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Muon spin relaxation in ferromagnets: II. Critical and paramagnetic magnetization fluctuations

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Abstract. Mechanisms for the relaxation of muon spin relaxation signals in paramagnets are analysed in terms of spin fluctuations in a Heisenberg magnet with a ferromagnetic exchange interaction. Explicit predictions provided for EuO are based on the assumption that the implanted muon occupies an interstitial site of high symmetry. Because the average dipolar field at the muon site is zero, and the material is ferromagnetic, the dipolar mechanism of relaxation is not enhanced by critical spin fluctuations. In consequence, the hyperfine mechanism dominates relaxation in the critical region, even though the ratio of the hyperfine and dipolar coupling constants might be small. Calculations of relaxation rates for both mechanisms, made on the basis of coupled-mode theory, are provided for critical and paramagnetic states.

1. Introduction

Numerous research groups have studied the relaxation rate of muon spin relaxation (μSR) signals in paramagnets; see, for example, reviews by Cox (1987), Hohenemser *et al* (1989) and Karlsson (1990). The magnetic materials used include ferromagnetically and antiferromagnetically coupled salts and metals. The consensus from the reported work is that the relaxation rate λ increases as the critical temperature T_c is approached from above. In some cases, a significant enhancement of λ is observed well above T_c. While some general statements have been made about the dependence of λ on critical exponents, no detailed calculations of λ in the critical and paramagnetic regions have appeared. As a first remedial step, we report the findings of an investigation of the temperature dependence of λ that is based on the Heisenberg model of a ferromagnetically coupled (simple) magnet. The present work complements our discussion of relaxation in an ordered ferromagnet (Lovesey *et al* 1992, hereafter referred to as I).

Our choice of the Heisenberg model means that we can draw on a wealth of previous theoretical work on critical and paramagnetic spin fluctuations. In particular, we use results obtained using dynamic scaling arguments, and the renormalization-group and coupled-mode theory. The Heisenberg model can be used with confidence to describe properties of magnetic salts. We illustrate the implications of our theoretical results by evaluating them for parameters appropriate to EuO, which has been extensively studied by many other experimental techniques. An outcome of our work is a clear statement of how λ is related to quantities observed with some of these techniques, e.g. neutron spectroscopy. Although we draw on material given in I, to avoid unnecessary repetition of data and formula, we strive to make the present paper more or less self-contained.

Attention is focused on two possible magnetic mechanisms for relaxation of the μSR signal. One arises from fluctuations in the dipolar field created by the atomic magnetic moments. If the implanted muon in EuO resides at a high-symmetry interstitial site, the average dipolar field it experiences is zero. The second mechanism considered is generated by the hyperfine interaction. While the ratio of the hyperfine and dipolar coupling constants is likely to be small in a magnetic salt, we argue that critical spin fluctuations enhance only the hyperfine mechanism, which therefore dominates in the critical region.

Formulae for the contributions to λ made by these two mechanisms are reviewed in section 2. These formulae are obtained from expressions derived in I using standard first-order perturbation theory. The relaxation rate is a bulk response function, in the sense that the specific heat, susceptibility, thermal conductivity, etc., are bulk response functions. In contrast, the response function observed in neutron scattering can be viewed as a differential response function that does contain direct evidence of elementary excitations (spin waves), critical fluctuations and collective modes. The formulae for λ in section 2 make explicit this fundamental difference, for λ is proportional to an integral over all wavevectors of the neutron scattering response function. A consequence of this relationship is that λ might not reflect strong features in the neutron scattering response because the integration negates their influence. Set against this, μSR permits measurements to be made that are not currently possible by other techniques, e.g. the measurement of spin fluctuations in zero magnetic field.

In section 3 the formulae for λ, from section 2, are expressed in terms of quantities that are natural vehicles for theoretical discussions. There is also a brief review of coupled-mode theory of spin fluctuations, which is the basis of calculations described in sections 4 and 5. Critical fluctuations are discussed in section 4; results of previous work enable us to provide a concrete prediction for the enhancement of λ. The temperature dependence of λ in the paramagnetic region is the subject of section 5, and specific results for EuO are gathered in section 6. Our findings are summarized in section 7.

