Fe₃O₄ THICK FILMS AS DESCRIBED BY SECOND ORDER PERTURBED HEISENBERG HAMILTONIAN

P. SAMARASEKARA

Department of Physics, University of Peradeniya, Peradeniya, Sri Lanka

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ABSTRACT

Modified second order perturbed Heisenberg Hamiltonian has been used to explain magnetic properties including magnetic easy axis orientation of Fe₃O₄ thick films up to 1000 spin layers. The variation of magnetic easy and hard directions with the number of spin layers and stress induced anisotropy was investigated. 3-D plot of energy versus angle and number of spin layers was plotted to find the values of number of spin layers corresponding to energy minima and maxima. By plotting the curve of energy versus angle at one selected value of number of layers, magnetic easy and hard directions were determined. Similarly 3-D plot of energy versus angle and stress induced anisotropy was plotted to find the values of stress induced anisotropy corresponding to energy minima and maxima. Then the curve of energy versus angle at one selected value of stress induced anisotropy was plotted to find the magnetic easy and hard directions. MATLAB software was employed to plot all the graphs.

Keywords: spin, stress induced anisotropy, second order perturbed Heisenberg Hamiltonian, ferrites, magnetic easy direction

*Corresponding author Email: pubudus@pdn.ac.lk;

<http://orcid.org/0000-0003-2806-0518>

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INTRODUCTION

Fe_3O_4 (magnetite) is a prime candidate in applications of magnetic storage, industrial catalysts, water purification and drug delivery. Fe_3O_4 is a non-uniaxial ferrite with inverse spinel structure. Magnetic easy axis of Fe_3O_4 is along one of the body diagonal of the cubic cell due to the spin arrangement parallel and antiparallel to the body diagonal. Spinel structure with tetrahedral and octahedral sites can be found in detail in some previous publications (Ahmed Farag *et al.*, 2001a, Ahmed Farag *et al.*, 2001b, Kahlenberg *et al.*, 2001, John Zhang *et al.*, 1998, Sickafus *et al.*, 1999). Five of Fe^{3+} ions occupy tetrahedral sites. Other five Fe^{3+} ions and four Fe^{2+} ions occupy octahedral sites. The spinel structure of this ferrite is represented by $\text{Fe}^{3+}(\text{Fe}^{2+}\text{Fe}^{3+})\text{O}_4$. The magnetic moments of Fe^{2+} and Fe^{3+} are $4 \mu_B$ and $5 \mu_B$, respectively. Because magnetic moments of Fe^{3+} in tetrahedral and octahedral sites cancel each other, the net magnetic moment of Fe_3O_4 is completely due to the magnetic moments of four Fe^{2+} ions. Therefore, the theoretical net magnetic moment of Fe_3O_4 is 4 Bohr magnetons.

Rietveld method has been employed to determine the cation distribution of ferrite like compounds (Ahmed Farag *et al.*, 2001a, Ahmed Farag *et al.*, 2001b). Surface spin waves in CsCl type ferrimagnet with a (001) surface has been studied by combining Green function theory with the transfer matrix method (Dai & Li, 1990). Anisotropy of ultrathin ferromagnetic films and the spin reorientation transition have been investigated using Heisenberg Hamiltonian with few terms (Usadel & Hucht, 2002). In addition, the surface magnetism of ferrimagnet thin films has been studied using Heisenberg method (Ding *et al.*, 1993). The surface spin wave spectra of both the simple cubic and body centered ferrimagnets have been theoretically studied using Heisenberg Hamiltonian (Hung *et al.*, 1975). The cation distribution and oxidation state of Mn-Fe spinel nanoparticles have been systematically studied at various temperatures by using neutron diffraction and electron

energy loss spectroscopy (John Zang *et al.*, 1998). The crystal structure of spinel type compounds has been found using single crystal X-ray diffraction data (Kahlenberg *et al.*, 2001). Surface spin waves on the (001) free surface of semi-infinite two lattice ferrimagnets on the Heisenberg model with nearest neighbor exchange interactions has been investigated (Zheng & Lin, 1988). The lattice parameter, anion parameter and the cation inversion parameter of spinel structures have been presented (Sickafus *et al.*, 1999).

