Derivation of the basic system of kinetic equations governing superparamagnetic relaxation by the use of the adjoint Fokker-Planck operator

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The basic equations governing the kinetics of superparamagnets (and dielectric lattices in the Debye approximation) are derived by taking advantage of the properties of the adjoint Fokker-Planck operator. The equations obtained, which duly reduce to those derived by direct averaging of the corresponding stochastic Landau-Lifshitz-type equations, are expressed in terms of the coefficients determining the action of the angular momentum operator and the spin variables on the spherical harmonics. The structure of the equations so obtained directly reflects that of the adjoint Fokker-Planck operator, which permits to trace back easily the origin of the different contributions (precession and relaxation). This makes the method specially suitable for generalization to more elaborate descriptions of rotational relaxation.

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The stochastic Landau-Lifshitz equation for classical spins and its associated Fokker-Planck equation (as introduced by Brown¹ and by Kubo and Hashitsume²) play an important role in condensed-matter physics, describing a variety of magnetic and also electric systems. Indeed, in the limit of zero precession these equations *formally* reduce to the equations in the Debye theory of noninertial dielectric relaxation, which has applications in permanent dipole molecules, nematic liquid crystals, and relaxor ferroelectrics. In magnetism, they describe the $T \neq 0$ spin dynamics of classical XY and Heisenberg models, thin films, and superparamagnets (nanoscale solids or clusters whose net spin $S \sim 10^2 - 10^5$ rotates thermally activated in the anisotropy potential).

In the calculation of the quantities of interest (e.g., dynamical susceptibilities or relaxation times) a variety of analytical and numerical methods are employed. A rigorous and powerful approach consists of casting the equations into an infinite hierarchy of *linear* kinetic equations for the averages of a complete set of appropriate functions (e.g., the spherical harmonics in rotational relaxation), which can be tackled by matrix or continued-fraction methods. Nevertheless, the problem of finding the coefficients of such a system for specific Hamiltonians is difficult, and it has not been until recently that a general solution has been found in the context of the Brown-Kubo-Hashitsume model.³

In this paper we present an alternative solution for the more modest problem of determining the kinetic equations for reasonably simple Hamiltonians. The method takes advantage of the properties of the adjoint Fokker-Planck operator. The structure of the equations obtained closely parallels the structure of this operator, permitting to trace back readily the origin of the different contributions (precession and relaxation) from the generally complex final expressions. This makes the method suitable for application to more elaborate descriptions of rotational relaxation, and we outline its application to the interesting generalization of the superparamagnetic model that also incorporates anisotropy-type fluctuations, as those generated by the modulation of the crystal field by the lattice vibrations.⁴

Let us begin with some general remarks about the equation for the average of an arbitrary quantity. The Fokker-Planck equation governing the time evolution of the nonequilibrium probability distribution of the system variables can be written generically as

$$\partial P / \partial t = \mathcal{L}_{\rm FP} P, \tag{1}$$

where \mathcal{L}_{FP} is the corresponding Fokker-Planck operator (which is linear but in general not self-adjoint). The average of a quantity *f* is given by $\langle f \rangle = \int d\Omega P f$, where $d\Omega$ is the differential element of phase space $(d\Omega \propto d^2 \vec{s})$ in rotational relaxation). When *f* is not explicitly time dependent, the equation governing the dynamics of $\langle f \rangle$ follows from

$$\frac{d}{dt}\langle f\rangle = \int d\Gamma \frac{\partial P}{\partial t} f = \int d\Gamma (\mathcal{L}_{\rm FP} P) f = \int d\Gamma P \mathcal{L}_{\rm FP}^{\dagger} f,$$

where we have passed the action of \mathcal{L}_{FP} from *P* to *f* by introducing the adjoint operator $\mathcal{L}_{\text{FP}}^{\dagger}$. Therefore, recalling the definition of average, we can write

$$d\langle f \rangle / dt = \langle \mathcal{L}_{\text{FP}}^{\dagger} f \rangle. \tag{2}$$

This result shows that the calculation of the dynamical equation for the average of any quantity can be reduced to the determination of the action of the adjoint Fokker-Planck operator on it.

