Skewed-field technique in muon-spin rotation: Kubo-Toyabe longitudinal and transverse relaxation functions

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(Received 17 February 1984; revised manuscript received 8 June 1984)

For any arbitrary external-field direction, three distinct muon-spin relaxation functions can be defined; that is, there is a longitudinal relaxation function in the direction of the external field and two transverse relaxation functions in the plane perpendicular to the field direction. One of these latter functions, termed the coplanar transverse relaxation function, lies in the plane defined by the field and the incoming muon-spin polarization, while the other, termed the perpendicular transverse relaxation function, is perpendicular to both the field direction and the incoming spin polarization. All three relaxation functions can be measured simultaneously if the applied external field is not in either of the standard geometries, namely longitudinal or transverse. This suggests that a skewed-field arrangement provides an experimental technique in which the traditional relaxation functions for a given sample may be determined in a single experiment with the use of one apparatus. Thus, at the very least, such an experimental alignment eliminates the down time required when changing from one geometry to the other in the determination of the relaxation functions for a given sample. With the use of the static Kubo-Toyabe theory as an illustrative example, explicit expressions for these relaxation functions are obtained in terms of a pair of one-dimensional integrals. In particular, an alternate, but equal, expression for the standard longitudinal Kubo-Toyabe relaxation function is obtained. An analytic expression for the coplanar transverse Kubo-Toyabe relaxation function is presented which agrees with the numerical results derived by Kubo, while an analytic expression for the new perpendicular transverse Kubo-Toyabe relaxation function is given.

I. INTRODUCTION

Muon-spin-rotation (μSR) experiments in condensed matter[1–4] probe the local magnetic environment experienced when spin-polarized positive muons thermalize in diamagnetic or paramagnetic states. That is, the dynamics of the spin polarization of an ensemble of muons is followed by observing the decay positrons that are emitted preferentially along the muon's spin direction. Histograms of these ensembles are fitted to a function of the form

$$N(t)=N_0\exp(-t/\tau_\mu)[1+G(t)]+B_0\ , \tag{1.1}$$

where $\tau_\mu=2.2\ \mu\text{sec}$ is the muon's lifetime, N_0 is a normalization constant, B_0 is a background constant, and $G(t)$ is the relaxation function. The experimental relaxation functions are related to theories for the spin dynamics of a single muon. Experiments are performed using two standard[1–4] geometries, namely longitudinal and transverse. In the longitudinal case, the incoming muon-spin polarization and the applied external magnetic field are collinear, while counters are placed normal to this direction. On the other hand, for the transverse case, the incoming polarization and the magnetic field are perpendicular, while counters are placed in the plane perpendicular to the field. Experiments with the same sample but different geometries are carried out separately. Indeed, there often are two completely different experimental apparatuses for the different geometries. On the other hand, if one apparatus is used, alterations are required to change the field direction. In both situations measurement of the longitudinal and transverse relaxation functions for the same sample requires a down time in which the apparatus is either changed or modified.

The purpose of the present paper is to point out that longitudinal and transverse experiments can be carried out simultaneously. Such a simultaneous measurement is possible using a skewed-field technique wherein the direction of the magnetic field and the incoming polarization are at an arbitrary angle to each other. At the very least there are two advantages to such a technique, namely that a single experimental apparatus is required and that no down time is needed for obtaining the longitudinal and transverse relaxation functions for a given sample. Indeed, the advent of spin rotators,[5,6] with which the muon's spin may be rotated with respect to its momentum, makes the single-apparatus experiment very attractive. Thus, given the appropriate apparatus, whether the skewed-field technique or one of the traditional alignments is used is then a matter of priorities determined by the particular experiment. For such an apparatus the counters define the laboratory reference frame, that is, an orthonormal coordinate system X,Y,Z with counters placed normal to these directions (in both the positive and negative sense). There are two other directions associated with this experiment, namely the external magnetic field direction and the direction of the incoming muon-spin polarization. These directions are associated with the muon's spin dynamics. They and the plane defined by them can be related to the laboratory frame. Indeed, this plane defines two orthonormal directions with the third spatial direction being perpendicular to it. Thus measurements of the muon po-

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larization in a general coordinate system can be expressed in terms of these dynamical directions. Here, in particular, the external magnetic field is taken to lie along the $\hat{Z}$ direction, while the incoming muon-spin polarization is assumed to lie in the XZ plane. When the magnitude of the external field is allowed to approach zero, the $\hat{Z}$ direction is still a well-defined quantity.

To illustrate that longitudinal and transverse experiments can be conducted simultaneously, three distinct relaxation functions associated with the dynamics of a diamagnetic muon are evaluated using standard static Kubo-Toyabe theory.[7–10] This model for the spin dynamics of a diamagnetic muon assumes that the effects of the environment on a thermalized static (nondiffusing) muon are well characterized by a classical random isotropic local magnetic field. That is, a statistical average of the dynamics associated with an isolated muon is taken using an isotropic distribution of classical local random fields about the given external magnetic field. The result is an expectation value for the spin polarization of a muon wherein the components in the appropriate directions are called relaxation functions. Analytic expressions[7–10] [see, for example, Eq. (A9)] for the longitudinal geometry using either a Gaussian or a Lorentzian random local field constitute the standard model, while numerical results[8] for one of the transverse relaxation functions have also been given. Here, for a Gaussian field distribution, an alternate analytic expression for the longitudinal static Kubo-Toyabe relaxation function is derived, while analytic expressions for the two transverse static Kubo-Toyabe relaxation functions are presented. In particular, each of these relaxation functions is written as a sum of three terms. That is, there is a static term and two time-dependent terms. The static term is due to the component of the incoming spin polarization along the appropriate direction, while the time-dependent terms are products of relaxation functions with either the cosine or sine of the product of the time and the diamagnetic Larmor frequency (ω_μ). Such a resolution of the full longitudinal, coplanar, and perpendicular transverse relaxation functions is more akin to the isolated dynamics than the previous results. Thus it is hoped that these new expressions are more illustrative of the physics. In addition, it is to be emphasized that all three Kubo-Toyabe relaxation functions can be observed simultaneously if the incoming polarization and the external field are neither parallel nor perpendicular.

