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We propose a functional integral framework for the derivation of hierarchies of Landau-Lifshitz-Bloch (LLB) equations that describe the flow toward equilibrium of the first and second moments of the magnetization. The short-scale description is defined by the stochastic Landau-Lifshitz-Gilbert equation, under both Markovian or non-Markovian noise, and takes into account interaction terms that are of practical relevance. Depending on the interactions, different hierarchies on the moments are obtained in the corresponding LLB equations. Two closure *Ansätze* are discussed and tested by numerical methods that are adapted to the symmetries of the problem. Our formalism provides a rigorous bridge between the atomistic spin dynamics simulations at short scales and micromagnetic descriptions at larger scales.

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I. INTRODUCTION

Thermal fluctuations of the magnetization can have a significant influence on the operating conditions of magnetic devices [1–3]. To describe them well is quite challenging, and the development of appropriate computational methods has a long history [4–6]. A textbook approach for their description is stochastic calculus [7,8]: The fluctuations are described by a thermal bath interacting with the magnetic degrees of freedom, namely, spins. The quantities of interest are the correlation functions of the magnetization, deduced from numerical simulations [9,10]. Former micromagnetic formulations do not account for a direct evaluation of the dynamics of those thermally activated correlation functions, which can be accessed naturally by experimental measurements.

The dynamics of those spins can be specified by a particular choice of a Langevin equation. A common and well-studied choice is the so-called stochastic form of the Landau-Lifshitz-Gilbert (SLLG) equation [11]. Up to a renormalization factor over the noise [12], the SLLG equation of motion for each spin component s_i can be written as

$$\frac{ds_i}{dt} = \frac{1}{1+\lambda^2}\epsilon_{ijk}s_k(\omega_j + \eta_j - \lambda\epsilon_{jlm}\omega_l s_m), \tag{1}$$

where the Einstein summation convention is adopted and ϵ_{ijk} describes the Levi-Cività fully antisymmetric pseudotensor. The vector $\boldsymbol{\omega}$ sets the precession frequency and is defined as [13]

$$\omega_i = -\frac{1}{\hbar}\frac{\partial \mathscr{H}}{\partial s_i}, \tag{2}$$

where $\mathscr{H}$ is the magnetic Hamiltonian of the system.

In Eq. (1), a random vector $\boldsymbol{\eta}$ defines the connection to the thermal bath. The components of $\boldsymbol{\eta}$ are assumed to be drawn from Gaussian distributions and are completely defined by the first two moments

$$\langle \eta_a(t)\rangle = 0, \quad \langle \eta_a(t)\eta_b(t')\rangle = \delta_{ab}C(t-t'), \tag{3}$$

with $C(t-t')$ a scalar correlation function of the bath. Here C can be any continuous function invariant by translation in time, assuming a weaker stationary form. The main advantage of such a weak stationary form is that it places any time series in the context of Hilbert spaces and as a consequence these series admit a Fourier-like decomposition on a spectral measure.

Within this framework, we mainly focus on the Markovian definition of the correlation function and we have $C(t-t') = 2D\,\delta(t-t')$, where D defines the amplitude of the noise. In the associated Supplemental Material (SM) [14], the invoked approach is more general and can also by applied to non-Markovian formulations, with the Ornstein-Uhlenbeck process as an example.

Equation (1) also describes a purely transverse damping with a nondimensional constant λ. Properly integrated [15,16], it ensures that the norm of the spin remains constant, which one can normalize to unity, $|\boldsymbol{s}| = 1$. This transverse damping term is responsible for the transfer of spin angular momentum from the magnetization to the environment, whereas the thermal bath allows energy to be pumped from the environment to the magnetization.

Many different mechanisms for damping are already known that include spin-orbit coupling, lattice vibrations, and spin waves [2]. Overall, this model is well suited for the simulation of an ensemble of interacting atomic magnetic spins including temperature effects [17,18].

When simulations of a spin ensemble (i.e., a finite-size magnetic grain) are at stake, it is well known that the norm of the average magnetization is not preserved and that longitudinal damping effects have to be taken into account [19–21].

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Although this mechanism is often accounted for in an empirical way, it has been shown that an appropriate averaging procedure over the bath can describe those longitudinal damping effects [22,23]. An equation describing the precession of the magnetization dynamics of nanomagnets and accounting for both transverse and longitudinal forms of damping is commonly referred to as a Landau-Lifshitz-Bloch (LLB) equation. Recent studies have been leveraging this model for the description of thermal fluctuations in different areas of magnetization dynamics, such as thermally assisted magnetic switching [24], ultrafast magnetism [25], or studies in chaotic magnetization dynamics [26].

In this work we present how a rigorous and systematic statistical averaging procedure, based on functional calculus, can be applied to the derivation of sets of LLB equations for the moments of the spin variables. Our approach differs quite significantly from previous attempts [22,23]. The form of the equilibrium distribution, for instance, is not postulated, but we try to deduce its properties from the evolution of the off-equilibrium dynamics of equal-time correlation functions. To this end, we explore the consequences of closure schemes on the evolution equations. We show how different magnetic interactions can be incorporated simply into the model. This makes the procedure particularly suitable and relevant for the derivation of effective micromagnetic equations from the knowledge of interactions at the atomic scale. This can more efficiently bridge the gap between atomistic methods, such as spin-dependent density-functional theory and classical spin dynamics, and macroequations used in micromagnetic simulations.

