A numerical method to calculate the muon relaxation function in the presence of diffusion

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Abstract

We present an accurate and efficient method to calculate the effect of random fluctuations of the local field at the muon, for instance in the case of muon diffusion, within the framework of the strong collision approximation. The method is based on a reformulation of the Markovian process over a discretized time base, leading to a summation equation for the muon polarization function which is solved by discrete Fourier transform. The latter is formally analogous, though not identical, to the integral equation of the original continuous-time model, solved by Laplace transform. With real-case parameter values, the solution of the discrete-time strong collision model is found to approximate the continuous-time solution with excellent accuracy even with a coarse-grained time sampling. Its calculation by the fast Fourier transform algorithm is very efficient and suitable for real time fitting of experimental data even on a slow computer.

Keywords: muon spin rotation and relaxation, numerical methods, Markov processes, disordered magnetism

1. Introduction

One of the greatest benefits of muon spin rotation ($\mu$SR) as a local probe of magnetism in condensed matter is its capability of detecting randomly distributed static magnetic fields even in the absence of a net bias field. This makes $\mu$SR the technique of choice for the study of disordered magnets or weakly magnetic systems, such as spin and cluster-spin glasses. In most cases, purely static magnetic disorder is adequately accounted for by a classical distribution of random fields, such as spin and cluster-spin glasses. In most cases, purely static magnetic disorder is adequately accounted for by a classical distribution of random fields, whence the longitudinal muon spin polarization function $G(t) \equiv \langle S_z(t)S_z(0) \rangle / \langle S_z(0)^2 \rangle$ is calculated by averaging the precession waveform of a muon in a random local field over the muon ensemble. A Gaussian distribution, yielding the well-known static Kubo–Toyabe function [1], usually describes the muon depolarization by the static dipolar fields from nuclei or paramagnetic ions with sufficient accuracy. In non-dilute magnetic alloys, however, both detailed calculations and $\mu$SR experiments revealed non-Gaussian field distributions at the muon sites [2–5]. A consistent quantum mechanical treatment of the coupled muon-nuclear spin system, on the other hand, also predicts appreciable deviations from the simple Kubo–Toyabe behaviour [6–8], which may become dramatic in some circumstances [9, 10].

The effect of time-dependent fluctuations on an otherwise static random distribution of fields at the muon can be easily accounted for in the limit of very rapid fluctuations (the so-called narrowing limit), whereby they produce simple exponential muon spin relaxations. The intermediate case between the narrowing limit and purely static fields requires, however, a detailed modelling of the dynamical processes perturbing the instantaneous field at the muon. The simplest dynamical model, suitable e.g. to describe the effect of muon diffusion, is based on a strong collision approximation [11], clearly a very crude simplification as it neglects, for instance, the quantum mechanical correlations among spins [12]. Such a model deals with the muon spin evolution in the form of a Markovian process, yielding a recursion series for $G(t)$. The latter is summed into an integral equation, which is solved in principle by Laplace transform [13]. However, an exact analytical solution of the strong collision model (SCM) with an arbitrary distribution of static fields cannot be obtained.
In the most general case, including that of a Gaussian distribution corresponding to the dynamical Kubo–Toyabe function, Laplace transforms have to be calculated and inverted numerically. This makes the Laplace transform method impractical, especially when the free parameters of the model have to be optimized in order to fit experimental data. To overcome these difficulties, approximate solutions of the SCM, valid in a limited range of parameters, have been obtained [14, 15].

In this paper we illustrate an effective method, alternative to both numerical quadrature and approximate solutions, to solve the SCM and calculate \( G(t) \) for a generic static field distribution. The basic idea underlying the method is replacing Laplace integrals with discrete Fourier transforms (DFT). In the past, Weber et al showed that the Laplace transform-based solution can be mapped into an expression involving the Fourier transform, suitable to be approximated by DFT [16]. However, the naïve replacement of continuous-variable Fourier integrals with discrete sums, though yielding acceptable approximate solutions for small scattering frequencies, eventually leads to badly inaccurate results for large values of such a parameter. In order to correctly transpose the original integral equation into a summation equation, we recalculated the Markov chains directly and self-consistently over a discretized time base \( t_n \), in a so-called discrete-time SCM (DTSCM). The solution of the resulting equation by DFT provides an efficient and accurate algorithm to calculate \( G(t_n) \), suitable for real time fitting of experimental data.

The paper is organized as follows. The continuous-time SCM is recalled for reference in section 2. Its reformulation into a DTSCM is detailed in section 3. The application of the DTSCM as an effective calculation method is discussed in section 4.

