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October 1994

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**MUON SPIN RELAXATION IN ANTIFERROMAGNETS:
A STUDY OF RbMnF_3 BASED ON THE COUPLED-
MODE THEORY OF PARAMAGNETIC AND CRITICAL
SPIN FLUCTUATIONS**

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Abstract

The relaxation rate for the depolarization of a positive muon implanted in an antiferromagnetically coupled Heisenberg magnet is studied on the basis of a coupled-mode theory of critical and paramagnetic spin fluctuations, which gives overall an unrivalled account of spin fluctuations. The paper includes the first comprehensive treatment of the dipole field that couples the muon and atomic (spin) magnetic moments. An analytical calculation of the relaxation rate is feasible in the vicinity of the critical temperature, because of the dominant rôle of the critical fluctuations, whereas in general numerical methods are required to calculate the spin response function and the dipole field. In the approach to the critical temperature, the muon relaxation rate increases as the square root of the correlation length. Results for the isotropic antiferromagnet RbMnF_3 demonstrate that, the relaxation rate is not a monotonic function of the temperature, and the magnitude and temperature variation of the relaxation rate depend to a significant degree on the site of the implanted muon. A theoretical framework for the interpretation of the muon relaxation rate is reviewed in a set of appendices.

1. Introduction

It is well established that, the depolarization of positive muons implanted in a magnetic material is a signal which contains useful information, at an atomic level of description, on magnetic fluctuations; for a review of the experimental technique see, for example, Cox (1987). Here, we

report the first comprehensive theoretical investigation of the muon relaxation rate in an antiferromagnetically coupled paramagnet. The dipole field which couples the implanted muon and the atomic moments is treated exactly. The critical and paramagnetic atomic spin fluctuations are described by the coupled-mode theory applied to an isotropic Heisenberg magnet, and the resulting equation for the spin response function is solved numerically for several temperatures. Our report is a significant contribution to the growing development of a theoretical framework for the interpretation of muon spin relaxation experiments (Lovesey 1992, Lovesey et al. 1992). In this instance, the inspiration for the work is provided by an interesting scientific conundrum in a much-studied magnetic salt.

In the history of the development of modern, magnetic critical phenomena, which dates from the late 1960s, experiments on the isotropic antiferromagnetic salt RbMnF_3 , and the coupled-mode theory of critical and paramagnetic spin fluctuations have each played a prominent rôle. The theory is consistent with dynamic scaling arguments and results from the renormalization group method, and it successfully explains a wide variety of experiments, not only on magnetic materials. With regard to the data gathered on the van Hove spin response function of RbMnF_3 it is widely reported to be in accord with the celebrated theories. But, recent work has demonstrated that, in at least one important aspect, coupled-mode theory is not in accord with the data. Our aim is to draw attention to the discrepancy, and, at the same time, provide a feasibility study for a muon spin relaxation experiment that might shed light on the apparent conspicuous shortcoming of the theory.

Let us denote the van Hove spin response function by $S(\mathbf{q}, \omega)$, where \mathbf{q} and ω are the wave vector and frequency variables, respectively. As the temperature is reduced toward the critical temperature the response function gradually bears the signature of critical spin fluctuations at the incipient magnetic ordering wave vector. For an antiferromagnet, the condensed phase is labelled by a wave vector $\mathbf{w} \neq 0$, whereas in a ferromagnet the corresponding wave vector is the zone centre since the chemical and magnetic ordering coincide.

The function $S(\mathbf{q}, \omega)$ can be extracted from the signal measured by inelastic magnetic neutron scattering. Tucciarone et al. (1971) found for RbMnF_3 at T_c that $S(\mathbf{q}, \omega)$ for \mathbf{q} in the vicinity of \mathbf{w} is a three peaked structure; a central ($\omega \sim 0$) component and two distinct peaks equidistant from the

elastic line. An intuitive description for these features is a non-propagating spin diffusion process, and collective (spin-wave) excitations. This picture of the spin fluctuations continues to hold in the paramagnetic phase, where it has been substantiated by computer simulation and neutron scattering experiments (Tucciarone et al. 1971, Evans and Windsor 1973).

Recently, Cuccoli et al. (1994) have shown, in a thorough investigation of coupled-mode theory applied to the standard model for RbMnF_3 , namely the isotropic Heisenberg magnet, that $S(\mathbf{q},\omega)$ in the vicinity of \mathbf{w} does not, at T_c , contain a diffusive contribution. Instead, $S(\mathbf{q},\omega)$ is a narrow function with a minimum at $\omega = 0$. The absence of a central peak in the coupled-mode prediction for the van Hove response function of an antiferromagnet at its critical temperature is apparent in seminal work by Wegner (1969). For some reason, unknown to the authors, the discrepancy between theory and experiment for RbMnF_3 is not mentioned in recent reviews of magnetic critical phenomena (Cowley 1987, Collins 1989, Privman et al. 1990).

In closing this survey of theoretical and experimental investigations of RbMnF_3 at its critical temperature, we remind the reader of the success to date achieved by the theory. To the best of our knowledge, the discrepancy referred to between coupled-mode theory and experimental investigations of magnetic materials is the only really significant one on record.

We turn now to the possible benefit of a muon beam experiment. It might be expected that, the relaxation rate for the depolarization of a positive muon implanted in a magnet can be expressed in terms of $S(\mathbf{q},\omega)$; a theoretical framework for the interpretation of muon relaxation rates is reviewed in appendices A, B and C. Our formula for the relaxation rate, λ , derived from Fermi's Golden Rule for transition rates, contains $S(\mathbf{q},\omega)$ weighted by a geometric structure factor (determined by the contact interaction and the dipole field of the atomic moments) integrated over all wave vectors. The value of ω in this formula is the Larmor precession frequency for the muon, ω_μ . But, this can safely be set to zero since $\omega_\mu = 0.56 \mu\text{eV T}^{-1}$, i.e. the relaxation rate in zero magnetic field is determined by the elastic value of the spin response function, which is fortunately just the component in question in the interpretation of the early neutron scattering experiments. (In a paramagnet the average magnetic field from the atomic moments is zero, of course, and the experiment can be performed without need of an applied magnetic field.)

In this paper we report our findings for λ calculated using coupled-mode theory for the standard model of RbMnF_3 . Before looking at our full calculation, and by way of orientation, we sketch a relatively simple calculation which is appropriate for the critical region.

If the geometric structure factor derived from the contact interaction and the dipole field is set to one side, assuming it is a benign function in the small range of wave vectors that dominate in the description of atomic spin fluctuations, the relaxation rate is simply,

$$\lambda \propto \int d\mathbf{q} S(\mathbf{q}, 0). \quad (1.1)$$

In the so-called Markovian approximation, $S(\mathbf{q}, 0)$ is characterized by a decay rate, $\Gamma(\mathbf{q})$, for the very slow, critical spin fluctuations at the antiferromagnetic Bragg point. For \mathbf{q} measured relative to \mathbf{w} , one finds,

$$\Gamma(q) \sim (\kappa^2 + q^2) / \kappa^{1/2}, \quad (1.2)$$

where κ is the inverse correlation length, and $\kappa = 0$ at T_c . To complete the description of $S(\mathbf{q}, 0)$ in the vicinity of T_c one needs the susceptibility, $\chi(q)$. A reasonable approximation is the form proposed by Ornstein and Zernike, namely (the critical exponent η is set equal to zero),

$$\chi(q) \sim (\kappa^2 + q^2)^{-1}.$$

Combining these factors in the Markovian relation,

$$S(q, 0) \propto \{\chi(q) / \Gamma(q)\}, \quad (1.3)$$

we find,

$$\lambda \propto (1 / \kappa^{1/2}). \quad (1.4)$$

Thus, as $T \rightarrow T_c$ the relaxation rate diverges with a power law temperature dependence $(T - T_c)^{-\nu}$, where ν is the critical exponent for κ .