The basic Fokker-Planck equation governing superparamagnetic relaxation can be written as

$$2\tau_D \frac{\partial P}{\partial t} = -\frac{\partial}{\partial \vec{s}} \cdot \left\{ \frac{1}{\lambda} \vec{s} \times \vec{B} - \vec{s} \times \left[\vec{s} \times \left(\vec{B} - \frac{\partial}{\partial \vec{s}} \right) \right] \right\} P,$$

where λ is the Landau-Lifshitz relaxation parameter, $\vec{B} = -(\partial \mathcal{H}/\partial \vec{s})/k_{\rm B}T$ is the effective field (in $k_{\rm B}$ units; $||\vec{s}|| = 1$), $\tau_D \propto T^{-1}$ is the characteristic time of free rotational diffusion ($\vec{B} = \vec{0}$), and the nabla operator ($\partial/\partial \vec{s}$) = $\sum_k \hat{x}_k (\partial/\partial s_k)$ acts on everything written on its right.

Note that taking formally the limit $\lambda \rightarrow \infty$ one gets the Fokker-Planck equation describing noninertial Debye relaxation (without translational degrees of freedom). Therefore,

although we shall use the language of magnetism, all the considerations that follow will also be applicable to the congeneric dielectric problem.

Let us now write the Fokker-Planck equation in terms of the angular momentum operator

$$\vec{\mathcal{J}} = -i\vec{s} \times (\partial/\partial\vec{s}), \qquad (3)$$

which is self-adjoint and directly related with the generator of infinitesimal rotations (Ref. 5, Sec. 26). Specifically, in a rotation of \vec{s} around $\delta \vec{\psi}$ a function $f(\vec{s})$ is transformed into $f(\vec{s} + \delta \vec{s}) \approx [1 + i \delta \vec{\psi} \cdot \vec{\mathcal{J}}] f(\vec{s})$. Besides, taking into account the permutability of the mixed product $\vec{a} \cdot (\vec{b} \times \vec{c})$ (with some care, as operators are involved), using $2\vec{s} = \partial(\vec{s}^2)/\partial \vec{s}$ and since the curl of a gradient is zero, the action of the divergence operator on any vector of the form $\vec{s} \times \vec{a}$ can be written as $(\partial/\partial \vec{s}) \cdot (\vec{s} \times \vec{a}) = -i \vec{\mathcal{J}} \cdot \vec{a}$. Consequently, we can cast the Fokker-Planck equation in the form

$$2\tau_D \partial P/\partial t = i\vec{\mathcal{J}} \cdot [(1/\lambda)\vec{B} - (\vec{s} \times \vec{B}) + i\vec{\mathcal{J}}]P, \qquad (4)$$

where the term acting on *P* on the right-hand side gives the Fokker-Planck operator in terms of $\mathcal{J}[cf. Eq. (1)]$. To get the adjoint operator we simply use $(\mathcal{MN})^{\dagger} = \mathcal{N}^{\dagger}\mathcal{M}^{\dagger}$, and since \mathcal{J} is self-adjoint, we get

$$\mathcal{L}_{\rm FP}^{\dagger} = -(i/\lambda) \left[\vec{B} - \lambda(\vec{s} \times \vec{B}) \right] \cdot \vec{\mathcal{J}} - \vec{\mathcal{J}}^2.$$

This result permits a transparent interpretation: (i) the term $\propto \vec{J}^2$ is the only term left when $\vec{B} = \vec{0}$, so it governs the free diffusion of the spins. (ii) As \vec{J} is the generator of infinitesimal rotations, the term $\vec{B} \cdot \vec{J}$ accounts for the precession of \vec{s} around the field \vec{B} , while (iii) $-\lambda(\vec{s} \times \vec{B}) \cdot \vec{J}$ generates the relaxational rotation of \vec{s} towards \vec{B} .

A final manipulation will cast $\mathcal{L}_{\text{FP}}^{\dagger}$ into a form more convenient for the subsequent calculation. Using again the permutation properties of the mixed product we have $-(\vec{s} \times \vec{B}) \cdot \vec{\mathcal{J}} = \vec{B} \cdot (\vec{s} \times \vec{\mathcal{J}})$, whence we get the key result

$$\mathcal{L}_{\rm FP}^{\dagger} = -(i/\lambda)\vec{B} \cdot [\vec{\mathcal{J}} + \lambda(\vec{s} \times \vec{\mathcal{J}})] - \vec{\mathcal{J}}^2, \tag{5}$$

where all the fields are gathered on the left, and the remaining spin variables and the operators on the right. Then, one can apply standard results for the action of $\vec{\mathcal{J}}$ and \vec{s} to get the basic equations for the averages.