2. The strong collision model

We recall here briefly the results of the original SCM applied to the muon spin evolution in a randomly distributed local field, due to Hayano et al [13]. The model postulates that after a ‘collision’ event, occurring with a probability \( \nu \) per unit time, the local field is a random variable totally uncorrelated with the local field before the collision, and governed by the same distribution (i.e. collisions map the static field distribution into itself). Based on these assumptions, it is legitimate to treat the muon spin ensemble, described by its polarization function \( G(t) \), as a single entity subject to collisions as a whole [13, 14]. \( G(t) \) will then evolve as the unperturbed static-field function \( G^{(0)}(t) \) with probability \( \exp(-\nu t) \), corresponding to no scattering event in the \((0, t)\) time interval; or it will resume as \( G^{(0)}(t_1)G^{(0)}(t-t_1) \exp(\nu(t-t_1)) \) with probability \( \nu \exp(-\nu t) \) \( dt_1 \) after a collision occurred in a time interval \( dt_1 \) around \( t_1 \), and so on. This leads to the following expansion in powers of \( \nu \):

\[
G(t) = e^{-\nu t} G^{(0)}(t) + \nu \int_0^t dt_1 e^{-\nu t_1} G^{(0)}(t_1) e^{-\nu(t-t_1)} G^{(0)}(t-t_1) \\
+ \nu^2 \int_0^t dt_1 e^{-\nu t_1} G^{(0)}(t_1) e^{-\nu(t-t_1)} G^{(0)}(t-t_1) \\
\cdot \int_t^{t_1} dt_2 e^{-\nu(t_2-t_1)} G^{(0)}(t_2-t_1) \\
\cdot e^{-\nu(t-t_2)} G^{(0)}(t-t_2) + \cdots ,
\]

which is rewritten into the following recursion series, upon changing the order of integration

\[
G(t) = e^{-\nu t} G^{(0)}(t) + \nu \int_0^t dt_1 H^{(0)}(t-t_1) G^{(0)}(t_1) \\
\cdot \left\{ e^{-\nu t_1} G^{(0)}(t_1) + \nu \int_0^{t_1} dt_2 e^{-\nu(t_2-t_1)} G^{(0)}(t_2-t_1) \\
\cdot \left\{ e^{-\nu t_2} G^{(0)}(t_2) + \nu \int_0^{t_2} dt_3 \cdots \right\} \right\} .
\]

From the comparison with the right hand side of (2), it is apparent that the expression in braces equals \( G(t_1) \). Defining \( H^{(0)}(t) \equiv G^{(0)}(t) \exp(-\nu t) \), the following one-dimensional Dyson-type equation is obtained for \( G(t) \):

\[
G(t) = H^{(0)}(t) + \nu \int_0^t dt_1 H^{(0)}(t-t_1) G^{(0)}(t_1)
\]

or

\[
G = H^{(0)} + \nu H^{(0)}G,
\]

where the convolution operator ‘*’ is defined as the integral in the rightmost term of (3a), i.e. as in the theory of Laplace transform. Equation (3b), actually a particular case of a Volterra integral equation of the second kind, is solved in principle by Laplace transformation. Let \( H^{(0)}(s) \equiv \mathcal{L}[H^{(0)}] \), \( G^{(0)}(s) \equiv \mathcal{L}[G^{(0)}] \), \( G(s) \equiv \mathcal{L}[G] \) be the Laplace transforms of \( H^{(0)}(t) \), \( H^{(0)}(t) \) and \( G(t) \), respectively; then [13]

\[
G(s) = \frac{H^{(0)}(s)}{1 - \nu H^{(0)}(s)} = \frac{G^{(0)}(s+v)}{1 - \nu G^{(0)}(s+v)} .
\]

An analytical expression for \( G(t) \) can be obtained from (4) in the case of a Lorentzian distribution of random static fields in zero external field, corresponding to a Lorentzian Kubo–Toyabe polarization function [17]

\[
G^{(L)}_L(t) = \frac{1}{3} + \frac{2}{3} (1-\Lambda t) e^{-\Lambda t},
\]

where \( \Lambda = \gamma_e/m_e \) is the distribution half width (\( \gamma_e \) is the muon gyromagnetic ratio). Its Laplace transform is straightforwardly calculated as

\[
G^{(L)}_L(s) = \frac{1}{3s} + \frac{2}{3(s+\Lambda)} - \frac{2\Lambda}{3(s+\Lambda)^2} .
\]

From (4) and (6), the \( s \)-domain dynamic function \( G_L(s) \) is a third-order rational function with non-degenerate poles \( \mathcal{R}(\nu, \Lambda) \) for \( \nu \neq 0 \), which is decomposed into a sum of simple
fractions of the form
\[ G_L(s) = \sum_{k=1}^{3} \frac{C_k(\nu, A)}{s - P_k(\nu, A)} \] (7)
whence the time-domain function \( G_L(t) \) is the superposition of three exponentials
\[ G_L(t) = \sum_{k=1}^{3} C_k e^{\nu_k t}. \] (8)

