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Theory of zero-field muon-spin relaxation in simple magnetic systems

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A theory is developed for the calculation of the distribution function of random fields for classical simple magnets (Ising, XY, and Heisenberg) in $D(=1,2,3)$ dimensions. The zero-field, muon-spin-relaxation functions for these simple magnetic systems are calculated with the distribution function of random field. The relaxation functions $G(t)$ are different from the prediction of the Kubo-Toyabe theory, except for three-dimensional Heisenberg system. The long-time limit $G(\infty)=G(t\rightarrow\infty)$ is 0 for the Ising magnet in 1, 2, and 3 dimensions. For the XY magnet, $G(\infty)=0.5$ for $D=1$ and 2, but $G(\infty)=0.356$ for $D=3$. For the Heisenberg magnet, $G(\infty)=0.1168$, 0.4608, and $\frac{1}{3}$ for $D=1$, 2, and 3. For Ising magnets, there are oscillations in $G(t)$ when the magnetization is nonzero. The effects of vortices as extra magnetic sources in the two-dimensional XY model are addressed and the possibility of detecting the Kosterlitz-Thouless transition by use of the zero-field, muon-spin-relaxation technique is suggested. The theoretical predictions are calculated numerically. The limitations, extensions of the formalism, as well as the applications of the theory to real magnetic systems, are discussed.

I. INTRODUCTION

Muon spin relaxation[1] (μSR) has become a widely used experimental technique to study the local magnetic environment experienced by muons, as well as the diffusion and localization process of muons. In zero external field, the μSR technique (ZF-μSR) is particularly important in the study of systems where external field perturbations are highly relevant, such as in spin-glass and two-dimensional XY systems.[2] This is especially true for systems near a phase-transition point. The muon spin dynamics is governed by, among other things, the local random field of the system. Since muons (with a lifetime $\tau_\mu\simeq 2.2$ μsec) decay into neutrinos and positrons, with the positrons emitted preferentially along the muon's spin direction, one can obtain the muon-spin-relaxation function $G(t)$ from a histogram of the decay particles. Furthermore, since a muon produced from pion decay has its polarization collinear with the muon momentum, μSR is very useful for the study of magnetism. For example, we can study one-dimensional magnets with the muon initial polarization along the chain direction by having the muon beam directed along the chain. Similarly, by having the beam parallel to the XY plane, one can study the relaxation of the muon spin in a two-dimensional magnet. In this paper, we address the problem of ZF-μSR in simple magnets in one, two, and three dimensions.

Almost all ZF-μSR works are based on the Kubo-Toyabe theory.[3] Since the spirit of our calculations follow the Kubo-Toyabe theory, we first list the basic assumptions made in this theory.

(i) The muon depolarization field is static. (The dynamical effects are taken care of by the strong-collision model of Kubo[4] and its generalizations[5,6] including muon trapping and escaping processes. The dipolar coupling of the muon spin and the nuclear spin can actually change the direction of both.)

(ii) The probability distribution of the depolarization field is isotropic. The actual calculations in the Kubo-Toyabe theory assumes further that this distribution is Gaussian. It is found that as long as the isotropy criterion and the next assumption are met, the ZF-μSR relaxation function at long time recovers to the value of $\frac{1}{3}$. The Gaussian distribution is only a simplification.

(iii) The environment of the muon can be described by a classical three-dimensional Heisenberg system. This is a crucial assumption as it implies a random field with three components, which is responsible for $G(t)\rightarrow\frac{1}{3}$ as $t\rightarrow\infty$. We find different results for other simple magnetic environments.

Our work is an attempt to provide a general framework for various simple magnetic models much in the same way as the Kubo-Toyabe theory does for three-dimensional Heisenberg magnetic systems. We have formulated the calculation so that modifications due to the special magnetic lattice structure and anisotropy in the spin Hamiltonian can be incorporated. The formalism is applied for model Ising, XY, and Heisenberg magnets in one, two, and three dimensions. We retain assumption (i) above, as well as the classical nature of the interaction between moments. The isotropy and Gaussian nature of the random-field distribution are not assumed.

We find that there are general formulas relating the ZF-μSR relaxation function for Ising, XY, and Heisenburg magnets to the fourier transform $A(\mathbf{p})$ of the distribution function $W(\mathbf{B})$ of the random field. We also find that there is a general form for $A(\mathbf{p})$ for a given magnetic interaction decreasing with separation r like $r^{-\beta}$ in a D-dimensional system. Namely, at small p, $A(\mathbf{p})\sim e^{-c_1 p^2}$ and for large p, $A(\mathbf{p})\sim e^{-c_2 p^{D/\beta}}$ with c_1 and c_2 positive constants. The details of the calculations of $G(t)$ for vari-

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ous model magnets in D dimension are carried out numerically. We also present an analysis of the effects of magnetic vortices on $G(t)$ for the two-dimensional XY model.

In Sec. II, the probability distribution of the random fields is calculated. The details are given in Appendixes A and B. In Sec. III, the sources of the random fields are discussed. Fairly general results on the fourier transform of the field distribution are obtained. In Sec. IV, the formalism of Secs. II and III is applied to calculate $G(t)$ for Ising, XY, and Heisenberg models for D dimensions $(D=1,2,3)$. Finally, in Sec. V, applications to model magnets, limitations, and possible extensions of the theory are discussed.

II. PROBABILITY DISTRIBUTION OF THE RANDOM FIELDS

For a muon precessing in a given magnetic field $\mathbf{B}$, the spin vector $\mathbf{s}(=\frac{1}{2}\boldsymbol{\sigma})$ evolves as

$$\boldsymbol{\sigma}(t)=\boldsymbol{\sigma}_{\parallel}(0)+\boldsymbol{\sigma}_{\perp}(0)\cos(\gamma_\mu B t)\ , \tag{1}$$

where γ_μ $(=2\pi\times 13.554\times 10^3$ $\mathrm{Oe}^{-1}\,\mathrm{sec}^{-1})$ is the gyromagnetic ratio of the muon. $\boldsymbol{\sigma}_{\parallel}(0)$ and $\boldsymbol{\sigma}_{\perp}(0)$ denote the parallel and perpendicular component with respect to the magnetic field at time 0. In the case of a distribution $W(\mathbf{B})$ of magnetic field, the muon-spin-time correlation function $\boldsymbol{\sigma}(t)\cdot\boldsymbol{\sigma}(0)$ is averaged over $W(\mathbf{B})$ to give the muon-spin-relaxation function $G(t)=\langle\boldsymbol{\sigma}(t)\cdot\boldsymbol{\sigma}(0)\rangle$. In this section, we discuss some general features of the relation of $W(\mathbf{B})$ with $G(t)$ and leave the details of the calculation on $W(\mathbf{B})$ to the following sections.

We first discuss the simple case of isotropic random-field distribution, $W(\mathbf{B})=W(|\mathbf{B}|)$.[7,8] There are three possibilities.

