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Brownian rotation of classical spins: dynamical equations for non-bilinear spin-environment couplings

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Abstract. Dynamical equations for a classical spin interacting with the surrounding medium are derived by means of the formalism of the *oscillator-bath environment*. The bilinear-coupling treatment of Jayannavar (Z. Phys. B **82**, 153 (1991)) is extended to couplings that depend arbitrarily on the spin variables and are linear or linear-plus-quadratic in the environment dynamical variables. The dynamical equations obtained have the structure of generalised Langevin equations, which, in the Markovian approach, formally reduce to known semi-phenomenological equations of motion for classical magnetic moments. Specifically, the generalisation of the stochastic Landau–Lifshitz equation effected by Garanin, Ishchenko, and Panina (Theor. Math. Phys. **82**, 169 (1990)) in order to incorporate fluctuations of the magnetic anisotropy of the spin, is obtained for spin-environment interactions including up to quadratic terms *in the spin variables*. On the other hand, the portion of the coupling quadratic *in the environment variables* introduces an explicit dependence of the effective damping coefficients on the temperature.

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1 Introduction

The phenomenon of spin relaxation plays a significant rôle in condensed matter systems. Due to the interaction of a spin with the surrounding medium (phonons, conducting electrons, nuclear spins, etc.) its dynamics is quite complicated. The complexity itself, however, allows an idealisation of the phenomenon, by replacing the effect of the environment by a magnetic field randomly varying in time. Nevertheless, in order to describe the environmental effects properly and to attain a thermodynamically consistent description, the fluctuating terms must be supplemented with the analogue of a *relaxation* (damping or dissipative) term, to which must be linked by *fluctuation-dissipation* relations.

1.1 Phenomenological equations

For classical spins, the aforementioned program was carried out by Brown [1], in order to model the thermally activated rotation of the magnetic moment of a nanometric particle over the magnetic-anisotropy potential barriers (Néel rotation). This solid-state relaxation process was described by augmenting the phenomenological equation of Gilbert, which already embodied a damping term, by a fluctuating field $\mathbf{b}_{\rm fl}(t)$ in the following way

$$\frac{\mathrm{d}\mathbf{S}}{\mathrm{d}t}=\gamma\mathbf{S}\wedge\left[\mathbf{B}_{\rm eff}+\mathbf{b}_{\rm fl}(t)-(\gamma S)^{-1}\lambda\frac{\mathrm{d}\mathbf{S}}{\mathrm{d}t}\right]. \qquad (1)$$

Here λ is a dimensionless damping coefficient and $\mathbf{B}_{\rm eff}=-\partial\mathcal{H}_{\rm S}/\partial\mathbf{S}$ is the (deterministic) effective field associated with the Hamiltonian of the spin $\mathcal{H}_{\rm S}(\mathbf{S})$. This typically includes Zeeman and magnetic-anisotropy energy terms, *e.g.*, $\mathcal{H}_{\rm S}=-\mathbf{S}\cdot\mathbf{B}-\frac{1}{2}\beta(\mathbf{S}\cdot\mathbf{n})^2$ for uniaxial anisotropy with symmetry axis $\mathbf{n}$, whence $\mathbf{B}_{\rm eff}=\mathbf{B}+\hat{K}\mathbf{S}$, where $\hat{K}$ is a second-rank tensor with elements $K_{ij}=\beta n_i n_j$.

Equation (1), which was independently introduced by Kubo and Hashitsume [2], can be solved for $\mathrm{d}\mathbf{S}/\mathrm{d}t$ and cast into the archetypal Landau–Lifshitz form, and *vice versa*. However, we shall name the *stochastic Landau–Lifshitz equation* to its $\lambda\ll 1$ form ($|\mathbf{b}_{\rm fl}|\sim\lambda^{1/2}$)

$$\frac{\mathrm{d}\mathbf{S}}{\mathrm{d}t}=\gamma\mathbf{S}\wedge[\mathbf{B}_{\rm eff}+\mathbf{b}_{\rm fl}(t)]-\lambda\frac{\gamma}{S}\mathbf{S}\wedge(\mathbf{S}\wedge\mathbf{B}_{\rm eff}), \qquad (2)$$

since, in accordance with the spirit of its original deterministic counterpart [3], it describes weakly damped precession. Besides, except for a global time scale renormalisation, the *average* quantities derived from equation (1) are equivalent [4] to those associated with equation (2), which was indeed the equation thoroughly studied by Kubo and Hashitsume.

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On assuming that $\mathbf{b}_{\rm fl}(t)$ is Gaussian-distributed, centred (zero-mean), isotropic, and delta-correlated in time, Brown derived the Fokker–Planck (diffusion) equation governing the time evolution of the non-equilibrium probability distribution of spin orientations. By *imposing* that the Boltzmann distribution $P_0(\mathbf{S}) \propto \exp[-\mathcal{H}_{\rm S}(\mathbf{S})/k_{\rm B}T]$ is a stationary solution of that Fokker–Planck equation, the amplitude (variance) of $\mathbf{b}_{\rm fl}(t)$ was determined in terms of λ and T (this ensures the thermodynamic consistency of the approach). By a different method Kubo and Hashitsume arrived at an equation for the probability distribution, which, when the auto-correlation times of $\mathbf{b}_{\rm fl}(t)$ are much shorter than the precession period of $\mathbf{S}$, coincides with the Fokker–Planck equation of Brown in the absence of anisotropy potential (they considered the case $\mathcal{H}_{\rm S} = -\mathbf{S}\cdot\mathbf{B}$).

The Brown–Kubo–Hashitsume stochastic model has been the basis of significant studies of the dynamics of classical magnetic moments (see, *e.g.*, Refs. [5–11], to cite but a few). Nonetheless, there exist important microscopic relaxation mechanisms that cannot be accommodated in the context of this model, inasmuch as they do not produce a field-type perturbation on the spin ("field-type" fluctuations). An important example is the coupling of the spin to the lattice vibrations, which modulate the crystal-field and the exchange and dipole-dipole interactions, and can produce fluctuations of the magnetic-anisotropy potential of the spin ("anisotropy-type" fluctuations).

In order to take into account this phenomenon, Garanin, Ishchenko, and Panina [12] generalised the above Langevin equations to $\mathrm{d}\mathbf{S}/\mathrm{d}t = \gamma\mathbf{S}\wedge[\mathbf{B}_{\rm eff}+\mathbf{b}(t)+\hat{k}(t)\mathbf{S}]-\mathbf{R}$. Here, $\mathbf{R}$ is a relaxation term to be determined and, in analogy with the expression $\mathbf{B}_{\rm eff} = \mathbf{B} + \hat{K}\mathbf{S}$ for the effective field, $\mathbf{b}(t)$ is a stochastic *vector* that introduces the field-type part of the thermal fluctuations, while $\hat{k}(t)$ is a stochastic *second-rank tensor*, so $\hat{k}(t)\mathbf{S}$ incorporates anisotropy-type fluctuations into the dynamical equation.

On assuming the correlation properties

$$\begin{aligned}\langle b_i(t)b_j(s)\rangle &= \frac{2\lambda_{ij}}{\gamma S}\,k_{\rm B}T\delta(t-s),\\ \langle b_i(t)k_{jk}(s)\rangle &= \frac{2\lambda_{i,jk}}{\gamma S}\,k_{\rm B}T\delta(t-s),\\ \langle k_{ik}(t)k_{j\ell}(s)\rangle &= \frac{2\lambda_{ik,j\ell}}{\gamma S}\,k_{\rm B}T\delta(t-s),\end{aligned} \tag{3}$$

the associated Fokker–Planck equation was constructed [12]. The relaxation term $\mathbf{R}$ was then determined by merely assuming that the Boltzmann distribution is a stationary solution of their Fokker–Planck equation, getting (*cf.* Eq. (2))

$$\frac{\mathrm{d}\mathbf{S}}{\mathrm{d}t} = \gamma\mathbf{S}\wedge\left[\mathbf{B}_{\rm eff}+\mathbf{b}(t)+\hat{k}(t)\mathbf{S}\right] - \frac{\gamma}{S}\mathbf{S}\wedge\hat{G}\,(\mathbf{S}\wedge\mathbf{B}_{\rm eff})\,, \tag{4}$$

where $\hat{G}$ is a symmetrical second-rank tensor related with the correlation coefficients of the fluctuating terms by

$$G_{ij} = \lambda_{ij} + \sum_k(\lambda_{i,jk}+\lambda_{j,ik})S_k + \sum_{k\ell}\lambda_{ik,j\ell}S_kS_\ell. \tag{5}$$

For an arbitrary form of $\hat{G}$, the relaxation term in equation (4) deviates from the form proposed by Landau and Lifshitz. Only for $G_{ij} = \lambda\delta_{ij}$, which for instance occurs when both the field-type and the anisotropy-type fluctuations are isotropic ($\lambda_{ij} \propto \delta_{ij}$ and $\lambda_{ik,j\ell} \propto \delta_{ij}\delta_{k\ell}$) and there is not interference between them ($\lambda_{i,jk} \equiv 0$), that archetypal relaxation term is recovered and the Fokker–Planck equation of Garanin, Ishchenko, and Panina reduces to that obtained by Brown.

1.2 Dynamical approaches to the phenomenological equations

There have been several attempts to justify, starting from dynamical descriptions of a spin coupled to its surroundings, phenomenological equations for the stochastic spin dynamics.

Smith and De Rozario [13] considered a classical magnetic moment $\mathbf{S}$ coupled to a field $\mathbf{b}(P,Q)$ depending on the canonical momenta and coordinates (P,Q) of the environment. They derived a master equation for $\mathbf{S}$ by "projecting out" the environment variables, which, when the modulation due to the surroundings is fast in comparison with the precession period of $\mathbf{S}$, reduces to the Fokker–Planck equation associated with equation (2).

Seshadri and Lindenberg [14] studied a test spin interacting through a Heisenberg-type Hamiltonian with an environment consisting of other spins. The interaction among the latter was treated by a mean field approach, and a dynamical equation for the test spin was obtained to second order in the spin-environment coupling. The equation derived has the form of a generalised (*i.e.*, containing "memory" terms) Langevin equation, whose fluctuating and relaxation terms naturally obey fluctuation-dissipation relations.

Jayannavar [15] employed the *oscillator-bath* representation of the environment [16–20], and assumed a coupling linear in both the spin variables and the oscillator coordinates (*bilinear coupling*). A generalised Langevin equation for the spin was derived, which, in the Markovian approach (no memory) and for isotropic fluctuations, formally reduces to the stochastic Gilbert equation (1). (A similar treatment was presented in Ref. [21].) Equations of Landau–Lifshitz form, akin to those derived by Seshadri and Lindenberg, were also obtained in the weak-coupling regime.

Nevertheless, since spin-environment interactions linear in $\mathbf{S}$ produce a field-type perturbation on the spin (see below), the treatments mentioned do not account for fluctuations of the magnetic anisotropy of the spin. In this article, in order to incorporate this phenomenon, we shall extend the bilinear-coupling treatment of Jayannavar by considering general dependences of the spin-environment

coupling on the spin variables. Furthermore, we shall also include interactions quadratic in the oscillator variables (the classical analogue of, *e.g.*, two-phonon or two-photon relaxation processes), which are essential at sufficiently high temperatures. Because the ordinary formalism of the environment of independent oscillators [16–20] is not directly applicable when such quadratic couplings are included, we shall resort to a perturbational expansion in the spin-environment coupling, which is inspired on that of Cortés, West, and Lindenberg [22].

We shall obtain dynamical equations for the spin that have the structure of generalised Langevin equations with fluctuating terms $\gamma\mathbf{S}\wedge\mathbf{b}_{\rm fl}(\mathbf{S},t)$ and concomitant relaxation terms. These will have the form of a vector product of $\mathbf{S}(t)$ with a memory integral, which includes $(\mathrm{d}\mathbf{S}/\mathrm{d}t)(s)$ or $(\mathbf{S}\wedge\mathbf{B}_{\rm eff})(s)$ for weak coupling, taken along the past history ($s\leq t$) of the spin. In the Markovian approach, the equations derived will reduce to the form $\mathrm{d}\mathbf{S}/\mathrm{d}t = \gamma\mathbf{S}\wedge[\mathbf{B}_{\rm eff}+\mathbf{b}_{\rm fl}(\mathbf{S},t)]-\mathbf{R}$, where for couplings *linear* in the environmental variables the relaxation term reads $\mathbf{R}=\frac{1}{S}\mathbf{S}\wedge\hat{\Lambda}^{(\mathrm{L})}(\mathrm{d}\mathbf{S}/\mathrm{d}t)$ or $\mathbf{R}=\frac{\gamma}{S}\mathbf{S}\wedge\hat{\Lambda}^{(\mathrm{L})}(\mathbf{S}\wedge\mathbf{B}_{\rm eff})$ for weak coupling, $\hat{\Lambda}^{(\mathrm{L})}$ being a second-rank tensor depending on the structure of the coupling. In addition, when interactions *quadratic* in the environment variables are also accounted for, the relaxation term will depend explicitly on the temperature and, in the Markovian approach, $\mathbf{R}$ will take the form $\mathbf{R}=\frac{\gamma}{S}\mathbf{S}\wedge\hat{\Lambda}(\mathbf{S}\wedge\mathbf{B}_{\rm eff})$, where $\hat{\Lambda}=\hat{\Lambda}^{(\mathrm{L})}+k_{\rm B}T\,\hat{\Lambda}^{(\mathrm{Q})}$ and the additional tensor $\hat{\Lambda}^{(\mathrm{Q})}$ is determined by the quadratic portion of the coupling.