For a skewed-magnetic-field technique the relaxation function in the direction of the external magnetic field is the product of the usual longitudinal relaxation function and the component, $\cos(\theta_{\rm in})$, of the muon-spin polarization in this direction. Here, $\theta_{\rm in}$ is the angle between the external field and the incoming polarization, the skew angle. Thus, for zero external fields, this longitudinal relaxation function reduces to the product of $\cos(\theta_{\rm in})$ and the standard zero-field Kubo-Toyabe relaxation function,[7–10] while for high longitudinal fields it becomes simply $\cos(\theta_{\rm in})$. Furthermore, it is possible to simultaneously observe two other relaxation functions which lie in the plane perpendicular to the external field. One of these lies in the plane defined by the external field and the incoming muon polarization. Thus it is termed the coplanar transverse relaxation function and it is the product of $\sin(\theta_{\rm in})$ and a transverse relaxation function. For zero fields this coplanar transverse relaxation function reduces to the product of $\sin(\theta_{\rm in})$ with the zero-field Kubo-Toyabe relaxation function, while for high fields it becomes the product of $\sin(\theta_{\rm in})$ with a Gaussian damping function and $\cos(\omega_\mu t)$. This transverse function has a nonzero signal for low fields since it contains a component of the initial polarization. On the other hand, the perpendicular transverse relaxation function, which is also the product of $\sin(\theta_{\rm in})$ and another transverse relaxation function, reduces to zero for low fields and is the product of $\sin(\theta_{\rm in})$ with a Gaussian damping function and $\sin(\omega_\mu t)$ for high fields. This perpendicular transverse relaxation function is zero for low fields since it does not contain any of the initial polarization. Finally, for high fields, the transverse motion becomes a rotation about the external magnetic field with a Gaussian damping due to the random local magnetic field.

The polarization dynamics of a spin-$\frac{1}{2}$ particle in a magnetic field are reviewed in Sec. II, while in Sec. III Kubo-Toyabe theory is presented for an arbitrary magnetic field direction. Gaussian relaxation functions are obtained in Sec. IV.

II. MUON-SPIN DYNAMICS IN A FIELD

For a diamagnetic muon in a magnetic field with magnitude B and direction $\hat{B}$, the Zeeman spin-Hamiltonian operator,[1] $H_{\rm op}=(-\frac{1}{2}\hbar\omega_\mu)\hat{B}\cdot\vec{\rm P}_{\rm op}$, involves the spin polarization of the muon, $\vec{\rm P}_{\rm op}=(2/\hbar)\vec{\rm I}_{\rm op}$ and the muon Larmor frequency $\omega_\mu=\bar{\gamma}_\mu B$, where $\gamma_\mu=0.0136$ MHz/G ($\bar{\gamma}_\mu=2\pi\gamma_\mu$) is the muon's gyromagnetic ratio. The time dependence of the expectation value of the polarization of an isolated muon is described by the Heisenberg equation,[11]

$$\begin{aligned} d\langle\vec{\rm P}_{\rm op}\rangle(t)/dt &= \langle(i/\hbar)[H_{\rm op},\vec{\rm P}_{\rm op}]\rangle(t) \\ &= -\omega_\mu\underline{\epsilon}{:}\hat{B}\langle\vec{\rm P}_{\rm op}\rangle=\omega_\mu\hat{B}\times\langle\vec{\rm P}_{\rm op}\rangle\ , \end{aligned} \tag{2.1}$$

where $\underline{\epsilon}$ is the Levi-Cività[11,12] third-rank antisymmetric tensor. The dot-product convention, $\overleftrightarrow{\rm F}{:}\overleftrightarrow{\rm G}=F_{ij}G_{ji}$, has been adopted here. There is no difference between quantal and classical motion for this spin-$\frac{1}{2}$ particle.[11] Quantum mechanics only enters in the discrete magnitude of the spin. The resulting motion of the normalized muon-spin polarization,

$$\langle\vec{\rm P}_{\rm op}\rangle_N(t)=\langle\vec{\rm P}_{\rm op}\rangle(t)/|\langle\vec{\rm P}_{\rm op}\rangle(0)|=\overleftrightarrow{\rm G}(t)\cdot\hat{P}_{\rm in}\ , \tag{2.2}$$

involves the initial polarization of the muon, $\langle\vec{\rm P}_{\rm op}\rangle(0)=\vec{\rm P}_{\rm in}$, and a second-rank motion tensor,

$$\begin{aligned} \overleftrightarrow{\rm G}(t) &= \exp(-\omega_\mu t\underline{\epsilon}\cdot\hat{B}) \\ &= \overleftrightarrow{\rm U}+(\overleftrightarrow{\rm U}-\hat{B}\hat{B})[\cos(\omega_\mu t)-1]-\underline{\epsilon}\cdot\hat{B}\sin(\omega_\mu t)\ , \end{aligned} \tag{2.3}$$

which describes the classical or quantal rotation dynamics of a spin-$\frac{1}{2}$ particle in a magnetic field $\vec{\rm B}$. Normalized polarizations are considered here since it is in this form that experimental relaxation functions are written. Using Eq. (2.3), the normalized muon-spin polarization becomes

$$\begin{aligned}\langle \vec{P}_{op}\rangle_N(t) &= [\hat{B}\hat{B}+(\overleftrightarrow{U}-\hat{B}\hat{B})\cos(\omega_\mu t)-\underline{\epsilon}\cdot\hat{B}\sin(\omega_\mu t)]\cdot\hat{P}_{in} \\ &= \hat{B}(\hat{B}\cdot\hat{P}_{in})+[\hat{P}_{in}-\hat{B}(\hat{B}\cdot\hat{P}_{in})]\cos(\bar{\gamma}_\mu Bt)+\hat{B}\times\hat{P}_{in}\sin(\bar{\gamma}_\mu Bt) \\ &= \hat{B}\cos\theta+\sin\theta[\hat{n}\cos(\bar{\gamma}_\mu Bt)+\hat{m}\sin(\bar{\gamma}_\mu Bt)] \ . \end{aligned} \tag{2.4}$$

The last form of Eq. (2.4) involves three contributions. One of these, in the direction of the magnetic field $\hat{B}$, is the longitudinal term, namely the component of the incoming polarization in the field direction, $\cos\theta=\hat{P}_{in}\cdot\hat{B}$, which is independent of the time. The other two are the transverse terms which lie in the plane perpendicular to the magnetic field. One is in the plane defined by $\hat{B}$ and $\hat{P}_{in}$, that is, along the direction

$$\hat{n}=(\hat{P}_{in}-\hat{B}\cos\theta)/\sin\theta \ .$$

Thus it is the coplanar transverse component. The other, in the direction $\hat{m}=\hat{B}\times\hat{P}_{in}/\sin\theta$, is perpendicular to both the magnetic field and the incoming polarization and, thus, is the perpendicular transverse component. This transverse motion is, of course, the usual rotation of the spin about the magnetic field axis with frequency $\bar{\gamma}_\mu B$. The magnitude of these terms is the component of the incoming polarization, $\sin\theta$, which is perpendicular to the magnetic field. On the other hand, the terms in the $\hat{B},\hat{P}_{in}$ plane may also be resolved along the direction of the incoming polarization and along the component of $\hat{B}$ perpendicular to $\hat{P}_{in}$. However, the dynamics of the isolated muon contains three functionalities of the time, namely a static term, a $\cos(\bar{\gamma}_\mu Bt)$ term, and a $\sin(\bar{\gamma}_\mu Bt)$ term. Because of this general behavior, the Kubo-Toyabe relaxation functions obtained for the Gaussian local random field are also resolved in terms of a static contribution, a $\cos(\bar{\gamma}_\mu B_{ex}t)$ contribution and a $\sin(\bar{\gamma}_\mu B_{ex}t)$ contribution, where B_{ex} is the magnitude of the external field.