The application of the SCM to a Lorentzian random field distribution is a rather academic exercise. In most situations of practical interest the field distribution is instead Gaussian, as in the case of the stray dipolar fields from nuclei, which produce a muon depolarization following the static Kubo–Toyabe function \([1]\) at low temperature
\[ G_{KT}^{(0)}(t) = \frac{1}{3} + \frac{2}{3} \left(1 - \Delta^2 r^2\right) e^{-\frac{1}{2} \lambda^2 t^2}, \] (9)
where \((\Delta / t_0)^2\) is the second moment of each Cartesian component of the local field \([18]\). In this context the SCM correctly describes the effect of the thermally activated diffusion of the muon on its spin relaxation, following a so-called dynamic Kubo–Toyabe function. On the other hand, a Lorentzian field distribution is typically encountered in dilute disordered magnetic systems, where it mimics a distribution of Gaussian widths \(\Delta\) due to a corresponding distribution of muon coupling constants to the magnetic centres. In these systems, the effect of a Markovian dynamics is incorrectly described by \((3a)\) with \(G^{(0)}\) given by \((5)\). Rather, the correct muon polarization function is obtained by averaging the dynamic Gaussian Kubo–Toyabe functions appropriate for each spatial configuration of the muon (in other words, dynamization and spatial averaging do not commute, and the former has to be applied first) \([19, 20]\). Despite its questionable physical significance, the main interest of \((8)\) is providing here a formally exact solution of the continuous-time SCM to be used as a benchmark for the DTSCM developed in the next section. An analytic expression for the dynamic Kubo–Toyabe function \(G_{KT}(t)\) analogous to \((8)\) cannot be obtained in fact from \((9)\) and \((4)\), therefore \(G_{KT}(t)\) has to be calculated numerically.

3. The discrete-time strong collision model

We now modify the original SCM sketched in the previous section 1, by imposing that scattering events may occur only at discrete times \(t_n = n \tau\), where \(n\) is a non-negative integer. Let \(\lambda\) be the probability that a collision occurs over the finite time lag \(\tau\), and \(q = 1 - \lambda\) the complementary probability. According to the above definitions, \(\lambda = 1 - \exp(-\nu \tau)\), while the probability that no collision occurs over a time \(t_n\) equals \(q^n = \exp(-n \nu \tau)\). Following a similar argument as for the continuous-time case, the muon spin polarization \(G_n \equiv G(t_n)\) will be the unperturbed \(G^{(0)}\) with probability \(q^n\); or it will be given by the free evolution to \(t_k\) with probability \(q^{k-1}\), followed by the free evolution to \(t_n\) with probability \(q^{n-k}\), in the case of a single collision occurring at a non-zero time \(t_k \leq t_n\) with probability \(\lambda\), each \(k\) thus contributing a term \(G^{(0)}_k G^{(0)}_k\) with probability \(\lambda q^{k-1}\); and so on. We are thus led to write the following equations for the Markov chain:

\[ G_0 = G^{(0)}_0 = 1 \]
\[ G_1 = q^{1} G^{(0)}_1 + \lambda q^{0} G^{(0)}_1 \cdot q^{0} G^{(0)}_0 \]
\[ G_2 = q^{2} G^{(0)}_2 + \lambda \left(q^{1} G^{(0)}_1 \cdot q^{1} G^{(0)}_1 + q^{2} G^{(0)}_2 \cdot q^{0} G^{(0)}_0\right) \]
\[ + \lambda^2 q^{0} G^{(0)}_1 \cdot q^{0} G^{(0)}_1 \cdot q^{0} G^{(0)}_0 \]
\[ \cdots \]
\[ G_n = q^{n} G^{(0)}_n + \lambda \sum_{k=0}^{n-1} q^{k} G^{(0)}_{n-k+1} \cdot q^{n-k-1} G^{(0)}_{n-k+1} \]
\[ + \lambda^2 q^{0} G^{(0)}_{n-k+1} \cdot q^{0} G^{(0)}_{n-k+1} \cdot q^{0} G^{(0)}_{n-k+1} \]
\[ \cdots \] (10)

Defining \(H^{(0)}_n \equiv q^{n} G^{(0)}_n = G^{(0)}_n \exp(-n \nu \tau)\) as in the continuous-time case, and taking into account that \(H^{(0)}_0 = 1\), equation \((10)\) is straightforwardly rewritten as

\[ G_n = H^{(0)}_n + \lambda \sum_{k=0}^{n-1} H^{(0)}_{k+1} + H^{(0)}_{n-k-1} \]
\[ + \lambda^2 \sum_{k=0}^{n-1} \sum_{h=0}^{n-k-2} H^{(0)}_{k+1} \cdot H^{(0)}_{k+1} \cdot H^{(0)}_{n-k-h-2} + \cdots \] (11)

whence, upon factoring the outermost summation, a recursive series is obtained, analogous to \((2)\)

\[ G_n = H^{(0)}_n + \frac{\lambda}{q} \sum_{k=1}^{n} H^{(0)}_{k+1} \left[H^{(0)}_{n-k-1} + \lambda \sum_{h=0}^{n-k-2} H^{(0)}_{h} + H^{(0)}_{n-k-h-2} \right] \]
\[ + \left(\frac{\lambda}{q}\right)^2 \sum_{k=1}^{n-1} \sum_{h=1}^{n-k-3} H^{(0)}_{k+1} + H^{(0)}_{k+1} \times H^{(0)}_{n-k-h-j-3} + \cdots \} \] (12)

