Heisenberg Hamiltonian with Second Order Perturbation for Spinel Ferrite Ultra-thin Films

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Received 1 April 2008, Accepted 15 August 2008, Published 20 February 2009

Abstract: The solution of Heisenberg Hamiltonian with second order perturbation will be described for non-oriented spinel cubic ferrimagnetic materials. The perturbation related to the change of angle at the interface of two cells will be considered. The energy peaks become sharper and peak position varies in energy-angle curve as N is increased from 2 to 3. But the separation between two consecutive major maximums remains same. The 3-D plot of total energy versus angle and stress becomes smoother as N is increased from 2 to 3. The energy decreases with number of layers indicating that the behavior of oriented and non-oriented films is different. In N=2 case, minor maximums next to major maximum can be observed. When second order anisotropy constant does not vary within the film with N=2, film behaves as an oriented film.

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Keywords: Heisenberg Hamiltonian; Ferrimagnetic Materials; Spinel Ferrites; Spin Models; Magnetic Anisotropy

PACS (2008): 75.10.Hk; 75.30.Gw; 75.70.-i

1. Introduction

In some early reports, the structure of spinel ferrites with the position of octahedral and tetrahedral sites is given in detail [1-5]. Only the occupied octahedral and tetrahedral sites were used for the calculation in this report, although there are many filled and vacant octahedral and tetrahedral sites in cubic spinel cell [1]. Only few previous reports could be found on the theoretical works of ferrites [6-9]. The solution of Heisenberg ferrites only with spin exchange interaction term has been found earlier by means of the retarded Green function equations [6].

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All the relevant energy terms such as spin exchange energy, dipole energy, second and fourth order anisotropy terms, interaction with magnetic field and stress induced anisotropy in Heisenberg Hamiltonian were taken into consideration. These equations derived here can be applied for spinel ferrites with unit cell AFe$_2$O$_4$ such as Fe$_3$O$_4$, NiFe$_2$O$_4$ and ZnFe$_2$O$_4$ only. The spin exchange interaction energy and dipole interaction have been calculated only between two nearest spin layers and within same spin plane. Also the azimuthal angle of spins within one cubic cell is assumed to be constant. The change of angle at the interface of cubic cell will be considered. According to some of our early experimental reports, the anisotropy energy of Nickel ferrite and Lithium mixed ferrite depends on the stress of the film induced during cooling or heating process of the film [15, 16].

2. The Model

Classical Heisenberg Hamiltonian of a thin film can be written as following.

$$H = -J \sum_{m,n} \vec{S}_m \cdot \vec{S}_n + \omega \sum_{m \neq n} \left( \frac{\vec{S}_m \cdot \vec{S}_n}{r_{mn}^3} - \frac{3(\vec{S}_m \cdot \vec{r}_{mn})(\vec{r}_{mn} \cdot \vec{S}_n)}{r_{mn}^5} \right) - \sum_m D_2^{(2)} (S^2_m) - \sum_m D_4^{(4)} (S^4_m) - \sum_m \vec{H} \cdot \vec{S}_m - \sum_m K_s \sin 2\theta_m$$

Here $J$, $\omega$, $\theta$, $D^{(2)}_m$, $D^{(4)}_m$, $H_{in}$, $H_{out}$, $K_s$, $m$, $n$ and $N$ are spin exchange interaction, strength of long range dipole interaction, azimuthal angle of spin, second and fourth order anisotropy constants, in plane and out of plane applied magnetic fields, stress induced anisotropy constant, spin plane indices and total number of layers in film, respectively. When the stress applies normal to the film plane, the angle between $m$th spin and the stress is $\theta_m$.

The cubic cell was divided into 8 spin layers with alternative A and Fe spins layers [1]. The spins of A and Fe will be taken as 1 and p, respectively. While the spins in one layer point in one direction, spins in adjacent layers point in opposite directions. A thin film with (001) spinel cubic cell orientation will be considered. The length of one side of unit cell will be taken as “a”. Within the cell the spins orient in one direction due to the super exchange interaction between spins (or magnetic moments). Therefore the results proven for oriented case in one of our early report [11] will be used for following equations. But the angle $\theta$ will vary from $\theta_m$ to $\theta_{m+1}$ at the interface between two cells.