Since the fluctuating effective field $\mathbf{b}_{\rm fl}(\mathbf{S},t)$ will depend in general on $\mathbf{S}$, it can incorporate fluctuations of the magnetic anisotropy of the spin. For instance, when the spin-environment interaction includes terms up to quadratic *in the spin variables*, $\mathbf{b}_{\rm fl}(\mathbf{S},t)$ can be written as $\mathbf{b}(t)+\hat{k}(t)\mathbf{S}$, with the correlation coefficients of the fluctuating terms being related to the tensors $\hat{\Lambda}$ by expressions identical with equation (5). In this way, the generalisation of the classic Brown–Kubo–Hashitsume model effected by Garanin, Ishchenko, and Panina will formally be obtained.

2 Free dynamics and canonical variables

The dynamical equation for an isolated classical spin with Hamiltonian $\mathcal{H}_{\rm S}(\mathbf{S})$ is

$$\frac{\mathrm{d}\mathbf{S}}{\mathrm{d}t}=\gamma\mathbf{S}\wedge\mathbf{B}_{\rm eff},\qquad \mathbf{B}_{\rm eff}=-\frac{\partial\mathcal{H}_{\rm S}}{\partial\mathbf{S}}\cdot \tag{6}$$

These vectorial equations, which merely express the precession of $\mathbf{S}$ about the instantaneous effective field, can be written as

$$\frac{\mathrm{d}\varphi}{\mathrm{d}t}=-\frac{\gamma}{S}\frac{1}{\sin\vartheta}\frac{\partial\mathcal{H}_{\rm S}}{\partial\vartheta},\qquad \frac{\mathrm{d}\vartheta}{\mathrm{d}t}=\frac{\gamma}{S}\frac{1}{\sin\vartheta}\frac{\partial\mathcal{H}_{\rm S}}{\partial\varphi},$$

where φ and ϑ are, respectively, the azimuthal and polar angles of $\mathbf{S}$. Furthermore, these formulae are equivalent to the Hamilton equations

$$\frac{\mathrm{d}q}{\mathrm{d}t}=\frac{\partial\mathcal{H}_{\rm S}}{\partial p},\qquad \frac{\mathrm{d}p}{\mathrm{d}t}=-\frac{\partial\mathcal{H}_{\rm S}}{\partial q},$$

with the conjugate canonical variables[1]

$$q=\varphi,\qquad p=S_z/\gamma. \tag{7}$$

In terms of these variables $\mathbf{S}$ is given by

$$\begin{aligned} S_x&=\sqrt{S^2-(\gamma p)^2}\cos q,\\ S_y&=\sqrt{S^2-(\gamma p)^2}\sin q,\\ S_z&=\gamma p, \end{aligned} \tag{8}$$

from which one can readily obtain the customary Poisson-bracket ("commutation") relations among the spin variables: $\{S_i,S_j\}=\gamma\sum_k\epsilon_{ijk}S_k$, where ϵ_{ijk} is the Levi–Civita symbol and the Poisson bracket is defined as $\{A,B\}\equiv[(\partial A/\partial q)(\partial B/\partial p)-(\partial A/\partial p)(\partial B/\partial q)]$. In addition, on using the *chain rule* of the Poisson bracket (*i.e.*, $\{f,g\}=\sum_{i,k}\frac{\partial f}{\partial x_i}\frac{\partial g}{\partial x_k}\{x_i,x_k\}$, where $x_i=x_i(p,q)$) one gets the useful relation (*cf.* Eq. (13) of Ref. [13])

$$\{S_i,V(\mathbf{S})\}=-\gamma\left(\mathbf{S}\wedge\frac{\partial V}{\partial\mathbf{S}}\right)_i, \tag{9}$$

which is valid for any function of the spin variables $V(\mathbf{S})$. Naturally, one can conversely postulate the relations $\{S_i,S_j\}=\gamma\sum_k\epsilon_{ijk}S_k$ and then derive at equation (6) starting from the basic Hamiltonian evolution equation $\mathrm{d}S_i/\mathrm{d}t=\{S_i,\mathcal{H}_{\rm S}\}$ and using equation (9).

3 Dynamical equations for couplings linear in the environment variables

In this section we shall study a classical spin surrounded by an environment that can be represented by a set of independent classical harmonic oscillators. These may correspond to the *normal modes* of an electromagnetic field, the lattice vibrations (in the harmonic approximation), or they can be an effective low-energy description of a more general surrounding medium [19]. We shall assume that the spin-environment interaction is linear in the coordinates of the oscillators but otherwise arbitrary in the spin variables. In this way, fluctuations of the magnetic anisotropy of the spin will be incorporated in the dynamical equations.

3.1 The spin-environment Hamiltonian: couplings linear in the environment variables

The total system consisting of the spin (the "system of interest") plus the oscillators representing the environment forms a *closed* dynamical system that we shall describe by augmenting the isolated-spin Hamiltonian as follows

$$\mathcal{H}=\mathcal{H}_{\rm S}(\mathbf{S})+\sum_\alpha\frac{1}{2}\Big\{P_\alpha^2+\omega_\alpha^2\Big[Q_\alpha+\frac{\varepsilon}{\omega_\alpha^2}F_\alpha(\mathbf{S})\Big]^2\Big\}. \tag{10}$$

[1] The alternative choice $\tilde{q}=S_z/\gamma$ and $\tilde{p}=-\varphi$ of, *e.g.*, reference [23] is equivalent to the one considered here through the *canonical* transformation $q=-\tilde{p}$ and $p=\tilde{q}$.

Here, α is an oscillator index (*e.g.*, the pair $(\mathbf{k}, s)$ formed by the wave-vector and branch index of a normal mode of the environment), and the coupling terms $F_\alpha(\mathbf{S})$ are arbitrary functions of the spin variables (typically polynomials in $\mathbf{S}$). These terms may depend on the parameters of the oscillators ω_α, but not on their dynamical variables P_α, Q_α. On the other hand, for the sake of convenience in keeping track of the various orders, we have introduced a spin-environment coupling constant ε, which in the weak-coupling approximation will be considered small.

The terms proportional to F_α^2, which emerge when squaring $Q_\alpha + (\varepsilon/\omega_\alpha^2)F_\alpha$, are "counter-terms" introduced to balance the coupling-induced renormalisation of the Hamiltonian of the spin. The formalism takes as previously considered whether such a renormalisation actually occurs for a given interaction [19], so that $\mathcal{H}_\mathrm{S}$ would already include it (whenever exists). An advantage of this convention is that one deals with the experimentally accessible energy of the spin, instead of the "bare" one, which might be difficult to determine.

The introduction of *non-linear* coupling terms $F_\alpha(\mathbf{S})$, as otherwise occur in various relevant situations ($F_\alpha \propto \sum S_k S_\ell$ for the magneto-elastic coupling of $\mathbf{S}$ to the lattice vibrations), will be essential to get fluctuations of the magnetic anisotropy of the spin. The starting Hamiltonian in the work of Jayannavar [15] was similar to (10) but with a special type of *linear* $F_\alpha(\mathbf{S})$: the component S_i of the magnetic moment was coupled to the ith Cartesian component $Q_{\alpha,i}$ of certain three-dimensional oscillators. This specific *bilinear* interaction yielded, not only field-type fluctuations, but also uncorrelated ones. (In Ref. [21] couplings non-linear in $\mathbf{S}$ were also considered, but in that work the focus was on the existence of thermal equilibrium in the Markovian limit.)

3.2 Dynamical equations: general case

For the sake of simplicity in notation but also of generality, we cast the Hamiltonian (10) into the form

$$\mathcal{H} = \mathcal{H}_\mathrm{S}^{(\mathrm{m})}(p,q) + \sum_\alpha \frac{1}{2}\left(P_\alpha^2 + \omega_\alpha^2 Q_\alpha^2\right) + \varepsilon \sum_\alpha Q_\alpha F_\alpha(p,q), \tag{11}$$

where q and p are the canonical coordinate and conjugate momentum of a "system" with Hamiltonian $\mathcal{H}_\mathrm{S}(p,q)$ (in the spin-dynamics case p and q are given by Eqs. (7)), and the "modified" system Hamiltonian $\mathcal{H}_\mathrm{S}^{(\mathrm{m})}$ augments $\mathcal{H}_\mathrm{S}$ by the aforementioned counter-terms

$$\mathcal{H}_\mathrm{S}^{(\mathrm{m})} = \mathcal{H}_\mathrm{S} + \frac{\varepsilon^2}{2}\sum_\alpha \frac{F_\alpha^2}{\omega_\alpha^2}\cdot \tag{12}$$

The equation of motion for any dynamical variable without explicit dependence on the time, $C(p,q;P,Q)$, where (P,Q) stands for the set of canonical variables of the environment, is given by the basic Hamiltonian evolution equation $\mathrm{d}C/\mathrm{d}t = \{C, \mathcal{H}\}$, where the whole Poisson bracket is given by

$$\{A,B\} \equiv \frac{\partial A}{\partial q}\frac{\partial B}{\partial p} - \frac{\partial A}{\partial p}\frac{\partial B}{\partial q} + \sum_\alpha \frac{\partial A}{\partial Q_\alpha}\frac{\partial B}{\partial P_\alpha} - \frac{\partial A}{\partial P_\alpha}\frac{\partial B}{\partial Q_\alpha}\cdot$$

One therefore gets from the Hamiltonian (11) the following coupled equations of motion for *any dynamical variable (observable) of the system* $A(p,q)$ and the environment variables ($C = A, P_\alpha$, and Q_α)

$$\frac{\mathrm{d}A}{\mathrm{d}t} = \{A, \mathcal{H}_\mathrm{S}^{(\mathrm{m})}\} + \varepsilon \sum_\alpha Q_\alpha \{A, F_\alpha\}, \tag{13}$$

$$\frac{\mathrm{d}Q_\alpha}{\mathrm{d}t} = P_\alpha, \qquad \frac{\mathrm{d}P_\alpha}{\mathrm{d}t} = -\omega_\alpha^2 Q_\alpha - \varepsilon F_\alpha. \tag{14}$$

The goal is to derive a dynamical equation for $A(p,q)$ involving the system variables only (*reduced* dynamical equation). Then, the corresponding equation for the spin will be obtained by replacing $A(p,q)$ in that equation by the Cartesian components of $\mathbf{S}$ (Eq. (8)).

On considering that in equations (14) the term $-\varepsilon F_\alpha(t) = -\varepsilon F_\alpha[p(t),q(t)]$ plays the rôle of a time-dependent forcing on the oscillators, those equations can be explicitly integrated, yielding

$$Q_\alpha(t) = Q_\alpha^\mathrm{h}(t) - \frac{\varepsilon}{\omega_\alpha}\int_{t_0}^t \mathrm{d}s\, \sin[\omega_\alpha(t-s)]F_\alpha(s), \tag{15}$$

where

$$Q_\alpha^\mathrm{h}(t) = Q_\alpha(t_0)\cos[\omega_\alpha(t-t_0)] + \frac{P_\alpha(t_0)}{\omega_\alpha}\sin[\omega_\alpha(t-t_0)], \tag{16}$$

are the solutions of the *homogeneous* system of equations for the oscillators in the absence of the system-environment interaction (proper modes of the environment). Then, on integrating by parts in equation (15) one gets for the combination εQ_α that appears in equation (13)

$$\varepsilon Q_\alpha(t) = f_\alpha(t) - [\mathcal{K}_\alpha(t-s)F_\alpha(s)]_{s=t_0}^{s=t} + \int_{t_0}^t \mathrm{d}s\, \mathcal{K}_\alpha(t-s)\frac{\mathrm{d}F_\alpha}{\mathrm{d}t}(s), \tag{17}$$

where

$$f_\alpha(t) = \varepsilon Q_\alpha^\mathrm{h}(t), \qquad \mathcal{K}_\alpha(\tau) = \frac{\varepsilon^2}{\omega_\alpha^2}\cos(\omega_\alpha\tau). \tag{18}$$

Next, in order to eliminate the environment variables from the equation for $A(p,q)$, one substitutes equation (17) back into equation (13). This yields a term $\sum_\alpha \{A, F_\alpha\}\mathcal{K}_\alpha(t-t_0)F_\alpha(t_0)$ that depends on the initial state of the system $(p(t_0), q(t_0))$ and produces a transient response that can be ignored in the long-time dynamics (we shall however return to this question

below)[2]. The parallel term $-\sum_\alpha \{A, F_\alpha\}\mathcal{K}_\alpha(0)F_\alpha(t)$, which is derivable from a Hamiltonian, is exactly balanced by the term originated from the counter-terms in $\{A, \mathcal{H}_{\rm S}^{(\rm m)}\}$. This can be shown by using $-\sum_\alpha \{A, F_\alpha\}\mathcal{K}_\alpha(0)F_\alpha = \{A, -\frac{1}{2}\sum_\alpha \mathcal{K}_\alpha(0)F_\alpha^2\}$, which follows from the *product rule* $\{A, BC\} = \{A, B\}C + \{A, C\}B$ of the Poisson bracket, and then accounting for $\mathcal{K}_\alpha(0) = \varepsilon^2/\omega_\alpha^2$ (see Eq. (18)).