Ferromagnetic ultra thin and thick films have been studied previously using third order perturbed Heisenberg Hamiltonian (Samarasekara, 2008, Samarasekara & Mendoza, 2010). Ferromagnetic ultra-thin and thick films have been investigated using second order perturbed Heisenberg Hamiltonian by us (Samarasekara & Gunawardhane, 2011). Furthermore, ferrite ultra-thin and thick films have been investigated using second order perturbed Heisenberg Hamiltonian by us (Samarasekara *et al.*, 2009, Samarasekara, 2010). Ferrite ultra-thin and thick films have been investigated using third order perturbed Heisenberg Hamiltonian (Samarasekara & Mendoza, 2011, Samarasekara, 2011). The magnetic properties of the spinel Fe₃O₄ thick films are described in this manuscript.

MODEL

The modified Heisenberg Hamiltonian of any thin film with N spin layers can be given as

$$\begin{aligned} H = & -J \sum_{m,n} \vec{S}_m \cdot \vec{S}_n + \omega \sum_{m \neq n} \left(\frac{\vec{S}_m \cdot \vec{S}_n}{r_{mn}^3} - \frac{3(\vec{S}_m \cdot \vec{r}_{mn})(\vec{r}_{mn} \cdot \vec{S}_n)}{r_{mn}^5} \right) - \sum_m D_{\lambda_m}^{(2)} (S_m^z)^2 - \sum_m D_{\lambda_m}^{(4)} (S_m^z)^4 \\ & - \sum_m \vec{H} \cdot \vec{S}_m - \sum_m K_s \sin 2\theta_m \end{aligned} \quad (1)$$

Here J is spin exchange interaction, ω is the strength of long range dipole interaction, θ is azimuthal angle of spin, $D_m^{(2)}$ and $D_m^{(4)}$ are second and fourth order

anisotropy constants, H_{in} and H_{out} are in plane and out of plane applied magnetic fields, K_s is stress induced anisotropy constant, n and m are spin plane indices and N is total number of layers in film. When the stress applies normal to the film plane, the angle between m^{th} spin and the stress is θ_m .

The spinel cubic cell can be divided into 8 spin layers with alternative Fe^{2+} and Fe^{3+} spins layers (Sickafus et al., 1999). The spins in one layer and adjacent layers point in one direction and opposite directions, respectively. The spins of Fe^{2+} and Fe^{3+} will be taken as 1 and p , respectively. A cubic unit cell with length a will be considered. Due to the super exchange interaction between spins, the spins are parallel or antiparallel to each other within the cell. Therefore the results proven for oriented case in one of our early report (Samarasekara *et al.*, 2009) will be used for following equations. But the angle θ will vary from θ_m to θ_{m+1} at the interface between two cells.

Following equations will be proven for a thin film with thickness Na .

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

$$\text{Dipole interaction energy} = E_{\text{dipole}}$$

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

The first and second term in each above equation represent the variation of energy within the cell (Samarasekara *et al.*, 2009) and the interface of the cell, respectively.

Total energy

$$\begin{aligned}
 E = & N(-10J+72Jp-22Jp^2)+8Jp \sum_{m=1}^{N-1} \cos(\theta_{m+1} - \theta_m) \\
 & - 48.415\omega \sum_{m=1}^N (1+3\cos 2\theta_m) + 20.41\omega p \sum_{m=1}^{N-1} [\cos(\theta_{m+1} - \theta_m) + 3\cos(\theta_{m+1} + \theta_m)] \\
 & - \sum_{m=1}^N [D_m^{(2)} \cos^2 \theta_m + D_m^{(4)} \cos^4 \theta_m] \\
 & - 4(1-p) \sum_{m=1}^N [H_{in} \sin \theta_m + H_{out} \cos \theta_m + K_s \sin 2\theta_m] \quad (2)
 \end{aligned}$$

The anisotropy energy term and the last term given in above equations have been explained in our previous report for oriented spinel ferrite (Samarasekara *et al.*, 2009). The angle can be given as $\theta_m = \theta + \varepsilon_m$ with perturbation ε_m .

