Langevin spin dynamics

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Microscopic stochastic Langevin-type spin dynamics equations provide a convenient and tractable model describing the relaxation of spin and spin-lattice ensembles. We develop a robust and numerically stable algorithm for integrating the Langevin spin dynamics equations, and explore, both numerically and analytically, a range of applications of the method. We show that the algorithm conserves the magnitude of the spin vector irrespectively of the amplitude of the thermal noise. Using the Furutsu-Novikov theorem, we derive a system of deterministic differential equations for the ensemble-average moments of solutions of the Langevin spin dynamics equations, and explore the dynamics of relaxation of a spin ensemble toward the equilibrium Gibbs distribution. Analytical solutions of the moments equations make it possible to estimate the time scales of spin thermalization and spin-spin self-correlation, which we investigate as functions of the damping parameter and temperature.

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I. INTRODUCTION

Langevin dynamics is a subject with long history, with the treatment of Brownian motion being among the well known applications of the method.[1–3] By introducing fluctuation and dissipation forces in the Hamiltonian equations of motion, one can model the dynamics of relaxation in an ensemble of interacting particles evolving toward thermal equilibrium. The relation between the stochastic and dissipative Langevin terms stems from the fluctuation-dissipation theorem (FDT),[1–3] in accord with the requirement that in thermal equilibrium the population of states in the phase space is given by the Gibbs distribution. There is an extensive literature on the numerical integration algorithms for Langevin dynamic systems of interacting particles[4–9] and on the statistical mechanics aspects of the problem.[1–3]

A similar approach can be applied to the investigation of a spin ensemble. In a method proposed by Brown,[10] a system of spins is driven toward equilibrium by suitably chosen fluctuation and dissipation terms. The treatment also makes it possible to model equilibrium thermal fluctuations associated with the dynamics of either individual atomic spins or the magnetic moments of small particles.[11–17] Thermal magnetic fluctuations are generic phenomena characterizing all nanoscale magnetic systems and devices.

The recently developed spin-*lattice* dynamics simulation approach,[18] incorporating both the atomic spin and lattice degrees of freedom, shows that mechanical properties of common materials such as steels, as well as dislocation and radiation defect structures formed in iron and steels under irradiation, are closely related to their temperature-dependent magnetic properties.[19–23] A striking example illustrating the interplay between magnetism and structural stability is the bcc-fcc α-γ structural phase transformation in iron-based alloys that, according to a recent study,[24] stems from the free energy change associated with thermal magnetic excitations, also investigated in Refs. 25 and 26.

Integrating Langevin spin dynamics equations proves more difficult than integrating Langevin equations for particles. Numerical integration algorithms described in the literature[11–17] are either subject to constraints limiting their application[11,12] or contain mathematical inconsistencies.[13,14] Some literature sources do not provide the necessary mathematical detail required for the implementation of an algorithm.[15–17] Furthermore, for all the algorithms it proves difficult to give a reliable assessment of the numerical error associated with the integration time step. Similarly, understanding of the connection between numerical realisations of stochastic spin dynamics and the statistical mechanics aspects of the method remains limited.[11] The difficulty partially stems from the fact that until recently there was no recipe for evaluating the temperature of a dynamic spin ensemble from the microscopic spin vector variables characterizing an instantaneous configuration of the spin system. A solution to the problem has now been found.[27]

In this paper, we derive a simple, robust, and numerically stable algorithm for integrating the stochastic Langevin spin dynamics equations. The algorithm is based on the Suzuki-Trotter decomposition (STD) method applied earlier to the integration of deterministic spin dynamics equations.[28–32] To investigate the microscopic aspects of spin relaxation and its dependence on the parameters characterizing the spin system, we derive a set of equations for the statistical moments of solutions of Langevin spin dynamics equations. Using the Furutsu-Novikov theorem,[33–36] we convert the stochastic Langevin equation for an individual spin into an infinite set of coupled deterministic first-order differential equations for the statistical moments of the evolving probability distributions. We prove equivalence between the stochastic and the moments approaches by exploring spin thermalization and the dynamics of relaxation of spin-spin self-correlation functions. We also give analytical estimates for the time scales of relaxation processes and show that they agree well with direct numerical solutions of Langevin spin dynamics equations.

II. INTEGRATION ALGORITHM

For a system of interacting spins described by a Hamiltonian $\mathcal{H}(\mathbf{S}_1,\mathbf{S}_2,\ldots,\mathbf{S}_N)$, the Langevin equation of motion for an individual spin $\mathbf{S}_i$ has the form[12,27,37]

$$\frac{d\mathbf{S}_i}{dt} = \frac{1}{\hbar}[\mathbf{S}_i \times (\mathbf{H}_i + \mathbf{h}_i) - \gamma \mathbf{S}_i \times (\mathbf{S}_i \times \mathbf{H}_i)], \qquad (1)$$

where $\mathbf{S}_i = \mathbf{S}_i(t)$ is a spin vector, $\mathbf{H}_i(t) = -\partial\mathcal{H}/\partial\mathbf{S}_i$ is the effective field acting on spin $\mathbf{S}_i$, and γ is a dimensionless damping parameter. $\mathbf{h}_i = \mathbf{h}_i(t)$ is a delta-correlated fluctuating

effective magnetic field, satisfying the conditions $\langle \mathbf{h}_i(t)\rangle = 0$ and $\langle h_{i\alpha}(t)h_{j\beta}(t')\rangle = \mu\delta_{ij}\delta_{\alpha\beta}\delta(t-t')$. Subscripts α and β denote the Cartesian components of a vector. Parameters γ and μ are related via the FDT.[1,2,10] In thermal equilibrium, by identifying the energy distribution for the spin system with the Gibbs distribution, we find the following fluctuation-dissipation relation (FDR)[10,27] between parameter μ characterizing the magnitude of random fluctuations of $\mathbf{h}_i(t)$ and the dissipation parameter γ, namely $\mu = 2\gamma\hbar k_B T$.

