnetic crystal EuO, which is ferromagnetic below 69 K. The position of the μ^+ in the crystal lattice has to be known for an evaluation of the spin fluctuation rates of the surrounding ions. Being positively charged the μ^+ has its lowest potential energy in an interstitial site of the unit cell. The actual crystallographic position can usually be identified by measuring the local static field created by the surrounding dipoles in the ordered state if an additional external field is applied along certain main axes of a single-crystal sample.

We will assume (no experiment has been carried out so far) that the μ^+ occupies the centre of the unit cell surrounded by four Eu^{2+} ions at the cube corners as illustrated in figure 1. For a completely ordered Eu^{2+} spin system a dipole sum carried out over the whole lattice would produce a zero magnetic dipole field at the muon position because of the high symmetry. A non-zero local field at the μ^+ can still exist through the action of the spin density of electrons at the same position, which produces a contact field along the magnetization direction,

$$B_c = -\tfrac{8}{3}\mu_B\eta[n_\downarrow(r) - n_\uparrow(r)] \tag{2.4}$$

where $n_\downarrow(r)$ and $n_\uparrow(r)$ are the interstitial electron densities for spin up and spin down, respectively (in the absence of the muon), and η is a spin-density enhancement factor caused by the muonic charge itself. We anticipate that B_c is small in an insulator unless a muonium-like (muon–electron bound) state is formed.

Below the transition temperature in zero applied field, the Eu spins are lined up along an easy magnetization axis z with an average value determined by the spontaneous magnetization curve. No orbital contribution needs to be considered for Eu^{2+}, which is well represented by a ^8S ionic term. It is important to observe that, even if the time average of the dipolar field produced by the surrounding dipoles may be zero due to the

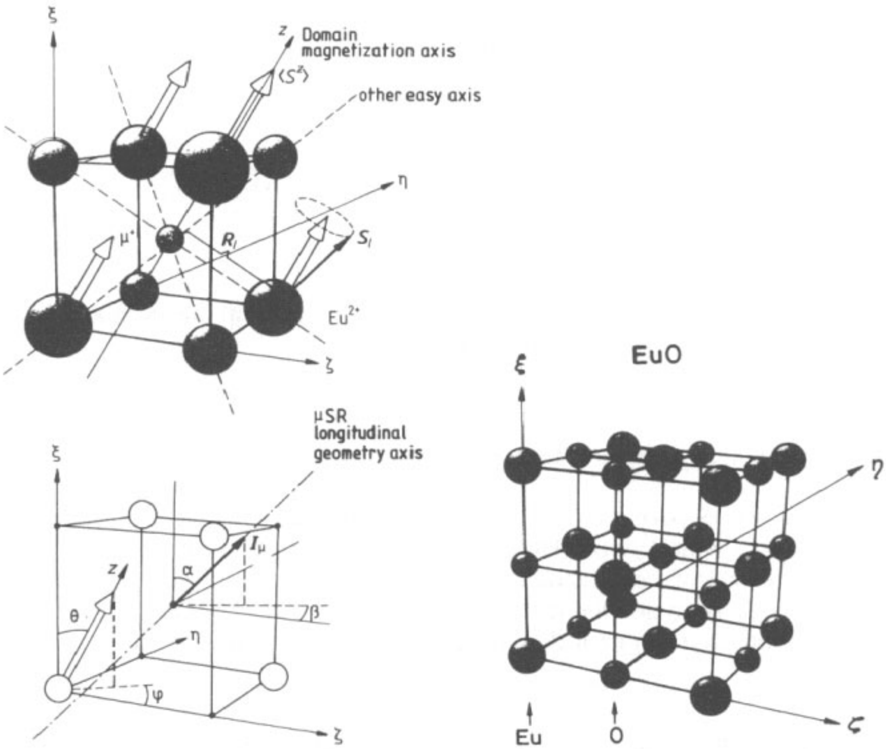

Figure 1. The notation for the atomic and muon spin orientations relative to the crystal axes is illustrated for EuO, together with the expected geometry of the implanted muon.

symmetry, the instantaneous local fields are not, unless the sample is at zero temperature and the dipolar interaction between atomic moments is neglected.

Now consider the geometrical arrangement of figure 1. The easy magnetization directions of the Eu crystal are (1, 1, 1), which happen to coincide with the atomic vectors if the μ^+ sits in the assumed position. Of course, only one of the directions (1, 1, 1) is the true preferential direction in each domain in the spontaneously ordered state. Let us put the quantization axis along this direction and express the sum of the dipolar fields from the four Eu dipoles,

$$\boldsymbol{B} = (g\mu_{\mathrm{B}}/d^3) \sum_l \{\boldsymbol{S}(l) - (3/d^2)\boldsymbol{l}[\boldsymbol{l} \cdot \boldsymbol{S}(l)]\} \qquad (2.5)$$

in which d is the distance between the muon and atomic spins in the first coordination shell.