The total energy can be given as $E(\theta) = E_0 + E(\varepsilon) + E(\varepsilon^2)$

Here the energy is given only up to the second order perturbation of ε . Then after expanding sin and cosine terms,

$$\begin{aligned}
 E_0 = & -10JN+72pNJ-22Jp^2N+8Jp(N-1)-48.415\omega N-145.245\omega N \cos(2\theta) \\
 & +20.41\omega p[(N-1)+3(N-1)\cos(2\theta)] \\
 & - \cos^2 \theta \sum_{m=1}^N D_m^{(2)} - \cos^4 \theta \sum_{m=1}^N D_m^{(4)} - 4(1-p)N(H_{in} \sin \theta + H_{out} \cos \theta + K_s \sin 2\theta) \quad (3) \\
 E(\varepsilon) = & 290.5\omega \sin(2\theta) \sum_{m=1}^N \varepsilon_m - 61.23\omega p \sin(2\theta) \sum_{m=1}^{N-1} (\varepsilon_m + \varepsilon_n) \\
 & + \sin 2\theta \sum_{m=1}^N D_m^{(2)} \varepsilon_m + 2 \cos^2 \theta \sin 2\theta \sum_{m=1}^N D_m^{(4)} \varepsilon_m
 \end{aligned}$$

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

$$\begin{aligned} E(\varepsilon^2) = & -4Jp \sum_{m=1}^{N-1} (\varepsilon_n - \varepsilon_m)^2 + 290.5\omega \cos(2\theta) \sum_{m=1}^N \varepsilon_m^2 - 10.2\omega p \sum_{m=1}^{N-1} (\varepsilon_n - \varepsilon_m)^2 \\ & - 30.6\omega p \cos(2\theta) \sum_{m=1}^{N-1} (\varepsilon_n + \varepsilon_m)^2 \\ & - (\sin^2 \theta - \cos^2 \theta) \sum_{m=1}^N D_m^{(2)} \varepsilon_m^2 + 2 \cos^2 \theta (\cos^2 \theta - 3 \sin^2 \theta) \sum_{m=1}^N D_m^{(4)} \varepsilon_m^2 \\ & + 4(1-p) \left[\frac{H_{in}}{2} \sin \theta \sum_{m=1}^N \varepsilon_m^2 + \frac{H_{out}}{2} \cos \theta \sum_{m=1}^N \varepsilon_m^2 + 2K_s \sin 2\theta \sum_{m=1}^N \varepsilon_m^2 \right] \end{aligned} \quad (5)$$

Here $n = m+1$.

After using constraint $\sum_{m=1}^N \varepsilon_m = 0$, first and last three terms of equation 4 are zero.

$$\text{Then, } E(\varepsilon) = \vec{\alpha} \cdot \vec{\varepsilon} \quad (6)$$

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

$$B_\lambda(\theta) = -122.46\omega p + D_\lambda^{(2)} + 2D_\lambda^{(4)} \cos^2 \theta$$

$$\text{Also } E(\varepsilon^2) = \frac{1}{2} \vec{\varepsilon} \cdot C \cdot \vec{\varepsilon}$$

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

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

For $m = 1$ and N ,

Fe₃O₄ thick films as described by second order perturbed Heisenberg Hamiltonian

$$C_{mm} = -8Jp - 20.4\omega p - 61.2p\alpha\cos(2\theta) + 581\alpha\cos(2\theta) - 2(\sin^2\theta - \cos^2\theta) D_m^{(2)} \\ + 4\cos^2\theta(\cos^2\theta - 3\sin^2\theta) D_m^{(4)} + 4(1-p)[H_{in}\sin\theta + H_{out}\cos\theta + 4K_s\sin(2\theta)] \quad (7)$$

For $m = 2, 3, \dots, N-1$

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

Otherwise, $C_{mm} = 0$

Finally the total energy can be given as

$$E(\theta) = E_0 + \vec{\alpha} \cdot \vec{\varepsilon} + \frac{1}{2} \vec{\varepsilon} \cdot C \cdot \vec{\varepsilon} = E_0 - \frac{1}{2} \vec{\alpha} \cdot C^+ \cdot \vec{\alpha} \quad (8)$$

Here C^+ is the pseudo-inverse given by

$$C \cdot C^+ = 1 - \frac{E}{N}. \quad (9)$$

For a thick film with $N = 10000$, the second term in equation number 9 can be neglected.

Then C^+ will be the normal inverse of matrix C .