III. KUBO-TOYABE THEORY

The above dynamics represents the motion of an isolated muon in a magnetic field $\vec{B}$. However, when a muon thermalizes in condensed matter, the effects of the environment on the muon's spin polarization must be taken into account. One model for the effects of the environment is static Kubo-Toyabe theory,[7–10] where a nondiffusing muon is assumed to be at a site which contains an isotropic classical random local magnetic field. That is, the muon experiences a classical distribution of local fields around the given external magnetic field. The expectation value of the muon's spin polarization is then assumed to be an average over this field distribution of the isolated dynamics given by Eq. (2.4), that is

$$\begin{aligned}\langle \vec{P}_{op}\rangle_N^{KT}(t) &= \int d\vec{B}\, f(\vec{B}-\vec{B}_{ex})\langle \vec{P}_{op}\rangle_N(t) \\ &= \overleftrightarrow{G}^{KT}(t;\vec{B}_{ex})\cdot\hat{P}_{in} \ , \end{aligned} \tag{3.1}$$

where the Kubo-Toyabe motion tensor is

$$\overleftrightarrow{G}^{KT}(t;\vec{B}_{ex}) = \int d\vec{B}\, f(\vec{B}-\vec{B}_{ex})\overleftrightarrow{G}(t) = \sum_{j=1}^{3}\overleftrightarrow{I}_j^{KT}(t;\vec{B}_{ex}) \ . \tag{3.2}$$

This may be written as a sum of three terms when Eq. (2.4) is used. Since there is only one direction ($\hat{B}_{ex}$) associated with this Kubo-Toyabe dynamics, the second-rank motion tensor can then only involve the isotropic unit tensor $\overleftrightarrow{U}$, the dyadic $\hat{B}_{ex}\hat{B}_{ex}$, and the combination of $\hat{B}_{ex}$ with the Levi-Cività antisymmetric tensor, $\underline{\epsilon}\cdot\hat{B}_{ex}$. In particular, the first term in Eq. (2.4) gives rise to a static contribution to the dynamics, namely

$$\begin{aligned}\overleftrightarrow{I}_1^{KT}(\vec{B}_{ex}) &= \int d\vec{B}\, f(\vec{B}-\vec{B}_{ex})\hat{B}\hat{B} \\ &= \tfrac{1}{2}A_1(B_{ex})(\overleftrightarrow{U}-\hat{B}_{ex}\hat{B}_{ex})+C_1(B_{ex})\hat{B}_{ex}\hat{B}_{ex} \ , \\ C_1(B_{ex}) &= \hat{B}_{ex}\hat{B}_{ex}\colon \int d\vec{B}\, f(\vec{B}-\vec{B}_{ex})\hat{B}\hat{B} \ , \\ A_1(B_{ex}) &= \overleftrightarrow{U}\colon \int d\vec{B}\, f(\vec{B}-\vec{B}_{ex})\hat{B}\hat{B}-C_1(B_{ex}) = 1-C_1(B_{ex}) \ . \end{aligned} \tag{3.3}$$

On the other hand, the second and third terms can be written as

$$\begin{aligned}\overleftrightarrow{I}_2^{KT}(t;\vec{B}_{ex}) &= \tfrac{1}{2}A_2(t;B_{ex})(\overleftrightarrow{U}-\hat{B}_{ex}\hat{B}_{ex})+C_2(t;B_{ex})\hat{B}_{ex}\hat{B}_{ex} \ , \\ C_2(t;B_{ex}) &= \hat{B}_{ex}\hat{B}_{ex}\colon \int d\vec{B}\, f(\vec{B}-\vec{B}_{ex})(\overleftrightarrow{U}-\hat{B}\hat{B})\cos(\bar{\gamma}_\mu Bt) \ , \\ A_2(t;B_{ex}) &= 2\int d\vec{B}\, f(\vec{B}-\vec{B}_{ex})\cos(\bar{\gamma}_\mu Bt)-C_2(t;B_{ex}) \ , \end{aligned} \tag{3.4}$$

and

$$\begin{aligned}\overleftrightarrow{I}_3^{KT}(t;\vec{B}_{ex}) &= C_3(t;B_{ex})\underline{\epsilon}\cdot\hat{B}_{ex} \ , \\ C_3(t;B_{ex}) &= \hat{B}_{ex}\cdot\int d\vec{B}\, f(\vec{B}-\vec{B}_{ex})\hat{B}\sin(\bar{\gamma}_\mu Bt) \ , \end{aligned} \tag{3.5}$$

respectively. The Kubo-Toyabe motion tensor,

$$\begin{aligned}\overleftrightarrow{G}^{KT}(t;\vec{B}_{ex}) &= \hat{B}_{ex}\hat{B}_{ex}G_L^{KT}(t;B_{ex}) \\ &\quad +(\overleftrightarrow{U}-\hat{B}_{ex}\hat{B}_{ex})G_{CT}^{KT}(t;B_{ex}) \\ &\quad -\underline{\epsilon}\cdot\hat{B}_{ex}G_{PT}^{KT}(t;B_{ex}) \ , \end{aligned} \tag{3.6}$$

may then be expressed in terms of the second-rank tensors, $\hat{B}_{ex}\hat{B}_{ex}$, $\overleftrightarrow{U}-\hat{B}_{ex}\hat{B}_{ex}$, and $\underline{\epsilon}\cdot\hat{B}_{ex}$, whose coefficients are the standard longitudinal (L) relaxation function,[7–10]

$$\begin{aligned}G_L^{KT}(t;B_{ex}) &= C_1(B_{ex})+C_2(t;B_{ex}) \\ &= \hat{B}_{ex}\hat{B}_{ex}\colon \int d\vec{B}\, f(\vec{B}-\vec{B}_{ex}) \\ &\qquad \times[\hat{B}\hat{B}+(\overleftrightarrow{U}-\hat{B}\hat{B})\cos(\bar{\gamma}_\mu Bt)] \ , \end{aligned} \tag{3.7}$$

the coplanar transverse (CT) relaxation function,[8]

$$G^{\mathrm{KT}}_{\mathrm{CT}}(t;B_{\mathrm{ex}})=\tfrac{1}{2}[A_1(B_{\mathrm{ex}})+A_2(t;B_{\mathrm{ex}})]$$
$$=\tfrac{1}{2}\hat{B}_{\mathrm{ex}}\hat{B}_{\mathrm{ex}}:\int d\vec{B}f(\vec{B}-\vec{B}_{\mathrm{ex}})[(\overleftrightarrow{U}-\hat{B}\hat{B})+(\overleftrightarrow{U}+\hat{B}\hat{B})\cos(\bar{\gamma}_\mu Bt)]\ , \tag{3.8}$$

and the perpendicular transverse (PT) relaxation function,

$$G^{\mathrm{KT}}_{\mathrm{PT}}(t;B_{\mathrm{ex}})=C_3(t;B_{\mathrm{ex}})\ . \tag{3.9}$$