When each Eu spin component is allowed to fluctuate, it produces a field fluctuation $\langle B(t)B(0)\rangle$, which can be expressed in terms of the Eu spin correlation functions $\langle S(t)S(0)\rangle$. These local field fluctuations in turn give rise to the relaxation of the muon spin according to equation (2.2). Recall that the direction of the initial polarization of the muon spin system can be chosen at will simply by placing the single-crystal sample at the desired angle with respect to the beam of incoming muons. Details of the geometric factors in Γ are provided in the appendix. In the calculation described there, the muon

Table 1. Values of spin-wave stiffness D and wavevector ζ[a].

	Fe	Ni	EuO	EuS
D (meV Å^2)	280	400	11.65	2.56
ζ (Å^{-1})	0.02	0.01	0.11	0.18

[a] After Keffer (1966) and Passell *et al* (1976).

is assumed to be equidistant from the atomic moments, but the method used lends itself to calculations for a lower spatial symmetry.

3. Ferromagnetic spin-wave theory

Spin operators $\{S_l\}$ of magnitude S are assigned to sites defined by vectors $\{l\}$ on a Bravais crystal lattice with N unit cells. A ferromagnetic state is achieved, at temperatures $T < T_c$, by an isotropic exchange interaction. The spatial Fourier transform of the exchange interactions is denoted by $J(k)$, and $J(k) = J(-k)$ because the lattice is Bravais. The exchange and magnetic field interactions lead to a spin-wave dispersion,

$$\varepsilon_k = h + 2S[J(0) - J(k)] = h + Dk^2 \qquad ak \ll 1. \tag{3.1}$$

Here, the Zeeman energy $h = g\mu_B H$, where g is the electronic gyromagnetic factor and H is the external field strength, and the second equality, valid for long wavelengths, defines the spin-wave stiffness D. Values of D for EuO, EuS, Fe and Ni are recorded in table 1.

With the addition of dipolar forces the spin-wave dispersion ω_k satisfies (Lovesey 1987)

$$\omega_k^2 = A_k^2 - |B_k|^2 \tag{3.2}$$

in which

$$A_k = \varepsilon_k + |B_k| = A_{-k} = A_k^* \tag{3.3}$$

and

$$B_k = B_{-k} \neq B_k^* \tag{3.4}$$

is the Fourier transform of part of the dipolar force field. For all but the extreme value $k = 0$, it has been shown that (Keffer 1966, Passell *et al* 1976)

$$B_k = D\zeta^2 \sin^2\theta_k \exp(-2\mathrm{i}\varphi_k) \tag{3.5}$$

where θ_k and φ_k are the angles in polar coordinates for k relative to the easy (z) axis. The strength of the dipolar force is expressed by the wavevector ζ, listed in table 1, defined through

$$D\zeta^2 = 2\pi g\mu_B M_0 \tag{3.6}$$

in which M_0 is the saturation magnetization.

Fluctuations in S^z, in linear spin-wave theory, are created by the operator product S^-S^+, where $S^\pm$ are spin raising and lowering operators, and

$$\Delta S_k^z = (-1/2SN) \sum_q S_{k+q}^- S_q^+. \tag{3.7}$$

It is usual to express $S_k^\mp$ in terms of Bose operators a_k and a_k^+, which reduce the Hamiltonian to a quadratic form,

$$S_k^+ = u_k a_k + v_k a_{-k}^+. \tag{3.8}$$

The coefficients in (3.8) are taken to be

$$u_k^2 = (2SN)(A_k + \omega_k)/2\omega_k \tag{3.9}$$

and

$$v_k = -u_k B_k^* /(A_k + \omega_k) \tag{3.10}$$

but this prescription is not unique. Finally, we need to note

$$a_k(t) = a_k(0) \exp(-it\omega_k) \tag{3.11}$$

which follows because the Hamiltonian in terms of a and a^+ has the harmonic oscillator form.

4. Muon relaxation rate

Relaxation processes that involve single spin-wave events, often called direct processes, are neglected in our calculation of the muon damping rate λ. The justification for this stems in part from the fact that they contribute only when the muon Larmor frequency ω_μ matches the energy of a spin wave. Hence, there are no direct processes in λ if ω_μ is less than the minimum spin-wave energy. In practice, ω_μ is likely to be very small on the scale of electronic spin-wave energies, since to begin with $\omega_\mu = 0.57\ \mu\text{eV T}^{-1}$. The field experienced by a muon implanted in a magnetic material is essentially the sum of the dipolar and hyperfine fields generated by electronic magnetic moments, and the external field, corrected by demagnetizing effects. Using the Lorentz technique, the dipolar field can be calculated for a given magnetic configuration (Kaplan *et al* 1973). It vanishes for some geometries, e.g. an interstitial site in a FCC magnet (Ni, EuO), while severe cancellation effects and muon diffusion contribute to the reduction of the dipolar field in most other cases. Finally, direct processes are very weak except at low temperatures. In consequence, the lowest-order relaxation process is assumed to come from two-spin-wave events, or Raman processes, generated by fluctuations in S^z.

There is a conceptual advantage in expressing λ in terms of the longitudinal spin response function $S(k, \omega)$ mentioned in the introduction, and directly related to the cross section for magnetic neutron scattering. Expressions for the dipolar and hyperfine contributions to λ are derived in the appendix in what amounts to a straightforward application of Fermi's golden rule for transition rates (Moriya 1956, Van Kranendonk and Bloom 1956).

