Quantum relaxation dynamics of magnetic moments in a radiative reservoir

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A quantum Langevin equation is derived to describe the relaxation dynamics of a magnetic moment in a static magnetic field and radiative reservoir. The damping and fluctuation forces are derived from the radiative interaction between magnetic moment and surrounding reservoir. Through the use of a symmetrized interaction Hamiltonian, the damping force is identified due to the combination of radiation self-reaction and reservoir fluctuations. The radiation self-reaction is a quantum version of its classical counterpart, whereas the reservoir fluctuations are solely a quantum effect resulting from the quantization of the electromagnetic field. The relative magnitude between these two effects changes during the relaxation process. This equation shows that the relative magnitude of the quantum correction to the classical Landau-Lifshitz description is inversely proportional to the system angular momentum quantum number.

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I. INTRODUCTION

With the recent advance in magnetic nanoparticle experiments[1] and the continuing increase in magnetic recording density, where magnetic grain is also on the nanometer scale, the dynamics of magnetic particles will soon be in the transition between classical and quantum regimes. When a magnetic particle is placed in an uniform static magnetic field, the magnetic moment M precesses about the magnetic field direction. At the same time, it also moves toward the field direction because of the damping effect from the interaction with the reservoir. Depending on the type of reservoir, the relaxation mechanism can be due to radiative interaction, phonon interaction, spin-spin interaction, and/or random collisions in fluid environment, etc. The internal degrees of freedom of the magnetic moment, e.g., the exchange coupling between constituent spins, is assumed to be not affected by the externally applied field. This assumption basically states that the magnitude of magnetic moment or the angular momentum quantum number is constant, which is normally true under most conditions. In the classical regime, this relaxation dynamics is described by the Landau-Lifshitz equation[2]

$$\frac{d\vec{M}}{dt} = \gamma(\vec{M}\times\vec{B}) - \frac{\alpha|\gamma|}{|\vec{M}|}\vec{M}\times(\vec{M}\times\vec{B}), \qquad (1)$$

where γ is the gyromagnetic constant. The first term on the right-hand side of Eq. (1) describes the familiar precession. The second term describes the phenomenological damping force with damping constant α. Based on the fluctuation-dissipation theorem,[3,4] a zero-mean random fluctuation noise $\vec{B}^f(t)$ is added to the magnetic field to describe the fluctuations associated with the damping process.[5–7] The random noise $\vec{B}^f(t)$ satisfies the correlation relation $\langle B_i^f(t)B_j^f(t')\rangle = 2\alpha k_B T/(\gamma M)\delta_{ij}\delta(t-t')$[5] where i and j are Cartesian indices. This relation is essential to ensure that the magnetic moments has a Boltzman distribution at thermal equilibrium. This Langevin form of the Landau-Lifshitz equation, also referred to as the Landau-Lifshitz-Gilbert equation, has been widely used to study various magnetic dynamics,[8–14] in particular, the thermally activated magnetization reversal in ferromagnetic particles. As the size of the magnetic particle in new technologies continues to decrease, the application of this classical equation becomes questionable.

In the quantum regime, this equation needs to be revised. It is interesting to see how the revised equation resembles or differs from its classical counterpart and the corresponding physical meanings. The Bloch equation[15] in quantum mechanics is the one that could be found to most closely resemble the Landau-Lifshitz equation. This equation was first introduced for nuclear induction study. It is basically also considered a damped magnetic moment problem and can be derived by using the density operator approach.[16] The Bloch equation is expressed as

$$\frac{d}{dt}\langle\vec{M}\rangle = \gamma\langle\vec{M}\rangle\times B_z\vec{z} - \frac{(\langle M_z\rangle - M_0)\vec{z}}{T_1} - \frac{\langle M_x\rangle\vec{x} + \langle M_y\rangle\vec{y}}{T_2}, \qquad (2)$$

where $\vec{x}$, $\vec{y}$, and $\vec{z}$ are the unit vectors, M_0 is the steady state magnetization, and T_1 and T_2 are the longitudinal and transverse relaxation time constants. The longitudinal relaxation describes the change of magnetization along the applied field direction. The transverse relaxation describes the loss of coherence of transverse magnetic moment due to random reservoir fluctuations.[15] It is interesting to see that this description of relaxation dynamics is very different from that in the classical Landau-Lifshitz Eq. (1). A quantum equation is expected to be able to reduce to its classical counterpart under certain approximations, e.g., the relevant quantum numbers become very large. It is important to derive an equation to resolve this discrepancy.

This paper presents a fully quantum-mechanical derivation for such an equation. The analysis considers only the radiative system-reservoir interaction, i.e., damping due to radiative energy decay. The system is defined as the magnetic moment in a dc magnetic field. The reservoir is the surrounding thermal radiation. This damping mechanism is chosen because the interaction Hamiltonian is well defined and depends only on fundamental parameters. Even though the radiative damping is not the primary contributing factor to the overall damping effect in a real system, the form of the interaction Hamiltonian is, in fact, rather general and can be applied to other damping mechanisms except with different interaction coefficients. The derived equation, therefore, provides general insights to the relaxation dynamics.

The analysis focuses on the dynamics of angular momentum operator $\hat{\mathbf{J}}$ because the magnetic moment is simply related to angular momentum by the gyromagnetic constant γ. This derivation leads to an angular momentum operator rate equation in the Langevin equation format, which includes a damping term and random fluctuation noise. Part of the damping term is in a form similar to the phenomenological damping force in the Landau-Lifshitz equation. It is contributed by the interaction of the magnetic moment with its own field (self-reaction). The other part of the damping term is more similar to that in the Bloch equation. It is contributed by the zero point vacuum fluctuations and thermal fluctuations of the surrounding reservoir (reservoir fluctuations). The equation not only provides a detailed physical picture for the relaxation dynamics, it also quantitatively describes the transition of the relaxation dynamics between classical and quantum regimes. The rest of the paper is organized as follows. The notation for the quantized electromagnetic field is briefly introduced in Sec. II. The derivation for the quantum Langevin equation is detailed in Sec. III, where the analysis is done in the Heisenberg picture. Finally, the physical meanings of the derived equation and its implications are discussed in Sec. IV, followed by a conclusion.

II. NOTATION FOR THE QUANTIZED ELECTROMAGNETIC FIELD

In the conventional quantization formulation, the electromagnetic radiation is often expressed in terms of a set of eigenmodes, where each mode is quantized as a simple harmonic oscillator. The quantized vector potential $\hat{A}$ in the plane wave mode expansion basis has the expression

$$\hat{\mathbf{A}} = \sum_{k,\varepsilon} \sqrt{\frac{\hbar}{2\epsilon_0\omega_k V}}\vec{\varepsilon}_k\{\hat{a}_{k\varepsilon}\exp(i\vec{k}\cdot\vec{x}) + \hat{a}^{\dagger}_{k\varepsilon}\exp(-i\vec{k}\cdot\vec{x})\}, \tag{3}$$

where $\vec{k}$ is the wave vector of each plane wave mode, ω_k is the corresponding oscillation frequency, V is the volume of the quantization box, and $\vec{\varepsilon}_k$ $(\varepsilon=1,2)$ are the two orthogonal polarization unit vectors associated with each vector $\vec{k}$. The time dependence is implicitly included in amplitude operator $\hat{a}_{k\varepsilon}$. Given this quantized vector potential $\hat{A}$, the quantized magnetic field operator $\hat{\mathbf{B}}$ is

$$\hat{\mathbf{B}} = \nabla\times\hat{\mathbf{A}} = \sum_{k,\varepsilon} i\sqrt{\frac{\hbar}{2\epsilon_0\omega_k V}}\vec{k}\times\vec{\varepsilon}_k\{\hat{a}_{k\varepsilon}\exp(i\vec{k}\cdot\vec{x}) - \hat{a}^{\dagger}_{k\varepsilon}\exp(-i\vec{k}\cdot\vec{x})\}. \tag{4}$$

The polarization vector $\vec{\varepsilon}_k$ will be implicitly included in the operators $\hat{a}_k$ and $\hat{a}^{\dagger}_k$ subsequently whenever it is convenient for notational simplicity. The creation and annihilation operators $\hat{a}^{\dagger}_k$ and $\hat{a}_k$ satisfy the commutation relations $[\hat{a}_n,\hat{a}^{\dagger}_m]=\delta_{nm}$ and $[\hat{a}_n,\hat{a}_m]=[\hat{a}^{\dagger}_n,\hat{a}^{\dagger}_m]=0$, where different polarization states are included in the the operator subfix. In this quantization formalism, a discrete summation over wave vector components $\vec{k}$ is used for mathematical simplicity. This does not compromise any important physics of interest in this paper.