Thus the normalized Kubo-Toyabe polarization,

$$\langle\vec{P}_{\mathrm{op}}\rangle^{\mathrm{KT}}_{N}(t)=\hat{B}_{\mathrm{ex}}(\hat{B}_{\mathrm{ex}}\cdot\hat{P}_{\mathrm{in}})G^{\mathrm{KT}}_{\mathrm{L}}(t;B_{\mathrm{ex}})+[\hat{P}_{\mathrm{in}}-\hat{B}_{\mathrm{ex}}(\hat{B}_{\mathrm{ex}}\cdot\hat{P}_{\mathrm{in}})]G^{\mathrm{KT}}_{\mathrm{CT}}(t;B_{\mathrm{ex}})+\hat{B}_{\mathrm{ex}}\times\hat{P}_{\mathrm{in}}G^{\mathrm{KT}}_{\mathrm{PT}}(t;B_{\mathrm{ex}})$$
$$=\hat{B}_{\mathrm{ex}}\bar{G}^{\mathrm{KT}}_{\mathrm{L}}(t;B_{\mathrm{ex}})+\hat{n}_{\mathrm{ex}}\bar{G}^{\mathrm{KT}}_{\mathrm{CT}}(t;B_{\mathrm{ex}})+\hat{m}_{\mathrm{ex}}\bar{G}^{\mathrm{KT}}_{\mathrm{PT}}(t;B_{\mathrm{ex}})\ , \tag{3.10}$$

involves the orthonormal coordinate system,

$$\hat{B}_{\mathrm{ex}},\hat{n}_{\mathrm{ex}}=(\hat{P}_{\mathrm{in}}-\hat{B}_{\mathrm{ex}}\cos\theta_{\mathrm{in}})/\sin\theta_{\mathrm{in}}\ ,$$

and

$$\hat{m}_{\mathrm{ex}}=\hat{B}_{\mathrm{ex}}\times\hat{P}_{\mathrm{in}}/\sin\theta_{\mathrm{in}}$$

(where $\hat{B}_{\mathrm{ex}}\cdot\hat{P}_{\mathrm{in}}=\cos\theta_{\mathrm{in}}$, $0\le\theta_{\mathrm{in}}\le\pi$); cf. Eq. (2.4). This coordinate system, $\hat{n}_{\mathrm{ex}},\hat{m}_{\mathrm{ex}},\hat{B}_{\mathrm{ex}}$, can be associated with the laboratory system $\hat{X},\hat{Y},\hat{Z}$, which is defined by the counter geometry. Equation (3.10) contains three relaxation functions,

$$\bar{G}^{\mathrm{KT}}_{\mathrm{L}}(t;B_{\mathrm{ex}})=\cos\theta_{\mathrm{in}}G^{\mathrm{KT}}_{\mathrm{L}}(t;B_{\mathrm{ex}})\ ,$$
$$\bar{G}^{\mathrm{KT}}_{\mathrm{CT}}(t;B_{\mathrm{ex}})=\sin\theta_{\mathrm{in}}G^{\mathrm{KT}}_{\mathrm{CT}}(t;B_{\mathrm{ex}})\ , \tag{3.11}$$
$$\bar{G}^{\mathrm{KT}}_{\mathrm{PT}}(t;B_{\mathrm{ex}})=\sin\theta_{\mathrm{in}}G^{\mathrm{KT}}_{\mathrm{PT}}(t;B_{\mathrm{ex}})\ ,$$

which are observable if counters are placed normal to the coordinates $\hat{B}_{\mathrm{ex}}$, $\hat{n}_{\mathrm{ex}}$, and $\hat{m}_{\mathrm{ex}}$, that is, if the dynamical and counter geometries are the same. Equation (3.10) holds for any isotropic classical random local magnetic field. In such a counter-defined geometry, the direction associated with the external field is well defined whether or not an external magnetic field is applied. In addition, longitudinal and transverse experiments can be performed simultaneously. However, the amplitudes of the signals will be reduced by the $\cos(\theta_{\mathrm{in}})$ or $\sin(\theta_{\mathrm{in}})$, respectively. Taking $\theta_{\mathrm{in}}=45°$ will reduce both signals by the same amount, namely 0.7071. This reduction in the signal amplitude is offset by the fact that all three relaxation functions are obtained from one experiment.

IV. GAUSSIAN RELAXATION FUNCTIONS

In standard Kubo-Toyabe theory,[7–10] the isotropic local-field distribution is taken to be either a Gaussian or a Lorentzian about the external field. Here, only the Gaussian distribution is considered, namely

$$f(\vec{B})=(2\pi B_0^2)^{-3/2}\exp(-B^2/2B_0^2)\ . \tag{4.1}$$

The three relaxation functions are now evaluated for this choice of the random local field. It is to be noted that the muon experiences a magnetic field which is the sum of the external field and the local random field.

A. Longitudinal relaxation function

When the integrals in Eq. (3.7) are performed using this Gaussian distribution function [see Eqs. (A1)–(A8)], the longitudinal relaxation function can be written in a form,

$$G^{\mathrm{KT}}_{\mathrm{L}}(t;B_{\mathrm{ex}})=G_{\mathrm{L}}(b)+\cos(bs)G^{C}_{\mathrm{L}}(s;b)+\sin(bs)G^{s}_{\mathrm{L}}(s;b)/b\ , \tag{4.2}$$

which emphasizes its oscillatory nature and its relation to the isolated dynamics given by Eq. (2.4). That is, it contains all three general dynamical time dependences. There is a static term since there is a component of the incoming polarization in the $\hat{B}_{\mathrm{ex}}$ direction, and there are cosine and sine terms since there are components of the random local field in the $\hat{n}_{\mathrm{ex}}$ and $\hat{m}_{\mathrm{ex}}$ directions. These latter transverse components give rise to the time dependence. Here, $b=B_{\mathrm{ex}}/\sqrt{2}B_0$ is the reduced external field and $s=\sqrt{2}B_0\bar{\gamma}_\mu t$ is the reduced time. In particular, for random-field spreads of a few gauss, this reduced time is of the order of microseconds. Equation (4.2) is an alternate, but equal, expression for the standard longitudinal Kubo-Toyabe relaxation function; see Eq. (A9). The static term,