For a thin film with thickness $Na$,

Spin exchange interaction energy=

$$E_{exchange} = N(-10J + 72Jp - 22Jp^2) + 8Jp \sum_{m=1}^{N-1} \cos(\theta_{m+1} - \theta_m)$$

Dipole interaction energy=$E_{dipole}$

$$E_{dipole} = -48.415 \omega \sum_{m=1}^{N} (1 + 3 \cos 2\theta_m) + 20.41 \omega p \sum_{m=1}^{N-1} [\cos(\theta_{m+1} - \theta_m) + 3 \cos(\theta_{m+1} + \theta_m)]$$
Here the first and second term in each above equation represent the variation of energy within the cell [11] and the interface of the cell, respectively.

Total energy

\[
E = N(-10J + 72Jp - 22Jp^2) + 8Jp \sum_{m=1}^{N-1} \cos(\theta_{m+1} - \theta_m)
\]

\[
- 48.415\omega \sum_{m=1}^{N} (1 + 3 \cos 2\theta_m) + 20.41\omega p \sum_{m=1}^{N-1} [\cos(\theta_{m+1} - \theta_m) + 3 \cos(\theta_{m+1} + \theta_m)]
\]

\[
- \sum_{m=1}^{N} [D_m^{(2)} \cos^2 \theta_m + D_m^{(4)} \cos^4 \theta_m] - 4(1 - p) \sum_{m=1}^{N} [H_{in} \sin \theta_m + H_{out} \cos \theta_m + K_s \sin 2\theta_m]
\]

(2)

Here the anisotropy energy term and the last term have been explained in our previous report for oriented spinel ferrite [11]. If the angle is given by \( \theta_m = \theta + \varepsilon_m \) with perturbation \( \varepsilon_m \), after taking the terms up to second order perturbation of \( \varepsilon \) only,

The total energy can be given as

\[
E(\theta) = E_0 + E(\varepsilon) + E(\varepsilon^2)
\]

Here

\[
E_0 = -10JN + 72pNJ - 22Jp^2N + 8Jp(N - 1) - 48.415\omega N - 145.245\omega N \cos(2\theta)
\]

\[
+ 20.41\omega p [[(N-1)+3(N-1)\cos(2\theta)]
\]

\[
- \cos^2 \theta \sum_{m=1}^{N} D_m^{(2)} - \cos^4 \theta \sum_{m=1}^{N} D_m^{(4)} - 4(1 - p)N(H_{in} \sin \theta + H_{out} \cos \theta + K_s \sin 2\theta)
\]

(3)

\[
E(\varepsilon) = 290.5\omega \sin(2\theta) \sum_{m=1}^{N} \varepsilon_m - 61.23\omega p \sin(2\theta) \sum_{m=1}^{N-1} (\varepsilon_m + \varepsilon_n)
\]

\[
+ \sin 2\theta \sum_{m=1}^{N} D_m^{(2)} \varepsilon_m + 2 \cos^2 \theta \sin 2\theta \sum_{m=1}^{N} D_m^{(4)} \varepsilon_m
\]

\[
+ 4(1 - p) [-H_{in} \cos \theta \sum_{m=1}^{N} \varepsilon_m + H_{out} \sin \theta \sum_{m=1}^{N} \varepsilon_m - 2K_s \cos 2\theta \sum_{m=1}^{N} \varepsilon_m]
\]

(4)
\[
E(\varepsilon^2) = -4Jp \sum_{m=1}^{N-1} (\varepsilon_n - \varepsilon_m)^2 + 290.5\omega \cos(2\theta) \sum_{m=1}^{N} \varepsilon_m^2 - 10.2\omega p \sum_{m=1}^{N-1} (\varepsilon_n - \varepsilon_m)^2 \\
- 30.6\omega p \cos(2\theta) \sum_{m=1}^{N-1} (\varepsilon_n + \varepsilon_m)^2 \\
- (\sin^2 \theta - \cos^2 \theta) \sum_{m=1}^{N} D^{(2)}_m \varepsilon_m^2 + 2\cos^2 \theta (\cos^2 \theta - 3\sin^2 \theta) \sum_{m=1}^{N} D^{(4)}_m \varepsilon_m^2 \\
+ 4(1-p)[\frac{H_{in}}{2} \sin \theta \sum_{m=1}^{N} \varepsilon_m^2 + \frac{H_{out}}{2} \cos \theta \sum_{m=1}^{N} \varepsilon_m^2 + 2K_s \sin 2\theta \sum_{m=1}^{N} \varepsilon_m^2] 
\]

The sin and cosine terms in equation number 2 have been expanded to obtain above equations. Here n=m+1.