III. THE QUANTUM LANGEVIN EQUATION OF ANGULAR MOMENTUM $\hat{J}$

The Hamiltonian for a magnetic moment placed in a static magnetic field $\mathbf{B}_{\mathrm{dc}}$ and radiative reservoir is

$$H = \sum_k \hbar\omega_k\left(\hat{a}^{\dagger}_k\hat{a}_k + \frac{1}{2}\right) - \gamma\hat{\mathbf{J}}\cdot\mathbf{B}_{\mathrm{dc}} - \gamma\hat{\mathbf{J}}\cdot\hat{\mathbf{B}}, \tag{5}$$

where the first term is the radiation field Hamiltonian of the reservoir, the second term is the system Hamiltonian of the magnetic moment in static magnetic field $\mathbf{B}_{\mathrm{dc}}$, and the last term is the radiative interaction Hamiltonian between magnetic moment and reservoir field. This interaction term assumes that the physical size of magnetic moment is much smaller than the wavelength of the electromagnetic field that can significantly contribute to the interaction. The magnetic moment, therefore, sees a spatially uniform electromagnetic field for all frequency components of interest. For wavelengths shorter than the physical size of magnetic moment, the interaction term has to take into account the spacial average of the field seen by magnetic moment. The short wavelength components, therefore, contribute little to the interaction term and can be neglected. The angular momentum and electromagnetic field observables are same-time commutable. The order of these two operators in the interaction term is undetermined, i.e., the interaction term can be written as $\hat{\mathbf{B}}\cdot\gamma\hat{\mathbf{J}}$ as well or even an arbitrary linear combination of these two different ordered expressions $\alpha\gamma\hat{\mathbf{J}}\cdot\hat{\mathbf{B}}+(1-\alpha)\hat{\mathbf{B}}\cdot\gamma\hat{\mathbf{J}}$, where $0\leqslant\alpha\leqslant 1$. The final derived rate equation for $\hat{\mathbf{J}}$ is independent of the order of $\hat{\mathbf{J}}$ and $\hat{\mathbf{B}}$ in the interaction term. However, it was pointed out that this interaction term has to be symmetrized in order to rationally identify the respective effects of radiation fluctuations (interaction of the system with the quantized radiation field) and self-reaction (interaction of the system with its own field) involved in the interaction process.[17] The total Hamiltonian is thus rewritten as

$$H = \sum_k \hbar\omega_k\left(\hat{a}^{\dagger}_k\hat{a}_k + \frac{1}{2}\right) - \gamma\hat{\mathbf{J}}\cdot\mathbf{B}_{\mathrm{dc}} - \frac{1}{2}\gamma(\hat{\mathbf{J}}\cdot\hat{\mathbf{B}} + \hat{\mathbf{B}}\cdot\hat{\mathbf{J}}). \tag{6}$$

From the above Hamiltonian, the equations of motion for angular momentum and field amplitude operators, in the Heisenberg picture, are

$$\frac{d\hat{\mathbf{J}}}{dt} = \frac{1}{i\hbar}[\hat{\mathbf{J}},H] = \gamma\hat{\mathbf{J}}\times\mathbf{B}_{\mathrm{dc}} + \frac{\gamma}{2}(\hat{\mathbf{J}}\times\hat{\mathbf{B}} - \hat{\mathbf{B}}\times\hat{\mathbf{J}}) \tag{7}$$

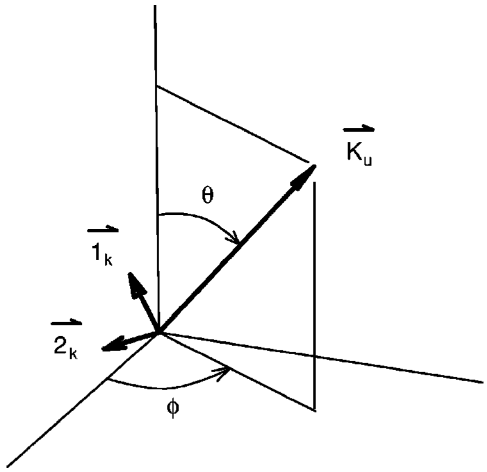

FIG. 1. The spherical coordinate representation for the unit wave vector $\vec{k}_u$, where $\vec{1}_k$ and $\vec{2}_k$ are the two orthogonal polarization states for each wave vector $\vec{k}$.

$$\frac{d\hat{a}_{k\varepsilon}}{dt} = \frac{1}{i\hbar}[\hat{a}_{k\varepsilon},H] = -i\omega_k\hat{a}_{k\varepsilon} + \frac{g_k}{\hbar}(\hat{\mathbf{J}} \times \vec{k}_u) \cdot \vec{\varepsilon}_k \exp(-i\vec{k} \cdot \vec{x}_J), \tag{8}$$

where $g_k = \gamma\sqrt{(\hbar/2\epsilon_0\omega_k V)}k$ is the coupling coefficient between angular momentum operator $\hat{\mathbf{J}}$ and the radiation field amplitude operator $\hat{a}_k$, $\vec{k}_u$ is the unit vector of $\vec{k}$ as shown in Fig. 1, and $\vec{x}_J$ is the position coordinate of the system.

Since the main interest is in the dynamics of the system angular momentum and no observation of the reservoir field is made except knowing that it is at temperature T, the goal is to derive a simplified equation of $\hat{\mathbf{J}}$. From Eqs. (7) and (8), the angular momentum and field operators are cross coupled. The field operator $\hat{a}_k$, in addition to its own free evolution, is driven by the angular momentum $\hat{\mathbf{J}}$ (magnetic moment $\hat{\mathbf{M}}$) source. Similarly, the angular momentum operator $\hat{J}$, in addition to its own precession due to the dc magnetic field $\mathbf{B}_{\mathrm{dc}}$, is driven by reservoir magnetic field operator $\hat{\mathbf{B}}$ $(\hat{a}_k)$. When the coupling constant g_k is small, which is the case since the size of the reservoir V is very large by definition, a second order perturbative calculation is sufficient. The simplification is to obtain the formal solutions of Eqs. (7) and (8) and then substitute these solutions into the interaction terms of Eq. (7) and keep the calculation up to the second order in g_k.

In order to further facilitate the derivation, the unit vector basis $(\vec{+},\vec{-},\vec{z})$ is introduced where

$$\vec{+} = \frac{1}{\sqrt{2}}(\vec{x} + i\vec{y}), \tag{9}$$

$$\vec{-} = \frac{1}{\sqrt{2}}(\vec{x} - i\vec{y}). \tag{10}$$

The angular momentum in this coordinate system is expressed as

$$\hat{\mathbf{J}} = \hat{J}_+\frac{\vec{-}}{\sqrt{2}} + \hat{J}_-\frac{\vec{+}}{\sqrt{2}} + \hat{J}_z\vec{z}, \tag{11}$$

where $\hat{J}_+ = \hat{J}_x + i\hat{J}_y$ and $\hat{J}_- = \hat{J}_x - i\hat{J}_y$ are the conventional raising and lowing angular momentum operators. Using this new basis, with the $\vec{z}$ vector chosen to be aligned with the dc magnetic field direction, Eq. (7) becomes

$$\frac{d}{dt}\hat{\mathbf{J}} = i\Omega\hat{J}_+\frac{\vec{-}}{\sqrt{2}} - i\Omega\hat{J}_-\frac{\vec{+}}{\sqrt{2}} + \frac{\gamma}{2}(\hat{\mathbf{J}} \times \hat{\mathbf{B}} - \hat{\mathbf{B}} \times \hat{\mathbf{J}}), \tag{12}$$

where $\Omega \equiv -\gamma|\mathbf{B}_{\mathrm{dc}}|$ is the precession frequency along the $\vec{z}$ axis. In the rest of paper, the gyromagnetic constant is assumed to be negative if not explicitly stated. The formal solution for each component of $\hat{\mathbf{J}}$ is

$$\hat{J}_z = \hat{J}_z(t_0) + \int_{t_0}^{t}\frac{\gamma}{2}(\hat{\mathbf{J}} \times \hat{\mathbf{B}} - \hat{\mathbf{B}} \times \hat{\mathbf{J}})_{t'} \cdot \vec{z}dt', \tag{13}$$

$$\hat{J}_+ = \hat{J}_+(t_0)e^{i\Omega(t-t_0)} + \int_{t_0}^{t}\frac{\gamma}{2}(\hat{\mathbf{J}} \times \hat{\mathbf{B}} - \hat{\mathbf{B}} \times \hat{\mathbf{J}})_{t'} \cdot \sqrt{2}\vec{+}\, e^{i\Omega(t-t')}dt', \tag{14}$$

$$\hat{J}_- = \hat{J}_-(t_0)e^{-i\Omega(t-t_0)} + \int_{t_0}^{t}\frac{\gamma}{2}(\hat{\mathbf{J}} \times \hat{\mathbf{B}} - \hat{\mathbf{B}} \times \hat{\mathbf{J}})_{t'} \cdot \sqrt{2}\,\vec{-}\, e^{-i\Omega(t-t')}dt'. \tag{15}$$

On the right-hand side of each of the last three equations, the first term describes the free evolution (precession) part of $\hat{\mathbf{J}}$, identified as $\hat{\mathbf{J}}^f$, and the second term describes the source driven part of $\hat{\mathbf{J}}$ by the reservoir field $\hat{\mathbf{B}}$, identified as $\hat{\mathbf{J}}^s$. The magnitude of the source term $\hat{\mathbf{J}}^s$, when expressed in terms of fundamental quantum operators $\hat{\mathbf{J}}$ and $\hat{a}_k$, is of the order of $O(g_k)$ compared to the free evolution part $\hat{\mathbf{J}}^f$. The formal solution of Eq. (8) is

$$\hat{a}_{k\varepsilon}(t) = \hat{a}_{k\varepsilon}(t_0)e^{-i\omega_k(t-t_0)} + \frac{g_k}{\hbar}\int_{t_0}^{t}(\hat{\mathbf{J}}(t') \times \vec{k}_u) \cdot \vec{\varepsilon}_k e^{-i\omega_k(t-t')-i\vec{k}\cdot\vec{x}_J}dt' \tag{16}$$

$$= \hat{a}_{k\varepsilon}^f(t) + \hat{a}_{k\varepsilon}^s(t), \tag{17}$$

where $\hat{a}_{k\varepsilon}^f(t)$ is the free evolution and $\hat{a}_{k\varepsilon}^s(t)$ is the contribution from the radiation source $\hat{\mathbf{J}}$. Similarly, the total reservoir field $\hat{\mathbf{B}}$ consists of a free evolution $\hat{\mathbf{B}}^f$ and a source driven part $\hat{\mathbf{B}}^s$. The magnitude of the source term $\hat{a}_{k\varepsilon}^s$ $(\hat{\mathbf{B}}^s)$ is also of the order of $O(g_k)$ compared to the free evolution part $\hat{a}_{k\varepsilon}^f$ $(\hat{\mathbf{B}}^f)$. The above formal solutions for $\hat{\mathbf{J}}$, Eqs. (13)–(15), and $\hat{a}_k$, Eq. (16), will be used to calculate the interaction terms in the angular momentum rate Eq. (7) up to the order of $O(g_k^2)$. The first interaction term $\hat{\mathbf{J}} \times \hat{\mathbf{B}}$ will be calculated first. The second interaction term $\hat{\mathbf{B}} \times \hat{\mathbf{J}}$ can be readily obtained by simply reversing the order of the operators therein.