$$G_{\mathrm{L}}(b)=1-[b-F(b)]/b^3\ , \tag{4.3}$$

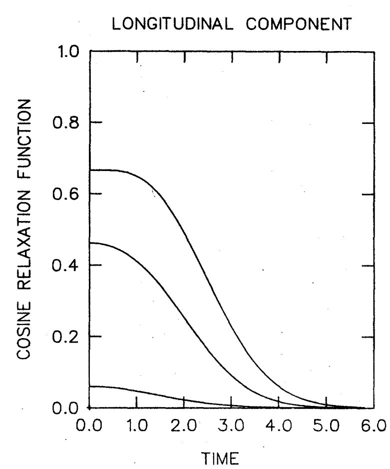

FIG. 1. Longitudinal cosine relaxation function. The reduced external magnetic fields b, for which these curves are evaluated, are 0.1 for the upper curve, 1.0 for the middle curve, and 4.0 for the lower curve. These values for the reduced external field are used in the subsequent figures. The relaxation functions and the times in this and subsequent figures are dimensionless.

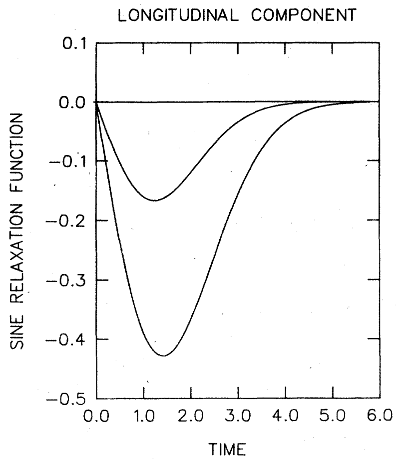

FIG. 2. Longitudinal sine relaxation function. The values of the reduced external fields increase from 0.1 through 1.0 to 4.0 from the lower to upper curves.

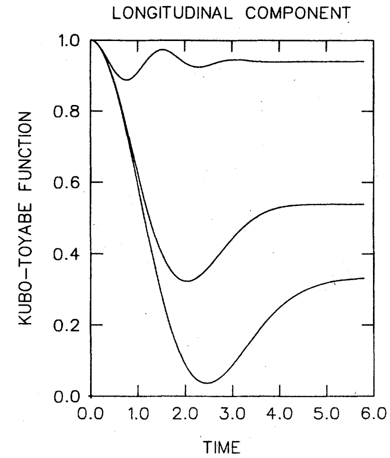

FIG. 3. Standard longitudinal Kubo-Toyabe relaxation function. Again, the values of the reduced external field increase from the lower to the upper curves. These are the standard longitudinal Kubo-Toyabe relaxation functions (Refs. 7—10) evaluated using Eq. (4.2).

involves Dawson's integral,[13]

$$F(b)=\exp(-b^2)\int_0^b dy\,\exp(y^2)\ , \tag{4.4}$$

which, for small b, reduces to $b-2b^2/3$. It is this term which is responsible for the recovery to one-third for zero fields. On the other hand, for large reduced external fields this static contribution approaches unity. The $\cos(bs)$ term involves a longitudinal cosine relaxation function.

$$G_L^C(s;b)=b^{-2}\exp(-s^2/4)[1-G_0^C(s;b)] \tag{4.5}$$

(see Fig. 1), where

$$G_0^C(s;b)=\int_0^1 dx\,\cos(bsx)\exp[-b^2x(2-x)] \tag{4.6}$$

is a function common to all three Kubo-Toyabe relaxation functions. This latter function reduces to

$$1-(2+\tfrac{1}{2}s^2)b^2/3$$

for small reduced external fields. Thus the longitudinal cosine relaxation function reduces to

$$\exp(-s^2/4)(2+\tfrac{1}{2}s^2)/3$$

for small external fields. On the other hand, this cosine function decays to zero for large reduced external fields. Equation (4.2) also involves the other general time dependence, namely $\sin(bs)$, with a longitudinal sine relaxation function

$$G_L^S(s;b)=-\exp(-s^2/4)G_0^S(s;b) \tag{4.7}$$

(see Fig. 2), where

$$G_0^S(s;b)=b^{-1}\int_0^1 dx\,\sin(bsx)\exp[-b^2x(2-x)] \tag{4.8}$$

is the other one-dimensional integral that is also common to all three Kubo-Toyabe relaxation functions. For small b this latter function approaches $\frac{1}{2}s$, while the longitudinal sine relaxation function becomes $-\frac{1}{2}s\exp(-s^2/4)$. The sine relaxation function also decays to zero for large reduced external fields.

In the limit of zero external field the longitudinal relaxation function, Eq. (4.2), reduces to the standard zero-field Kubo-Toyabe result,[7—10] namely

$$G_L^{KT}(t;B_{ex})=\tfrac{1}{3}+\tfrac{2}{3}(1-\tfrac{1}{2}s^2)\exp(-s^2/4)\quad\text{when } b=0\ . \tag{4.9}$$

All three terms in Eq. (4.2) contribute to this result. The longitudinal relaxation function is plotted for various values of the reduced external field in Fig. 3. For large fields, since both the sine and cosine relaxation functions approach zero, the longitudinal Kubo-Toyabe relaxation function approaches 1, as does the static contribution to it. When measured in the skewed coordinate frame defined by the counter geometry $\hat{X},\hat{Y},\hat{Z}$ $(\hat{n}_{ex},\hat{m}_{ex},\hat{B}_{ex})$, the longitudinal Kubo-Toyabe relaxation function $\overline{G}_L^{KT}(t;B_{ex})$, Eq. (3.11), is the product of Eq. (4.2) and $\cos\theta_{in}$. Thus, while the overall signal is reduced by $\cos\theta_{in}$, the relaxation function remains the same.