Therefore, one is finally left with the *reduced* dynamical equation

$$\frac{{\rm d}A}{{\rm d}t} = \{A, \mathcal{H}_{\rm S}\} + \sum_\alpha \{A, F_\alpha\}\left[f_\alpha(t) + \int_{t_0}^t {\rm d}s\, \mathcal{K}_\alpha(t-s)\frac{{\rm d}F_\alpha}{{\rm d}t}(s)\right], \quad (19)$$

where the first term yields the free (conservative) time evolution of the system, whereas the second term incorporates the effects of the interaction of the system with its environment. The terms $f_\alpha(t)$ are customarily interpreted as *fluctuating* "forces" (or "fields"), while the integral term, which keeps in general memory of the previous history of the system, provides the *relaxation* due to the interaction with the surrounding medium[3].

The origin of both types of terms can be traced back as follows. Recall that in equation (15) the time evolution of the oscillators has formally been written as if they were driven by (time-dependent) forces $-\varepsilon F_\alpha[p(s), q(s)]$ depending on the state of the system. Therefore, $Q_\alpha(t)$ consists of the sum of the proper (free) mode $Q_\alpha^{\rm h}(t)$ and the driven-type term, which naturally depends on the "forcing" (state of the system) at previous times. Then, the replacement of Q_α in the equation for the system variables by the driven-oscillator solution incorporates:

(i) The time-dependent modulation due to the proper modes of the environment.
(ii) The "back-reaction" on the system of its preceding action on the surrounding medium.

Thus, the formalism leads to a description in terms of a reduced number of dynamical variables at the expense of both explicitly time-dependent (fluctuating) terms and history-dependent (relaxation) terms.

Archetypal example: Brownian particle

In order to particularise these general expressions to definite situations, the structure of the coupling terms F_α needs to be specified. For instance, on setting $F_\alpha(p, q) = -c_\alpha q$ (bilinear coupling), where the $c_\alpha = c_\alpha(\omega_\alpha)$ are coupling constants, and writing down equation (19) for $A = q$ and $A = p$, one gets the celebrated generalised Langevin equation for a "Brownian" particle [18]

$$\frac{{\rm d}q}{{\rm d}t} = \frac{\partial \mathcal{H}_{\rm S}}{\partial p}, \quad \frac{{\rm d}p}{{\rm d}t} = -\frac{\partial \mathcal{H}_{\rm S}}{\partial q} + f(t) - \int_{t_0}^t {\rm d}s\, \mathcal{K}(t-s)\frac{{\rm d}q}{{\rm d}t}(s). \quad (20)$$

Here, $f(t) = \sum_\alpha c_\alpha f_\alpha(t)$ is the fluctuating force and $\mathcal{K}(\tau) = \sum_\alpha c_\alpha^2 \mathcal{K}_\alpha(\tau)$ is the memory kernel, the relaxation term associated with which comprises minus the velocity $-({\rm d}q/{\rm d}t)(s)$ of the particle (*viscous damping*).

In general, when $\{A, F_\alpha\}$ in equation (19) is not constant, the fluctuating terms $f_\alpha(t)$ enter multiplying the system variables (*multiplicative* fluctuations). In this example, owing to $\{q, -c_\alpha q\} = 0$ and $\{p, -c_\alpha q\} = c_\alpha$, the fluctuations are *additive*.

3.3 Dynamical equations: the spin-dynamics case

Let us now particularise the above results to the dynamics of a classical spin. Here, we introduce the coupling functions

$$F_\alpha(\mathbf{S}) = \sum_l c_\alpha^l V_l(\mathbf{S}), \quad (21)$$

where l stands for a general index depending on the type of interaction, the coefficients c_α^l are spin-environment coupling constants, and the terms $V_l(\mathbf{S})$ are certain functions of the spin variables. In order to motivate this expression, consider, *e.g.*, the magneto-elastic coupling of $\mathbf{S}$ to the lattice vibrations. The index l then stands for a pair of Cartesian indices (ij) and $V_l \to V_{ij} = \sum_{k\ell} a_{ij,k\ell} S_k S_\ell$, where the $a_{ij,k\ell}$ are magneto-elastic coefficients.

In order to derive the reduced dynamical equation for the spin, we merely put $A = S_i$, $i = x, y, z$, in equation (19), and then use equation (9) to calculate the Poisson brackets required. On gathering the results so-obtained in vectorial form and using $\mathbf{B}_{\rm eff} = -\partial \mathcal{H}_{\rm S}/\partial \mathbf{S}$ and ${\rm d}V_{l'}/{\rm d}t = (\partial V_{l'}/\partial \mathbf{S}) \cdot ({\rm d}\mathbf{S}/{\rm d}t)$, we arrive at

$$\frac{{\rm d}\mathbf{S}}{{\rm d}t} = \gamma \mathbf{S} \wedge \left\{ \mathbf{B}_{\rm eff} + \mathbf{b}_{\rm fl}(\mathbf{S}, t) - \int_{t_0}^t {\rm d}s\, \hat{\Gamma}^{(\rm L)}(\mathbf{S}; t, s)\frac{{\rm d}\mathbf{S}}{{\rm d}t}(s) \right\}. \quad (22)$$

In this equation the *fluctuating magnetic field* is given by

$$\mathbf{b}_{\rm fl}(\mathbf{S}, t) = -\sum_l f_l(t)\frac{\partial V_l}{\partial \mathbf{S}}, \quad (23)$$

which involves the environmental proper modes *via* the fluctuating sources

$$f_l(t) = \varepsilon \sum_\alpha c_\alpha^l Q_\alpha^{\rm h}(t). \quad (24)$$

[2] In the ordinary independent oscillator model, one considers $F_\alpha(p, q) \propto q$ and the corresponding terms can formally be removed from the dynamical equations by choosing the origin of the "coordinate frame" to lay at the "position" of the system at $t = t_0$, that is, $F_\alpha(t_0) \propto q(t_0) = 0$. However, this frame-dependent procedure cannot be employed if the system comprises different entities. In addition, in the spin-dynamics case with, *e.g.*, $F_\alpha(\mathbf{S})$ linear in $\mathbf{S}$, one cannot set $\mathbf{S}(t_0) = \mathbf{0}$ due to the conservation of the length of the spin.

[3] Note that without the integration by parts yielding equation (17), the Hamiltonian (renormalisation) terms would occur inconveniently mixed in the integral term.

On the other hand, the relaxation tensor in equation (22) reads[4]

$$\hat{\Gamma}^{(\mathrm{L})}(\mathbf{S};t,s)=\sum_{l,l'}\mathcal{K}_{ll'}(t-s)\frac{\partial V_l}{\partial \mathbf{S}}(t)\frac{\partial V_{l'}}{\partial \mathbf{S}}(s), \tag{25}$$

where the *memory kernel* is given by

$$\mathcal{K}_{ll'}(\tau)=\varepsilon^2\sum_{\alpha}\frac{C_\alpha^l C_\alpha^{l'}}{\omega_\alpha^2}\cos(\omega_\alpha\tau). \tag{26}$$

Equation (22) contains $\mathrm{d}\mathbf{S}/\mathrm{d}t$ on its right-hand side, so it will be referred to as a *Gilbert-type* equation (*cf.* Eq. (1)). For $\varepsilon \ll 1$, on substituting perturbatively that derivative by its conservative part, $\mathrm{d}\mathbf{S}/\mathrm{d}t \simeq \gamma\mathbf{S}\wedge\mathbf{B}_{\mathrm{eff}}$, one gets the weak-coupling *Landau–Lifshitz-type* equation

$$\frac{\mathrm{d}\mathbf{S}}{\mathrm{d}t}=\gamma\mathbf{S}\wedge[\mathbf{B}_{\mathrm{eff}}+\mathbf{b}_{\mathrm{fl}}(\mathbf{S},t)] - \gamma\mathbf{S}\wedge\left\{\int_{t_0}^{t}\mathrm{d}s\,\gamma\hat{\Gamma}^{(\mathrm{L})}(\mathbf{S};t,s)\,(\mathbf{S}\wedge\mathbf{B}_{\mathrm{eff}})\,(s)\right\}, \tag{27}$$

which describes weakly damped precession.

For spin-environment interactions *linear* in the environment variables but being otherwise *arbitrary* functions of $\mathbf{S}$, equations (22, 27) are the desired reduced dynamical equations for the spin. They have the structure of generalised Langevin equations with *fluctuating* terms $\gamma\mathbf{S}\wedge\mathbf{b}_{\mathrm{fl}}(\mathbf{S},t)$ (associated with the modulation by the proper modes of the environment) and history-dependent *relaxation* terms (corresponding to the back-reaction on the spin of its previous action on the surrounding medium).

Note that $f_l(t)$ (Eq. (24)) is a sum of a large number of sinusoidal terms with different frequencies and phases; this can give to $f_l(t)$ the form of a highly irregular function of t that is expected for a fluctuating term. However, for a general form of the coupling functions $V_l(\mathbf{S})$, the term $\mathbf{b}_{\mathrm{fl}}(\mathbf{S},t)$ *cannot* be interpreted as a fluctuating *ordinary* field, since it may depend on $\mathbf{S}$, but it is rather a fluctuating *effective* field to be added to the deterministic effective field $\mathbf{B}_{\mathrm{eff}}=-\partial\mathcal{H}_{\mathrm{S}}/\partial\mathbf{S}$ (Eq. (6)). This can be illustrated by phrasing the discussion in terms of the *fluctuating part* of the energy of the spin, namely (see Hamiltonian (11)): $\mathcal{H}_{\mathrm{fl}}=\varepsilon\sum_\alpha Q_\alpha^{\mathrm{h}}(t)F_\alpha(\mathbf{S})$. From this definition one first gets

$$\mathcal{H}_{\mathrm{fl}}(\mathbf{S},t)=\sum_l f_l(t)V_l(\mathbf{S}),\quad \mathbf{b}_{\mathrm{fl}}(\mathbf{S},t)=-\frac{\partial\mathcal{H}_{\mathrm{fl}}}{\partial\mathbf{S}}\cdot \tag{28}$$

So that $\mathbf{b}_{\mathrm{fl}}(\mathbf{S},t)$ can be derived from $\mathcal{H}_{\mathrm{fl}}(\mathbf{S},t)$ in the same way as $\mathbf{B}_{\mathrm{eff}}(\mathbf{S})$ is obtained from $\mathcal{H}_{\mathrm{S}}(\mathbf{S})$. Next, recall that the non-linear part of $\mathcal{H}_{\mathrm{S}}(\mathbf{S})$ carries the anisotropy-energy terms, *e.g.*, $\mathcal{H}_{\mathrm{S}}=-\mathbf{S}\cdot\mathbf{B}-\frac{1}{2}\beta(\mathbf{S}\cdot\mathbf{n})^2$ in a uniaxial crystal. Analogously, $\mathcal{H}_{\mathrm{fl}}$ has the form $\mathcal{H}_{\mathrm{fl}}(\mathbf{S},t)=-\mathbf{S}\cdot\mathbf{b}_{\mathrm{fl}}(t)$, with $\mathbf{b}_{\mathrm{fl}}$ independent of $\mathbf{S}$, only for linear $V_l(\mathbf{S})$ (bilinear coupling), so that *the non-linear part of* $V_l(\mathbf{S})$ *incorporates fluctuations of the magnetic anisotropy of the spin*. This resembles the scenario encountered for a mechanical oscillator [24], where the portion of the oscillator-environment coupling quadratic in the coordinate of the test oscillator yields, instead of a fluctuating force, a fluctuating contribution to its harmonic potential (*frequency-type* fluctuations). Finally, if $V_l(\mathbf{S})$ only comprises non-linear terms, such as those occurring in the magneto-elastic coupling mentioned ($V_l \propto \sum S_k S_\ell$), no field-type fluctuating terms emerge and only anisotropy-type fluctuations remain.

We remark in closing that, even for couplings linear in the spin variables, and hence for $\mathbf{b}_{\mathrm{fl}}(t)$ independent of $\mathbf{S}$, the occurrence of the vector *product* $\mathbf{S}\wedge\mathbf{b}_{\mathrm{fl}}$ in the dynamical equations entails that the fluctuating terms enter in a *multiplicative* way. This is at variance with the situation encountered in ordinary mechanical systems [24], where couplings linear in the system variables lead to additive fluctuations (see, *e.g.*, Eq. (20)), whereas multiplicative fluctuating terms only emerge for couplings non-linear in the system variables (*e.g.*, for $F_\alpha(p,q)=-c_\alpha q^2$). In the spin-dynamics case, in analogy with the results obtained for mechanical rigid rotators [25], the multiplicative character of the fluctuations is a consequence of the Poisson bracket relations $\{S_i,S_j\}=\gamma\sum_k\epsilon_{ijk}S_k$ for angular-momentum-type dynamical variables, which, even for F_α linear in $\mathbf{S}$, lead to non-constant $\{A,F_\alpha\}$ in equation (19). In our derivation, this can straightly be traced back by virtue of the Poisson-bracket formalism employed.