An important fact in rotational relaxation (and one of the major sources of mathematical difficulties) is that the equations for the averages are *not* closed. The reason is that the underlying Landau-Lifshitz-type relaxation term $-\vec{s} \times (\vec{s} \times \vec{B})$ is nonlinear in the spin variables and hence contributes terms of the form $\langle s_i s_j \rangle$ to the equation for $\langle s_i \rangle$. Then, an additional equation for $\langle s_i s_j \rangle$ is required to close the system, but that equation involves $\langle s_i s_j s_k \rangle$, and so on.

To handle such an infinite hierarchy of equations a very convenient approach is to introduce the equations for the averages of the spherical harmonics (recall that $||\vec{s}|| = 1$),

$$X_l^m = e^{im\varphi} P_l^m(s_z), \quad |m| \le l, \tag{6}$$

where φ is the azimuth of *s* and the P_l^m are the associated Legendre functions. The X_l^m are related with the normalized spherical harmonics Y_l^m (Condon-Shortley phase) by

$$Y_l^m = n_{l,m} X_l^m, \quad n_{l,m} = (-1)^m \sqrt{\frac{2l+1}{4\pi} \frac{(l-m)!}{(l+m)!}}.$$
 (7)

All the averages can be written in terms of these quantities: $\langle s_z \rangle = \langle X_1^0 \rangle$, $\langle s_x \rangle + i \langle s_y \rangle = \langle X_1^1 \rangle$, $\langle 3s_z^2 - 1 \rangle / 2 = \langle X_2^0 \rangle$, etc. Besides, the use of the recurrence relations satisfied by the P_l^m simplifies considerably the manipulation of the formulas.

In order to get the equations for the averages of the spherical harmonics, one needs to determine the action of the angular momentum operators and the spin variables on them. The action of $\vec{\mathcal{J}}$ can be written compactly as

$$\mathcal{J}_{z}X_{l}^{m} = \alpha_{l,m}^{z}X_{l}^{m}, \quad \mathcal{J}_{\pm}X_{l}^{m} = \alpha_{l,m}^{\pm}X_{l}^{m\pm 1}, \quad (8)$$

where the $\mathcal{J}_{\pm} = \mathcal{J}_x \pm i \mathcal{J}_y$ are the corresponding circular components (ladder operators). When the Y_l^m are used, one has $\alpha_{l,m}^z = m$ and $\alpha_{l,m}^{\pm} = \sqrt{(l \pm m)(l \pm m + 1)}$, while for the X_l^m they can be obtained directly from these and the relation (7), and read $\alpha_{l,m}^+ = -1$, $\alpha_{l,m}^z = m$, $\alpha_{l,m}^- = -(l+m)(l-m+1)$.

Concerning the "action" of \vec{s} , recall that the product of the spin variables with the X_l^m can in turn be expanded in spherical harmonics by using the recurrence relations for the P_l^m . This yields expressions linear in the spherical harmonics, which is essential for the eventual application of standard numerical techniques. Thus, the spin variables can formally be considered as "operators" on the X_l^m and their action be expressed as $(s_{\pm} = s_x \pm i s_y)$

$$s_{z}X_{l}^{m} = \beta_{l-1,m}^{z}X_{l-1}^{m} + \beta_{l+1,m}^{z}X_{l+1}^{m}, \qquad (9)$$

$$s_{\pm}X_{l}^{m} = \beta_{l-1,m}^{\pm}X_{l-1}^{m\pm 1} + \beta_{l+1,m}^{\pm}X_{l+1}^{m\pm 1}.$$
 (10)

When the Y_l^m are used, the coefficients β are readily obtained, for instance, from the corresponding expressions of Ref. 3. The coefficients for the X_l^m can be extracted from the known explicit versions of Eqs. (9) and (10) (see, for example, Ref. 6, Appendix A), and can be written as

$$\beta_{l\pm 1,m}^{+} = \pm \frac{1}{(2l+1)}, \quad \beta_{l\pm 1,m}^{z} = \frac{(l+\frac{1}{2})\mp (m-\frac{1}{2})}{(2l+1)},$$
$$\beta_{l\pm 1,m}^{-} = \mp \frac{[(l+\frac{1}{2})\mp (m-\frac{1}{2})][(l+\frac{1}{2})\mp (m-\frac{3}{2})]}{(2l+1)}.$$

Nevertheless, leaving the coefficients α and β unspecified has, as we shall see below, a number of advantages.