The foregoing line of reasoning cannot be applied with confidence in the paramagnetic phase because fluctuations with all wave vectors come into play. Hence, outside the critical region, the geometric (dipole) structure factor must be calculated at all points in the Brillouin zone, and in the calculation of λ convoluted with the spin response function. As mentioned already, the latter is calculated from the coupled-mode theory of spin fluctuations, following the study of RbMnF_3 by Cuccoli et al. (1994). Results from this calculation provide reliable values for λ that can be used to study its temperature dependence, and its dependence on the location of the implanted positive muon.

The next section contains a recap of coupled-mode theory applied to the Heisenberg antiferromagnet; we adhere to the notation and formulation used by Cuccoli et al. (1994). In §3 we provide the complete expression for the relaxation rate in terms of the spin response function by drawing on the theoretical framework found in appendices A - C. Results created from the numerical solution of the coupled-mode theory, and the numerical evaluation of the dipole field (appendix D) are presented in §4. Such calculations are computationally very demanding. However, it is the only secure way to assess the temperature dependence of λ . In the immediate vicinity of the critical temperature the calculation of λ simplifies because of the increasing importance of long wavelength fluctuations near the incipient ordering wave vector. An appropriate calculation is given in §5. Our results are discussed in §6.

2. Magnetic Model and Coupled-Mode Theory

Spin operators \mathbf{S}_a are placed on a lattice with N sites labelled by the index a . The spins interact through a Heisenberg interaction of strength J , so the model Hamiltonian is,

$$\mathcal{H} = J \sum_{a,b}^{\text{n.n.}} \mathbf{S}_a \cdot \mathbf{S}_b, \quad (2.1)$$

where the sum is over all nearest-neighbour pairs on the lattice.

We will study the time development of spatial Fourier components $S(\mathbf{k})$ defined through,

$$S_a = (1/N) \sum_{\mathbf{k}} \exp(-i\mathbf{k} \cdot \mathbf{a}) S(\mathbf{k}). \quad (2.2)$$

The isothermal susceptibility is,

$$\chi(\mathbf{k}) = \frac{1}{3} (S(\mathbf{k}), \cdot S(-\mathbf{k})). \quad (2.3)$$

Here, $(,)$ denotes a Kubo relaxation function; for classical variables $(A,B) = (\langle AB \rangle / T)$ where the angular brackets denote a thermal average, and T is the temperature ($k_B = \hbar = 1$). The dynamic properties of (2.1) are studied in terms of the normalized relaxation function,

$$F(\mathbf{k}, t) = \frac{1}{3} (S(\mathbf{k}, t), \cdot S(\mathbf{k})) / \chi(\mathbf{k}), \quad (2.4)$$

where $S(\mathbf{k}, t)$ is the standard Heisenberg time-dependent operator.

The spectrum of neutrons inelastically scattered by spin fluctuations is proportional to,

$$F(\mathbf{k}, \omega) = (1/2\pi) \int_{-\infty}^{\infty} dt \exp(-i\omega t) F(\mathbf{k}, t). \quad (2.5)$$

In the context of neutron scattering, ω is the energy transferred from the primary beam to the spin fluctuations. The concomitant change in the wave vector of the neutrons is $\mathbf{k} = \mathbf{w} + \mathbf{q}$, where \mathbf{w} is an antiferromagnetic ordering wave vector.

Coupled-mode theory is a closed set of equations for $F(\mathbf{k}, t)$. The latter is determined by,

$$\partial_t F(\mathbf{k}, t) = - \int_0^t dt' F(\mathbf{k}, t-t') K(\mathbf{k}, t'), \quad (2.6)$$

and the so-called memory function, $K(\mathbf{k}, t)$, is approximated by,

$$K(\mathbf{k}, t) = (4rJT / N\chi(\mathbf{k})) \sum_{\mathbf{p}} \{\gamma_{\mathbf{p}-\mathbf{k}} - \gamma_{\mathbf{p}}\} F(\mathbf{p}, t) F(\mathbf{p} - \mathbf{k}, t) \chi(\mathbf{p}). \quad (2.7)$$

Here, $\gamma_{\mathbf{k}}$ is a geometric factor that depends on the point group symmetry of the lattice; for a simple cubic lattice with a cell length a ,

$$\gamma_{\mathbf{k}} = \frac{1}{3} (\cos(ak_x) + \cos(ak_y) + \cos(ak_z)) = 1 - \rho^2 k^2 + \dots.$$

The spherical model susceptibility is,

$$\chi(\mathbf{k}) = \{N / 2rJ(\mu_o + \gamma_{\mathbf{k}})\}. \quad (2.8)$$

In the key equations (2.6), (2.7) and (2.8) the wave vector variables \mathbf{p} and \mathbf{k} are general wave vectors in the Brillouin zone for the reciprocal lattice of the chemical structure. The quantity μ_o varies with temperature. In fact, the temperature scale is determined by,

$$(2rJS(S+1)/3T) = (1/N) \sum_{\mathbf{p}} (\mu_o - \gamma_{\mathbf{p}})^{-1} = I(\mu_o), \quad (2.9)$$

in which r is the number of nearest neighbours ($r = 6$, s.c.), and the integral on the right-hand side is the standard extended Watson integral. For a simple cubic lattice, the critical temperature, T_c , satisfies,

$$(4JS(S+1)/T_c) = 1.5164. \quad (2.10)$$

As the critical temperature is approached from above $\mu_o \rightarrow 1$, and the susceptibility has a maximum at the antiferromagnetic ordering wave vector, \mathbf{w} , for which $\gamma_{\mathbf{w}} = -1$. Hence, for $(\mu_o - 1) \ll 1$ we expand the geometric factor $\gamma_{\mathbf{k}}$ in the susceptibility about \mathbf{w} using the small argument expansion, and find an Ornstein-Zernike form,

$$\chi(\mathbf{k}) = \{N / 2rJ\rho^2(\kappa^2 + q^2)\}, \quad (2.11)$$

in which the inverse correlation length, κ , satisfies,

$$\rho^2 \kappa^2 = (\mu_o - 1), \quad (2.12)$$

and \mathbf{q} is measured relative to \mathbf{w} . For the spherical model, (2.12) leads to $\kappa \sim (T - T_c)^\nu$ where the critical exponent $\nu = 1$.

3. Muon Relaxation Rate

A theory of the rate for depolarization of a positive muon implanted in a magnetic material is reviewed in appendices A, B and C. Here we apply the theory to RbMnF_3 which is an isotropic antiferromagnet (Evans and Windsor 1973, Collins 1989). The relaxation rate, λ , averaged over the orientations of the muon polarization with respect to the crystal axes is given by (C.1).