The plan of this work is the following. In the SM [14], the general framework based on functional calculus is presented. It allows us to take the statistical average over the equations of motion of the spins and to derive hierarchies of LLB equations. In Sec. II we show how this procedure can be applied to obtain the LLB equations for the first and second moments. The formalism is then applied to the case of a Markovian process and to three common magnetic interactions: a Zeeman interaction, a magnetic uniaxial anisotropy, and the exchange interaction. These equations are part of an open hierarchy. To solve them, we must impose closure conditions. In Sec. III we explore Gaussian as well as non-Gaussian closure conditions, derived from the theory of chaotic dynamical systems. To test their validity, we compare the results against those of a reference model, studied within the framework of stochastic atomistic spin dynamics simulations. A summary and our conclusions are presented in Sec. IV.

II. DERIVATION OF FIRST- AND SECOND-ORDER LLB EQUATIONS

In this section, the application of the formalism detailed in the SM [14] allows us to derive first- and second-order LLB equations. The derivation is particularized to three essential magnetic interactions, useful to describe realistic nanomagnets. An open hierarchy on the moments of the spin variables is generated. The hierarchy of LLB equations for the dynamics of the spin moments could easily be extended to higher orders, but is limited to the first- and second-order moments only for practical reasons.

A. LLB equations for paramagnetic spins in an external magnetic field

Atomic spins submitted to an external magnetic field are considered first. The contribution to the magnetic energy consists in a Zeeman term only. For a given magnetic spin $\boldsymbol{s}$, we have the magnetic Hamiltonian and magnetic precession vector

$$\mathscr{H}_{\text{Zeeman}} = -g\mu_B s_i B_i, \quad \omega_i^{\text{ext}} = \frac{g\mu_B}{\hbar} B_i, \tag{4}$$

respectively, with g the gyromagnetic ratio, μ_B the Bohr magneton, and $\boldsymbol{B}$ the external magnetic induction (in tesla). This contribution can be seen as an ultralocal expression, as it only accounts for the spin $\boldsymbol{s}$.

The corresponding equation of motion is obtained by inserting the expression of the precession vector ω_i^{ext} in Eq. (1):

$$\frac{ds_i}{dt} = \frac{1}{1+\lambda^2}\epsilon_{ijk}s_k\left(\omega_j^{\text{ext}} + \eta_j - \lambda\epsilon_{jlm}\omega_l^{\text{ext}}s_m\right). \tag{5}$$

Even if the external magnetic field $\boldsymbol{\omega}^{\text{ext}}$ may be time dependent, it is assumed to be independent of the noise and therefore can be taken out of any statistical average over the noise (see the SM [14] for clarification).

Applying the statistical averaging procedure described in the SM [14] to Eq. (5), we obtain the set of LLB-type equations on the first and second spin moments

$$\frac{d\langle s_i\rangle}{dt} = \frac{1}{1+\lambda^2}\epsilon_{ijk}\left(\omega_j^{\text{ext}}\langle s_k\rangle - \lambda\epsilon_{jlm}\omega_l^{\text{ext}}\langle s_k s_m\rangle\right) - \frac{2D}{(1+\lambda^2)^2}\langle s_i\rangle, \tag{6}$$

$$\frac{d\langle s_i s_j\rangle}{dt} = \frac{1}{1+\lambda^2}\epsilon_{ikl}\left(\omega_k^{\text{ext}}\langle s_j s_l\rangle - \lambda\epsilon_{kmn}\omega_m^{\text{ext}}\langle s_j s_l s_n\rangle\right) + \frac{D}{(1+\lambda^2)^2}(\delta_{ij}\langle s_n s_n\rangle - 3\langle s_i s_j\rangle), +(i \leftrightarrow j), \tag{7}$$

with $\langle s_i\rangle$ and $\langle s_i s_j\rangle$ denoting the first- and second-order statistical averages over the noise of the spin variables. Note that $\langle \boldsymbol{s}\rangle$ is also the average magnetization, a quantity commonly measured. The first terms on the right-hand sides of Eqs. (6) and (7) encapsulate the effects of the transverse dynamics (arising from the LLG equation), whereas the second term, which is proportional to the amplitude of the noise D, generates longitudinal damping effects and is a direct consequence of the statistical average over the noise. This first set of equations is very close to the one obtained by Garanin *et al.* from a similar approach, but with different hypothesis [22]. We also observe that this set of equations is not closed. Indeed, the right-hand side of Eq. (7) contains three-point moments $\langle s_j s_l s_n\rangle$, which are not defined yet.

B. LLB equations for anisotropic magnetic spins

The situation of spins in an anisotropic environment is now considered. The expression of the magnetic Hamiltonian accounting for the interaction between a given magnetic spin and a uniform axial magnetic anisotropy, defined by an

easy-axis $\boldsymbol{n}$ and an intensity K_a (in eV), is

$$\mathscr{H}_{\text{aniso}} = -\frac{K_a}{2}[(n_i s_i)^2 - 1]. \tag{8}$$

Again, this expression is ultralocal. Also, an uniaxial form of magnetic anisotropic contribution was considered, but other local forms (such as a cubic anisotropy) could have been considered the same way. The corresponding magnetic precession vector is given by

$$\omega_i^{\text{aniso}} = \omega_a(\boldsymbol{n}\cdot\boldsymbol{s})n_i, \tag{9}$$

which inserted into Eq. (1) gives

$$\frac{ds_i}{dt} = \frac{1}{1+\lambda^2}\epsilon_{ijk}s_k\left(\omega_j^{\text{aniso}} + \eta_j - \lambda\epsilon_{jlm}\omega_l^{\text{aniso}}s_m\right), \tag{10}$$

where $\omega_a \equiv 2K_a/\hbar$ is the effective field corresponding to the anisotropy.