Equation (1) is a part of a set of N coupled first-order differential equations describing the evolution of an ensemble of interacting spins.[18,27] In this paper we focus on the development of an integration algorithm for spin and spin-lattice dynamics, and in what follows consider a case where Eq. (1) describes the dynamics of an individual spin $\mathbf{S}_i$, interacting with external magnetic field $\mathbf{H}_i$. Omitting index i, we assume that the magnitude and the direction of $\mathbf{H}$ do not depend on time t. In this limit Eq. (1) acquires the form

$$\frac{d\mathbf{S}}{dt} = \frac{1}{\hbar}[\mathbf{S}\times(\mathbf{H}+\mathbf{h}) - \gamma\mathbf{S}\times(\mathbf{S}\times\mathbf{H})]. \tag{2}$$

While this equation provides the simplest possible model for Langevin spin dynamics, its solution still cannot be found in an explicit analytical form similar to that available for the case of Langevin dynamics of particles.[3,38] Fortunately, it appears possible to develop an efficient and accurate numerical approach to solving Eq. (2), which is based on the Suzuki-Trotter decomposition (STD).[28] Integration algorithms involving the STD are known to accumulate small numerical error over long intervals of time, due to their symplectic nature.[4,5,28]

Consider a first-order differential equation of the form

$$\frac{d\mathbf{x}}{dt} = (\hat{A}+\hat{B})\mathbf{x}, \tag{3}$$

where $\mathbf{x}$ is a vector variable, and $\hat{A}$ and $\hat{B}$ are operators acting on $\mathbf{x}$. The formal solution of the above equation is

$$\mathbf{x}(\tau) = \exp[(\hat{A}+\hat{B})\tau]\mathbf{x}(0). \tag{4}$$

Using the STD, we transform the evolution operator $\exp[(\hat{A}+\hat{B})\tau]$ as follows:[28]

$$\exp[(\hat{A}+\hat{B})\tau] \approx \exp(\hat{A}\tau/2)\exp(\hat{B}\tau)\exp(\hat{A}\tau/2). \tag{5}$$

The numerical error associated with this transformation is of the order of $O(\tau^3)$. The computational benefit of this decomposition is that it reduces the problem of finding the full evolution operator $\exp[(\hat{A}+\hat{B})\tau]$ to the problem of finding two simpler evolution operators $\exp(\hat{A}\tau)$ and $\exp(\hat{B}\tau)$, both of which in the case of Langevin spin dynamics can be found analytically, leaving the only source of numerical error to be the STD itself.[4,5,28]

In our case, we separate Eq. (2) into the deterministic and stochastic parts by rewriting it in the following form:

$$\frac{d\mathbf{S}}{dt} = (\hat{L}_d + \hat{L}_s)\mathbf{S}, \tag{6}$$

where

$$\hat{L}_d\mathbf{S} = \frac{1}{\hbar}[\mathbf{S}\times\mathbf{H} - \gamma\mathbf{S}\times(\mathbf{S}\times\mathbf{H})], \tag{7}$$

$$\hat{L}_s\mathbf{S} = \frac{1}{\hbar}[\mathbf{S}\times\mathbf{h}]. \tag{8}$$

The deterministic part $\hat{L}_d$ contains the spin rotation and the dissipation terms, whereas the stochastic part $\hat{L}_s$ contains only the fluctuation field term.

First, we solve the deterministic equation (7). Without loss of generality, we choose the orientation of the Cartesian system of coordinates in such a way that the external field $\mathbf{H}$ points in the z direction. Transforming the equation

$$\frac{d\mathbf{S}}{dt} = \frac{1}{\hbar}[\mathbf{S}\times\mathbf{H} - \gamma\mathbf{S}\times(\mathbf{S}\times\mathbf{H})] \tag{9}$$

into spherical coordinates,[11] we arrive at

$$d\theta/dt = -HS\gamma\sin\theta/\hbar, \tag{10}$$

$$d\phi/dt = -H/\hbar, \tag{11}$$

where $\mathbf{S} = (S_x, S_y, S_z) = S(\sin\theta\cos\phi, \sin\theta\sin\phi, \cos\theta)$, $S = |\mathbf{S}|$, and $H = |\mathbf{H}|$.

Equations (10) and (11) for the polar and azimuthal angles are not coupled to each other, and their solutions have the form

$$\tan(\theta(\tau)/2) = \tan(\theta(0)/2)\exp(-HS\gamma\tau/\hbar), \tag{12}$$

$$\phi(\tau) = \phi(0) - H\tau/\hbar. \tag{13}$$

Transforming these solutions back into the Cartesian representation, and rotating the system of coordinates, we find a covariant expression for the deterministic spin evolution operator valid for an arbitrary orientation of the external magnetic field $\mathbf{H}$, namely

$$\mathbf{S}(\tau) = \exp(\hat{L}_d\tau)\mathbf{S}(0) = \frac{2\mathbf{S}(0)He^{\zeta}\cos\xi + 2[\mathbf{S}(0)\times\mathbf{H}]e^{\zeta}\sin\xi + \mathbf{H}S[1-e^{2\zeta}+\chi(1+e^{2\zeta}-2e^{\zeta}\cos\xi)]}{H[1+e^{2\zeta}+\chi(1-e^{2\zeta})]}, \tag{14}$$

where $\xi = H\tau/\hbar$, $\zeta = -HS\gamma\tau/\hbar$, and $\chi = (\mathbf{S}(0)\cdot\mathbf{H})/SH$. We note that equation (9) conserves the magnitude of the spin vector, and hence $S = |\mathbf{S}(t)| = |\mathbf{S}(0)|$.

For the stochastic part (8), provided that the magnitude of $\mathbf{h}$ remains constant over an infinitesimal interval of time τ, we use an analytical solution of equation

$$\frac{d\mathbf{S}}{dt} = \frac{1}{\hbar}[\mathbf{S}\times\mathbf{h}]. \tag{15}$$

This solution has the form[29–32]

$$\mathbf{S}(\tau) = \exp(\hat{L}_s\tau)\mathbf{S}(0) = \mathbf{D}(\tau)\mathbf{S}(0), \tag{16}$$

where $\mathbf{D}(\tau) = \mathbf{I} + \boldsymbol{\Omega}\sin(\omega\tau) + \boldsymbol{\Omega}^2[1-\cos(\omega\tau)]$. Here $\boldsymbol{I}$ is the identity matrix, and $\boldsymbol{\Omega}$ is an antisymmetric matrix with components $\Omega_{xy} = -\omega_z/\omega$, $\Omega_{zx} = -\omega_y/\omega$, and $\Omega_{yz} = -\omega_x/\omega$, where $\omega = |\boldsymbol{\omega}|$ and $\boldsymbol{\omega} = -\mathbf{h}/\hbar$.

In the practical implementation of the method, at each integration step we evaluate the stochastic magnetic field using the expression $h_\alpha = \eta\sqrt{\mu/\tau}$, where τ is the time step and η is a random variable sampled from the standard normal distribution.

We now have explicit analytical expressions for both the deterministic and the stochastic evolution operators (7) and (8), which we evaluate *exactly* using Eqs. (14) and (16). Substituting these expressions into (5), we arrive at a formula for the integration time step for our numerical Langevin spin dynamics algorithm. The only source of error associated with our formula is the error of the STD decomposition itself, which is a quantity of the order of $O(\tau^3)$.