With the definition (l, l' run over all N magnetic cells in the sample)

$$S(k, \omega) = \frac{1}{N}\sum_{l,l'}\int_{-\infty}^{\infty}\frac{dt}{2\pi\hbar}\,e^{-i\omega t}\,e^{ik\cdot(l-l')}\langle S^z(l,t)S^z(l',0)\rangle \tag{4.1}$$

we obtain from (A10) and (A17)

$$\lambda_d = \left(\frac{2\pi\Gamma\hbar}{N}\right)\sum_k S(k,0)[1-\cos(k\cdot\delta)] \tag{4.2}$$

and

$$\lambda_h = \left(\frac{\pi\Gamma_0\hbar}{3N}\right)\sum_k S(k,0)[1+3\cos(k\cdot\delta)]. \tag{4.3}$$

These two expressions apply to an FCC material in which the muon is assumed to occupy an interstitial site, e.g. EuO for which $\delta = (a/2)(1,1,0)$. The coupling constants have the dimension of $(\text{time})^{-2}$, and for EuO $\Gamma = 0.055\times 10^6\ \mu s^{-2}$ and while $\Gamma_0 = 8(A/\hbar)^2$ is not known for this magnetic salt, it is expected that $\Gamma \gg \Gamma_0$.

Equations (4.2) *and* (4.3) *are properly viewed as relations between bulk* ($\lambda = \lambda_d + \lambda_h$) and differential $\{S(k,\omega)\}$ response functions. From this point of view, λ is the ratio of a transport coefficient, which describes the coupling mechanism, and magnetic specific heat. Far away from the critical region, the temperature dependence of λ comes from the specific heat. By and large, bulk response functions provide a very limited amount of information. However, as in the case with μSR, they may provide information that is not obtainable directly by other experimental techniques.

The spin-wave approximation for $S(k,\omega)$ contains two spin-wave annihilation and creation events, induced by dipolar forces, and difference events (Lovesey and Trohidou 1991). For $\omega = 0$ only the latter survive because they can conserve the total energy, whereas simultaneous annihilation, or creation, of two spin waves does not. A compact expression for λ is obtained with the aid of a structure factor (Lovesey and Trohidou 1991).

$$F(k,q) = \{A_kA_q + \omega_k\omega_q + |B_k||B_q|\cos[2(\varphi_k - \varphi_q)]\}/2\omega_k\omega_q. \tag{4.4}$$

Observe that $F \geqslant 0$, and $F \to 1$ in the absence of dipolar forces.

From the result (3.7*b*) in Lovesey and Trohidou (1991), we obtain the results,

$$\lambda_d = \left(\frac{2\pi\Gamma}{N^2}\right)\sum_{k,q} n_q(1+n_q)F(k,q)\delta(\omega_k - \omega_q)\{1-\cos[\delta\cdot(k-q)]\} \tag{4.5}$$

and

$$\lambda_h = \left(\frac{\pi\Gamma_0}{3N^2}\right)\sum_{k,q} n_q(1+n_q)F(k,q)\delta(\omega_k - \omega_q)\{1+3\cos[\delta\cdot(k-q)]\} \tag{4.6}$$

where n_q is the standard Bose occupation factor ($k_B = 1$),

$$n_q = [\exp(\omega_q/T) - 1]^{-1}. \tag{4.7}$$

The geometric factors in these expressions play an important role in determining the properties of the relaxation rates. For simple ferromagnetic spin waves λ_h is not defined because the integration in (4.6) is divergent. This behaviour stems from properties of the integrand at the centre of the Brillouin zone. Finite results are obtained with the

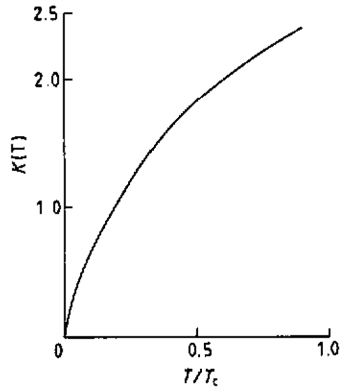

Figure 2. The integral $K(T)$ in equation (4.9) for the dipolar relaxation rate is displayed as a function of T/T_c. Parameters employed are appropriate for EuO.

inclusion of either a magnetic field or dipolar forces between atoms, as shown later. Since dipolar forces exist in all magnetic materials to some degree, these, or some other anisotropic interaction, are always responsible for the value of λ_h. In view of the fact that the integral in (4.6) is sensitive to behaviour of the integrand near the zero centre, it is at once evident on looking at (4.5) that λ_d can have quite different properties. Indeed, as we now show, it has a finite value for simple ferromagnetic spin waves, i.e. anisotropic interactions do not play a crucial role.