3.4 Statistical properties of the fluctuating terms

In order to determine the statistical properties of the fluctuating sources $f_l(t)$, one usually assumes that the environment was in thermodynamic equilibrium at the *initial* time (recall that no statistical assumption has been explicitly introduced until this point). This initial state is customarily chosen in two different ways.

Decoupled initial conditions

The environment variables are distributed at $t=t_0$ according to the Boltzmann law associated with the environment Hamiltonian alone

$$P_0(P(t_0),Q(t_0))=\mathcal{Z}^{-1}\exp[-\mathcal{H}_{\mathrm{E}}(t_0)/k_{\mathrm{B}}T], \tag{29}$$
$$\mathcal{H}_{\mathrm{E}}(t_0)=\sum_\alpha\frac{1}{2}\left[P_\alpha(t_0)^2+\omega_\alpha^2Q_\alpha(t_0)^2\right],$$

where $\mathcal{Z}$ is the corresponding partition function. One then has for the first two moments of the environmental variables $\langle Q_\alpha(t_0)\rangle=\langle P_\alpha(t_0)\rangle=0$, as well as $\langle Q_\alpha(t_0)Q_\beta(t_0)\rangle=\delta_{\alpha\beta}k_{\mathrm{B}}T/\omega_\alpha^2$, $\langle Q_\alpha(t_0)P_\beta(t_0)\rangle=0$, and $\langle P_\alpha(t_0)P_\beta(t_0)\rangle=\delta_{\alpha\beta}k_{\mathrm{B}}T$. Thus, the $f_l(t)$ (Eq. (24)) are Gaussian stochastic processes and the relevant averages over initial states of the environment (ensemble averages) are given by

$$\langle f_l(t)\rangle=0, \tag{30}$$
$$\langle f_l(t)f_{l'}(s)\rangle=k_{\mathrm{B}}T\,\mathcal{K}_{ll'}(t-s). \tag{31}$$

[4] Although we omit the symbol of scalar product, the action of a dyadic $\mathbf{AB}$ on a vector $\mathbf{C}$ is the standard one: $(\mathbf{AB})\mathbf{C}\equiv\mathbf{A}(\mathbf{B}\cdot\mathbf{C})$.

Equation (31) relates the statistical time correlation of the fluctuating terms $f_l(t)$ with the relaxation memory kernels $\mathcal{K}_{ll'}(\tau)$ occurring in the dynamical equations (*fluctuation-dissipation* relations). Short (long) correlation times of the fluctuating terms entail short-range (long-range) memory effects in the relaxation term, and *vice versa*.

Coupled initial conditions

The environment is assumed to be at $t = t_0$ in thermal equilibrium *in the presence of the system*, which is however taken as *fastened* in its initial state [20]. Therefore, the corresponding initial distribution of the environment variables is

$$P_0(P(t_0), Q(t_0)) = \mathcal{Z}^{-1} \exp[-\mathcal{H}_{\rm SE}(t_0)/k_{\rm B}T],$$
$$\mathcal{H}_{\rm SE}(t_0) = \sum_\alpha \frac{1}{2}\Big\{P_\alpha(t_0)^2 + \omega_\alpha^2\Big[Q_\alpha(t_0) + \frac{\varepsilon}{\omega_\alpha^2}F_\alpha(t_0)\Big]^2\Big\}.$$

In this case, the dropped terms depending on the initial state of the system $\mathcal{K}_\alpha(t-t_0)F_\alpha(t_0)$ (recall the remarks before Eq. (19)), which for $F_\alpha = \sum_l c_\alpha^l V_l$ lead to the terms $\sum_{l'}\mathcal{K}_{ll'}(t-t_0)V_{l'}(t_0)$, are not omitted but they are included into an alternative definition of the fluctuating sources, namely $\tilde{f}_l(t) = f_l(t) + \sum_{l'}\mathcal{K}_{ll'}(t-t_0)V_{l'}(t_0)$. The statistical properties of these terms, as determined by the above distribution, are given by expressions *identical* with equations (30, 31).

Note that the recourse to the "process" of initial fastening (and subsequent releasing) of the system by an external agency can, to a certain extent, be circumvented on noting that the concomitant initial statistical properties of the environment are consistent with the notion of a time-scale separation between the system and the surrounding medium, *i.e.*, the latter adjust rapidly to the state of the former [26].

Note finally that the differences associated with assuming decoupled initial conditions or the more physically motivated coupled initial conditions diminish as long as the weak-coupling condition is met. Anyhow, with both types of initial conditions one obtains the *same* Langevin equation after a time, measured from t_0, of the order of the width of the memory kernels $\mathcal{K}_{ll'}(\tau)$, which is the characteristic time for the "transient" terms $\sum_{l'}\mathcal{K}_{ll'}(t-t_0)V_{l'}(t_0)$ to die out.

4 Dynamical equations for couplings linear-plus-quadratic in the environment variables

In this Section we shall introduce interactions non-linear in the environment variables. This is mandatory when relaxation mechanisms involving more than one environmental normal mode (*e.g.*, multi-phonon or multi-photon processes) become relevant, as it occurs at sufficiently high temperatures. When such non-linear couplings are accounted for, one must resort to approximate methods to derive a reduced equation of motion for the spin. Here, we shall tackle the important weak-coupling case by a perturbational treatment.

4.1 The spin-environment Hamiltonian: couplings linear-plus-quadratic in the environment variables

Let us consider the following generalisation of the Hamiltonian (10)

$$\mathcal{H} = \mathcal{H}_{\rm S}(\mathbf{S}) + \sum_\alpha \frac{1}{2}\Big\{P_\alpha^2 + \omega_\alpha^2\Big[Q_\alpha + \frac{\varepsilon}{\omega_\alpha^2}F_\alpha(\mathbf{S})\Big]^2\Big\}$$
$$+ \frac{1}{2}\sum_{\alpha\beta}\Big[\varepsilon Q_\alpha Q_\beta F_{\alpha\beta}(\mathbf{S}) + k_{\rm B}T\frac{\varepsilon^2}{2\omega_\alpha^2\omega_\beta^2}F_{\alpha\beta}(\mathbf{S})^2\Big], \quad (32)$$

where couplings quadratic in the coordinates of the oscillators representing the environment have been included. The part of this interaction depending on the spin variables is introduced *via* the functions $F_{\alpha\beta}$. On the other hand, embodying the additional counter-terms (those proportional to $F_{\alpha\beta}^2$), the coupling-induced renormalisation of the energy of the spin is balanced to order ε^2. This renormalisation results to be explicitly dependent on the temperature for interactions non-linear in the environment variables (see below).

4.2 Dynamical equations: general case

Again, for the sake of simplicity and generality, we rewrite the Hamiltonian (32) as (*cf.* Eq. (11))

$$\mathcal{H} = \mathcal{H}_{\rm S}^{(\rm m)}(p,q) + \sum_\alpha \frac{1}{2}\left(P_\alpha^2 + \omega_\alpha^2 Q_\alpha^2\right)$$
$$+ \varepsilon\Big[\sum_\alpha Q_\alpha F_\alpha(p,q) + \frac{1}{2}\sum_{\alpha\beta}Q_\alpha Q_\beta F_{\alpha\beta}(p,q)\Big], \quad (33)$$

where $\mathcal{H}_{\rm S}^{(\rm m)}$ augments the system Hamiltonian $\mathcal{H}_{\rm S}$ by the counter-terms (*cf.* Eq. (12))

$$\mathcal{H}_{\rm S}^{(\rm m)} = \mathcal{H}_{\rm S} + \frac{\varepsilon^2}{2}\Big(\sum_\alpha \frac{F_\alpha^2}{\omega_\alpha^2} + k_{\rm B}T\sum_{\alpha\beta}\frac{F_{\alpha\beta}^2}{2\omega_\alpha^2\omega_\beta^2}\Big). \quad (34)$$

The ordinary formalism of the environment of *independent* oscillators [16–20] is not directly applicable when couplings non-linear in the environment variables are included. For instance, $Q_\alpha Q_\beta F_{\alpha\beta}$ brings about an indirect interaction among the oscillators so that these are no longer independent. Because a reduced equation of motion for the system cannot be easily derived for an arbitrary strength of the coupling, we shall perform a perturbational treatment in the weak-coupling regime by means of simple extensions of that developed by Cortés, West, and Lindenberg [22].

In Appendix A the corresponding calculations are detailed for a class of Hamiltonians with quite general non-linear couplings in both the system and the environment

variables. The results obtained permit the incorporation of relaxation mechanisms involving any number of normal modes into the dynamical equations of the system (under the weak-coupling condition mentioned). In the linear-plus-quadratic case considered here, we find the following *reduced* dynamical equation for any observable of the system $A(p,q)$ (*cf.* Eq. (19))

$$\begin{aligned}\frac{\mathrm{d}A}{\mathrm{d}t} &= \{A,\mathcal{H}_{\mathrm{S}}\} \\ &+ \sum_{\alpha}\{A,F_{\alpha}\}\Big[f_{\alpha}(t)+\int_{t_0}^{t}\mathrm{d}s\,\mathcal{K}_{\alpha}(t-s)\frac{\mathrm{d}F_{\alpha}}{\mathrm{d}t}(s)\Big] \\ &+ \sum_{\alpha\beta}\{A,F_{\alpha\beta}\}\Big[f_{\alpha\beta}(t)+\int_{t_0}^{t}\mathrm{d}s\,\mathcal{K}_{\alpha\beta}(t-s)\frac{\mathrm{d}F_{\alpha\beta}}{\mathrm{d}t}(s)\Big].\end{aligned} \tag{35}$$

Here, the fluctuating terms $f_{\alpha}(t)$ and the corresponding kernels $\mathcal{K}_{\alpha}(\tau)$ are again given by equations (18), whereas their counterparts for the quadratic portion of the coupling read

$$f_{\alpha\beta}(t) = \frac{\varepsilon}{2}Q^{\mathrm{h}}_{\alpha}(t)Q^{\mathrm{h}}_{\beta}(t), \tag{36}$$

$$\mathcal{K}_{\alpha\beta}(\tau) = \frac{\varepsilon^2}{2}\frac{k_{\mathrm{B}}T}{2\omega_{\alpha}^2\omega_{\beta}^2}\big\{\cos[(\omega_{\alpha}-\omega_{\beta})\tau]+\cos[(\omega_{\alpha}+\omega_{\beta})\tau]\big\}, \tag{37}$$

where the $Q^{\mathrm{h}}_{\alpha}(t)$ are the environmental proper modes (16).

The treatment leading to equation (35) can be summarised in terms of the driven-oscillator picture discussed in Section 3. One part of the driving from the system now depends on the state of the oscillators (*cf.* Eqs. (14) with (A.3)); this state is perturbatively replaced by the free evolution terms $Q^{\mathrm{h}}_{\alpha}(t)$, and the back-reaction on the system is averaged over initial states of the oscillators. This averaging yields the explicit dependence of the kernels $\mathcal{K}_{\alpha\beta}(\tau)$ on the temperature (and that of the associated counter-term $\frac{1}{2}\sum_{\alpha\beta}\mathcal{K}_{\alpha\beta}(0)F^2_{\alpha\beta}$).

4.3 Dynamical equations: the spin-dynamics case

In order to particularise the result (35) to the dynamics of a classical spin, the additional coupling functions $F_{\alpha\beta}$ are expressed as

$$F_{\alpha\beta}(\mathbf{S}) = \sum_{q} C^{q}_{\alpha\beta}V_q(\mathbf{S}),$$

where the general index q is analogous to that introduced in the linear case (Eq. (21)), the coefficients $C^{q}_{\alpha\beta}$ are the spin-environment coupling constants for the quadratic part of the interaction, and the terms $V_q(\mathbf{S})$ are certain functions of the spin variables. To illustrate, for the coupling of $\mathbf{S}$ to the lattice vibrations including quadratic terms in the strain tensor ("two-phonon" processes), the index q stands for *two* pairs of Cartesian indices and, *e.g.*, $V_q \to V_{ij,k\ell} = \sum_{mn} r_{ijk\ell,mn}S_mS_n$, where the $r_{ijk\ell,mn}$ are second-order magneto-elastic coefficients.