Comparing the above recipe with the available integration algorithms[11–14] we note the advantages offered by our method. First, our formula does not require consideration of the effects of multiplicative noise, which are automatically included in the algorithm via the Suzuki-Trotter decomposition of the evolution operators. The difficulty associated with the existing treatments of multiplicative noise is associated with the nonlinear nature of Eq. (2).[10,11] In Ref. 11 the authors adopted a fairly elaborate treatment of the multiplicative noise, which involved the derivation of a Fokker-Planck equation. Indeed, if one attempts to treat the evolution of a spin vector using a single operation at each time step, then one needs to include the noise-induced drift (the Stratonovich calculus) as a correction. If such a correction is not introduced, the spin trajectory converges to the Ito solution[11] and becomes inconsistent with the FDR (see e.g., Refs. 13 and 14) derived within the framework of the Stratonovich calculus.

Second, our algorithm guarantees the preservation of the magnitude of the spin vector, since both the deterministic [Eq. (14)] and stochastic [Eq. (16)] evolution operators conserve the magnitude of the spin vector exactly. There is no truncation error in Eqs. (14) and (16) associated with the higher order terms $\sim\tau^2$, $\tau^3, \ldots,$ as opposed to the often used linear addition[11] or Runge-Kutta methods.[12] Furthermore, our method imposes no constraint on the magnitude of the fluctuation term, which appears difficult to treat within the Runge-Kutta integration algorithm,[12] where numerical accuracy is guaranteed only in the weak noise limit.

III. EQUATIONS FOR STATISTICAL MOMENTS

A question that one can pose in relation to Eq. (2) is whether it is possible to evaluate the statistical ensemble average of the spin vector $\langle\mathbf{S}\rangle$ as a function of time. It is not immediately obvious how to answer the question since the Langevin spin dynamics equation, as opposed to the conventional Langevin equation,[3] is nonlinear. Earlier[27] we noted that the nonlinear nature of Eq. (2) allows the noise occurring at a preceding moment of time to affect the solution at the current moment of time through the nonlinear coupling involving all three components of the spin vector.

Still, it appears possible to address the issue. To do this, we first need to develop an auxiliary formalism. Our treatment involves applying the Furutsu-Novikov theorem (FNT).[33–36] The FNT states that the ensemble average of a product of a functional $f(t)$, which depends on Gaussian noise $\epsilon(t)$, and the noise itself, equals

$$\langle\epsilon(t)f(t)\rangle = \int dt'\langle\epsilon(t)\epsilon(t')\rangle\left\langle\frac{\delta f(t)}{\delta\epsilon(t')}\right\rangle, \tag{17}$$

where $\delta f(t)/\delta\epsilon(t')$ denotes the functional derivative. In the case of Langevin spin dynamics, the Furutsu-Novikov formula has the form

$$\langle h_\alpha(t)f(t)\rangle = \mu\left\langle\frac{\delta f(t)}{\delta h_\alpha(t)}\right\rangle. \tag{18}$$

This expression has a broad range of validity and is only subject to the constraint that the random field $\mathbf{h}(t)$ is Gaussian and delta-correlated.

Since the spin vector $\mathbf{S}$ at time t can be found by formally integrating the Langevin spin equation, i.e.,

$$\mathbf{S}(t) = \mathbf{S}(0) + \frac{1}{\hbar}\int_0^t d\tau[\mathbf{S}\times(\mathbf{H}+\mathbf{h}) - \gamma\mathbf{S}\times(\mathbf{S}\times\mathbf{H})], \tag{19}$$

we can evaluate the functional derivative as

$$\frac{\delta S_i(t)}{\delta h_j(t)} = \frac{1}{2\hbar}\epsilon_{ikj}S_k, \tag{20}$$

where ϵ_{ikj} is the Levi-Civita completely antisymmetric tensor.

Now, we generalize the treatment to the case of statistical moments of the spin vector $\langle(\mathbf{S}\cdot\mathbf{H})^N\mathbf{S}\rangle$ treated as a function of time, where N is a nonnegative integer. After some algebra, we find that

$$\begin{aligned}
&\frac{d}{dt}\langle(\mathbf{S}\cdot\mathbf{H})^N\mathbf{S}\rangle \\
&\quad= \left\langle(\mathbf{S}\cdot\mathbf{H})^N\frac{d\mathbf{S}}{dt}\right\rangle + \left\langle N(\mathbf{S}\cdot\mathbf{H})^{N-1}\left(\frac{d\mathbf{S}}{dt}\cdot\mathbf{H}\right)\mathbf{S}\right\rangle \\
&\quad= \frac{1}{\hbar}\{\langle(\mathbf{S}\cdot\mathbf{H})^N\mathbf{S}\rangle\times\mathbf{H} + \langle(\mathbf{S}\cdot\mathbf{H})^N(\mathbf{S}\times\mathbf{h})\rangle \\
&\qquad- \gamma(N+1)\langle(\mathbf{S}\cdot\mathbf{H})^{N+1}\mathbf{S}\rangle + \gamma S^2\langle(\mathbf{S}\cdot\mathbf{H})^N\rangle\mathbf{H} \\
&\qquad- N\langle(\mathbf{S}\cdot\mathbf{H})^{N-1}[(\mathbf{S}\times\mathbf{H})\cdot\mathbf{h}]\mathbf{S}\rangle \\
&\qquad+ N\gamma S^2H^2\langle(\mathbf{S}\cdot\mathbf{H})^{N-1}\mathbf{S}\rangle\}.
\end{aligned} \tag{21}$$

In Eq. (21), there are two terms that explicitly contain the fluctuating field $\mathbf{h}$. Using Eqs. (18) and (20), we find

$$\begin{aligned}
&\langle(\mathbf{S}\cdot\mathbf{H})^N(\mathbf{S}\times\mathbf{h})\rangle \\
&\quad= \frac{\mu}{\hbar}\left[\frac{N}{2}S^2\langle(\mathbf{S}\cdot\mathbf{H})^{N-1}\rangle\mathbf{H} - \left(\frac{N}{2}+1\right)\langle(\mathbf{S}\cdot\mathbf{H})^N\mathbf{S}\rangle\right],
\end{aligned} \tag{22}$$

$$\begin{aligned}
&\langle(\mathbf{S}\cdot\mathbf{H})^N[(\mathbf{S}\times\mathbf{H})\cdot\mathbf{h}]\mathbf{S}\rangle \\
&\quad= \frac{\mu}{2\hbar}[(N+3)\langle(\mathbf{S}\cdot\mathbf{H})^{N+1}\mathbf{S}\rangle \\
&\qquad- NS^2H^2\langle(\mathbf{S}\cdot\mathbf{H})^{N-1}\mathbf{S}\rangle - S^2\langle(\mathbf{S}\cdot\mathbf{H})^N\rangle\mathbf{H}].
\end{aligned} \tag{23}$$