The same statistical averaging procedure is repeated and applied to Eq. (10). The following set of anisotropic LLB equations is obtained:

$$\frac{d\langle s_i\rangle}{dt} = \frac{\omega_a}{1+\lambda^2}\epsilon_{ijk}(n_j n_l\langle s_k s_l\rangle - \lambda\epsilon_{jlm}n_l n_p\langle s_k s_m s_p\rangle) - \frac{2D}{(1+\lambda^2)^2}\langle s_i\rangle, \tag{11}$$

$$\frac{d\langle s_i s_j\rangle}{dt} = \frac{\omega_a}{1+\lambda^2}\epsilon_{imn}(n_m n_p\langle s_j s_n s_p\rangle - \lambda\epsilon_{mpq}n_p n_r\langle s_j s_n s_r s_q\rangle) + \frac{D}{(1+\lambda^2)^2}(\delta_{ij}\langle s_n s_n\rangle - 3\langle s_i s_j\rangle) + (i\leftrightarrow j). \tag{12}$$

Equations (11) and (12) present very similar features to Eqs. (6) and (7). The main difference lies in the open hierarchy on the moments. Indeed, the quadratic term on the right-hand side of Eq. (10) implies that Eq. (11) depends on three-point moments and Eq. (12) on four-point moments.

We observe that for the local interactions considered so far, the average-over-the-noise procedure does not affect the final expression of the longitudinal damping. Only the precession expression is modified.

C. LLB equations for ferromagnetic nanomagnets

The situation of a ferromagnetic medium, where a spin is submitted to local interactions, is now considered. Typically, the exchange interaction, which controls the local alignment and order of spins, is defined by the expression [17,18]

$$\mathscr{A}_{\text{ex}} = -\sum_{I,J\neq I}^{N}\mathsf{J}_{IJ}\boldsymbol{s}^I(t)\cdot\boldsymbol{s}^J(t), \tag{13}$$

where $\boldsymbol{s}^I(t)$ and $\boldsymbol{s}^J(t)$ are the values of neighboring spins (with I and J labeling different sites) at time t and J_{IJ} is the strength of the exchange interaction between these spins.

When working out of equilibrium, the dynamics described by a two-spin interaction can be reduced by an averaging method [27,28] to that of one spin in an effective field. The exchange interaction is then described by

$$\mathscr{A}_{\text{ex}}^{\text{MF}} = -\mathsf{J}_{\text{ex}}s_i(t)\cdot\langle s_i(t)\rangle, \tag{14}$$

where $\mathsf{J}_{\text{ex}} = n_v\mathsf{J}_{IJ}$, with n_v the number of spins in the considered neighbor shells.

This mean-field approximation allows us to remain within a local formulation. The statistical averaging procedure is only applied to a given spin, submitted to an average magnetic field, and connected to a thermal bath. Under these conditions, and injecting the precession vector resulting from Eq. (14) into Eq. (1), we have the equation of motion

$$\frac{ds_i}{dt} = \frac{\omega_{\text{ex}}}{1+\lambda^2}\epsilon_{ijk}s_k(\langle s_j\rangle + \eta_j - \lambda\epsilon_{jlm}\langle s_l\rangle s_m), \tag{15}$$

with $\omega_{\text{ex}} = \mathsf{J}_{\text{ex}}/\hbar$ the intensity of the exchange pulsation. Then the exchange interaction contributes to the LLB equations as

$$\frac{d\langle s_i\rangle}{dt} = \frac{\lambda\omega_{\text{ex}}}{1+\lambda^2}(\langle s_i\rangle\langle s_k s_k\rangle - \langle s_k\rangle\langle s_k s_i\rangle) - \frac{2D}{(1+\lambda^2)^2}\langle s_i\rangle, \tag{16}$$

$$\frac{d\langle s_i s_j\rangle}{dt} = \frac{\omega_{\text{ex}}}{1+\lambda^2}\epsilon_{ikl}(\langle s_k\rangle\langle s_j s_l\rangle - \lambda\epsilon_{kmn}\langle s_m\rangle\langle s_j s_l s_n\rangle) + \frac{D}{(1+\lambda^2)^2}(\delta_{ij}\langle s_n s_n\rangle - 3\langle s_i s_j\rangle) + (i\leftrightarrow j). \tag{17}$$

Simplifications have been performed in Eq. (16), as $\epsilon_{ijk}\langle s_j\rangle\langle s_k\rangle = 0$ within the mean-field approximation.

