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EXPLAINING μ^+ SPIN-LATTICE RELAXATION IN MnF_2, BELOW T_N, BY SCATTERING FUNCTION

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A relation between the spin-lattice relaxation rate $(1/T_1)$ of positive muons and the scattering function $S_\perp^T(q,\omega)$ is derived for the antiferromagnet MnF_2. We find good agreement between previous μSR measurements of $1/T_1$ and our calculation using neutron scattering measurements of $S_\perp^T(q,\omega)$.

It is very well known [1] that linewidth $(\delta\nu)$ and spin-lattice relaxation of local probes like NMR and μSR, are related to the sum of spin fluctuations of all wavevectors q; namely

$$1/T_1 \text{ or } \delta\nu \sim \frac{T}{\omega_s}\int \chi''(q,\omega_s)\mathrm{d}^3q, \text{ and } \chi''(q,\omega) \propto S^\xi(q,\omega)/\langle n+1\rangle. \qquad (1)$$

In this expression: ω_s is the precession frequency, ξ is the correlation length, χ'' is the imaginary part of the susceptibility, and $\langle n+1\rangle = (1-\exp(\hbar\omega/k_BT))^{-1}$; $S(q,\omega)$ is the magnetic scattering function which is measurable directly by neutron scattering. Whether most of the contribution to the relaxation rate comes from longitudinal $S_{\|}(q,\omega_s)$ or transverse $S_\perp(q,\omega_s)$ fluctuation is determined by the geometry of the probe site. In studies of the critical region of magnetic materials, Eq. 1 is mostly used to deduce critical exponents from $\delta\nu$ or $1/T_1$ measurements [2–4]. $1/T_1$ measurements are especially sensitive to dynamical effects since they are independent of static field inhomogeneities. This feature of spin-lattice relaxation becomes even more important below T_N where there are static fields from electronic sources.

In this work we show that the muon $1/T_1$ in the ordered state of an antiferromagnet can be accounted for by the sum of host spin fluctuations, taken over all q vectors. The μSR technique is a very sensitive way of measuring spin-lattice relaxation times; values of $1/T_1$ up to 10^2 μsec^{-1} can be obtained. In comparison, NMR is limited to relaxation rate smaller then 10^{-3} μsec^{-1}. This, in turn, allows one to extend μSR measurements to temperatures closer to the critical temperature. In fact, in the compound we discuss here (MnF_2), $1/T_1$ of ^{19}F-NMR in the ordered state reaches its sensitivity limit before the temperature reaches T_N [5]. Using data from previous neutron scattering and μSR experiments we achieve good agreement between our model and the data within and outside the critical region.

For this discussion we first adopt Moriya's [1] ideas to the case of positive muon in MnF_2. The starting point is his expression for the spin-lattice relaxation rate

$$1/T_1 = (1/2\hbar^2)\int_{-\infty}^{\infty} \mathrm{d}t\cos(\omega_s t)\sum_{j,j'}\sum_{\nu,\nu'}[(F_{pj})_{x\nu} + i(F_{pj})_{y\nu}] \times[(F_{pj'})_{x\nu'} + i(F_{pj'})_{y\nu'}]\langle\{\delta S_{j\nu}(t)\delta S_{j'\nu'}(0)\}\rangle. \tag{2}$$

which is derived from the generalized spin-spin interaction

$$\vec{I}_p\cdot \overleftrightarrow{F}_{pj}\cdot\vec{S}_j, \tag{3}$$

between the probe moment $\vec{I}_p$ and the ionic spins $\vec{S}_j$. In this expression $\omega_s = \gamma_\mu B_s$ is the Zeeman frequency due to the average static local field B_s at the muon site, and

$$\delta\vec{S}_j = \vec{S}_j - \langle\vec{S}_j\rangle; \tag{4}$$

$\langle\ \rangle$ denotes the thermal average and $\{AB\} = (AB + BA)/2$. The direction $\hat{z}$ is taken along the average local field at the muon site, which in MnF_2 coincides with the easy $\hat{c}$ axis [6]. The index j of the tensor $\overleftrightarrow{F}_{pj}$ runs over neighbors of the muon while $p = 1$ in a time differential μSR experiment since there is only one muon at a time in the sample. The indices ν and ν' represent x, y, and z. This expression is valid in the so-called narrowing limit where the dynamical local field ω_d is much smaller then the inverse of the correlation time $1/\tau_c$ [7]. This condition is related to the spin lattice relaxation time via $1/T_1 \sim \omega_d^2\tau_c$. The dynamical field ω_d near the critical temperature can be evaluated from the static field ω_s at zero temperature.