Then, on merely replacing $A(p,q)$ in equation (35) by the Cartesian components of the magnetic moment and using equation (9) to calculate the corresponding Poisson brackets, one arrives at the following reduced equation of motion for $\mathbf{S}$ (*cf.* Eq. (27))

$$\begin{aligned}\frac{\mathrm{d}\mathbf{S}}{\mathrm{d}t} &= \gamma\mathbf{S}\wedge[\mathbf{B}_{\mathrm{eff}}+\mathbf{b}_{\mathrm{fl}}(\mathbf{S},t)] \\ &- \gamma\mathbf{S}\wedge\Big\{\int_{t_0}^{t}\mathrm{d}s\,\gamma\left[\hat{\Gamma}^{(\mathrm{L})}+k_{\mathrm{B}}T\hat{\Gamma}^{(\mathrm{Q})}\right]_{(\mathbf{S};t,s)}(\mathbf{S}\wedge\mathbf{B}_{\mathrm{eff}})(s)\Big\}.\end{aligned} \tag{38}$$

Here, the fluctuating effective field generalises the expression (23) to

$$\mathbf{b}_{\mathrm{fl}}(\mathbf{S},t) = -\Big[\sum_{l}f_l(t)\frac{\partial V_l}{\partial\mathbf{S}}+\sum_{q}f_q(t)\frac{\partial V_q}{\partial\mathbf{S}}\Big], \tag{39}$$

where the $f_l(t)$ are given by equation (24) and the $f_q(t) = \sum_{\alpha\beta}C^{q}_{\alpha\beta}f_{\alpha\beta}(t)$ are additional fluctuating terms

$$f_q(t) = \frac{\varepsilon}{2}\sum_{\alpha\beta}C^{q}_{\alpha\beta}Q^{\mathrm{h}}_{\alpha}(t)Q^{\mathrm{h}}_{\beta}(t). \tag{40}$$

Concerning the relaxation terms, $\hat{\Gamma}^{(\mathrm{L})}$ is again given by equation (25), while the part of the relaxation tensor associated with the quadratic part of the coupling is given by

$$k_{\mathrm{B}}T\hat{\Gamma}^{(\mathrm{Q})}(\mathbf{S};t,s) = \sum_{q,q'}\mathcal{K}_{qq'}(t-s)\frac{\partial V_q}{\partial\mathbf{S}}(t)\frac{\partial V_{q'}}{\partial\mathbf{S}}(s)\ , \tag{41}$$

where $\mathcal{K}_{qq'}(\tau) = \sum_{\alpha\beta}C^{q}_{\alpha\beta}C^{q'}_{\alpha\beta}\mathcal{K}_{\alpha\beta}(\tau)$ or, explicitly

$$\begin{aligned}\mathcal{K}_{qq'}(\tau) = k_{\mathrm{B}}T\frac{\varepsilon^2}{2}\sum_{\alpha\beta}\frac{C^{q}_{\alpha\beta}C^{q'}_{\alpha\beta}}{2\omega_{\alpha}^2\omega_{\beta}^2}\Big\{&\cos[(\omega_{\alpha}-\omega_{\beta})\tau] \\ &+\cos[(\omega_{\alpha}+\omega_{\beta})\tau]\Big\}.\end{aligned} \tag{42}$$

Note that equation (38) is of Landau–Lifshitz type since the derivative $\mathrm{d}\mathbf{S}/\mathrm{d}t$ that would appear in the relaxation term has been replaced, within the approximation used ($\varepsilon \ll 1$), by its free evolution (conservative) part $\mathrm{d}\mathbf{S}/\mathrm{d}t \simeq \gamma\mathbf{S}\wedge\mathbf{B}_{\mathrm{eff}}$ (see the remarks after Eq. (A.10)). Notice also that we have explicitly shown the temperature dependence of the relaxation term, which is caused by the quadratic portion of the coupling.

For weak spin-environment interactions being arbitrary functions of the spin variables and embodying linear-plus-quadratic terms in the coordinates of the oscillators representing the environment, equation (38) is the desired dynamical equation for $\mathbf{S}$. Note that, in quantum-mechanical language, the term comprising $\cos(\omega_{\alpha}\tau)$ in the memory kernel (26) would correspond to a relaxation mechanism (transition) *via* the emission or absorption of a vibrational quantum of energy $\hbar\omega_{\alpha}$. Similarly, $\cos[(\omega_{\alpha}+\omega_{\beta})\tau]$ in the kernel (42) would be

associated with relaxation mechanisms with either the emission or the absorption of two vibrational quanta, whereas $\cos[(\omega_\alpha - \omega_\beta)\tau]$ would correspond to the absorption of one quantum and the emission of a second one (scattering processes).

Finally, the definition (28) of the fluctuating part of the energy of the spin can be generalised to

$$\mathcal{H}_{\rm fl}(\mathbf{S},t) = \sum_l f_l(t) V_l(\mathbf{S}) + \sum_q f_q(t) V_q(\mathbf{S}), \tag{43}$$

whence $\mathbf{b}_{\rm fl}(\mathbf{S},t) = -\partial\mathcal{H}_{\rm fl}/\partial\mathbf{S}$, in correspondence with $\mathbf{B}_{\rm eff} = -\partial\mathcal{H}_{\rm S}/\partial\mathbf{S}$. Remarks similar to those made after equation (28) concerning the structure of $\mathcal{H}_{\rm fl}(\mathbf{S},t)$ for linear and non-linear (in the spin variables) spin-environment interactions, and the corresponding nature of the fluctuations (field- and/or anisotropy-type), are in order here.

4.4 Statistical properties of the fluctuating terms

The statistical properties of the $f_l(t)$ (Eq. (24)), as determined by the initial distribution (29) of the environment variables (*decoupled initial conditions*), are given by equations (30, 31), whereas those of $f_q(t)$ (Eq. (40)) and their cross-correlations read

$$\langle f_q(t)\rangle = 0, \tag{44}$$

$$\langle f_l(t) f_q(s)\rangle = 0, \tag{45}$$

$$\langle f_q(t) f_{q'}(s)\rangle = k_{\rm B}T\,\mathcal{K}_{qq'}(t-s). \tag{46}$$

In order to obtain equation (44), *i.e.*, centred fluctuating sources, as well as equation (46), we have assumed that $c^q_{\alpha\beta} \equiv 0$ for $\alpha = \beta$. If such a restriction is not applied, one has, *e.g.*, $\langle f_q(t)\rangle \neq 0$, which represents a non-vanishing average forcing of the spin. Note however that to retain those terms must cause no harm since, when the double sums over oscillators $\sum_{\alpha\beta}(\cdot)$ are transformed into double integrals for (quasi-) continuous distributions of oscillators, such $\alpha = \beta$ terms constitute a zero-measure set, whose contribution can therefore be ignored.

The Gaussian property of the $f_q(t)$ can then be established on the basis that these terms are sums over a large number of contributions $c^q_{\alpha\beta}\,Q^{\rm h}_\alpha(t)Q^{\rm h}_\beta(t)$ (Eq. (40)) with mean zero and equivalent statistical properties (Central Limit Theorem). On the other hand, equation (46) expresses that the fluctuating sources $f_q(t)$ and the relaxation memory kernels $\mathcal{K}_{qq'}(\tau)$ associated with the quadratic portion of the coupling also obey fluctuation-dissipation relations. In addition, the zero cross-correlations of equation (45) are also fluctuation-dissipation relations involving null kernels (see Eq. (A.11)).

We finally remark that on assuming *coupled initial conditions*, without modifying the definitions of the fluctuating terms, the corrections to equations (30, 44), and to the relations (31, 45, 46), are of order ε^2 and ε^3, respectively; these corrections are of order higher than the terms retained in the weak-coupling approximation used (see Appendix A).

5 Markovian regime and phenomenological equations

In this section we shall study the form that the dynamical equations derived exhibit in the absence of memory effects. Then, we shall consider some specific spin-environment interactions, formally obtaining the Langevin equations discussed in the Introduction.

5.1 Markovian regime

The Markovian regime arises when the relaxation memory kernels are sharply peaked about $\tau = 0$, the remainder terms in the memory integrals change slowly enough in the relevant range, and the kernels enclose a finite non-zero algebraic area. Under these conditions, one can substitute the kernels by Dirac deltas and no memory effects occur.

5.1.1 Langevin equations

Let us assume that the memory kernel (26) can be replaced by a Dirac delta

$$\mathcal{K}_{ll'}(\tau) = 2(\lambda_{ll'}/\gamma S)\delta(\tau), \tag{47}$$

where the $\lambda_{ll'}$ are damping coefficients related with the strength and characteristics of the coupling (see below). Then, on using $\int_0^\infty {\rm d}\tau\,\delta(\tau)h(\tau) = h(0)/2$, equation (22) reduces to the *Gilbert-type* equation (*cf.* Eq. (1))

$$\frac{{\rm d}\mathbf{S}}{{\rm d}t} = \gamma\mathbf{S}\wedge\left[\mathbf{B}_{\rm eff} + \mathbf{b}_{\rm fl}(\mathbf{S},t) - (\gamma S)^{-1}\hat{\Lambda}^{(\rm L)}\frac{{\rm d}\mathbf{S}}{{\rm d}t}\right], \tag{48}$$

where $\hat{\Lambda}^{(\rm L)}(\mathbf{S})$ is a dimensionless second-rank tensor with elements

$$\Lambda^{(\rm L)}_{ij}(\mathbf{S}) = \sum_{l,l'}\lambda_{ll'}\frac{\partial V_l}{\partial S_i}\frac{\partial V_{l'}}{\partial S_j}\,. \tag{49}$$

Likewise, on inserting equation (47) in the weak-coupling equation (27) we get the following *Landau–Lifshitz-type* equation (*cf.* Eq. (2))

$$\frac{{\rm d}\mathbf{S}}{{\rm d}t} = \gamma\mathbf{S}\wedge[\mathbf{B}_{\rm eff} + \mathbf{b}_{\rm fl}(\mathbf{S},t)] - \frac{\gamma}{S}\mathbf{S}\wedge\hat{\Lambda}^{(\rm L)}\left(\mathbf{S}\wedge\mathbf{B}_{\rm eff}\right). \tag{50}$$

Note that the tensor $\hat{\Lambda}^{(\rm L)}$, the precursor of which is the tensor $\hat{\Gamma}^{(\rm L)}$ (Eq. (25)) in the memory integrals, is symmetrical since $\lambda_{ll'}$ is so (see Eq. (62) below).

On the other hand, the Markovian case of the dynamical equation for couplings linear-plus-quadratic in the environment coordinates (Eq. (38)) arises when the additional memory kernel can also be replaced by a Dirac delta

$$\mathcal{K}_{qq'}(\tau) = 2(\lambda_{qq'}k_{\rm B}T/\gamma S)\delta(\tau), \tag{51}$$

where we have explicitly shown the temperature dependence of the kernel (42). Then, equation (38) reduces to the *Landau–Lifshitz-type* equation

$$\frac{\mathrm{d}\mathbf{S}}{\mathrm{d}t} = \gamma\mathbf{S}\wedge[\mathbf{B}_{\mathrm{eff}} + \mathbf{b}_{\mathrm{fl}}(\mathbf{S},t)] - \frac{\gamma}{S}\mathbf{S}\wedge\hat{\Lambda}\,(\mathbf{S}\wedge\mathbf{B}_{\mathrm{eff}})\,, \quad (52)$$

where $\mathbf{b}_{\mathrm{fl}}(\mathbf{S},t)$ is now given by equation (39). In the above equation the relaxation tensor

$$\hat{\Lambda} = \hat{\Lambda}^{(\mathrm{L})} + k_{\mathrm{B}}T\,\hat{\Lambda}^{(\mathrm{Q})}, \quad (53)$$

where

$$\Lambda^{(\mathrm{Q})}_{ij}(\mathbf{S}) = \sum_{q,q'}\lambda_{qq'}\frac{\partial V_q}{\partial S_i}\frac{\partial V_{q'}}{\partial S_j}, \quad (54)$$

introduces an explicit dependence on the temperature rooted in the quadratic portion of the coupling.

For a general form of the spin-environment interaction, due to the occurrence of the tensors $\hat{\Lambda}^{(\mathrm{L})}$ and $\hat{\Lambda}^{(\mathrm{Q})}$ in the above equations, the structure of the relaxation terms deviates from the forms proposed by Gilbert and Landau and Lifshitz. Such deviations can be produced by couplings non-linear in $\mathbf{S}$, for which $\hat{\Lambda}^{(\mathrm{L})}_{ij}$ and $\hat{\Lambda}^{(\mathrm{Q})}_{ij}$ depend in general on the spin variables, but they also emerge when these tensors are independent of $\mathbf{S}$ (*e.g.*, for couplings linear in $\mathbf{S}$) but they are not proportional to δ_{ij}. The relaxation is then anisotropic because, *e.g.*, $-\mathbf{S}\wedge\hat{\Lambda}(\mathbf{S}\wedge\mathbf{B}_{\mathrm{eff}})$ no longer points from $\mathbf{S}$ to the direction of $\mathbf{B}_{\mathrm{eff}}$.

Finally, owing to the fluctuation-dissipation relations (31, 46), the fluctuating terms corresponding to the Markovian memory kernels are naturally delta-correlated in time, so we have the following statistical properties

$$\langle f_l(t)\rangle = 0, \quad (55)$$

$$\langle f_l(t)f_{l'}(s)\rangle = \frac{2\lambda_{ll'}}{\gamma S}\,k_{\mathrm{B}}T\delta(t-s), \quad (56)$$

and

$$\langle f_q(t)\rangle = 0, \quad (57)$$

$$\langle f_l(t)f_q(s)\rangle = 0, \quad (58)$$

$$\langle f_q(t)f_{q'}(s)\rangle = \frac{2(\lambda_{qq'}k_{\mathrm{B}}T)}{\gamma S}\,k_{\mathrm{B}}T\delta(t-s). \quad (59)$$

Notice the double occurrence of $k_{\mathrm{B}}T$ in the last relation.

