Intermode depolarization correlation of magnons

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Multimode magnon dynamics affected by demagnetizing fields has been discussed in terms of a Hamiltonian approach. In the multimode dynamics involving magnetization relaxation, inter-mode correlation of magnons is found to be formed spontaneously. The correlation suppresses the transverse magnetization dynamics, giving rise to depolarization correlation (DPC) of magnons. DPC may subsist in equilibrium distribution at finite temperatures.

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I. INTRODUCTION

Magnons, quanta of spin waves, play fundamental roles in properties of magnetic materials [1,2]. Magnon dynamics has not only been attracting much interest in various areas of fundamental physics but also providing indispensable tools in the recent developments of spintronics and magnonics [3,4]. In particular, magnons in magnetic materials can be utilized as an information carrier and a logic device in magnonics, without the need for electric currents which lead to substantial energy consumption. However, magnon dynamics and transport are strongly affected by magnetization relaxation [5].

Main relaxation processes for magnon dynamics can be classified by two concepts. One is the microscopic scattering process between magnons and other elementary excitations such as conduction electrons and phonons [6–9]. This type of microscopic spin relaxation is universal and inevitable in condensed matters, and leads to (quantum) decoherence of magnon states in magnets. The other type is phenomenology described by the so-called Gilbert damping term, which gives an effective description for real ferromagnetic (FM) materials at good accuracy [10,11]. The origin of the phenomenological Gilbert damping, in addition to the microscopic relaxation, has been discussed from macroscopic viewpoints, such as energy losses from eddy currents and electromagnetic radiations due to magnetization dynamics, frictions from deformation of sample shapes, and scatterings from defects and impurities in magnets (see Ref. [11] and references therein). The Gilbert damping term phenomenologically summarizes all the contributions. From its macroscopic origin, the Gilbert damping mechanism is considered to work on the local magnetization $\boldsymbol{M}(x)$, which is defined as the local order parameter, i.e., the expectation value of the averaged magnetic moment operator projected onto the atomic position x, rather than directly on the individual microscopic spins.

In this paper, we revisit magnon dynamics under the phenomenological Gilbert damping. So far, in the analysis of FM materials, dynamics of (almost) coherent modes of magnons has been mainly discussed within macrospin models. Here, we discuss the magnon dynamics involving the effects of inhomogeneous demagnetizing fields and the Gilbert damping in small FM samples, and investigate a nontrivial correlation among magnon modes arising from such effects beyond macrospin approximation.

The organization of this paper is as follows. In Sec. II, we illustrate the basic setup to realize the intermode correlation of magnons in general. After the brief summary of the magnetization dynamics in FM films in Sec. III, we concretely discuss the emergence of the two-mode correlation of magnons in finite-size FM films as an example in Sec IV. The temporal evolution of the magnon quantum states under the intermode correlation is discussed in a quantum-optics-like approach in Sec. V. Finally, we summarize our results in Sec. VI.

II. INTERMODE CORRELATION OF MAGNONS: BASIC SETUP

The dynamics of local magnetization $\boldsymbol{M}(x)$ in ferromagnets is phenomenologically described by the Landau-Lifshitz-Gilbert (LLG) equation [10,11] with the Gilbert damping term

$$-\lambda \boldsymbol{M}(x) \times (\boldsymbol{M}(x) \times \boldsymbol{H}_{\text{eff}}(x)), \tag{1}$$

where λ is the Gilbert damping constant and $\boldsymbol{H}_{\text{eff}}(x)$ is the effective magnetic field. Due to the Gilbert damping term, local magnetization dynamics is quickly relaxed in the Gilbert timescale $\tau_G \propto 1/\lambda$. On the basis of the linearized LLG equation in the equilibrium state, such as $\langle M_z \rangle = M_s =$ const and $\langle M_x \rangle = \langle M_y \rangle = 0$, each magnon, corresponding to each spin-wave mode of transverse magnetization fluctuations ($m_\pm = m_x \pm i m_y$), is usually described as an independent boson without interactions [1,2]. Thus, each magnon dynamics undergoes the Gilbert-type relaxation independently and simultaneously. This simultaneous relaxation essentially originates from the conservation of the wave-number k, which characterizes the spin-wave mode due to (discrete) translational invariance, much like the Bloch electrons.

In this paper, we consider magnon dynamics in small FM samples, such as small FM films with a finite area. In small samples, the finite-size effects and edge effects cannot be ignored, and the magnon dynamics is not expected to reflect the original translational invariance of crystals in the small k region. In particular, internal effective magnetic fields, demagnetizing fields, and surface anisotropies lead to the disturbance of homogeneity in the thermal equilibrium configuration and violate the translational invariance in small FM samples [1,2]. Thus, mixings among the different spin-wave modes can appear under such circumstances, with the aid of coupling to external fields (and nonlinear self-fields).

In addition, as discussed in Sec. I, the phenomenological Gilbert damping Eq. (1) works on the local magnetization, which corresponds to the macroscopic FM order parameter. As seen below, the transverse magnetization fluctuations can be generically expanded in terms of the contribution from each magnon mode: $m_\pm(x) = \sum_k m_k^\pm(x)$ (k is the label of modes). This implies that, in the Gilbert damping process, only the sum (or average) of the magnon contributions $m_\pm(x)$ should be relaxed in principle. It should be noted that there exist other microscopic spin relaxation processes, such as multimagnon and magnon-phonon scattering, characterized by the spin relaxation time τ_S. In this paper, we assume that the macroscopic Gilbert damping leads to the main relaxation process, and thus $\tau_G < \tau_S$ throughout.

Here, for concreteness, we consider the following process in small FM samples: A coherent magnon state of a single mode is assumed to be excited selectively by external pumping (such as microwave pumping) in a thermal ensemble with multiple magnon modes, and then relaxed by the Gilbert damping after turning off the pumping. In the Gilbert damping process, in addition to the finite-size effects, the damping effect leading to broadening of spectral width allows the mixing among almost degenerate magnon modes. From the above arguments, through this process, a nontrivial intermode correlation of magnons can be transiently formed in a finite time $\tau_G < t < \tau_S$.[1] In the following sections, we closely examine the intermode magnon correlation.