D. LLB equations for combined interactions

If a classical ferromagnet in a simultaneous anisotropic and external magnetic field is considered, the contribution of each interaction to the moment equations has to be computed. The final form can be obtained by straightforwardly taking the sum of the right-hand sides. The only subtle point is that the longitudinal damping contribution should not be overcounted. The full expressions are not very illuminating as such; it suffices to stress that they have been obtained under few and tightly controlled assumptions.

III. CLOSING THE HIERARCHY

In order to solve the sets of LLB equations for the first- and second-order moments of the spin variables obtained in the preceding section and to deduce the consequences for the magnetization dynamics itself (i.e., the first-order moments), the hierarchies must be closed. In this section, two closure methods inspired from turbulence theory [29,30] and dynamical systems [31] are reviewed and applied. A purely numerical reference model is presented first, which allows us to check the consistency and to assess the range of validity of the applied closures.

A. Reference model

Atomistic spin dynamics (ASD) simulations are commonly performed by solving sets of interacting SLLG equations [Eq. (1)], coupled to white-noise processes. For a single magnetic moment, getting that equation for ASD simulations has been considered from a quantum perspective [18,32,33], and several numerical implementations have been reported

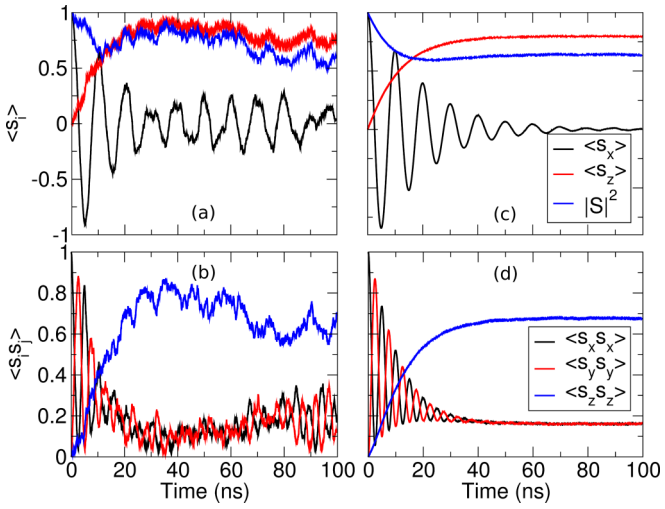

FIG. 1. Random magnetization dynamics of paramagnetic spins in a constant magnetic field: (a) and (c) some of the first-order moments and the norm of the averaged magnetization and (b) and (d) the diagonal elements only of the matrix of the second-order moments. The averages over ten paramagnetic spins only are shown in (a) and (b), whereas 10^4 spins are shown in (c) and (d). The parameters of the simulations are $D = 5 \times 10^{-2}$ rad GHz, $\lambda = 0.1$, $\vec{\omega} = (0, 0, 0.63)$ rad GHz, and time step $\Delta t = 10^{-3}$ ns. The initial conditions are $\vec{s}(0) = (1, 0, 0)$ and $\langle s_i(0)s_j(0)\rangle = 0$ except for $\langle s_x(0)s_x(0)\rangle = 1$.

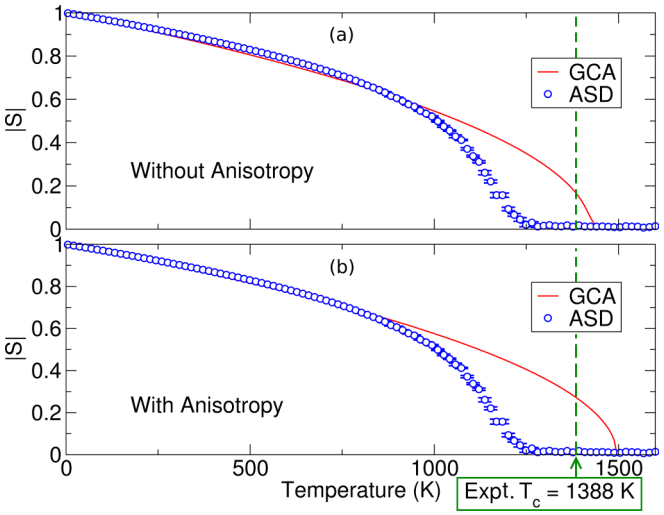

FIG. 2. Equilibrium magnetization norm vs temperature for hcp Co (a) without and (b) with the anisotropic interaction. The solid line plots the result of the GCA applied on the third-order moments. Open circles (with error bars) plot the ASD results, performed with the SLLG equation. The experimental Curie temperature T_C for hcp Co is also reported.

[17,34,35], including the exchange interaction, the treatment of external and anisotropic magnetic fields, and temperature.

For each individual spin, a SLLG equation (1) is integrated by a third-order Omelyan algorithm, which preserves the symplectic structure of the set of SLLG equations [36–38]. More details of this integration method have been provided in previous works [35,39]. These ASD simulations are performed for different noise realizations and averages are taken.

In practice, we find that it is possible to generate a sufficient number of noise configurations so that the map induced by the stochastic equations, as a result of this averaging procedure, realizes the exact statistical average over the noise [40,41]. These averages therefore define our reference model.

Figure 1 presents how effective this averaging procedure can be. The example of convergence toward a statistical average for paramagnetic spins is shown. From Fig. 1 we readily grasp that increasing the number of spins (or, equivalently, realizations) does accelerate the convergence toward the true averaged dynamics. For practical purposes, 10^4, interacting or not, spins are considered to be enough. This fixes statistical errors to a sufficiently low level to draw accurately the desired average quantities. From now on, this averaging procedure is used in order to check the consistency of the closure assumptions presented hereafter.