(i) Ising random field, $\mathbf{B}=B\hat{\mathbf{z}}$,

$$G_I^0(t)=\sigma_z^2(0)+[1-\sigma_z^2(0)]\langle\cos(B\tau)\rangle\ . \tag{2}$$

(ii) XY random field, $\mathbf{B}=B_x\hat{\mathbf{x}}+B_y\hat{\mathbf{y}}$,

$$G_{XY}^0(t)=\langle\sigma_{\parallel}^2(0)\rangle+\langle\sigma_{\perp}^2(0)\cos(B\tau)\rangle$$
$$=\tfrac{1}{2}+\tfrac{1}{2}\langle\cos(B\tau)\rangle\ . \tag{3}$$

(iii) Heisenberg random field, $\mathbf{B}=B_x\hat{\mathbf{x}}+B_y\hat{\mathbf{y}}+B_z\hat{\mathbf{z}}$,

$$G_H^0(t)=\tfrac{1}{3}+\tfrac{2}{3}\langle\cos(B\tau)\rangle\ . \tag{4}$$

Here the superscript zero reminds us that $W(\mathbf{B})$ is isotropic and $\tau=\gamma_\mu t$. As $\tau\to\infty$, we have $\langle\cos(B\tau)\rangle\to 0$ and $G_I^0(\infty)=\sigma_z^2(0)$, $G_{XY}^0(\infty)=\frac{1}{2}$, and $G_H^0(\infty)=\frac{1}{3}$. Note that the classical theory of Kubo and Toyabe[3] deals with Heisenberg spin with a Gaussian random-field distribution, which is a special case considered here. The average $\langle\cos(B\tau)\rangle$ can be calculated conveniently by means of the fourier transform $A(\mathbf{p})$ of $W(\mathbf{B})$. The calculations of $\langle\cos(B\tau)\rangle$ are presented in Appendix A.

For anisotropic random-field distribution, we consider $A(\mathbf{p})=\exp(-ap_\perp^2-bp_\parallel^2)$ with $\mathbf{p}=p_\parallel\hat{\mathbf{z}}+p_\perp\hat{\mathbf{x}}$. Then, the random-field distribution is

$$W^A(\mathbf{B})=\frac{1}{8\pi^{3/2}}\frac{1}{a\sqrt{b}}\exp\left[-\frac{B_\perp^2}{4a}-\frac{B_\parallel^2}{4b}\right]\ , \tag{5}$$

where $\mathbf{B}=B_\parallel\hat{\mathbf{z}}+B_\perp(\hat{\mathbf{x}}\cos\phi_B+\hat{\mathbf{y}}\sin\phi_B)$. Such an anisotropic random-field distribution arises from the Gaussian approximation for the Heisenberg spin model in one, two, and three dimensions and for the three-dimensional XY spin model. The relaxation is given by (Appendix B)

$$G^A(t)=\left\langle\frac{B_\parallel^2}{B^2}\right\rangle+\left\langle\frac{B_\perp^2}{B^2}\cos(B\tau)\right\rangle$$
$$=\frac{1}{8a\sqrt{b}}\int_0^1\left[x^2z^{3/2}+(1-x^2)\left[z^{3/2}-\frac{\tau^2z^{5/2}}{2}\right]\times e^{-(\tau^2/4)z}\right]dx \tag{6}$$

with

$$z^{-1}=\frac{1}{4a}+\left[\frac{1}{4a}-\frac{1}{4b}\right]x^2\ . \tag{7}$$

The long-time limit of $G^A(t)$ is no longer the same as the isotropic case. $G^A(\infty)$ is a function of a and b.

III. SOURCES OF THE RANDOM FIELDS

In this section a model calculation of $W(\mathbf{B})$ is presented for a given magnetic interaction between the muon and the magnetic sources. We assume that the small distance behavior of the magnetic field $\mathbf{b}$ of the source is $h_0\hat{\mathbf{h}}_\alpha r^\alpha$ and for far field $(r>r_0)$, $\mathbf{b}$ is given by $h_0\hat{\mathbf{h}}_\beta r^{-\beta}$, where $\hat{\mathbf{h}}_\alpha$ and $\hat{\mathbf{h}}_\beta$ are function of the $\hat{\mathbf{r}}$ and $\hat{\boldsymbol{\mu}}$ with $\mathbf{r}$ being the position of the source of magnetic moment $\boldsymbol{\mu}$. Here r_0 is a short distance cutoff parameter. Following Chandrasekhar,[9] we write $W(\mathbf{B})$ as the average of $\delta(\mathbf{B}-\sum\mathbf{b}(\mathbf{r},\boldsymbol{\mu}))$ over all possible position $\mathbf{r}$ and orientation $\boldsymbol{\mu}$ of the sources. Here the sum in the δ functional is over all the sources. By introducing the Fourier representation of δ, we have

$$W(\mathbf{B})=\overline{\langle\delta(\mathbf{B}-\sum\mathbf{b}(\mathbf{r},\boldsymbol{\mu}))\rangle}$$
$$=\int\frac{d^3\mathbf{p}}{(2\pi)^3}e^{i\mathbf{p}\cdot\mathbf{B}}\overline{\langle\prod e^{-i\mathbf{p}\cdot\mathbf{b}}\rangle}\ . \tag{8}$$

We distinguish here the positional average, denoted by a bar, and the magnetic moment orientational average, denoted by $\langle\ \rangle$. By assuming that the sources are evenly distributed over the volume with density N/V and that the orientational average over $\boldsymbol{\mu}$ is positionally independent (a mean-field assumption), we can write

$$A(\mathbf{p})=Y(\mathbf{p})^N\ . \tag{9}$$

Here, $Y(\mathbf{p})$ is the single-particle average,

$$Y(\mathbf{p})=\overline{\langle e^{-i\mathbf{p}\cdot\mathbf{b}}\rangle}$$
$$=1-\left\langle\frac{1}{V}\int d^D\mathbf{r}(1-e^{-i\mathbf{p}\cdot\mathbf{b}(\mathbf{r},\boldsymbol{\mu})})\right\rangle\ , \tag{10}$$

where D is the dimensionality of space. Since V is large, we can exponentiate equation (9) to obtain

$$A(\mathbf{p})=\exp\left[-\frac{N}{V}\langle g(k_\alpha\hat{\mathbf{h}}_\alpha)+g(k_\beta\hat{\mathbf{h}}_\beta)\rangle\right]\ , \tag{11}$$

where these g functions are given by

$$g(k_\alpha \hat{\mathbf{h}}_\alpha)=\frac{D}{\alpha}V_0 k_\alpha^{-D/\alpha} \times \int_0^{k_\alpha} d\omega\, \omega^{-1+D/\alpha}(1-[e^{-i\omega\hat{\mathbf{p}}\cdot\hat{\mathbf{h}}_\alpha}]) \quad (12)$$

and

$$g(k_\beta \hat{\mathbf{h}}_\beta)=\frac{D}{\beta}V_0 k_\beta^{D/\beta} \times \int_0^{k_\beta} d\omega\, \omega^{-1-D/\beta}(1-[e^{-i\omega\hat{\mathbf{p}}\cdot\hat{\mathbf{h}}_\beta}]) \; . \quad (13)$$

Here $\Omega_D r_0^D$ is the volume V_0 of a D-dimensional sphere of radius r_0. The [ ] bracket refers to the angular average $\int d^D\hat{\mathbf{r}}/(D\Omega_D)$ of its argument. Here $k_\alpha = ph_0 r_0^\alpha$ and $k_\beta = ph_0 r_0^{-\beta}$.