Under the constraint \(\sum_{m=1}^{N} \varepsilon_m = 0\), first and last three terms of equation 4 are zero.

Therefore, \(E(\varepsilon) = \vec{\alpha}.\vec{\varepsilon}\)

Here \(\vec{\alpha}(\varepsilon) = \vec{B}(\theta)\sin 2\theta\) are the terms of matrices with

\[
B_\lambda(\theta) = -122.46\omega p + D^{(2)}_\lambda + 2D^{(4)}_\lambda \cos^2 \theta
\]

Also \(E(\varepsilon^2) = \frac{1}{2}\vec{\varepsilon}.C.\vec{\varepsilon}\), and matrix C is assumed to be symmetric (\(C_{mn}=C_{nm}\)).

Here the elements of matrix C can be given as following,

\[
C_{m,m+1} = 8Jp + 20.4\omega p - 61.2p\omega \cos(2\theta)
\]

For m=1 and N,

\[
C_{mm} = -8Jp - 20.4\omega p - 61.2p\omega \cos(2\theta) + 581\omega \cos(2\theta) - 2(\sin^2 \theta - \cos^2 \theta)D^{(2)}_m \\
+ 4\cos^2 \theta (\cos^2 \theta - 3\sin^2 \theta)D^{(4)}_m + 4(1-p)[H_{in} \sin \theta + H_{out} \cos \theta + 4K_s \sin(2\theta)]
\]

For m=2, 3, —-, N-1

\[
C_{mm} = -16Jp - 40.8\omega p - 122.4p\omega \cos(2\theta) + 581\omega \cos(2\theta) - 2(\sin^2 \theta - \cos^2 \theta)D^{(2)}_m \\
+ 4\cos^2 \theta (\cos^2 \theta - 3\sin^2 \theta)D^{(4)}_m + 4(1-p)[H_{in} \sin \theta + H_{out} \cos \theta + 4K_s \sin(2\theta)]
\]

Otherwise, \(C_{mn}=0\)

Therefore, the total energy can be given as

\[
E(\theta) = E_0 + \vec{\alpha}.\vec{\varepsilon} + \frac{1}{2}\vec{\varepsilon}.C.\vec{\varepsilon} = E_0 - \frac{1}{2}\vec{\alpha}.C^+.\vec{\alpha}
\]

Here \(C^+\) is the pseudo-inverse given by

\[
C.C^+ = 1 - \frac{E}{N}
\]

Here E is the matrix with all elements \(E_{mn}=1\).
3. Results and Discussion

First a film with N=2 will be considered. When anisotropy constants $D_m^{(2)}$ and $D_m^{(4)}$ do not vary within the film $\alpha_1 = \alpha_2$ from equation 6.

Then from equation 7, $C_{11}=C_{22}$ and $C_{12}=C_{21}$.

From equation 9, $C_{12}^+ = C_{21}^+ = \frac{1}{2(C_{21}-C_{22})} = -C_{11}^+ = -C_{22}^+$

Therefore, $\vec{\alpha}.C^+.\vec{\alpha} = (\alpha_1 - \alpha_2)^2 C_{11}^+=0$

Finally the energy $E(\theta) = E_0$ and film behaves as an oriented film. The exactly same result was obtained for thin ferromagnetic films earlier [12].

If the anisotropy constants vary within the film, then $C_{12}=C_{21}$ and $C_{11} \neq C_{22}$.

Then $C_{11}^+ = -C_{12}^+ = \frac{C_{22}+C_{21}}{2(C_{22}-C_{21})}$ and $C_{21}^+ = -C_{22}^+ = \frac{C_{22}+C_{21}}{2(C_{22}-C_{21})}$.

Hence, $\vec{\alpha}.C^+.\vec{\alpha} = (\alpha_1 - \alpha_2)^2(C_{11}^+ \alpha_2 - C_{12}^+ \alpha_1)$

When all the terms in equation 7 are taken into account, the $C_{mn}^+$ is contained more than 80 terms. Therefore only the spin exchange interaction, spin dipole interaction, second order anisotropy and stress induced anisotropy have been considered to avoid tedious calculations.