Substituting Eqs. (22) and (23) back into Eq. (21), we arrive at

$$\begin{aligned}
&\frac{d}{dt}\langle(\mathbf{S}\cdot\mathbf{H})^N\mathbf{S}\rangle \\
&\quad= \frac{1}{\hbar}\left[\langle(\mathbf{S}\cdot\mathbf{H})^N\mathbf{S}\rangle\times\mathbf{H} + \frac{\mu}{2\hbar}N(N-1)S^2H^2\langle(\mathbf{S}\cdot\mathbf{H})^{N-2}\mathbf{S}\rangle\right. \\
&\qquad\left.+ N\gamma S^2H^2\langle(\mathbf{S}\cdot\mathbf{H})^{N-1}\mathbf{S}\rangle - \frac{\mu}{2\hbar}(N+2)(N+1)\right.
\end{aligned}$$

$$\times \langle (\mathbf{S}\cdot\mathbf{H})^N \mathbf{S}\rangle - \gamma(N+1)\langle (\mathbf{S}\cdot\mathbf{H})^{N+1}\mathbf{S}\rangle + \frac{\mu N}{\hbar} S^2 \langle (\mathbf{S}\cdot\mathbf{H})^{N-1}\rangle \mathbf{H} + \gamma S^2 \langle (\mathbf{S}\cdot\mathbf{H})^N\rangle \mathbf{H}\Big]. \quad (24)$$

Note that this equation contains no stochastic terms. It is also consistent with the treatment developed by García-Palacios and Lázaro,[11] who investigated the case $N = 0$ using the Fokker-Planck equation and derived an equation, similar to (24), for the case corresponding to $N = 0$. One should note that Eq. (24) is in fact an infinite set of differential equations. The covariant vector form of this equation makes it possible to project it onto an arbitrary constant vector. For example, we can form a scalar product of (24) with the external field $\mathbf{H}$, and derive a somewhat more compact set of differential equations for the moments $\langle (\mathbf{S}\cdot\mathbf{H})^N\rangle$, namely

$$\frac{d\langle (\mathbf{S}\cdot\mathbf{H})^N\rangle}{dt} = \frac{N}{\hbar}\Big[S^2H^2\Big(\frac{\mu(N-1)}{2\hbar}\langle (\mathbf{S}\cdot\mathbf{H})^{N-2}\rangle + \gamma\langle (\mathbf{S}\cdot\mathbf{H})^{N-1}\rangle\Big) - \frac{\mu(N+1)}{2\hbar}\langle (\mathbf{S}\cdot\mathbf{H})^N\rangle - \gamma\langle (\mathbf{S}\cdot\mathbf{H})^{N+1}\rangle\Big]. \quad (25)$$

This set of coupled equations makes it possible to follow the evolution of the Nth moment of $(\mathbf{S}\cdot\mathbf{H})$, which is the negative of the energy of interaction between the spin and the external magnetic field.

Using Eq. (25) we find that the energy distribution for an ensemble of spins evolving according to Langevin spin dynamics converges to the equilibrium Gibbs distribution. Indeed, if the energy distribution is given by the Gibbs formula, the expectation value for the statistical moment $\langle (\mathbf{S}\cdot\mathbf{H})^N\rangle_{\rm eq}$ at equilibrium can be evaluated analytically as

$$\langle (\mathbf{S}\cdot\mathbf{H})^N\rangle_{\rm eq} = \frac{(SH)^{N+1}\Lambda - \langle (\mathbf{S}\cdot\mathbf{H})^{N+1}\rangle_{\rm eq}}{(N+1)k_BT}, \quad (26)$$

where

$$\Lambda = \begin{cases} 1 & \text{where } N \text{ is odd,} \\ \coth(SH/k_BT) & \text{where } N \text{ is even.} \end{cases} \quad (27)$$

Substituting Eq. (26) into Eq. (25) and using the FDR $\mu = 2\gamma\hbar k_BT$ we find that the right-hand side of Eq. (25) vanishes. This applies to all the nonnegative integer values of N, proving that the asymptotic equilibrium state of a spin ensemble that evolves according to the Langevin spin dynamics equation is described by the Gibbs distribution. This finding is consistent with analysis based on the Fokker-Planck equation approach.[27]

Concluding this section, we note that the system of equations (24) offers a straightforward and relatively simple means of investigating the dynamics of spin relaxation, which is alternative to the more elaborate approaches involving either the integration of stochastic equations of Langevin spin dynamics, or the derivation and analysis of the Fokker-Planck equations.

IV. NUMERICAL EXAMPLES

In this section, we investigate two examples illustrating the equivalence between the Langevin spin dynamics treatment based on Eq. (2), and the set of coupled differential equations for the statistical moments (24) and (25). For convenience, in what follows we refer to the differential equations (24) and (25) as the moments equations.

First, we investigate the dynamics of the thermalization process for an ensemble of noninteracting spins. This requires studying the variation of $\langle \mathbf{S}\cdot\mathbf{H}\rangle$ treated as a function of time. We also investigate the evolution of higher order moments $\langle (\mathbf{S}\cdot\mathbf{H})^N\rangle$. Second, we analyze the evolution of the spin-spin self-correlation function $\langle \mathbf{S}(t)\cdot\mathbf{S}(0)\rangle$, which is a quantity related to the structure factor for neutron scattering.[39,40]

Using the integration algorithm developed in Sec. II above, we have followed the evolution of a spin ensemble containing 5000 spins. The ensemble average of any quantity was estimated by averaging over all the 5000 spins during the simulation. The spin evolution followed Eq. (2). The fluctuating fields $\mathbf{h}$ for each spin were generated independently using different random seeds. We use the following values of parameters $S = 2.2/g$, $H = 0.35$ eV, and $\gamma = 0.1$, where g is the electron g factor. These parameters approximately describe bcc iron in the low temperature limit. The external field $\mathbf{H}$ is assumed to be pointing in the z direction.

For the purpose of our analysis, the choice of parameters is largely immaterial, although the fact that we can choose them freely illustrates the robustness and numerical stability of the Langevin spin dynamics integration algorithm. Similarly, the values of temperature T and time t are significant only in relation to the above choice of $\mathbf{H}$ and γ.

In a numerical simulation, we cannot integrate an infinite number of equations, and hence the moments equations (24) and (25) need to be terminated at a certain N. We note that if we consider the equations up to the Nth one, we lose information about the $(N+1)$th moment. On the other hand, we know the asymptotic equilibrium values of all the moments derived analytically from the Gibbs distribution. Using the equilibrium limit values of moments for terminating the time-dependent moments equations has the advantage of guaranteeing the correct asymptotic behavior of these quantities as $t \to \infty$. To assess the accuracy of this approximation we proceed as follows.