There are two types of standard approximations used in the evaluation of these g functions. The first one, assuming a small-p expansion, leads to the Gaussian approximation. For small x,

$$\frac{g(x\hat{\mathbf{h}}_\gamma)}{V_0}=\frac{ix}{\gamma+\epsilon D}\left[\int \frac{d^D\hat{\mathbf{r}}}{\Omega_D}\hat{\mathbf{p}}\cdot\hat{\mathbf{h}}_\gamma\right] + \frac{x^2}{2\gamma+\epsilon D}\left[\int \frac{d^D\hat{\mathbf{r}}}{\Omega_D}(\hat{\mathbf{p}}\cdot\hat{\mathbf{h}}_\gamma)^2\right]+O(x^3) \; , \quad (14)$$

where $\epsilon=+1$ for $\gamma=\alpha$ and $\epsilon=-1$ for $\gamma=\beta$. One sees that, unless from some symmetry consideration the angular average of $\hat{\mathbf{p}}\cdot\hat{\mathbf{h}}_\gamma$ vanishes, the function g is not well defined for $\beta=D$. And in general, the function g is divergent for $\beta \le D/2$. The Gaussian approximation is widely used because the small p limit of $A(\mathbf{p})$ corresponds to the large B limit of $W(\mathbf{B})$.

A second approximation concerns the beginning of $W(\mathbf{B})$, or the large p approximation. Traditionally, for dipole-dipole interaction in three dimensions such a large p approximation of $A(\mathbf{p})$ is the Lorentzian approximation. We can write for large x

$$\frac{g(x\hat{\mathbf{h}}_\alpha)}{V_0}\simeq 1-\frac{D}{\alpha}x^{-D/\alpha}C_1 \quad (15)$$

and

$$\frac{g(x\hat{\mathbf{h}}_\beta)}{V_0}\simeq \frac{D}{\beta}x^{D/\beta}C_2-1 \; , \quad (16)$$

where C_1 and C_2 are given by

$$C_1=\int_0^\infty d\omega\, \omega^{-1+D/\alpha}[e^{-i\omega\hat{\mathbf{p}}\cdot\hat{\mathbf{h}}_\alpha}] \quad (17)$$

and

$$C_2=\int_0^\infty d\omega\, \omega^{-1-D/\beta}(1-[e^{-i\omega\hat{\mathbf{p}}\cdot\hat{\mathbf{h}}_\beta}]) \; . \quad (18)$$

Here, C_1 and C_2 are assumed convergent. This large p approximation gives $A(p)\sim \exp(-cp^{D/\beta})$ where c is some constant. For the special case where $D=\beta$, the corresponding $W(B)$ is Lorentzian.

In magnetic systems describable with localized moment, the magnetic field is dipolar in origin so that $\beta=3$. From Eq. (13), as long as $0<D/\beta\le 1$, the integral is convergent and one can extend the upper limit to infinity. This corresponds to the case of taking the cutoff parameter r_0 to zero and we can ignore the short-range-interaction ($r<r_0$) contribution $g(k\hat{\mathbf{h}}_\alpha)$. Under such circumstances ($0<D/\beta\le 1$), the expression for $A(\mathbf{p})$ is

$$A(\mathbf{p})\sim \exp\left[-\frac{N}{V}Cp^{D/\beta}\right] \quad (19)$$

with

$$C=\Omega_D\frac{D}{\beta}h_0^{D/\beta}\int_0^\infty d\omega\, \omega^{-1-D/\beta}(1-[e^{-i\omega\hat{\mathbf{p}}\cdot\hat{\mathbf{h}}_\beta}]) \; . \quad (20)$$

In the numerical calculations presented in the following sections, we assume $r_0 \sim a$, the lattice constant, and the interaction is dipolar in nature down to $r=r_0$. From an experimental point of view, r_0 is a fitting parameter.

IV. EXAMPLES

The formalism developed in Secs. II and III is applied to different magnetic systems describable with localized moments. We assume the magnetic field is dipolar in origin and the lattice constant is the length unit. In the case of $D=1$ and 2, the microscopic field b has exactly only one component for the Ising spin system and exactly two components for the XY spin system. (See Appendix C.) This is because the term $3(\boldsymbol{\mu}\cdot\hat{\mathbf{r}})\hat{\mathbf{r}}$ in the dipolar interaction is exactly zero for Ising spins and XY spins in one and two dimensions. Therefore, in $D=1$ and 2, the notions of Ising field from Ising spin and XY field from XY spin using the dipolar interaction are workable concepts, whereas for three dimensions, there are in general three components of microscopic field for both Ising spin and XY spin, and one can use the anisotropic field distribution for calculation. These simplifications allow a more general discussion of different magnetic systems, at the price of not having a particular lattice structure and consequently no mention of the location of the muon. The applicability of these general examples is similar to that of Kubo-Toyabe. Specific magnetic systems, such as an Ising chain with some interchain coupling, must be worked out in detail with due respect to lattice structure and muon site. Encouraged by the similarity of the Kubo-Toyabe theory with the detailed few-body quantum-mechanical calculations of Celio and Meier,[10] we hope that these examples will provide a good approximation to more detailed quantum mechanical calculations on a given lattice.

A. One-dimensional Ising magnet

Consider a chain of length $L=Na$ of Ising spins $\boldsymbol{\mu}=\pm\mu\hat{\mathbf{x}}$ located along the $\hat{\mathbf{z}}$ axis. Assume that N_+ of them are up ($\boldsymbol{\mu}=\mu\hat{\mathbf{x}}$) and N_- of them are down, ($\boldsymbol{\mu}=-\mu\hat{\mathbf{x}}$). Then $A(\mathbf{p})=A_+(\mathbf{p})A_-(\mathbf{p})$ where $A_\pm(\mathbf{p})$ are given by

$$A_\pm(\rho)=\exp\left[-\frac{r_0}{3}n_\pm \xi^{1/3}\int_0^\xi dy\, y^{-4/3}(1-e^{\pm iy})\right] \; , \quad (21)$$

so that according to Eq. (A1),

$$\langle \cos(B\tau)\rangle = \{ \exp[-\kappa g_R(\xi)]\} \cos[\kappa m g_I(\xi)] \ . \quad (22)$$

Here $n_\pm = N_\pm/(N_+ + N_-)$, $\xi = \mu\tau/(r_0)^3$, m is the magnetization given by $m=(n_+ - n_-)$, and $\kappa = \frac{1}{3}(r_0/a)$. g_R and g_I are the real and imaginary part of $g(\xi)$ given by

$$g(\xi) = \xi^{1/3} \int_0^\xi dy\, y^{-4/3}(1-e^{-iy}) \ . \quad (23)$$

If the muon beam has its polarization transverse to the $\hat{\mathbf{z}}$ axis, then $\sigma_z(0)=0$ and $G_I^0(t)$ is given by Eq. (21), which is plotted in Fig. 1. By expanding for small τ for $m=0$ case, we obtain the second moment of the Ising relaxation function to be

$$\Delta^2 = \frac{1}{10}\frac{r_0}{a}\frac{\mu^2}{r_0^6}\gamma_\mu^2 \ , \quad (24)$$

where Δ^2 is defined to be the coefficient of t^2 term in the small-t expansion of G_I^0,

$$G_I^0(t, m=0) = 1 - \Delta^2 t^2 + \cdots \ . \quad (25)$$

From Fig. 1, the oscillatory term in $G_I^0(t)$ due to the nonvanishing of m is damped out by the exponential decay. Since the one-dimensional Ising model never exhibits ferromagnetism, m is zero in the absence of external field.