In the present case, the field experienced by the muon, \mathbf{B} , satisfies $\langle \mathbf{B} \rangle = 0$, i.e. there is no steady magnetic field. Moreover, it is assumed to come solely from the dipole-dipole interaction between the muon and atomic magnetic moments. A Cartesian component of the field, labelled by α , is,

$$\begin{aligned} B_\alpha &= -g\mu_B \sum_{a\beta} (1/R_a^3) (3\hat{R}_a^\alpha \hat{R}_a^\beta - \delta_{\alpha\beta}) S_a^\beta \\ &= -(g\mu_B / N) \sum_{\mathbf{k}\beta} D^{\alpha\beta}(\mathbf{k}) S^\beta(\mathbf{k}), \end{aligned} \quad (3.1)$$

where,

$$D^{\alpha\beta}(\mathbf{k}) = \sum_{\mathbf{a}} \exp(i\mathbf{k} \cdot \mathbf{a}) (3\hat{R}_a^\alpha \hat{R}_a^\beta - \delta_{\alpha\beta}) / R_a^3. \quad (3.2)$$

In these expressions, $\mathbf{R}_a = \boldsymbol{\delta} + \mathbf{a}$ where $\{\mathbf{a}\}$ are lattice vectors that define the positions of the atomic (manganese) moments, and $\boldsymbol{\delta}$ defines the position of the implanted muon. The calculation of $D^{\alpha\beta}(\mathbf{k})$, the lattice Fourier transform of the dipole field, is the subject of appendix D.

For an isotropic spin system, like RbMnF_3 ,

$$\langle \mathbf{B}^+ \cdot \mathbf{B}(t) \rangle = \frac{1}{3} (g\mu_B / N)^2 \sum_{\mathbf{k}} \langle \mathbf{S}(-\mathbf{k}) \cdot \mathbf{S}(\mathbf{k}, t) \rangle \sum_{\alpha\beta} |D^{\alpha\beta}(\mathbf{k})|^2. \quad (3.3)$$

Now, the standard definition of the van Hove spin response function, $S(\mathbf{k}, \omega)$, is,

$$S(\mathbf{k}, \omega) = (1 / 6\pi N) \int_{-\infty}^{\infty} dt \exp(-i\omega t) \langle \mathbf{S}(-\mathbf{k}) \cdot \mathbf{S}(\mathbf{k}, t) \rangle, \quad (3.4)$$

whereupon,

$$\lambda = \frac{1}{3} (g_\mu \mu_N / \hbar)^2 \int_{-\infty}^{\infty} dt \langle \mathbf{B}^+ \cdot \mathbf{B}(t) \rangle = \frac{1}{3} (gg_\mu \mu_B \mu_N / \hbar)^2 (2\pi / N) \sum_{\mathbf{k}} S(\mathbf{k}, 0) \sum_{\alpha\beta} |D^{\alpha\beta}(\mathbf{k})|^2. \quad (3.5)$$

The relation between the van Hove response function and the time Fourier transform of the Kubo relaxation function, evaluated for zero frequency, is,

$$S(\mathbf{k}, 0) = \{T \chi(\mathbf{k}) / N\} F(\mathbf{k}, \omega = 0), \quad (3.6)$$

where T is the temperature.

The expression (3.5) relates the muon relaxation rate, λ , to the response function which is usually extracted from the signal measured in inelastic neutron scattering. For various reasons, it is not convenient to couch theoretical developments in terms of $S(\mathbf{k}, \omega)$. Instead, it is usual to employ an auxiliary function which, in the present case, is chosen to be Kubo's relaxation function. The latter is obtained from the coupled-mode theory introduced in §2.

In applications of (3.5) a useful figure is,

$$(2 g_{\mu} \mu_B \mu_N / \hbar a_o^3) = 10.66 \text{ nsec}^{-1}, \quad (3.7)$$

where a_o is the Bohr radius, and we have $g = 2$ for the gyromagnetic factor of the atomic moment.

4. Results from Coupled-Mode Theory

Various properties of RbMnF_3 are gathered in Table (1). Here we report our findings for the relaxation rate calculated for this material using the coupled-mode theory described in §2. A review of coupled-mode theory applied to the isotropic, antiferromagnetically coupled Heisenberg magnet is provided by Cuccoli et al. (1994), and so we focus here on results for λ , and do not dwell on the nature of the spin fluctuations observed in the van Hove spin response function.

Values for λ are given in Table (2) for four temperatures in the interval $(T/T_c) = 1.125$ to 3.55 . These values are based on the coupled-mode theory of the time-dependent fluctuations, and the spherical model of static fluctuations. The latter sets a temperature scale which is reviewed in Table (3) and compared with one determined by neutron diffraction data for κ . A second important feature of the results appearing in Table (2) is the choice of the site for the implanted muon, defined by the vector δ . Results in Table (2) are for the choice $\delta = (\frac{1}{4}, \frac{1}{4}, 0)$, in units of the lattice spacing, a . For a perovskite structure, this choice for δ seems a reasonable first guess.

The increase in λ seen in Table (2) as the temperature approaches T_c was anticipated in the introduction. For T closer to T_c than $T = 1.125T_c$ the calculation sketched in §1 becomes increasingly reliable. At the higher temperatures included in Table (2) the relaxation rate begins to slowly increase.

To obtain an independent estimate of λ at infinite temperature we have approximated $F(\mathbf{k}, \omega = 0)$ by its value obtained from a gaussian model of $F(\mathbf{k}, t)$ with the exact value of the second frequency moment, namely,

$$F(\mathbf{k}, t) = \exp \left\{ -\frac{1}{2} t^2 \omega_0^2 \right\},$$

with,

$$\omega_0^2 = \frac{8J^2}{3} r S(S+1) (1 - \gamma_{\mathbf{k}}).$$

This leads to ($T = \infty$),

$$\lambda = (1 / 6\hbar J) (g g_{\mu} \mu_N \mu_B / \nu_o)^2 \{ \pi S(S+1) / 3r \}^{1/2} \frac{1}{N} \sum_{\mathbf{k}} (1 - \gamma_{\mathbf{k}})^{-1/2} \sum_{\alpha\beta} \left| \nu_o D^{\alpha\beta}(\mathbf{k}) \right|^2.$$

The value of the wave vector sum is 3471.35, and this taken together with data in Table (1) leads to $\lambda (T = \infty) = 0.695 \mu\text{s}^{-1}$. It is satisfying to find this totally independent, realistic estimate of λ is reasonably consistent with the value found at $T = \infty$ from the full theory and shown in Table (2).

Also shown in Table (2) is the ratio of λ to its temperature dependence in the critical region, given by (5.8), namely $(T/T_c a\kappa)^{1/2}$, and κ obtained from the spherical model. The mild variation of this ratio between $T = 1.125T_c$ and $T = 1.25T_c$ is also satisfying, since it is further evidence that data from the full theory is understood.

The final topic in this section is the variation of λ with δ . Table (4) shows the temperature dependence of the ratio of values obtained with two δ 's. One vector, labelled δ_1 , is the same as the one used in the calculations reported in Table (2). The second choice for the muon site, $\delta_2 = (0.4, 0.3, 0.2)$, is a non-symmetric site of the type which could be occupied if the muon distorts the crystal lattice, i.e. should the positive muon not be a passive, environmentally friendly observer of spin fluctuations. (Data in appendix (D) shows that the dipole field varies quite dramatically as δ is moved around the site δ_1). From the data given in Table (4) we conclude that not only is the magnitude of λ a strong function of δ but also the temperature dependence of λ varies with δ . This

latter finding does not bode well for the use of μ SR as a valuable probe of critical and paramagnetic spin fluctuations.