Upon recognizing that the expression within braces in \((12)\) equals the expansion for \(G_{n-k-1}\) as of \((11)\), we obtain the following summation equation

\[ G_n = H^{(0)}_n + \frac{\lambda}{q} \sum_{k=1}^{n} H^{(0)}_{k+1} \]
\[ \equiv H^{(0)}_n + \frac{\lambda}{q} \sum_{k=1}^{n} \left( H^{(0)}_{k+1} - \delta_{k,0} \right) G_{n-k} \] (13a)

or

\[ G = H^{(0)} + \frac{\lambda}{q} \text{conv}\left((H^{(0)} - \delta), G\right), \] (13b)

where \(\delta_n \equiv \delta_{n,0}\), and the \(\text{conv()}\) operator is defined as

\[ \text{conv}(A, B)_n \equiv \sum_{k=0}^{n} A_k B_{n-k}, \] (14)

formally analogous to the convolution operator \(*\) defined in section 2.

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Equation (13a) (as well as its continuous-variable counterpart (3a)) exhibits a remarkable invariance by exponential weighting. Let $G$ be the solution of (13a); then, the same equation holds also for the exponentially weighted quantities $\tilde{G}^{(0)}_n \equiv H^{(0)}_n \exp (-\alpha n)$, $\tilde{G}_n \equiv G_n \exp (-\alpha n)$:

$$\tilde{G} = \tilde{H}^{(0)} + \frac{2}{q} \text{conv}\left( \tilde{H}^{(0)} - \delta, \tilde{G} \right).$$ (15)

Despite the formal similarity between equations (3b) and (13b), an exact closed expression in terms of $H^{(0)}$, analogous to (4), cannot be obtained in the discrete time case. Indeed, the $\text{conv}()$ operator defined in (14) is not transformed into a product by DFT (denoted hereafter as $F()$). Rather, it is circular convolution, defined as

$$(A \ast B)_n \equiv \sum_{k=0}^{N-1} A_k B_{(n-k) \mod \mathbb{N}},$$ (16)

where $N$ is the dimension of the discretized time base and $r = \text{mod} (m, N)$ is the remainder of $m$ modulo $N$ ($0 \leq r < N$), which is transformed into a product [21]: $F\{A \ast B\} = F\{A\} F\{B\}$. Nonetheless, non-circular convolution (14) may be reduced to circular convolution (16) by doubling the space dimension and padding vectors with $N$ trailing zeros. Let $A, B$ be arbitrary vectors, $A^p, B^p$ the corresponding zero-padded vectors, defined as

$$A^p_n = \begin{cases} A_n & 0 \leq n < N \\ 0 & N \leq n < 2N \end{cases},$$

etc, and let $U$ be the zero-padded unit: $U_n = 1$ for $n < N$, $U_n = 0$ for $n \geq N$. Then

$$U_n \text{conv}(A^p, B^p)_n = U_n(A^p \ast B^p)_n.$$ (17)

Henceforth we implicitly consider a doubled space dimension and zero-padded $G^{(0)}, H^{(0)}$ vectors, with the $P$ superscript dropped for simplicity of notation. It is intended that only vector elements with indices $n < N$ are physically meaningful. Multiplying both sides of (13b) by $U$, substituting (17) therein, and taking into account that $U_n A^p_n = A^p_n$, we then obtain

$$G - qH^{(0)} = \lambda \left( H^{(0)} \ast G \right) + R$$

$$\left[ R_n \equiv (1 - U_n) \left( G_n - \lambda \left( H^{(0)} \ast G \right)_n \right) \right]$$ (18a)

and a similar equation for the exponentially weighted quantity $\tilde{G}$

$$\tilde{G} - \tilde{q}H^{(0)} = \lambda \left( \tilde{H}^{(0)} \ast \tilde{G} \right) + R'$$

$$\left[ R'_n \equiv (1 - U_n) \left( \tilde{G}_n - \lambda \left( \tilde{H}^{(0)} \ast \tilde{G} \right)_n \right) \right].$$ (18b)