It is customary to introduce at this point the Fourier transform in space of the ionic spin variable. In MnF_2 there is only one magnetic ion per unit cell, and we can define

$$\vec{S}_k = N^{1/2}\sum_j \vec{S}_j\exp(i\vec{k}\cdot\vec{R}_j), \tag{5}$$

where $\vec{k}$ is a wavevector and N the number of magnetic ions. The correlation function between the j th and the j' th ionic spins can then be expressed as follows:

$$\langle\{\delta S_{j\nu}(t)\delta S_{j'\nu'}(0)\}\rangle = N^{-1}\sum_k \langle\{\delta S_{k\nu}(t)\delta S_{-k\nu'}(0)\}\rangle \exp[-i\vec{k}\cdot(\vec{R}_j - \vec{R}'_j)]. \quad (6)$$

This equation is then used to write $1/T_1$ of Eq. 2 in terms of the correlation function as

$$1/T_1 = (1/2N\hbar^2)\int_{-\infty}^{\infty} \mathrm{d}t \cos(\omega_s t)\sum_k \sum_{\nu\nu'} D_{\nu\nu'}(k)\langle\{\delta S_{k\nu}(t)\delta S_{-k\nu'}(0)\}\rangle \quad (7)$$

where

$$D_{\nu\nu'}(k) = \sum_{j,j'}[(F_{pj})_{x\nu} + i(F_{pj})_{y\nu}] \quad (8)$$
$$\times[(F_{pj'})_{x\nu'} + i(F_{pj'})_{y\nu'}]\exp[-i\vec{k}\cdot(\vec{R}_j - \vec{R}'_j)].$$

Most of the contribution to the sum in Eq. 7 is from $\vec{k}$ near the staggered magnetization wave vector $\vec{k}_0$ of the antiferromagnetic order. We therefore shift the origin of the Brillouin zone to the point $\vec{k}_0$, and define $\vec{q} = \vec{k} - \vec{k}_0$. In the most general case, both longitudinal and transverse fluctuations contribute to $1/T_1$. However, as we will show, longitudinal fluctuations do not contribute to $1/T_1$ in the case of MnF_2. We therefore conclude that in this case $D_{33}(q) = 0$. Since $\overleftrightarrow{F}_{pj}$ is very short range, as a function of j, and since the correlation function peaks at $q = 0$ [2], we can approximate $D_{\nu\nu'}(q)$ in the sum of Eq. 7 as independent of q for small q. Similar approximations were used by De Renzi *et al.* [2] in their work on μSR linewidth at $T > T_N$ of MnF_2.

Defining the scattering function

$$S_\perp(\vec{q},\omega) = \int_{-\infty}^{\infty} \mathrm{d}t \cos(\omega t)\langle\{\delta S_{q+}(t)\delta S_{-q-}(0)\}\rangle \quad (9)$$

and using the symmetry of MnF_2 under the interchange of x and y we arrive at

$$1/T_1 = D\int S_\perp(q,\omega_s)\mathrm{d}^3q \quad (10)$$

where D is a constant independent of temperature, and the sum over q is replaced by an integral. The temperature dependence of the right hand side enters in two places: one is the temperature dependence of the scattering function $S_\perp^T(q,\omega)$, and the other is the temperature dependence of the static local field $\omega_s(T)$.

μSR measurements in MnF_2 were preformed by Uemura *et al.*, and are fully described in Ref. [6]. They used two experimental configurations both in zero external field: (a) the transverse configuration (TC) in which the initial muon polarization was perpendicular to the $\hat{c}$ axis, and (b) the longitudinal configuration (LC) in which the initial polarization was parallel to $\hat{c}$. In TC they measured the precession frequency of the muon moment in the static local field at $T < T_N$, and thus obtained $\omega_s(T)$. Two frequencies were found in the μSR spectra and assigned to two different muon sites. In LC they measured the relaxation rate of the muon polarization in temperatures both above and below T_N. Here again two relaxation time-scales were observed. The fast relaxation was attributed to muons at the high field site. We will be concerned here only with the spin lattice relaxation of muons in the site with the higher field since the data is less scarce. In Fig. 1a we show the fast $1/T_1$ as a function of temperature [6]. The relaxation rate of the muon at the Néel temperature is ~ 14 μsec^{-1}; the precession frequency at $T = 0$ in the TC is $\omega_s = 2\pi \times 1.3$ GHz. Therefore $\omega_s \tau_c \sim 10^{-3}$, and we can safely say that Eq. 2 is valid in this case. It is obvious from the figure that at temperatures higher then the Néel temperature $1/T_1$ is independent of T. This is a clear evidence that $1/T_1$ is independent of the longitudinal spin fluctuation since these fluctuations are known to undergo critical slowing down as discussed in [6,8]. The same conclusion was drawn from the study at $T > T_N$ of De Renzi *et al.* [2].