III. MAGNETIZATION DYNAMICS IN FM THIN FILMS

As an example of realizing the intermode magnon correlation, we consider the phenomenological Hamiltonian [12,13], describing the local magnetization dynamics in a thin FM film with a parallel external field $H_{\rm ex}$ in the z direction (see Fig. 1):

$$\hat{\mathcal{H}} = \int d^3x \left(-H_{\rm ex}\hat{M}_z + \frac{D}{2}\sum_a [\nabla \hat{M}_a]^2 + \frac{N}{2}\hat{M}_x^2 \right). \tag{2}$$

Here, $\hat{M}_a(x)$ $(a = x, y, z)$ is the local magnetization vector in the continuum approximation, D is the exchange stiffness constant, and N $(\sim O(1))$ is the demagnetizing factor in the

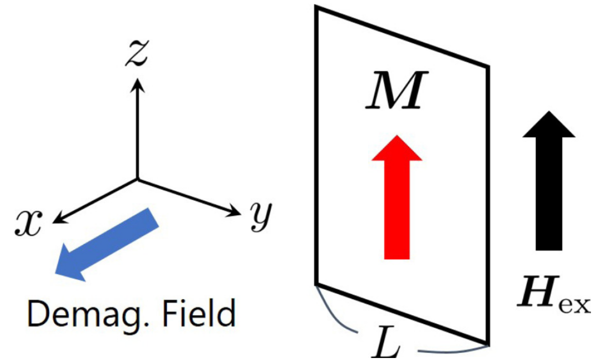

FIG. 1. Configuration of ferromagnetic film with finite area.

perpendicular x direction to the film. The magnetization vector is proportional to the spin density operators of electrons, $\hat{M}_a = -\gamma\,\hat{S}_a$, where $\gamma\ (> 0)$ is the gyromagnetic ratio, and thus we treat the magnetization vector as a quantum mechanical operator (indicated by the hat) in the following discussion. In this section, we briefly summarize magnetization dynamics in the FM film on the basis of the Hamiltonian Eq. (2), for this paper to be self-contained. (See Ref. [4] for details.)

A. Uniform magnetization dynamics in FM thin films

First, we summarize the dynamics of a unform precession mode of the magnetization vector. From the SU(2) algebra of spin, the uniform magnetization vector $\hat{M}_a$ satisfies the commutation relation,[2]

$$[\hat{M}_a,\, \hat{M}_b\,] = -i\gamma\,\epsilon_{abc}\,\hat{M}_c/V, \tag{3}$$

where ϵ_{abc} is the totally antisymmetric tensor with $\epsilon_{xyz} = 1$ and V is the volume of the FM sample.

The equations of motion for $\hat{M}_a(t)$ are derived from the Hamiltonian Eq. (2) (without spatial derivatives) using the commutation relation:

$$\frac{d\,\hat{M}_a}{dt} = -i[\hat{M}_a, \hat{\mathcal{H}}] = -\gamma\,\epsilon_{abc}\,\hat{M}_b\hat{H}_{\rm eff}^c\,, \tag{4}$$

where the effective magnetic field is given by $\hat{H}_{\rm eff}^a \equiv -\delta\hat{\mathcal{H}}/\delta\hat{M}_a = (-N\hat{M}_x, 0, H_{\rm ex})$. Note that, replacing the commutator with the Poisson bracket, we can obtain the classical equations of motion for magnetic texture from Eq. (2) and the following discussion also holds in the classical Hamiltonian mechanics.

In the following discussion, we consider the FM ordered phase where the magnetization almost aligns along the external magnetic field $\boldsymbol{H}_{\rm ex}$ in the z direction. Then, the expectation values of magnetization vector in the thermal equilibrium are given by $\langle M_z\rangle = M_s$ (= const) and $\langle M_x\rangle = \langle M_y\rangle = 0$, where M_s is the saturation magnetization. Note that, in this paper, we define the expectation values in the sense of quantum statistical mechanics as $\langle\mathcal{O}\rangle = {\rm tr}\,(\hat{\mathcal{O}}\,\hat{\rho})$ with the density matrix operator $\hat{\rho}$.

[1]The transient process in a finite time interval also allows the mixing among the modes with almost degenerate frequencies by uncertainty principle.

[2]We set $\hbar = 1$ and repeated indices are summed in the following discussion.

Under this situation, we can approximate dynamics of the local magnetization $\hat{M}_a(x)$ in terms of the fluctuation $\hat{m}_a(x)$ measured from the thermal equilibrium $\langle M_a\rangle$:

$$\hat{M}_x = \langle M_x\rangle + \hat{m}_x = \hat{m}_x, \quad \hat{M}_y = \langle M_y\rangle + \hat{m}_y = \hat{m}_y,$$
$$\hat{M}_z = \langle M_z\rangle + \hat{m}_z \simeq M_s \qquad (|\hat{m}_x|, |\hat{m}_y| \ll M_s). \tag{5}$$

Here, the fluctuation of $\hat{M}_z$ becomes higher order of $\hat{m}_x/M_s$ and $\hat{m}_y/M_s$ due to the constraint $\sum_a \hat{M}_a^2 = M_s^2$, and thus we neglect the parallel fluctuation $\hat{m}_z$.

At the linear order of the transverse fluctuations $(\hat{m}_x, \hat{m}_y)$, the equations of motion are reduced:

$$\frac{d\,\hat{m}_x}{dt} = -\gamma H_{\rm ex}\,\hat{m}_y,$$
$$\frac{d\,\hat{m}_y}{dt} = \gamma H_{\rm ex}\,\hat{m}_x + \gamma N M_s\,\hat{m}_x, \tag{6}$$

where the higher order terms of $\hat{m}_x/M_s$ and $\hat{m}_y/M_s$ are neglected. We focus on the linearized dynamics of transverse magnetization described by Eqs. (6).