2. Relaxation mechanisms

An implanted muon interacts with the atomic moments in a magnetic material largely through dipolar and hyperfine mechanisms. Both these mechanisms are thoroughly discussed in I. Here we record the expressions for λ appropriate to the paramagnetic state in which the atomic spins are disordered. Hence, the results of immediate interest are obtained from results in I for the special case when we average over the orientation of the muon spin relative to the atomic spins.

Following the discussion of EuO given in I, the muon is taken to be equidistant from four Eu ions, located at sites defined by lattice vectors $\{\boldsymbol{l}\}$. For this arrangement, illustrated in figure 1, the average dipolar field at the muon is zero. In the calculation of λ, using standard first-order perturbation theory, this value of the average dipolar field is manifest in the identity

$$\sum_{l} Y_Q^2(\boldsymbol{l}) = 0 \tag{2.1}$$

for all components of the second-order spherical harmonic. From results in I we obtain the following contribution to λ due to fluctuations in the dipolar field,

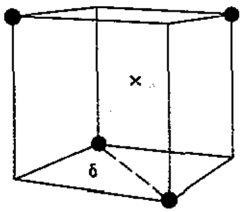

Figure 1. The interstitial site assumed for the implanted μ^+ in EuO is illustrated relative to the four nearest-neighbour Eu ions. Units are such that the length of one side of the cube in the figure is $a/2$, where a is the FCC cell dimension.

$$\lambda_{\rm d} = 3\Gamma \int_{-\infty}^{\infty} {\rm d}t \langle S^z(l,t)S^z(l,0) - S^z(l+\delta,t)S^z(l,0)\rangle. \tag{2.2}$$

Here,

$$\Gamma = \tfrac{8}{3}(2g_\mu \mu_{\rm B}\mu_{\rm N}/\hbar d^3)^2 = 0.055 \times 10^6\,(\mu{\rm s})^{-2} \tag{2.3}$$

in which $d = a\sqrt{3}/4$ is the distance between the muon site and a Eu ion, expressed in terms of the cell length $a = 5.14$ Å. The factor 3 in (2.2) arises because $\langle S^\alpha S^\alpha \rangle$ is independent of the Cartesian label $\alpha = x,\ y,\ z$ in the paramagnetic state, and $\boldsymbol{\delta} = (a/2)(1,1,0)$ connects nearest-neighbour Eu ions. The fact that λ is related to the difference of two spin correlation functions is a direct consequence of (2.1).

Turning now to the hyperfine mechanism, we find

$$\begin{aligned} \lambda_{\rm h} &= (1/\hbar^2) \sum_{l,l'} A_l A_{l'} \int_{-\infty}^{\infty} {\rm d}t \langle S^z(l,t)S^z(l',0)\rangle \\ &= (4A^2/\hbar^2) \int_{-\infty}^{\infty} {\rm d}t \langle S^z(l,t)S^z(l,0) + 3S^z(l+\delta,t)S^z(l,0)\rangle \end{aligned} \tag{2.4}$$

where the second equality is obtained when the coupling constant A is the same for all sites. The expectation is that $4(A/\hbar)^2 \ll 3\Gamma$.

It is very helpful to express the spin correlation functions in (2.2) and (2.4) in terms of the Van Hove response function,

$$S(\boldsymbol{k},\omega) = \frac{1}{N} \sum_{m,n} \int_{-\infty}^{\infty} \frac{{\rm d}t\, {\rm e}^{-{\rm i}\omega t}}{2\pi\hbar} {\rm e}^{{\rm i}\boldsymbol{k}\cdot(\boldsymbol{m}-\boldsymbol{n})} \langle S^z(m,0)S^z(n,t)\rangle. \tag{2.5}$$