B. Gaussian closure assumption

A first closure assumption that is consistent with Gaussian statistics is a direct application of Wick's theorem [42]. This approach, referred to as the Gaussian closure assumption (GCA) in this work, has been briefly explored in previous studies [10,43].

Denoting the cumulant of any stochastic spin vector variable $\boldsymbol{s}$ by double angular brackets $\langle\langle\cdot\rangle\rangle$ [8], one has

$$\begin{aligned}\langle\langle s_i s_j s_l\rangle\rangle = {} & \langle s_i s_j s_l\rangle - \langle s_i s_j\rangle\langle s_l\rangle - \langle s_i s_l\rangle\langle s_j\rangle \\ & - \langle s_j s_l\rangle\langle s_i\rangle + 2\langle s_i\rangle\langle s_j\rangle\langle s_l\rangle \end{aligned} \tag{18}$$

for any combination of the space indices for the third-order cumulant and

$$\begin{aligned}\langle\langle s_i s_j s_l s_m\rangle\rangle = {} & \langle s_i s_j s_l s_m\rangle - 6\langle s_i\rangle\langle s_j\rangle\langle s_l\rangle\langle s_m\rangle - \langle s_i s_j\rangle\langle s_l s_m\rangle \\ & - \langle s_i s_l\rangle\langle s_j s_m\rangle - \langle s_i s_m\rangle\langle s_j s_l\rangle - \langle s_i\rangle\langle s_j s_l s_m\rangle \\ & - \langle s_j\rangle\langle s_i s_l s_m\rangle - \langle s_l\rangle\langle s_i s_j s_m\rangle - \langle s_m\rangle\langle s_i s_j s_l\rangle \\ & + 2[\langle s_i\rangle\langle s_j\rangle\langle s_l s_m\rangle + \langle s_i\rangle\langle s_l\rangle\langle s_j s_m\rangle \\ & + \langle s_i\rangle\langle s_m\rangle\langle s_j s_l\rangle + \langle s_j\rangle\langle s_l\rangle\langle s_i s_m\rangle \\ & + \langle s_j\rangle\langle s_m\rangle\langle s_i s_l\rangle + \langle s_l\rangle\langle s_m\rangle\langle s_i s_j\rangle] \end{aligned} \tag{19}$$

for any combination of the space indices for the fourth-order cumulant. The GCA implies that, for every time t, $\langle\langle s_i s_j s_k\rangle\rangle = 0$ and $\langle\langle s_i s_j s_k s_l\rangle\rangle = 0$. Thus the relationships

$$\langle s_i s_j s_k\rangle = \langle s_i\rangle\langle s_j s_k\rangle + \langle s_j\rangle\langle s_i s_k\rangle + \langle s_k\rangle\langle s_i s_j\rangle - 2\langle s_i\rangle\langle s_j\rangle\langle s_k\rangle, \tag{20}$$

$$\begin{aligned}\langle s_i s_j s_k s_l\rangle = {} & \langle s_i s_j\rangle\langle s_k s_l\rangle + \langle s_i s_k\rangle\langle s_j s_l\rangle \\ & + \langle s_i s_l\rangle\langle s_j s_k\rangle - 2\langle s_i\rangle\langle s_j\rangle\langle s_k\rangle\langle s_l\rangle, \end{aligned} \tag{21}$$

apply, relating thereby the third and fourth moments with the first and second ones only. Equations (20) and (21) have to be substituted into Eqs. (7), (11), (12), and (17). Because of the form these equations assume, they are referred to as dynamical Landau-Lifshitz-Bloch (DLLB) models.

Simulations of hcp cobalt are performed and the results are depicted in Figs. 2–4. These figures compare the GCA, applied to the third and fourth moments according to Eqs. (20)

and (21), to the ASD calculations performed for a hexagonal $22 \times 22 \times 22$ supercell. The nearest-neighbor and next-nearest-neighbor shells are taken into account for the exchange interaction, and its value, which is directly taken from *ab initio* calculations [44,45], is $J_{IJ} = 29.79$ meV for each atomic bond of the nearest neighbors and $J_{IJ} = 3.572$ meV for the next-nearest neighbors. The anisotropy energy for hcp Co is given to $K_a = 4.17 \times 10^{-2}$ meV for each spin, according to the same references [44,45].

The magnetic analog of the Einstein relation can be introduced in order to relate the amplitude D of the noise to the temperature of the bath [4,5]:

$$D = \frac{\lambda k_B T}{\hbar(1+\lambda^2)}. \tag{22}$$

The conditions for the validity of such an expression, which are not immediately obvious, are beyond the scope of this work and Eq. (22) is assumed to be valid. Thus, the averages over the noise can be replaced by the corresponding thermal averages. This allows us to have effective sets of DLLB micromagnetic equations at practical temperatures, which can be directly compared to experimental measurements.

Figure 2 plots the average magnetization norm versus the temperature for hcp Co with and without the anisotropic contribution, over a long simulation time, assuming the system to be at equilibrium. The application of the GCA matches well the ASD calculations, without requiring prior knowledge of the equilibrium magnetization value. Thus, the GCA can be considered to be a valid assumption for a broad range of temperatures (up to approximately two-thirds of the Curie temperature $2T_c/3$).