4.1. Dipolar mechanism

We will evaluate the integral in (4.5) using simple ferromagnetic spin waves, and then it is appropriate to set $F(\boldsymbol{k}, \boldsymbol{q}) = 1$. From the material gathered in section 3 we obtain from (4.5) the expression

$$\lambda_d = \left(\frac{v_0 \Gamma \hbar}{4\pi^3 D}\right) \int_0^\infty q^3 \, dq \, n_q(1 + n_q)\left[1 - \left(\frac{\sin(\delta q)}{\delta q}\right)^2\right] \tag{4.8}$$

in which $v_0 = a^3/4$ (FCC) is the unit-cell volume, the Bose factors are evaluated with (4.7) and $\omega_q = Dq^2$, and $\delta^2 = a^2/2$. In the limit of low temperatures the integral in (4.8) approaches the value

$$(\delta^2 \pi^2 T^3 / 18 D^3).$$

When seeking the general dependence on temperature it is convenient to write (4.8) in the form that factors out the trivial T^2 dependence generated by the quasi-classical approximation for the Bose occupation factors,

$$\lambda_d = 3.91 \times 10^{-3} (\mu s)^{-1} (T/T_c)^2 K(T). \tag{4.9}$$

The coefficient in (4.9) applies to EuO ($a = 5.14$ Å and $T_c = 69.5$ K), assuming that the muon is at an interstitial site; and the integral $K(T)$ is displayed in figure 2 and it initially increases linearly with temperature.

Our estimate of λ_d for EuO at $T = T_c/2$ is

$$\lambda_d = 0.002 \, (\mu s)^{-1}.$$

At higher temperatures the actual spin-wave stiffness departs significantly from the value quoted in table 1 and used in obtaining this estimate. From Passell *et al* (1976) we obtain

$$D(T)/D(0) = 0.64 \qquad T = 0.8T_c.$$

The reduction of $D(T)$ with temperature, due to non-linear spin-wave events, has a significant effect on our estimates, largely because the coefficient in (4.9) is proportional to $(1/D)^3$. For example, using the value $D(0)$ and $T = 0.8T_c$ gives

$$\lambda_d = 0.006\,(\mu s)^{-1}$$

whereas with the value of D determined by experiment ($K = 2.71$ is 18% larger than for $D(0)$),

$$\lambda_d = 0.026\,(\mu s)^{-1}$$

which is well within the range of values that can be measured. Application of a magnetic field will depress λ_d.

The use of $D(T)$ in our formulae is not strictly a consistent method of accounting for non-linear spin-wave events. A theory of $S(\boldsymbol{k}, \omega)$ that accounts for these events reveals not only the well known renormalization of $D(T)$ but also an interaction that generates an integral equation for the response function. This implies that the relaxation of μSR signals has the potential to probe non-linear spin-wave events beyond what is presently established.

We conclude by remarking that spin waves with a dispersion $\omega_k = h + Dk^2$ lead to an $S(\boldsymbol{k}, \omega)$ that is identical to the density autocorrelation response of an ideal Bose fluid in which the chemical potential is $-h$. The expression to be used in (4.2) is

$$S(k, 0) = (Tv_0/16\pi^2D^2k)\{\exp[(4h + Dk^2)/4T] - 1\}^{-1}$$

and the result for λ_d with $h = 0$ can be shown to be the same as (4.8).

4.2. Hyperfine mechanism

When the energy-conserving delta function in (4.6) demands $\boldsymbol{k} = \boldsymbol{q}$, which is the case for small wavevectors, the expression reduces to

$$\lambda_h = \left(\frac{4\pi\Gamma_0}{3N}\right) \sum_q n_q(1 + n_q)F(\boldsymbol{q}, \boldsymbol{q})Z(\omega_q) \tag{4.10}$$

in which $Z(\omega)$ is the spin-wave density of states,

$$Z(\omega) = \frac{1}{N}\sum_k \delta(\omega - \omega_k) \equiv (2\omega/N) \sum_k \delta(\omega^2 - \omega_k^2). \tag{4.11}$$

In the absence of dipolar forces $F(\boldsymbol{q}, \boldsymbol{q}) = 1$, and (4.10) diverges because when $q \to 0$, $Z(\omega_q) \sim q$, the product of Bose factors $\sim (1/q^4)$ and the density of q-states $\sim q^2$, i.e. the integral diverges logarithmically, cf (4.15).

The second form for $Z(\omega)$ is useful in view of the nature of the expression for the spin-wave dispersion (3.2). From (4.4),

$$F(\boldsymbol{q}, \boldsymbol{q}) = A_q^2/\omega_q^2 = 1 + |B_q|^2/\omega_q^2. \tag{4.12}$$

In order to gain an orientation for the behaviour of λ_h as a function of temperature, magnetic field and dipolar forces, it is reasonable to start by evaluating the density of states with the long-wavelength approximation for the exchange contribution to ω_k. Replacing in (4.11) the sum over $\boldsymbol{k}$ in a Brillouin zone by an integration over all $\boldsymbol{k}$, the density of states is

$$Z(\omega) = \left(\frac{\omega v_0}{4\pi^2\sqrt{2}\,D^2\zeta}\right)\int_{t_1}^{t_2} \frac{\mathrm{d}t\,(t-\frac{1}{2}-h/2D\zeta^2)^{1/2}}{[(t-\frac{1}{2})(t^2-t_1^2)]^{1/2}} \tag{4.13}$$

where the integration limits are

$$t_1 = \tfrac{1}{2}[1+(\omega/D\zeta^2)^2]^{1/2}$$

and

$$t_2^2 = t_1^2 + (\omega/2\zeta^2 D) = \tfrac{1}{4}[1+(\omega/D\zeta^2)]^2$$

with

$$\omega^2 \geqslant h^2 + 2\zeta^2 Dh$$

and it is zero otherwise. An expression for $Z(\omega)$ in terms of an elliptic integral of the third kind has not proved useful; its structure as a function of ω is illustrated in figure 3. Let us now evaluate λ_h from (4.10) and (4.13) for some special cases.