Due to the $R$ term on the right hand side of (18a), a closed exact expression for $F\{G\}$ as a function of $F\{H^{(0)}\}$ cannot be obtained yet. In the analogous (18b) for $\tilde{G}$, however, the ‘error’ $R'$ can be made arbitrarily small by an arbitrarily large weighting exponent $\alpha$. This suggests that the approximate solution $\tilde{G}'$ of (18b) obtained by dropping $R'$ is asymptotically exact. A more rigorous proof that $G_n \equiv \tilde{G}'_n \exp (\alpha n)$ actually tends to $G_n$ for $\alpha \to \infty$ is outlined in appendix A. The approximate solution $F\{\tilde{G}'_n\}$ is then straightforwardly written in the following closed form

$$F\{\tilde{G}'\} = \frac{qF\{\tilde{H}^{(0)}\}}{1 - q\lambda F\{\tilde{H}^{(0)}\}} \approx F\{G\}.$$ (19)

Equation (19), which is the discrete-time analogous to (4), is the main result of this paper. Its practical implementation in an effective calculation algorithm for $G(t)$ is discussed in the next section.

4. Application of the DTSCM method

Summarizing the above results, the muon longitudinal polarization function $G_n = G(n)$, $0 \leq n < N$ in the DTSCM approximation is calculated as

$$G_n \approx G'_n = e^{\alpha n} F\left\{ \frac{e^{-i\tau n} F(e^{-i\lambda \tau n} G^{(0)}_m)}{1 - (1 - e^{-i\lambda \tau}) F(e^{-i\tau n} G^{(0)}_m)} \right\}_n$$ (20)

with $G^{(0)}_m, 0 \leq m < 2N$ being the zero-padded static relaxation function defined such that $G^{(0)}_m = 0$ for $m \geq N$, and $F^{-1}\{\}$ the inverse DFT. The weighting coefficient $\alpha$ is a large-enough positive quantity, whose practical choice is discussed in the following. In high-level mathematics-oriented computer programming languages such as Matlab or Octave, which provide the fast Fourier transform (FFT) built-in or in a standard library, (20) is implemented by just a few lines of code.

The accuracy of $G'_n$ as an approximation for the exact solution $G_n$ of (13b) depends critically on the proper tuning of the exponential weighting. While in principle $G'_n \to G_n$ tends to zeros as $\exp (-2\pi N\alpha)$ for $\alpha \to \infty$ (see appendix A), an exceedingly large value of the weighting coefficient $\alpha$ leads in practice to numerical overflow. On the other hand, a too small $\alpha$ brings about an error which may become very large in some particular cases. In order to guide a convenient choice of $\alpha$ in (20), we compared $G'$ with the exact solution $G$ of the DTSCM corresponding to the static Kubo–Toyabe function (9) for several values of $\alpha, \Delta$, and $\nu$. The exact $G \equiv G_{\text{KT}}$ can be calculated in the general case from the expansion (10) (or equivalently (A3)) in powers of $\lambda/\tau$. We stress however that the summation of the series (A3) constitutes a quite inefficient algorithm, as its numerical convergence requires up to nearly as many terms as $N$ for large $\nu$ values. In the limit case $\Delta = 0$ (i.e. $G^{(0)}_n \equiv 1$) it is apparent from (10) that identically $G_n = 1$, in agreement with the observation that dynamics cannot alter the muon polarization in the absence of an internal field. The numerical tests were performed in standard double precision IEEE-754 floating point arithmetics [22] on an Intel-based personal computer running Matlab. The mean value and

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1 Sample routines running under Matlab and Octave are made available online in the supplemental materials accompanying this paper.
standard deviation of $G'_n - G_n$, $0 \leq n < N = 8192$, are listed in table 1 for a few representative parameter values. The best accuracy, approximately $10^{-10}$, is obtained for $\alpha$ in the order of $10/N$, while for $\alpha > 20/N$ the calculation of $G'_n$ is increasingly afflicted by floating point truncation errors, up to a numerical divergence at $\alpha \geq 40/N$. This trend was reproduced in all our simulations. Based on these figures, we chose $\alpha = 10/N$ as a convenient setting in all the following examples.

If implemented by the FFT algorithm, the DTSCM-based method is inherently very fast. The arithmetic complexity of FFT is in fact in the order of $N \log(N)$, while the direct numerical integration of equation (3a) scale as $N^2$. On large datasets, our method is therefore expected to outperform the latter. We benchmarked the computational efficiency of (20) by calculating the zero-field (ZF) dynamic Kubo–Toyabe function for the same $\nu/\Delta$ values as in figure 3(a) of Hayano et al [13] on several personal computers. The $G_{\text{KT}}(t)$ curves, calculated on an array of $N=8192$ sampling points (the typical histogram length e.g. of the datasets from the Paul Scherrer Institute muon facility), are plotted in figure 1. The calculation in Matlab took a 0.4 s overall CPU time on an AMD Athlon processor at 750 MHz dating back to year 2000, and approximately one-tenth of that on a recent PC (Pentium G2030 CPU at 3.0 GHz). A fit of real $\mu$SR data, requiring typically several hundred function calls, can therefore be performed by means of (20) virtually in real time even on a very low-end computer. For reference, the numerical solver of general Volterra equations from the Numerical Recipes [23], employed for instance by Keren [24], was found to run nearly 1000 times slower on the same time base, without, however, producing more accurate results.

