

# Resonant Frequency Shift in Anisotropic Exchange Ferromagnets

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The resonant frequency shift  $\Delta\omega$  in anisotropic exchange ferromagnets at high power is calculated by using magnon theory. It is found that the presence of dynamically excited magnons at high power may reduce the z component of magnetization and results in a shift of the resonant frequency through a change of the exchange anisotropy. The resonance condition for a fixed resonance frequency changes towards higher fields with increasing the microwave power in the spherical sample.

This note is concerned with the effect of an exchange anisotropy on the FMR frequency in anisotropic exchange ferromagnets at high power. In the ferro- and antiferromagnets, there have been many studies of high power effects on the FMR<sup>1,2)</sup> and AFMR<sup>3-6)</sup>. It has been shown that the surface demagnetization and uniaxial anisotropy fields have a profound influence on the frequency shifts  $\Delta\omega$  at high power resonances. A review of this topic was given by Damon<sup>7)</sup>. We shall consider that the present ferromagnet consists of anisotropic exchange terms, plus Zeeman energy terms under an external field applied along the axis of magnetization, i.e., z-axis. The Hamiltonian of the system may be represented as

$$\mathcal{H} = -J^z \sum_j \sum_{\rho} S_j^z S_{j+\rho}^z - \frac{1}{2} J^{\perp} \sum_j \sum_{\rho} (S_j^+ S_{j+\rho}^- + S_j^- S_{j+\rho}^+) - g\mu_B H_0 \sum_j S_j^z \quad (1)$$

where  $S_j^{\pm} = S_j^x \pm iS_j^y$  and j denotes the lattice sites, the subscript  $\rho$  specifies the nearest neighboring site, and  $\sum_j$  means the summation of all lattice site.  $J^z(>0)$  and  $J^{\perp}(>0)$  are the longitudinal and perpendicular exchange integrals respectively, g the spectroscopic splitting factor, and  $\mu_B$  the Bohr magneton. We now assume that the longitudinal exchange integral  $J^z$  is larger than the perpendicular one  $J^{\perp}$ . Therefore the ground state of the system is the complete spin alignment.

Following the previous paper<sup>6)</sup>, we shall calculate the frequency of the uniform magnon mode in a spherical sample, retaining terms in the Hamiltonian up to fourth order in magnon creation and annihilation operators. Then the Hamiltonian can be expressed as

$$\begin{aligned} \mathcal{H} = & 2J^z S_z \sum_{\mathbf{k}} [1 - \gamma_{\mathbf{k}} J^{\perp}/J^z - g\mu_B H_0/2J^z S_z] a_{\mathbf{k}}^+ a_{\mathbf{k}} \\ & + \frac{J^z}{2N} \sum_{\mathbf{k}_1} \sum_{\mathbf{k}_2} \sum_{\mathbf{k}_3} \sum_{\mathbf{k}_4} [(\gamma_{\mathbf{k}_1} + \gamma_{\mathbf{k}_2}) J^{\perp}/J^z - 2\gamma_{\mathbf{k}_2 - \mathbf{k}_3}] a_{\mathbf{k}_1}^+ a_{\mathbf{k}_2}^+ a_{\mathbf{k}_3} a_{\mathbf{k}_4} \delta(\mathbf{k}_1 + \mathbf{k}_2 - \mathbf{k}_3 - \mathbf{k}_4) \end{aligned} \quad (2)$$

where

$$\gamma_{\mathbf{k}} = \frac{1}{z} \sum_{\rho} e^{i\mathbf{k}\cdot\boldsymbol{\rho}} \quad (3)$$

Here  $S$  is the magnitude of spin,  $N$  the number of spins and  $z$  the coordination number. To diagonalize the Hamiltonian (2), we shall use a random phase approximation (RPA) for the quartic terms of the magnon operators of eq. (2) as follows;

$$\begin{aligned} a_{\mathbf{k}_1}^+ a_{\mathbf{k}_2}^+ a_{\mathbf{k}_3} a_{\mathbf{k}_4} \simeq & \langle a_{\mathbf{k}_1}^+ a_{\mathbf{k}_3} \rangle a_{\mathbf{k}_2}^+ a_{\mathbf{k}_4} + \langle a_{\mathbf{k}_1}^+ a_{\mathbf{k}_4} \rangle a_{\mathbf{k}_2}^+ a_{\mathbf{k}_3} + \langle a_{\mathbf{k}_2}^+ a_{\mathbf{k}_3} \rangle a_{\mathbf{k}_1}^+ a_{\mathbf{k}_4} \\ & + \langle a_{\mathbf{k}_2}^+ a_{\mathbf{k}_4} \rangle a_{\mathbf{k}_1}^+ a_{\mathbf{k}_3} - \langle a_{\mathbf{k}_1}^+ a_{\mathbf{k}_3} \rangle \langle a_{\mathbf{k}_2}^+ a_{\mathbf{k}_4} \rangle - \langle a_{\mathbf{k}_1}^+ a_{\mathbf{k}_4} \rangle \langle a_{\mathbf{k}_2}^+ a_{\mathbf{k}_3} \rangle \end{aligned} \quad (4)$$