In the first case study, we initiate all the spin vectors assuming that they point in the same direction as the external field. Figure 1 shows the variation of the normalized first moment $\langle \mathbf{S}\cdot\mathbf{H}\rangle$ as a function of time, found by integrating the Langevin equation (2) and the moments equations (25). For $T = 300$ K, we find that about 30 moments equations are required to arrive at a fully converged result, whereas at $T = 3000$ K it is sufficient to retain just 5 equations for the moments. The choice of the terminator does not have any visible effect on the simulated dynamics of the relaxation process if the number of equations is large enough. Since the computational effort required for integrating fewer than a hundred coupled differential equations is negligible, this ensures the practical viability of the moments equations approach. It is interesting that while evaluating the terminator accurately is more difficult in the limit of high T, far fewer equations need to be retained in (24) and (25) in this limit, solving the problem of numerical stability of the method over the entire temperature interval.

Figure 2 shows the variation of the normalized 1st, 2nd, 3rd, and 4th moments of $(\mathbf{S}\cdot\mathbf{H})$ at various

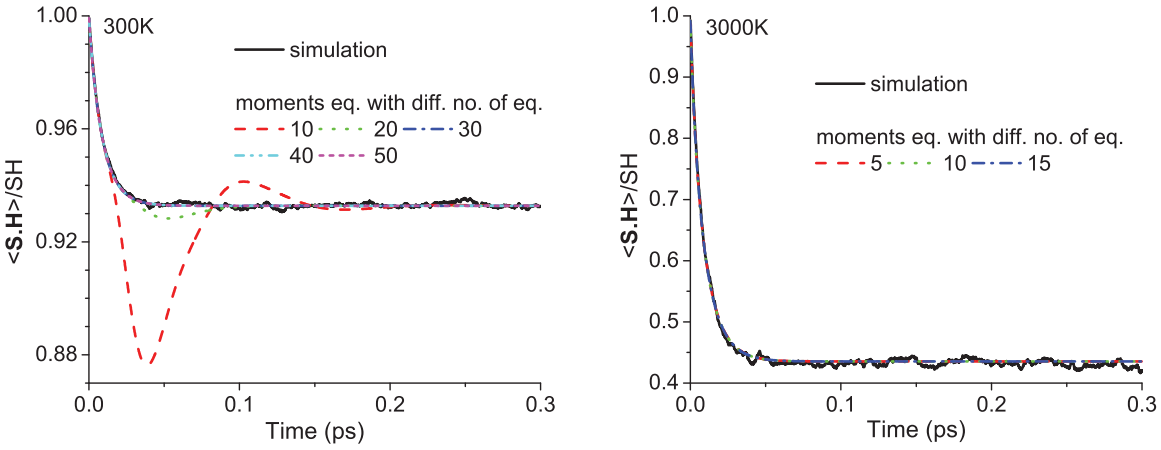

FIG. 1. (Color online) The normalized first moment $\langle \mathbf{S} \cdot \mathbf{H} \rangle$ evaluated as a function of time by integrating Eq. (2) and by using the moments equations (25). At low temperature $T = 300$ K, as many as 30 equations are required for convergence, while at a higher temperature $T = 3000$ K just 5 equations are sufficient. The solutions to the Langevin equation and the moments equations follow the same trend, confirming the equivalence of the two methods in the treatment of spin relaxation.

temperatures, ranging from relatively low to extremely high. Only the convergent curves evaluated using the moments equations are shown for comparison. We see that not only for the 1st moment, but also for the higher order moments, the deterministic equations (24) and (25) are fully consistent with the stochastic Langevin spin dynamics simulations. All the relaxation modes characterizing a spin ensemble evolving under the action of a stochastic

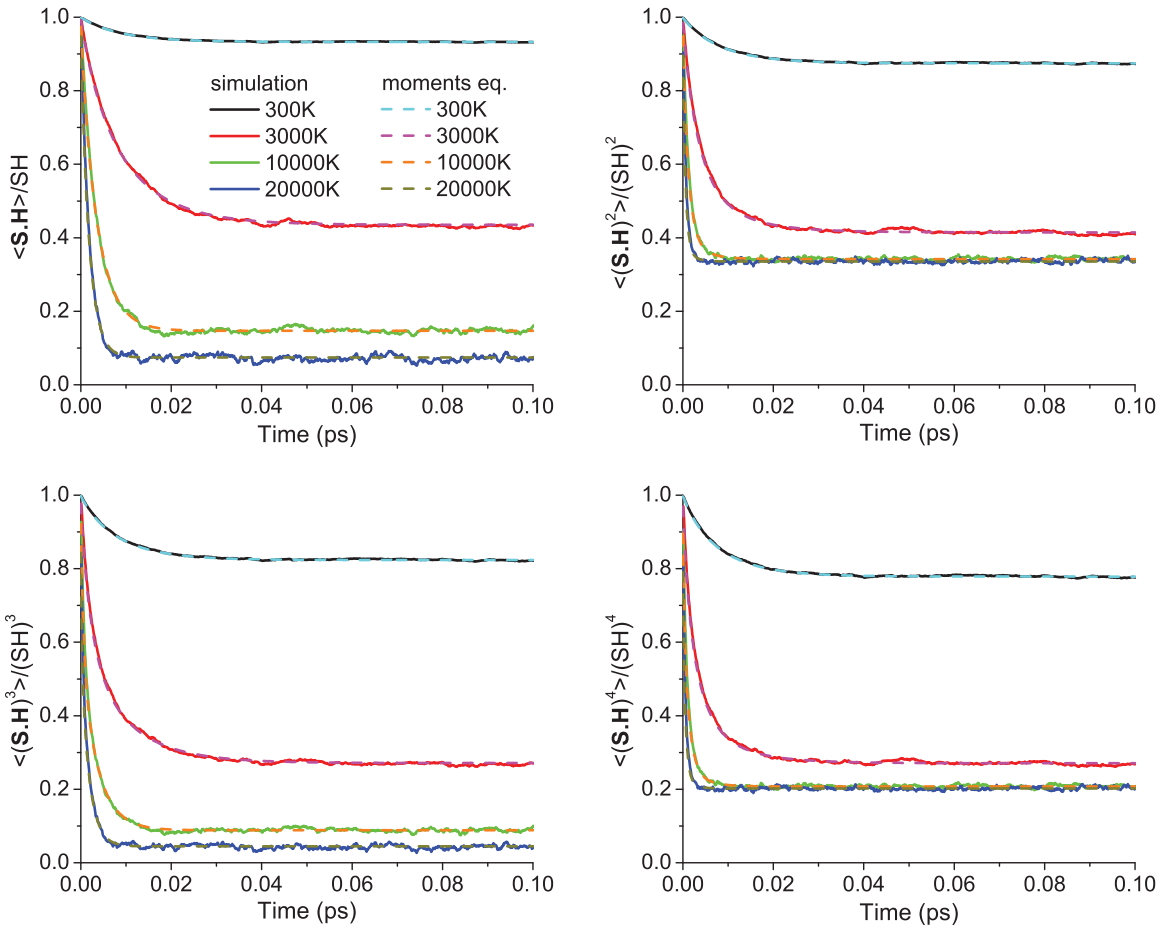

FIG. 2. (Color online) The normalized 1st, 2nd, 3rd, and 4th moments of $(\mathbf{S} \cdot \mathbf{H})$ plotted as functions of time for $T = 300$ K, 3000 K, 10000 K, and 20000 K. The solutions of the moments equations are fully consistent with stochastic simulations based on the Langevin spin dynamics equations.