Using the free evolution and source driven terms of field operator $\hat{\mathbf{B}}$, the first interaction term in Eq. (7) is separated into two parts

$$\gamma(\hat{\mathbf{J}} \times \hat{\mathbf{B}}) = \gamma(\hat{\mathbf{J}} \times \hat{\mathbf{B}}^f) + \gamma(\hat{\mathbf{J}} \times \hat{\mathbf{B}}^s) \quad (18)$$

$$= \mathbf{R}(t) + \mathbf{S}(t). \quad (19)$$

The part $\mathbf{R}(t)$ describes the change rate of angular momentum due to its interaction with the free evolution of reservoir field $\hat{\mathbf{B}}^f$. This contribution is often called reservoir fluctuations. The part $\mathbf{S}(t)$ describes the change rate of angular momentum due to the interaction with its self-generated field $\hat{\mathbf{B}}^s$. This contribution is often called self-reaction. Using the result for $\hat{a}^s_{k\varepsilon}$ in Eq. (16), $\mathbf{S}(t)$ is

$$\mathbf{S}(t) = \hat{\mathbf{J}} \times \sum_k \frac{g_k^2}{\hbar} i \int_{t_0}^{t} \{\vec{k}_u \times [\hat{\mathbf{J}}(t') \times \vec{k}_u] e^{-i\omega_k(t-t')} - \text{H.c.}\} dt', \quad (20)$$

where H.c. stands for Hermitian conjugate. This self-reaction term is of the order of $O(g_k^2)$. The summation is taken over those k values satisfying the periodic boundary condition of the quantization box, i.e., $|k_i| = 2\pi n/L$, where $i = x, y, z$, n is a positive integer and L is the length of the quantization box. In $\vec{k}$ space representation, these $\vec{k}$ values sit on the cubic lattice points where the cubic volume is $8\pi^3/L^3$. Therefore, the quantization mode density is $\rho(k) = V/(8\pi^3)$, where $V = L^3$. The summation over quantization mode component k can be mathematically approximated by an integral in k space[18]

$$\sum_k \simeq \int_{k=0}^{\infty} \int_{\phi=0}^{2\pi} \int_{\theta=0}^{\pi} \rho(k) k^2 \sin\theta d\theta d\phi dk, \quad (21)$$

where ϕ and θ are the polar coordinates in the $\vec{k}$ vector space. After carrying out the integral over ϕ and θ, the self-reaction term is reduced to

$$\mathbf{S}(t) = \hat{\mathbf{J}}(t) \times i \int_{t_0}^{t} dt' \int_0^{\infty} dk \frac{g_k^2}{\hbar} \rho(k) \frac{8}{3} \pi k^2 \hat{\mathbf{J}}_{t'} (e^{-i\omega_k(t-t')} - \text{c.c.}) \quad (22)$$

$$= -\frac{1}{2} \int_{t_0}^{t} dt' \int_0^{\infty} dk G_k (\hat{J}_{+t} \hat{J}_{-t'} - \hat{J}_{-t} \hat{J}_{+t'}) (e^{-i\omega_k(t-t')} - \text{c.c.}) \vec{z}$$
$$+ \int_{t_0}^{t} dt' \int_0^{\infty} dk G_k (\hat{J}_{zt} \hat{J}_{-t'} - \hat{J}_{-t} \hat{J}_{zt'}) (e^{-i\omega_k(t-t')} - \text{c.c.}) \frac{\vec{+}}{\sqrt{2}}$$
$$- \int_{t_0}^{t} dt' \int_0^{\infty} dk G_k (\hat{J}_{zt} \hat{J}_{+t'} - \hat{J}_{+t} \hat{J}_{zt'}) (e^{-i\omega_k(t-t')} - \text{c.c.}) \frac{\vec{-}}{\sqrt{2}}, \quad (23)$$

where $G_k \equiv (g_k^2/\hbar)\rho(k)\frac{8}{3}\pi k^2$ and c.c. stands for complex conjugate. To evaluate the integral, the precession frequency components of $\hat{J}_\pm$ are factored out by expressions

$$\hat{J}_+(t) = \hat{\tilde{J}}_+(t) e^{i\Omega t}, \quad (24)$$

$$\hat{J}_-(t) = \hat{\tilde{J}}_-(t) e^{-i\Omega t}, \quad (25)$$

where $\hat{\tilde{J}}_+(t)$ and $\hat{\tilde{J}}_-(t)$ are slowly time varying functions due to the system-reservoir interaction. First, let us examine the $\vec{z}$ component of $\mathbf{S}(t)$ in Eq. (23),

$$S_z(t) = -\frac{1}{2} \int_{t_0}^{t} dt' \int_0^{\infty} dk G_k (\hat{\tilde{J}}_{+t} \hat{\tilde{J}}_{-t'} e^{i(\Omega - \omega_k)(t-t')}$$
$$- \hat{\tilde{J}}_{-t} \hat{\tilde{J}}_{+t'} e^{-i(\Omega + \omega_k)(t-t')} - \hat{\tilde{J}}_{+t} \hat{\tilde{J}}_{-t'} e^{i(\Omega + \omega_k)(t-t')}$$
$$+ \hat{\tilde{J}}_{-t} \hat{\tilde{J}}_{+t'} e^{-i(\Omega - \omega_k)(t-t')}). \quad (26)$$

Since G_k is a slow function of k and is modulated by sinusoidal functions $e^{\pm i(\Omega \pm \omega_k)(t-t')}$, the integral over k is significant only for $t' \to t$ and becomes negligible for $t - t' \gg 1/\Delta$, where Δ is the spectral width of G_k. As a result, the integral over t' can be approximated by the substitution $\hat{\tilde{J}}(t') \simeq \hat{\tilde{J}}(t)$ and replacing t_0 by $-\infty$.[19,20]

To better illustrate this approximation, the integral of the first term in the parenthesis is rewritten as

$$\int_{t_0}^{t} dt' h(\Omega, t - t') \hat{\tilde{J}}_+(t) \hat{\tilde{J}}_-(t'), \quad (27)$$

where $h(\Omega, t-t') = \int_0^\infty dk G_k e^{i(\Omega - \omega_k)(t-t')}$. The function G_k has a k^3 dependence. This is derived for radiation wavelengths significantly greater than the physical size of magnetic moment. As mentioned earlier, the interaction Hamiltonian becomes negligible for wavelengths much smaller than the physical size of magnetic moment. This implies that G_k is a decreasing function of k when k is very large. The spectrum of G_k, therefore, has a finite width Δ. The time constant $\tau_c \equiv 1/\Delta$ can be regarded as the reservoir correlation time because G_k describes the spectrum of the reservoir field components that can significantly interact with magnetic moment. When $t - t' \gg 1/\Delta$, the product of G_k and $e^{i(\Omega - \omega_k)(t-t')}$ is a fast oscillation function of k and the integral over k, i.e., $h(\Omega, t-t')$, is negligible. When $t - t' \simeq 0$, it is clear that the integral is significant. This property of $h(\Omega, t-t')$ means that the integral in Eq. (27) is mostly contributed by those values of $\hat{\tilde{J}}_-(t')$ when t' is very close to t. If $\hat{\tilde{J}}_-(t)$ varies very little over a time interval of the order of τ_c, the integral in Eq. (27) can be approximated by replacing $\hat{\tilde{J}}_-(t')$ by $\hat{\tilde{J}}_-(t)$ and t_0 by $-\infty$. This approximation basically says that the system relaxation time is much longer than the reservoir correlation time. This condition is satisfied under most circumstances because the reservoir, by definition, consists of a very large number of degrees of freedom and therefore has a very short correlation time τ_c. Physically, this approximation implies that the change rate of $\hat{\mathbf{J}}$ depends only on its present state and has no memory of its past history. This is in fact the so-called

Markoffian approximation in the context of stochastic processes.