4.2.1. Non-dipolar magnet. On taking $\zeta = 0$ in (4.13),

$$Z(\omega) = \begin{cases} 0 & \omega < h \\ (v_0/4\pi^2 D^{3/2})(\omega-h)^{1/2} & \omega \geqslant h. \end{cases} \tag{4.14}$$

Using this in (4.10), together with $F(\boldsymbol{q},\boldsymbol{q}) = 1$, leads to

$$\lambda_h = \Gamma\int_0^\infty \mathrm{d}\omega\, n(\omega)[1+n(\omega)]Z^2(\omega) = (\Gamma_0 v_0^2 T^2\hbar/12\pi^3 D^3)\ln(1-\mathrm{e}^{-h/T})^{-1} \tag{4.15}$$

in which $n(\omega)$ is the function (4.7) with $\omega_q = \omega$. Thus in the limit of small magnetic fields λ diverges logarithmically, and for a fixed field it has a pronounced temperature dependence. As should be expected, the dependence of λ on T and h is the same as in $1/T_1$ calculated for NMR experiments performed on insulating ferromagnets (Mitchell 1957).

4.2.2. Zero field. Taking $h = 0$ in (4.13) leads to ($\omega \geqslant 0$)

$$Z(\omega) = (\sqrt{2}\,\zeta v_0 x/4\pi^2 D)\ln[(1+2x^{1/2}+2x)/(1+4x^2)^{1/2}] \tag{4.16}$$

in which the dimensionless variable

$$x = (\omega/2\zeta^2 D).$$

The expression for λ_h that results from using (4.16) and $h = 0$ in (4.6) is finite, and not expressible in a simple closed form. It can be shown that, in the limit of high temperatures,

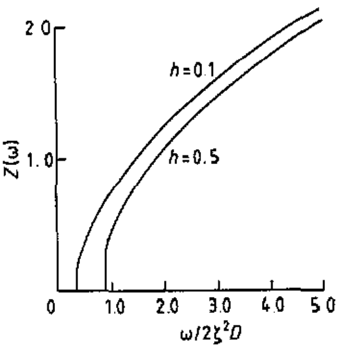

Figure 3. The quantity displayed is $Z(\omega)$, equation (4.13), in units of $v_0/4\pi^2\sqrt{2}\,D^2\zeta$, and ω and h are expressed in units of $2\zeta^2 D$. The density of states is zero for $0 \leqslant \omega \leqslant \omega_0 = (h+h^2)^{1/2}$. At ω_0 it achieves a finite value, proportional to $h^{3/4}$ for $h \ll 1$, and then increases with ω as $(\omega-\omega_0)^{1/2}$. Results are displayed for $h = 0.1$ and 0.5.

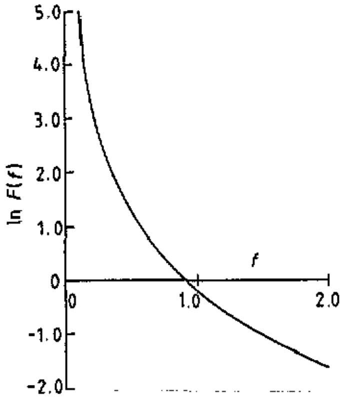

Figure 4. The integral $F(f)$ defined through (4.18) and (4.19) is illustrated in the form ln $F(f)$ as a function of f.

$\lambda_h \propto T^2$, and the proportionality constant is independent of ζ. For the opposite limiting case,

$$\lambda_h = (\Gamma_0 \zeta(\tfrac{3}{2}) v_0^2 T^{5/2} \hbar / 64\sqrt{2}\,\pi^{3/2} D^{7/2} \zeta) \qquad h = 0, T \ll T_c \qquad (4.17)$$

where $\zeta(n)$ is the Riemann zeta function of order n. For intermediate temperatures λ has been obtained by numerical integration.

The complete formula for the relaxation rate ($h = 0$) is

$$\lambda_h = (\Gamma_0 v_0 \hbar \zeta^3 \sqrt{2}/3\pi^2) \int \mathrm{d}x\, Z(\varepsilon) n(\varepsilon)[1 + n(\varepsilon)][(x^2 + \tfrac{1}{2}\sin^2\theta)/\varepsilon]^2 \qquad (4.18)$$

where the three-dimensional vector x is dimensionless,

$$\varepsilon = (x^4 + x^2 \sin^2\theta)^{1/2}$$

and

$$n(\varepsilon) = [\exp(2\zeta^2 D\varepsilon/T) - 1]^{-1}.$$

Notice that all parameters in the integral appear together as a single factor in the thermal distribution function.