In this expression, the lattice sums run over all N magnetic sites in the sample. We now find

$$\lambda_{\rm d} = (6\pi\hbar\Gamma/N) \sum_{\boldsymbol{k}} S(\boldsymbol{k},0)[1 - \cos(\boldsymbol{k}\cdot\boldsymbol{\delta})] \tag{2.6}$$

and

$$\lambda_{\rm h} = (\pi\hbar\Gamma_0/N) \sum_{\boldsymbol{k}} S(\boldsymbol{k},0)[1 + 3\cos(\boldsymbol{k}\cdot\boldsymbol{\delta})] \tag{2.7}$$

with

$$\Gamma_0 = 8(A/\hbar)^2. \tag{2.8}$$

We recall that the relaxation rates are related to $S(\boldsymbol{k},\omega)$ evaluated at $\omega = 0$; the muon Larmor frequency is very small compared with characteristic electronic frequencies, so it may be taken to be zero to a very good approximation.

The expressions (2.6) and (2.7) are the basis of subsequent discussions. As mentioned in the introduction, they suggest that λ is quite properly regarded as a bulk response function akin to more familiar bulk response functions such as the specific heat, susceptibility, etc. By the same token, $S(\boldsymbol{k}, \omega)$ is a differential response function that does contain direct signatures of spin waves, collective modes and critical fluctuations. The formula for the nuclear magnetic resonance (NMR) linewidth ($1/T_2$) is essentially the same as (2.7) apart from the geometric factor (Heller 1967).

3. Response functions

The Van Hove response function $S(\boldsymbol{k}, \omega)$, introduced in the previous section, is related to the spectrum of neutrons scattered by a paramagnet. Knowledge of its properties has been obtained from a variety of calculations using one of several techniques. To facilitate contact with this body of theoretical work, it is necessary to record the relation between $S(\boldsymbol{k}, \omega)$ and the Kubo relaxation function, denoted here by $F(\boldsymbol{k}, t)$ (background material is covered by Lovesey (1986)). The latter satisfies a generalized Langevin equation

$$\partial_t F(\boldsymbol{k}, t) = -\int_0^t \mathrm{d}t'\, K(\boldsymbol{k}, t-t') F(\boldsymbol{k}, t') \tag{3.1}$$

in which $K(\boldsymbol{k}, t)$ is usually called the memory function, and its time–Laplace transform the collisional self-energy. Equation (3.1) is the common starting point for a coupled-mode theory, based on (3.8), which gives results for $F(\boldsymbol{k}, t)$ that have been demonstrated to be good in the critical and paramagnetic regions (Cuccoli *et al* 1989, 1990). For the critical region the results are consistent with dynamic scaling arguments and calculations based on the method of the renormalization group. Static correlations are consistent with the spherical model.

With our set of definitions,

$$F(\boldsymbol{k}, t=0) = \chi(\boldsymbol{k}) \tag{3.2}$$

is the isothermal susceptibility. If $\bar{K}(\boldsymbol{k}, s)$ denotes the Laplace transform of $K(\boldsymbol{k}, t)$, the relaxation rates of interest are

$$\lambda_{\mathrm{d}} = (6\Gamma T/N) \sum_{\boldsymbol{k}} [\chi(\boldsymbol{k})/\bar{K}(\boldsymbol{k}, 0)][1 - \cos(\boldsymbol{k}\cdot\boldsymbol{\delta})] \tag{3.3}$$

and

$$\lambda_{\mathrm{h}} = (\Gamma_0 T/N) \sum_{\boldsymbol{k}} [\chi(\boldsymbol{k})/\bar{K}(\boldsymbol{k}, 0)][1 + 3\cos(\boldsymbol{k}\cdot\boldsymbol{\delta})] \tag{3.4}$$

in which T is the absolute temperature ($k_{\mathrm{B}} = 1$).