With the above approximation, the integral in Eq. (26) can be readily carried out by first integrating over t', which results in $\delta(\Omega \pm \omega_k)$ functions, and then integrating over k. The overall integral is mainly contributed by the first and fourth terms in the parenthesis for a negative gyromagnetic constant and the self radiation reaction $S_z(t)$ is reduced to

$$S_z(t) = -\frac{1}{2}\frac{\pi}{c}G_{k_\Omega}[\hat{J}_+(t)\hat{J}_-(t) + \hat{J}_-(t)\hat{J}_+(t)], \qquad (28)$$

where the subscript $k_\Omega = \Omega/c$. (For a positive gyromagnetic constant, the above equation changes sign because the integral is instead contributed by the second and third terms.) In addition to the above result, there is actually also an imaginary part in the integral calculation. This is in fact very much like the Lamb shift[21] in the atomic energy level due to the atom-field interaction. Because this shift is usually rather small and is not the main interest of this paper, it is neglected here. Applying similar calculations, the other two components of $\mathbf{S}(t)$ are reduced to

$$S_-(t) = \frac{\pi}{c}G_{k_\Omega}\hat{J}_z(t)\hat{J}_-(t), \qquad (29)$$

$$S_+(t) = \frac{\pi}{c}G_{k_\Omega}\hat{J}_z(t)\hat{J}_+(t). \qquad (30)$$

(The above two equations also change sign for a positive gyromagnetic constant.) The above self-reaction results are carried out from the interaction expression $\gamma\hat{\mathbf{J}} \times \hat{\mathbf{B}}^s$. It is clear that S_- and S_+ are not Hermitian operators, therefore, it is improper to associate them with a physical meaning. In order to obtain a Hermitian expression, the symmetrized interaction Hamiltonian needs to be used. The self-reaction from $\gamma\hat{\mathbf{B}}^s \times \hat{\mathbf{J}}$ can be readily obtained by simply reversing the order of the operators in the above results. The Hermitian self-reaction term $\mathbf{S}_H(t)$ becomes

$$\mathbf{S}_H(t) = \frac{\gamma}{2}(\hat{\mathbf{J}} \times \hat{\mathbf{B}}^s - \hat{\mathbf{B}}^s \times \hat{\mathbf{J}}) \qquad (31)$$

$$= -\frac{1}{2}\frac{\pi}{c}G_{k_\Omega}(\hat{J}_+\hat{J}_- + \hat{J}_-\hat{J}_+)\vec{z} + \frac{1}{2}\frac{\pi}{c}G_{k_\Omega}(\hat{J}_z\hat{J}_- + \hat{J}_-\hat{J}_z)\frac{\vec{+}}{\sqrt{2}} + \frac{1}{2}\frac{\pi}{c}G_{k_\Omega}(\hat{J}_z\hat{J}_+ + \hat{J}_+\hat{J}_z)\frac{\vec{-}}{\sqrt{2}} \qquad (32)$$

$$= \frac{1}{2}\frac{\pi}{c}G_{k_\Omega}[\hat{\mathbf{J}} \times (\hat{\mathbf{J}} \times \vec{z}) - (\hat{\mathbf{J}} \times \vec{z}) \times \hat{\mathbf{J}}]. \qquad (33)$$

(The above equation changes sign for a positive gyromagnetic constant.) The last expression shows that the effect of the self radiation reaction always moves the magnetic moment toward the applied dc magnetic field direction. This description exactly matches to the phenomenological damping term in the classical Landau-Lifshitz Eq. (1).

The self-radiation reaction, however, only accounts for part of the energy dissipation processes experienced by the magnetic moment. The other part of energy dissipation mechanism is due to the reservoir fluctuation term $\mathbf{R}(t)$ in Eq. (19). To evaluate the reservoir fluctuations $\mathbf{R}(t) = \gamma\hat{\mathbf{J}} \times \hat{\mathbf{B}}^f$, substitute $\hat{\mathbf{J}}$ by its free evolution and source driven parts obtained from Eqs. (13)–(15),

$$\mathbf{R}(t) = \gamma\hat{\mathbf{J}}(t) \times \hat{\mathbf{B}}^f(t) \qquad (34)$$

$$= \gamma\hat{\mathbf{J}}^f(t) \times \hat{\mathbf{B}}^f(t) + \gamma\hat{\mathbf{J}}^s(t) \times \hat{\mathbf{B}}^f(t) \qquad (35)$$

$$= \mathbf{R}^f(t) + \mathbf{R}^B(t). \qquad (36)$$

The reservoir average of the first term $\mathbf{R}^f = \gamma\hat{\mathbf{J}}^f(t) \times \hat{\mathbf{B}}^f(t)$ is zero because free evolution $\hat{\mathbf{J}}^f$ is independent of $\hat{\mathbf{B}}^f$ and the reservoir average of $\hat{\mathbf{B}}^f$ is zero. This $\mathbf{R}^f$ term corresponds to the zero-mean random fluctuation noise in the Langevin equation context. The reservoir average of the second term $\mathbf{R}^B = \gamma\hat{\mathbf{J}}^s(t) \times \hat{\mathbf{B}}^f(t)$ is not zero because $\hat{\mathbf{J}}^s(t)$ is driven by the reservoir field $\hat{\mathbf{B}}$. This second term partially contributes to the damping and is proportional to the fluctuation noise power, which will be calculated shortly. The zero-mean random fluctuation force $\mathbf{R}^f$ is of order $O(g_k)$, and the nonzero mean reservoir fluctuation induced damping $\mathbf{R}^B$ is of order $O(g_k^2)$. This is a general characteristic of Langevin equation. To carry the calculation further, the field operator $\hat{a}_k$ in $\hat{\mathbf{J}}^s$ is approximated by its free evolution part $\hat{a}_k^f$. Since the reservoir field, by definition, is insignificantly perturbed by a small system, the difference between $\hat{a}_k$ and $\hat{a}_k^f$ is rather small and is only of order $O(g_k)$. This approximation leads to a deviation of order $O(g_k^3)$ for the calculated $\mathbf{R}^B$ and is, therefore, justified because we calculate $\mathbf{S}(t)$ and $\mathbf{R}(t)$ only up to order $O(g_k^2)$. After expanding the cross products and applying the similar Markoffian approximation used to calculate the self-reaction term $\mathbf{S}(t)$ in Eq. (26), the symmetrized Hermitian reservoir fluctuations $\mathbf{R}_H(t)$ become (see Appendix A for details)

$$\mathbf{R}_H(t) = \gamma\hat{\mathbf{J}}^f(t) \times \hat{\mathbf{B}}^f(t) - \frac{\pi}{c}G_{k_\Omega}\hbar\left(\hat{J}_z\vec{z} + \frac{\hat{J}_-}{2}\frac{\vec{+}}{\sqrt{2}} + \frac{\hat{J}_+}{2}\frac{\vec{-}}{\sqrt{2}}\right) \times (1 + 2\hat{n}_\Omega) \qquad (37)$$

$$= \mathbf{R}_H^f + \mathbf{R}_H^B, \qquad (38)$$

where $\mathbf{R}_H^f$ stands for the zero-mean random fluctuation force and $\mathbf{R}_H^B$ stands for the reservoir fluctuation induced dissipation. From the above result, we see that the reservoir fluctuations contribute to the decay of all three angular momentum components. The magnitude of this decay rate is proportional to $1 + 2\hat{n}_\Omega$, where the unity is a result of the electromagnetic field quantization and stands for the zero point vacuum fluctuations and $\hat{n}_\Omega$ is the thermal photon number operator. The reservoir average of thermal photon number $\hat{n}_{k_\Omega}$ at temperature T is $\langle \hat{n}_{k_\Omega} \rangle = 1/[\exp(\hbar\Omega/k_B T) - 1]$. Due to the existence of the zero point vacuum fluctuations, the reservoir fluctuation induced dissipation is inescapable even at zero temperature.

Given the results of the self-reaction $\mathbf{S}_H$, Eq. (33), and radiation fluctuations $\mathbf{R}_H$ Eq. (38), we are now ready to rewrite the angular momentum rate equation in a quantum Langevin format

$$\frac{d}{dt}\hat{\mathbf{J}} = \gamma\hat{\mathbf{J}} \times B_{\mathrm{dc}}\vec{z} + \mathbf{S}_H(t) + \mathbf{R}_H^B(t) + \mathbf{R}_H^f(t), \tag{39}$$

where the dissipation force $\mathbf{S}_H(t)+\mathbf{R}_H^B(t)$ is

$$\mathbf{S}_H(t) + \mathbf{R}_H^B(t) = \pm\frac{1}{2}\frac{\pi}{c}G_{k_\Omega}[\hat{\mathbf{J}} \times (\hat{\mathbf{J}} \times \vec{z}) - (\hat{\mathbf{J}} \times \vec{z}) \times \hat{\mathbf{J}}] - \frac{1}{2}\frac{\pi}{c}G_{k_\Omega}\hbar(1 + 2\hat{n}_\Omega)\left(2\hat{J}_z\vec{z} + \hat{J}_-\frac{\vec{+}}{\sqrt{2}} + \hat{J}_+\frac{\vec{-}}{\sqrt{2}}\right) \tag{40}$$

and the zero-mean random fluctuation force is $\mathbf{R}_H^f(t) = \gamma\hat{\mathbf{J}}^f(t)\times\hat{\mathbf{B}}^f(t)$. The $\pm$ signs are for negative and positive gyromagnetic constants, respectively. The dissipation process is identified as partly due to the interaction of the magnetic moment with its self-generated field (self-reaction) $\mathbf{S}_H$ and partly due to its interaction with the fluctuating reservoir field (reservoir fluctuations) $\mathbf{R}_H^B$. The self-reaction term is in a form similar to the classical damping term in the Landau-Lifshitz equation. Physically, this damping force is the result of the radiative energy loss due to magnetic moment precession and can be viewed as a quantum version of its classical counterpart. The reservoir fluctuations are similar to the damping terms in the Bloch equation. This damping effect is the result of the magnetic moment dephasing due to the surrounding reservoir random fluctuations. They include contributions from random thermal photon fluctuations and zero point vacuum fluctuations. One further interesting aspect of this damping force is that the derived longitudinal decay rate is twice that of the transverse decay rate. This stands in contrast to the two independent decay rates in the conventional Bloch equation. The zero-mean random fluctuation noise $\mathbf{R}_H^f$ describes the fluctuation force underlying the dissipation process and completes the full description of the relaxation dynamics.

The zero-mean random field fluctuations $\hat{\mathbf{B}}^f(t)$ are not only important from the view point of fluctuation-dissipation theorem, it is also essential to assure that the derived equation is self consistent, i.e., to preserve the angular momentum commutation relation. In order to demonstrate the quantum self consistency, it is necessary to verify if the derived angular momentum equation still satisfies the fundamental commutation relation $[\hat{J}_i,\hat{J}_j]=\epsilon_{ijk}i\hbar\hat{J}_k$, where ϵ_{ijk} is the permutation constant, or in a vector format $\hat{\mathbf{J}}\times\hat{\mathbf{J}}=i\hbar\hat{\mathbf{J}}$. Instead of solving for the rate equation and check if the solution satisfies the above commutation relation, an alternative approach is to take time derivative of the commutation relation

$$\frac{d\hat{\mathbf{J}}}{dt} \times \hat{\mathbf{J}} + \hat{\mathbf{J}} \times \frac{d\hat{\mathbf{J}}}{dt} = i\hbar\frac{d\hat{\mathbf{J}}}{dt} \tag{41}$$

and verify that the rate Eq. (39) for $\hat{\mathbf{J}}$ indeed satisfies the above relation. This proof is provided in Appendix B.