B. Two-dimensional Ising magnet

The spins are defined on the xy plane with moments $\mu = \pm\mu\hat{\mathbf{z}}$. For $\sigma_z(0)=0$, the relaxation $G_I^0(t)$ is given by

$$\langle \cos(B\tau)\rangle = \{ \exp[-\kappa g_R(\xi\Delta t)]\} \times \cos[\kappa m g_I(\xi \Delta t)] \ , \quad (26)$$

with $\kappa = (2\pi/3)(r_0/a)^2$ and $\xi = \mu\tau/r_0^3$. Here the second moment is given by

$$\Delta^2 = \frac{\pi}{4}\frac{\mu^2\gamma_\mu^2}{r_0^6}\left[\frac{r_0}{a}\right]^2 \ . \quad (27)$$

g_R and g_I are the real and imaginary part of

$$g(\xi) = \xi^{2/3}\int_0^\xi dy\, y^{-5/3}(1-e^{-iy}) \ . \quad (28)$$

Since the two-dimensional Ising model exhibits ferromagnetism at finite temperature, the magnetization m is nonzero below the critical temperature T_c. We can use Eq. (26) to probe the magnetization as the system approaches T_c. A graph of Eq. (26) for different m is shown in Fig. 2.

C. Three-dimensional Ising magnet

If we assume that we have Ising field from Ising spins in $D=3$, then we can repeat the analysis for $D=1$ and 2. However, in general, one would have to use the anisotropic field distribution for the calculation of the relaxation function. Assuming $\sigma_z(0)=0$, we have $G_I^0(t)$ given by

$$\langle \cos(B\tau)\rangle = \{ \exp[\kappa g_R(\xi)]\} \cos[\kappa m g_I(\xi)] \quad (29)$$

with $\kappa = (4\pi/3)(r_0/a)^3$, $\xi = \mu\tau/r_0^3$, and the second moment Δ is given by

$$\Delta^2 = \frac{8\pi}{15}\left[\frac{r_0}{a}\right]^3\left[\frac{\mu^2\gamma_\mu^2}{r_0^6}\right] . \quad (30)$$

$g_R(\xi)$ and $g_I(\xi)$ are the real and imaginary part of

$$g(\xi) = \xi\int_0^\xi d\omega\, \omega^{-2}\left[1 - \int_0^1 dx\, e^{-i(3x^2-1)\omega}\right] . \quad (31)$$

In this calculation of $G_I^0(t)$, we have assumed that the transverse fields (B_x, B_y) are zero.

A plot of the relaxation is shown in Fig. 3. The relaxation is damped out in roughly one unit of second moment. A comparison of Figs. 1, 2, and 3 on the Ising relaxation function indicates that the damping (in units of second moment) is bigger as the spatial dimensionality increases.

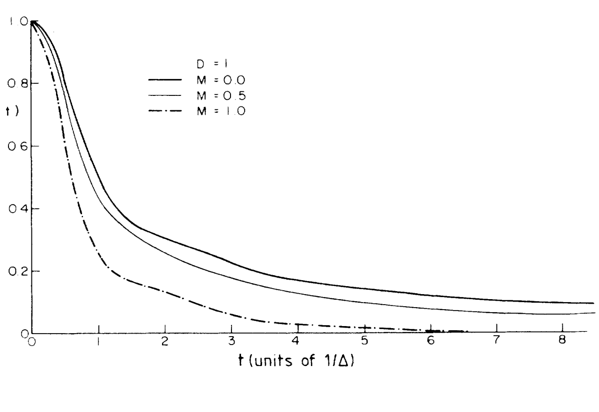

FIG. 1. The relaxation function $G_I^0(t)$ vs time t in unit $(1/\Delta)$ for the one-dimensional Ising model, with different magnetization $m=0.0$, 0.5, 1.0. [$\sigma_z(0)=0$.]

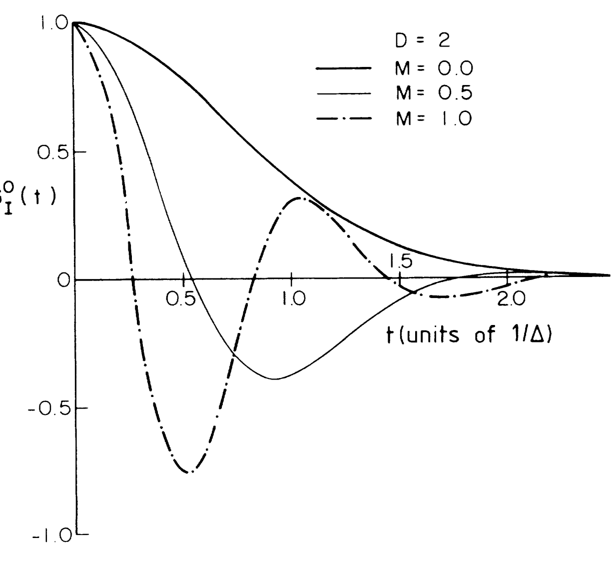

FIG. 2. The relaxation function $G_I^0(t)$ vs time t in unit $(1/\Delta)$ for the two-dimensional Ising model, with different magnetization $m=0.0$, 0.5, 1.0. [$\sigma z(0)=0$.]

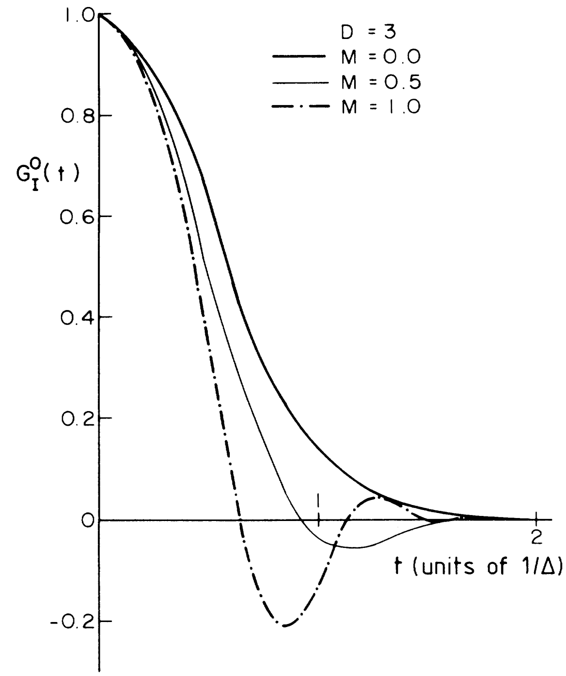

FIG. 3. The relaxation function $G_I^0(t)$ vs time t in unit $(1/\Delta)$ for the three-dimensional Ising model, with different magnetization $m=0{,}0$, 0,5, 1.0. $[\sigma_z(0)=0.]$

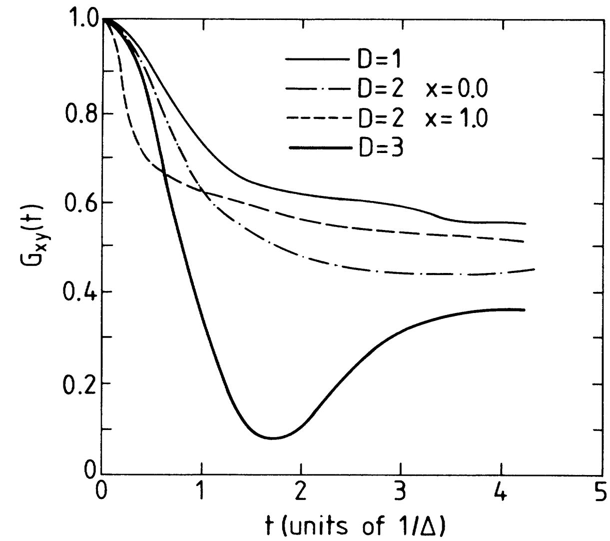

FIG. 4. The relaxation function $G_{xy}(t)$ versus time t for the XY model in one, two, and three dimensions. The unit of time is $1/\Delta$ with Δ given by Eqs. (35) or (43) for the XY model in one and two dimensions. For the three-dimensional XY model, computed with an anisotropic distribution, the unit of time is $1/\Delta$ with $\Delta=\sqrt{2\kappa}$ and κ given by Eq. (45). For the two-dimensional XY model, $X=0$ and 1, corresponding to the case of vortex free and vortex saturated systems.