For higher temperatures, a departure from the ASD calculations is observed. This is not surprising as the correlation length of the connected real-space two-point correlation function at equilibrium grows without limit when T approaches T_c. Interestingly, the GCA allows us to observe the critical transition from a ferromagnetic to a paramagnetic phase, driven by the temperature.

Figure 3 plots the time dependence of the average magnetization for hcp Co for an external magnetic field of 10 T along the z axis, without any internal anisotropic contribution. The value of the external magnetic field is chosen to hasten the convergence of large ASD simulations. In addition, as the closure assumption does not rely on the intensity of the Zeeman interaction, any value of the field can be used. For T = 500 K, the GCA appears to be a good approximation and the two models are in good agreement. For T = 1000 K, the validity of the GCA becomes more questionable, in particular regarding the norm of the average magnetization and $\langle s_z \rangle$ at equilibrium.

Another interesting feature of this figure is the presence of two regimes for the magnetization dynamics. The first one is an extremely short thermalization regime. Because the exchange pulsation is the fastest pulsation in the system, the magnetization norm sharply decreases in order to balance the exchange energy with the thermal agitation. The second regime is the relaxation around the Zeeman field itself. The GCA model and the ASD simulations are in good agreement concerning the characteristic times of these two regimes.

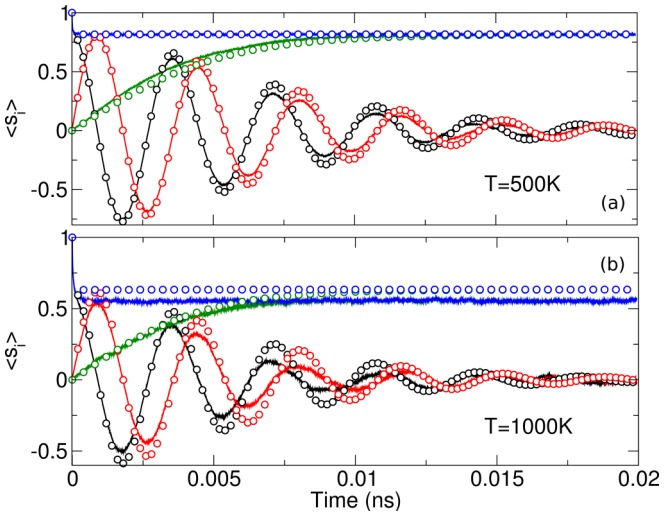

FIG. 3. Relaxation of the average dynamics up to 20 ps for (a) 500 K and (b) 1000 K, under a constant external magnetic induction of 10 T, applied on the z axis for initial conditions $s_y(0) = 1$ on each spin and $\lambda = 0.1$. The ASD results are shown as solid lines ($\langle s_x \rangle$ in black, $\langle s_y \rangle$ in red, $\langle s_z \rangle$ in blue, and $|s|$ in green), whereas the DLLB results with the GCA are shown as open circles.

Figure 4 displays the nonequilibrium profile of the average magnetization for hcp Co assuming axial anisotropy, oriented along the z axis, along with a small Zeeman field, also along the z axis, which ensures that the average magnetization aligns itself along the $+z$ direction. For $T = 500$ K, the GCA leads to the same equilibrium magnetization as the ASD, whereas for $T = 1000$ K, the average magnetization norm and the average magnetization along the z axis, calculated by the GCA, show deviations from the ASD calculations.

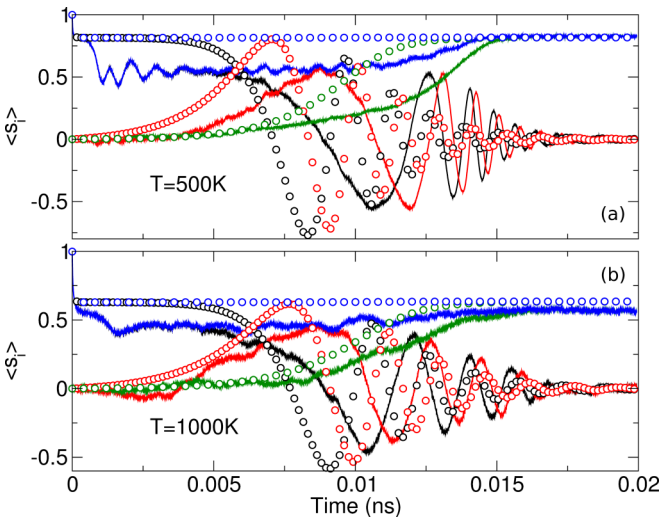

FIG. 4. Relaxation of the average dynamics up to 20 ps for (a) 500 K and (b) 1000 K, under an axial anisotropic field ($K_a = 4.17$ meV) and a constant external magnetic induction of 0.1 T, both applied along the z axis. The initial conditions are $s_x(0) = 1$ on each spin and $\lambda = 0.1$. The ASD results are depicted by solid lines ($\langle s_x \rangle$ in black, $\langle s_y \rangle$ in red, $\langle s_z \rangle$ in blue, and $|s|$ in green), whereas the DLLB results with the GCA are shown as open circles (see the text).