The initial value of $K(\boldsymbol{k}, t)$ is the second frequency moment of $S(\boldsymbol{k}, \omega)$. For a Heisenberg magnet with a nearest-neighbour exchange of strength J between spins of magnitude S,

$$K(\boldsymbol{k}, t=0) = \omega_0^2(\boldsymbol{k}) = (4rJ/\chi(\boldsymbol{k}))(1-\gamma_{\boldsymbol{k}})\langle S^z(0)S^z(\delta)\rangle. \tag{3.5}$$

Here, the static correlation function involves nearest-neighbour spins separated by a distance δ, r is the number of nearest neighbours, and the geometric factor

$$\gamma_k = (1/r) \sum_{\delta} \cos(\boldsymbol{k} \cdot \boldsymbol{\delta}). \tag{3.6}$$

Note that in (3.3) and (3.4) we can replace $\cos(\boldsymbol{k} \cdot \boldsymbol{\delta})$ by γ_k. For an FCC lattice, appropriate for EuO, $r = 12$, $\delta^2 = a^2/12$ and

$$3\gamma_k = \cos\left(\frac{a}{2}k_x\right)\cos\left(\frac{a}{2}k_y\right) + \cos\left(\frac{a}{2}k_y\right)\cos\left(\frac{a}{2}k_z\right) + \cos\left(\frac{a}{2}k_z\right)\cos\left(\frac{a}{2}k_x\right). \tag{3.7}$$

The memory function $K(\boldsymbol{k}, t)$ is not known exactly. Probably the best description available today is obtained from coupled-mode theory, in which

$$K(\boldsymbol{k}, t) = (Q_k/N) \sum_{\boldsymbol{q}} (\gamma_{k+q} - \gamma_q)^2 F(\boldsymbol{k} + \boldsymbol{q}, t) F(\boldsymbol{q}, t). \tag{3.8}$$

Taken together with (3.1) this prescription for $K(\boldsymbol{k}, t)$ provides an integro-differential equation for $F(\boldsymbol{k}, t)$, and thus results for $K(\boldsymbol{k}, t)$. The function Q_k in (3.8) is

$$Q_k = (2rJ)^2 T/\chi(\boldsymbol{k}). \tag{3.9}$$

It is relatively easy to demonstrate that (3.8) for $K(\boldsymbol{k}, t)$ is consistent with the spherical model result for $\chi(\boldsymbol{k})$. One finds

$$2rJ\chi(\boldsymbol{k}) = (1/\mu - \gamma_k)^{-1} \tag{3.10}$$

in which the temperature factor μ is related to the critical temperature T_c through

$$T/T_c = I(1)/I(\mu) \tag{3.11}$$

with the standard definition of Watson's extended integral

$$I(\mu) = \left(\frac{1}{2\pi}\right)^3 \int_{-\pi}^{\pi} \mathrm{d}\boldsymbol{k} \left(\frac{1}{\mu} - \gamma_k\right)^{-1}. \tag{3.12}$$

For an FCC lattice, $I(1) = 1.3446\ldots$ (Joyce 1972).

Returning to $\omega_0(\boldsymbol{k})$ defined in (3.5), the explicit value, consistent with mode-coupling theory,

$$\omega_0^2(\boldsymbol{k}) = [2(2rJ)^2/3\mu^2\hbar^2] S(S+1)(1-\gamma_k)(1-\mu\gamma_k)[1-\mu/I(\mu)]. \tag{3.13}$$

Finally, we record the relation between T_c and J, namely

$$\tfrac{1}{3}(2rJ)S(S+1) = I(1)T_c. \tag{3.14}$$

Here, as elsewhere in this section, the relation is valid for a nearest-neighbour exchange interaction. This model is not entirely satisfactory for EuO. Measurements by Passell *et al* (1976) show that a second-nearest-neighbour coupling is significant in this material. We employ a single exchange interaction because of the simplifications it brings to the calculations. However, it should be noted that coupled-mode theory can be prepared for an arbitrary range of the exchange interaction (Cuccoli *et al* 1989).

4. Critical fluctuations

To deduce the properties of the relaxation rates λ_d and λ_h from (3.3) and (3.4) it is evident that we need expressions for $\chi(\boldsymbol{k})$ and $\tilde{K}(\boldsymbol{k}, 0)$ in the critical region, where the inverse correlation length κ satisfies $a\kappa \ll 1$ and $\kappa = 0$ at T_c. For $\chi(k)$ the appropriate expression is derived from (3.10),

$$2a^2 J\chi(k) = (\kappa^2 + k^2)^{-1} \tag{4.1}$$

where we neglect the critical exponent η because it is very small for the Heisenberg model in three dimensions.