D. One-dimensional XY magnet

The spins are free rotators on the xy plane but are located along the z axis. $[\boldsymbol{\mu}=\mu(\cos\phi_\mu\hat{\mathbf{x}}+\sin\phi_\mu\hat{\mathbf{y}}).]$ The relaxation function G_{xy}^0 is given by

$$G_{xy}^0(t)=\tfrac{1}{2}+\tfrac{1}{2}\int_0^{\pi/2}d\theta\sin\theta\left[A(p)+\frac{d}{dp}A(p)\right], \tag{32}$$

with $p=\tau\sin\theta$. $A(p)$ is given by

$$A(p)=\exp\left[-\frac{1}{3}\left[\frac{r_0}{a}\right]g(\xi)\right], \tag{33}$$

where $\xi=\mu p/r_0^3$ and $g(\xi)$ is given by

$$g(\xi)=\xi^{1/3}\int_0^{\xi}dy\,y^{-4/3}[1-J_0(y)]\ . \tag{34}$$

The result of $G_{xy}^0(t)$ is calculated numerically, shown in Fig. 4. The second moment Δ is given by

$$\Delta^2=\frac{1}{20}\left[\frac{r_0}{a}\right]\left[\frac{\mu\gamma_\mu}{r_0^3}\right]^2 . \tag{35}$$

Since there is no phase transition in the model, one does not expect any oscillatory behavior in the relaxation. However, correlated region of spins will modify the form of G_{XY}^0, computed here assuming a random angular distribution of the moment $\boldsymbol{\mu}(\phi_\mu)$.

E. Two-dimensional XY model

The spins are free rotators on the xy plane and are also located on a two-dimensional square lattice defined on the xy plane. $(\boldsymbol{\mu}(\mathbf{r})=\mu[\cos\phi_\mu(\mathbf{r})\hat{\mathbf{x}}+\sin\phi_\mu(\mathbf{r})\hat{\mathbf{y}}]$ with $\mathbf{r}=r_x\hat{\mathbf{x}}+r_y\hat{\mathbf{y}}.)$ There are special topological configurations of spins with definite vorticity that must be discussed separately, as these vortices will give a different random-field contribution to $W(\mathbf{B})$ from ordinary random nonvortex spins. Besides this complication of the presence of extra magnetic random sources, the formalism can be applied in a straightforward manner to give the relaxation function.

Let there be f spins associated with a vortex (+ or − vorticity) and let there be $N_\pm$ of $\pm$ vortices. Then the total number of spins are $N=N_0+f(N_++N_-)$, where N_0 are the number of nonvortex spins. Let $n_\pm=N_\pm/N$ and the vortex density (n_++n_-) defined by X/f. Then $n_0=N_0/N=1-X$. From the dipole field contributions of vortices, shown in Fig. 5, one can show the magnetic fields of the $\pm$ vortex are

$$\mathbf{b}_+\simeq+\mu\frac{6a_0r}{r_>^5}\sin\phi\hat{\mathbf{r}}\ , \tag{36}$$

$$\mathbf{b}_-\simeq+\mu\frac{a_0r}{r_>^5}[18\sin(\phi'-\epsilon\theta)\hat{\rho}+12\cos(\phi'-\epsilon\theta)\hat{\rho}^*]\ , \tag{37}$$

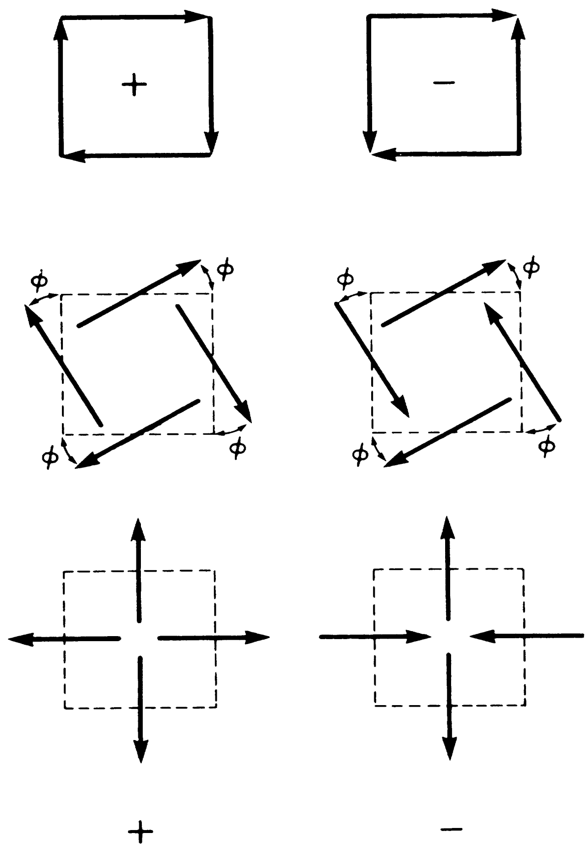

FIG. 5. Vortices of + and − vorticity. The vorticity of each vortex is fixed, but their dipole fields are different for different angle ϕ.

with $\phi'=\phi-\theta$, $r_>=\max(r,a_0)$, $\hat{\mathbf{r}}=(\cos\phi,\sin\phi)$, $\hat{\boldsymbol{\rho}}=(\cos\theta,\epsilon\sin\theta)$, $\hat{\boldsymbol{\rho}}^*=(-\epsilon\sin\theta,\cos\theta)$, and $\epsilon=\pm 1$ for $r \gtrless a$. These expressions for $\mathbf{b}_\pm$ are valid when r is not close to $a_0=a/\sqrt{2}$. For nonvortex spins, the magnetic field is $\mathbf{b}_0$, given by

$$\mathbf{b}_0=\frac{3(\boldsymbol{\mu}\cdot\hat{\mathbf{r}})\hat{\mathbf{r}}-\boldsymbol{\mu}}{r^3}\ . \tag{38}$$