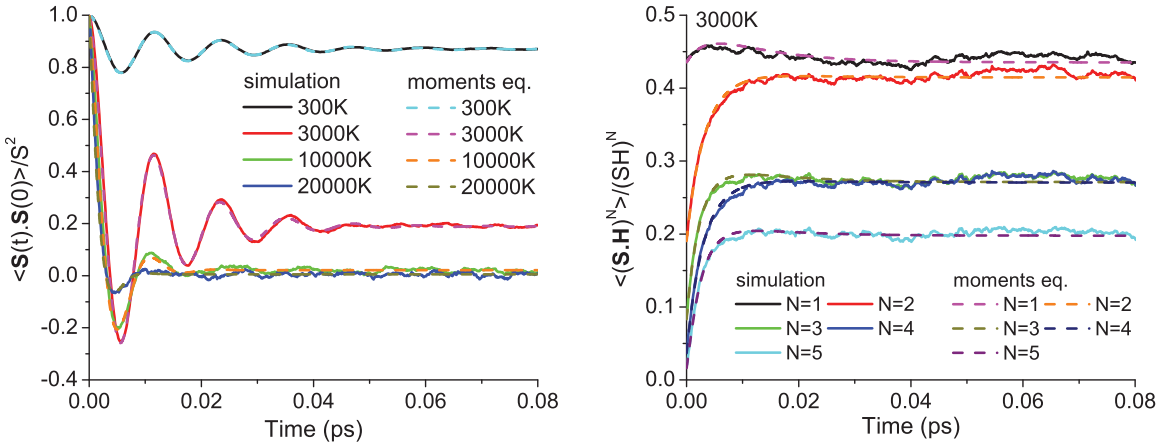

FIG. 3. (Color online) The normalized spin-spin self-correlation function plotted as a function of time for $T = 300$ K, 3000 K, 10000 K, and 20000 K. All the spins were initiated assuming $\mathbf{S} = (S_x, 0, S_z)$, where $S_z = \langle \mathbf{S} \cdot \mathbf{H} \rangle_{\text{eq}} / H$. The simulation data are in very good agreement with the moments equations. The normalized moments $\langle (\mathbf{S} \cdot \mathbf{H})^N \rangle$ for $N = 1, \ldots, 5$ at $T = 3000$ K are also shown for reference.

magnetic field can be reproduced by solving the moments equations.

In the second case study illustrated in Fig. 3 we analyze the variation of the normalized spin-spin self-correlation function $\langle \mathbf{S}(t) \cdot \mathbf{S}(0) \rangle$ treated as a function of time t. At $t = 0$ all the spins were assigned the same value $\mathbf{S} = (S_x, 0, S_z)$, where $S_z = \langle \mathbf{S} \cdot \mathbf{H} \rangle_{\text{eq}} / H$. Since the energy of a spin in an external magnetic field is degenerate with respect to its rotation in the plane normal to the field, the choice of S_x and S_y is unimportant, although it needs to be consistent with the principle of conservation of the magnitude of the spin vector. As we noted before, Eq. (24) can be projected onto an arbitrary vector. The spin-spin self-correlation function $\langle \mathbf{S}(t) \cdot \mathbf{S}(0) \rangle$ is in fact a projection of $\langle \mathbf{S}(t) \rangle$ onto $\mathbf{S}(0)$. By examining Fig. 3 we conclude that the moments equations reproduce almost exactly the behavior found in the Langevin spin dynamics simulations. The oscillations reflect the rotational nature of spin evolution. Asymptotically, the spin-spin self-correlation function relaxes to an equilibrium value. The variation of the normalized moments $\langle (\mathbf{S} \cdot \mathbf{H})^N \rangle$ for $N = 1, \ldots, 5$ at $T = 3000$ K is also shown for reference.

V. ANALYTICAL ESTIMATES FOR THE RELAXATION TIME SCALES

In order to investigate the dynamics of relaxation of a spin ensemble evolving under the action of a stochastic field, one can either directly integrate the Langevin spin dynamics equation (2) or use the moments equations (24). In this section we develop an approximate analytical treatment that makes it possible to estimate the relaxation time scales.

If the spin ensemble is close to thermal equilibrium, we may investigate the dynamics of thermalization by studying the relaxation of a small perturbation of the spin configuration from equilibrium. Since the reference equilibrium configuration of the ensemble depends on temperature, we expect that the relaxation time scales will also be temperature dependent. Using Eq. (25) for $N = 1$, we find

$$\frac{d\langle \mathbf{S} \cdot \mathbf{H} \rangle}{dt} = \frac{\gamma}{\hbar} [S^2 H^2 - 2 k_B T \langle \mathbf{S} \cdot \mathbf{H} \rangle + \langle (\mathbf{S} \cdot \mathbf{H})^2 \rangle]. \quad (28)$$

A small perturbation $\delta\mathbf{S}$ introduced into Eq. (28) evolves according to

$$\frac{d(\delta \mathbf{S} \cdot \mathbf{H})}{dt} = -\frac{2\gamma}{\hbar} [k_B T + \langle \mathbf{S} \cdot \mathbf{H} \rangle] (\delta \mathbf{S} \cdot \mathbf{H}). \quad (29)$$

The prefactor on the right-hand side of this equation equals the inverse of the relaxation time scale,

$$\tau_{\text{th}}^{-1} = \frac{2\gamma}{\hbar} [k_B T + \langle \mathbf{S} \cdot \mathbf{H} \rangle]. \quad (30)$$

The equilibrium value of $\langle \mathbf{S} \cdot \mathbf{H} \rangle$ is given by Eq. (26) for $N = 0$,

$$\langle \mathbf{S} \cdot \mathbf{H} \rangle = SH \coth \left(\frac{SH}{k_B T} \right) - k_B T. \quad (31)$$

Using this equation, we find that the inverse of the thermalization time scale approximately equals

$$\tau_{\text{th}}^{-1} = \frac{2\gamma}{\hbar} SH \coth \left(\frac{SH}{k_B T} \right). \quad (32)$$

At low temperatures (relative to the strength of $\mathbf{H}$) we have $\coth(SH/k_B T) \approx 1$, whereas in the high temperature limit $\coth(SH/k_B T) \approx k_B T / SH$. The interval of variation of the inverse of the relaxation time scale is constrained by the two limits

$$\tau_{\text{th}}^{-1} \approx \frac{2\gamma}{\hbar} \times \begin{cases} SH, & \text{if} \quad k_B T \ll SH. \\ k_B T, & \text{if} \quad k_B T \gg SH. \end{cases} \quad (33)$$

This equation shows that at low temperature the dominant factor influencing the thermalization time scale is the strength of external magnetic field $\mathbf{H}$, whereas at high temperature the main factor determining the relaxation time scale is temperature.