To discuss the magnetization relaxation by the phenomenological LLG equation, we should incorporate the Gilbert damping term Eq. (1), which describes the relaxation of local magnetization dynamics. The Gilbert damping term can be similarly evaluated at the linear order of the transverse fluctuations:

$$-\lambda(\hat{\boldsymbol{M}} \times (\hat{\boldsymbol{M}} \times \hat{\boldsymbol{H}}_{\rm eff}))_x \simeq -\lambda\, H_{\rm ex} M_s\,\hat{m}_x - \lambda\, N M_s^2\,\hat{m}_x,$$
$$-\lambda(\hat{\boldsymbol{M}} \times (\hat{\boldsymbol{M}} \times \hat{\boldsymbol{H}}_{\rm eff}))_y \simeq -\lambda\, H_{\rm ex} M_s\,\hat{m}_y. \tag{7}$$

Using these expressions, we obtain the linearized LLG equations:

$$\frac{d\,\hat{m}_x}{dt} = -\gamma\, H_{\rm ex}\,\hat{m}_y - \lambda\, M_s(H_{\rm ex} + N M_s)\hat{m}_x,$$
$$\frac{d\,\hat{m}_y}{dt} = \gamma(H_{\rm ex} + N M_s)\hat{m}_x - \lambda\, M_s H_{\rm ex}\,\hat{m}_y. \tag{8}$$

In terms of the (circular) variables $\hat{m}_\pm = \hat{m}_x \pm i\hat{m}_y$, the linearized LLG equations can be compactly expressed as follows:

$$\frac{d\,\hat{m}_-}{dt} = -i\tilde{\gamma}\left(H_{\rm ex} + \frac{NM_s}{2}\right)\hat{m}_- - \frac{i\tilde{\gamma}NM_s}{2}\,\hat{m}_+,$$
$$\frac{d\,\hat{m}_+}{dt} = i\tilde{\gamma}^*\left(H_{\rm ex} + \frac{NM_s}{2}\right)\hat{m}_+ + \frac{i\tilde{\gamma}^*NM_s}{2}\,\hat{m}_-, \tag{9}$$

where $\tilde{\gamma} = \gamma - i\lambda M_s$ is a complexified gyromagnetic ratio and $\tilde{\gamma}^* = \gamma + i\lambda M_s$. The resulting Eqs. (9) imply that the effects of the Gilbert damping term can be incorporated by complexifying the parameters $\gamma \to \tilde{\gamma} = \gamma - i\lambda M_s$ in the equations of motion Eqs. (6) at the linear order of $\hat{m}_\pm$. In our linearized analysis, we assume the situation with small thermal fluctuations at sufficiently low temperatures, and the finite temperature effects are included in the effective damping term and the equilibrium distribution function.[3]

To describe magnon dynamics by the creation-annihilation operators using the Holstein-Primakoff (HP) transformation [18], we rewrite Eqs. (9) in terms of the spin operator $\hat{s}_\pm = -\hat{m}_\pm/\gamma$:

$$\frac{d\,\hat{s}_-}{dt} = -i\tilde{\gamma}\left(H_{\rm ex} + \frac{NM_s}{2}\right)\hat{s}_- - \frac{i\tilde{\gamma}NM_s}{2}\,\hat{s}_+,$$
$$\frac{d\,\hat{s}_+}{dt} = i\tilde{\gamma}^*\left(H_{\rm ex} + \frac{NM_s}{2}\right)\hat{s}_+ + \frac{i\tilde{\gamma}^*NM_s}{2}\,\hat{s}_-. \tag{10}$$

The linearized equations can be derived from the quadratic spin Hamiltonian for the fluctuations,

$$\hat{\mathcal{H}}_{\rm L} = \tilde{\gamma}\int d^3x\left[\left(\frac{H_{\rm ex}}{2S_0} + \frac{\gamma N}{4}\right)\hat{s}_+\hat{s}_- + \frac{\gamma N}{8}(\hat{s}_+^2 + \hat{s}_-^2)\right], \tag{11}$$

using the linearized spin algebra $[\hat{s}_+, \hat{s}_-] = -2S_0/V$ with $S_0 = M_s/\gamma$. Here, we introduce the HP representation at the linear order,

$$\hat{S}_- = \hat{s}_- = \sqrt{\frac{2S_0}{V}}\,\hat{a}, \quad \hat{S}_+ = \hat{s}_+ = \sqrt{\frac{2S_0}{V}}\,\hat{a}^\dagger,$$
$$\hat{S}_z = -S_0 + \frac{\hat{a}^\dagger\hat{a}}{V}, \tag{12}$$

where the creation-annihilation operators satisfy the commutation relation, $[\hat{a}, \hat{a}^\dagger] = 1$. Inserting the HP representation into the linearized Hamiltonian Eq. (11), we obtain the quadratic Hamiltonian with $\hat{a}$ and $\hat{a}^\dagger$:

$$\hat{\mathcal{H}}_{\rm L} = \tilde{\gamma}\left[\left(H_{\rm ex} + \frac{NM_s}{2}\right)\hat{a}^\dagger\hat{a} + \frac{NM_s}{4}(\hat{a}^{\dagger 2} + \hat{a}^2)\right]. \tag{13}$$

As is well-known, this Hamiltonian can be diagonalized by the Bogoliubov transformation [1,2]:

$$\hat{c} = \mu\,\hat{a} + \nu\,\hat{a}^\dagger \quad \text{with} \quad |\mu|^2 - |\nu|^2 = 1, \tag{14}$$

where the coefficients are given by

$$\mu = \cosh r, \qquad \nu = \sinh r,$$
$$\text{with} \quad e^r = \left(\frac{H_{\rm ex} + NM_s}{H_{\rm ex}}\right)^{1/4}. \tag{15}$$

Note that the transformed operators also satisfy the commutation relation, $[\hat{c}, \hat{c}^\dagger] = 1$. Using the transformed operators, $\hat{c}$ and $\hat{c}^\dagger$, the linearized Hamiltonian $\hat{\mathcal{H}}_L$ is diagonalized,

$$\hat{\mathcal{H}}_{\rm L} = \tilde{\gamma}\,\omega_0\left(\hat{c}^\dagger\hat{c} + \tfrac{1}{2}\right), \tag{16}$$

with $\omega_0 = \sqrt{H_{\rm ex}(H_{\rm ex} + NM_s)}$. This gives the eigenstates and FM resonance frequency of magnetization dynamics in thin films with an in-plane magnetic field $H_{\rm ex}$.

