

## Coherent Radiation in A Very Thin Ferromagnetic Film

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Relaxation of magnon in a very thin ferromagnetic film through spontaneous emission of photon shows an enhancement of the decay rate due to the phase coherence between the magnon and the planar component of wave vector of photon. The coupling between magnon and photon under a strong external magnetic field is considered only at the lowest order one-magnon one-photon process, which we believe the most dominant channel for the radiation from the system. Theoretical understanding related to the geometric confinement is pursued; the phase coherence due to the crystal symmetry in the film plane gives rise to superradiative emission on one hand, but the symmetry breaking along the direction perpendicular to the film renders the possibility of emission itself, providing the increased degrees of freedom for the photon.

**Key words :** coherent radiation, magnon-photon interaction, superradiance, magnetic film

### 1. Introduction

Relaxation of magnetic excitation in magnetic systems has been investigated for a long time [1, 2]. Spin waves are coupled with various types of mechanisms as dipole-dipole interaction [3], impurity potential and surface roughness [4], magnon-phonon coupling [5], and/or magnon-photon coupling [6, 7]. These channels are usually associated with multi-magnon scattering processes, involving more than two magnon operators [8]. However, we can think of lower order of interaction for a magnon decaying into a photon only through one-magnon one-photon process, which is generally stronger than multi-channel scattering processes. The reason that the previous works ignore this process is that the interaction potential composed of one-magnon one-photon operators is simply diagonalized to form new eigenstates, therefore preventing the system from radiation: Magnons coupled with photons in ferromagnetic and antiferromagnetic materials form a hybrid of the quanta in the usual eigenvalue problem, in which the reflectivity spectrum manifests the details of the coupled dispersion curves. [9] Dispersion relation of the magnon-photon coupled system including the phonon involvement is also well known for ferromagnetic insulators. [10] However, the existence of new eigen modes due to

the couplings naturally restricts the possibility of transition between the quanta, but the excitation energy is trapped. This effect is just an analog of non-radiation of exciton in bulk semiconductors already pointed out by Hopfield [11], where the formation of polariton is suggested. Physical origin of the diagonalization is due to the periodic boundary condition in bulk, that is, perfect translational symmetry. Formation of a new eigen mode limits to have an increase of degrees of freedom, so that the emission of excitation energy is highly restricted. If there could be any decay process in bulk, there must be corresponding channel for breaking of the symmetry [7]. Recently, an induced radiation damping in ferromagnetic resonance situation is suggested by introducing complex involvement of heat production of lattice motion [12].

More effective and active means of symmetry breaking is possible when we artificially confine excitations in the restricted geometry of the sample. As a matter of fact, conspicuous growth of fine technology for fabricating nano-scale specimens for the magnetic devices already provides the practical environment to study the radiation from magnetic excitation of confined systems. Though the experimental advances are enormous these days in the field of spintronics and magnetic storage devices of the form of very thin film, theoretical understanding for the relaxation mechanism of these systems is not profound yet, since the exchange related interactions are coupled with various channels in a complicated way in general,

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especially when non-bulk geometry is involved. On the other hand, relaxation of exciton in the geometrically confined systems is well known, and further the radiation is coherent, in that the phases of exciton and photon are well matched to enhance the radiation rate and frequency shift [13, 14]. This effect is observed in semiconductor systems of films, quantum wires and dots, or heterostructures [15]. But, the original introduction of the coherence and the superradiance in the radiation is indebted to Dicke for the atomic gas system [16]. Magnon-photon coupling in thin magnetic film under an external field is also investigated [17, 18], giving rise to circularly polarized photon modes with energy gap, but the radiation mechanism is not clear.