B. Coplanar transverse relaxation function

With the skewed coordinate frame it is also possible to simultaneously measure the two transverse relaxation functions. The first to be considered is the coplanar transverse relaxation function which lies in the plane of the external field and the incoming muon polarization, that is, in the $\hat{n}_{ex}$ $(\hat{X})$ direction. Again, using Eq. (4.1) in Eq. (3.8) [see Eq. (A10)], this coplanar transverse relaxation function,

$$G_{CT}^{KT}(t;B_{ex})=G_{CT}(b)+\cos(bs)G_{CT}^C(s;b)+\sin(bs)G_{CT}^S(s;b)/b\ , \tag{4.10}$$

can also be written in terms of all three time functionalities of the isolated motion; see Eq. (2.4). Since there is a

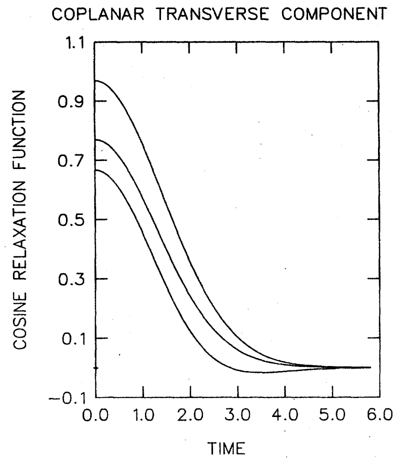

FIG. 4. Coplanar transverse cosine relaxation function. The values of the reduced external field increase from the lower to upper curve.

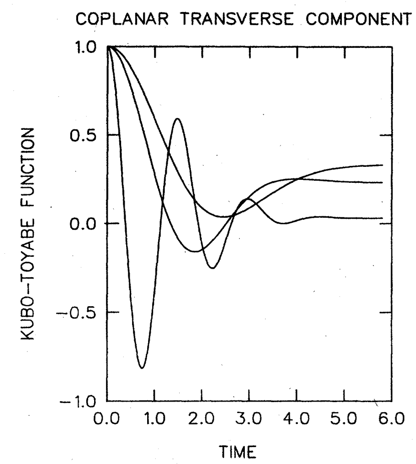

FIG. 6. Coplanar transverse Kubo-Toyabe relaxation function. For large times the reduced fields decrease from the lower to upper curves. The 0.1 reduced-field curve is essentially the standard zero-field Kubo-Toyabe relaxation function (Refs. 7–10).

component of the incoming polarization in the $\hat{n}_{ex}$ direction, then there is a static term, namely

$$G_{CT}(b)=\tfrac{1}{2}b^{-3}[b-F(b)] \ . \tag{4.11}$$

This function approaches one-third for small reduced external fields and zero for large reduced external fields. There is a $\cos(bs)$ term,

$$G^{C}_{CT}(s;b)=\tfrac{1}{2}b^{-2}\exp(-s^2/4)[2b^2-1+G^{C}_{0}(s;b)] \tag{4.12}$$

(see Fig. 4), and a $\sin(bs)$ term,

$$G^{S}_{CT}(s;b)=\tfrac{1}{2}\exp(-s^2/4)[G^{S}_{0}(s;b)-s] \tag{4.13}$$

(see Fig. 5), due to the external magnetic field.

The full coplanar transverse relaxation function is plotted for various values of the external reduced field in Fig. 6. It agrees with the numerical results of Kubo.[8] For small reduced fields this relaxation function reduces to the standard zero-field Kubo-Toyabe result, Eq. (4.9), while for large fields it has a Gaussian-damped cosine time dependence, namely

$$G^{KT}_{CT}(t;B_{ex})\sim\exp(-s^2/4)\cos(bs) \quad \text{as } b\rightarrow\infty \ . \tag{4.14}$$

When measured in the skewed coordinate frame the coplanar transverse Kubo-Tobaye relaxation function $\overline{G}^{KT}_{CT}(t;B_{ex})$, Eq. (3.11), is the product of Eq. (4.10) and $\sin\theta_{in}$. Thus, while the overall signal is reduced by $\sin\theta_{in}$, this transverse relaxation function varies from the standard zero-field Kubo-Toyabe function to the expected high-field transverse behavior, namely a Gaussian-damped cosine time dependence.

C. Perpendicular transverse relaxation function

The other transverse Kubo-Toyabe relaxation function lies in the $\hat{m}_{ex}(\hat{Y})$ direction, that is, in the direction perpendicular to the plane defined by the external field and

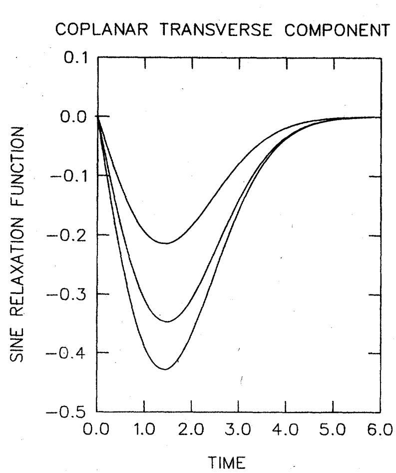

FIG. 5. Coplanar transverse sine relaxation function. The reduced field decreases from the lower to upper curve.

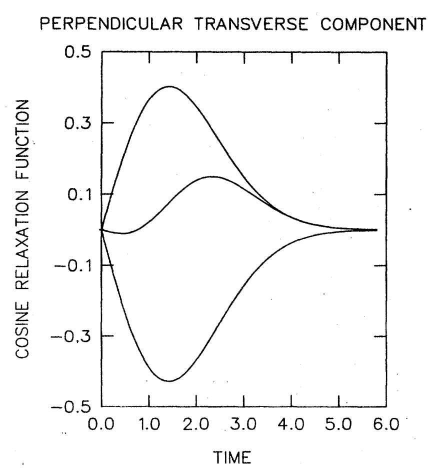

FIG. 7. Perpendicular transverse cosine relaxation function. The lower curve has $b=0.1$, while the upper curve has $b=4.0$ and the middle curve has $b=1.0$.

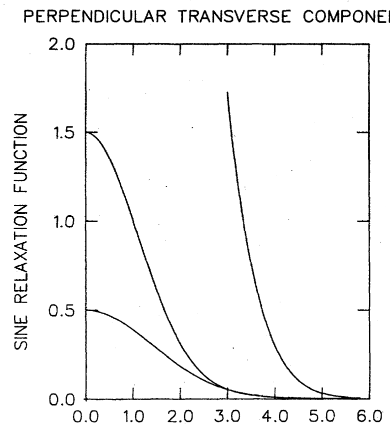

FIG. 8. Perpendicular transverse sine relaxation function. The values of the reduced field increase from the lower to upper curves. The value of the sine relaxation function for $b=4.0$ at zero time is 16.5

the incoming muon-spin polarization. This perpendicular transverse relaxation function can be written as

$$G_{\mathrm{PT}}^{\mathrm{KT}}(t;B_{\mathrm{ex}})=\cos(bs)G_{\mathrm{PT}}^{C}(s;b)/b+\sin(bs)G_{\mathrm{PT}}^{S}(s;b)/b^2 \tag{4.15}$$