RESULTS AND DISCUSSION

If anisotropy constants $D_m^{(2)}$ and $D_m^{(4)}$ do not vary within the film, then $C_{11} = C_{NN}$ and $C_{22} = C_{33} = \dots = C_{N-1, N-1}$. When H_{in} , H_{out} and K_s are very large, $C_{22} \gg C_{12}$. If $C_{m, m+1}$ is zero, then the matrix C will be diagonal. Then the elements of inverse matrix (C^+)

is given by $C_{mm}^+ = \frac{1}{C_{mm}}$. To avoid tedious calculations, the solution will be found under

assumption $C_{m,m+1} = 0$.

Then $\alpha_1 = \dots = \alpha_n = [-122.46\omega p + D_{\lambda}^{(2)} + 2D_{\lambda}^{(4)} \cos^2 \theta] \sin(2\theta)$

$$\alpha.C^+.\alpha = 2C_{11}^+ \alpha_1^2 + \alpha_1^2 (N-2)C_{22}^+ = \frac{2\alpha_1^2}{C_{11}} + \frac{\alpha_1^2 (N-2)}{C_{22}}$$

For Fe_3O_4 , $p = 5/4 = 1.25$.

From equation 3,

$$E_0 = 55.625NJ - 10J - 48.415\omega N - 145.245\omega N \cos(2\theta) + 25.5125\omega(N-1)[1 + 3\cos(2\theta)] \\ - N[\cos^2 \theta D_m^{(2)} + \cos^4 \theta D_m^{(4)} - (H_{in} \sin \theta + H_{out} \cos \theta + K_s \sin 2\theta)]$$

From equation 7,

$$C_{11} = C_{NN} = -10J - 25.5\omega + 504.5\omega \cos(2\theta) + 2\cos 2\theta D_m^{(2)} + 4\cos^2 \theta (\cos^2 \theta - 3\sin^2 \theta) D_m^{(4)} \\ - [H_{in} \sin \theta + H_{out} \cos \theta + 4K_s \sin(2\theta)]$$

$$C_{22} = C_{33} = \dots = C_{N-1, N-1} = -20J - 51\omega + 428\omega \cos(2\theta) + 2(\cos 2\theta) D_m^{(2)}$$

$$+ 4\cos^2 \theta (\cos^2 \theta - 3\sin^2 \theta) D_m^{(4)} - [H_{in} \sin \theta + H_{out} \cos \theta + 4K_s \sin(2\theta)]$$

$$\alpha_1 = [-153.08\omega + D_{\lambda}^{(2)} + 2D_{\lambda}^{(4)} \cos^2 \theta] \sin(2\theta)$$

From equation 8,

$$\text{Total energy} = E(\theta) = E_0 - \frac{\alpha_1^2}{C_{11}} - \frac{\alpha_1^2 (N-2)}{2C_{22}}$$

Figure 1 shows the 3-D plot of $\frac{E(\theta)}{\omega}$ versus θ and N , for

$$\frac{J}{\omega} = \frac{D_m^{(2)}}{\omega} = \frac{H_{in}}{\omega} = \frac{H_{out}}{\omega} = \frac{K_s}{\omega} = 10 \text{ and } \frac{D_m^{(4)}}{\omega} = 5.$$

Several energy minima and maxima can be observed at different values of N . According to this graph, the magnetic easy direction depends on the number of spin layers or the thickness of the film. In addition, the variation of magnetic easy direction with the deposition temperature has been explained using Heisenberg Hamiltonian coupled with spin reorientation by us (Samarasekara & Gunawardhane, 2011, Samarasekara & Saparamadu, 2012, Samarasekara & Saparamadu, 2013). This graph is similar to the same graph plotted for thick ferromagnetic films using third order perturbed Heisenberg Hamiltonian, and the maximum value of energy is also the same in the both cases (Samarasekara, 2008). The same graph plotted for thick nickel ferrite films using second order perturbed Heisenberg Hamiltonian is similar to this graph (Samarasekara, 2010).

Figure 1: 3-D plot of $\frac{E(\theta)}{\omega}$ versus θ and N

The graph of $\frac{E(\theta)}{\omega}$ versus N is given in figure 2 for $\theta = 0$ radians. Major energy minima can be observed at $N = 5, 12, 20, \dots$ etc. Major energy maxima can be observed at $N = 15, 19, 24, \dots$ etc. This graph can be obtained by rotating above 3-D plot in MATLAB. In addition to major minima and maxima, some minor maxima and minima can be observed in figure 2.