Then

$C_{11} = -8Jp-20.4p_1 \omega p+61.2p_1 \omega \cos(2\theta) +581p_1 \omega \cos(2\theta) + 2(\cos 2\theta)D_{1}^{(2)} +16(1-p)K_{s} \sin(2\theta)$

$C_{22} = -8Jp-20.4p_1 \omega p+61.2p_1 \omega \cos(2\theta) +581p_1 \omega \cos(2\theta) + 2(\cos 2\theta)D_{2}^{(2)} +16(1-p)K_{s} \sin(2\theta)$

$C_{12} = 8Jp+20.4p_1 \omega p-61.2p_1 \omega \cos(2\theta)$

$\alpha_1 = [-122.46p_1 + D_{1}^{(2)}] \sin(2\theta)$

$\alpha_2 = [-122.46p_1 + D_{2}^{(2)}] \sin(2\theta)$

$E(\theta)=E_0 = \frac{(\alpha_1 - \alpha_2) (C_{11}^+ \alpha_2 - C_{12}^+ \alpha_1)}{2}$

$E_0 = -20J+144p_1 \omega +44Jp^2 +8Jp-96.83p_1 \omega -290.5p_1 \omega \cos(2\theta) +20.41p_1 \omega \sin(2\theta)$

This simulation will be performed for Nickel ferrite with $p=2.5$.

Then

$C_{11} = -20J-51\omega +428\omega \cos(2\theta) + 2(\cos 2\theta)D_{1}^{(2)} - 24K_{s} \sin(2\theta)$

$C_{22} = -20J-51\omega +428\omega \cos(2\theta) + 2(\cos 2\theta)D_{2}^{(2)} - 24K_{s} \sin(2\theta)$

$C_{12} = 20J+51\omega -153\omega \cos(2\theta)$

$E_0 = 85J-96.83\omega -290.5\omega \cos(2\theta) + 51.03\omega \sin(2\theta) - 20.41\omega \sin(2\theta) - 12K_{s} \sin(2\theta)$

$+ 51.03\omega \sin(2\theta)$

Then $E(\theta)$ can be found using equation 8. When $\theta = 0^0$ and $\theta = 90^0$, the second order perturbation energy term is zero and film behaves as an oriented film. The graph between $E(\theta)$ and $\theta$ is given in figure 1, for $\frac{J}{\omega} = \frac{D_{1}^{(2)}}{\omega} = \frac{K_{s}}{\omega} = 10, \frac{D_{2}^{(2)}}{\omega} = 5$. Nearest maximum and minimum can be observed at $63^0$ and $86^0$, respectively. Therefore the angle between easy and hard directions is not $90^0$ in this case. Two consecutive maximum can be observed at $63^0$ and $240.7^0$.

When $\frac{K_{s}}{\omega}$ is a variable, the 3-D plot of $E(\theta)$ versus $\theta$ and $\frac{K_{s}}{\omega}$ is given in figure 2. This graph shows a variation similar to oriented spinel ferrite films [11]. The graph indicates several minimums indicating that film can be easily oriented in some particular directions by applying a stress.
When N=3, the each $C^+_{mn}$ element found using equation 9 is contained more than 20 terms. To avoid this problem, matrix elements were found using C.C$^+_{mn}$=1. Then $C^+_{mn}$ is given by $C^+_{mn} = \frac{\text{cofactor}_{C_{mn}}}{\det C}$. Under this condition, $\bar{E}$ is zero, and the average value of first order perturbation is zero [12]. The second order anisotropy constant is assumed to be an invariant for the convenience.

Then $C_{11}=C_{33}$, $C_{12}=C_{21}=C_{23}=C_{32}$, $C_{13}=C_{31}=0$, $\alpha_1 = \alpha_2 = \alpha_3$. Then $C_{11}=C_{33} = -20\omega + 428\omega \cos(2\theta) + 2(\cos 2\theta) D_m(2) - 24K_s \sin(2\theta)$

$C_{22} = -40\omega - 102\omega + 275\omega \cos(2\theta) + 2(\cos 2\theta) D_m(2) - 24K_s \sin(2\theta)$

Therefore[12], $C^+_{11} = \frac{C_{11}C_{22}-C_{21}^2}{C_{11}C_{22}-2C_{32}C_{31}} = C^+_{33}, C^+_{13} = \frac{C_{13}^2}{C_{13}C_{32}+C_{11}C_{33}-2C_{32}C_{11}} = C^+_{31}$

$C^+_{12} = \frac{-C_{32}C_{11}}{C_{11}C_{22}-2C_{32}C_{11}} = C^+_{21} = C^+_{23} = C^+_{32}, C^+_{22} = \frac{C^2_{11}}{C_{11}C_{22}-2C_{32}C_{11}}$

The total energy can be found using following equation.