In deriving an expression for $\bar{K}(k, 0)$ we begin by noting that, according to the coupled-mode theory, $K(k, t)$ is a generalized homogeneous function at $T = T_c$ of the form

$$K(k, t) = \zeta K(\zeta^{-1/5}k, \zeta^{1/2}t). \tag{4.2}$$

The choice $\zeta = \xi^2 k^5$, where ξ is a non-universal energy constant, is consistent with coupled-mode theory, dynamic scaling and calculations based on the renormalization group (Hohenberg 1981, Balucani *et al* 1987). From these results it follows that $\bar{K}(k, 0)$ is proportional to $\xi k^{5/2}$. The proportionality constant is obtained from an explicit calculation using one of the two types of theory. Here we follow Lovesey and Williams (1986), who obtain from the theory reviewed in section 3,

$$\bar{K}(k, 0) = \xi k^{5/2} \times 4.510. \tag{4.3}$$

Armed with (4.1) and (4.3) we proceed to a description of the relaxation rates in the critical region.

From (3.3) it follows by inspection that λ_d is not divergent at the critical temperature. The potential divergence of the integral in (3.3) is killed by the geometric factor.

The same fate does not befall λ_h; it is straightforward to demonstrate that λ_h diverges like $\kappa^{-3/2}$ as $T \rightarrow T_c$. On performing the k integration in (3.4),

$$\lambda_h = (0.40 A^2 T_c v_0/\hbar a^2 J\xi)(1/\kappa)^{3/2}. \tag{4.4}$$

For EuO, $\xi = 1.31$ meV Å$^{5/2}$, $a = 5.14$ Å, $v_0 = a^3/4$ and $T_c = 69.5$ K is consistent in the spherical model with $J = 0.74$ K. For these parameters the factor in (4.4) has the value

$$36.85(A^2/\hbar)\,(\text{meV Å}^{3/2})^{-1}.$$

The power-law divergence predicted by (4.4) is most usefully written in the form

$$(1/\kappa)^{2+z-d-\eta}$$

where d is the spatial dimensionality, and $z = 5/2$ and η are the usual critical exponents, taken together with the definition

$$\kappa \propto (T/T_c - 1)^{\nu}.$$

Recall that the exponent for the static susceptibility γ satisfies $\gamma = \nu(2 - \eta)$. A first-order renormalization-group analysis yields $z = 1 + d/2$ on both sides of the critical temperature.

The results for λ_d and λ_h demonstrate that critical fluctuations enhance the hyperfine mechanism for muon relaxation, but not the dipolar mechanism. So sufficiently close to T_c we predict $\lambda_d \ll \lambda_h$. The absence of an enhancement of λ_d stems from the spatial property of the dipolar interaction for a special high-symmetry environment (the average dipolar field is zero at the position occupied by the implanted muon), and the union of this property with a ferromagnetic susceptibility that peaks at $k = 0$.

The estimate (4.4) does not take account of the influence of dipolar interactions between the atomic moments (Huber 1971, Frey and Schwabl 1988), which have been shown to have a significant effect on $F(k, t)$ at very small wavevectors (Mezei 1986). A dipolar-induced cross-over to a critical exponent $z = 2.0$ occurs, in the theory, at a k-value that is about an order of magnitude smaller than the value for cross-over in static properties and time dependence, and confirmation remains a challenge to the experimentalist. Note that for λ_d to display an enhancement the value of z must exceed $d = 3$.

5. Paramagnetic fluctuations: theory

Our discussion of the relaxation rates in the paramagnetic phase is based on the coupled-mode theory result (3.8). Hence, the susceptibility is taken to be (3.10), i.e. the spherical model susceptibility (Joyce 1972).