The last two terms are required if we are not to count the remaining terms twice, as is made evident if the average of the whole expression is taken. However these terms results only in additional constant energy in the Hamiltonian. Neglecting the constant term, eq. (4) can be expressed as

$$\begin{aligned} a_{\mathbf{k}_1}^+ a_{\mathbf{k}_2}^+ a_{\mathbf{k}_3} a_{\mathbf{k}_4} \simeq & \langle a_{\mathbf{k}_1}^+ a_{\mathbf{k}_1} \rangle (\delta_{\mathbf{k}_1 \mathbf{k}_3} a_{\mathbf{k}_2}^+ a_{\mathbf{k}_4} + \delta_{\mathbf{k}_1 \mathbf{k}_4} a_{\mathbf{k}_2}^+ a_{\mathbf{k}_3}) \\ & + \langle a_{\mathbf{k}_2}^+ a_{\mathbf{k}_2} \rangle (\delta_{\mathbf{k}_2 \mathbf{k}_3} a_{\mathbf{k}_1}^+ a_{\mathbf{k}_4} + \delta_{\mathbf{k}_2 \mathbf{k}_4} a_{\mathbf{k}_1}^+ a_{\mathbf{k}_3}) \end{aligned} \quad (5)$$

where the average of  $\langle a_{\mathbf{k}_1}^+ a_{\mathbf{k}_1} \rangle$  denotes a Hartree field and does not mean the thermal average only. Within the framework of a RPA, the magnon frequency of anisotropic exchange ferromagnets is obtained

$$\omega_{\mathbf{k}} = 2J^z S_z [1 - \gamma_{\mathbf{k}} J^{\perp}/J^z - g\mu_B H_0/2J^z S_z - \frac{1}{NS} \sum_{\mathbf{q}} (1 + \gamma_{\mathbf{q}}) (1 - J^{\perp}/J^z) \langle a_{\mathbf{q}}^+ a_{\mathbf{q}} \rangle] \quad (6)$$

For a small amplitude excitation, the magnon system is in thermal equilibrium. In this case the average  $\langle a_{\mathbf{q}}^+ a_{\mathbf{q}} \rangle$  may be taken as the occupation number of the thermal magnon,  $n_{\mathbf{q}}$  which is defined by the Bose factor. The FMR frequency  $\omega_0$  of anisotropic exchange ferromagnets is given by

$$\frac{\omega_0}{\gamma} = H_E (1 - J^{\perp}/J^z) [1 - \frac{1}{NS} \sum_{\mathbf{q}} (1 + \gamma_{\mathbf{q}}) n_{\mathbf{q}}] - H_0 \quad (7)$$

where

$$H_E = 2J^z S_z / g\mu_B \quad (8)$$

$\gamma$  is the gyromagnetic ratio.

At high power resonance, a large number of uniform mode magnons are excited. As mentioned in the previous paper<sup>6)</sup>, we assume that these magnons decay very rapidly via  $T_2$ -like process such as imperfection scatterings into the manifold of degenerate

$\mathbf{q} \neq 0$  magnon states and these magnons then relax into the whole spectrum of non-degenerate magnons with a characteristic time  $T_q$ . We also assume that the non-degenerate magnons relax rapidly to the heat bath with magnon-phonon relaxation time, i.e.,  $T_L < T_q$ , so that all magnons not degenerate with the uniform mode are in thermal equilibrium with lattice. Since the occupation numbers of the uniform magnon and degenerate  $\mathbf{q} \neq 0$  magnon modes change from their thermal values, we may put  $\langle a_{\mathbf{q}}^\dagger a_{\mathbf{q}} \rangle$  in eq. (6) as follows

$$\langle a_{\mathbf{q}}^\dagger a_{\mathbf{q}} \rangle = n_q + \Delta n_q \quad (9)$$

where  $\Delta n_q$  is the magnon population in excess of the thermal number. It is noted that  $\Delta n_q$  represents the number of dynamically excited magnons. Thus the frequency shift  $\Delta\omega$  at high power FMR is expressed by

$$\frac{\Delta\omega}{\gamma} = -H_E(1-J/J^z) \frac{\gamma\hbar}{M} [2\Delta n_0 + \sum_q (1+\gamma_q) \Delta n_q] \quad (10)$$

where  $M = g\mu_B NS$  is the saturation magnetization and  $\hbar$  is Planck's constant. In eq. (10) the prime in the summation denotes the sum over wave vectors  $\mathbf{q}$  whose energies are degenerate with that of the uniform mode.

Thus the shift is obtained in terms of population of the uniform mode ( $\mathbf{q}=0$ ) and degenerate ( $\mathbf{q} \neq 0$ ) mode magnons. In order to derive eqs. (7) and (10), we have used a random phase approximation (RPA). It might be reasonable to approximate the magnon interactions by means of a RPA because of the high density of the uniform and degenerate magnons. However the interference effect among the thermal and dynamical magnons on the shift is ignored in our discussion. It is shown that the shift  $\Delta\omega$  depends on the degree of exchange anisotropy and its sign is negative, so that the resonance field shifts towards higher fields with increasing the microwave power. Similar effects may be expected for anisotropic exchange antiferromagnets. It is hoped that the present note will stimulate the observation of the frequency shift at high power in anisotropic exchange magnets as  $\text{CoCl}_2 \cdot 2\text{H}_2\text{O}$ ,  $\text{FeCl}_2$  and  $\text{K}_2\text{CoF}_4$ .

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