5.1.2 Damping coefficients

On accounting for equations (47, 51), one can calculate the damping coefficients from the area enclosed by the memory kernels, namely

$$\frac{\lambda_{ll'}}{\gamma S} = \int_0^\infty \mathrm{d}\tau\,\mathcal{K}_{ll'}(\tau), \quad (60)$$

$$\frac{\lambda_{qq'}}{\gamma S}k_{\mathrm{B}}T = \int_0^\infty \mathrm{d}\tau\,\mathcal{K}_{qq'}(\tau). \quad (61)$$

These areas must be: (i) *finite* and (ii) *different from zero*, for the Markovian approximation to work.

On the other hand, as it could be difficult to find the kernels exactly in some cases, it is convenient to have alternative means for calculating only the areas required. Thus, on inserting the definitions of the kernels (26, 42) into the above integrals and using $\int_0^\infty \mathrm{d}\tau\,\cos(\omega\tau) = \pi\delta(\omega)$, we arrive at the following expressions for the damping coefficients in terms of the distribution of the normal modes and spin-environment coupling constants

$$\frac{\lambda_{ll'}}{\gamma S} = \pi\varepsilon^2\sum_\alpha \frac{C^l_\alpha C^{l'}_\alpha}{\omega^2_\alpha}\delta(\omega_\alpha), \quad (62)$$

$$\frac{\lambda_{qq'}}{\gamma S} = \pi\frac{\varepsilon^2}{2}\sum_{\alpha\beta}\frac{C^q_{\alpha\beta}C^{q'}_{\alpha\beta}}{2\omega^2_\alpha\omega^2_\beta} \times\big[\delta(\omega_\alpha-\omega_\beta)+\delta(\omega_\alpha+\omega_\beta)\big]. \quad (63)$$

Note that the Dirac deltas in these formulae make sense under integral signs for (quasi-) continuous distributions of environmental modes.

5.1.3 Fokker–Planck equations

The Markovian Langevin equations can be employed to construct the corresponding Fokker–Planck equations governing the time evolution of the non-equilibrium probability distribution of spin orientations $P(\mathbf{S},t)$. The details of the derivations of the Fokker–Planck equations associated with the Landau–Lifshitz-type equations (50, 52) are given in Appendix B. For the latter we get

$$\frac{\partial P}{\partial t} = -\frac{\partial}{\partial\mathbf{S}}\cdot\left\{\gamma\mathbf{S}\wedge\mathbf{B}_{\mathrm{eff}} - \frac{\gamma}{S}\mathbf{S}\wedge\hat{\Lambda}\left[\mathbf{S}\wedge\left(\mathbf{B}_{\mathrm{eff}} - k_{\mathrm{B}}T\frac{\partial}{\partial\mathbf{S}}\right)\right]\right\}P, \quad (64)$$

where $\frac{\partial}{\partial\mathbf{S}}\cdot$ stands for the divergence operator ($\frac{\partial}{\partial\mathbf{S}}\cdot\mathbf{J} = \sum_i \frac{\partial J_i}{\partial S_i}$). Similarly, the Fokker–Planck equation corresponding to equation (50) is analogous to equation (64) with $\hat{\Lambda} = \hat{\Lambda}^{(\mathrm{L})} + k_{\mathrm{B}}T\,\hat{\Lambda}^{(\mathrm{Q})}$ replaced by $\hat{\Lambda}^{(\mathrm{L})}$.

Stationary solution

By using $\mathbf{B}_{\mathrm{eff}} = -\partial\mathcal{H}_{\mathrm{S}}/\partial\mathbf{S}$ and $\frac{\partial}{\partial\mathbf{S}}\cdot\big(\mathbf{S}\wedge\mathbf{B}_{\mathrm{eff}}P_0\big) = 0$, one readily demonstrates that the Boltzmann distribution, $P_0(\mathbf{S}) \propto \exp[-\mathcal{H}_{\mathrm{S}}(\mathbf{S})/k_{\mathrm{B}}T]$, is a stationary solution of these Fokker–Planck equations. This entails that under external stationary conditions $P(\mathbf{S},t) \overset{t\to\infty}{\longrightarrow} P_0(\mathbf{S})$, that is, the spin eventually reaches the thermal equilibrium distribution of orientations. Note that this is a consequence of the formalism employed, instead of a constrain imposed separately, as it is usually done in the (semi-) phenomenological approaches.

Note nevertheless that we have only proved the thermal equilibration for equations (50, 52), *i.e.*, in the weak-coupling regime. In this connection, it is to be recalled that, inasmuch as the spin-environment coupling Hamiltonians themselves are commonly obtained *via* perturbation theory (so they are "small" in some sense), the study of the arbitrary-coupling case of such Hamiltonians is mainly of an academic interest.

5.2 Brown–Kubo–Hashitsume model

Let us first consider the case of couplings linear in the environment coordinates. Then, if $V_l(\mathbf{S})$ is linear in $\mathbf{S}$, both the relaxation tensor $\hat{\Lambda}^{(\mathrm{L})}$ and the fluctuating field $\mathbf{b}_{\mathrm{fl}}$ are independent of $\mathbf{S}$ (see Eqs. (49, 23)). From the statistical properties (55, 56) of the fluctuating sources $f_l(t)$, one then gets

$$\langle b_{\mathrm{fl},i}(t)\rangle = 0, \quad \langle b_{\mathrm{fl},i}(t) b_{\mathrm{fl},j}(s)\rangle = \frac{2\Lambda_{ij}^{(\mathrm{L})}}{\gamma S} k_{\mathrm{B}} T \delta(t-s),$$

where the last result establishes the relation between the structure of the correlations among the components of $\mathbf{b}_{\mathrm{fl}}(t)$ and the form of the relaxation tensor $\hat{\Lambda}^{(\mathrm{L})}$ [5]. The corresponding result by Jayannavar [15] comprised an uncorrelated $\mathbf{b}_{\mathrm{fl}}(t)$ (a diagonal $\Lambda_{ij}^{(\mathrm{L})}$ in our formulation) due to special bilinear interaction that he considered (recall the discussion after Eq. (10)).

On the other hand, if the spin-environment interaction yields uncorrelated *and* isotropic fluctuations ($\Lambda_{ij}^{(\mathrm{L})} = \lambda\delta_{ij}$), the Langevin equations (48, 50) reduce, respectively, to the stochastic Gilbert (Eq. (1)) and Landau–Lifshitz (Eq. (2)) equations. Thus, the phenomenological Brown–Kubo–Hashitsume model [1,2] is formally obtained.

Note finally that these results also hold when couplings quadratic in the environment variables are included (Eq. (52)), with the difference that the relaxation terms (effective damping coefficients) are then explicitly dependent on the temperature.

5.3 Garanin, Ishchenko, and Panina model

We shall now show that the weak-coupling Landau–Lifshitz-type equations (50, 52), formally reduce to the Langevin equation (4) of Garanin, Ishchenko, and Panina when the spin-environment interaction includes up to quadratic terms in the *spin variables*. In this case, the coupling functions $V_l(\mathbf{S})$ and $V_q(\mathbf{S})$ can be written as

$$V_l(\mathbf{S}) = \sum_i v_{l,i} S_i + \frac{1}{2}\sum_{ij} w_{l,ij} S_i S_j, \tag{65}$$

$$V_q(\mathbf{S}) = \sum_i v_{q,i} S_i + \frac{1}{2}\sum_{ij} w_{q,ij} S_i S_j, \tag{66}$$

[5] Note that for $\mathbf{b}_{\mathrm{fl}}(\mathbf{S},t)$ depending on $\mathbf{S}$, one cannot merely employ equations (55, 56) to derive the statistical properties of $\mathbf{b}_{\mathrm{fl}}(\mathbf{S},t)$, since $\mathbf{S}(t)$ and $f_l(t)$ are not independent.

where the constants $v_{l,i}$, $w_{l,ij}$, $v_{q,i}$, and $w_{q,ij}$ incorporate the symmetry of the interaction. Then, the fluctuating effective field (39) can be cast into the form (*cf.* Eq. (4))

$$\mathbf{b}_{\mathrm{fl}}(\mathbf{S},t) = \mathbf{b}(t) + \hat{k}(t)\mathbf{S},$$

with the following expressions for the fluctuating sources $\mathbf{b}(t)$ and $\hat{k}(t)$ in terms of the coupling constants

$$b_i(t) = -\Big[\sum_l f_l(t) v_{l,i} + \sum_q f_q(t) v_{q,i}\Big],$$

$$k_{ij}(t) = -\Big[\sum_l f_l(t) w_{l,ij} + \sum_q f_q(t) w_{q,ij}\Big].$$

As $\mathbf{b}(t)$ does not depend on $\mathbf{S}$, it can be interpreted as a fluctuating *ordinary* field (the $\mathbf{b}_{\mathrm{fl}}(t)$ of the previous subsection). The fluctuations of $\hat{k}(t)$, however, do not enter in this way, since they occur *via* $\sum_j k_{ij}(t) S_j$, but they produce fluctuations of the magnetic-anisotropy potential of the spin, both of the direction of the anisotropy axes and of the magnitudes of the anisotropy constants. This is neatly perceived on considering that the fluctuating part of the energy of the spin (43), which gives $\mathbf{b}_{\mathrm{fl}}(\mathbf{S},t) = -\partial\mathcal{H}_{\mathrm{fl}}/\partial\mathbf{S}$, takes in this case the form

$$\mathcal{H}_{\mathrm{fl}}(\mathbf{S},t) = -\mathbf{S}\cdot\mathbf{b}(t) - \frac{1}{2}\mathbf{S}\cdot\hat{k}(t)\mathbf{S}.$$

In the Markovian regime, the auto- and cross-correlations of $\mathbf{b}(t)$ and $\hat{k}(t)$ can be obtained by dint of equations (56, 58, 59). Such correlations can be cast into the form proposed by Garanin, Ishchenko, and Panina (Eq. (3))

$$\langle b_i(t) b_j(s)\rangle = \frac{2\lambda_{ij}}{\gamma S}\, k_{\mathrm{B}} T \delta(t-s),$$

$$\langle b_i(t) k_{jk}(s)\rangle = \frac{2\lambda_{i,jk}}{\gamma S}\, k_{\mathrm{B}} T \delta(t-s),$$

$$\langle k_{ik}(t) k_{j\ell}(s)\rangle = \frac{2\lambda_{ik,j\ell}}{\gamma S}\, k_{\mathrm{B}} T \delta(t-s),$$

with the following expressions for the correlation coefficients

$$\lambda_{ij} = \sum_{l,l'} \lambda_{ll'} v_{l,i} v_{l',j} + k_{\mathrm{B}} T \sum_{q,q'} \lambda_{qq'} v_{q,i} v_{q',j},$$

$$\lambda_{i,jk} = \sum_{l,l'} \lambda_{ll'} v_{l,i} w_{l',jk} + k_{\mathrm{B}} T \sum_{q,q'} \lambda_{qq'} v_{q,i} w_{q',jk},$$

$$\lambda_{ik,j\ell} = \sum_{l,l'} \lambda_{ll'} w_{l,ik} w_{l',j\ell} + k_{\mathrm{B}} T \sum_{q,q'} \lambda_{qq'} w_{q,ik} w_{q',j\ell}.$$

Concerning the relaxation term, the tensor $\hat{\Lambda} = \hat{\Lambda}^{(\mathrm{L})} + k_{\mathrm{B}} T\, \hat{\Lambda}^{(\mathrm{Q})}$ (Eq. (53)) associated with the coupling functions (65, 66), is given by

$$\Lambda_{ij} = \sum_{l,l'} \lambda_{ll'} \Big(v_{l,i} + \sum_k w_{l,ik} S_k\Big)\Big(v_{l',j} + \sum_\ell w_{l',j\ell} S_\ell\Big)$$
$$+ k_{\mathrm{B}} T \sum_{q,q'} \lambda_{qq'} \Big(v_{q,i} + \sum_k w_{q,ik} S_k\Big)\Big(v_{q',j} + \sum_\ell w_{q',j\ell} S_\ell\Big).$$

However, this expression can be written in terms of the *above* correlation coefficients as

$$\Lambda_{ij} = \lambda_{ij} + \sum_k (\lambda_{i,jk} + \lambda_{j,ik}) S_k + \sum_{k\ell} \lambda_{ik,j\ell} S_k S_\ell,$$

which is identical with the relation (5) between the tensor $\hat{G}$ in equation (4) and the correlation coefficients in equation (3).

Therefore, we find that when the spin-environment coupling includes up to quadratic terms in the spin variables, *the structures of the fluctuating effective field* $\mathbf{b}_{\rm fl}(\mathbf{S},t)$ *and of the relaxation term* $\mathbf{R} = \frac{\gamma}{S}\mathbf{S} \wedge \hat{\Lambda}(\mathbf{S} \wedge \mathbf{B}_{\rm eff})$ *in the Landau–Lifshitz-type equation (52), as well as the relation between them, are identical with those of the corresponding terms in the Langevin equation (4) of Garanin, Ishchenko, and Panina.* Naturally, the Fokker–Planck equation (64), then reduces to that constructed by them [12].