5. Critical values of λ

In the introduction we sketched an argument by which to estimate the muon relaxation rate in terms of the decay rate for critical fluctuations. Let us now fill in the various steps in the argument, which amounts to no more than determining the constant of proportionality in the relation (1.4). We shall then judge if the argument is efficacious by comparing the outcome to results reported in the previous section. The expectation is that the estimate is good very close to the critical temperature.

Combining (3.5) and (3.6),

$$\lambda = (2\pi/3) (g g_\mu \mu_N \mu_B / \hbar)^2 (T/N^2) \sum_{\mathbf{k}} \chi(\mathbf{k}) F(\mathbf{k}, \omega=0) \sum_{\alpha\beta} |D^{\alpha\beta}(\mathbf{k})|^2. \quad (5.1)$$

For a temperature, T , close to T_c the susceptibility $\chi(\mathbf{k})$ is sharply peaked at $\mathbf{k} = \mathbf{w}$, and it is well described by the Ornstein-Zernike form (2.11). Using the latter in the exact relation (5.1), and taking the dipole field evaluated for $\mathbf{k} = \mathbf{w}$ outside the sum,

$$\lambda \sim (\pi/3rJ\rho^2) (g g_\mu \mu_N \mu_B / \hbar)^2 \sum_{\alpha\beta} |D^{\alpha\beta}(\mathbf{w})|^2 (T/N) \sum_{\mathbf{q}} \{F(\mathbf{q} + \mathbf{w}, \omega=0) / (\kappa^2 + q^2)\}. \quad (5.2)$$

Two further steps are taken. First, the Markovian approximation is invoked to describe $F(\mathbf{q} + \mathbf{w}, 0)$, and this means,

$$F(\mathbf{q} + \mathbf{w}, 0) \sim \{1/\pi \Gamma(\mathbf{q})\}. \quad (5.3)$$

Cuccoli et al. (1994) find for the decay rate of the critical spin fluctuations,

$$\Gamma(q) = 0.711(A/\kappa)^{1/2} (q^2 + \kappa^2) / \hbar, \quad (5.4)$$

in which the material constant,

$$A = (4T r J v_0 / \pi^2), \quad (5.5)$$

and r is the number of nearest neighbours and v_0 is the volume of a unit cell. The second step is to replace the sum over the Brillouin zone in (5.2) by an integral over all \mathbf{q} , viz.,

$$(1/N) \sum_{\mathbf{q}} \rightarrow (v_0 / 2\pi^2) \int_0^\infty q^2 dq. \quad (5.6)$$

Assembling the various pieces we arrive, finally, at the result,

$$\lambda \sim (0.019T / r \hbar J \rho^2 v_0) (A\kappa)^{-1/2} (g g_\mu \mu_B \mu_N)^2 \sum_{\alpha\beta} |v_0 D^{\alpha\beta}(\mathbf{w})|^2. \quad (5.7)$$

This can be evaluated for RbMnF_3 with the aid of the material properties gathered in Table (1), and the value for the dipole sum provided in appendix D.

We find for RbMnF_3 , in the limit $T \rightarrow T_c$,

$$\lambda \sim 55.94 (T / T_c a\kappa)^{1/2} \sum_{\alpha\beta} |v_0 D^{\alpha\beta}(\mathbf{w})|^2 \text{ sec}^{-1}. \quad (5.8)$$

Values of the dipole field for various positions of the implanted muon are given in appendix D. In Table (3) we compare values of the temperature dependent factor in (5.8) obtained in one case from the spherical model, and secondly from experimental data reviewed by Als-Nielsen (1974). In the latter work data are well represented by,

$$a\kappa = 2.0 \left(\frac{T}{T_c} - 1 \right)^v,$$

where $v = 0.70$. For T close to T_c the spherical model predicts (s.c. lattice),

$$a\kappa = 3.176 \left(1 - \frac{T_c}{T} \right).$$

This result, and tabulated values of the Watson integral have been used to construct the entries in Table (3).

Returning to (5.8), on using the value,

$$\sum_{\alpha\beta} \left| v_{\alpha} D^{\alpha\beta}(\mathbf{w}) \right|^2 = 3357; \quad \delta = (\frac{1}{4}, \frac{1}{4}, 0),$$

and the spherical model estimate of κ , cf. Table (3), we find $\lambda = 0.34 \mu s^{-1}$ at $T = 1.125T_c$. This is about a factor of 2 smaller than the value listed in Table (2) obtained from the full theory. Our view is that the temperature dependence predicted by (5.8) is soundly based for $T \rightarrow T_c$, even though the estimate of the prefactor is not reliable at the relatively high temperature $T \approx 1.125T_c$. (The Ornstein-Zernike approximation for the susceptibility is not seriously in question. The main sources of the discrepancy are approximation of the wave vector sum by an integral over all wave vectors, and factorization of the dipole field from the spectrum of spin fluctuations.)

6. Discussion

Several of our findings merit comment. First, the depolarization relaxation rate, λ , for a positive muon in RbMnF_3 is not a monotonic function of the temperature. Near the critical temperature λ is proportional to the square-root of the correlation length. Moving away from the critical region λ passes through a minimum and eventually saturates deep in the paramagnetic phase (a similar behaviour has been observed in estimates for a ferromagnetically coupled magnet, Lovesey et al. 1992). We have verified that the entire source of the non-monotonic temperature dependence of λ is the atomic spin dynamics, i.e. a similar temperature dependence is found if the dipole field in (5.1) is replaced by a constant. Secondly, we present incontrovertible evidence that λ depends to a significant degree on the position of the implanted muon. Not only the magnitude but, also the temperature dependence of the relaxation rate depends on the position. Finally, our work shows that a realistic interpretation of λ must include a full treatment of the dipole field (unless, of course, this is completely dominated by a contact interaction, cf. appendix C).

The results presented have been subjected to several checks to ensure accuracy in the various numerical procedures. These checks include: calculations of the dipole field by independent methods; a test of the Brillouin sum integration in (5.1) by calculating the extended Watson integral; a test of $S(\mathbf{k}, 0)$ and λ for sensitivity to the q -mesh size used in the numerical solution of the coupled-mode equations and Brillouin zone integration.

With regard to the nature of spin fluctuations in RbMnF_3 two recent neutron scattering studies are noted. Data reported by Yazaki et al. (1994) for the paramagnetic phase concurs with the first data, published by Tucciarone et al. (1971); it is interesting that the two groups of authors use different experimental methods. On the other hand, an experiment performed on the ordered state by Cox et al. (1989) reveals features that are significantly different from those at T_c reported by Tucciarone et al. (1971). Regarding the conundrum mentioned in §1, the diffusive component of the spectrum reported by Cox et al. (1989) is a very weak feature; weaker than in the spectrum at T_c from Tucciarone et al. (1971), to a degree larger than might reasonably be anticipated from the difference in the thermodynamic states of the samples.

Acknowledgements

Two of us (E.B. and A.C.) thank the Associate Director for Science and his staff at DRAL, Rutherford Appleton Laboratory for support during the course of the reported work. Dr. S. R. Brown and Dr. P. Delmas de Reotier advised us on aspects concerned with the muon beam technique, and Dr. T. G. Perring likewise on the neutron beam technique. Finally, we are grateful to Dr. E. Engdahl for contributions to some of the numerical work.

