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THIRD ORDER PERTURBED HEISENBERG HAMILTONIAN OF SPINEL FERRITE ULTRA-THIN FILMS

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Abstract

The classical Heisenberg Hamiltonian equation of spinel ferrite ultra-thin films will be solved for third order perturbation. When second order anisotropy constant do not vary within the film of N=2, the film behaves as an oriented film. But the film of N=3 does not behave as an oriented film even for invariant second order anisotropy. Also the second and third order perturbations become zero in perpendicular and in plane directions, indicating that films behave as oriented films. For N-2 film, nearest maximum and minimum can be observed at 45° and 135° , respectively. For N=3, the first nearest maximum and minimum are observed at 47° and 137° , respectively. In both cases, the angle between easy and hard direction is 90° , and the energy at hard or easy directions does not vary with angle. The 3-D plot of total energy versus angle and stress induced anisotropy indicates some energy minimums. Fourth order anisotropy slightly destroys the smoothness of the energy curve with N=3.

Keywords: Heisenberg Hamiltonian, spin, ferrites, thin films

1. Introduction:

For the first time the effect of stress induced anisotropy on total energy of spinel ferrite thin films using classical model of Heisenberg Hamiltonian with third order perturbation was investigated in detail. 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 account. The spin exchange interaction energy and dipole interaction only between two nearest spin layers and within same spin plane were considered for this simulation. Although these equations derived here can be applied for spinel ferrites such as Fe_3O_4 , $NiFe_2O_4$ and $ZnFe_2O_4$ only, these equations can not be applied for ferrites such as Lithium ferrite.

The position of octahedral and tetrahedral sites in the structure of spinel ferrites is given in detail in some early report ¹⁻⁵. Although there are many filled and vacant octahedral and tetrahedral sites in cubic spinel cell ¹, only the occupied octahedral and tetrahedral sited were used for the calculation in this report. Only few previous reports could be found on the theoretical works of ferrites ⁶. The solution of Heisenberg ferrites consist of spin exchange interaction term only has been found earlier using the retarded Green function equations ⁶. The dipole matrix elements, energy of oriented films and energy of second order perturbed ultra-thin films were derived in some of our early reports have been used for the simulations given in this report ^{7,8}.

$$H = -J \sum_{m,n} \vec{S}_{m} \cdot \vec{S}_{n} + \omega \sum_{m \neq n} \left(\frac{\vec{S}_{m} \cdot \vec{S}_{n}}{r_{mn}^{3}} - \frac{3(\vec{S}_{m} \cdot \vec{r}_{mn})(\vec{r}_{mn} \cdot \vec{S}_{n})}{r_{mn}^{5}} \right) - \sum_{m} D_{\lambda_{m}}^{(2)} (S_{m}^{z})^{2} - \sum_{m} D_{\lambda_{m}}^{(4)} (S_{m}^{z})^{4} - \sum_{m} \vec{H} \cdot \cdot \vec{S}_{m} - \sum_{m} K_{s} Sin2\theta_{m}$$
(1)

Here J, \dots $D_m^{(2)}$, $D_m^{(4)}$, 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 mth spin and the stress is m.

The cubic cell was divided into 8 spin layers with alternative A and Fe spins layers. 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 report⁷ will be used for following equations. But the angle will vary from $_{m}$ to $_{m+1}$ 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 cell⁷ and the interface of the cell, respectively. Then total energy is given by

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

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

$$-\sum_{m=1}^{N}[D_{m}^{(2)}\cos^{2}\theta_{m} + D_{m}^{(4)}\cos^{4}\theta_{m}]$$

$$-4(1-p)\sum_{m=1}^{N}[H_{in}\sin\theta_{m} + H_{out}\cos\theta_{m} + K_{s}\sin 2\theta_{m}]$$
(2)

Here the anisotropy energy term and the last term have been explained in our previous report for oriented spinel ferrite⁷. If the angle is given by $m^{=} + m$ with perturbation m, after taking the terms up to third order perturbation of ,

The total energy can be given as
$$E()=E_0+E()+E()+E()^3)$$

Here
 $E_0=-10JN+72pNJ-22Jp^2N+8Jp(N-1)-48.$ 1 -145. $\cos(2)$
+20.41 p[(N-1)+3(N-1)\cos(2]]

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$$-\cos^{2}\theta \sum_{m=1}^{N} D_{m}^{(2)} - \cos^{4}\theta \sum_{m=1}^{N} D_{m}^{(4)} - 4(1-p)N(H_{in}\sin\theta + H_{out}\cos\theta + K_{s}\sin2\theta)$$
(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)
$$E(\varepsilon^{2}) = -4Jp\sum_{m=1}^{N-1}(\varepsilon_{n} - \varepsilon_{m})^{2} + 290.5\omega\cos(2\theta)\sum_{m=1}^{N}\varepsilon_{m}^{2} - 10.2\omega p\sum_{m=1}^{N-1}(\varepsilon_{n} - \varepsilon_{m})^{2} - 30.6\omega p\cos(2\theta)\sum_{m=1}^{N-1}(\varepsilon_{n} + \varepsilon_{m})^{2} - (\sin^{2}\theta - \cos^{2}\theta)\sum_{m=1}^{N}D_{m}^{(2)}\varepsilon_{m}^{2} + 2\cos^{2}\theta(\cos^{2}\theta - 3\sin^{2}\theta)\sum_{m=1}^{N}D_{m}^{(4)}\varepsilon_{m}^{2} + 4(1-p)[\frac{H_{in}}{2}\sin\theta\sum_{m=1}^{N}\varepsilon_{m}^{2} + \frac{H_{out}}{2}\cos\theta\sum_{m=1}^{N}\varepsilon_{m}^{2} + 2K_{s}\sin 2\theta\sum_{m=1}^{N}\varepsilon_{m}^{2}]$$
(5)