We consider in the present study the coherent decay of magnetic excitation in a very thin ferromagnetic film when an external field is applied. The film as a structure with broken symmetry is expected to have a good eligibility of emission itself, but further the symmetry remained on the film assures the phase coherence between the wave vector of two-dimensional magnon and the planar component of the wave vector of a photon. The coherence gives rise to an enhancement of radiation features, the decay rate and the frequency shift of the radiation spectrum. In section 2, we will derive the interaction Hamiltonian for the coupling of two-dimensional magnon and photon propagating in an arbitrary direction as well as unperturbed Hamiltonian for two-dimensional magnetic energy and for the usual photon term. Decay rate in the very thin ferromagnetic film is calculated in section 3, and the enhancement of the decay rate is emphasized. Section 4 is devoted to discussion related to the role of coherence in this system.

## 2. Interaction Hamiltonian

We consider a ferromagnetic film on xy-plane, and a uniform external field is applied along y-axis, which is one of the easy axes for the magnetic crystal. The elementary magnetic excitation in the film, the magnon or the spin wave, propagates along the plane, so that the spin wave is described by two-dimensional (2D) wave vectors. Once a magnon exists in the film, the excitation energy is supposed to be released by emitting a photon. We are going to find expressions for 2D magnon and 3D photon terms along with the interaction Hamiltonian in this section. Since the spontaneous decay of the magnon conserves energy in the long wavelength range during the process of emitting photons, as will be discussed later, the system may be treated as a continuum of magnetic moments [1, 2]. The magnetization of the film is written

in terms of 2D magnon creation and annihilation operators,  $a_k^+$  and  $a_k$ , by introducing the Holstein-Primakoff transformation. Components of the magnetization are written as

$$\begin{aligned} M_x(\boldsymbol{\rho}) &= \sqrt{\frac{\mu_m M_s}{\Omega}} \sum_{\mathbf{k}} (a_{\mathbf{k}} + a_{-\mathbf{k}}^+) e^{i\mathbf{k} \cdot \boldsymbol{\rho}} \\ M_z(\boldsymbol{\rho}) &= i \sqrt{\frac{\mu_m M_s}{\Omega}} \sum_{\mathbf{k}} (a_{-\mathbf{k}}^+ - a_{\mathbf{k}}) e^{i\mathbf{k} \cdot \boldsymbol{\rho}} \\ M_y &\approx M_s - \frac{2\mu_m}{\Omega} \sum_{\mathbf{k}\mathbf{k}'} a_{\mathbf{k}}^+ a_{\mathbf{k}'}, \end{aligned} \quad (1)$$

where  $\boldsymbol{\rho}$  and  $\mathbf{k}$  are 2D position vector and wave vector in the plane respectively,  $\mu_m = g\mu_B/2$  is magnetic moment in a unit cell with  $g$  the gyromagnetic ratio and  $\mu_B$  the Bohr magneton. Saturation magnetization  $M_s$  and the area of the film  $\Omega$  guarantee the commutation relation between the magnon operators,  $[a_{\mathbf{k}}, a_{\mathbf{k}'}^+] = \delta_{\mathbf{k}, \mathbf{k}'}$ . It is assumed that the external field is strong enough to align spins almost in one direction.

In the macroscopic formalism, the exchange energy is expressed phenomenologically by adopting the Landau free energy

$$H_{ex} = \int \frac{A}{M_s} \{ (\nabla M_x)^2 + (\nabla M_z)^2 \} d\boldsymbol{\rho}, \quad (2)$$

where only the first order for magnetization components is kept.  $A$  is the macroscopic exchange energy constant, which is known to be proportional to the microscopic exchange overlap integral  $J$ ;  $A \propto JS^2$  with  $S$  the spin quantum number. The second quantization is elementary and gives rise to the 2D exchange Hamiltonian;

$$H_{ex} = \frac{4A\mu_m}{M_s} \sum_{\mathbf{k}} \left( \frac{1}{2} + a_{\mathbf{k}}^+ a_{\mathbf{k}} \right) k^2. \quad (3)$$

Note that the quadratic behavior is only due to the usage of continuum model, and is consistent with the long wavelength limit of the microscopic results. The Zeeman energy for the applied field is similarly obtained

$$H_z = \frac{H_{ext}}{2M_s} \int \{ (M_x)^2 + (M_z)^2 \} d\boldsymbol{\rho} = 2H_{ext}\mu_m \sum_{\mathbf{k}} \left( \frac{1}{2} + a_{\mathbf{k}}^+ a_{\mathbf{k}} \right), \quad (4)$$

where the y-component is omitted since it is a constant in the first order.  $H_{ext}$  is the magnitude of applied magnetic field.