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Figure Captions

- 1.) Axes that define the magnetic moment of the muon (x' , y' , z') relative to the crystal axes (a , b , c).
- 2.) Representative values of the dipole field $\sum_{\alpha\beta} \left| \nu_{\alpha} D^{\alpha\beta}(\mathbf{w}) \right|^2$ are shown for various values of the muon position, defined by the vector δ . The selected δ 's are in the vicinity of $\delta = a$ ($1/4$, $1/4$, 0) at which the dipole field has the value 3357.0. The other δ that features in the main text is $\delta = a$ (0.4, 0.3, 0.2), and here $\sum_{\alpha\beta} \left| \nu_{\alpha} D^{\alpha\beta}(\mathbf{w}) \right|^2 = 444.7$. All values have been verified in a completely independent check performed by E. Engdahl using a program that is based on the standard Ewald summation method.

Appendix A

The muon relaxation rate λ , which is at the centre of our discussion in the main text, is proportional to the probability per unit time for the implanted muon to make a transition from the state with magnetic quantum number $m = -\frac{1}{2}$ to the state with $m = +\frac{1}{2}$. We denote this probability by W . In the next appendix, W is related, using first-order perturbation theory, to properties of the host material. For the moment, we note the relation $\lambda = 2W$, and provide a justifying argument.

To this end, let N_{\pm} denote the average number of muons in the state labelled by $m = \pm \frac{1}{2}$. The average polarization at time t is,

$$\frac{1}{2}\{N_{-}(t) - N_{+}(t)\} = \frac{1}{2}n(t) = \frac{1}{2}\exp(-\lambda t). \quad (\text{A.1})$$

The final equality defines λ in terms of the average polarization. The expression for $n(t)$ that supports the use of an exponential decay, as in (A.1), is obtained from the standard rate equations for N_{\pm} , namely,

$$\frac{d}{dt}N_{+}(t) = W_{-+}N_{-}(t) - W_{+-}N_{+}(t) = -\frac{d}{dt}N_{-}(t). \quad (\text{A.2})$$

With our chosen definitions, $W \equiv W_{-+}$ and,

$$\frac{dn}{dt} = 2(W_{+-}N_{+} - WN_{-}). \quad (\text{A.3})$$

The steady-state condition for $N_{+}(t)$ provides the relation,

$$W_{+-} = W \exp(\hbar\omega_{\mu} / k_{\text{B}}T),$$

where ω_{μ} is the Larmor frequency of the muon. If $\hbar\omega_{\mu} \ll k_{\text{B}}T$, then (A.3) provides the estimate,

$$\frac{dn}{dt} \sim -2Wn, \quad (\text{A.5})$$

which is consistent with (A.1) if $\lambda = 2W$.

Appendix B

Here, the goal is to obtain a relation for $\lambda = 2W$ in terms of the properties of the material in which the observed muon is implanted.

The muon and host material are described by a Hamiltonian \mathcal{H} which is the sum of a Hamiltonian for two independent systems, \mathcal{H}_0 , the material and the free muon in a steady field, and the Hamiltonian \mathcal{H}_1 which describes their mutual interaction, so,

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_1 \quad . \quad (\text{B.1})$$

In the subsequent argument, it is assumed that \mathcal{H}_1 is weak, so the effect of the muon-material interaction is adequately described by first-order perturbation theory, or, what is the same thing, Fermi's Golden Rule for transition rates.

The wave function, $\psi(t)$, associated with \mathcal{H} satisfies the Schrödinger equation,

$$(i\hbar(\partial/\partial t) - \mathcal{H}_0 - \mathcal{H}_1)\psi(t) = 0. \quad (\text{B.2})$$

Properties described by \mathcal{H}_0 are to be fully taken into account, and to this end $\psi(t)$ is expressed in the form,

$$\psi(t) = \exp(-it\mathcal{H}_0/\hbar)\phi(t),$$

and the equation,

$$i\hbar(\partial/\partial t)\phi(t) = \mathcal{H}_1(t)\phi(t),$$

with,

$$\mathcal{H}_1(t) = \exp(it\mathcal{H}_0/\hbar)\mathcal{H}_1\exp(-it\mathcal{H}_0/\hbar),$$

follows directly from (B.2). The value of $\phi(t)$ correct to first-order in $\mathcal{H}_1(t)$ is,

$$\phi(t) = \phi(0) - (i/\hbar) \int_0^t dt' \mathcal{H}_1(t')\phi(0). \quad (\text{B.3})$$

In this expression, it is assumed that \mathcal{H}_1 is switched on at time $t = 0$.

The second term in (B.3) determines the probability that the system changes on application of \mathcal{H}_1 from the state $\phi(0) = |m, \nu\rangle$ to a state $|m', \nu'\rangle$, where $m = -m' = -\frac{1}{2}$ and the labels $\{\nu\}$ specify states of the material. The required probability is, of course,

$$\begin{aligned} & |\langle m', \nu' | (-i/\hbar) \exp(-it\mathcal{H}_0/\hbar) \int_0^t dt' \mathcal{H}_1(t') |m, \nu\rangle|^2 \\ &= (1/\hbar^2) |\langle m', \nu' | \int_0^t dt' \mathcal{H}_1(t') |m, \nu\rangle|^2. \end{aligned} \quad (\text{B.4})$$

The probability observed in an experiment is the sum of this expression over all final states, and averaged over all initial states. Let us define,

$$p_\nu = \exp(-E_\nu / k_B T) / Z, \quad (\text{B.5})$$

where E_ν is the energy of the ν -th eigenstate of the material, and the partition function Z is obtained from $\sum p_\nu = 1$. The expression for W is then,

$$W = \lim_{t \rightarrow \infty} \frac{1}{t} (1/\hbar^2) \sum_{\nu\nu'} p_\nu |\langle m', \nu' | \int_0^t dt' \mathcal{H}_1(t') |m, \nu\rangle|^2. \quad (\text{B.6})$$

In order to develop this key expression for W for the case of muon relaxation it is necessary to introduce the specific form for \mathcal{H}_1 .

The material creates a fluctuating magnetic field, \mathbf{B} , at the site occupied by the muon (the steady, including applied, magnetic field is contained in \mathcal{H}_0). If the muon spin angular momentum operator is denoted by \mathbf{I} then,

$$\mathcal{H}_1 = -g_\mu \mu_N \mathbf{I} \cdot \mathbf{B}, \quad (\text{B.7})$$

where μ_N is the nuclear Bohr magneton. The quantization axes for the muon are labelled (x', y', z') , in which case ($m = -m' = -\frac{1}{2}$, and $I = \frac{1}{2}$),

$$\begin{aligned} \langle m' | I_{x'} | m \rangle &= \frac{1}{2} \\ \langle m' | I_{y'} | m \rangle &= -i/2 \\ \langle m' | I_{z'} | m \rangle &= 0. \end{aligned}$$