In Fig. 4, we plot the normalized first moment $\langle \mathbf{S} \cdot \mathbf{H} \rangle$ as a function of time for the cases where the spin ensemble was thermalized from 0 K to 300 K, and from 0 K to 20000 K. The red curves in the same figure were calculated using the equations

$$\langle \mathbf{S}(t) \rangle = [\langle \mathbf{S}(0) \rangle - \langle \mathbf{S} \rangle_{\text{eq}}] e^{-t/\tau_{\text{th}}} + \langle \mathbf{S} \rangle_{\text{eq}}, \quad (34)$$

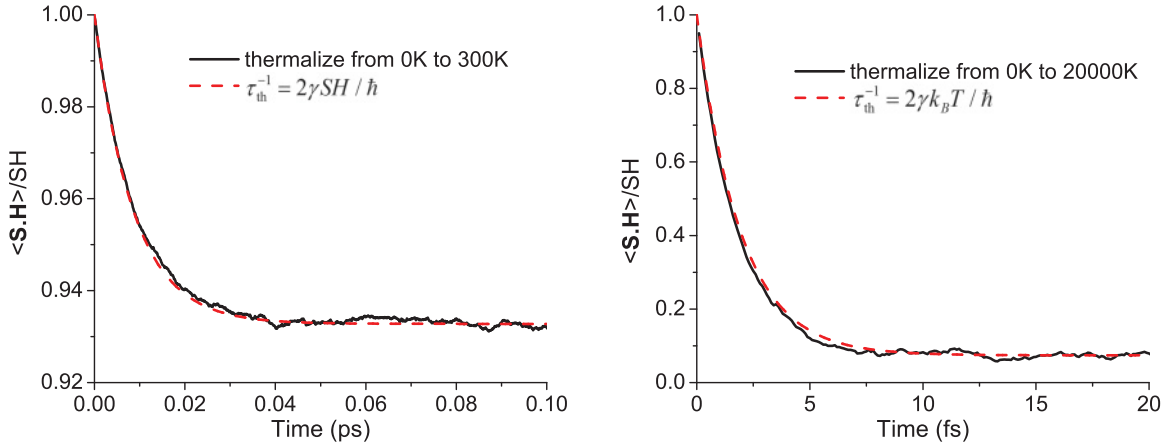

FIG. 4. (Color online) The black curves show the normalized values of $\langle \mathbf{S}\cdot\mathbf{H}\rangle$ plotted as functions of time for spin ensembles thermalized by stochastic fields from 0 K to 300 K, and from 0 K to 20000 K. The red curves were calculated using the equations $\langle \mathbf{S}(t)\rangle = [\langle \mathbf{S}(0)\rangle - \langle \mathbf{S}\rangle_{\rm eq}]e^{-t/\tau_{\rm th}} + \langle \mathbf{S}\rangle_{\rm eq}$, where $\tau_{\rm th} = 2\gamma SH/\hbar$ and $\tau_{\rm th} = 2\gamma k_B T/\hbar$ for 300 K and 20000 K, respectively.

where $\tau_{\rm th}^{-1} = 2\gamma SH/\hbar$ and $\tau_{\rm th}^{-1} = 2\gamma k_B T/\hbar$ for 300 K and 20000 K, respectively.

It is surprising that the black and the red curves match each other so well, given that the conditions of validity of the perturbation treatment based on Eq. (29) are obviously not satisfied. The origin of the unexpectedly high accuracy of the perturbation treatment may be understood if we reexamine Eq. (30).

Defining the reservoir temperature T_R and the system temperature T_S, we rewrite Eq. (32) as follows:

$$\tau_{\rm th}^{-1} = \frac{2\gamma}{\hbar}\left[k_B T_R + SH\coth\left(\frac{SH}{k_B T_S}\right) - k_B T_S\right]. \quad (35)$$

If the spin ensemble is thermalized starting from 0 K, where both the reservoir and the system temperatures remain low $SH \gg k_B T_S$, $SH \gg k_B T_R$, the relaxation of the system follows the low temperature limit of Eq. (33). On the other hand, if the reservoir is at very high temperature, the term $k_B T_R$ initially dominates the dynamics of relaxation. During the thermalization process, T_S increases, and the terms $SH\coth(SH/k_B T_S)$ and $k_B T_S$ cancel each other, with the result that $k_B T_R$ dominates the entire relaxation history. In this case, relaxation follows the high temperature limit of Eq. (33).

In the case of relaxation of spin-spin self-correlations, the situation is more complicated. First, we write Eq. (24) for $N = 0$ as

$$\frac{d\langle \mathbf{S}\rangle}{dt} = \frac{1}{\hbar}\left[\langle \mathbf{S}\rangle \times \mathbf{H} - \frac{\mu}{\hbar}\langle \mathbf{S}\rangle - \gamma\langle(\mathbf{S}\cdot\mathbf{H})\mathbf{S}\rangle + \gamma S^2\mathbf{H}\right]. \quad (36)$$

In cartesian component form this becomes

$$\frac{d\langle S_x\rangle}{dt} = \frac{1}{\hbar}\left[\langle S_y\rangle H - \frac{\mu}{\hbar}\langle S_x\rangle - \gamma\langle S_z S_x\rangle H\right], \quad (37)$$

$$\frac{d\langle S_y\rangle}{dt} = \frac{1}{\hbar}\left[-\langle S_x\rangle H - \frac{\mu}{\hbar}\langle S_y\rangle - \gamma\langle S_z S_y\rangle H\right], \quad (38)$$

$$\frac{d\langle S_z\rangle}{dt} = \frac{1}{\hbar}\left[-\frac{\mu}{\hbar}\langle S_z\rangle - \gamma\langle S_z^2\rangle H + \gamma S^2 H\right]. \quad (39)$$

If a spin ensemble is in equilibrium, the probability of finding a certain value of S_z is given by the Gibbs distribution. We introduce the approximation $S_z \approx \overline{S}_z$, where $\overline{S}_z = \langle \mathbf{S}\cdot\mathbf{H}\rangle_{\rm eq}/H$. Using this approximation, we find $\langle S_z S_x\rangle \approx \overline{S}_z\langle S_x\rangle$ and $\langle S_z S_y\rangle \approx \overline{S}_z\langle S_y\rangle$. Applying the FDR, from Eqs. (37) and (38) we find

$$\frac{d\langle S_x\rangle}{dt} = \frac{1}{\hbar}[H\langle S_y\rangle - \gamma(2k_B T + \overline{S}_z H)\langle S_x\rangle], \quad (40)$$

$$\frac{d\langle S_y\rangle}{dt} = \frac{1}{\hbar}[-H\langle S_x\rangle - \gamma(2k_B T + \overline{S}_z H)\langle S_y\rangle]. \quad (41)$$