When magnetic energy is transferred to electromagnetic wave, it might not necessarily be restricted to propagate in the film plane. The electromagnetic wave is described in terms of usual 3D creation and annihilation photon operators,  $b_{\mathbf{Q}\lambda}^+$  and  $b_{\mathbf{Q}\lambda}$ , which satisfy the commutation relation

$[b_{Q\lambda}, b_{Q\lambda'}^\dagger] = \delta_{\lambda,\lambda'} \delta_{Q,Q'}$ , where  $Q$  is 3D photon wave vector and  $\lambda$  is the index for two transverse modes of photon polarization. The vector field can be written in SI units as

$$\mathbf{A}(\mathbf{r}, t) = \frac{c\sqrt{\hbar\mu_0}}{(2\pi)^{3/2}\sqrt{V}} \sum_{Q\lambda} \hat{\varepsilon}_{Q\lambda} \frac{1}{\sqrt{2\omega_Q}} \{b_{Q\lambda} + b_{Q\lambda}^\dagger\} e^{iQ\cdot\mathbf{r}}, \quad (5)$$

where  $\hat{\varepsilon}_{Q\lambda}$  is the unit vector for the polarization and  $\mu_0$  is the permeability of free space. Photon part of the Hamiltonian is written as usual

$$H_p = \sum_{q q_z \lambda} \hbar c \sqrt{q^2 + q_z^2} \left( b_{q q_z \lambda}^\dagger b_{q q_z \lambda} + \frac{1}{2} \right), \quad (6)$$

where the wave vector is decomposed of 2D component  $q$  and the perpendicular component to the film plane  $q_z$ .

Among the various channels for the magnon relaxation processes, we assume that the spontaneous decay of magnon by the interaction

$$H_{mp} = -\int d\mathbf{r} \mathbf{M}(\boldsymbol{\rho}) \cdot \mathbf{B}(\mathbf{r}) \rightarrow -\int d\mathbf{r} \{M_x B_x + M_z B_z\} \quad (7)$$

is the most important for the case that spins are almost aligned along one direction. As a matter of fact, magnons can be scattered by emitting photons, phonons, or heat, but these multiple magnon processes conserving energy and momentum has small transition probability, comparing with the process that doesn't conserve momentum. Especially, it is very interesting that the momentum non-conserving radiation possesses of coherence during the transition. It is the purpose of the present study to elucidate the role of coherent radiation through the interaction, eq. (7). We will find the enhanced decay rate of magnon because of the coherence in the next section. The form of the interaction is the mixture of 2D and 3D geometry, therefore, a careful consideration for the spatial integral is required: Since the interaction is acting only in the film plane, the integral for the perpendicular coordinate ( $z$ , here) to the film plane is separated, but for a few layers of crystal planes we rewrite the integral as a summation over layers;

$$\int dz e^{iq_z z} \rightarrow na \sum_{n=1} e^{iq_z(n-1)a}, \quad (8)$$

where  $a$  is the lattice constant along  $z$ -direction. For film of one layer the spatial integral becomes

$$\int d\mathbf{r} e^{iQ\cdot\mathbf{r}} e^{ik\cdot\boldsymbol{\rho}} \rightarrow a\Omega \delta_{Q,-k} \quad (9)$$