The (x', y', z') and crystal axes (a, b, c) are related by angles α and β depicted in Fig. (1). The matrix elements of \mathbf{I} with respect to the crystal axes are,

$$\begin{aligned}
\langle m' | I_a | m \rangle &= \frac{1}{2} (\sin \beta - i \cos \alpha \cos \beta) = \frac{1}{2} \xi \\
\langle m' | I_b | m \rangle &= -\frac{1}{2} (\cos \beta + i \cos \alpha \sin \beta) = \frac{1}{2} \eta \\
\langle m' | I_c | m \rangle &= (i/2) \sin \alpha = \frac{1}{2} \zeta,
\end{aligned} \tag{B.8}$$

in which the parameters ξ , η and ζ are convenient for subsequent use. The free muon spin dynamics is described by its Larmor frequency ω_μ which is determined by the steady magnetic field in \mathcal{H}_0 . From (B.7) and (B.8) we then have,

$$\begin{aligned}
\langle m', v' | \mathcal{H}_1(t) | m, v \rangle &= -g_\mu \mu_N \langle m' | \mathbf{I}(t) | m \rangle \cdot \langle v' | \mathbf{B}(t) | v \rangle \\
&= -\frac{1}{2} g_\mu \mu_N \exp(-it\omega_\mu) \{ \xi \langle v' | B_a(t) | v \rangle + \eta \langle v' | B_b(t) | v \rangle + \zeta \langle v' | B_c(t) | v \rangle \},
\end{aligned} \tag{B.9}$$

in which,

$$\mathbf{B}(t) = \exp(it\mathcal{H}_m / \hbar) \mathbf{B} \exp(-it\mathcal{H}_m / \hbar),$$

and \mathcal{H}_m is the Hamiltonian which described the material in which the muon is implanted, with, of course,

$$\mathcal{H}_m |v\rangle = E_v |v\rangle. \tag{B.10}$$

Inserting (B.9) in the key formula (B.6), the latter reduces to the master formula,

$$W = (g_\mu \mu_N / 2\hbar)^2 \int_{-\infty}^{\infty} dt \exp(-it\omega_\mu) \langle Y^+ Y(t) \rangle, \tag{B.11}$$

with,

$$Y(t) = \xi B_a(t) + \eta B_b(t) + \zeta B_c(t). \tag{B.12}$$

The components B_a , B_b and B_c expressed in terms of B_x , B_y and B_z that refer to the appropriate coordinate system for the magnetic ions in the material are,

$$\begin{aligned}
B_a &= B_x \sin \phi + B_y \cos \theta \cos \phi + B_z \sin \theta \cos \phi \\
B_b &= -B_x \cos \phi + B_y \cos \theta \sin \phi + B_z \sin \theta \sin \phi \\
B_c &= -B_y \sin \theta + B_z \cos \theta.
\end{aligned} \tag{B.13}$$

The angles θ and ϕ relate the (x, y, z) and (a, b, c) axes as depicted in Fig. (1) with α and β replaced by θ and ϕ , respectively. The result (B.11) together with (B.12) is the basis of the work reported in the text. Several special cases that merit attention are provided in the next appendix.

Appendix C

In some instances, such as a polycrystal or a domain structure with perfect cubic symmetry, it is appropriate to average the result (B.11) for W over the orientations of the muon polarization with respect to the crystal axes. In practice this means averaging the products of the ξ , η and ζ over the angles α and β ; denoting the spatial average by a horizontal bar, all off-diagonal products vanish, e.g. $\overline{\xi\zeta} = 0$, and all diagonal products have the same value, namely, $2/3$. The corresponding value of the relaxation rate is,

$$\lambda = 2\overline{W} = \frac{1}{3}(g_\mu \mu_N / \hbar)^2 \int_{-\infty}^{\infty} dt \exp(-it\omega_\mu) \langle \mathbf{B}^+ \cdot \mathbf{B}(t) \rangle. \quad (\text{C.1})$$

Thus, in this special case, the relaxation rate is simply the time Fourier transform of the scalar product correlation function of the fluctuating magnetic field at the site of the implanted muon.

A second special case is that of a pure contact interaction, i.e.,

$$\mathbf{B} = \sum_{\ell} A(\ell) \mathbf{S}(\ell), \quad (\text{C.2})$$

where \mathbf{S} are spin angular momentum operators associated with magnetic ions at sites defined by the set of indices $\{\ell\}$, and $\{A\}$ are interaction parameters. In an ordered magnetic material, the dominant contribution to λ if $\omega_\mu \sim 0$ arises from longitudinal spin fluctuations, i.e. the components parallel to the magnetic axis of quantization, denoted here by the z -axis. For this case one keeps just the z -component of \mathbf{B} in (C.2). From (B.11), (B.12), (B.13) and (C.2) we arrive at the result, on setting $\omega_\mu = 0$,

$$\lambda = 2W = \frac{1}{2}(g_\mu \mu_N / \hbar)^2 |\xi \sin \theta \cos \phi + \eta \sin \theta \sin \phi + \zeta \cos \theta|^2 \quad (\text{C.3})$$

$$\sum_{\ell, \ell'} \int_{-\infty}^{\infty} dt A^*(\ell) A(\ell') \langle S^z(\ell) S^z(\ell', t) \rangle.$$

Here, ξ , η and ζ are defined in (B.8). Inspection of these factors in conjunction with the form of the geometric prefactor in (C.3) shows that this prefactor vanishes if the muon polarization and magnetic quantization axes coincide, i.e. $\alpha = \theta$ and $\beta = \phi$. So, as expected for this special geometry, there is

no relaxation due to longitudinal spin fluctuations mediated by the contact interaction. However, it is to be noted that, for a uniaxial magnet at the critical temperature it is only these fluctuations that can generate a critical enhancement of the relaxation rate.

In genral, the muon is not depolarized if \mathbf{B} is parallel to the muon polarization; this intuitively obvious result, of course, is borne out by the general expression (B.11).

Appendix D: Dipole Sums

The methods described here have been used to obtain our results, quoted in the main text, for the spatial Fourier transform of the dipole operator, $D^{\alpha\beta}(\mathbf{k})$. It is well known that the lattice sums involved can converge slowly and some care is needed in the numerical analysis to obtain reliable results. Both the methods used exploit the fact that the implanted muon does not occupy a lattice site. This fact results in an incoherence in the lattice Fourier transform which can be used to good effect in providing rapid convergence of the lattice sums. The methods used differ in several respects from the conventional Ewald summation technique, e.g. no auxiliary functions and introduced.

The first method we describe makes use of formulae in which the convergence induced by the incoherence is explicit. The second method is a straightforward spatial Fourier transform which is coded to produce a fast and accurate computational method. Results from the two, distinct numerical methods are in complete agreement.

To illustrate the first method we consider the calculation of the diagonal components of $D^{\alpha\beta}(\mathbf{k})$. We employ two identities:

$$(3R_{\alpha}R_{\beta} - R^2) / R^5 = \nabla_{\alpha} \nabla_{\beta} (1 / R),$$

and,

(D.1)

$$(1 / R) = (1 / \sqrt{\pi}) \int_0^{\infty} d\xi \xi^{-1/2} \exp(-\xi R^2).$$

In the calculation of $D^{\alpha\beta}(\mathbf{k})$, the position variable $\mathbf{R} = (R_1, R_2, R_3)$ is,

$$\mathbf{R} = \mathbf{a} + \delta,$$

where $\{\mathbf{a}\}$ defines the magnetic lattice, and δ defines the position of the implanted muon. Since δ does not coincide with a position on the lattice the dipole operator is well defined, and non-singular.