6 Summary and discussion

Starting from a Hamiltonian description, reduced equations of motion for a classical spin interacting with the surrounding medium, have been derived. The oscillator-bath representation of the environment has been employed and couplings depending arbitrarily on the spin variables and being linear or linear-plus-quadratic in the environment variables, have been considered (higher-order non-linearities in the environment variables can be incorporated by the treatment of Appendix A).

The dynamical equations obtained (Eqs. (22, 27, 38)) have the structure of generalised Langevin equations with fluctuating terms $\gamma\mathbf{S} \wedge \mathbf{b}_{\rm fl}(\mathbf{S},t)$ (associated with the modulation by the proper modes of the environment), and relaxation terms (corresponding to the back-reaction on the spin of its previous action on the surrounding medium) obeying fluctuation-dissipation relations (Eqs. (31, 45, 46)). For couplings non-linear in the spin variables, the fluctuating effective field $\mathbf{b}_{\rm fl}(\mathbf{S},t)$ (Eqs. (23, 39)) depends in general on $\mathbf{S}$, so that it can incorporate fluctuations of the magnetic anisotropy of the spin.

In the Markovian approach, we have obtained generalisations of the phenomenological Langevin equations with the general form $\mathrm{d}\mathbf{S}/\mathrm{d}t = \gamma\mathbf{S} \wedge [\mathbf{B}_{\rm eff} + \mathbf{b}_{\rm fl}(\mathbf{S},t)] - \mathbf{R}$, where for couplings linear *in the environment coordinates* the relaxation term reads $\mathbf{R} = \frac{1}{S}\mathbf{S} \wedge \hat{\Lambda}^{(\mathrm{L})}(\mathrm{d}\mathbf{S}/\mathrm{d}t)$ (Gilbert-type; Eq. (48)) and $\mathbf{R} = \frac{\gamma}{S}\mathbf{S} \wedge \hat{\Lambda}^{(\mathrm{L})}(\mathbf{S} \wedge \mathbf{B}_{\rm eff})$ for weak coupling (Landau–Lifshitz-type; Eq. (50)). When interactions quadratic in the environment variables are accounted for, the tensor $\hat{\Lambda}^{(\mathrm{L})}$ in the weak-coupling relaxation term results to be augmented to $\hat{\Lambda} = \hat{\Lambda}^{(\mathrm{L})} + k_{\rm B}T\,\hat{\Lambda}^{(\mathrm{Q})}$, incorporating an explicit dependence of the effective damping coefficients on the temperature (Eq. (52)). Finally, when the spin-environment interaction consists of linear and quadratic terms *in the spin variables*, the weak-coupling Landau–Lifshitz-type equations obtained and the corresponding Fokker–Planck equations, formally reduce to those of Garanin, Ishchenko, and Panina.

Note however that the presented derivation of the equations of Garanin, Ishchenko, and Panina and, similarly, the previous derivations [13–15,21] of the equations occurring in the Brown–Kubo–Hashitsume stochastic model, are formal in the sense that one must still investigate specific realizations of the spin-plus-environment whole system, and then prove that the assumptions employed (mainly that of Markovian behavior) are at least approximately met. A paradigmatic case where this does not occur is that of a magneto-elastic coupling of the spin to the lattice vibrations (in two or three dimensions) *linear* in the corresponding normal modes [27,28]. The associated memory kernel crosses zero, changes it sign, and tends to zero from negative values as $\tau \to \infty$, *enclosing a zero algebraic area*. One then gets identically zero $\lambda_{ll'}$ by equation (60) and hence a zero tensor $\hat{\Lambda}^{(\mathrm{L})}$ by equation (49). Therefore, on replacing such a kernel by a Dirac delta, one looses the relaxational effects associated with the portion of the coupling *linear* in the environment variables ("one-phonon" processes), which are dominant at sufficiently low temperatures.

On the other hand, we have considered the classical regime of the environment and the spin. A classical description of the environment is adequate, *e.g.*, for the coupling to low-frequency ($\hbar\omega_\alpha/k_{\rm B}T \ll 1$) normal modes, while, for instance, the magnetic moment of a nanometric particle ($S \sim 10^3$–$10^5\,\mu_{\rm B}$) behaves, except for very low temperatures, as a classical spin. In addition, the equations derived might also serve as a limit description of the semi-classical dynamics of molecular magnetic clusters with high spin ($\gtrsim 10$) in their ground state. Finally, the Hamiltonian formalism employed could be suitable to attempt the generalisation of some of the results presented to the quantum case.

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Appendix A: Equations of motion for couplings non-linear in the environment variables

In this appendix we shall derive a reduced equation of motion for any dynamical variable $A(p,q)$ whose time evolution is determined by the Hamiltonian (33), by means of a perturbational expansion in the coupling parameter ε. Nevertheless, we shall first study the weak-coupling dynamics associated with a larger class of Hamiltonians of the form

$$\mathcal{H} = \mathcal{H}_{\rm S}^{(\mathrm{m})} + \sum_\alpha \frac{1}{2}\left(P_\alpha^2 + \omega_\alpha^2 Q_\alpha^2\right) + \varepsilon \sum_N \mathcal{B}^N(Q) F_N(p,q), \tag{A.1}$$

where the coupling terms $\mathcal{B}^N(Q)$ are *arbitrary* functions of the environment coordinates Q and N stands for a general index, which can run, *e.g.*, over single oscillator indices, pairs, triplets, etc. $(\alpha, \alpha\beta, \alpha\beta\gamma, \dots)$. On the other hand, the modified system Hamiltonian $\mathcal{H}_{\rm S}^{(\rm m)}$ augments the system Hamiltonian $\mathcal{H}_{\rm S}$ by appropriate counter-terms, which will be determined below.

We shall first derive the reduced dynamical equations associated with the Hamiltonian (A.1), so that one could incorporate the effects of relaxation mechanisms involving any number of environmental normal modes. This will be done by a perturbational treatment, which is an extension of that carried out by Cortés, West, and Lindenberg [22] to deal with a system-environment coupling *linear* in the system coordinate (the case $F_N(p,q) \propto q$ of the Hamiltonian (A.1)), but otherwise arbitrary in the environment coordinates (Brun [29] also treated rather general non-bilinear interactions by perturbation theory). Eventually, we shall particularise the results obtained to the Hamiltonian (33), which is recovered when:

1. N only runs over single oscillator indices α and pairs $\alpha\beta$.
2. The corresponding coupling terms are $\mathcal{B}^{\alpha}(Q) = Q_\alpha$ and $\mathcal{B}^{\alpha\beta}(Q) = \frac{1}{2}Q_\alpha Q_\beta$.

The coupled dynamical equations for $A(p,q)$ and the environment variables associated with the Hamiltonian (A.1) are (*cf.* Eqs. (13, 14))

$$\frac{{\rm d}A}{{\rm d}t} = \big\{A, \mathcal{H}_{\rm S}^{(\rm m)}\big\} + \varepsilon \sum_N \mathcal{B}^N(Q)\big\{A, F_N\big\}, \tag{A.2}$$

$$\frac{{\rm d}Q_\alpha}{{\rm d}t} = P_\alpha, \quad \frac{{\rm d}P_\alpha}{{\rm d}t} = -\omega_\alpha^2 Q_\alpha - \varepsilon \sum_N \mathcal{B}_\alpha^N(Q) F_N, \tag{A.3}$$

where $\mathcal{B}_\alpha^N = \partial\mathcal{B}^N/\partial Q_\alpha$. Equations (A.3) can *formally* be integrated, yielding an equation akin to equation (15) with $F_\alpha(s) \to \sum_N \mathcal{B}_\alpha^N[Q(s)]F_N(s)$, where $F_N(s) = F_N[p(s), q(s)]$. On integrating by parts in such an equation one gets (*cf.* Eq. (17))

$$Q_\alpha(t) = Q_\alpha^{\rm h}(t) - \varepsilon \sum_N \big[D_\alpha^N(Q;t,s)F_N(s)\big]_{s=t_0}^{s=t} + \varepsilon \int_{t_0}^t {\rm d}s \sum_N D_\alpha^N(Q;t,s)\frac{{\rm d}F_N}{{\rm d}t}(s), \tag{A.4}$$

where $Q_\alpha^{\rm h}(t)$ is the familiar solution (16) for the free oscillator and we have introduced the indefinite integral

$$D_\alpha^N(Q;t,s) = \frac{1}{\omega_\alpha}\int^s {\rm d}s' \, \sin[\omega_\alpha(t-s')]\mathcal{B}_\alpha^N[Q(s')]. \tag{A.5}$$

Recall that on writing $Q_\alpha(t)$ in the form (A.4) by an integration by parts, permits the separation of the Hamiltonian (renormalisation) and relaxational terms. However, equation (A.4) gives $Q_\alpha(t)$ in implicit form, since this also appears on the right-hand side *via* $\mathcal{B}_\alpha^N(Q)$ (Eq. (A.4) is an explicit solution only in the linear $\mathcal{B}^N(Q)$ case of the Hamiltonian (11)).

For weak system-environment interactions, we shall solve equation (A.4) for $Q_\alpha(t)$ perturbatively in ε. However, as pointed out in reference [22], in order to get eventually a thermodynamically consistent description, the expansion cannot be uniform in ε. If one keeps fluctuating terms up to order ε^k, the relaxation terms must be retained up to order ε^{2k}, in order to obtain proper fluctuation-dissipation relations (see, *e.g.*, Eqs. (24, 26, 31)).

The ε-expansion of $Q_\alpha(t)$ reads

$$Q_\alpha(t) = Q_\alpha^{\rm h}(t) + \varepsilon\, \delta Q_\alpha(t) + \dots,$$

where $\varepsilon\,\delta Q_\alpha(t)$ is given by the second plus third terms on the right-hand side of equation (A.4) when $Q^{\rm h}$ (the zeroth-order term) is substituted for Q in $D_\alpha^N(Q;t,s)$ (that is, we iterate Eq. (A.4) into itself). The corresponding expansion of the coupling functions is given by

$$\varepsilon\mathcal{B}^N(Q) = \varepsilon\mathcal{B}^N(Q^{\rm h}) + \varepsilon^2 \sum_\alpha \mathcal{B}_\alpha^N(Q^{\rm h})\delta Q_\alpha + \dots, \tag{A.6}$$

which enters into equation (A.2). The term

$$f_N(t) = \varepsilon\mathcal{B}^N[Q^{\rm h}(t)], \tag{A.7}$$

per analogy with $f_\alpha(t) = \varepsilon Q_\alpha^{\rm h}(t)$ (Eq. (18)), is interpreted as the lowest order fluctuation. Following the program of reference [22], we shall retain fluctuations only to this order. On the other hand, in order to ensure $\langle f_N(t)\rangle = 0$, where the angular brackets denote average over initial states of the oscillators, one could assume that, *e.g.*, at least one coordinate enters in $\mathcal{B}^N(Q)$ an odd number of times [22]. Nevertheless, as discussed after equations (44, 45, 46), such a restriction is not needed when the frequency spectrum of the oscillators is sufficiently dense.

Concerning the back-reaction part on introducing

$$\mathcal{K}^{N,M}(t,s) = \varepsilon^2\Big\langle \sum_\alpha \mathcal{B}_\alpha^N[Q^{\rm h}(t)]D_\alpha^M(Q^{\rm h};t,s)\Big\rangle, \tag{A.8}$$

$$\delta\mathcal{K}^{N,M}(t,s) = \varepsilon^2 \sum_\alpha \mathcal{B}_\alpha^N[Q^{\rm h}(t)]D_\alpha^M(Q^{\rm h};t,s) - \mathcal{K}^{N,M}(t,s),$$

the second term in the expansion (A.6) can be decomposed as

$$\begin{aligned}\varepsilon^2 \sum_\alpha \mathcal{B}_\alpha^N(Q^{\rm h})\delta Q_\alpha &= -\Big[\sum_M \big[\mathcal{K}^{N,M}(t,s) + \delta\mathcal{K}^{N,M}(t,s)\big]\, F_M(s)\Big]_{s=t_0}^{s=t} \\ &\quad + \int_{t_0}^t {\rm d}s \sum_M \big[\mathcal{K}^{N,M}(t,s) + \delta\mathcal{K}^{N,M}(t,s)\big]\,\frac{{\rm d}F_M}{{\rm d}t}(s).\end{aligned}$$