The simple result gives rise to the conservation of wave vectors between 2D magnon and planar component of

photon, which will render the coherence effect for the decay mechanism of magnon. Now, the interaction Hamiltonian turns out to be

$$H_{mp} = \frac{ia}{(2\pi)^{3/2}} \sqrt{\frac{\Omega}{V}} \sqrt{\mu_0 g \mu_B M_s} \\ \times \sum_{\lambda k q_z} \sqrt{\frac{\hbar\omega_{-kq_z}}{2}} \{ \eta_x (a_k + a_{-k}^\dagger) (b_{\lambda-kq_z} + b_{\lambda k q_z}^\dagger) \\ + i\eta_z (a_{-k}^\dagger - a_k) (b_{\lambda-kq_z} + b_{\lambda k q_z}^\dagger) \} \quad (10)$$

where  $\boldsymbol{\eta} = \hat{Q} \times \hat{\varepsilon}_{\lambda Q}$  with  $\hat{Q}$  the unit vector along the propagation direction of photon. In the resonance approximation of taking the exact energy conservation between magnon and photon, terms are collected only for one-creation one-annihilation process;

$$H_{mp} = \sum_{\lambda k q_z} D_{\lambda k q_z} \{ \eta_{\lambda k q_z} a_k^\dagger b_{\lambda k q_z} + h.c. \}, \\ D_{\lambda k q_z} = \frac{ia}{(2\pi)^{3/2}} \sqrt{\frac{\Omega}{V}} \sqrt{\mu_0 g \mu_B M_s} \sqrt{\frac{\hbar c (k^2 + q_z^2)^{1/2}}{2}}, \quad (11) \\ \eta_{\lambda k q_z} = \eta_x + i\eta_z.$$

### 3. Coherent Decay of Magnon

Relaxation feature of magnetic excitation could be a mixture of complex processes in general as is true for relaxation in atomic gas, or excitonic relaxation in semiconductors. Although the existence of boundary in atomic gas system renders a fundamental possibility of relaxation, the emission itself in a bulk of crystal is very restricted due to a formation of new eigenstates, polariton, which originates from the establishment of the periodic boundary condition. In the actual situation, however, a variety of interaction mechanisms make it possible to emit photons from bulks, e.g., transition between magnon and photon by impurity potential, or the magnon scattering by surface related mechanisms. Geometric confinement of 2D film system is a special example for observing the spontaneous emission of photons, which is strongly related to the increase of degrees of freedom when a 2D magnon takes a transition to 3D photon; a specific 2D magnon is converted to a photon with arbitrary propagation direction.

To study the transition between 2D magnon and the photon, we consider an initial magnon of  $k$ . If the spontaneous radiation is allowed by emitting a photon, the state of the system would be written at later time as

$$|\Psi(t)\rangle = f_o(t) |k; 0\rangle + \sum_{q,s} f_{kq_s}(t) |G; k q_s\rangle, \quad (12)$$

where  $f_o(t)$  represents the probability amplitude that the initial magnon of  $\mathbf{k}$  is found without any photon in the system and  $f_{kq,s}(t)$  is the probability amplitude that the initial magnon disappears but a photon with momentum  $\mathbf{Q}=\mathbf{q}+q_z\hat{k}$  is found. Note that the momentum conservation is already applied for the planar component of photon wave vector, but z-component is arbitrary. The probability amplitudes can be obtained by solving time dependent Schrödinger equation

$$i\hbar \frac{\partial}{\partial t} |\Psi(t)\rangle = H |\Psi(t)\rangle, \quad (13)$$

where

$$H = H_m + H_p + H_{mp} \quad (14)$$

with the magnetic energy including the Zeeman energy

$$H_m = \sum_{\mathbf{k}} B(k) a_{\mathbf{k}}^+ a_{\mathbf{k}}, \quad B(k) = \frac{4A\mu_m}{M_s} k^2 + 2\mu_m H_{ext}. \quad (15)$$