[see Eq. (A11)]. It has no static component since there is no component of the incoming polarization in the $\hat{m}_{\mathrm{ex}}$ direction. The cosine and sine terms have the following relaxation functions:

$$G_{\mathrm{PT}}^{C}(s;b)=\tfrac{1}{2}s\exp(-s^2/4)[1-2G_0^C(s;b)] \tag{4.16}$$

and

$$G_{\mathrm{PT}}^{S}(s;b)=\exp(-s^2/4)\{\tfrac{1}{2}+b^2[1-sG_0^S(s;b)]\}\ , \tag{4.17}$$

respectively. Reduced external field dependences of these

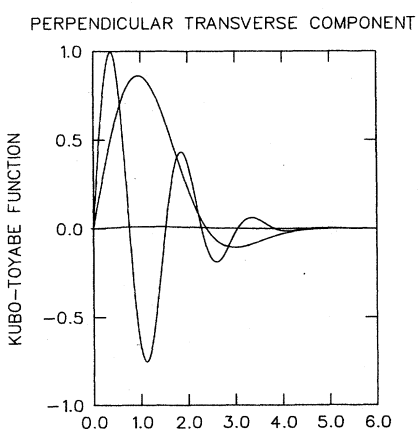

FIG. 9. Perpendicular transverse Kubo-Toyabe relaxation function. For $b=0.1$ the perpendicular transverse relaxation function is essentially zero. The curve whose first maximum appears at about unit time has $b=1.0$, while the highly oscillating damped curve has $b=4.0$.

functions are plotted in Figs. 7 and 8, while plots of the full perpendicular transverse relaxation function appear in Fig. 9. This full relaxation function reduces to zero for small fields and to a Gaussian-damped sine for high fields, namely

$$G_{\mathrm{PT}}^{\mathrm{KT}}(t;B_{\mathrm{ex}})\sim\exp(-s^2/4)\sin(bs)\ \text{ as } b\to\infty\ . \tag{4.18}$$

When measured in the skewed coordinate frame, the perpendicular transverse Kubo-Toyabe relaxation function $\overline{G}_{\mathrm{PT}}^{\mathrm{KT}}(t;B_{\mathrm{ex}})$, Eq. (3.11), is the product of Eq. (4.15) and $\sin\theta_{\mathrm{in}}$. Thus the overall signal is reduced by $\sin\theta_{\mathrm{in}}$ in this geometry. For large external fields the muon precesses about B_{ex} with the appropriate Larmor frequency. However, its magnitude is reduced by the Gaussian-damping function. In low or intermediate external fields the precession about B_{ex} is modulated by the local random field.

V. DISCUSSION

The advent of spin rotators,[5,6] with which the angle between the muon's spin polarization and its momentum can be varied, makes feasible the construction of a single apparatus that is suitable (without modification) for both longitudinal and transverse experiments. In such an apparatus, counters would be placed normal to the laboratory coordinates X,Y,Z with the external field aligned in the Z direction and with the incoming muon-spin polarization in the XZ plane. Three relaxation functions are observable, namely the longitudinal relaxation function (normal to the Z direction), the coplanar transverse relaxation function (normal to the X direction), and the perpendicular transverse relaxation function (normal to the Y direction). Traditional longitudinal experiments can be conducted when the incoming polarization lies in the Z direction, while, in separate experiments, traditional transverse experiments can be performed when this polarization lies in the X direction. On the other hand, both the longitudinal and transverse experiments can be performed simultaneously if the incoming polarization is set at an arbitrary angle in the XZ plane. Which of these alignments is chosen will depend on the particular sample of interest and on the information that is required.

To illustrate that the skewed-field alignment is of interest, new alternate analytic expressions, but equivalent to the standard literature formulas[7–10] and numerical results,[8] for the various relaxation functions, have been presented within static isotropic Kubo-Toyabe theory. These expressions, which are of the form

$$G(t;B_{\mathrm{ex}})=G(B_{\mathrm{ex}})+G^{C}(B_{\mathrm{ex}})\cos(\overline{\gamma}_{\mu}B_{\mathrm{ex}}t)+G^{S}(B_{\mathrm{ex}})\sin(\overline{\gamma}_{\mu}B_{\mathrm{ex}}t) \tag{5.1}$$

[see Eqs. (4.2), (4.10), and (4.15)], emphasize the physics. That is, they are combinations of appropriate isolated dynamical time dependencies multiplied by relaxation functions. For zero-field experiments with an arbitrary skew angle, the longitudinal and coplanar transverse observed relaxation functions will be $\cos\theta_{\mathrm{in}}$ and $\sin\theta_{\mathrm{in}}$ multiplied by the standard zero-field Kubo-Toyabe function, respectively. The perpendicular transverse relaxation function will be zero. For intermediate fields all three relaxation functions are different, while for high fields (much greater than B_0) the longitudinal relaxation function repolarizes and the two transverse relaxation functions are equal except for a simple phase shift of 90°. Such a situation then provides either a rather stringent test of isotropic static Kubo-Toyabe theory or a test of the accuracy of the apparatus for samples that obey isotropic static Kubo-Toyabe theory.

ACKNOWLEDGMENTS

I would like to thank the Schweizerisches Institut für Nuklearforschung (SIN) and the Physik-Institut der Universität Zürich for their financial assistance during my visit to Switzerland. I would also like to thank Professor Robert F. Snider and Professor Peter F. Meier for their help in writing this manuscript.

APPENDIX

The static contribution to the longitudinal Kubo-Toyabe relaxation function, Eq. (3.7), involves the function $C_1(B_{\rm ex})$, Eq. (3.3). Taking $\hat{B}_{\rm ex}$ as the $\hat{z}$ direction with $\cos\theta=\hat{B}\cdot\hat{B}_{\rm ex}$, this integral becomes

$$\begin{aligned} C_1(B_{\rm ex})&=(2\pi B_0^2)^{-3/2}\int_0^\infty dB\,B^2\int_0^\pi d\theta\sin\theta\int_0^{2\pi}d\phi\cos^2\theta\exp\{-[(B\cos\theta-B_{\rm ex})^2+B^2\sin^2\theta]/2B_0^2\}\\ &=(2/\sqrt{\pi})\int_{-1}^1 dx\,x^2\exp[-(1-x^2)b^2]\int_{-xb}^\infty dy\exp(-y^2)(y+bx)^2\,, \end{aligned}\tag{A1}$$

where the variables have been changed to $x=\cos\theta$ and $y=B/\sqrt{2}B_0-bx$. Here, $b=B_{\rm ex}/\sqrt{2}B_0$ is the reduced external field. Now the range of the x integral is separated into positive and negative regions. In the latter region, x is replaced by $-x$, while in the former, y is replaced by $-y$. The two integrals are summed so that the range of the y integral covers all space, that is,

$$\begin{aligned} C_1(B_{\rm ex})&=(2/\sqrt{\pi})\int_0^1 dx\,x^2\exp[-(1-x^2)b^2]\int_{-\infty}^\infty dy\exp(-y^2)(y-bx)^2\\ &=\int_0^1 dx\exp[-(1-x^2)b^2]x^2(1+2x^2b^2)=1-b^{-3}[b-F(b)]\,, \end{aligned}\tag{A2}$$

where $F(b)$ is Dawson's integral, Eq. (4.4).