Before proceeding to a discussion of numerical results derived from (4.8), we mention a pitfall in making a seemingly good approximation that brings (4.8) to the same appealing form as (4.15), in which λ is just a simple integral over Z^2. For $\zeta \neq 0$ we are prevented from obtaining such a result by the factor $\sin^2\theta$ in the integrand, which arises from A_q^2, in $F(q, q)$. It is then tempting to replace this angular factor by its spherical average. If this is done, λ is an integral over Z^2, given in (4.16), but the integral is divergent owing to the small-ω behaviour of the integrand.

It is convenient to express the result (4.18) in the form

$$\lambda_h = (2\Gamma_0 v_0^2 \zeta^4 \hbar / 3\pi^3 D) F(f) \qquad (4.19)$$

where the dimensionless variable

$$f = (2\zeta^2 D/T).$$

The dependence of the integral $F(f)$ on f is illustrated in figure 4. For small values of f it increases like $(1/f)^2$, which renders λ independent of ζ and proportional to T^2; we find for $f < 0.10$ that to an excellent approximation

$$F(f) = 0.862/f^2$$

which leads to

$$\lambda_h = (\Gamma_0 v_0^2 \hbar T^2 / D^3) \times 4.63 \times 10^{-3}. \qquad (4.20)$$

In the other extreme, $F(f)$ decays like $(1/f)^{5/2}$, and the result

$$F(f) = (1.36/f^{5/2}) \qquad f \to \infty$$

for $f = 10$ agrees with the numerical value to within 2%. In view of the strong variation of $F(f)$ we have chosen in figure 4 to display $\ln F(f)$ as a function of f.

We close this subsection with the observation that a fair estimate of λ_h for the case when dipolar forces and an external field are important can be obtained from (4.15). Away from extremely low temperatures it is reasonable to estimate the spin-wave dispersion relation by

$$\omega_k = Dk^2 + h + \tfrac{2}{3}(2\pi M_0 g \mu_B). \qquad (4.21)$$

The last term in this expression, obtained by averaging the dipolar contribution in (3.2) over the directions of $\boldsymbol{k}$ (Keffer 1966), is just an addition to the Zeeman energy in (4.15). Part of the justification behind such an approximate handling of the dipolar contribution to λ_h is that we have proved, in this subsection, that dipolar forces on their own lead to a finite relaxation rate, i.e. they do control the potential divergence of the integral in (4.6). Yet more reassurance comes from the fact that, at high temperatures, λ_h does not explicitly depend on the strength of the dipolar force between the magnetic ions. Indeed, the estimates (4.15) and (4.20) are quite similar. For EuO the dipolar contribution to the spin-wave dispersion (4.21) is only 6% of T_c.

The result provided in (4.15) is based on a density of states (4.14) derived using the long-wavelength approximation for the spin-wave energy, namely

$$\int_0^\infty d\omega\, n(\omega)[1 + n(\omega)] Z^2(\omega) = \left(\frac{v_0^2 T^2}{D^3 (2\pi)^4}\right) \ln(1 - e^{-h/T})^{-1}. \qquad (4.22)$$

To gauge the influence of the approximation to the density of states the integral has been evaluated with the full density of states for EuO. In this exercise, dipolar forces were not explicitly included but the field was chosen to have the value

$$h = \tfrac{2}{3}(2\pi M_0 g \mu_B) = 0.093\,\text{meV}.$$

The ratio of the result obtained with the full density of states and the right-hand side of (4.22), evaluated for the same h, is larger than unity for all temperatures, i.e. the approximate density of states (4.14) provides an underestimate of the integral. The ratio is 1.83 at $T = 0.1T_c$, and saturates beyond $0.5T_c$ at an average value of 7.2. Hence, the correction is significant at higher temperatures. To account partially for the effect of spin-wave collisions, which are important at high temperatures (cf section 4.1), the calculation of the integral was repeated at $T = 0.8T_c$ for a field obtained from the measured magnetization $M = 0.67M_0$ (Passell *et al* 1976) and low-temperature exchange interactions; there was next to no change in the exact numerical value and the ratio decreased to the value 6.78. Softening the spin waves is not expected to influence the ratio very much since the spin-wave stiffness appears as a factor in the approximate expression provided in (4.22).

5. Conclusions

A muon spin relaxation experiment on an ordered ferromagnet has been proposed with the aim of improving knowledge of spin fluctuations parallel to the easy axis of magnetization. Since many previous experiments have used neutron beam techniques, the relation between response functions observed in these and μSR experiments has been carefully established.

Predictions for the dependence of the μSR relaxation rate on temperature, magnetic field and dipolar interactions have been obtained from spin-wave theory. The latter should be perfectly reliable at temperatures small compared with the critical temperature. But there are reasons why it might be accurate over a considerably wider temperature range, which stem from the fact that the relaxation rate is dominated by longitudinal spin fluctuations (Raman processes) and these involve two-spin-wave events, i.e. non-linear events. Hence, the evaluation of Raman processes in lowest order, as reported here, actually takes the level of calculation of the relaxation rate beyond the domain covered by linear spin-wave theory.