B. Spin waves in FM thin films

In this subsection, we briefly describe the dynamics of nonuniform magnetization, such as spin waves, in the FM thin film. The analysis in the previous subsection is naturally extended to the nonuniform magnetization $\hat{M}_a(x)$ which depends on spatial coordinates. In the nonuniform case, the commutation relation of magnetization components becomes

$$[\hat{M}_a(x),\, \hat{M}_b(x')] = -i\gamma\,\epsilon_{abc}\,\hat{M}_c(x)\,\delta^3(x - x'). \tag{17}$$

[3]More precisely, at finite temperatures, the LLG equation should be extended to the Langevin-type equation including stochastic noise term, and the distribution function is given by the Fokker-Planck equation [14–17].

At the linear order of the transverse fluctuations, $(\hat{m}_x, \hat{m}_y)$, in the FM ordered phase, the linearized LLG equations for nonuniform modes are similarly given by

$$\begin{aligned}\frac{d\,\hat{m}_x}{dt} &= -\gamma(H_{\rm ex} - DM_s\nabla^2)\hat{m}_y \\ &\quad -\lambda M_s(H_{\rm ex} - DM_s\nabla^2 + NM_s)\hat{m}_x, \\ \frac{d\,\hat{m}_y}{dt} &= \gamma(H_{\rm ex} - DM_s\nabla^2 + NM_s)\hat{m}_x \\ &\quad -\lambda M_s(H_{\rm ex} - DM_s\nabla^2)\hat{m}_y, \end{aligned} \tag{18}$$

where we used the linearized form of the damping term (7) with $\hat{\boldsymbol{H}}_{\rm eff} = (-N\hat{M}_x, 0, H_{\rm ex}) + D\,\nabla^2\hat{\boldsymbol{M}}$.

In terms of the spin variables $\hat{s}_{\pm}(x) = -\hat{m}_{\pm}(x)/\gamma$, the linearized LLG equations can also be compactly expressed as follows:

$$\begin{aligned}\frac{d\,\hat{s}_-}{dt} &= -i\tilde{\gamma}\left(H_{\rm ex} - DM_s\nabla^2 + \frac{NM_s}{2}\right)\hat{s}_- - \frac{i\tilde{\gamma}NM_s}{2}\,\hat{s}_+, \\ \frac{d\,\hat{s}_+}{dt} &= i\tilde{\gamma}^*\left(H_{\rm ex} - DM_s\nabla^2 + \frac{NM_s}{2}\right)\hat{s}_+ + \frac{i\tilde{\gamma}^*NM_s}{2}\,\hat{s}_-, \end{aligned} \tag{19}$$

where $\tilde{\gamma} = \gamma - i\lambda M_s$ is the same complexified gyromagnetic ratio in Eqs. (9). These equations can be derived by the following quadratic Hamiltonian for the fluctuations $\hat{s}_{\pm}(x)$:

$$\begin{aligned}\hat{\mathcal{H}}_{\rm L} &= \tilde{\gamma}\int d^3x\left\{\left(\frac{H_{\rm ex}}{2S_0} + \frac{\gamma N}{4}\right)\hat{s}_+\hat{s}_- + \frac{\gamma D}{2}\nabla\hat{s}_+\nabla\hat{s}_- \right.\\ &\quad \left. + \frac{\gamma N}{8}(\hat{s}_+^2 + \hat{s}_-^2)\right\}, \end{aligned} \tag{20}$$

with the linearized spin algebra $[\hat{s}_+(x), \hat{s}_-(x')] = -2S_0\,\delta^3(x - x')$. For the nonuniform modes, we introduce the linearized HP representation [1,2],

$$\begin{aligned}\hat{S}_-(x) &= \hat{s}_-(x) = \sqrt{\frac{2S_0}{V}}\sum_k e^{ikx}\hat{a}_k, \\ \hat{S}_+(x) &= \hat{s}_+(x) = \sqrt{\frac{2S_0}{V}}\sum_k e^{-ikx}\hat{a}_k^\dagger, \\ \hat{S}_z(x) &= -S_0 + \frac{\sum_{k,k'} e^{i(k-k')x}\hat{a}_{k'}^\dagger\hat{a}_k}{V}, \end{aligned} \tag{21}$$

where the mode operators satisfy the commutation relation $[\hat{a}_k, \hat{a}_{k'}^\dagger] = \delta_{k,k'}$. Inserting this expression into the linearized Hamiltonian Eq. (20), we can obtain the quadratic Hamiltonian for the nonuniform mode operators, $\hat{a}_k$ and $\hat{a}_k^\dagger$, and diagonalize for each mode of spin waves by the Bogoliubov transformation similar to the uniform case, which is concretely discussed in the next section.