From statistical mechanics, when a system reaches thermal equilibrium with its surrounding reservoir at temperature T, it will be in a Boltzmann statistical mixture of its energy eigenstates. It is straightforward to verify that the angular momentum operator rate (39) indeed reaches thermal equilibrium at this state, i.e., $\mathrm{Tr}\{\sigma_S\dot{\hat{\mathbf{J}}}\}=0$, where the density operator

$$\sigma_S = \sum_{m=-J}^{m=J}\frac{e^{-m\beta}}{\sum e^{-m\beta}}|J,m\rangle\langle m,J|, \tag{42}$$

and $\beta=\hbar\Omega/k_BT$. The quantum magnetic moment rate equation can be easily obtained by $\hat{\mathbf{M}}\equiv\gamma\hat{\mathbf{J}}$. The damping constant due to radiative decay becomes $\alpha=\pi G_{k_\Omega}M/(c\gamma\Omega)$. It can also be shown that the precessing magnetic moment sees a zero-mean random fluctuation field $\hat{\mathbf{B}}^f(t)$ with the statistical property

$$\langle\hat{B}_i^f(t)\hat{B}_j^f(t')\rangle = 2\alpha\frac{\hbar\Omega}{\gamma M}\left(\langle\hat{n}_{k_\Omega}\rangle + \frac{1}{2}\right)\delta_{ij}\delta(t-t') \tag{43}$$

$$\simeq 2\alpha\frac{k_BT}{\gamma M}\left(1 + \frac{1}{2}\frac{\hbar\Omega}{k_BT}\right)\delta_{ij}\delta(t-t'), \tag{44}$$

where i and j are Cartesian indices and the approximation $\langle\hat{n}_{k_\Omega}\rangle\simeq k_BT/(\hbar\Omega)$ for $\hbar\Omega\ll k_BT$ is used in the second equality. The last expression is identical to the classical expression except for the additional second term in the parenthesis, which represents the contribution from zero point vacuum fluctuations. One final note, even though the rate Eq. (39) is derived specifically for the relaxation of a magnetic moment, this result is also applicable to the electric dipole radiation of a collective $2J$ two level systems. This is the so-called superradiance, where the system state can be modeled as an angular momentum state $|J,m\rangle$.[22]

IV. DISCUSSION

Let us consider a system with a negative gyromagnetic constant and initially in an angular momentum state $|j,m=j\rangle$. The reservoir is assumed to be at temperature $T=0$, i.e., thermal photon number $\langle\hat{n}_\Omega\rangle=0$. Physically, the magnetic moment is at its highest-energy state and will gradually move toward the lowest energy state. From the derived rate equation, the initial system energy decay rate is

$$\Omega\left\langle\frac{d\hat{J}_z}{dt}\right\rangle = \Omega\left\langle -\frac{1}{2}\frac{\pi}{c}G_{k_\Omega}(\hat{J}_+\hat{J}_- + \hat{J}_-\hat{J}_+ + 2\hbar\hat{J}_z)\right\rangle_{j,j} \tag{45}$$

$$=-\frac{\pi}{c}\Omega G_{k_\Omega}\hbar^2 j-\frac{\pi}{c}\Omega G_{k_\Omega}\hbar^2 j, \qquad (46)$$

where $\hat{J}_+\hat{J}_-+\hat{J}_-\hat{J}_+$ and $2\hbar\hat{J}_z$ are due to self-reaction and vacuum fluctuations, respectively. These two effects equally contribute to the initial system energy decay rate. The vacuum reservoir fluctuations play a very crucial role in starting the relaxation process because it not only introduces an initial tipping force to $\vec{J}$ but also provides a path for the self-reaction decay to start. After the decay process is completed, the angular momentum eventually reaches the lowest-energy state $|j,-j\rangle$. The energy decay rate at this state is

$$\Omega\left\langle\frac{d\hat{J}_z}{dt}\right\rangle=\Omega\left\langle-\frac{1}{2}\frac{\pi}{c}G_{k_\Omega}(\hat{J}_+\hat{J}_-+\hat{J}_-\hat{J}_++2\hbar\hat{J}_z)\right\rangle_{j,-j} \qquad (47)$$

$$=-\frac{\pi}{c}G_{k_\Omega}\hbar^2 j+\frac{\pi}{c}G_{k_\Omega}\hbar^2 j \qquad (48)$$

$$=0. \qquad (49)$$

The vacuum fluctuations play a crucial role here, again. The energy loss due to self-reaction is exactly balanced out by the energy gained from vacuum fluctuations. This ensures the stability of the ground state.

Another interesting point to check is the relative magnitude between the self-reaction and vacuum fluctuation effects during the relaxation process. Since it is rather difficult to obtain the detail evolution of angular momentum operator, for illustrative purpose, one can instead evaluate the relative magnitudes for different coherent Bloch states,[23,24]

$$|\theta,\phi\rangle=\sum_{m=-j}^{j}\left(\frac{(2j)!}{(j+m)!\,(2j-j-m)!}\right)^{1/2}\frac{\tau^{j+m}}{(1+|\tau|^2)^j}|j,m\rangle, \qquad (50)$$

where $\tau=e^{-i\phi}/\tan(\theta/2)$. This state is chosen because the expectation value of angular momentum for this state closely resembles a classical angular momentum vector with magnitude $\hbar j$ and polar coordinate (θ,ϕ), i.e., $\langle\hat{\mathbf{J}}\rangle_{\theta,\phi}=\hbar\vec{j}$. The damping force experienced by this angular momentum state is

$$\langle\mathbf{S}_H+\mathbf{R}_H^B\rangle=\frac{1}{2}\frac{\pi}{c}G_{k_\Omega}\langle\hat{\mathbf{J}}\times(\hat{\mathbf{J}}\times\vec{z})-(\hat{\mathbf{J}}\times\vec{z})\times\hat{\mathbf{J}}\rangle_{\theta,\phi}$$
$$-\frac{1}{2}\frac{\pi}{c}G_{k_\Omega}\hbar\left\langle 2\hat{J}_z\vec{z}+\hat{J}_-\frac{\vec{+}}{\sqrt{2}}+\hat{J}_+\frac{\vec{-}}{\sqrt{2}}\right\rangle_{\theta,\phi}, \qquad (51)$$

$$=\frac{\pi}{c}G_{k_\Omega}\hbar^2\left(-j^2\sin^2\theta-j\left(1-\frac{\sin^2\theta}{2}\right)\right)\vec{z}$$
$$+\frac{\pi}{c}G_{k_\Omega}\hbar^2\left(j^2-\frac{j}{2}\right)e^{-i\phi}\sin\theta\cos\theta\frac{\vec{+}}{\sqrt{2}}$$
$$+\frac{\pi}{c}G_{k_\Omega}\hbar^2\left(j^2-\frac{j}{2}\right)e^{i\phi}\sin\theta\cos\theta\frac{\vec{-}}{\sqrt{2}}-\frac{\pi}{c}G_{k_\Omega}\hbar^2$$
$$\times j(1+2\hat{n}_\Omega)\left(\cos\theta\vec{z}+\frac{1}{2}\sin\theta\left(e^{-i\phi}\frac{\vec{+}}{\sqrt{2}}+e^{i\phi}\frac{\vec{-}}{\sqrt{2}}\right)\right), \qquad (52)$$

where, in the last equality, the first three terms are due to the self-reaction effect and the last one is due to the reservoir fluctuation effect. The above expression shows how the magnitudes of self-reaction and reservoir fluctuation effects change for different coherent angular momentum state $|\theta,\phi\rangle$. It also shows how the dynamics transits from quantum to classical regimes as angular momentum number j increases.

The magnitudes of self-reaction and vacuum fluctuation contributions to magnetic moment relaxation are determined by the $\vec{z}$ components in Eq. (52). The magnitudes of these two effects are comparable when θ is close to zero or π. As θ approaches $\pi/2$, the self-reaction effect becomes dominant over the vacuum fluctuation effect. In fact, the self-reaction effect is always greater or equal to the vacuum fluctuation effect with equality at $\theta=0$ or π and the reservoir fluctuation effect makes no contribution to system energy decay at $\theta=0$. The transition of relaxation dynamics from quantum to classical regimes is determined by the j dependence in Eq. (52). When the angular momentum number becomes very large, the energy decay is mainly determined by the self-reaction terms with j^2 dependence and Eq. (52) can be reduced to

$$\langle\mathbf{S}_H+\mathbf{R}_H^B\rangle\simeq\frac{\pi}{c}G_{k_\Omega}\hbar^2 j^2\left(-\sin^2\theta\vec{z}+e^{-i\phi}\sin\theta\cos\theta\frac{\vec{+}}{\sqrt{2}}+e^{i\phi}\sin\theta\cos\theta\frac{\vec{-}}{\sqrt{2}}\right) \qquad (53)$$

$$=\frac{\pi}{c}G_{k_\Omega}\hbar^2\vec{j}\times(\vec{j}\times\vec{z}). \qquad (54)$$