**Table 1.** Average value and standard deviation of $G'_n - G_n$, where $G'_n$ and $G_n$ are, respectively, the approximate (20) and exact (10) dynamic functions, calculated on $N = 8192$ sampling points from the static Kubo–Toyabe function (9) in the DTSCM, for selected $\alpha$, $\nu$, and $\Delta$ values.

| $\alpha N$ | $\nu \tau$ | $\Delta \tau$ | $G'_n - G_n$ | $\sigma_{G'_n - G_n}$ |
|------------|------------|--------------|---------------|----------------------|
| 0          | 0.001 0    | 0            | $2.2 \times 10^2$ | $1.4 \times 10^{-1}$ |
| 0          | 0.01 0.001 | 0            | $2.0 \times 10^2$ | $9.4 \times 10^{-3}$ |
| 0          | 0.1 0      | 0            | $8.7 \times 10^1$ | $1.4 \times 10^{-1}$ |
| 2          | 0.001 0    | 0            | $1.9 \times 10^2$ | $1.2 \times 10^{-3}$ |
| 2          | 0.01 0.001 | 0            | $3.6 \times 10^4$ | $1.7 \times 10^{-7}$ |
| 2          | 0.1 0      | 0            | $1.9 \times 10^2$ | $1.4 \times 10^{-13}$ |
| 5          | 0.001 0    | 0            | $4.5 \times 10^3$ | $3.0 \times 10^{-8}$ |
| 5          | 0.01 0.001 | 0            | $8.9 \times 10^{-7}$ | $4.1 \times 10^{-7}$ |
| 5          | 0.1 0      | 0            | $4.5 \times 10^{-5}$ | $2.0 \times 10^{-13}$ |
| 10         | 0.001 0.001| 0            | $4.0 \times 10^{-11}$ | $1.9 \times 10^{-11}$ |
| 10         | 0.1 0      | 0            | $2.1 \times 10^{-9}$ | $1.2 \times 10^{-11}$ |
| 20         | 0.001 0    | 0            | $4.1 \times 10^{-10}$ | $3.1 \times 10^{-9}$ |
| 20         | 0.01 0.001 | 0            | $3.9 \times 10^{-30}$ | $2.5 \times 10^{-9}$ |
| 20         | 0.1 0      | 0            | $1.5 \times 10^{-9}$ | $7.2 \times 10^{-9}$ |
| 50         | 0.001 0    | 0            | $7.1 \times 10^{-2}$ | $1.1 \times 10^{-4}$ |
| 50         | 0.01 0.001 | 0            | $1.4 \times 10^{-1}$ | $2.6 \times 10^{-4}$ |
| 50         | 0.1 0      | 0            | $5.9 \times 10^{-3}$ | $3.3 \times 10^{-4}$ |

The applicability of (20) is clearly not limited to the ZF Gaussian Kubo–Toyabe case. Like the original continuous-time SCM, the DTSCM-based method can be applied to calculating the longitudinal muon spin polarization in the presence of any static field distribution, in either zero or a finite external longitudinal field (LF). An example is provided by figure 2, plotting the dynamic LF Gaussian Kubo–Toyabe function, calculated from the corresponding static function [13]

$$G^{(0)}(t) = 1 - \frac{2 \Delta \omega}{\omega_L^2} \left( 1 - e^{-\frac{\Delta \omega^2}{2 \omega_L^2} \cos \omega_L t} \right)$$

$$+ \frac{2 \Delta \omega}{\omega_L^2} \int_0^t dt' e^{-\frac{\Delta \omega^2}{2 \omega_L^2} \sin \omega_L t'}$$

for the same LF values $B_0 = \omega_L / \gamma_\mu$ as in figure 2 of Keren.
In the present figure, the DTSCM results are overlaid to the Keren approximant

\[ G^{(0)}(t) = \exp \left\{ -\frac{2\Delta^2}{(\omega_L^2 + \nu^2)} \left[ (\omega_L^2 + \nu^2) \nu t + \left( \omega_L^2 - \nu^2 \right) \left( 1 - e^{-i\nu t} \cos \omega_L t \right) - 2\nu \omega_L e^{-i\nu t} \sin \omega_L t \right] \right\} \]

(22)

which is known to tend to the exact SCM solution at early times [15]. The numerical evaluation of the analytical expression (22) is obviously faster than (20), by just a factor of ten in this example \((N = 8192)\), however. Incidentally, it is apparent from figure 2 that, in large applied fields, (22) is a globally better approximation of the exact polarization function than it appeared in [15], clearly due to an inaccurate numerical calculation of the LF dynamic Kubo–Toyabe function therein.