Finally, let us write the action of $\vec{s} \times \vec{\mathcal{J}}$ on the spherical harmonics in terms of the coefficients α and β . This is easily done by expressing the vector product in terms of circular components $(\vec{s} \times \vec{\mathcal{J}})_z = i/2(s_+\mathcal{J}_- - s_-\mathcal{J}_+)$ and $(\vec{s} \times \vec{\mathcal{J}})_{\pm} = \pm i(s_z\mathcal{J}_{\pm} - s_{\pm}\mathcal{J}_z)$,

$$-i(\vec{s} \times \vec{\mathcal{J}})_{z} X_{l}^{m} = \frac{1}{2} (\alpha_{l,m}^{-} \beta_{l-1,m-1}^{+} - \alpha_{l,m}^{+} \beta_{l-1,m+1}^{-}) X_{l-1}^{m} + \frac{1}{2} (\alpha_{l,m}^{-} \beta_{l+1,m-1}^{+} - \alpha_{l,m}^{+} \beta_{l+1,m+1}^{-}) X_{l+1}^{m}, -i(\vec{s} \times \vec{\mathcal{J}})_{\pm} X_{l}^{m} = \pm (\alpha_{l,m}^{\pm} \beta_{l-1,m\pm1}^{z} - \alpha_{l,m}^{z} \beta_{l-1,m}^{\pm}) X_{l-1}^{m\pm1} \pm (\alpha_{l,m}^{\pm} \beta_{l+1,m\pm1}^{z} - \alpha_{l,m}^{z} \beta_{l+1,m}^{\pm}) X_{l+1}^{m\pm1}.$$

From these results we can readily derive the required action of the adjoint Fokker-Planck operator $\mathcal{L}_{FP}^{\dagger}$ on the spherical harmonics. Let us pass $\tilde{\mathcal{J}}^2$ to the left-hand side of Eq. (5) and write explicitly the scalar product on the right-hand side in terms of circular components as $\vec{a} \cdot \vec{b} = a_z b_z + \frac{1}{2}(a_+b_- + a_-b_+)$; this yields

$$(\mathcal{L}_{\rm FP}^{\dagger}+\vec{\mathcal{J}}^2)X_l^m = -\frac{1}{2}B_+[(i/\lambda)\mathcal{J}_-+i(\vec{s}\times\vec{\mathcal{J}})_-]X_l^m$$
$$-B_z[(i/\lambda)\mathcal{J}_z+i(\vec{s}\times\vec{\mathcal{J}})_z]X_l^m$$
$$-\frac{1}{2}B_-[(i/\lambda)\mathcal{J}_++i(\vec{s}\times\vec{\mathcal{J}})_+]X_l^m.$$

Introducing the calculated actions on the spherical harmonics, we finally obtain

$$\mathcal{L}_{FP}^{\dagger}X_{l}^{m} + l(l+1)X_{l}^{m} = \frac{1}{2}B_{+}[(\alpha_{l,m}^{z}\beta_{l-1,m}^{-} - \alpha_{l,m}^{-}\beta_{l-1,m-1}^{z})X_{l-1}^{m-1} - (i/\lambda)\alpha_{l,m}^{-}X_{l}^{m-1} + (\alpha_{l,m}^{z}\beta_{l+1,m}^{-} - \alpha_{l,m}^{-}\beta_{l+1,m-1}^{z})X_{l+1}^{m-1}] \\ + B_{z}[\frac{1}{2}(\alpha_{l,m}^{-}\beta_{l-1,m-1}^{+} - \alpha_{l,m}^{+}\beta_{l-1,m+1}^{-})X_{l-1}^{m} - (i/\lambda)\alpha_{l,m}^{z}X_{l}^{m} + \frac{1}{2}(\alpha_{l,m}^{-}\beta_{l+1,m-1}^{+} - \alpha_{l,m}^{+}\beta_{l+1,m+1}^{-})X_{l+1}^{m}] \\ + \frac{1}{2}B_{-}[(\alpha_{l,m}^{+}\beta_{l-1,m+1}^{z} - \alpha_{l,m}^{z}\beta_{l-1,m}^{+})X_{l-1}^{m+1} - (i/\lambda)\alpha_{l,m}^{+}X_{l}^{m+1} + (\alpha_{l,m}^{+}\beta_{l+1,m+1}^{z} - \alpha_{l,m}^{z}\beta_{l+1,m}^{+})X_{l+1}^{m+1}].$$

$$(11)$$

When the effective field $\vec{B} \propto (\partial \mathcal{H}/\partial \vec{s})$ does depend on \vec{s} one can again use Eqs. (9) and (10) to derive via Eq. (2) an expression for $d\langle X_l^m \rangle/dt$ linear in the $\langle X_{l'}^{m'} \rangle$ with constant (independent of \vec{s}) coefficients. In the general case this, although straightforward, can be very laborious and it would be advantageous to use the general expression for the coefficients derived by Kalmykov and Titov.³ However, for reasonably simple Hamiltonians (say uniaxial and biaxial) the effective field is linear in \vec{s} and only one further application of \vec{s} on the X_l^m is required.