We assume that this form holds down to a short distance r_0. After averaging over the orientation of $\hat{\boldsymbol{\mu}}(\phi)$ and $\hat{\mathbf{r}}$, we have[11]

$$A(\mathbf{p})=\exp\left\{-\pi\left[n_+g_+(\xi)+n_-g_-(\xi)+n_0\left[\frac{r_0}{a_0}\right]^2 g_0(\xi_0)\right]\right\}, \tag{39}$$

with the following g functions,

$$g_+(\xi)=\xi^{-2}\int_0^{\xi}d\omega\omega[1-J_0^2(6\omega)]+\frac{\xi^{1/2}}{4}\int_0^{\xi}d\omega\,\omega^{-3/2}[1-J_0^2(6\omega)]\ , \tag{40}$$

$$g_-(\xi)=\xi^{-2}\int_0^{\xi}d\omega\omega[1-J_0(15\omega)J_0(3\omega)]+\frac{\xi^{1/2}}{4}\int_0^{\xi}d\omega\omega^{-3/2}[1-J_0(15\omega)J_0(3\omega)]\ , \tag{41}$$

and

$$g_0(\xi_0)=\frac{\xi_0^{2/3}}{3}\int_0^{\xi_0}d\omega\,\omega^{-5/3}\left[1-J_0\left[\frac{3}{2}\omega\right]J_0\left[\frac{\omega}{2}\right]\right]\ . \tag{42}$$

Here $\xi=p\mu/a_0^3$ and $\xi_0=p\mu/r_0^3$. Using Eq. (39) we can compute (31), with the results shown in Fig. 6 for various vortex density X. The second moment Δ is given by

$$\Delta^2(X)=\frac{5\pi}{32}\left[\frac{\mu^2\gamma_\mu^2}{a_0^6}\right]\left[\frac{a_0}{r_0}\right]^4\left\{1+X\left[\frac{357}{20}\left[\frac{r_0}{a_0}\right]^4-1\right]\right\}. \tag{43}$$

Note the linear dependence of the square of the second moment of $G_{XY}(t)$ on the density of vortex. By the measurement of the zero field μSR relaxation function, one can test the prediction of the Kosterlitz-Thouless theory,[12] which give a definite prediction on the temperature dependence of the density of vortex near the vortex unbinding transition to be

$$X\sim\exp[-2b/(T-T_c)^{1/2}],\quad T>T_c\ , \tag{44}$$

with b a positive constant. Therefore, the quantity $1-[\Delta(T)/\Delta(T_c)]^2$, which is proportional to X will exhibit the exponential temperature dependence near T_c. In Fig. 6, $G_{xy}^0(t)$ is calculated for the case with $r_0=a_0/\sqrt{2}$ and $f=4$, suitable for a square lattice.

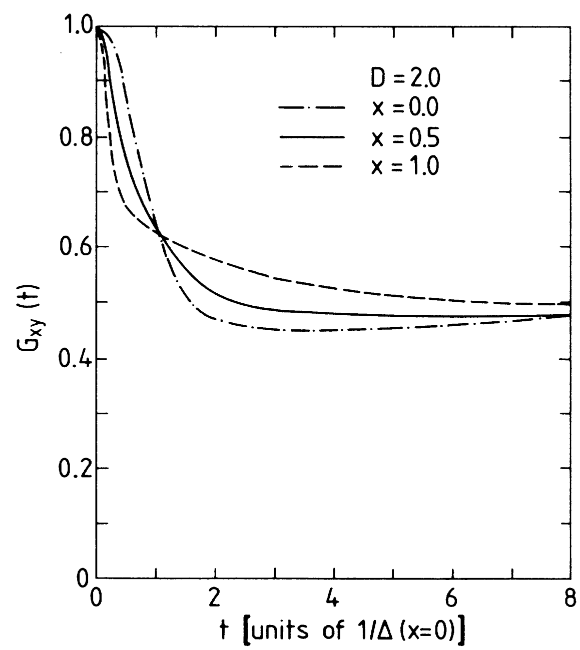

FIG. 6. The relaxation function $G_{xy}(t)$ versus time in units of $1/\Delta(X=0)$ for the two-dimension XY model. X is the vortex density $G_{xy}(t\to\infty)\to 0.5$.

F. Three-dimensional XY magnet

Here μ is confined to rotate on the xy plane. The angular average of $\hat{\mu}$ and $\hat{r}$ produce an anisotropic $A(\mathbf{p})$ given for small $\mathbf{p}$ by

$$A(\mathbf{p})=\exp[-\kappa(\tfrac{6}{5}p_{\parallel}^2+p_{\perp}^2)]\ , \tag{45}$$

where $\kappa=(4\pi/3)(r_0/a)^3$ and $\mathbf{p}=p_{\parallel}\hat{\mathbf{z}}+p_{\perp}(\cos\phi_p\hat{\mathbf{x}}+\sin\phi_p\hat{\mathbf{y}})$.

Making use of Appendix B, we can calculated the relaxation function $G^A(t)$. The results are shown in Fig. 4. The value of $G^A(t)$ approaches 0.359 as t approaches infinity.

G. Heisenberg magnet in 1, 2, and 3 dimensions

The magnetic moment of a Heisenberg magnet has three components. Generally the angular average of $\hat{\mu}$ for a dipolar field is complicated and one can simplify the calculation by the Gaussian approximation. The resulting magnetic field distribution $W(\mathbf{B})$ is generally anistropic.

In one dimension, we have

$$A(\mathbf{p})=\exp[-\kappa_{\parallel}p_{\parallel}^2-\kappa_{\perp}p_{\perp}^2]\ , \tag{46}$$

where $\kappa_{\parallel}=\kappa_1$ and $\kappa_{\perp}=8\kappa_1$ with

$$\kappa_1=\frac{32}{15}\left[\frac{r_0}{a}\right]\left[\frac{\mu^2\gamma_\mu^2}{r_0^6}\right]. \tag{47}$$

In two dimensions, $\kappa_{\parallel}=\frac{5}{2}\kappa_2$ and $\kappa_{\perp}=\kappa_2$ with $\kappa_2=(\pi/12)(r_0/a)^2(\mu^2\gamma_\mu^2/r_0^6)$. In three dimensions, the distribution is isotropic with $\kappa_{\parallel}=\kappa_{\perp}=\kappa_3=20\pi/27(r_0/a)^3(\mu^2\gamma_\mu^2/r_0^6)$. The fact that it is isotropic implies $G_H(t\to\infty)\to\frac{1}{3}$. This is the prediction of the Kubo-Toyabe theory.

For the two-dimensional Heisenberg model, $G_H(t\to\infty)\to 0.4608$ and in one dimension, $G_H(t\to\infty)\to 0.1168$. These features of the Heisenberg relaxation functions are shown in Fig. 7.

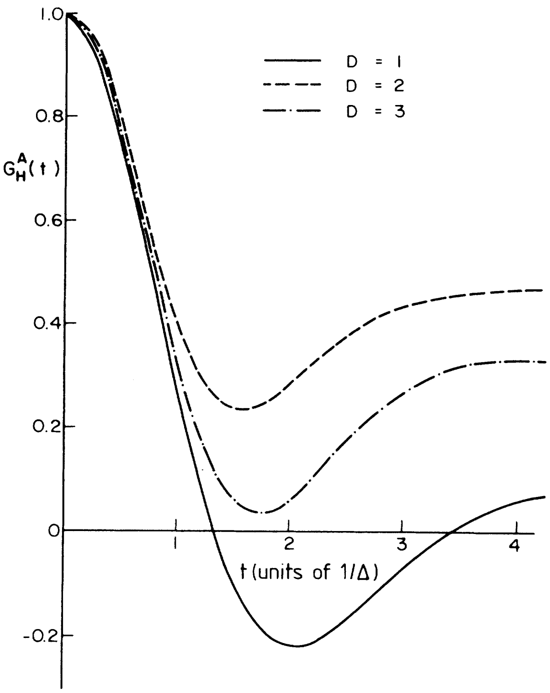

FIG. 7. The relaxation function $G_H^A(t)$ versus time T for Heisenberg systems in one, two, and three dimensions. The Gaussian approximation is used. The unit of time is $1/\Delta$ with $\Delta=(16\kappa_1)^{1/2},(2\kappa_2)^{1/2},(2\kappa_3)^{1/2}$ for one, two, and three dimensions.