Schulhof *et al.* [8] measured the scattering function $S_\perp^T(q,\omega)$ in MnF_2 near T_N by neutron scatting. They showed that near T_N, $S_\perp^T(q,\omega)$ could be approximated by

$$S_T(q,\omega) \propto \frac{1}{\kappa_\perp^2 + q^{*2}} \left(\frac{\Gamma_\perp}{\Gamma_\perp^2 + (\omega - \omega_0)^2} + \frac{\Gamma_\perp}{\Gamma_\perp^2 + (\omega + \omega_0)^2} \right) + \mathrm{O}((\omega_s/T)^2) \tag{11}$$

where ω_0 and $\Gamma_\perp$ are functions of the temperature and are given by

$$\begin{aligned} \omega_0(T,q) &= a_0(T) + b_0(T)(q^*)^2, \\ \Gamma_\perp(T,q) &= a_\perp(T) + b_\perp(T)(q^*)^2, \\ q*^2 &= q_x^2 + q_y^2 + (c/a)^2 q_z^2, \end{aligned} \tag{12}$$

where a and c are the lattice parameters. From their data we derive $b_0(T)$, $a_\perp(T)$, and $b_\perp(T)$, as shown in Fig. 1b. The solid lines represent fits to power laws made just for interpolating the neutron data. The functions describing these lines are shown in the figures. The gap energy $a_0(T) =$

$1.36(1 - T/T_N)^{0.37}$, and $\kappa_\perp = 0.054(5)$ Å^{-1} was explicitly given by the authors [8]. This form of $S^T_\perp(q,\omega)$, and $\omega_s(T)$ is then used to numerically integrate

$$\int S^T_\perp(q,\omega_s(T))\mathrm{d}^3q \tag{13}$$

at various temperatures. The integral was preformed in a cube, and the range of integration was limited by the available neutron data ($q < 0.3$ Å^{-1}). The temperature dependence of this integral was then scaled by a factor D so that both sides of Eq. 10 agree at T_N. The result of the computed $1/T_1$ is shown in Fig. 1a by the solid line. The calculated line agrees very well with the measured data. The broken line in this figure represents a fit to a power law. It is clear that such a power law fits the data very poorly. However,the agreement with Eq. 10 extends over a wide temperature range.

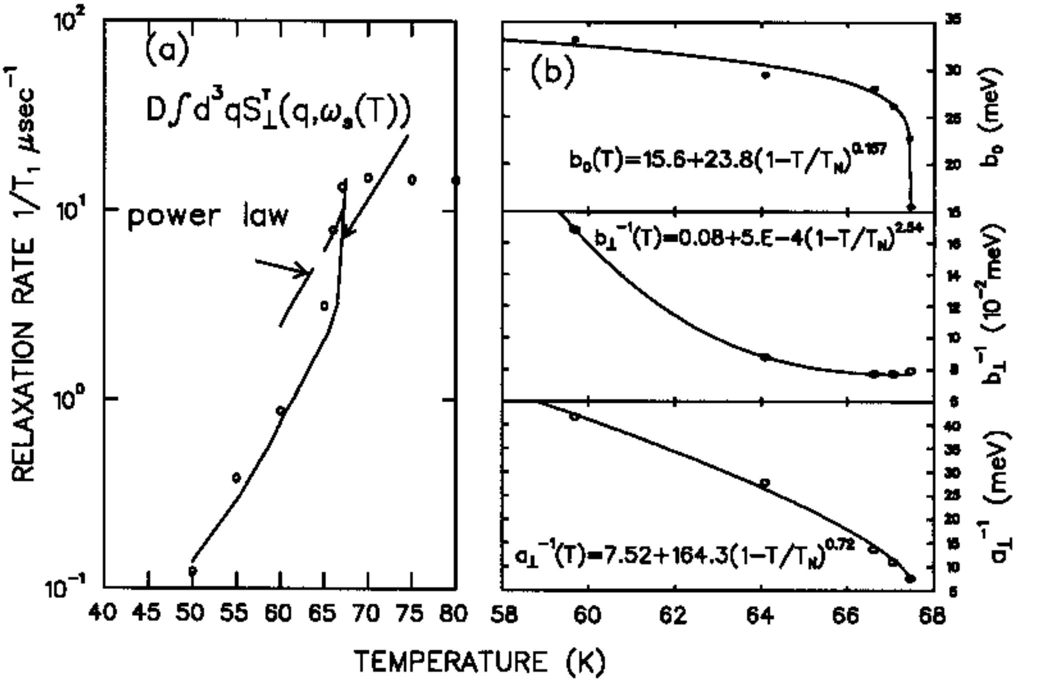

Fig. 1. (a) μSR relaxation rate measurements compared with model calculation and power law as described in the text. (b) Fitted parameters from neutron scattering measurements.

In conclusion, we showed that by using Eq. 10, one can relate dynamical spin fluctuations of MnF_2 to the scattering function in a wide range of temperatures below T_N. The scattering function, which is directly related to correlation functions, is a frequently calculated quantity, both numerically and analytically. Thus, Eq. 10 provides us with a powerful tool to check theories of magnetic interaction, since we can obtain more information than merely critical exponents.

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