The memory function (3.8) has been obtained for EuO, and other materials, at selected wavevectors by numerically solving the coupled equations (3.1) and (3.8) (Cuccoli *et al* 1989, 1990). To proceed along the same lines with the calculation of the relaxation rates (3.3) and (3.4) demands a knowledge of $K(k, t)$ at every point used in the k integration routine. This clearly amounts to a very large computation, which is probably not justified at present. Instead, we use the result (3.8) to generate an approximate analytic expression for $K(k, t)$ that provides a tolerable result for $\bar{K}(k, 0)$; the method adopted is closely related to ideas described by Lovesey and Meserve (1973).

The function $\bar{K}(k, 0)$ is the integral of $K(k, t)$ over all t. Hence, deep in the paramagnetic region it is not very sensitive to fine details of $K(k, t)$. Moreover, when $T > T_c$, $F(q, t)$ is a relatively benign function of its arguments, so $K(k, t)$, which is a joint function of $F(k + q, t)$ and $F(q, t)$, is also quite benign. In view of this, we will for the purpose of calculating $\bar{K}(k, 0)$ represent $K(k, t)$ by a simple gaussian function of t,

$$\bar{K}(k, 0) \simeq \int_0^\infty dt\, \omega_0^2(k) \exp[-\tfrac{1}{2}t^2\delta^2(k)] = (\omega_0^2/\delta)\surd(\pi/2). \tag{5.1}$$

Here, ω_0^2 is the second moment of $S(k, \omega)$ and it arises in (5.1) because we require the approximate $K(k, t)$ to satisfy (3.5). The function $\delta(k)$ is derived from (3.8) by noting that it is proportional to the second time derivative of $K(k, t)$ evaluated at $t = 0$. In this way we find

$$\delta^2(k) = 8JTL(k) \tag{5.2}$$

with

$$(2/r)(1-\gamma_k)L(k) = \frac{1}{N}\sum_q (\gamma_{k+q} - \gamma_q)^2(1-\gamma_{k+q})(1/\mu - \gamma_q)^{-1}. \tag{5.3}$$

The reasoning that leads us to propose (5.1) is vindicated by the fact that $L(k)$, obtained by numerical integration, is a mild function of k and T. In fact, the following analytic representation for an FCC lattice, obtained by making an expansion in μ, is good over the range of variables of immediate interest,

$$L(k) = \mu[1 + (\mu/48)(4 - 9\gamma_k) + (\mu^2/96)(19 - 9\gamma_k)]. \tag{5.4}$$

Using the expression in (5.2) the values obtained for $\delta(k)$ differ from the exact numerical values by a few per cent for $\mu < 0.8$, and at $\mu = 0.9$ ($T/T_c = 1.30$) the error is 9%. This level of accuracy is deemed to be quite consistent with the general tenor of our calculation, and fully justified by the simplification it brings to the calculation of the integrals involved in the expressions (3.3) and (3.4).

In order to assess the Gaussian approximation for the memory function, we compare it to the exact result, obtained by a full numerical solution of the coupled-mode equation (Cuccoli *et al* 1989). Figure 2 shows results for EuO, $T = 1.5\,T_c$ and $\boldsymbol{k} = \pi(1, 1, 1)/2a$. The difference in the initial values is attributed to the use in the couple-mode calculation of nearest and next-nearest exchange interactions. At much longer times (>34.0 reduced time steps) the exact memory function changes sign, but the magnitude does not exceed $\sim 10^{-4}$ of the initial value. The long tail in the exact memory function is expected, and it becomes a stronger feature as $T \to T_c$. Although the Gaussian approximation to the memory function is but tolerable, its integral, which occurs in the relaxation rate, differs from the exact result by a mere 3.5%. This finding adds weight to our confidence in using the Gaussian approximation to estimate the paramagnetic relaxation rate.