The accuracy of the discrete-time approximation with a reasonable sampling interval, possibly identical to the native experimental resolution in the time-differential data, is another issue of our method. To this end, we tested the DTSCM solution against analytical or approximate solutions of the continuous-time SCM in two cases. The first benchmark is provided by the SCM in the presence of a Lorentzian field distribution, whose exact solution is given by (8). The polarization function \(G_L(t)\) is plotted in figure 3 for several values of the scattering frequency \(\nu\). The discrete-time \((N = 512)\) and continuous-time solutions \(G_L^{(DT)}, G_L^{(CT)}\) are practically overlapped and undistinguished in the plot. In spite of the rather coarse time sampling, their difference \(G_L^{(DT)}(t) - G_L^{(CT)}(t)\) (figure inset) is negligible for practical purposes even at comparatively high \(\nu\). For reference, the experimental relative uncertainty on the muon polarization in very-high-statistics measurements is considerably smaller than a few permil.

Another well-known case is the Gaussian field distribution in the extreme narrowing limit \(\nu \gg \Delta\). Its relaxation function is approximated by the so-called Abragam formula [14, 15, 25]

\[ G^{AF}(t) = \exp \left\{ -2\Delta^2 \nu^{-2} \left( \exp \left( -\nu t \right) - 1 + \nu t \right) \right\} \]

(23)

(indeed, a limiting case of (22) for \(\omega_L = 0\)) which is asymptotically exact for \(\nu/\Delta \to \infty\). The accuracy of (20) in reproducing (23) is exemplified in figure 4, showing simulations obtained by the DTSCM at \(\nu = 320\Delta\) and different time resolutions. Here again, the difference \(G^{(DT)}(t) - G^{AF}\) between the discrete-time and the continuous-time solution given by the Abragam approximate formula is negligible even with a relatively coarse-grained sampling, corresponding to a cumulative scattering probability over a time bin \(1 - \exp \left( -\nu t \right)\) on the order of 10%.

5. Conclusions

In conclusion, we have demonstrated an accurate and efficient numerical method to calculate the dynamical Kubo–Toyabe function describing the longitudinal muon polarization function \(G(t)\) versus time in the presence of muon diffusion as well as, in principle, the solution of the SCM for an arbitrary distribution of static internal fields. The error on \(G(t)\) produced by time discretization is found to be much smaller than the experimental uncertainty even with very coarse time resolutions, and data oversampling is not needed. If implemented by means of the FFT algorithm, the method requires negligible CPU resources, which makes it suitable to fit experimental data in real time even on a slow computer.
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Appendix A. Proof that $G'$ tends to $G$ for $\alpha \to \infty$

We sketch here the proof that $G'$, defined by (20), is an asymptotically exact solution of (13b) for $\alpha \to \infty$. It is easily shown that the exponentially weighted function $G'_n$ (0 $\leq n < 2N$) defined by (19) obeys the equation

$$G' = \hat{H}^{(0)} + \frac{\lambda}{q} (\hat{H}^{(0)} - \hat{\delta}) + G'$$  \hspace{1cm} (A.1)

formally identical to (15) but for the replacement of non-circular with circular convolution. Upon defining $\hat{K}^{(0)} = \hat{H}^{(0)} - \hat{\delta}$, the following series expansion is straightforwardly obtained from (A1)

$$G' = \hat{H}^{(0)} + \frac{\lambda}{q} \text{conv} \left( \hat{K}^{(0)}, \hat{H}^{(0)} \right)$$

$$+ \frac{\lambda^2}{q^2} \text{conv} \left( \hat{K}^{(0)}, \text{conv} \left( \hat{K}^{(0)}, \hat{H}^{(0)} \right) \right)$$

$$+ \frac{\lambda^3}{q^3} \text{conv} \left( \hat{K}^{(0)}, \text{conv} \left( \hat{K}^{(0)}, \text{conv} \left( \hat{K}^{(0)}, \hat{H}^{(0)} \right) \right) \right) + \cdots$$

$$\equiv \sum_{m=0}^{\infty} \tilde{C}^{(m)}$$  \hspace{1cm} (A.2)

where the summation indices are upper-limited to $N - 1$ since $\hat{K}^{(0)}$ is a zero-padded vector. On the other hand, $\tilde{C}^{(m)}$ is calculated from (A3) as

$$\tilde{C}^{(m)} = \frac{\lambda^m}{q^m} \sum_{k=0}^{N-1} \sum_{s=0}^{N-1} \sum_{r=0}^{N-1} \hat{K}^{(0)} \cdots \hat{K}^{(0)} \hat{H}^{(0)} \delta_{n-k-s-t}$$  \hspace{1cm} (A.8)

for $m > N$ (see (10)).