V. DISCUSSIONS

The formalism developed in Secs. II and III provides a classical method of calculating the probability distribution of the random fields of the muon site for a given magnetic system. It extends the Kubo-Toyabe theory beyond the classical three-dimensional Heisenberg spin model to different magnets in one, two and three dimensions. The formalism itself is very versatile in dealing with various magnetic arrangements, in that the details of the calculation can be categorized into Ising, XY, and Heisenberg cases, corresponding to one, two, and three components of the random fields. On the other hand, the details of specific magnetic system enter into the calculation of $W(\mathbf{B})$ and correlation function $G(t)$ in the form of numerical integration. The model calculation presented in Sec. IV provide a framework where all corrections due to various perturbations can be made, just like the role the Kubo-Toyabe theory plays in the study of three dimensional Heisenberg system.

The examples we worked out in Sec. IV can be applied to various real systems. But before discussing the applications, we first note that in real magnetic systems, there are modifications on the nature of the spin involved, as well as the spatial dimensionality. In our model calculations, we discuss purely Ising, XY, and Heisenberg spins. In real magnets, the moments μ are usually not so ideal in its orientational degree of freedom. Generally, μ behaves more like a Heisenberg moment $\mu(\theta_\mu,\phi_\mu)$ with certain constraints on θ_μ and ϕ_μ. These constraints will affect the average of the orientation of μ. Such nonideal behavior of the moment μ in real magnetic systems usually comes from the anisotropy of the nearest neighbor spin model Hamiltonian. Another common consideration is the nonideal spatial dimensionality of real magnets. It is usually necessary to consider interchain coupling in the case of one dimensional system and interplanar interaction in the case of the two-dimensional system. Generally, the real system is three dimensional but with a nonrandom distribution of its moments μ. Thus, in the average over all the possible positions of the moments, the magnetic lattice structure must be taken into consideration. These modifications on our model calculations are necessary in the application of the theory, and they must be dealt with for each individual experimental system.[13,14] Nevertheless, the model calculations in Sec. IV provide the basic predictions of ideal magnetic system.

In one dimension, $[(CH_3)_4N][N_iCl_3]$ is an example of Heisenberg magnet.[14] Susceptibility measurements indicates a spin 1, ferromagnetic and Heisenberglike intrachain interaction with weak ferromagnetic interchain interactions. CsN_iF_3 is an example of quasi-XY magnet.[15] The easy plane of magnetization is perpendicular to the chain axis. For the Ising magnet, $CoCl_2 \cdot 2NC_5H_5$ (Ref. 17) is an example. It undergoes a crossover to three dimensional ordering of an ensemble of ferromagnetic chain below ~ 4 K.

In two dimension, graphite intercalation compounds provides a large category of quasi-two dimensional magnets.[18] Depending on the number of graphite layers between intercalants, the effect of the interplanar coupling can be monitored to yield an approximation to ideal two-dimensional magnets. Intercalation of different magnetic species provides quasi-two-dimensional Ising, XY, and Heisenberg magnets. However, this subject is still fairly new and deserves much more exploration. A successful example is the intercalation of $CoCl_2$ into graphite.[19] It provides one of the best model two-dimensional XY magnet and the possibility of observing the vortex unbinding transition predicted by the Kosterlitz-Thouless theory. Intercalation of $FeCl_2$ into graphite provides an example of quasi-two dimensional Ising magnet,[20] where as the intercalation of $FeCl_3$ yields a quasi-two-dimensional Heisenberg magnet.[21] There are other quasi-two-dimensional magnetic systems that are of interest. The family of $CuCl_4$ compounds $(C_nH_{2n+1}NH_3)_2CuCl_4$ for $n=1,2,3\ldots$ are examples of quasi-two-dimensional Heisenberg magnets,[22] while $FeCl_2$ is an example of Ising magnet.[23] Since in two dimension, Ising magnet has a finite temperature phase transition with the magnetization $m \propto (1-T/T_c)^{\beta}$ with $\beta=\frac{1}{8}$, one may measure the critical exponents through the magnetization dependence of the correlation function $G_I^0(t)$, given in Eq. (25).

For three dimensions, there are plenty of Heisenberg magnets, such as $Cu(NH_4)Br_4 2H_2O$,[24] EuO,[25] etc. There are some ferromagnetic Ising systems, such as $Tb(OH)_3$,[26] $Dy(OH)_3$, and $HO(OH)_3$.[27] We do not know of a good example of ferromagnetic XY magnets in three dimension.

The above magnetic systems are all ferromagnetic. Our formalism can be extended to antiferromagnetic systems. However, the fundamental limitations are more subtle. The description of a muon spin precessing in a random field with a distribution computed classically assuming no correlation between spins and purely dipolar interaction is a great simplification. In real situations, we expect that the spins in the immediate neighborhood of the muon will play an important role in the dynamics of the muon. Futhermore, there are indications[10,28,29] that quantum effects are important. The consideration of the effects of nearest neighbors on the muons necessitates a specification of the lattice structure and the muon site. All these microscopic details are ignored in our calculation. It is desirable to extend our investigation to include these microscopic details, at least semiclassically. However, such an extension necessarily requires specifications of the details of the system under investigation, thereby a general statement on the behavior of the muon relaxation function is likely to be very difficult. An important point about our formalism is that it tries to take into account all the spins in the systems. This is almost impossible to achieve quantum mechanically on a lattice. In one point of view, we can consider that the calculations made here are the required limits of the corresponding refined quantum treatments as the number of particles goes to infinity.[30]

Our calculations can be used as a basis for the analysis of experimental results, provided that we take into account the muon diffusion process. Such an analysis is fairly straightforward, but the details of the muon trapping and escaping processes require some knowledge in the material under studied. If we let the ϵ be the muon hopping rate and ν the muon trapping rate, and denote our relaxation function calculated assuming no dynamical processes as $G(t)$, then the measured relaxation function, $G_d(t)$, is[31]

$$G_d(t)=e^{-\nu t}+e^{-gt}\nu\int_0^t dt' e^{-\bar{\nu}t'}G(t-t') +\epsilon\nu\int_0^t dt''\int_0^{t''} dt' e^{-gt''-\bar{\nu}t'}G(t''-t')G_d(t-t'') , \tag{48}$$

with $\bar{\nu}=\nu-\epsilon$.