For a simple cubic lattice, with a unit lattice spacing $\mathbf{a} = (\ell_1, \ell_2, \ell_3)$ and the integers $\{\ell_i\}$ take all values. Thus,

$$\sum_{\mathbf{a}} (1/R) \exp(\mathbf{i}\mathbf{k} \cdot \mathbf{a}) = (1/\sqrt{\pi}) \int_0^\infty d\xi \xi^{-1/2} \sum_{\{\ell\}} \exp\{ik_1\ell_1 + ik_2\ell_2 + ik_3\ell_3 - \xi R^2\} \quad (\text{D.2})$$

$$= \sqrt{\pi} \int_0^\infty d\xi \xi^{-3/2} \sum_{\ell_3} \exp\{-\xi a + ik_3\ell_3\} \sum_{q_1 q_2} \exp\{-i\beta_1\delta_1 - i\beta_2\delta_2 - b/\xi\},$$

where $a = (\ell_3 + \delta_3)^2$, $\beta_i = (2\pi q_i + k_i)$, and ,

$$b = \frac{1}{4} (\beta_1^2 + \beta_2^2).$$

In arriving at the second equality in (D.2), use is made of the identity,

$$\sum_{l=-\infty}^{\infty} \delta(x-l) = \sum_{q=-\infty}^{\infty} \exp(2\pi i q x), \quad (\text{D.3})$$

to re-write the sums over ℓ_1 and ℓ_2 ; this amounts to use of the Poisson formula. We have chosen to re-write these two sums, rather than some other combination of $\{\ell_i\}$, with a view to calculating $D^{33}(\mathbf{k})$ by an application of (D.1). In terms of the variables used in the second equality in (D.2), the derivatives in (D.1) with respect to R_i are easily cast as derivatives with respect to the variable a , viz.,

$$(\partial^2 / \partial R^2) = 2(\partial / \partial a) + 4a(\partial^2 / \partial a^2). \quad (\text{D.4})$$

The integral in (D.2) is standard,

$$\int_0^\infty d\xi \xi^{-3/2} \exp(-a\xi - b/\xi) = (\pi/b)^{1/2} \exp(-2(ab)^{1/2}), \quad (\text{D.5})$$

for $a, b > 0$. Using this result together with (D.4) one arrives from (D.2) at the desired expression,

$$D^{33}(\mathbf{k}) = \sum_{\ell} \exp(\mathbf{i}\mathbf{k} \cdot \ell) (3R_z^2 - R^2) / R^5 \quad (\text{D.6})$$

$$= 4\pi \sum_{\ell} \exp(ik_3\ell) \sum_{qq'} \exp(-i\delta_1\beta_1 - i\delta_2\beta_2') b^{1/2} \exp(-2(a_3b)^{1/2}),$$

where the integers ℓ , q and q' take all values. The remaining two diagonal components of $D^{\alpha\alpha}(\mathbf{k})$ are obtained from (D.6) by a suitable choice of the component δ and \mathbf{k} in $\beta_\alpha = (2\pi q + k_\alpha)$, $a_\alpha = (\ell + \delta_\alpha)^2$, and b .

A similar bag of tricks can be applied with advantage to the off-diagonal components of $D^{\alpha\beta}(\mathbf{k})$. We find, for example,

$$D^{12}(\mathbf{k}) = -\pi \sum_{\ell} \exp(ik_3\ell) \sum_{qq'} \beta_1\beta_2' \exp(-i\delta_1\beta_1 - i\delta_2\beta_2') b^{-1/2} \exp(-2(a_\alpha b)^{1/2}). \quad (\text{D.7})$$

The remaining off-diagonal components are obtained from (D.7) by a suitable choice of the components of δ and \mathbf{k} . The same notation is used in (D.6) and (D.7).

In the second method we perform a direct (straightforward) spatial Fourier transform of the dipole operator as given in (3.2). The code was set up in such a way as to monitor the convergence of the lattice sums.

Each sum value is obtained from separate contributions in which a subsum over all points lying on a cubical surface in lattice space is accumulated. In almost all cases considered the subsums showed a rapid decrease, eventually falling below a preset value. For a few combinations of wave vector and muon position vector the convergence is slow and the lattice summation has to be taken to a large limit. However, changing the critical values by a small amount immediately improves the speed of convergence, while the result for the corresponding dipole field hardly varies. This demonstrates the benign behaviour of the quantity under study. In all cases, the two numerical methods described are in complete agreement.

As an indication of the strong dependence of the dipole field on the muon position vector δ , we display in Fig. (2) the results for several sites around the most likely position of the muon, (0.25, 0.25, 0.), for one selected wave vector, $\mathbf{k} = (1, 1, 1)$, the incipient ordering wave vector. The values of the dipole field for other wave vectors will be of the same order since the variation across the Brillouin zone is rather weak. The value at $\delta = (0.25, 0.25, 0.)$ is 3357 and is not given in Fig. 2.

Table (1): Properties of RbMnF₃

Quantity	Symbol	Value
Chemical unit cell dimension	a	4.24Å
Critical temperature ⁽¹⁾	T_c	83K
Nearest neighbour exchange interaction	J	0.28 meV
Non-universal material constant in the damping rate	A	361.0 meV ² Å ³
Superlattice wave vector	\mathbf{w}	$(\pi/a)(1,1,1)$
Geometrical factor ($\alpha = x,y,z$)	$\gamma_{\mathbf{k}}$	$\frac{1}{3} \sum_{\alpha} \cos(ak_{\alpha})$
Number of nearest neighbours	r	6
Spin magnitude	S	5/2

- (1) The quoted value of J and the spherical model relation (2.9) produce a critical temperature = 78K. Evans and Windsor (1973) report the value $J = 0.28 \pm 0.03$ meV obtained from an analysis of the spin wave dispersion. All our results are provided as a function of the reduced temperature (T/T_c), and A is calculated with $T_c = 78$ K.

Table (2)

Values of the relaxation rate (in units of μs^{-1}) as a function of temperature calculated for a muon at $\delta = (0.25, 0.25, 0)$.

T/T_c	$\lambda(\mu\text{s}^{-1})$	$\lambda(\mu\text{s}^{-1})/\zeta^{(b)}$
1.125	0.661	0.363
1.25	0.573	0.399
2.0	0.584	0.530
3.55	0.653	0.591
∞	0.805 ^(a)	

(a) We are grateful to E. Engdahl for calculating this result. The gaussian model described in §4 gives the result 0.695.

(b) The quantity $\zeta = (T/T_c a \kappa)^{1/2}$ is the temperature variation of λ in the critical region, cf. (5.8); values of κ are calculated from the spherical model of spin correlations.

Table (3)

Spherical model and measured temperature scales.