IV. TWO-MODE CORRELATION OF MAGNONS IN FM THIN FILMS

In this section, we discuss the coupled dynamics of multiple spin wave modes, which are almost degenerate, in small FM thin films. For the short wavelength (and high-frequency) region, where the finite-size effect can be neglected, the mode expansions of the spin variables are generally given by the standard plane-wave form Eq. (21). In the following discussion, we focus on the low-energy magnetization dynamics, and assume that the low-energy dynamics is governed by several low-frequency modes with the long wavelength, such as the uniform mode ($k = 0$) and the long wavelength modes with $k \sim 0$.[4]

For concreteness and simplicity, we consider the coupled dynamics of two modes, i.e., the uniform mode $\hat{a}$ and the second-lowest energy mode, $\hat{b} \equiv \hat{a}_k$ with $k \sim 0$, which are almost degenerate. Using the operators $\hat{a}$ and $\hat{b}$, we approximate the linearized spin densities in the eigenmode expansion,

$$\begin{aligned}\hat{s}_-(x) &= \sqrt{\frac{2S_0}{V}}(\hat{a} + f_k(x)\,\hat{a}_k + f_q(x)\,\hat{a}_q + \cdots) \\ &\simeq \sqrt{\frac{2S_0}{V}}(\hat{a} + f_k(x)\,\hat{b}), \\ \hat{s}_+(x) &= \sqrt{\frac{2S_0}{V}}(\hat{a}^\dagger + f_k^*(x)\,\hat{a}_k^\dagger + f_q^*(x)\,\hat{a}_q^\dagger + \cdots) \\ &\simeq \sqrt{\frac{2S_0}{V}}(\hat{a}^\dagger + f_k^*(x)\,\hat{b}^\dagger), \end{aligned} \tag{22}$$

where $f_{k(q)}(x)$ is the mode function of the eigenmode of the spin-wave equations Eqs. (19), and the mode operators satisfy the following commutation relations:

$$[\hat{a}, \hat{a}^\dagger] = 1, \quad [\hat{b}, \hat{b}^\dagger] = 1, \quad [\hat{a}, \hat{b}] = [\hat{a}, \hat{b}^\dagger] = 0. \tag{23}$$

The extension to the multimode dynamics including several long wavelength modes, such as the third-lowest mode $\hat{a}_q$, is straightforward and the results are almost similar. It is worth noting that the mode functions $f_{k(q)}(x)$ of the long wavelength eigenmodes depend on the details of the sample such as the geometry, bulk inhomogeneity, and boundary conditions, and do not have a simple plane-wave form in general.

Inserting the two-mode expression Eqs. (22) into the linearized Hamiltonian Eq. (20), we obtain the quadratic two-mode Hamiltonian:

$$\hat{\mathcal{H}}_{\rm tot} = \hat{\mathcal{H}}_a + \hat{\mathcal{H}}_b + \hat{\mathcal{H}}_{\rm mix}, \tag{24}$$

$$\hat{\mathcal{H}}_a = \tilde{\gamma}\left\{\left(H_{\rm ex} + \frac{NM_s}{2}\right)\hat{a}^\dagger\hat{a} + \frac{NM_s}{4}(\hat{a}^{\dagger 2} + \hat{a}^2)\right\}, \tag{25}$$

$$\begin{aligned}\hat{\mathcal{H}}_b &= \tilde{\gamma}\int\frac{d^3x}{V}\left\{\left(H_{\rm ex} + \frac{NM_s}{2}\right)|f_k|^2\,\hat{b}^\dagger\hat{b} \right.\\ &\quad \left. + DM_s\,|\nabla f_k|^2\,\hat{b}^\dagger\hat{b} + \frac{NM_s}{4}\left(f_k^{*2}\,\hat{b}^{\dagger 2} + f_k^2\,\hat{b}^2\right)\right\}, \end{aligned} \tag{26}$$

$$\begin{aligned}\hat{\mathcal{H}}_{\rm mix} &= \tilde{\gamma}\left\{\left(H_{\rm ex} + \frac{NM_s}{2}\right)(F_k\,\hat{a}^\dagger\hat{b} + F_k^*\,\hat{b}^\dagger\hat{a}) \right.\\ &\quad \left. + \frac{NM_s}{2}(F_k^*\,\hat{a}^\dagger\hat{b}^\dagger + F_k\,\hat{a}\,\hat{b})\right\}, \end{aligned} \tag{27}$$

[4]For an example, such modes appear as low-lying modes in spin wave resonance in thin films [19,20].

where $F_k = \int d^3x\, f_k(x)/V$ gives the form factor which has been discussed in the context of a two-magnon scattering process [21].

Here, we assume the real mode function $f_k(x) = f_k^*(x)$ for simplicity, and normalize the mode function as

$$\int \frac{d^3x}{V}\, |f_k(x)|^2 = \int \frac{d^3x}{V}\, f_k^2(x) = 1, \tag{28}$$

which is consistent with the linearized spin commutator $[\hat{s}_+(x), \hat{s}_-(x')] = -2S_0\, \delta^3(x - x')$. We also define the wave number of the $\hat{b}$ mode:

$$k^2 = \int \frac{d^3x}{V}\, |\nabla f_k(x)|^2. \tag{29}$$

Then, the Hamiltonian of the nonuniform mode, $\hat{\mathcal{H}}_b$, can be diagonalized by the Bogoliubov transformation,

$$\hat{d} = \alpha\, \hat{b} + \beta\, \hat{b}^\dagger \quad \text{with} \quad |\alpha|^2 - |\beta|^2 = 1, \tag{30}$$

where the coefficients are given by

$$\alpha = \cosh s, \quad \beta = \sinh s, \\ \text{with} \quad e^s = \left(\frac{H_{\text{ex}} + NM_s + DM_s k^2}{H_{\text{ex}} + DM_s k^2} \right)^{1/4}. \tag{31}$$

The diagonalized Hamiltonian becomes

$$\hat{\mathcal{H}}_b = \tilde{\gamma}\, \omega_k \big(\hat{d}^\dagger \hat{d} + \tfrac{1}{2}\big), \\ \text{with } \omega_k = \sqrt{(H_{\text{ex}} + DM_s k^2)(H_{\text{ex}} + NM_s + DM_s k^2)}. \tag{32}$$