The reservoir averaged angular momentum rate equation for a coherent state $|\theta,\phi\rangle$ is then simplified to

$$\frac{d}{dt}\hbar\vec{j}\simeq\gamma\hbar\vec{j}\times\vec{B}_{\mathrm{dc}}\pm\frac{\pi}{c}G_{k_\Omega}\hbar^2\vec{j}\times(\vec{j}\times\vec{z}), \qquad (55)$$

for $j\gg 1$. The $\pm$ signs of damping term are for negative and positive gyromagnetic constants. Since j is assumed to be a very large quantum number, $\hbar\vec{j}$ can be considered as a classical angular momentum vector $\vec{J}$. After explicitly spelling out the G_{k_Ω} coefficient defined earlier, the rate equation for angular momentum $\vec{J}$ becomes

$$\frac{d}{dt}\vec{J} = \gamma\vec{J}\times\vec{B}_{dc} \pm \frac{|\Omega|^3\gamma^2}{6\pi\epsilon_0 c^5}\vec{J}\times(\vec{J}\times\vec{z}), \tag{56}$$

or for magnetic moment $\vec{M}$ with negative gyromagnetic constant

$$\frac{d}{dt}\vec{M} = \gamma\vec{M}\times\vec{B}_{dc} - \frac{\gamma^4|\vec{B}_{dc}|^2}{6\pi\epsilon_0 c^5}\vec{M}\times(\vec{M}\times\vec{B}_{dc}). \tag{57}$$

This equation matches in form to the Landau-Lifshitz Eq. (1). The last term basically describes the radiative decay of a precessing magnetic moment.[25] Because the damping effect from j^2-dependent terms is similar to the classical description as demonstrated, all the other j-dependent terms in Eq. (52) can be viewed as quantum correction. It is important to note that the relative magnitude of quantum correction to classical $\hat{\mathbf{J}}\times(\hat{\mathbf{J}}\times\vec{B})$ damping description is of the order of $1/j$.

The Landau-Lifshitz damping constant α due to this radiative decay is $\alpha=\gamma^3B_{dc}^2M/(6\pi\epsilon_0c^5)$. For a magnetic moment $M=4\times10^{-19}$A m^2 [equivalent to a (10 nm)^3 magnetic grain with 400 $e$mu/cm^3], $B_{dc}=0.4T$, and $\gamma=1.76\times10^{11}$ s^{-1} T^{-1}, the damping constant is $\alpha\simeq8.6\times10^{-19}$. The order of magnitude does not change much even with the thermal photon $\langle\hat{n}_\Omega\rangle$ taken into account. This is much smaller than the often quoted values $\alpha\sim0.01-1$ in literatures for magnetic recording material. It is clear that the decay mechanism for the quoted material system is mostly contributed by other damping mechanisms, e.g., phonon and spin-spin interactions, rather than radiative decay. Nevertheless, the study of radiative decay mechanism at the quantum level provides useful insights for other damping mechanisms. In general, the system-reservoir interaction Hamiltonian H_{int} for other types of damping mechanisms can, at least to the first order, be expressed as $\vec{M}\cdot\Sigma_i\vec{R}_i$, where $\Sigma_i\vec{R}_i$ includes all reservoir variables and can be obtained by $\Sigma_i\vec{R}_i=\partial H_{int}/\partial\vec{M}$. This is similar to the radiative interaction Hamiltonian $H_{int}=\vec{M}\cdot\vec{B}$. The derived self-reaction and reservoir-fluctuation relaxation dynamics is therefore a rather general result. Equations (39) and (40) can be applied to other damping mechanisms except that the interaction coefficient G_k varies depending on the damping mechanism.

V. CONCLUSION

Starting from the Hamiltonian with symmetrized radiative interaction terms, a quantum Langevin equation is derived to describe the relaxation dynamics of a magnetic moment in static magnetic field. This analysis results in an angular momentum operator rate equation, which includes a dissipation force and a zero-mean random fluctuation noise along with the familiar precession motion. The derived dissipation force is contributed by both self-reaction and reservoir fluctuations. The reservoir fluctuation induced dissipation is uniquely obtained from the quantized electromagnetic field. The self-reaction induced dissipation, on the other hand, is a quantum version of its classical counterpart. The zero mean random noise force in the rate equation is essential for describing the existence of the random fluctuations underlying the dissipation process. This fluctuation noise also plays a very important role in preserving the conventional angular momentum commutation relation. The derived operator rate equation quantitatively describes the transition of the relaxation dynamics between classical and quantum regimes.

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APPENDIX A: RESERVOIR FLUCTUATION INDUCED DAMPING

The detailed calculation for the reservoir fluctuation induced dissipation term $\mathbf{R}_H^B(t)$ is carried out in this appendix. For the convenience of reference, $\mathbf{R}_H^B(t)$ is rewritten here

$$\mathbf{R}_H^B(t) = \frac{\gamma}{2}\{\hat{\mathbf{J}}^s(t)\times\hat{\mathbf{B}}^f(t) - \hat{\mathbf{B}}^f(t)\times\hat{\mathbf{J}}^s(t)\}, \tag{A1}$$

where $\hat{\mathbf{J}}^s(t)$ is the integral part of Eqs. (13)–(15). The interaction term $\mathbf{R}^B=\gamma\hat{\mathbf{J}}^s\times\hat{\mathbf{B}}^f$ is first calculated and then the result is symmetrized to obtain $\mathbf{R}_H^B$.

To facilitate the calculation, the coordinate system as shown in Fig. 1 is used, where $\vec{k}_u=\cos\theta\vec{z}+\sin\theta\cos\phi\vec{x}+\sin\theta\sin\phi\vec{y}$ is the unit vector of the quantized plane wave mode and $\vec{1}_k=\sin\theta\vec{z}-\cos\theta\cos\phi\vec{x}-\cos\theta\sin\phi\vec{y}$ and $\vec{2}_k=\sin\phi\vec{x}-\cos\phi\vec{y}$ are the unit vectors of the two orthogonal polarization states for each mode vector $\vec{k}$. For the convenience of calculation, $\vec{1}_k$ and $\vec{2}_k$ are expressed in the vector basis $(\vec{+},\vec{-},\vec{z})$

$$\vec{1}_k = \sin\theta\vec{z} - \cos\theta\left(e^{-i\phi}\frac{\vec{+}}{\sqrt{2}} + e^{i\phi}\frac{\vec{-}}{\sqrt{2}}\right), \tag{A2}$$

$$\vec{2}_k = ie^{-i\phi}\frac{\vec{+}}{\sqrt{2}} - ie^{i\phi}\frac{\vec{-}}{\sqrt{2}}. \tag{A3}$$

In this vector basis, the three components of the source driven angular momentum operator $\hat{\mathbf{J}}^s(t)=\hat{J}_z^s\vec{z}+\hat{J}_-^s(\vec{+}/\sqrt{2})+\hat{J}_+^s(\vec{-}/\sqrt{2})$ are

$$\hat{J}_z^s = \sum_k i\frac{g_k}{2}\int_{t_0}^{t}\{(-\hat{J}_+e^{-i\phi}-\hat{J}_-e^{i\phi})A_{k1} + i(\hat{J}_+e^{-i\phi} - \hat{J}_-e^{i\phi})\cos\theta A_{k2}\}_{t'}dt', \tag{A4}$$

$$\hat{J}_+^s = \sum_k ig_k\int_{t_0}^{t}\{\hat{J}_ze^{i\phi}A_{k1} + i(\hat{J}_z\cos\theta e^{i\phi} + \hat{J}_+\sin\theta)A_{k2}\}_{t'}e^{i\Omega(t-t')}dt', \tag{A5}$$

$$\hat{J}_-^s = \sum_k i g_k \int_{t_0}^{t} \{\hat{J}_z e^{-i\phi} A_{k1} - i(\hat{J}_z \cos\theta e^{-i\phi} + \hat{J}_- \sin\theta) A_{k2}\}_{t'} e^{-i\Omega(t-t')} dt', \tag{A6}$$

where the shorthand notation for the quantized field operator is $A_{k\varepsilon} \equiv \hat{a}_{k\varepsilon}^f e^{-i\omega_k t} - \hat{a}_{k\varepsilon}^{f\dagger} e^{i\omega_k t}$. Using these notations, $\mathbf{R}^B$ becomes

$$\mathbf{R}^B(t) = \gamma \hat{\mathbf{J}}^s(t) \times \hat{\mathbf{B}}^f(t) \tag{A7}$$

$$= \sum_{k\varepsilon} i g_k \hat{\mathbf{J}}^s(t) \times (\vec{k}_u \times \vec{\varepsilon}_k) A_{k\varepsilon} \tag{A8}$$

$$= \sum_{k\varepsilon} i g_k \hat{\mathbf{J}}^s(t) \times (\vec{2}_k A_{k1} - \vec{1}_k A_{k2}) \tag{A9}$$

$$= R_z^B(t)\vec{z} + R_-^B(t)\frac{\vec{+}}{\sqrt{2}} + R_+^B(t)\frac{\vec{-}}{\sqrt{2}}. \tag{A10}$$