The dipolar and hyperfine mechanisms for relaxation possess different characteristics. A potential divergence of the $\boldsymbol{k}$ integration in the formula for λ_h is controlled by the dipolar forces between the magnetic ions. However, at high temperatures λ_h does not explicitly depend on the strength of the dipolar forces. There are no such subtle features about the dipolar contribution, at least when the muon occupies a site at which the average value of the dipolar field from the surrounding magnetic ions is zero. This property introduces in the $\boldsymbol{k}$ integral a function that controls the potential divergence seen in the corresponding formula for λ_h. In consequence, λ_d has been estimated with simple ferromagnetic spin waves, and dipolar forces between magnetic ions are not included. If the ratio of the hyperfine and dipolar coupling constants for the muon is small, as it is expected to be in EuO, then the observed relaxation rate will be dominated by the dipolar contribution λ_d, i.e. spin dynamics are not strongly revealed in the hyperfine contribution.

The longitudinal spin fluctuations of interest are created by fluctuations in the dipolar field experienced by the implanted muon (which is assumed to be a perfectly passive probe of the ordered magnetic state). Given that the muon is equidistant from each coordination shell of atomic moments and zero-point motion of the muon is not influential, our theory should cover all experimental geometries. The theory is presented in a form that makes a generalization to non-equidistant configurations relatively straightforward.

The magnetic salt EuO is of particular interest, and predictions for μSR specific to it are presented. Many properties of EuO are firmly established, but neutron beam data on longitudinal fluctuations are not in accord with existing theoretical models. It is well suited for μSR experiments for several reasons, including the fact that Eu^{2+} is an S-state ion, and thus quadrupole interactions will not complicate this interpretation of data. It is shown how the μSR signal depends on the orientation of the muon beam relative to one of the easy axes, but this dependence is completely averaged out in a multi-domain sample. A predominance of one domain can possibly be achieved with a magnetic field of a strength so small that the Larmor frequency of the muon is negligible compared to electronic frequencies, in which case a properly shaped sample is required to avoid demagnetizing field in homogeneities.

Experiments on such a sample will permit stringent tests of the influence of spin fluctuations on the muon spin relaxation in an ordered ferromagnet, and evidence for fluctuations in the dipolar field at the muon as the principal relaxation mechanism.

Acknowledgments

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Appendix

Here we provide formulae for the dipolar and hyperfine relaxation rates. It is assumed that identical magnetic ions are located at positions $\{\boldsymbol{l}\}$ with respect to the implanted muon and $|\boldsymbol{l}| = d$. The magnetic moment carried by each ion is $g\mu_B \boldsymbol{S}(\boldsymbol{l})$. Figure 1 illustrates the configuration of the muon and neighbouring spins in EuO.

The dipolar field at the muon is expressed in terms of spherical components of the spin operators by means of the standard expression

$$B_Q = \left(\frac{g\mu_B}{d^3}\right) \sum_{l} \left\{ S(l) - \frac{3}{d^2} l[l \cdot S(l)] \right\}_Q = \left(\frac{g\mu_B}{d^3}\right) (8\pi)^{1/2} \sum_{l} \sum_{qq'} S_q(l) Y^2_{q'}(l) (1q2q'|1Q). \tag{A1}$$

In the last equality the spin operator is coupled to a spherical harmonic of rank two by a Clebsch–Gordon coefficient. We use the phase conventions employed by Edmonds (1960).

Fluctuations in the dipolar field away from its average value produce a relaxation of the muon spin signal. For simple magnets it is likely that $\langle \boldsymbol{B} \rangle = 0$, and EuO is one such material.

The relaxation rate λ_d is obtained by a straightforward application of Fermi's golden rule for the perturbation

$$\mathcal{H} = g_\mu \mu_N \boldsymbol{I} \cdot \boldsymbol{B} \tag{A2}$$

where $\boldsymbol{I}$ is the muon spin operator. The muon Larmor frequency is set equal to zero in the following calculations, for reasons explained in the text, and the initial and final muon spin states are labelled by m and m', respectively. From (A2) we obtain

$$\lambda_d = 2\left(\frac{g_\mu \mu_N}{\hbar}\right)^2 \int_{-\infty}^{\infty} dt \sum_{QQ'} (-1)^{Q+Q'} \langle m|I_Q|m'\rangle \langle m'|I_{Q'}|m\rangle \langle B_{-Q}(t) B_{-Q'}\rangle / \langle m|I^-|m'\rangle^2 \tag{A3}$$

where the normalization is chosen such that λ_d is the same as the conventional definition of $1/T_1$.

If the muon spin axis is defined by polar angles (α, β) with respect to the crystal axes, the single spin-flip matrix elements required in (A3) are

$$
\begin{aligned}
\langle m|I_0|m'\rangle &= -\tfrac{1}{2}\sin\alpha\langle m|I^-|m'\rangle \\
\langle m|I_{+1}|m'\rangle &= (\mathrm{e}^{\mathrm{i}\beta}/\sqrt{2})\sin^2(\alpha/2)\langle m|I^-|m'\rangle \\
\langle m|I_{-1}|m'\rangle &= (\mathrm{e}^{-\mathrm{i}\beta}/\sqrt{2})\cos^2(\alpha/2)\langle m|I^-|m'\rangle
\end{aligned}
\tag{A4}
$$

in which

$$\langle m|I^-|m'\rangle^2 = (I+m+1)(I-m). \tag{A5}$$

In fact, it is unlikely that the sample will be a single domain, or even possess a predominant domain. Hence, for most cases of interest it is appropriate to average (A3) over the orientations of the muon spin. Using the result (A4) we obtain

$$\lambda_d = \tfrac{1}{3}\left(\frac{g_\mu \mu_N}{\hbar}\right)^2 \int_{-\infty}^{\infty} \mathrm{d}t \sum_Q (-1)^Q \langle B_Q(t) B_{-Q}\rangle \tag{A6}$$

in which it remains to isolate the spin components that affect relaxation.