Each kernel $\mathcal{K}^{N,M}$ gives a different type of contribution whereas the contribution of $\delta\mathcal{K}^{N,M}$ can be interpreted as fluctuations around the former [22]. As these fluctuations are of order higher (ε^2) than the fluctuations that we are retaining in the present treatment,

the terms $\delta\mathcal{K}^{N,M}$ will henceforth be omitted. On the other hand, the terms $\sum_M \mathcal{K}^{N,M}(t,t_0)F_M(t_0)$ in $\varepsilon^2\sum_\alpha \mathcal{B}^N_\alpha \delta Q_\alpha$ will also be ignored as they are the generalisation of those terms that give a transient in the response (see Sect. 3; recall however that they could be incorporated into an alternative definition of the fluctuating sources but, as they are of order ε^2, they would anyhow be ignored). Finally, the parallel terms $-\sum_M \mathcal{K}^{N,M}(t,t)F_M(t)$ give the Hamiltonian contributions. In order to prove this, note first that, since $\mathcal{K}^{N,M}(t,s)$ comprises equilibrium averages (Eq. (A.8)), it depends on $(t-s)$ and, hence, $\mathcal{K}^{N,M}(t,t)$ is independent of t. By the same reasoning one can demonstrate the symmetry property $\mathcal{K}^{N,M} = \mathcal{K}^{M,N}$ (we shall anyway verify explicitly these two results for the Hamiltonian (33)). Then, by using the product rule of the Poisson bracket, $\{A,BC\} = \{A,B\}C + \{A,C\}B$, one finds that the contribution originating from $-\sum_M \mathcal{K}^{N,M}(t,t)F_M(t)$ in the equation for $A(p,q)$, is given by $-\sum_{NM}\mathcal{K}^{N,M}(0)\{A,F_N\}F_M = \{A,-\frac{1}{2}\sum_{NM}\mathcal{K}^{N,M}(0)F_NF_M\}$, which is indeed derivable from a (time-independent) Hamiltonian. This term is associated with the coupling-induced renormalisation of the energy of the system and is balanced by the counter-terms incorporated into $\mathcal{H}_{\rm S}^{(\rm m)}$, now explicitly identified as (*cf.* Eq. (12))

$$\mathcal{H}_{\rm S}^{(\rm m)} = \mathcal{H}_{\rm S} + \frac{1}{2}\sum_{NM}\mathcal{K}^{N,M}(0)F_NF_M. \tag{A.9}$$

On collecting the terms whose retention has hitherto been argued and introducing them into equation (A.2), one finally gets the (approximate) reduced equation of motion for any dynamical variable $A(p,q)$ (*cf.* Eq. (19))

$$\frac{{\rm d}A}{{\rm d}t} = \{A,\mathcal{H}_{\rm S}\} + \sum_N \{A,F_N\}\left[f_N(t) + \int_{t_0}^t {\rm d}s \sum_M \mathcal{K}^{N,M}(t-s)\frac{{\rm d}F_M}{{\rm d}t}(s)\right]. \tag{A.10}$$

In addition, within the approximation used (fluctuating and relaxation terms to order ε and ε^2, respectively), one can substitute ${\rm d}F_M/{\rm d}t$ in the memory integral by its conservative part ${\rm d}F_M/{\rm d}t \simeq \{F_M,\mathcal{H}_{\rm S}\}$. On the other hand, one can establish fluctuation-dissipation relations by means of arguments parallel to those presented in reference [22].

To conclude, we shall particularise these results to the linear-plus-quadratic couplings of the Hamiltonian (33). This is recovered when N runs over single oscillator indices α, with $\mathcal{B}^\alpha = Q_\alpha$, and pairs $\alpha\beta$, with $\mathcal{B}^{\alpha\beta} = \frac{1}{2}Q_\alpha Q_\beta$. Then, the fluctuating terms $f_N(t) = \varepsilon\mathcal{B}^N[Q^{\rm h}(t)]$ (Eq. (A.7)) are given by $f_\alpha(t) = \varepsilon Q^{\rm h}_\alpha(t)$ (Eq. (18)) and $f_{\alpha\beta}(t) = \frac{\varepsilon}{2}Q^{\rm h}_\alpha(t)Q^{\rm h}_\beta(t)$ (Eq. (36)). On the other hand, by inserting the derivatives $\mathcal{B}^\alpha_\gamma = \partial\mathcal{B}^\alpha/\partial Q_\gamma = \delta_{\alpha\gamma}$ and $\mathcal{B}^{\alpha\beta}_\gamma = \partial\mathcal{B}^{\alpha\beta}/\partial Q_\gamma = \frac{1}{2}(\delta_{\alpha\gamma}Q_\beta + \delta_{\beta\gamma}Q_\alpha)$ in equation (A.5), the functions $D^N_\gamma(Q;t,s)$ emerge in the form ($N = \alpha,\ \alpha\beta$)

$$D^\alpha_\gamma(Q;t,s) = \frac{\delta_{\alpha\gamma}}{\omega_\alpha^2}\cos[\omega_\alpha(t-s)],$$
$$D^{\alpha\beta}_\gamma(Q;t,s) = \frac{1}{\omega_\gamma}\int^s {\rm d}s'\,\sin[\omega_\gamma(t-s')] \times\frac{1}{2}[\delta_{\alpha\gamma}Q_\beta(s') + \delta_{\beta\gamma}Q_\alpha(s')].$$

Therefore, on taking the averages in equation (A.8) with respect to the distribution (29) (decoupled initial conditions) by using $\langle Q^{\rm h}_\alpha(t)\rangle = 0$ and $\langle Q^{\rm h}_\alpha(t)Q^{\rm h}_\beta(s)\rangle = \delta_{\alpha\beta}(k_{\rm B}T/\omega_\alpha^2)\cos[\omega_\alpha(t-s)]$, we get for the kernels $\mathcal{K}^{N,M}$

$$\mathcal{K}^{\alpha,\beta}(\tau) = \delta_{\alpha\beta}\frac{\varepsilon^2}{\omega_\alpha^2}\cos(\omega_\alpha\tau),$$
$$\mathcal{K}^{\alpha,\beta\gamma}(\tau) = \mathcal{K}^{\alpha\beta,\gamma}(\tau) = 0, \tag{A.11}$$
$$\mathcal{K}^{\alpha\beta,\gamma\delta}(\tau) = \frac{1}{2}(\delta_{\alpha\gamma}\delta_{\beta\delta} + \delta_{\alpha\delta}\delta_{\beta\gamma})\frac{\varepsilon^2}{2}\frac{k_{\rm B}T}{2\omega_\alpha^2\omega_\beta^2} \times\left\{\cos[(\omega_\alpha-\omega_\beta)\tau] + \cos[(\omega_\alpha+\omega_\beta)\tau]\right\}.$$

These kernels satisfy the properties mentioned above: they depend on $\tau = t-s$ and are symmetrical with respect to the indices separated by commas, which correspond to the general indices N, M.

On introducing all these results in equation (A.10), the resulting dynamical equation for $A(p,q)$ is given by equation (35), where, for the sake of simplicity, we have introduced the kernels $\mathcal{K}_\alpha(\tau)$ and $\mathcal{K}_{\alpha\beta}(\tau)$, which are defined in terms of the above kernels by

$$\mathcal{K}^{\alpha,\beta}(\tau) = \delta_{\alpha\beta}\mathcal{K}_\alpha(\tau),$$
$$\mathcal{K}^{\alpha\beta,\gamma\delta}(\tau) = \frac{1}{2}(\delta_{\alpha\gamma}\delta_{\beta\delta} + \delta_{\alpha\delta}\delta_{\beta\gamma})\mathcal{K}_{\alpha\beta}(\tau).$$

Besides, on explicitly writing the counter-term of equation (A.9) in this linear-plus-quadratic case, one arrives at equation (34).

Note finally that, owing to $\mathcal{B}^\alpha_\gamma(Q^{\rm h})D^\beta_\gamma(Q^{\rm h};t,s)$ does not depend on $Q^{\rm h}$, the kernel $\mathcal{K}_\alpha(\tau)$ is not affected by the averaging procedure, whereas this renders $\mathcal{K}_{\alpha\beta}(\tau)$ explicitly dependent on the temperature. In this connection, we remark that the modifications of this last kernel, obtained when one assumes coupled initial conditions, begin at order ε^3.

Appendix B: Derivation of the Fokker–Planck equations

In this appendix we shall derive the Fokker–Planck equations associated with the Landau–Lifshitz-type equations (50, 52). On examining the statistical properties (56, 59), one realizes that Langevin equations where the noise terms are not statistically independent need to be considered.

Let us consider the general system of Langevin equations

$$\frac{\mathrm{d}y_i}{\mathrm{d}t} = A_i(y,t) + \sum_k B_{ik}(y,t)L_k(t), \qquad \text{(B.1)}$$

where $i = 1,\ldots,n$, $y = (y_1,\ldots,y_n)$, and k runs over a given set of indices. The Langevin sources $L_k(t)$ are Gaussian stochastic processes satisfying

$$\langle L_k(t)\rangle = 0, \qquad \langle L_k(t)L_\ell(s)\rangle = 2D_{k\ell}\delta(t-s), \qquad \text{(B.2)}$$

where the constant matrix $D_{k\ell}$ accounts for the possible correlations among the $L_k(t)$.

The time evolution of $P(y,t)$, the non-equilibrium probability distribution for y at time t, is given by the Fokker–Planck equation

$$\frac{\partial P}{\partial t} = -\sum_i \frac{\partial}{\partial y_i}\left[\left(A_i + \sum_{jk\ell} B_{j\ell}D_{\ell k}\frac{\partial B_{ik}}{\partial y_j}\right)P\right] + \sum_{ij}\frac{\partial^2}{\partial y_i \partial y_j}\left[\left(\sum_{k\ell} B_{ik}D_{k\ell}B_{j\ell}\right)P\right],$$

where the Stratonovich stochastic calculus [30] has been used to treat the (in general) multiplicative fluctuating terms in the Langevin equations (B.1). On taking the y_j-derivatives of the second term on the right-hand side, one alternatively gets the Fokker–Planck equation in the form of a continuity equation for the probability distribution, namely

$$\frac{\partial P}{\partial t} = -\sum_i \frac{\partial}{\partial y_i}\left\{\left[A_i - \sum_{k\ell} B_{ik}D_{k\ell}\left(\sum_j \frac{\partial B_{j\ell}}{\partial y_j}\right) - \sum_{jk\ell} B_{ik}D_{k\ell}B_{j\ell}\frac{\partial}{\partial y_j}\right]P\right\}. \qquad \text{(B.3)}$$

The Stratonovich calculus has been chosen on the basis of two points. On the one hand, the results in the context of this calculus coincide with those formally obtained in the limit of small but finite width of the autocorrelation functions of the fluctuating sources, which is precisely the way one takes the Markovian limit. On the other hand, the generalised Langevin equations obtained in this article are indeed dynamical equations to which a stochastic interpretation has been added. Besides, owing to $0 = \mathbf{S}\cdot(\mathrm{d}\mathbf{S}/\mathrm{d}t) = \frac{1}{2}\mathrm{d}(\mathbf{S}^2)/\mathrm{d}t$, the time evolution that they determine rigorously conserve the length of the spin. Nevertheless, when passing from ordinary to stochastic differential equations, specific rules of calculus (integration and differentiation) are required. In the context of the Stratonovich calculus, such rules are formally identical with those of the ordinary calculus, so that equations (48, 50, 52) also yield $d(\mathbf{S}^2)/\mathrm{d}t = 0$. However, when using the rules of differentiation of the Itô calculus, one finds that those equations *do not* conserve the length of $\mathbf{S}$.

Now, on considering the Landau–Lifshitz-type equation (50), supplemented by the statistical properties (55, 56), the substitutions

$$(k,\ell) = (l,l'), \qquad (y_1,y_2,y_3) = (S_x,S_y,S_z),$$
$$L_l(t) = f_l(t), \qquad D_{ll'} = \frac{\lambda_{ll'}}{\gamma S}k_{\mathrm{B}}T,$$
$$A_i = \left[\gamma\mathbf{S}\wedge\mathbf{B}_{\mathrm{eff}} - \frac{\gamma}{S}\mathbf{S}\wedge\hat{\Lambda}^{(\mathrm{L})}(\mathbf{S}\wedge\mathbf{B}_{\mathrm{eff}})\right]_i,$$
$$B_{il} = -\gamma\sum_{rs}\epsilon_{irs}S_r\frac{\partial V_l}{\partial S_s},$$

cast them into the form of the general system of Langevin equations (B.1) supplemented by equations (B.2). Therefore, on using

$$\frac{\partial B_{il}}{\partial S_j} = -\gamma\left(\sum_s \epsilon_{ijs}\frac{\partial V_l}{\partial S_s} + \sum_{rs}\epsilon_{irs}S_r\frac{\partial^2 V_l}{\partial S_j\partial S_s}\right),$$

one finds that $\sum_j \partial B_{jl}/\partial S_j \equiv 0$, $\forall l$, due to $\epsilon_{jjs} = 0$ and the vanishing of the contraction of symmetrical tensors with antisymmetrical tensors. Consequently, the second term on the right-hand side of the general Fokker–Planck equation (B.3) vanishes identically in this case. For the third term, by repeated use of $(\mathbf{J}\wedge\mathbf{J}')_i = \sum_{rs}\epsilon_{irs}J_rJ'_s$ and recalling the definition (49), we get

$$-\sum_{jll'} B_{il}D_{ll'}B_{jl'}\frac{\partial P}{\partial S_j} = \frac{\gamma}{S}k_{\mathrm{B}}T\left[\mathbf{S}\wedge\hat{\Lambda}^{(\mathrm{L})}\left(\mathbf{S}\wedge\frac{\partial P}{\partial\mathbf{S}}\right)\right]_i.$$

On introducing these results into equation (B.3) one eventually arrives at a Fokker–Planck equation analogous to (64) with $\hat{\Lambda}$ replaced by $\hat{\Lambda}^{(\mathrm{L})}$. In addition, by means of similar considerations and allowing the index in the Langevin sources $L_k(t)$ to run also over the indices q, the Landau–Lifshitz-type equation (52) leads to the Fokker–Planck equation (64).

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