After carrying out the cross product and collecting all the $\vec{z}$ components, R_z^B is

$$R_z^B = \sum_k g_k^2 \frac{1}{2} \int_{t_0}^{t} \{\hat{J}_{zt'} A_{k2t'} A_{k2t} + (\hat{J}_z \cos\theta e^{i\phi} + \hat{J}_+ \sin\theta)_{t'} \cos\theta e^{-i\phi} A_{k1t'} A_{k1t}\} e^{i\Omega(t-t')} dt' + \sum_k g_k^2 \frac{1}{2} \int_{t_0}^{t} \{\hat{J}_{zt'} A_{k2t'} A_{k2t} + (\hat{J}_z \cos\theta e^{-i\phi} + \hat{J}_- \sin\theta)_{t'} \cos\theta e^{i\phi} A_{k1t'} A_{k1t}\} e^{-i\Omega(t-t')} dt' = -\int dk G_k \int_{t_0}^{t} dt' \hbar \hat{J}_{zt'} (\hat{a}_k^{f\dagger} \hat{a}_k^f e^{-i\omega_k(t-t')} + \hat{a}_k^f \hat{a}_k^{f\dagger} e^{i\omega_k(t-t')})(e^{i\Omega(t-t')} + \text{c.c.}) \tag{A11}$$

$$= -\frac{\pi}{c} G_{k_\Omega} \hbar \hat{J}_z(t) (\hat{a}_{k_\Omega}^{f\dagger} \hat{a}_{k_\Omega}^f + \hat{a}_{k_\Omega}^f \hat{a}_{k_\Omega}^{f\dagger}), \tag{A12}$$

where Eq. (21) is used to approximate the discrete summation over $\vec{k}$ by an integral in $\vec{k}$ space in the second equality. Because of the sinusoidal modulation $e^{\pm i\omega_k(t-t')}$ in the integral, $\hat{J}_z(t')$ makes significant contribution to the integral with respect to k only for $t' \to t$. Therefore, similar Markoffian approximation used to calculate the self-reaction term is used again to obtain the last equality. Applying the similar calculation, the $\vec{+}$ component is

$$R_-^B = \sum_k g_k^2 \int_{t_0}^{t} (\hat{J}_z \cos\theta e^{-i\phi} + \hat{J}_- \sin\theta)_{t'} \sin\theta A_{k1t'} A_{k1t} e^{-i\Omega(t-t')} dt' + \sum_k g_k^2 \int_{t_0}^{t} \left(\frac{\hat{J}_+}{2} e^{-i\phi} + \frac{\hat{J}_-}{2} e^{i\phi}\right)_{t'} e^{-i\phi} A_{k2t'} A_{k2t} dt' + \sum_k g_k^2 \int_{t_0}^{t} \left(-\frac{\hat{J}_+}{2} e^{-i\phi} + \frac{\hat{J}_-}{2} e^{i\phi}\right)_{t'} \cos^2\theta e^{-i\phi} A_{k1t'} A_{k1t} dt' \tag{A13}$$

$$= -\int dk G_k \int_{t_0}^{t} dt' \hbar \hat{J}_{-t'} (\hat{a}_k^{f\dagger} \hat{a}_k^f e^{-i\omega_k(t-t')} + \hat{a}_k^f \hat{a}_k^{f\dagger} e^{i\omega_k(t-t')})(e^{-i\Omega(t-t')} + 1) \tag{A14}$$

$$= -\int dk G_k \int_{t_0}^{t} dt' \hbar \hat{\tilde{J}}_{-t'} (\hat{a}_k^{f\dagger} \hat{a}_k^f e^{-i\omega_k(t-t')} + \hat{a}_k^f \hat{a}_k^{f\dagger} e^{i\omega_k(t-t')})(1 + e^{i\Omega(t-t')}) e^{-i\Omega t} \tag{A15}$$

$$= -\frac{\pi}{c} G_{k_\Omega} \hbar \hat{J}_-(t) \hat{a}_{k_\Omega}^{f\dagger} \hat{a}_{k_\Omega}^f. \tag{A16}$$

To properly apply the Markoffian approximation to the above two calculations, $\hat{J}_+(t') = \hat{\tilde{J}}_+(t') e^{i\Omega(t'-t)}$ and $\hat{J}_-(t') = \hat{\tilde{J}}_-(t') e^{-i\Omega(t'-t)}$ are used in the third equality to calculate the integral. A concern might be raised here regarding the commutability problem between operators $\hat{\mathbf{J}}(t')$ and $\hat{\mathbf{B}}^f(t)$ $[\hat{a}_k^f(t)]$ in the above integral calculation. Strictly speaking $\hat{\mathbf{J}}$ is only same time commutable with $\hat{\mathbf{B}}$ $(\hat{a}_k)$ and $\hat{a}_k$ and $\hat{a}_k^f$ are different by a source driven term $\hat{a}_k^s$. This problem is resolved by the following two reasons. First, because the above integral is significant only for operators at $t' \to t$, the different time commutability problem between $\hat{\mathbf{J}}(t')$ and $\hat{a}_k^f(t)$ is reduced to a same time commutability problem after applying the Markoffian approximation. Secondly, the difference between $\hat{a}_k$ and $\hat{a}_k^f$ is of the order of $O(g_k)$ from Eq. (16). $\hat{\mathbf{J}}$ and $\hat{a}_k^f$ can be treated as commutable up to the order of $O(g_k)$. This approximation introduces a correction term of order $O(g_k^3)$ to the above final expression and is, therefore, justified because our calculation is only up to the order of $O(g_k^2)$. The $\vec{-}$ component is simply the Hermitian conjugate of R_-^B.

Finally, the symmetrized Hermitian reservoir-fluctuation induced damping term $\mathbf{R}_H^B$ is reduced to

$$\mathbf{R}_H^B = -\frac{\pi}{c}G_{k_\Omega}\hbar\left(\hat{J}_z\vec{z} + \frac{\hat{J}_-}{2}\frac{\vec{+}}{\sqrt{2}} + \frac{\hat{J}_+}{2}\frac{\vec{-}}{\sqrt{2}}\right)(1+2\hat{n}_\Omega), \tag{A17}$$

where $\hat{a}^{f\dagger}_{k_\Omega}\hat{a}^f_{k_\Omega} + \hat{a}^f_{k_\Omega}\hat{a}^{f\dagger}_{k_\Omega} = 1 + 2\hat{n}_\Omega$ is used.

APPENDIX B: CONSERVATION OF THE ANGULAR MOMENTUM COMMUTATION RELATION

This appendix is to show that the angular momentum operator in the derived operator rate (39) satisfies the conventional angular momentum commutation relation, i.e., it satisfies Eq. (41). The proof is done for a negative gyromagnetic constant and can be easily applied to a positive value. For the convenience of calculation, the dissipation term Eq. (40) is further reduced to

$$\mathbf{S}_H(t) + \mathbf{R}_H^B(t) = -\frac{\pi}{c}G_{k_\Omega}(\hat{J}_+\hat{J}_- + 2\hbar\hat{J}_z\hat{n}_{k_\Omega})\vec{z} + \frac{\pi}{c}G_{k_\Omega}(\hat{J}_z\hat{J}_- - \hbar\hat{J}_-\hat{n}_{k_\Omega})\frac{\vec{+}}{\sqrt{2}} + \frac{\pi}{c}G_{k_\Omega}(\hat{J}_+\hat{J}_z - \hbar\hat{J}_+\hat{n}_{k_\Omega})\frac{\vec{-}}{\sqrt{2}}. \tag{B1}$$

Also, the shorthand notation for time derivative $d\hat{\mathbf{J}}/dt = \dot{\hat{\mathbf{J}}}$, is use interchangeably. Since the derived angular momentum rate equation is calculated up to the order of $O(g_k^2)$, the proof is also carried out up to the same order.

The cross product of $\dot{\hat{\mathbf{J}}}\times\hat{\mathbf{J}} + \hat{\mathbf{J}}\times\dot{\hat{\mathbf{J}}}$ is carried out first term by term and then verified if it is equal to $i\hbar\dot{\hat{\mathbf{J}}}$. From the precession term, $-\Omega\hat{\mathbf{J}}\times\vec{z}$ in $\dot{\hat{\mathbf{J}}}$, we have

$$(\dot{\hat{\mathbf{J}}}\times\hat{\mathbf{J}} + \hat{\mathbf{J}}\times\dot{\hat{\mathbf{J}}})_{\text{pre}} = (-\Omega\hat{\mathbf{J}}\times\vec{z})\times\hat{\mathbf{J}} + \hat{\mathbf{J}}\times(-\Omega\hat{\mathbf{J}}\times\vec{z}) \tag{B2}$$

$$= -i\hbar\Omega\hat{\mathbf{J}}\times\vec{z}. \tag{B3}$$

From the damping terms due to the self-reaction and vacuum fluctuations $(\pi/c)G_{k_\Omega}[-\hat{J}_+\hat{J}_-\vec{z} + \hat{J}_z\hat{J}_-(\vec{+}/\sqrt{2}) + \hat{J}_+\hat{J}_z(\vec{-}/\sqrt{2})]$ in $\dot{\hat{\mathbf{J}}}$, we have

$$(\dot{\hat{\mathbf{J}}}\times\hat{\mathbf{J}} + \hat{\mathbf{J}}\times\dot{\hat{\mathbf{J}}})_{SR\text{-}VF} = \frac{\pi}{c}G_{k_\Omega}\left(-\hat{J}_+\hat{J}_-\vec{z} + \hat{J}_z\hat{J}_-\frac{\vec{+}}{\sqrt{2}} + \hat{J}_+\hat{J}_z\frac{\vec{-}}{\sqrt{2}}\right)\times\hat{\mathbf{J}} + \hat{\mathbf{J}}\times\frac{\pi}{c}G_{k_\Omega}\left(-\hat{J}_+\hat{J}_-\vec{z} + \hat{J}_z\hat{J}_-\frac{\vec{+}}{\sqrt{2}} + \hat{J}_+\hat{J}_z\frac{\vec{-}}{\sqrt{2}}\right) \tag{B4}$$

$$= i\hbar\frac{\pi}{c}G_{k_\Omega}\left\{(-\hat{J}_+\hat{J}_- + 2\hat{J}_z^2)\vec{z} + 3\hat{J}_z\hat{J}_-\frac{\vec{+}}{\sqrt{2}} + 3\hat{J}_+\hat{J}_z\frac{\vec{-}}{\sqrt{2}}\right\}. \tag{B5}$$