Figure 2: Graph of $\frac{E(\theta)}{\omega}$ versus N for $\theta = 0$ radians

Figure 3 shows the $\frac{E(\theta)}{\omega}$ versus θ for $N = 5$. Energy minima of this graph can be observed at $\theta = 2.6, 5.6$ radians, ----- etc. Energy maxima of this graph can be observed at $\theta = 3.6, 5.7$ radians,----- etc. Therefore, magnetic easy directions are $\theta = 2.6, 5.6$ radians, --- etc with respect to a normal line drawn to the film plane. The same study was done for ultra thin nickel ferrite films with 2 and 3 spin layers using second order perturbed Heisenberg Hamiltonian by the authors (Samarasekara et al., 2009). Broad peaks could be observed for films with 2 spin layers (Samarasekara *et al.*, 2009). However, the same graph plotted for ferromagnetic ultra thin films with three spin layers using third order perturbed Heisenberg Hamiltonian is somewhat similar to the plot given in this case (Samarasekara & Mendoza, 2010).

Figure 3: Graph of $\frac{E(\theta)}{\omega}$ versus θ for $N = 5$

Figure 4 shows the 3-D plot of $\frac{E(\theta)}{\omega}$ versus θ and $\frac{K_s}{\omega}$ for $N = 10000$. The other

parameters were kept at $\frac{J}{\omega} = \frac{D_m^{(2)}}{\omega} = \frac{H_{in}}{\omega} = \frac{H_{out}}{\omega} = 10$ and $\frac{D_m^{(4)}}{\omega} = 5$. Energy minima

can be observed at $\frac{K_s}{\omega} = 17, 21, 25$ ----- etc. Energy maxima can be observed at $\frac{K_s}{\omega} = 19,$

23, 27, ---- etc. The same kind of 3-D graph plotted for ultra thin nickel ferrite films using third order perturbed Heisenberg Hamiltonian is entirely different from the 3-D plot given in this manuscript (Samarasekara & Mendoza, 2011). However, this graph is similar to the same kind of 3-D graph plotted for nickel ferrite thick films using third order perturbed

Heisenberg Hamiltonian (Samarasekara, 2011). Figure 5 shows the graph of $\frac{E(\theta)}{\omega}$ versus θ

for $\frac{K_s}{\omega} = 17$. Energy minima of this graph can be observed at $\theta = 2.6$ radians. Energy

maxima of this graph can be observed at $\theta = 3.6$ radians. Therefore, magnetic easy direction is $\theta = 2.6$ radians with respect to a normal line drawn to the film plane. According to our

experimental studies, magnetic properties of ferrite films depend on the stress induced anisotropy (Samarasekara & Cadieu, 2001).

Figure 4: 3-D plot of $\frac{E(\theta)}{\omega}$ versus θ and $\frac{K_s}{\omega}$ for $N = 10000$

Figure 5: Graph of $\frac{E(\theta)}{\omega}$ versus θ for $\frac{K_s}{\omega} = 17$

CONCLUSIONS

When $\frac{J}{\omega} = \frac{D_m^{(2)}}{\omega} = \frac{H_{in}}{\omega} = \frac{H_{out}}{\omega} = \frac{K_s}{\omega} = 10$, $\frac{D_m^{(4)}}{\omega} = 5$ and $N = 5$, magnetic

easy directions were found to be $\theta = 2.6$ radians, 5.6 radians, ----- etc. The angle between two consecutive easy directions is 3 radians (172 degrees).

When $\frac{J}{\omega} = \frac{D_m^{(2)}}{\omega} = \frac{H_{in}}{\omega} = \frac{H_{out}}{\omega} = 10$, $N = 10000$ and $\frac{K_s}{\omega} = 17$, magnetic easy direction

was $\theta = 2.6$ radians. Several magnetic easy and hard directions could be found in first

three graphs. According to the 3-D plot of $\frac{E(\theta)}{\omega}$ versus θ and N , the magnetic easy and

hard directions vary with the number of spin layers. Compared to this energy versus angle

curve for $N = 5$, broad peaks could be observed for films with 2 spin layers. The 3-D graph of $\frac{E(\theta)}{\omega}$ versus θ and $\frac{K_s}{\omega}$ is similar to the same kind of 3-D graph plotted for spinel nickel ferrite thick films using third order perturbed Heisenberg Hamiltonian.

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