For the sufficiently long wavelength mode ($k \sim 0$), the resonance frequency ω_k is almost degenerate with the frequency of the uniform mode ω_0:

$$\omega_k \simeq \omega_0 + \cosh 2r\, (DM_s k^2). \tag{33}$$

As for the mode-mixing Hamiltonian $\hat{\mathcal{H}}_{\text{mix}}$, we assume that the form factor is nonvanishing, $F_k = F_k^* \simeq O(1)\, (\neq 0)$. The physical origin of the nonvanishing form factor is discussed below. Using the form factor F_k, we can rewrite the mixing Hamiltonian in terms of the diagonalizing operators, $\hat{c}$ and $\hat{d}$:

$$\hat{\mathcal{H}}_{\text{mix}} = F_k\, \tilde{\gamma}\, \omega_0[\cosh(r - s)(\hat{c}^\dagger \hat{d} + \hat{d}^\dagger \hat{c}) \\ + \sinh(r - s)(\hat{c}^\dagger \hat{d}^\dagger + \hat{d}\, \hat{c})], \tag{34}$$

where the Bogoliubov coefficients in Eqs. (15) and (31) have been used. In the small k limit, the coefficients are given by

$$\cosh(r - s) \simeq 1 + O\left(\frac{D^2 M_s^2 k^4}{\omega_0^2} \right), \\ \sinh(r - s) \simeq \frac{\sinh 2r}{2\omega_0}(DM_s k^2). \tag{35}$$

For the first approximation, we consider the degeneration limit with $\omega_k \simeq \omega_0$, which corresponds to $k \simeq 0$, and approximate the coefficients in $\hat{\mathcal{H}}_{\text{mix}}$ as $\cosh(r-s) \simeq 1$ and $\sinh(r-s) \simeq 0$. In this limit, we obtain the total two-mode Hamiltonian:

$$\hat{\mathcal{H}}_{\text{tot}} = \tilde{\gamma}\, \omega_0[\hat{c}^\dagger \hat{c} + \hat{d}^\dagger \hat{d} + F_k(\hat{c}^\dagger \hat{d} + \hat{d}^\dagger \hat{c})] \\ = \tilde{\gamma}\, \omega_0\big[(\hat{c}^\dagger + F_k\, \hat{d}^\dagger)(\hat{c} + F_k\, \hat{d}) + \big(1 - F_k^2\big)\hat{d}^\dagger \hat{d}\big], \tag{36}$$

with the complexified gyromagnetic ratio $\tilde{\gamma} = \gamma - i\lambda M_s$. Here, we assume that the uniform $\hat{c}$ mode and the nonuniform $\hat{d}$ mode can be selectively excited by external pumping.

This two-mode Hamiltonian leads to the following Heisenberg equations of motion for the mode operators $\hat{c}$ and $\hat{d}$:

$$i\frac{d}{dt}\, \hat{c} = [\hat{c},\, \hat{\mathcal{H}}_{\text{tot}}] = \tilde{\gamma}\, \omega_0\, (\hat{c} + F_k\, \hat{d}), \tag{37}$$

$$i\frac{d}{dt}\, \hat{d} = [\hat{d},\, \hat{\mathcal{H}}_{\text{tot}}] \\ = F_k\, \tilde{\gamma}\, \omega_0\, (\hat{c} + F_k\, \hat{d}) + \big(1 - F_k^2\big)\tilde{\gamma}\, \omega_0\, \hat{d}. \tag{38}$$

Introducing the normalized quantum states $|\psi\rangle_c \otimes |\phi\rangle_d$, where $|\psi\rangle_c$ and $|\phi\rangle_d$ are the elements in the Fock spaces of $\hat{c}$ and $\hat{d}$, respectively, the temporal evolution of the expectation value of each mode operator can be derived:

$$\frac{d}{dt}\, \langle c \rangle = -i\, \gamma\, \omega_0\, (\langle c \rangle + F_k\, \langle d \rangle) - \lambda\, M_s \omega_0\, (\langle c \rangle + F_k\, \langle d \rangle), \tag{39}$$

$$\frac{d}{dt}\, \langle d \rangle = -i\, F_k\, \gamma\, \omega_0\, (\langle c \rangle + F_k\, \langle d \rangle) \\ - F_k \lambda\, M_s \omega_0\, (\langle c \rangle + F_k\, \langle d \rangle) \\ - i\big(1 - F_k^2\big)\gamma\, \omega_0\, \langle d \rangle - \big(1 - F_k^2\big)\lambda\, M_s \omega_0\, \langle d \rangle. \tag{40}$$

Although we consider the expectation values for pure states (such as products of coherent states) here, the temporal evolution of more general expectation values including (statistical) mixed states using density matrix operators is discussed in the next section.

In the following discussion, the form factor is assumed to be $F_k \simeq 1$, for simplicity. In this case, these equations imply the existence of magnetization dynamics with two different timescales. One is the (coalesced) local magnetization sum governed by the usual Gilbert damping term $\propto \lambda$,

$$\frac{d}{dt}(\langle c \rangle + F_k\, \langle d \rangle) \simeq -i\big(1 + F_k^2\big)\gamma\, \omega_0\, (\langle c \rangle + F_k\, \langle d \rangle) \\ - \big(1 + F_k^2\big)\lambda\, M_s \omega_0\, (\langle c \rangle + F_k\, \langle d \rangle), \tag{41}$$

where the expectation value (or amplitude) of transverse magnetization is given by

$$\int \frac{d^3x}{V}\, \langle m_-(x) \rangle \;\propto\; \langle a \rangle + F_k \langle b \rangle \\ = \cosh r\, (\langle c \rangle + F_k \langle d \rangle) + \sinh r\, (\langle c^\dagger \rangle + F_k \langle d^\dagger \rangle). \tag{42}$$

The other is relatively slow dynamics[5] governed by a reduced damping constant $\lambda \to \lambda_r = (1 - F_k^2)\, \lambda$:

$$\frac{d}{dt}\langle d \rangle \simeq -i\big(1 - F_k^2\big)\gamma\, \omega_0\, \langle d \rangle - \big(1 - F_k^2\big)\lambda\, M_s \omega_0\, \langle d \rangle. \tag{43}$$

From the temporal evolutions with two different timescales, we can conclude that, even though the coalesced magnetization undergoes the relaxation by the standard Gilbert