Since for negative *m* the P_l^m in X_l^m [Eq. (6)] are implicitly defined through a Rodriges formula, the recurrence relations on which Eqs. (9) and (10) are based, and in turn Eq. (11), are valid for all *m* (Ref. 7, Sec. 12.6). Therefore, in our derivation we have not needed to examine the cases m>0and m<0 separately. Anyway, we can directly verify Eq. (11) by introducing the explicit expressions for the coefficients α and β . This yields (see the appendix) an equation for X_l^m in complete agreement with the result obtained by direct averaging of the stochastic Landau-Lifshitz-type equations⁸ and the corresponding equation for Y_l^m .³

It is not evident, however, how those equations for X_l^m and Y_l^m can be extended to more general descriptions of rotational relaxation. The form (11), on the other hand, directly reflects the structure of the adjoint Fokker-Planck operator (5). Indeed, the gyromagnetic terms (those proportional to i/λ) simply reflect the action of \vec{J} , while the relaxation terms explicitly preserve the form of the Landau-Lifshitz "damping term" $\vec{s} \times \vec{J}$ through the coefficients α (angular momentum action) and β (spin-variables action). Thus, B_+ is accompanied by $(\alpha^z \beta^- - \alpha^- \beta^z)$, the component B_z by $\frac{1}{2}(\alpha^- \beta^+ - \alpha^+ \beta^-)$, and B_- by $(\alpha^+ \beta^z - \alpha^z \beta^+)$, which reflect the vector-product structure of the relaxation term. This suggests that the adjoint operator method could also be used in cases in which the underlying Langevin equation might be more complex but one can still derive a reasonably simple expression for $\mathcal{L}_{FP}^{\dagger}$ in terms of \vec{s} and $\vec{\mathcal{J}}$.

To illustrate, let us briefly consider the important generalization of the Brown-Kubo-Hashitsume model developed by Garanin *et al.*⁴ to incorporate fluctuations of the magnetic anisotropy of the spin, such as those created by the phonon modulation of the spin-orbit coupling. In this case the Langevin equation is much more involved as it includes fluctuating fields and fluctuating anisotropy constants (fluctuations that can in addition be correlated). The associated Fokker-Planck equation, however, simplifies to

$$2\tau_D \frac{\partial P}{\partial t} = -\frac{\partial}{\partial \vec{s}} \cdot \left\{ \frac{1}{\lambda} \vec{s} \times \vec{B} - \vec{s} \times \hat{\Gamma} \left[\vec{s} \times \left(\vec{B} - \frac{\partial}{\partial \vec{s}} \right) \right] \right\} P,$$

where the *symmetric* matrix $\hat{\Gamma}$ summarizes the effect of the mentioned mechanisms $(\lambda \Gamma_{ij} = \eta_{ij} + \Sigma_k (\eta_{i,jk} + \eta_{j,ik}) s_k + \Sigma_{kl} \eta_{ik,jl} s_k s_l$, where η are the correlation coefficients of the fluctuating terms).

Manipulations analogous to those leading to Eq. (5) yield in this case

$$\mathcal{L}_{\rm FP}^{\dagger} = -(i/\lambda)\vec{B} \cdot [\vec{\mathcal{J}} + \lambda(\vec{s} \times \hat{\Gamma} \vec{\mathcal{J}})] - \vec{\mathcal{J}}\hat{\Gamma}\vec{\mathcal{J}}, \qquad (12)$$

where the symmetry of $\hat{\Gamma}$ has been taken into account when taking the adjoint. Therefore, we see that the key result (5) for $\mathcal{L}_{FP}^{\dagger}$, which permitted to perform readily the derivation of the dynamical Eq. (11), is also attainable in this more elaborate model.