To obtain the time-dependent terms in the longitudinal relaxation function, the integral $C_2(t;B_{\rm ex})$, Eq. (3.4), must be evaluated. Following the procedure for $C_1(B_{\rm ex})$, this integral becomes

$$\begin{aligned} C_2(t;B_{\rm ex})&=(2\pi B_0^2)^{-3/2}\int_0^\infty dB\,B^2\int_0^\pi d\theta\sin\theta\int_0^{2\pi}d\phi(1-\cos^2\theta)\cos(\overline{\gamma}_\mu Bt)\exp\{-[(B\cos\theta-B_{\rm ex})^2+B^2\sin^2\theta]/2B_0^2\}\\ &=(2/\sqrt{\pi})\int_0^1 dx(1-x^2)\exp[-(1-x^2)b^2]\int_{-\infty}^\infty dy\exp(-y^2)(y-bx)^2\cos[s(y-bx)]\\ &=\exp(-s^2/4)\int_0^1 dx\exp[-(1-x^2)b^2](1-x^2)\{\cos(sbx)[(1-\tfrac{1}{2}s^2)+2b^2x^2]-2sbx\sin(sbx)\}\\ &=b^{-3}\exp(-s^2/4)\{(1-\tfrac{1}{2}s^2)[b^2F_0^C(s;b)-F_2^C(s;b)]+2[b^2F_2^C(s;b)-F_4^C(s;b)]-2s[b^2F_1^S(s;b)-F_3^S(s;b)]\}\,, \end{aligned}\tag{A3}$$

where $s=\sqrt{2}B_0\overline{\gamma}_\mu t$ is the reduced time. The last form of this integral involves a series of integrals of the type

$$\begin{aligned} F_n^C(s;b)&=\exp(-b^2)\int_0^b dy\,y^n\exp(y^2)\cos(sy)\\ &=\tfrac{1}{2}\exp(-b^2)\int_0^b dy\,y^{n-1}\cos(sy)[d\exp(y^2)/dy]\\ &=\tfrac{1}{2}b^{n-1}\cos(bs)+\tfrac{1}{2}sF_{n-1}^S(s;b)-\tfrac{1}{2}(n-1)F_{n-2}^C(s;b) \end{aligned}\tag{A4}$$

and

$$\begin{aligned} F_n^S(s;b)&=\exp(-b^2)\int_0^b dy\,y^n\exp(y^2)\sin(sy)\\ &=\tfrac{1}{2}b^{n-1}\sin(bs)-\tfrac{1}{2}sF_{n-1}^C(s;b)-\tfrac{1}{2}(n-1)F_{n-2}^S(s;b)\,. \end{aligned}\tag{A5}$$

Making use of these definitions, Eq. (A3) becomes

$$C_2(t;B_{\rm ex})=b^{-3}\exp(-s^2/4)[b\cos(sb)-F_0^C(s;b)]\,.\tag{A6}$$

Furthermore, the integral $F_0^C(s;b)$ can be expressed in terms of $\cos(bs)$ and $\sin(bs)$, that is,

$$\begin{aligned} F_0^C(s;b)&=\exp(-b^2)\int_0^b dy\cos(sy)\exp(y^2)\\ &=\exp(-b^2)\int_0^b dx\cos[s(x-b)]\exp[(x-b)^2]\\ &=b\cos(bs)G_0^C(s;b)+b^2\sin(bs)G_0^S(s;b)\,, \end{aligned}\tag{A7}$$

where $G_0^C(s;b)$ and $G_0^S(s;b)$ are the cosine and sine relaxation functions defined in Eqs. (4.6) and (4.8), respectively. Thus the function $C_2(t;B_{\rm ex})$ becomes

$$C_2(t;B_{ex})=b^{-2}\exp(-s^2/4)\{\cos(bs)[1-G_0^C(s;b)]-b\sin(bs)G_0^S(s;b)]\} . \tag{A8}$$

The standard longitudinal Kubo-Toyabe expression,[7–10]

$$G_L^{KT}(t;B_{ex})=1-b^{-2}[1-\cos(bs)\exp(-s^2/4)]+(2b^3)^{-1}\int_0^s ds_1\exp(-s_1^2/4)\sin(bs_1) , \tag{A9}$$

can be derived from Eqs. (A1) and (A2) when the x integrals are performed before the y integrals.

To evaluate the coplanar transverse relaxation function, Eq. (3.8), the function $A_2(t;B_{ex})$, Eq. (3.4), is required. As with the previous integrals, this function can be written as

$$\begin{aligned}A_2(t;B_{ex})+C_2(t;B_{ex})&=2(2\pi B_0^2)^{-3/2}\int_0^\infty dB\,B^2\int_0^\pi d\theta\sin\theta\int_0^{2\pi}d\phi\cos(\overline{\gamma}_\mu Bt)\exp\{-[(B\cos\theta-B_{ex})^2+B^2\sin^2\theta]/2B_0^2\}\\&=2b^{-1}\exp(-s^2/4)[(1-\tfrac{1}{2}s^2)F_0^C(s;b)-2sF_1^S(s;b)+2F_2^C(s;b)]\\&=2b^{-1}\exp(-s^2/4)[b\cos(bs)-\tfrac{1}{2}s\sin(bs)] .\end{aligned} \tag{A10}$$

Finally, the perpendicular transverse relaxation function, Eq. (3.9), requires the $C_3(t;B_{ex})$ integral, Eq. (3.5), namely

$$\begin{aligned}C_3(t;B_{ex})&=(2\pi B_0^2)^{-3/2}\int_0^\infty dB\,B^2\int_0^\pi d\theta\sin\theta\int_0^{2\pi}d\phi\cos\theta\sin(\overline{\gamma}_\mu Bt)\exp\{-[(B\cos\theta-B_{ex})^2+B^2\sin^2\theta]/2B_0^2\}\\&=b^{-2}\exp(-s^2/4)[(1-\tfrac{1}{2}s^2)F_1^S(s;b)+2sF_2^C(s;b)+2F_3^S(s;b)]\\&=b^{-2}\exp(-s^2/4)\{\cos(bs)(\tfrac{1}{2}bs)[1-2G_0^C(s;b)]+\sin(bs)[b^2+\tfrac{1}{2}-sb^2G_0^S(s;b)]\} .\end{aligned} \tag{A11}$$

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