Formalism of Heitler-Ma is used for the present calculation. Substituting eq. (12) into the Schrödinger equation, the probability amplitudes are expressed in the form

$$f_o(t) = -\frac{1}{2\pi i} \int d\omega \frac{e^{i(B(k)/\hbar - \omega)t}}{\omega - B(k)/\hbar + (i/2)\Gamma_k(\omega)} \quad (16)$$

$$f_{kq,s}(t) = -\frac{1}{2\pi i} \int d\omega \frac{(J_{kq,s}^*(\omega)/\hbar) \zeta(\omega - c_s \sqrt{k^2 + q_z^2}) e^{i(c_s \sqrt{k^2 + q_z^2} - \omega)t}}{\omega - B(k)/\hbar + (i/2)\Gamma_k(B(k)/\hbar)}, \quad (17)$$

where

$$\frac{1}{2}\Gamma_k(\omega) \equiv \frac{i}{\hbar} \sum_{kq,s} J_{kq,s} U_{kq,s}(\omega) \zeta(\omega - c_s \sqrt{k^2 + q_z^2}) \quad (18)$$

with  $J_{kq,s} = D_{kq,s} \eta_{kq,s}$  and the zeta function  $\zeta(x)$ . Noticing that major contribution to the integral occurs for  $\Gamma_k(\omega)$  around  $\omega = B(k)/\hbar$ , eq. (16) can be approximated by a simple residue integral

$$f_o(t) = e^{\Gamma_k(B(k)/\hbar)t/2} = e^{-i\Omega_k t} e^{-\gamma_k t/2}. \quad (19)$$

Here, the frequency shift  $\Omega_k$  and the decay rate  $\gamma_k$  for the radiation

$$\begin{aligned} \Omega_k &= \frac{\text{Im}\Gamma_k(B(k)/\hbar)}{2} = P \sum_{q_z,s} \frac{|J_{kq,s}/\hbar|^2}{B(k)/\hbar - c_s \sqrt{k^2 + q_z^2}} \\ &= P \sum_{q_z,s} \frac{|J_{kq,s}/\hbar|^2}{\omega_{kq,s}} \\ \gamma_k &= \frac{\text{Re}\Gamma_k(B(k)/\hbar)}{2} = 2\pi \sum_{q_z,s} |J_{kq,s}/\hbar|^2 \delta(\omega_{kq,s}) \end{aligned} \quad (20)$$

describe the characteristic properties of the spontaneous emission in the present formulation. Here,  $P$  stands for the principal value and  $\omega_{kq,s} = B(k)/\hbar - c \sqrt{k^2 + q_z^2}$ . Energy is conserved when a magnon is decayed by emitting a photon. On the other hand, the radiation field probability amplitude can be approximated for  $t \rightarrow \infty$

$$f_{kq,s}(t \rightarrow \infty) = \frac{J_{kq,s}^*(\omega)/\hbar}{-\omega_{kq,s} + (i/2)\Gamma_k(B(k)/\hbar)}. \quad (21)$$

In this paper we would like to examine the coherence concealed in the decay rate expression. The coupling function in eq. (11) is already written after taking the momentum conservation according to eq. (9); however, if we leave the spatial integral in the decay rate expression, we can write it in the form

$$\gamma_k = \Lambda_k(0) + \sum_{\mathbf{l} \neq 0} e^{-i\mathbf{k} \cdot \mathbf{l}} \Lambda_k(\mathbf{l}), \quad (22)$$

$$\Lambda_k(\mathbf{l}) = \sum_{\mathbf{q}} G_k(\mathbf{q}) e^{i\mathbf{q} \cdot \mathbf{l}} \quad (23)$$

where  $\mathbf{l}$  is the separation vector between two spin sites, and

$$G_k(\mathbf{q}) = \sum_{q_z} \frac{2\pi a^2}{\hbar^2} \left[ \frac{\mu_o g \mu_B M_s \Omega}{(2\pi)^3 V} \right] \frac{\hbar c \sqrt{q^2 + q_z^2}}{2} \delta(\omega_{qq,k}). \quad (24)$$

Here,  $|G_k(\mathbf{q})|^{1/2}$  represents magnitude of the contribution of magnetic moment in the collective mode of  $\mathbf{k}$  magnon to the radiation amplitude, as the magnon is coupled to photon of fixed planar wave vector of  $\mathbf{q}$  but arbitrary z-component  $q_z$ .  $G_k(\mathbf{q})$  is almost independent of magnon mode of  $\mathbf{k}$ , so we can approximate it as  $G_k(\mathbf{q}) \approx G(\mathbf{q})$  therefore, the decay rate, now taking account of the momentum conservation for the spatial integral, becomes

$$\gamma_k \approx \sum_{\mathbf{q}} G(\mathbf{q}) N \delta_{\mathbf{q},\mathbf{k}} = N G(\mathbf{k}). \quad (25)$$