We can extend our analysis to the general problem of muon spin relaxation in a finite field. The formalism readily accommodates these situations with the total field as the sum of the local random field plus an external field. Finally, we can also employ the formalism developed in this paper to deal with the relaxation of muon in a spin-glass environment.[32]

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APPENDIX A

For the Ising case,

$$\langle \cos(B\tau)\rangle = \int_{-\infty}^{\infty} dB \int_{-\infty}^{\infty} \frac{dp}{2\pi} A(p) e^{ipB}\cos(B\tau) = \tfrac{1}{2}[A(\tau)+A(-\tau)] . \tag{A1}$$

For the XY case,

$$\begin{aligned}\langle \cos(B\tau)\rangle &= \int_0^{\infty} B\,dB \int_0^{\infty} p\,dp J_0(pB)A(p)\cos(B\tau)\\ &= \frac{d}{d\tau}\int_0^{\infty} pA(p)dp\int_0^{\infty}\sin(B\tau)J_0(pB)dB\\ &= \frac{d}{d\tau}\int_0^{\tau}\frac{pA(p)}{(\tau^2-p^2)^{1/2}}dp\\ &= \int_0^{\pi/2} d\theta\sin\theta\left[A(p)+p\frac{dA(p)}{dp}\right]_{p=\tau\sin\theta} .\end{aligned} \tag{A2}$$

Finally, for the Heisenberg case,

$$\langle \cos(B\tau)\rangle = \int d^3\mathbf{B} \int \frac{d^3\mathbf{p}}{(2\pi)^3} A(\mathbf{p}) e^{i\mathbf{pB}} \cos(B\tau)$$
$$= \frac{1}{\pi}\int_0^\infty B\,dB \int_0^\infty p\,dpA(p)[\sin(p+\tau)B + \sin(p-\tau)B]$$
$$= \frac{d}{d\tau}\int_0^\infty p\,dpA(p)[\delta(p-\tau)-\delta(p+\tau)]$$
$$= \frac{d}{d\tau}[\tau A(\tau)] = \left[A(\tau)+\tau\frac{dA(\tau)}{d\tau}\right] . \quad \text{(A3)}$$

APPENDIX B

For anisotropic $W(\mathbf{B})$ given by (5), we introduce $\eta = 1/4a$, $\lambda = 1/4b$, $\xi = \lambda - \eta$ and write

$$W(\mathbf{B}) = \frac{\eta\sqrt{\lambda}}{\pi^{3/2}} \exp(-\eta B_\perp^2 - \lambda B_\parallel^2) . \quad \text{(B1)}$$

Then,

$$\left\langle \frac{B_\parallel^2}{B^2} \right\rangle = \frac{\eta\sqrt{\lambda}}{\pi^{3/2}} \int_0^\infty B^2 dB \int_0^{2\pi} d\phi \times \int_{-1}^1 d\cos\theta \cos^2\theta e^{-B^2(\eta+\xi\cos^2\theta)}$$
$$= -4\pi\frac{\eta\sqrt{\lambda}}{\pi^{3/2}} \frac{\partial}{\partial \xi} \int_0^1 dx \int_0^\infty dB e^{-B^2(\eta+\xi x^2)}$$
$$= \eta\sqrt{\lambda}\int_0^1 dx x^2 z^{3/2} , \quad \text{(B2)}$$

where $z = (\eta+\xi x^2)^{-1}$:

$$\left\langle \frac{B_\perp^2}{B^2}\cos B\tau \right\rangle = \frac{\eta\sqrt{\lambda}}{\pi^{3/2}} \int_0^\infty dB \int_0^{2\pi} d\phi \int_{-1}^1 dx B^2(1-x^2) e^{-B^2(\eta+\xi x^2)} \cos(B\tau)$$
$$= -4\pi\frac{\eta\sqrt{\lambda}}{\pi^{3/2}} \left[\frac{\partial}{\partial \eta} - \frac{\partial}{\partial \xi}\right] \int_0^\infty dB \int_0^1 dx e^{-B^2(\eta+\xi x^2)} \cos(B\tau)$$
$$= \eta\sqrt{\lambda}\int_0^1 dx(1-x^2)\left[z^{3/2} - \frac{\tau^2}{2} z^{5/2}\right] e^{-z\tau^2/4}, \text{ as } \tau\to\infty,\ G^A(t)\to\left\langle \frac{B_\parallel^2}{B^2}\right\rangle .$$

For $\xi > 0$,

$$G^A(\infty) = -\sqrt{\frac{\lambda}{\eta}} \frac{1}{\alpha^2} \left[\frac{1}{(1+\alpha^2)^{1/2}} - \frac{1}{\alpha}\ln[\alpha+(1+\alpha^2)^{1/2}]\right] , \quad \text{(B3)}$$

with $\alpha = \sqrt{\xi/\eta}$ and, for $\xi < 0$,

$$G^A(\infty) = \sqrt{\frac{\gamma}{\eta}} \frac{1}{\beta^2} \left[\frac{1}{(1-\beta^2)^{1/2}} - \frac{1}{\beta}\sin^{-1}\beta\right] \quad \text{(B4)}$$

with $\beta = \sqrt{-\xi/\eta}$. The second moment for the anisotropic distribution is $\Delta = \sqrt{2a}$.

APPENDIX C: ISING FIELD AND XY FIELD FROM CORRESPONDING SPIN SYSTEM

1. One-dimensional Ising spin system

The Ising spin moments ($\boldsymbol{\mu} = \mu\hat{\mathbf{x}}$) are located along the $\hat{\mathbf{z}}$ axis. The microscopic dipole field at a distance $\mathbf{r} = r\hat{\mathbf{z}}$ from the moment is

$$\mathbf{b} = \frac{3(\boldsymbol{\mu}\cdot\hat{\mathbf{r}})\hat{\mathbf{r}} - \boldsymbol{\mu}}{r^3} = -\frac{\mu\hat{\mathbf{x}}}{r^3}$$

so that there is only one component of the dipolar field, thus the name Ising field.

2. Two-dimensional Ising spin system

The Ising spin moments ($\boldsymbol{\mu} = \mu\hat{\mathbf{z}}$) are located on the xy plane. The microscopic dipolar field at $\mathbf{r} = x\hat{\mathbf{x}} + y\hat{\mathbf{y}}$ from the moment is

$$\mathbf{b} = \frac{3(\boldsymbol{\mu}\cdot\hat{\mathbf{r}})\hat{\mathbf{r}} - \boldsymbol{\mu}}{r^3} = -\frac{\mu\hat{\mathbf{z}}}{(x^2+y^2)^{3/2}}$$

so again there is only one component of the field.

3. Three-dimensional Ising spin system

For three dimensional Ising spin system, the microscopic field has three components and the idea of Ising field is an approximation.

4. One-dimensional xy spin system

The moments of the XY spins are $\boldsymbol{\mu} = \mu_x\hat{\mathbf{x}} + \mu_y\hat{\mathbf{y}}$ located along the $\hat{\mathbf{z}}$ axis, so that the microscopic field $\mathbf{b}$ at a position $\mathbf{r}$ along the $\hat{\mathbf{z}}$ axis is

$$\mathbf{b} = \frac{3(\boldsymbol{\mu}\cdot\hat{\mathbf{r}})\hat{\mathbf{r}} - \boldsymbol{\mu}}{r^3} = -\frac{\boldsymbol{\mu}}{r^3} .$$

There are only two components of the field $\mathbf{b}$, which is called the XY field.

5. Two-dimensional XY spin system

The moments are the XY spins $\boldsymbol{\mu}=\mu_x\hat{\mathbf{x}}+\mu_y\hat{\mathbf{y}}$ located on the XY plane. The microscopic field $\mathbf{b}$ for a position $\mathbf{r}$ on the XY plane is

$$\mathbf{b}=\frac{3(\boldsymbol{\mu}\cdot\hat{\mathbf{r}})\hat{\mathbf{r}}-\boldsymbol{\mu}}{r^3}=(b_x,b_y)$$

and there are only two components.

6. Three-dimensional XY spin system

The moments are XY spins in three-dimensional space. In general there are three components of the microscopic field and the anisotropic distribution function can be used.

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