In closing this section we note that $\delta^2(\boldsymbol{k})$ is formally the second cumulant of $S(\boldsymbol{k}, \omega)$, i.e. the mean-square width of the neutron scattering response. Hence, (5.2) and (5.3) give the coupled-mode theory approximation to the mean-square width of $S(\boldsymbol{k}, \omega)$. This quantity is known exactly in the extreme limit $T = \infty$, and the result is (FCC)

$$32J^2S(S+1)[\tfrac{7}{3} - \tfrac{1}{2}\gamma_k - 1/16S(S+1)].$$

The corresponding limiting value of $\delta^2(\boldsymbol{k})$ is

$$\delta^2(\boldsymbol{k}) = 64J^2S(S+1).$$

It is not surprising that $\delta(\boldsymbol{k})$ contains less information than the exact expression, since it is an approximate result. The close agreement in the numerical values of the two expressions (for $S = \tfrac{7}{2}$) is added support for our confidence in (5.4) as a tolerable estimate.

6. Paramagnetic fluctuations: numerical results

Here we report the findings for the relaxation rates λ_d and λ_h obtained from the theory described in the previous section. To this end it is useful to gather the various components that appear in the expressions we have evaluated.

Let us write

$$\lambda_d = (3\hbar\Gamma/r^2J)(T_c/\pi J)^{1/2}J_d(\mu) \tag{6.1}$$

where the temperature dependence is solely contained in

$$J_d(\mu) = \left(\frac{I(1)}{I(\mu)}\right)^{1/2}\left(-1 + \frac{1}{\mu}I(\mu)\right)^{-1}\frac{1}{N}\sum_{k}[L(\boldsymbol{k})]^{1/2}\left(\frac{1}{\mu} - \gamma_k\right)^{-2}. \tag{6.2}$$

For EuO the prefactor in (6.1) has the value 0.063 $(\mu s)^{-1}$. Turning attention to the hyperfine contribution to the relaxation rate, the analogue of (6.1) is

$$\lambda_h = (\hbar\Gamma_0/2r^2J)(T_c/\pi J)^{1/2}J_h(\mu) \tag{6.3}$$

with

$$J_h(\mu) = \left(\frac{I(1)}{I(\mu)}\right)^{1/2}\left(-1 + \frac{1}{\mu}I(\mu)\right)^{-1} \times \frac{1}{N}\sum_{k}[L(\boldsymbol{k})]^{1/2}(1 + 3\gamma_k)\left[(1 - \gamma_k)\left(\frac{1}{\mu} - \gamma_k\right)^2\right]^{-1}. \tag{6.4}$$

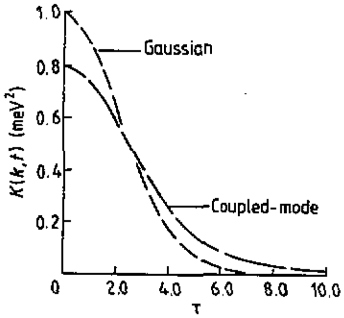

Figure 2. The exact and Gaussian approximation for the memory function are displayed as a function of the reduced time step $\tau = 6.51t$ where t is units of picoseconds (EuO, $T = 1.5\,T_c$ and $k = \pi(1, 1, 1)/2a$). The area under the exact and approximate curves is 0.650 and 0.628 meV, respectively.

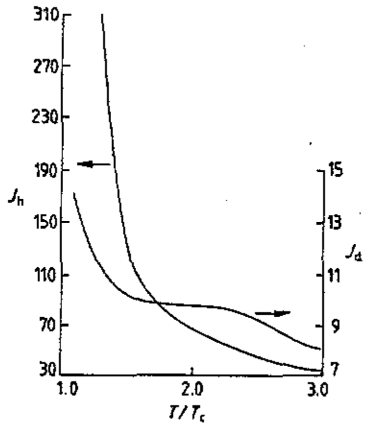

Figure 3. The functions J_d and J_h that determine the temperature dependence of the dipolar and hyperfine contributions to the relaxation rate deep in the paramagnetic phase, equations (6.1)–(6.4), are displayed as a function of the reduced temperature T/T_c. The results are appropriate for a ferromagnetically coupled (nearest-neighbour) FCC material.