[5]A small detuning effect, $\delta\omega_k = \omega_k - \omega_0 \neq 0$, also leads to similar slow dynamics with a reduced damping constant, $\lambda' = \delta\omega_k \lambda$.

damping, $\langle m_\pm \rangle \to 0$, relative correlation between $\langle c \rangle$ and $\langle d \rangle$ does not need to vanish and survives in an equilibrium state with finite-temperature thermal fluctuations, within a different timescale specified by the reduced damping constant, $\lambda_r = (1 - F_k^2)\lambda \ll \lambda$. More explicitly, due to the commutativity, $[\hat{c} - \hat{d}/F_k,\ \hat{c}^\dagger + F_k \hat{d}^\dagger] = 0$, two orthogonal modes, one is the coalesced mode and the other is the depolarized mode, can have the distinct temporal evolution: $\langle c \rangle + F_k \langle d \rangle$ and $\langle c \rangle - \langle d \rangle / F_k$ exhibit relaxation with different timescales. Note that such an intermode phase correlation does not alter the energy of the system as long as $\langle m_\pm \rangle = 0$.

This is a two-mode example of the spontaneous intermode depolarization correlation (DPC) of magnons in the small FM film,

$$\langle m_k^\pm \rangle \neq 0 \quad \text{with} \ \langle m_\pm \rangle = \sum_k \langle m_k^\pm \rangle \simeq 0, \tag{44}$$

where $\langle m_k^\pm \rangle$ is each mode contribution to local magnetization and the expectation value is defined as the trace over the (statistical) mixed states of magnon modes in quantum statistical mechanics. If we can access the individual dynamics of each mode, some information for the states of each mode can be extracted and manipulated from the DPC. Here, we note that the expectation value (or amplitude) of each mode should be reduced separately through the microscopic spin relaxations [6–9] and the DPC vanishes after the spin relaxation time τ_S.

The above argument for the DPC can be applicable for more general situations other than the original setup discussed in Sec. II. For example, two almost degenerate coherent excitations of different modes (for $\hat{c}$ and $\hat{d}$), which are prepared via multimode excitation or nonlinear magnon processes, can similarly form the DPC after the Gilbert damping.

It should be noted that, although the two-mode DPC has been discussed so far in terms of the uniform mode and the second-lowest energy mode of the spin wave, the discussion above can be straightforwardly generalized to the cases between the various types of spin-wave modes.

To summarize, we found that the depolarization correlation among the almost degenerate magnon modes can be transiently formed in the small FM samples due to the intermode mixing, which has been overlooked in macrospin model analysis. As discussed below, the Gilbert damping and the inhomogeneous demagnetizing fields (or dipolar fields), which originate from the edge effects and nonlinear magnetization dynamics in finite-size samples, actually give rise to the intermode mixing resulting from energy and momentum nonconservation.

Origin of nonvanishing form factor

In this subsection, we discuss the possible origin of the nonvanishing form factor F_k in Eq. (27). In the case possessing (discrete) translational invariance, where the wave number k is a good quantum number, i.e., a conserved quantity, the intermode mixings between different k are forbidden and the form factor F_k should vanish.

However, as discussed in Sec. II, we consider small FM films (or dots), where the translational invariance is generically broken by the boundary conditions, and thus the momentum conservation is violated in the order of $k \sim 1/L$ (L is a typical size of the sample). Furthermore, in such small FM media, we cannot ignore various finite-size effects in all directions of the film, such as the inhomogeneous demagnetizing field in the film, surface anisotropy near the edge, and so on [1,2]. These effects generally induce the inhomogeneous background magnetization, $\langle M_z \rangle = M_z(x) \neq \text{const}$ [20,22,23], and this leads to an inhomogeneous effective magnetic field for $\hat{m}_\pm$ (or magnons) inside the film.[6] When considering the excitation of magnon modes by the external pumping, nonlinear magnetization effects come to the surface and also generate an inhomogeneous effective magnetic field through the nontrivial mean field $\langle m_\pm(x) \rangle \neq \text{const}$. All these finite-size effects violate the conservation of wave number, and the mode function $f_k(x)$ does not become a simple plane-wave form in the small k region.

In addition, as discussed so far, the relaxation process is crucial for the DPC to correlate different frequency modes, and thus we should incorporate the effects of the Gilbert damping term $\propto \lambda$. The damping term causes the relaxation of not only energy but also momentum of magnons and the effects also violate the mode orthogonality: As discussed in Refs. [24,25], dispersion relations derived from wave equations with the damping term generally imply the complex frequency (with $\mathrm{Im}\,\omega \propto \lambda$) and the complex wave-number (with $\mathrm{Im}\,k \propto \lambda$).

Taking into account the finite-size effects and relaxation effects, the wave number is neither a good quantum number nor a conserved quantity in realistic small FM samples, and therefore we expect a nonvanishing form factor, $F_k \neq 0$. Note that similar form factors originated from crystal imperfections and pores in FM samples have been discussed in the context of two-magnon scatterings [1,21].

V. INTERMODE DPC BASED ON DENSITY MATRIX OPERATOR

In this section, we investigate the dynamics of the two-mode DPC of magnons using the simple model Eq. (36) by a quantum-optics-like approach [26]. The two-mode Hamiltonian leads to the Schrödinger equation for the quantum states $|\Psi\rangle = |\psi\rangle_c \otimes |\phi\rangle_d$:

$$\begin{aligned} i\frac{\partial}{\partial t}|\Psi\rangle &= \hat{\mathcal{H}}_{\text{tot}}|\Psi\rangle \\ &= \tilde{\gamma}\,\omega_0\big[(\hat{c}^\dagger + F_k\,\hat{d}^\dagger)(\hat{c} + F_k\,\hat{d}) + \big(1 - F_k^2\big)\hat{d}^\dagger\hat{d}\big]|\Psi\rangle \\ &\simeq \tilde{\gamma}\,\omega_0(\hat{c}^\dagger + F_k\,\hat{d}^\dagger)(\hat{c} + F_k\,\hat{d})|\Psi\rangle. \end{aligned} \tag{45}$$