$E(\theta)=E_0 - 0.5[C_{11}(2\alpha_1^2)+C_{32}(4\alpha_1^2)+C_{31}(2\alpha_2^2)+\alpha_1^2C_{22}^2]$

Here $E_0 = 137.5J-145.253\omega-0.5\omega \cos(2\theta)+102.05\omega[1+3\cos(2\theta)]$ - $\cos^2 \theta[D_{11}(2)+D_{22}(2)+D_{33}(2)] + 18K_s \sin(2\theta)$

The graph between $\frac{E(\theta)}{\omega}$ and $\theta$ is given in figure 3, for $\frac{K}{\omega} = \frac{D_{22}(2)}{\omega} = K_s = 10$.

These peaks become sharper compared with those given figure 1 for spinel ferrite films with N=2 and ferromagnetic thin films of N=3 with 2$^{nd}$ order perturbation described in one of our early report [12]. Two consecutive maxima can be observed at 23° and 97°. But the peak positions are different from those given in figure 1. Energy is smaller compared with N=2 spinel ferrite films given in figure 1. But energy is higher compared with ferromagnetic thin films [12] with N=3. Although it is difficult to find any experimental reports related to the thickness or angle dependence of magnetic energy of Nickel ferrite thin films, this kind of phenomena has been observed for Co-ferrite and Mn-Zn ferrite thin films. The energy of these Co- ferrite films depends on the thickness [13]. Also the easy direction (or preferred orientation direction) of Mn-Zn ferrite thin films varies with the thickness of the film [14]. According to figure 1 and 3, the angle corresponding to energy minima varies with the thickness or number of layers (N). But the numerical values of Nickel ferrite films obtained here can not be compared with the numerical values obtained for Co-ferrite or Mn-Zn ferrite films.

When $\frac{K_s}{\omega}$ is a variable, the 3-D plot of $\frac{E(\theta)}{\omega}$ versus $\theta$ and $\frac{K_s}{\omega}$ is given in figure 4. This graph shows a higher variation of energy and higher maximum energy compared with ferromagnetic thin films with N=3. Compared with figure 2, energy is less in this case. Some experimental evidences confirm that the energy of Co-ferrite thin films depend on the residual stress [13]. Also the anisotropy energy of Nickel ferrite and Lithium mixed ferrite films depends on the induced stress [15, 16].

**Conclusion**

The energy peaks become sharper and peak position varies in energy-angle curve as N is increased from 2 to 3. But the separation between two consecutive major maxima
remains same. The angle between easy and hard directions is not \(90^\circ\). The 3-D plot of \(E(\theta)\) versus \(\theta\) and \(K_{s}\) becomes smoother as \(N\) is increased from 2 to 3. Unlike the oriented spinel ferrite films, the energy decreases with number of layers in this case according to figure 1, 2, 3 and 4. Some experimental results also indicate that the anisotropy energy depends on the thickness and stress inside the film, and the easy direction depends on the thickness of the ferrite films. In case \(N=2\), minor maximums next to major maximum can be observed. These simulations can be performed for any other ferrite and some other values of \(\frac{J}{\omega}, \frac{D^{(2)}}{\omega}\) and \(K_{s}\).

References

Fig. 1 Graph between $E(\theta)/\omega$ and $\theta$ for $K_\omega^2=10$ with the effect of variable second order anisotropy for $N=2$. 
Fig. 2 3-D plot of $\frac{E(\theta)}{\omega}$ versus $\frac{K_s}{\omega}$ and $\theta$ with the effect of variable second order anisotropy for $N=2$. 
Fig. 3 Graph between $\frac{E(\theta)}{\omega}$ and $\theta$ for $\frac{K_s}{\omega}=10$ with the effect of invariant second order anisotropy for $N=3$. 
Fig. 4 3-D plot of $\frac{E(\theta)}{\omega}$ versus $\frac{K_s}{\omega}$ and $\theta$ with the effect of invariant second order anisotropy for $N=3$. 