The series (A2) is absolutely convergent for any positive $\alpha$. Its mth term $\tilde{A}^{(m)}$ clearly obeys the recursion relation

$$\tilde{A}^{(m+1)} = \frac{\lambda}{q} \hat{K}^{(0)} \tilde{A}^{(m)}.$$  \hspace{1cm} (A.4)

Taking into account that $|\hat{K}^{(0)}| \leq \exp (-\pi \alpha\nu \tau_0)$, from (A4) we can set the following upper bound

$$|\tilde{A}^{(m+1)}| \leq \frac{\lambda}{q} \sum_{k=0}^{N-1} \sum_{s=0}^{N-1} \max \tilde{A}^{(m)}$$

whereby, by induction, $|\tilde{A}^{(m)}| < A \exp (-\alpha m)$ with $A$ being a suitable positive constant. This ensures the absolute convergence of the series.

We now evaluate the difference $\tilde{E}_n \equiv \tilde{G}_n - \tilde{G}_n$ (0 $\leq n < N$) term by term from (A2) and (A3)

$$\tilde{E}_n = \sum_{m=2}^{\infty} \tilde{C}^{(m)} = \sum_{m=2}^{\infty} \tilde{A}^{(m)} - \tilde{A}^{(m)},$$  \hspace{1cm} (A.6)

where the first non-zero term in the sum is $m = 2$, since $\tilde{A}_n^{(1)} = \tilde{C}_n^{(1)}$ owing to (17). The mth term $\tilde{A}_n^{(m)}$ in (A2) is expressed as

$$\tilde{A}_n^{(m)} = \frac{\lambda^m}{q^m} \sum_{k=0}^{N-1} \sum_{s=0}^{N-1} \sum_{r=0}^{N-1} \hat{K}^{(0)} \cdots \hat{K}^{(0)} \hat{H}_n^{(0)}$$  \hspace{1cm} (A.7)

on the other hand, $\tilde{C}_n^{(m)}$ is calculated from (A3) as

$$\tilde{C}_n^{(m)} = \frac{\lambda^m}{q^m} \sum_{k=0}^{N-1} \sum_{s=0}^{N-1} \sum_{r=0}^{N-1} \hat{K}^{(0)} \cdots \hat{K}^{(0)} \hat{H}_n^{(0)}$$  \hspace{1cm} (A.8)

such that

$$\tilde{\alpha}^{(m)} = \tilde{\alpha}^{(m)}$$

$$\tilde{\alpha}^{(m+1)} = \frac{\lambda}{q} \hat{K}^{(0)} \tilde{\alpha}^{(m)}.$$  \hspace{1cm} (A.4)

Taking into account that $|\hat{K}^{(0)}| \leq \exp (-\pi \alpha\nu \tau_0)$, from (A4) we can set the following upper bound

$$|\tilde{\alpha}^{(m+1)}| \leq \frac{\lambda}{q} \sum_{k=0}^{N-1} \sum_{s=0}^{N-1} \max \tilde{\alpha}^{(m)}$$

whereby, by induction, $|\tilde{\alpha}^{(m)}| < A \exp (-\alpha m)$ with $A$ being a suitable positive constant. This ensures the absolute convergence of the series.
\\[\sum_{\lambda \delta} \sum_{\alpha \nu} \left( \sum_{\lambda \delta} \sum_{\alpha \nu} \right) \]

\( \delta_{4N+n-k, \alpha+1} \sum_{\lambda \delta} \sum_{\alpha \nu} \sum_{\lambda \delta} \sum_{\alpha \nu} \)

\( \frac{q^m}{m^2} \exp(-(\alpha+2N)(\alpha+1)) \)

\( \times \sum_{h=1}^{nN} N(m, n + 2hN) \exp(-2(\alpha+1)(\alpha+1)) \),

(A.12)

where \( N(m, n) \) denotes the number of combinations whereby the sum of \( m + 1 \) integer addends each in the range \([0, N-1]\) may yield \( n \). It follows that the unweighted difference \( E_n = G_n - G_n = \tilde{E}_n \exp(an) \) is bounded as

\[ |E_n| \leq \exp(-2N\alpha) \exp(-2N\alpha) \]

\( \times \sum_{m=2}^{\infty} \frac{2^m}{m^2} \sum_{h=1}^{m/2} N(m, n + 2hN) \exp(-2(\alpha+1)(\alpha+1)) \)

(A.13)

where the expression on the right hand side tends to zero for \( \alpha \to \infty \), since the series (which is convergent in view of (A5)) is a decreasing function of \( \alpha \), while its prefactor vanishes. Therefore \( \lim_{\alpha \to \infty} E_n = 0 \).

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