Solutions of these coupled equations are

$$\langle S_x\rangle = S_{xy}\cos\left(-\frac{H}{\hbar}t + \phi\right)e^{-t/\tau_{\rm ss}}, \quad (42)$$

$$\langle S_y\rangle = S_{xy}\sin\left(-\frac{H}{\hbar}t + \phi\right)e^{-t/\tau_{\rm ss}}, \quad (43)$$

where ϕ is a phase defined by the initial conditions, and $S_{xy} = \sqrt{S^2 - \overline{S}_z^2}$. The inverse of the relaxation time is now given by

$$\begin{aligned}\tau_{\rm ss}^{-1} &= \frac{\gamma}{\hbar}(2k_B T + \overline{S}_z H)\\ &= \frac{\gamma}{\hbar}(2k_B T + \langle \mathbf{S}\cdot\mathbf{H}\rangle_{\rm eq})\\ &= \frac{\gamma}{\hbar}\left[k_B T + SH\coth\left(\frac{SH}{k_B T}\right)\right]. \end{aligned} \quad (44)$$

Therefore, the spin-spin self-correlation function evolves approximately according to

$$\langle \mathbf{S}(t)\cdot\mathbf{S}(0)\rangle = \left(S^2 - \overline{S}_z^2\right)\cos\left(-\frac{H}{\hbar}t\right)e^{-t/\tau_{\rm ss}} + \overline{S}_z^2. \quad (45)$$

Proceeding similarly to the thermalization time (33), we write the relaxation time for the spin-spin self-correlation function in the low and high temperature limits as

$$\tau_{\rm ss}^{-1} \approx \frac{\gamma}{\hbar}\times\begin{cases} k_B T + SH & \text{if} \quad k_B T \ll SH,\\ 2k_B T & \text{if} \quad k_B T \gg SH. \end{cases} \quad (46)$$

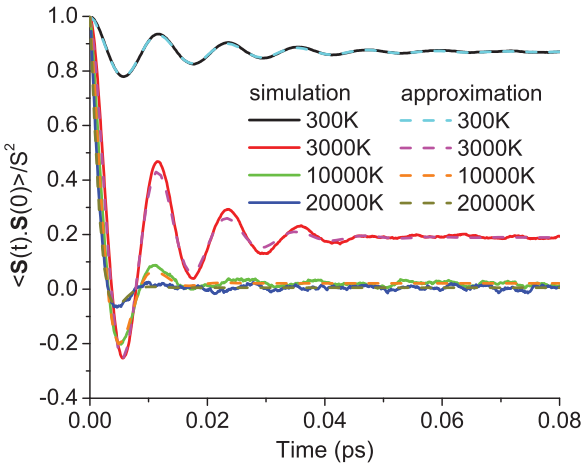

FIG. 5. (Color online) The normalized spin-spin self-correlation function plotted as a function of time. The simulation data points are the same as in Fig. 3. The approximate treatment [Eqs. (44) and (45)] agrees well with the simulation data.

In Fig. 5, we plot the relaxation curves calculated using Eqs. (44) and (45) together with the data given in Fig. 3. We see that the curves agree fairly well, although the agreement is not as good as in the case shown in Fig. 3, where numerical simulations are compared with the solutions of the moments equations. From Eq. (45), we see that the oscillatory behavior in Fig. 5 is associated with the cosine term, whereas the exponential function describes the irreversible part of the relaxation process.

In reality, the spin vectors do not initially point all in the same direction. In thermal equilibrium, the directions of the spin vectors are fairly random, subject to the Gibbs distribution, meaning that $\mathbf{S}(0)$ does not have the same value for all the spins. Figure 6 shows data for the spin-spin self-correlation function evaluated starting from a thermalized

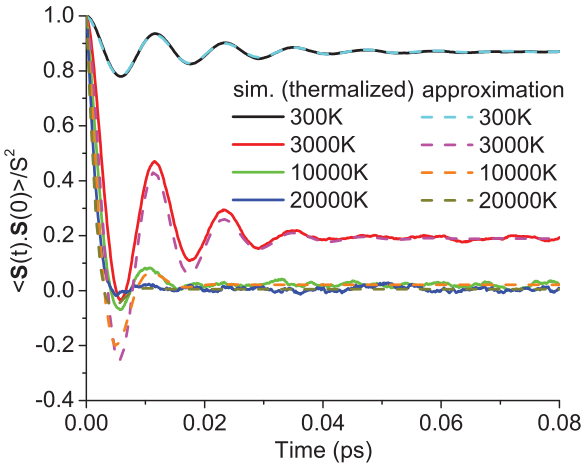

FIG. 6. (Color online) The normalized spin-spin self-correlation function plotted as a function of time for the case where the simulation starts from a thermalized spin ensemble. The approximate treatment based on Eqs. (44) and (45) still matches the simulation well, especially the relaxation behavior.

spin ensemble. We see that Eq. (45) still provides a fairly good description of the relaxation process. Although the oscillatory behavior predicted by Eq. (45) does not follow the simulation exactly, the overall trend still matches the simulation quite well.

VI. CONCLUSION

In this paper, we have derived a numerical algorithm for integrating the Langevin spin dynamics equations. The algorithm is based on the Suzuki-Trotter decomposition, and guarantees the preservation of the magnitude of the spin vector. Also the algorithm imposes no constraint on the magnitude of the fluctuation term, and does not require correcting the solution for a noise-induced drift term similar to that involved in the derivation of the Fokker-Planck equation, or similar methods.

We carried out the investigation starting from a set of equations of motion (1) for a system of interacting spins, and developed a rigorous integration algorithm considering, as an example, the case of a single spin (2) interacting with an external magnetic field. Of course, the algorithm applies fully to the treatment of a system of interacting spins.[18] The analysis given above opens an avenue for further study associated with the possibility of developing a self-consistent dynamic mean field treatment of spin evolution for a fully interacting system, where interaction with neighboring spins is treated self-consistently as being equivalent to the effect of an external time-dependent magnetic field.

To investigate spin relaxation, we derived a set of coupled deterministic equations for the statistical moments of solutions of the Langevin spin dynamics equations, and proved equivalence between the moments method and direct integration of the Langevin equation. Using the moments method, we showed that a spin ensemble interacting with delta-correlated stochastic noise evolves toward the equilibrium Gibbs distribution. By investigating two case studies we showed that both the thermalization process and the relaxation of spin-spin self-correlations are equally well described by the direct stochastic simulations and by the moments equations, which are in excellent agreement with each other.

Finally, we derived explicit analytical estimates for the relaxation time scales and showed that these estimates make it possible to identify the microscopic processes that dominate the relaxation of spin ensembles in the low and high temperature limits.

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