However, the GCA also fails to recover the transient dynamics of the relaxation found in the ASD simulations. The ASD calculations indicate a lag for the magnetization, compared to the results obtained by the GCA, and even if the precession frequency of the two models is the same, their dynamics are correspondingly shifted.

The origin of these departures for the transient regimes does not lie in the validity of the closure assumption itself, but in the fact that an averaged truncated (to the second-order spin moments) model is unable to recover the interacting magnon modes that are generated inside the large 22^3 ASD cell. Indeed, with a local anisotropy field only, the energetics of the spins is less constrained because individual spins may equilibrate along the two directions of the anisotropy axis. For a large but finite ASD cell, with periodic boundary conditions, this consequently generates local spin configurations (small subcells inside the large cell) due to the different realizations of the noise. In order to dissipate these subcells, additional internal magnon modes are produced. Their collective motion cannot be described by average thermal modes only, as we can see in the very beginning of the transient regimes of both graphs of Fig. 4. As a consequence, the effective precession around the anisotropy field is not shifted and delayed within the DLLB model with the GCA closure, whereas it is in the ASD simulation. In order to enhance the agreement for the equilibrium magnetization state of both ASD and averaged models, another closure method, more sophisticated than the GCA, is now considered.

C. Non-Gaussian closure

This method is inspired by studies in chaotic dynamical systems, where elaborate moment hierarchies are typically encountered [46,47]. Closure relations can be derived for the hierarchy of moments for the invariant measure of dynamical systems [48]. The proof relies on properties of the Fokker-Planck equation and on the assumption of ergodicity [49]. However, we saw in the preceding section that, depending on the magnetic interactions that are at stake, departures from ergodicity can be observed in the ASD simulations of large cells. Therefore, since the non-Gaussian closure assumption (NGCA) presented below is only expected to hold for ergodic situations, only the exchange and the Zeeman interactions will be considered, or cases when the Zeeman interaction is stronger than the anisotropic interaction, forcing each individual spin toward one possible equilibrium position.

The formalism can be presented as follows. Assuming ergodicity and with the cumulant notation at hand, such a NGCA relation can be parametrized for a stochastic variable $\boldsymbol{s}$ as

$$\begin{aligned}\langle\langle s_i s_j s_k\rangle\rangle = a_i^{(1)}\langle\langle s_j s_k\rangle\rangle + a_j^{(1)}\langle\langle s_i s_k\rangle\rangle + a_k^{(1)}\langle\langle s_i s_j\rangle\rangle \\ + a_{ij}^{(2)}\langle s_k\rangle + a_{ik}^{(2)}\langle s_j\rangle + a_{jk}^{(2)}\langle s_i\rangle \qquad (23)\end{aligned}$$

where the coefficients $a_i^{(1)}$ and $a_{ij}^{(2)}$ are assumed to depend not on time, but only on the system parameters, such as D, $\boldsymbol{\omega}$, and λ. These coefficients are assumed to be exactly zero when $D = 0$, hence matching the GCA. These NGCAs are tested with Eq. (7). The next logical step is to determine the values of these coefficients. As the third-order cumulants are symmetric under permutation of the coordinate indices, the coefficients $a_{ij}^{(2)}$ are symmetric too, and only nine coefficients have to be determined.

According to Nicolis *et al.* [49], these coefficients satisfy constraining identities that express physical properties of the considered spin systems. However, finding the general corresponding identities is quite nontrivial. To circumvent this difficulty, a fully computational approach was chosen. It is useful to stress that this approach is not without its proper theoretical basis: These identities indeed express properties of the functional integral [42].

Atomistic spin dynamics calculations are used to fit the coefficients $a_i^{(1)}$ and $a_{ij}^{(2)}$ for a given set of system parameters. At a given time, a distance function d, defined from the results of ASD simulations and our closed model as

$$\begin{aligned}d^2(t) \equiv \sum_{i=1}^{3}[\langle s_i(t)\rangle^{\mathrm{ASD}} - \langle s_i(t)\rangle]^2 \\ + \sum_{i,j=1}^{3}[\langle s_i s_j\rangle^{\mathrm{ASD}}(t) - \langle s_i s_j\rangle(t)]^2, \qquad (24)\end{aligned}$$

is computed and a least-squares-fitting method is applied. In this distance expression, each term is weighted equally to avoid any bias. At each step of the solver, a solution of the system of equations (6), (7), (11), (12), (16), and (17), closed by Eq. (23), is computed and the distance function is evaluated. From the evolution of this distance, the method determines another guess for the coefficients $a_i^{(1)}$ and $a_{ij}^{(2)}$. When the distance reaches a minimum, the hierarchy is assumed to be closed with the corresponding coefficients.

In order to check the validity of the NGCA, this was applied for the simulation performed at $T = 1000$ K presented in the preceding section, as in these situations neither the equilibrium nor the transient regimes of the ASD simulations were recovered by the GCA.

Another trial is carried out by performing again the simulation of Fig. 3(b). At the equilibration time, a minimum distance is found by considering a restriction to the third values only; thus we find $a_3^{(1)} = 0.145$ and $a_{33}^{(2)} = 0.145$. All the other coefficients are assumed to be zero. As expected, these dimensionless coefficients are small, demonstrating a slight departure of the GCA, which has to increase when the temperature increases. The uniqueness of these coefficients is not obvious and may depend on the choice of the distance function and its corresponding weights.