From the thermal photon induced damping terms $-(\pi/c)G_{k_\Omega}[2\hbar\hat{J}_z\vec{z} + \hbar\hat{J}_-(\vec{+}/\sqrt{2}) + \hbar\hat{J}_+(\vec{-}/\sqrt{2})]\hat{n}_{k_\Omega}$ in $\dot{\hat{\mathbf{J}}}$, we have

$$(\dot{\hat{\mathbf{J}}}\times\hat{\mathbf{J}} + \hat{\mathbf{J}}\times\dot{\hat{\mathbf{J}}})_{th} = -\frac{\pi}{c}G_{k_\Omega}\hbar\left(2\hat{J}_z\vec{z} + \hat{J}_-\frac{\vec{+}}{\sqrt{2}} + \hat{J}_+\frac{\vec{-}}{\sqrt{2}}\right)\hat{n}_{k_\Omega}\times\hat{\mathbf{J}} - \hat{\mathbf{J}}\times\frac{\pi}{c}G_{k_\Omega}\hbar\left(2\hat{J}_z\vec{z} + \hat{J}_-\frac{\vec{+}}{\sqrt{2}} + \hat{J}_+\frac{\vec{-}}{\sqrt{2}}\right)\hat{n}_{k_\Omega} \tag{B6}$$

$$= i\hbar\frac{\pi}{c}G_{k_\Omega}\left\{-2\hbar\hat{J}_z^2\hat{n}_{k_\Omega}\vec{z} - 3\hbar\hat{J}_-\hat{n}_{k_\Omega}\frac{\vec{+}}{\sqrt{2}} - 3\hbar\hat{J}_+\hat{n}_{k_\Omega}\frac{\vec{-}}{\sqrt{2}}\right\}. \tag{B7}$$

From the zero mean fluctuation term $\gamma\hat{\mathbf{J}}^f\times\hat{\mathbf{B}}^f$ in $\dot{\hat{\mathbf{J}}}$, we have

$$(\dot{\hat{\mathbf{J}}}\times\hat{\mathbf{J}} + \hat{\mathbf{J}}\times\dot{\hat{\mathbf{J}}})_{\text{noise}} = \gamma(\hat{\mathbf{J}}^f\times\hat{\mathbf{B}}^f)\times(\hat{\mathbf{J}}^f + \hat{\mathbf{J}}^s) + (\hat{\mathbf{J}}^f + \hat{\mathbf{J}}^s)\times\gamma(\hat{\mathbf{J}}^f\times\hat{\mathbf{B}}^f) \tag{B8}$$

$$= i\hbar\gamma(\hat{\mathbf{J}}^f\times\hat{\mathbf{B}}^f) + \gamma(\hat{\mathbf{J}}^f\times\hat{\mathbf{B}}^f)\times\hat{\mathbf{J}}^s + \hat{\mathbf{J}}^s\times\gamma(\hat{\mathbf{J}}^f\times\hat{\mathbf{B}}^f), \tag{B9}$$

where $\hat{\mathbf{J}}$ is explicitly expressed by its free evolution $\hat{\mathbf{J}}^f$ and source driven $\hat{\mathbf{J}}^s$ parts.

To calculate the cross product terms involving $\hat{\mathbf{J}}^s$ in the above equation, we use the coordinate system introduced in Appendix A and carry out the computation in basis $(\vec{+}, \vec{-}, \vec{z})$. The three components of $\gamma\hat{\mathbf{J}}^f\times\hat{\mathbf{B}}^f = (\gamma\hat{\mathbf{J}}^f\times\hat{\mathbf{B}}^f)_z\vec{z} + (\gamma\hat{\mathbf{J}}^f\times\hat{\mathbf{B}}^f)_-(\vec{+}/\sqrt{2}) + (\gamma\hat{\mathbf{J}}^f\times\hat{\mathbf{B}}^f)_+(\vec{-}/\sqrt{2})$ are

$$\gamma(\hat{\mathbf{J}}^f\times\hat{\mathbf{B}}^f)_z = \sum_k i\frac{g_k}{2}\{(-\hat{J}_+^f e^{-i\phi} - \hat{J}_-^f e^{i\phi})A_{k1}^f + i(\hat{J}_+^f e^{-i\phi} - \hat{J}_-^f e^{i\phi})\cos\theta A_{k2}^f\}, \tag{B10}$$

$$\gamma(\hat{\mathbf{J}}^f\times\hat{\mathbf{B}}^f)_+ = \sum_k ig_k\{\hat{J}_z^f e^{i\phi}A_{k1}^f + i(\hat{J}_z^f\cos\theta e^{i\phi} + \hat{J}_+^f\sin\theta)A_{k2}^f\}, \tag{B11}$$

$$\gamma(\hat{\mathbf{J}}^f\times\hat{\mathbf{B}}^f)_- = \sum_k ig_k\{\hat{J}_z^f e^{-i\phi}A_{k1}^f - i(\hat{J}_z^f\cos\theta e^{-i\phi} + \hat{J}_-^f\sin\theta)A_{k2}^f\}. \tag{B12}$$

Using these equations and the Eqs. (A4) and (A5) of $\hat{\mathbf{J}}^s$, one can calculate each vector component for $\hat{\mathbf{J}}^s\times\gamma(\hat{\mathbf{J}}^f\times\hat{\mathbf{B}}^f) + \gamma(\hat{\mathbf{J}}^f\times\hat{\mathbf{B}}^f)\times\hat{\mathbf{J}}^s$. The $\vec{z}$ component is

$$\{\hat{\mathbf{J}}^s \times \gamma(\hat{\mathbf{J}}^f \times \hat{\mathbf{B}}^f) + \gamma(\hat{\mathbf{J}}^f \times \hat{\mathbf{B}}^f) \times \hat{\mathbf{J}}^s\}_z \vec{z} = \{[\hat{J}_+^s, \gamma(\hat{\mathbf{J}}^f \times \hat{\mathbf{B}}^f)_-] - [\hat{J}_-^s, \gamma(\hat{\mathbf{J}}^f \times \hat{\mathbf{B}}^f)_+]\} i \frac{\vec{z}}{2}, \quad (B13)$$

$$= \Big\{ - \sum_k g_k^2 \int_{t_0}^t \hat{J}_z(t')\hat{J}_z^f(t)(1+\cos^2\theta)[A(t'),A^f(t)]e^{i\Omega(t-t')}dt' - \sum_k g_k^2 \int_{t_0}^t \sin^2\theta[\hat{J}_+(t')A(t'),\hat{J}_-^f(t)A^f(t)]e^{i\Omega(t-t')}dt' + \sum_k g_k^2 \int_{t_0}^t \hat{J}_z(t')\hat{J}_z^f(t)(1+\cos^2\theta)[A(t'),A^f(t)]e^{-i\Omega(t-t')}dt' + \sum_k g_k^2 \int_{t_0}^t \sin^2\theta[\hat{J}_-(t')A(t'),\hat{J}_+^f(t)A^f(t)]e^{-i\Omega(t-t')}dt' \Big\} i\frac{\vec{z}}{2}, \quad (B14)$$

$$= i\hbar\frac{\pi}{c}G_{k_\Omega}(-2\hat{J}_z^2)\vec{z}, \quad (B15)$$

where the integral approximation Eq. (21) for the discrete summation over $\vec{k}$ and the Markoffian approximation are used to lead to the final equality. The approximations $\hat{\mathbf{J}}^f \approx \hat{\mathbf{J}}$ and $A^f \approx A$ are used to calculate the commutation terms [$\ldots$, $\ldots$] in the above last second equality because they only introduce correction terms of order $O(g_k^3)$ and the analysis in this paper is done up to the order of $O(g_k^2)$. The same approach can be used to calculate the $\vec{+}$ and $\vec{-}$ components

$$\{\hat{\mathbf{J}}^s \times \gamma(\hat{\mathbf{J}}^f \times \hat{\mathbf{B}}^f) + \gamma(\hat{\mathbf{J}}^f \times \hat{\mathbf{B}}^f) \times \hat{\mathbf{J}}^s\}_- \frac{\vec{+}}{\sqrt{2}} = \{[\hat{J}_-^s, \gamma(\hat{\mathbf{J}}^f \times \hat{\mathbf{B}}^f)_z] - [\hat{J}_z^s, \gamma(\hat{\mathbf{J}}^f \times \hat{\mathbf{B}}^f)_-]\} i \frac{\vec{+}}{\sqrt{2}} \quad (B16)$$

$$= i\hbar\frac{\pi}{c}G_{k_\Omega}(-2\hat{J}_z\hat{J}_- + 2\hbar\hat{J}_-\hat{n}_{k_\Omega})\frac{\vec{+}}{\sqrt{2}}, \quad (B17)$$

$$\{\hat{\mathbf{J}}^s \times \gamma(\hat{\mathbf{J}}^f \times \hat{\mathbf{B}}^f) + \gamma(\hat{\mathbf{J}}^f \times \hat{\mathbf{B}}^f) \times \hat{\mathbf{J}}^s\}_+ \frac{\vec{-}}{\sqrt{2}} = \{-[\hat{J}_+^s, \gamma(\hat{\mathbf{J}}^f \times \hat{\mathbf{B}}^f)_z] + [\hat{J}_z^s, \gamma(\hat{\mathbf{J}}^f \times \hat{\mathbf{B}}^f)_+]\} i \frac{\vec{-}}{\sqrt{2}} \quad (B18)$$

$$= i\hbar\frac{\pi}{c}G_{k_\Omega}(-2\hat{J}_+\hat{J}_z + 2\hbar\hat{J}_+\hat{n}_{k_\Omega})\frac{\vec{-}}{\sqrt{2}}. \quad (B19)$$

Combining the results from Eqs. (B3), (B5), (B7), (B9), (B15), (B17), and (B19), we obtain

$$\frac{d\hat{\mathbf{J}}}{dt} \times \hat{\mathbf{J}} + \hat{\mathbf{J}} \times \frac{d\hat{\mathbf{J}}}{dt} = i\hbar\frac{d\hat{\mathbf{J}}}{dt}. \quad (B20)$$

Thus, it is shown that the angular momentum rate (39) indeed preserves the angular momentum commutation relation.

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