While the precession term remains unaltered, $\hat{\Gamma}\vec{\mathcal{J}}$ replaces $\vec{\mathcal{J}}$ in both the relaxation and free-diffusion terms, which are in general anisotropic. However, the matrix $\hat{\Gamma}$ is symmetric and can be diagonalized by choosing an appropriate reference system (indicated by the symmetry of the problem); let us assume then that $\Gamma_{xx} = \Gamma_{yy}$, as it holds for uniaxial or cubic anisotropies. Then, rewriting the action of $\vec{\mathcal{J}}$ as $\mathcal{J}_{\nu}X_{l}^{m} = \alpha_{l,m}^{\nu}X_{l}^{m_{\nu}}$, we have $(\hat{\Gamma}\vec{\mathcal{J}})_{\nu}X_{l}^{m} = (\gamma_{\nu}\alpha_{l,m}^{\nu})X_{l}^{m_{\nu}}$, with $\gamma_{\pm} = \Gamma_{xx} (=\Gamma_{yy})$ and $\gamma_{z} = \Gamma_{zz}$. This shows that the replacement of $\vec{\mathcal{J}}$ by $\hat{\Gamma}\vec{\mathcal{J}}$ implies that the effective coefficients $\tilde{\alpha}_{l,m}^{\nu} = \gamma_{\nu}\alpha_{l,m}^{\nu}$ now play the role of the $\alpha_{l,m}^{\nu}$ in the original model. Consequently, we can write *at once* the basic kinetic equation accounting for anisotropy-type fluctuations by just replacing in Eq. (11) the diffusion term by $\Sigma_{\nu}\mathcal{J}_{\nu}\tilde{\alpha}_{l,m}^{\nu}X_{l}^{m_{\nu}}$ and

plainly substituting $\tilde{\alpha}_{l,m}^{\nu}$ for $\alpha_{l,m}^{\nu}$ in the relaxation terms, to get contributions of the form $(\tilde{\alpha}^{z}\beta^{-} - \tilde{\alpha}^{-}\beta^{z}), \frac{1}{2}(\tilde{\alpha}^{-}\beta^{+} - \tilde{\alpha}^{+}\beta^{-}), \text{ and } (\tilde{\alpha}^{+}\beta^{z} - \tilde{\alpha}^{z}\beta^{+}).$

In summary, we have presented a method for the derivation of the basic kinetic equations governing the dynamics of rotational relaxation, based on a suitable representation of the adjoint Fokker-Planck operator in terms of the angular momentum operator. It has been shown that the structure of the kinetic equations derived [Eq. (11)] directly reflects that of the adjoint Fokker-Planck operator [Eq. (5)], which permits the derivation of the basic system of kinetic equations in more elaborate descriptions of rotational relaxation once the adjoint Fokker-Planck operator is constructed.

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APPENDIX: EXPLICIT EQUATIONS FOR X_l^m AND Y_l^m

Introducing the expressions for the coefficients α and β in Eq. (11), we get the explicit result

where we have formally replaced $\mathcal{L}_{FP}^{\dagger}$ by $2\tau_D(d/dt)$ [cf. Eq. (2)]. Since the order of magnitude of the coefficients can be very different for large *l*, and this can produce numerical instability, it will be in some cases convenient to use the equation for the Y_l^m . This can be obtained by multiplying across the above equation by $n_{l,m}$ [Eq. (7)], and calculating quotients of the form $n_{l,m}/n_{l',m'}$ to get

$$\begin{split} 2\,\tau_D \frac{d}{dt}\,Y_l^m + l(l+1)\,Y_l^m &= \frac{B_+}{2} \bigg[-(l+1)\,\sqrt{\frac{(l+m)(l+m-1)}{(2l-1)(2l+1)}} Y_{l-1}^{m-1} - \frac{i}{\lambda}\sqrt{(l+m)(l-m+1)}Y_l^{m-1} \\ &\quad -l\,\sqrt{\frac{(l-m+2)(l-m+1)}{(2l+1)(2l+3)}} Y_{l+1}^{m-1} \bigg] + B_z \bigg[(l+1)\,\sqrt{\frac{l^2-m^2}{(2l-1)(2l+1)}} Y_{l-1}^m \\ &\quad -\frac{i}{\lambda}mY_l^m - l\,\sqrt{\frac{(l+1)^2-m^2}{(2l+1)(2l+3)}} Y_{l+1}^m \bigg] + \frac{B_-}{2} \bigg[(l+1)\,\sqrt{\frac{(l-m)(l-m-1)}{(2l-1)(2l+1)}} Y_{l-1}^{m+1} \\ &\quad -\frac{i}{\lambda}\sqrt{(l-m)(l+m+1)}Y_l^{m+1} + l\,\sqrt{\frac{(l+m+2)(l+m+1)}{(2l+1)(2l+3)}} Y_{l+1}^{m+1} \bigg]. \end{split}$$

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