We notice an important fact concerning the coherence involved here: the magnon amplitude at each site is exactly in phase with the corresponding photon amplitude at that site. The contributions from all sites to the radiation field amplitude add up coherently, from which the factor  $N$  comes about. On the other hand, in the limit of isolated spin, neglecting the second term in eq. (22) by cancellation due to off-phase contribution between spins at different sites, the magnon would couple not only to various  $q_z$ , but also to all possible  $\mathbf{q}$ . According to the energy conserving delta-function,  $\mathbf{q}$  cannot exceed  $k_o$ , which is determined by  $B(k_o) = c\hbar \sqrt{k_o^2 + q_z^2}$ . Since the number of  $\mathbf{q}$  modes participating in relaxation process of a single spin is approximately  $k_o^2 \Omega / 4\pi$ , the decay rate of

the uncorrelated spin becomes

$$\Lambda_k(0) \equiv \gamma^{(o)} \approx (k_o^2 \Omega / 4\pi) G(k_o). \quad (26)$$

Comparing eqs. (25) and (26), we can see the effect of coherent effect on the decay rate of magnon in a very thin magnetic film. While an isolated spin may couple to as many as  $k_o^2 \Omega / 4\pi$  planar radiation modes, a magnon in the crystal film can only couple to one such radiation mode. However, all  $N$  spins in the film couple to photons coherently, therefore, the increase of the decay rate results. The enhancement factor, as the ratio of these two factors, is obtained easily as

$$\frac{\gamma_k}{\gamma_o} = \frac{N}{k_o^2 \Omega / 4\pi} \sim \frac{1}{(k_o d)^2}, \quad (27)$$

where  $d$  is the linear dimension of a unit cell. The magnitude of the enhancement factor is very large for the typical field strength. The physical origin of the enhancement is obviously due to the crystal symmetry in the film, which is purely solid state effect.

#### 4. Discussion

Although the phase coherence between magnon and the photon in the film plane is essential for determining the enhanced relaxation feature in this system, geometrical confinement is also a key to the spontaneous emission itself. While the degree of freedom for the 2D magnon is frozen out simply due to the reduced dimensionality, the degree of freedom for the emitted 3D phonon increases enormously because the z-component wave vector could be any value permitted in the range satisfying the energy conservation. Actually, the bulk Hamiltonian with crystal symmetry can be diagonalized [1], yielding new eigen states, the energy of which is maintained in the system without leaking. The coherent radiation related to the symmetry breaking is well understood in the case of exciton relaxation process of semiconductors, [13] and we can see a close analog in the present magnetic film case. As a matter of fact, the coherent radiation is studied for the atomic gas system, and the enhancement of radiation rate is called superradiance. [16]

Finally, we would like to point out some details that could affect the results of the present study. The role of the external field can be found in the energy conserving condition; emission of photon is possible by raising the magnon part of the dispersion curve over the photon curve, which is satisfied in the long wavelength regime. The usage of long wavelength also guarantees the application of the continuum model that we introduced

for magnon and photon from the beginning. The direction of the external field is chosen to point the usual direction of easy axis on the plane. The magnitude of the external field can control the range of magnon wave vector to decay into photon, permitted by the energy conservation. We actually assumed one layer of ferromagnetic film, but the multi-layer system would give rise to a change when we take spatial integration for z-coordinate in eq. (8); there would be an oscillation over the variation of number of layers. The effect of phonon in the film might be small, since the transition of magnon into phonon is very restricted due to a formation of hybrid of states in 2D again. However, if we supply a situation of bulk elastic system around the magnetic film, then a coherent radiation of magnon through phonon is also possible. This process is expected to be substantial in so called the sandwich structure, and is our future target.

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