In the following discussion, we focus on the fast dynamics and ignore the slow dynamics, and thus we set the vanishing slowing factor, $(1 - F_k^2) \simeq 0$, in the last line. Note that the complex coefficient $\tilde{\gamma} = \gamma - i\lambda M_s$ in $\hat{\mathcal{H}}_{\text{tot}}$ gives the non-Hermitian Hamiltonian, which describes magnetization dynamics involving the Gilbert damping, and the norm of states is not conserved under the nonunitary evolution given by Eq. (45). For such a non-Hermitian Hamiltonian, the

[6] In such cases, the form factor F_k should be generalized to the integral form including the inhomogeneities $M_z(x)$ and $H_{\text{eff}}(x)$, such as $\int d^3x\, M_z(x) f_k(x)/V$ in Eq. (27).

phenomenological Schrödinger equation for quantum mechanics with energy relaxation (or dissipation) has been discussed in the description of damped harmonic oscillators and magnetization dynamics [27–29] and in the Monte Carlo wave-function approach in quantum optics [26,30,31].

Following the phenomenological approach, we consider the norm-preserving Schrödinger equation for normalized states ($\langle\Psi|\Psi\rangle = 1$) instead of Eq. (45),

$$i\frac{\partial}{\partial t}|\Psi\rangle = \gamma\,\omega_0\,\hat{\mathcal{N}}\,|\Psi\rangle - i\,\lambda\,M_s\,\omega_0(\hat{\mathcal{N}} - \langle\mathcal{N}\rangle)|\Psi\rangle, \tag{46}$$

with $\hat{\mathcal{N}} = (\hat{c}^\dagger + F_k\,\hat{d}^\dagger)(\hat{c} + F_k\,\hat{d})$ and $\langle\mathcal{N}\rangle = \langle\Psi|\hat{\mathcal{N}}|\Psi\rangle$. From the phenomenological Schrödinger equation, the temporal evolution of the density matrix operator $\hat{\rho} = \sum_i p_i|\Psi_i\rangle\langle\Psi_i|$ is given by the following master equation which involves energy relaxation [27–29,32]:

$$\frac{d\hat{\rho}}{dt} = i\,\gamma\,\omega_0[\hat{\rho},\,\hat{\mathcal{N}}] - \lambda\,M_s\,\omega_0(\hat{\mathcal{N}}\,\hat{\rho} + \hat{\rho}\,\hat{\mathcal{N}} - 2\,\langle\mathcal{N}\rangle\,\hat{\rho}), \tag{47}$$

with $\langle\mathcal{N}\rangle = \mathrm{tr}(\hat{\mathcal{N}}\hat{\rho})$.

We can find the equilibrium state solution ($d\hat{\rho}/dt = 0$) of the master equation from the conditions

$$(\hat{c} + F_k\,\hat{d})\,\hat{\rho} = 0 \quad \text{and} \quad \hat{\rho}\,(\hat{c}^\dagger + F_k\,\hat{d}^\dagger) = 0. \tag{48}$$

The resulting equilibrium state is given by a product of the coherent states [33,34],

$$\hat{\rho}_{\mathrm{eq}} = (|C\rangle \otimes |D\rangle)(\langle C| \otimes \langle D|), \quad \text{with} \quad \hat{c}\,|C\rangle = C\,|C\rangle \quad \text{and} \quad \hat{d}\,|D\rangle = D\,|D\rangle, \tag{49}$$

whose eigenvalues are constrained by

$$C + F_k\,D = 0. \tag{50}$$

As discussed in Sec. IV, if the depolarization operator, $\hat{c} - \hat{d}/F_k$, initially has nonvanishing expectation value,

$$\langle c\rangle - \frac{\langle d\rangle}{F_k} = R, \tag{51}$$

this expectation value is conserved under the temporal evolution given by Eq. (47) in the absence of microscopic relaxation. Using Eqs. (50) and (51), the expectation values are given in the equilibrium state:

$$C = -F_k\,D = \left(\frac{F_k^2}{1 + F_k^2}\right)R. \tag{52}$$

In the original setup discussed in Sec. II, the initial state (or density matrix) corresponds to the mixed state of a coherent state of the $\hat{c}$ mode, which is excited by the external pumping, and a thermal state of the $\hat{d}$ mode: $\hat{\rho}_{\mathrm{ini}} = {}_c|\alpha\rangle\langle\alpha|_c \otimes \hat{\rho}_d^{\mathrm{th}}$. In this case, the expectation value of the depolarization mode is given by the eigenvalue of the initial coherent state of the $\hat{c}$ mode, i.e., $R = \alpha$. For another initial state with two coherent states, $\hat{\rho}_{\mathrm{ini}} = {}_c|\alpha\rangle\langle\alpha|_c \otimes {}_d|\beta\rangle\langle\beta|_d$, the expectation value of the depolarization mode is given by $R = \alpha - \beta/F_k$, which corresponds to the difference between the amplitudes of the coherent states.

The state Eqs. (49) with Eq. (50) shows the intermode DPC between the $\hat{c}$ and $\hat{d}$ modes with the relation

$$\langle c\rangle = -F_k\langle d\rangle. \tag{53}$$

Actually, the intermode DPC is a transient state and, including the effects of the detuning ($\delta\omega_k \neq 0$), nonvanishing slowing factor ($1 - F_k^2 \neq 0$), and microscopic spin relaxation, both amplitudes of the expectation values are relaxed eventually.

VI. SUMMARY

We calculated the intermode correlation of magnons in small FM films with finite area. The result shows a possibility of intermode DPC among magnon modes in an equilibrium state. With the assumptions of the existence of almost degenerate magnon modes and the nontrivial mixing among the modes due to the demagnetizing fields and damping effects, we have shown that the intermode DPC can be realized as a transient state. Although our discussion is based on the linearized analysis of a continuum model of FM films, the analysis of the magnon DPC in other approaches, such as micromagnetic simulation [3] and the stochastic LLG approach [14–17], is also interesting and will be discussed in a future work.

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