Figure 5 displays the result of this closure, with and without the anisotropic interaction. The two models present some slight differences in the beginning of the transient regime, but quickly match. This could be surely managed by increasing the number of distance points to match and by relaxing all the coefficients.

We now investigate the situation of including all interactions. For a different set of equations, a similar situation has already been investigated [43], with a slightly different closure method.

To close the hierarchy of moments in that case, an expression for the fourth-order moments $\langle s_i s_j s_k s_l\rangle$ is required. This is performed by assuming that the fourth-order cumulants

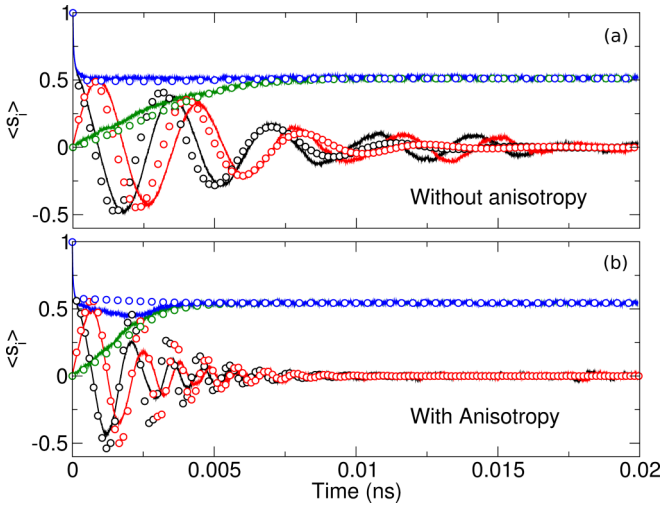

FIG. 5. Average dynamics up to 20 ps for $T = 1000$ K, with $\lambda = 0.1$ and initial conditions $\langle s_x(0)\rangle = 1$ for (a) a constant external field of 10 T applied along the z axis and (b) a uniaxial anisotropic field ($K_a = 4.17$ meV) and a constant external field of 10 T, both applied along the z axis. The ASD results are shown by solid lines ($\langle s_x\rangle$ in black, $\langle s_y\rangle$ in red, $\langle s_z\rangle$ in blue, and $|s|$ in green), whereas the DLLB results with the NGCA approximation are shown as open circles.

are negligible (i.e., $\langle\langle s_i s_j s_k s_l\rangle\rangle = 0$) and that each of the third-order moments is computed by Eq. (23). Yet again, one can systematically improve this hypothesis by increasing the number of desired coefficients up to this order such as

$$\begin{aligned}\langle\langle s_i s_j s_k s_l\rangle\rangle = b_i^{(1)}\langle\langle s_j s_k s_l\rangle\rangle + \text{permutation}\\ + b_{ij}^{(2)}\langle\langle s_k s_l\rangle\rangle + \text{permutation}\\ + b_{ijk}^{(3)}\langle s_l\rangle + \text{permutation}.\end{aligned}$$

Once again, invariance under permutation of indices enhances the symmetries of the $b^{(2)}$ and $b^{(3)}$ tensors and reduces the number of independent coefficients.

As a test, if we take all these coefficients b to be zero, at equilibrium, a minimum is found with $a_3^{(1)} = 0.07$ in the case where the z axis is preferred. Figure 5 displays the results of the application of the NGCA in that case. Again, the equilibrium state is recovered, even if some differences remain in the transient regime. By making educated guesses, we saw that the NGCA allows us to recover the equilibrium state of the magnetization, and much better agreement between ASD and DLLB models is also observed during the transient regimes.

IV. CONCLUSION

A framework based on a functional calculus approach was presented in the SM [14] associated with this article.

In Sec. II we showed that under controlled assumptions, this formalism can be applied to the derivation of open hierarchies of LLB equations on the moments of the magnetization dynamics. Within this study, the approach was limited to the situation of Markovian processes, even if it can be applied to more general correlation functions of the noise.

In Sec. III we saw that when the magnetic interactions include the exchange (within a mean-field approximation) and the Zeeman energy, a Gaussian closure assumption proved sufficient to recover both the transient regime and the equilibrium state of the average magnetization, for temperatures up to two-thirds of the Curie temperature. For higher temperatures, or when a magnetic anisotropy is taken into account, departures from the GCA can be observed. In those situations, a non-Gaussian closure method was proposed. It proved to be efficient at recovering the equilibrium state and improving the transient regime.

Finally, it should be emphasized that those sets of DLLB equations can be used to directly recover macroscopic magnetic quantities in micromagnetic simulations, such as the norm of the effective magnetization, the effective magnetic anisotropy, or the magnetic stiffness due to the exchange interaction, as well as performing direct simulations of the dynamics of nanomagnets. Only a prior knowledge of atomic-scale interactions (such as the interatomic exchange interaction or the per-atom magnetic anisotropy) is necessary. The implementation of those DLLB models as fundamental equations of micromagnetic numerical tools could greatly improve the accuracy of the latter and lead to new methodologies allowing us to bridge the gap between atomistic magnetic methods and micromagnetic simulations. Furthermore, whether they can describe deterministic chaos in micromagnetic systems deserves further study [26].

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