$\left(\frac{T}{T_c} - 1\right)$	$(T/T_c \ a\kappa)^{1/2}$	
	Spherical model	Measured values
0.001	17.76	7.94
0.01	5.67	3.56
0.10	1.98	1.66
1.0	1.10	1.0

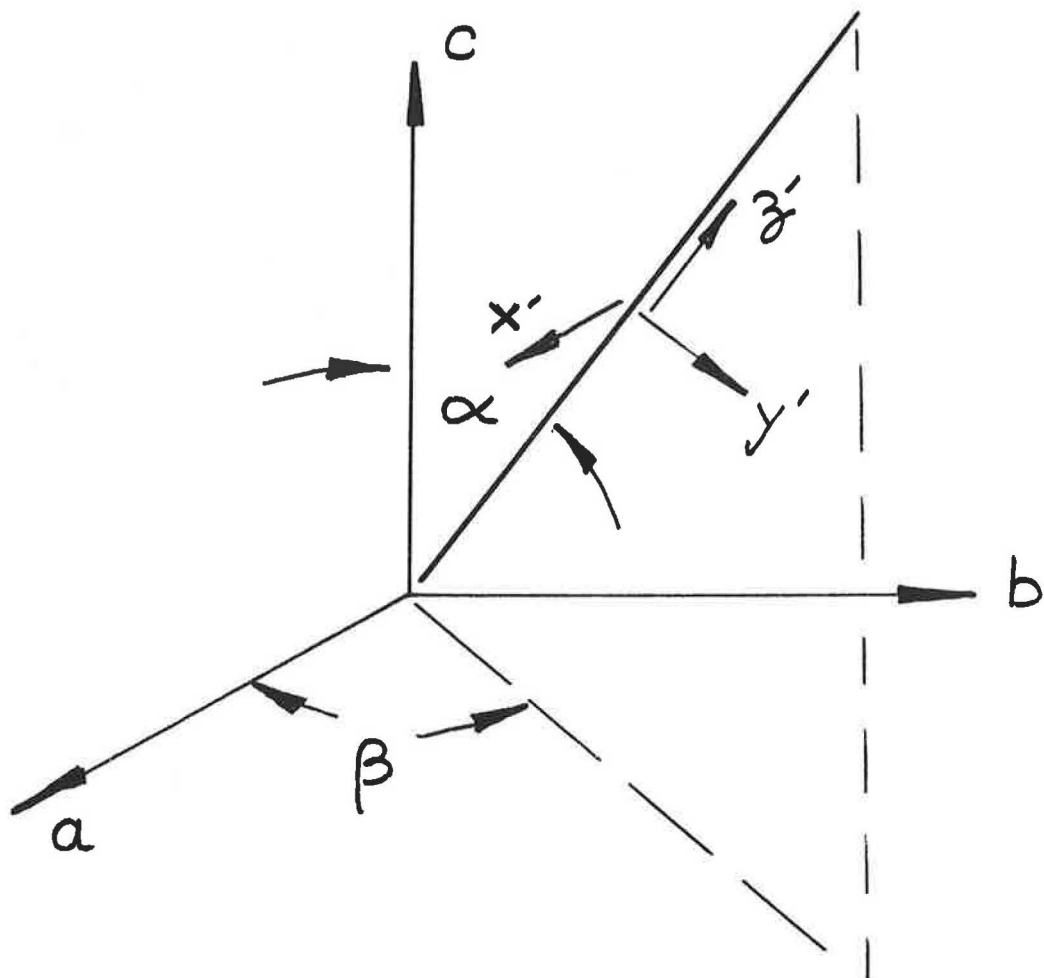
The measured values for RbMnF_3 are obtained from the relation $a\kappa = 2.0 \left(\frac{T}{T_c} - 1\right)^{0.70}$; after Als-Nielsen (1974).

Table (4)

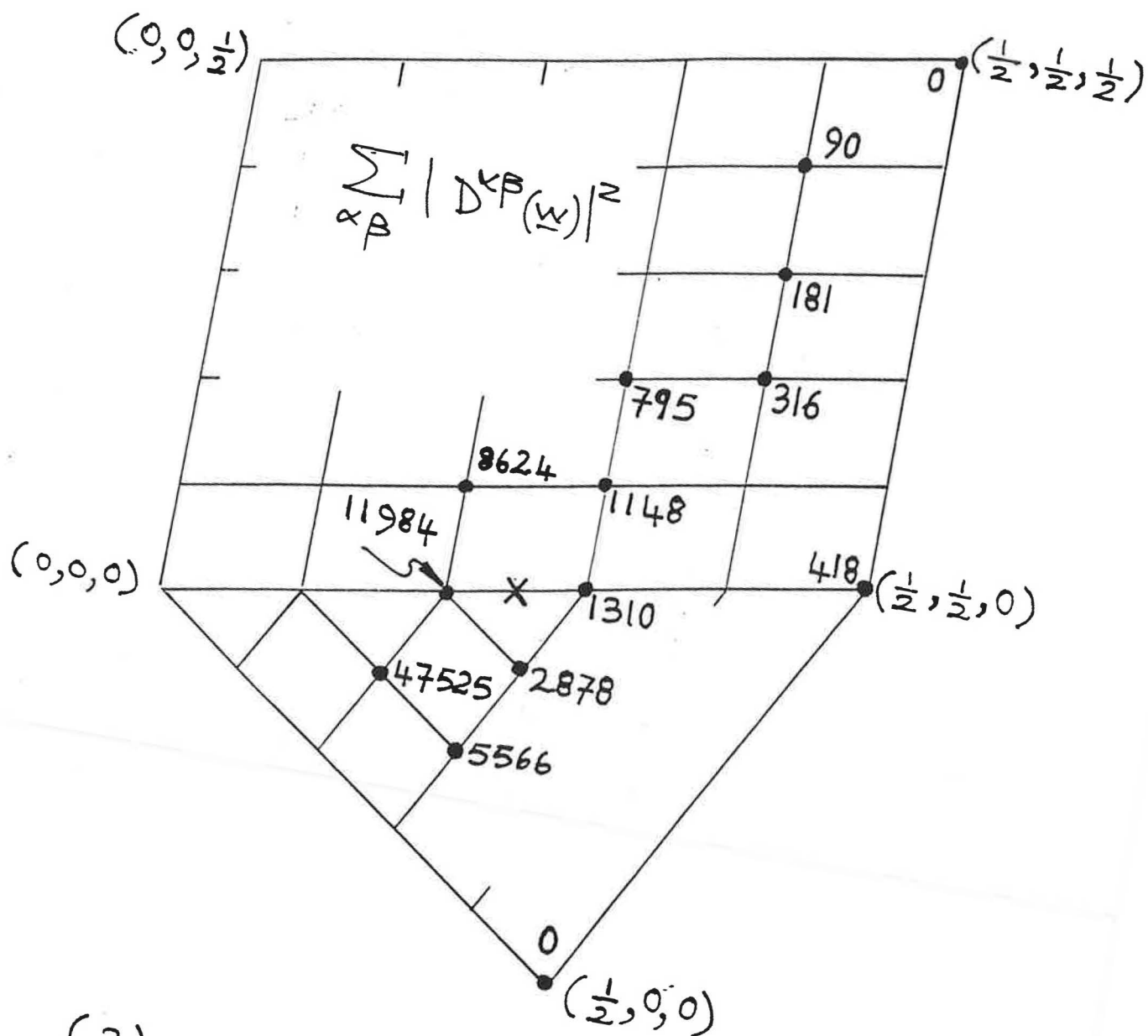
Ratio of the relaxation rates for two sites of the implanted muon, $\delta_1 = (0.25, 0.25, 0)$ and $\delta_2 = (0.4, 0.3, 0.2)$

T/T_c	$\lambda(2)/\lambda(1)$
1.0	0.132 ^(a)
1.125	0.122
1.25	0.119
2.0	0.113
3.55	0.109
∞	0.109 ^(b)

- (a) The listed value is the ratio of the dipole field $\sum_{\alpha\beta} |\nu_\alpha D^{\alpha\beta}(\mathbf{w})|^2$ for the two sites which gives the ratio of the relaxation rates evaluated within the theory described in §5 which is appropriate very close to T_c , cf. fig. 2.
- (b) The value is derived from the gaussian model for $F(k, \omega)$ described in §4 which is appropriate for infinite temperature.



(1)



(2)