The coupling constant Γ_0 is related to the hyperfine constant A, as in (2.8). In (6.2) and (6.4) the function $L(\boldsymbol{k})$ is given for an FCC lattice by (5.4), the Watson integral is defined in (3.12), and γ_k found from (3.7).

Taken at their face value in the critical region these expressions yield λ_d and λ_h proportional to $1/\kappa$ and $(1/\kappa)^3$, respectively. Failure to obtain the correct behaviour, provided in section 4, is hardly surprising, but it serves to remind us that the expressions we are considering apply to the paramagnetic region ($T > T_c$). The failure for $T \to T_c$ has something in common with the breakdown of the so-called conventional (or classical) theory of critical slowing-down of spin fluctuations. For this theory is based on the behaviour of frequency moments, and the erroneous assumption that the relevant transport coefficient is largely independent of temperature. At T_c it predicts a decay rate proportional to k^4, whereas it is firmly established experimentally that the dependence is $k^{5/2}$. The latter result is in accord with theory; indeed the decay rate is $\tilde{K}(\boldsymbol{k}, 0)$ given in (4.3).

The high-temperature limits for λ_d and λ_h are readily obtained with the aid of the following expansion for $I(\mu)$,

$$I(\mu) = \mu[1 + (\mu^2/r) + \ldots]$$

together with ($T \to \infty$)

$$\mu \to 2rJS(S+1)/3T.$$

Thus,

$$\lambda_d \to (\hbar\Gamma/J)(S(S+1)/2\pi)^{1/2} \tag{6.5}$$

and

$$(\lambda_h/\lambda_d) \to 0.396(\Gamma_0/\Gamma) \tag{6.6}$$

in which the factors involved are appropriate to an FCC lattice. The results (6.5) and (6.6) resemble Moriya's estimate of the nuclear magnetic relaxation rate (Moriya 1956), although it must be stressed that our theory is much more sophisticated and reliable.

Values of J_d and J_h are provided in figure 3 for temperatures up to $3T_c$, which for an FCC lattice corresponds to $\mu = 0.44$. Beyond this value J_d and J_h continue to decrease, and reach minimum values at $\mu \sim 0.25$ before achieving their limiting values of 13.92 and 33.10, respectively. Our paramagnetic theory begins to be suspect when $1/\kappa$ and $(1/\kappa)^3$ behaviour for λ_d and λ_h, respectively, becomes dominant, at which point the results in section 4 are used with complete confidence.

7. Conclusions

Relaxations of μSR signals by the dipolar and hyperfine mechanisms are radically different. The hyperfine relaxation rate diverges on approaching the critical temperature where its temperature dependence is $(T/T_c - 1)^{-3\nu/2}$. In contrast to this, the dipolar relaxation rate does not benefit from critical fluctuations. This behaviour reflects, to some extent, the model geometry used for the implanted muon, which is probably appropriate for EuO, where it experiences no net average dipolar field. Similar effects are unlikely in an antiferromagnetically coupled material because the chemical and magnetic structures do not coincide (Lovesey 1992). The temperature dependence of λ_d in MnF_2, for example, must be similar to the NMR linewidth data reported by Heller (1967); cf. De Renzi *et al* (1984).

Predictions regarding critical effects are surely based, since in our formulation we have direct recourse to the theory of dynamic critical phenomena. For the paramagnetic phase our theory is to some extent inchoate. An obvious development is to invest computer time and utilize the full memory function derived from coupled-mode theory, illustrated in figure 2, instead of the convenient parametrization we have exploited. However, the evidence documented in section 5 strongly supports the view that there will be a minimal difference between the two calculations and no new features will emerge. In view of this, the huge cost of computer time is not justified at the present stage in the development of theory and experiment.

We find that the hyperfine relaxation rate increases by about a factor 4 as the temperature is reduced by a half from $T/T_c = 3.0$. On reaching $T/T_c = 1.5$ a very rapid growth sets in as the precursor to the divergence encountered as $T \rightarrow T_c$. Set against this behaviour, the temperature dependence of the dipolar relaxation rate is feeble.

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