For reasons given in the text, we select Raman processes that are described by the longitudinal spin correlation function $\langle S^z(t)S^z\rangle$, i.e. fluctuations along the easy axis. On inserting in (A6) the expression (A1) for B_Q, and introducing the definition

$$\Gamma = \tfrac{8}{3}(g g_\mu \mu_B \mu_N/\hbar d^3)^2 \tag{A7}$$

we are led to the result

$$\lambda_d = \left(\frac{\pi\Gamma}{10}\right) \int_{-\infty}^{\infty} \mathrm{d}t \sum_{\substack{ll'\\Q}} (4-Q^2) Y_Q^2(l)[Y_Q^2(l')]\langle S^z(l,t)S^z(l',0)\rangle \tag{A8}$$

in which $Q = 0, \pm 1$.

Up to this point no mention of the configuration of the atomic spins has entered the discussion, other than that they are identical and equidistant from the muon. We continue for the special case when the average dipolar field at the muon is zero, achieved for a configuration $\{l\}$ that satisfies the condition

$$\sum_l Y_q^2(l) = 0. \tag{A9}$$

We further assume that, as in the case of EuO, there are just two distinct spin correlation functions, namely the site-diagonal and nearest-neighbour functions. In this case (A8) reduces to

$$\lambda_d = \left(\frac{\pi\Gamma}{10}\right) \int_{-\infty}^{\infty} \mathrm{d}t \langle S^z(m,t)S^z(m,0) - S^z(m+\delta,t)S^z(m,0)\rangle \sum_{l,Q} (4-Q^2)|Y_Q^2(l)|^2. \tag{A10}$$

For the configuration of spins illustrated in figure 1, which is believed to be appropriate for EuO, we find

$$\sum_l |Y_0^2(l)|^2 = (5/3\pi) \tag{A11}$$

and

$$\sum_l |Y_1^2(l)|^2 = (5/9\pi).$$

We conclude this discussion of the dipolar relaxation mechanism by providing the value of the coupling constant when the muon spin has a definite orientation specified by (α, β) with respect to the crystal axes, and the easy axis (z axis) is along the EuO crystal (1 1 1) direction:

$$\Gamma' = (\Gamma/4)[3 + \cos^2\alpha + 2\sin^2\alpha\cos^2(\pi/4 - \beta) - \sqrt{2}\sin(2\alpha)\cos(\pi/4 + \beta)]. \tag{A12}$$

Upon averaging Γ' over the polar angles (α, β), $\Gamma' = \Gamma$ as expected. Note that if the muon and electronic spins are parallel, $\Gamma' = 3.5\Gamma$.

For the discussion of the hyperfine relaxation mechanism, let the easy axis (z axis) be defined by polar angles (θ, φ) with respect to the crystal axes. The perturbation is then of the form

$$\begin{aligned}\mathcal{H} = \sum_l A_l \boldsymbol{I}\cdot\boldsymbol{S}(l) = \tfrac{1}{2}\sum_l A_l S^z(l) I^-[&\sin\theta_l\cos\varphi_l(\cos\alpha\cos\beta - \mathrm{i}\sin\beta) \\ &+ \sin\theta_l\sin\varphi_l(\cos\alpha\sin\beta + \mathrm{i}\cos\beta) - \cos\theta_l\sin\alpha]\end{aligned} \tag{A13}$$

where the second equality is achieved by selecting the longitudinal spin components. In calculating the relaxation rate λ_h we begin by retaining just the site-diagonal spin correlation functions, and this gives

$$\lambda_h = \left(\frac{\Gamma_0'}{16}\right)\int_{-\infty}^{\infty} \mathrm{d}t\,\langle S^z(m,t)S^z(m,0)\rangle \tag{A14}$$

where

$$\Gamma_0' = (N_0\Gamma_0/16)\{1 - [\cos\theta\cos\alpha + \sin\theta\sin\alpha\cos(\varphi - \beta)]^2\} \tag{A15}$$

in which N_0 is the number of magnetic ions and

$$\Gamma_0 = 8(A/\hbar)^2. \tag{A16}$$

Note that Γ_0' vanishes for a collinear arrangement of the atomic and muon spins. For a multi-domain EuO sample ($N_0 = 4$)

$$\Gamma_0' = \tfrac{4}{3}(A/\hbar)^2.$$

It is relatively easy for this special case to calculate the contribution to λ_h made by the next-nearest-neighbour spin correlation functions. The result

$$\lambda_h = \left(\frac{\Gamma_0}{6}\right)\int_{-\infty}^{\infty} \mathrm{d}t\,\langle S^z(m,t)S^z(m,0) + 3S^z(m+\delta,t)S^z(m,0)\rangle \tag{A17}$$

corresponds to (A10) for the dipolar mechanism.

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