$$E(\varepsilon^{3}) = 10.2 p \omega \sin 2\theta \sum_{m,n=1}^{N} (\varepsilon_{m} + \varepsilon_{n})^{3} - 193.66 \omega \sin 2\theta \sum_{m=1}^{N} \varepsilon_{m}^{3} - \frac{4}{3} \cos \theta \sin \theta \sum_{m=1}^{N} D_{m}^{(2)} \varepsilon_{m}^{3} - 4 \cos \theta \sin \theta (\frac{5}{3} \cos^{2} \theta - \sin^{2} \theta) \sum_{m=1}^{N} D_{m}^{(4)} \varepsilon_{m}^{3} + 4(1-p) [\frac{H_{in}}{6} \cos \theta \sum_{m=1}^{N} \varepsilon_{m}^{3} - \frac{H_{out}}{6} \sin \theta \sum_{m=1}^{N} \varepsilon_{m}^{3} + \frac{4K_{s}}{3} \cos 2\theta \sum_{m=1}^{N} \varepsilon_{m}^{3}]$$

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()= $\vec{\alpha}.\vec{\varepsilon}$ Here $\vec{\alpha}(\varepsilon) = \vec{B}(\theta) \sin 2\theta$ are the terms of matrices with $B_{\lambda}(\theta) = -122.46\omega p + D_{\lambda}^{(2)} + 2D_{\lambda}^{(4)}\cos^{2}\theta$ (6) 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, $C_{m, m+1} = 8Jp + 20.4 p - 61.2p cos(2)$ For m=1 and N, $C_{mm} = -8Jp-20.4 \quad p-61.2p \quad \cos(2) + 581 \quad \cos(2) - 2(\sin^2\theta - \cos^2\theta) D_m^{(2)}$

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 $+4\cos^{2}\theta(\cos^{2}\theta-3\sin^{2}\theta) D_{m}^{(4)}+4(1-p)[H_{in}\sin\theta+H_{out}\cos\theta+4K_{s}\sin(2\theta)]$ (7) For m=2, 3, ----, N-1 $C_{mm}=-16Jp-40.8 \quad p-122.4p \quad \cos(2)+581 \quad \cos(2) - 2(\sin^{2}\theta-\cos^{2}\theta) D_{m}^{(2)}$ $+4\cos^{2}\theta(\cos^{2}\theta-3\sin^{2}\theta) D_{m}^{(4)}+4(1-p)[H_{in}\sin\theta+H_{out}\cos\theta+4K_{s}\sin(2\theta)]$ Otherwise, $C_{mn}=0$ Also $E(\varepsilon^{3}) = \varepsilon^{2}\beta.\overline{\varepsilon}$ Here matrix elements of matrix can be given as following. When m=1 and N, $\beta_{mm} = -193.66\omega\sin 2\theta + 10.2p\omega\sin 2\theta - \frac{4}{3}\cos\theta\sin\theta D_{m}^{(2)}$

$$-4\cos\theta\sin\theta(\frac{5}{3}\cos^2\theta - \sin^2\theta)D_m^{(4)} + 4(1-p)\left[\frac{H_{in}}{6}\cos\theta - \frac{H_{out}}{6}\sin\theta + \frac{4K_s}{3}\cos2\theta\right]$$

When m=2, 3, -----, N-1

$$\beta_{mm} = -193.66\omega \sin 2\theta + 20.4p\omega \sin 2\theta - \frac{4}{3}\cos\theta \sin\theta D_m^{(2)}$$
$$-4\cos\theta \sin\theta (\frac{5}{3}\cos^2\theta - \sin^2\theta) D_m^{(4)} + 4(1-p) [\frac{H_{in}}{6}\cos\theta - \frac{H_{out}}{6}\sin\theta + \frac{4K_s}{3}\cos 2\theta]$$
$$\beta_{m,m+1} = 30.6p\omega \sin 2\theta$$
(8)

Otherwise $_{nm}=0$. Also $_{nm}=_{mn}$ and matrix is symmetric.

Therefore, the total magnetic energy given in equation 2 can be deduced to

$$E() = E_0 + \vec{\alpha}.\vec{\varepsilon} + \frac{1}{2}\vec{\varepsilon}.C.\vec{\varepsilon} + \varepsilon^2 \beta.\vec{\varepsilon}$$
(9)

Because the derivation of a final equation for with the third order of in above equation is tedious, only the second order of will be considered for following derivation.

Then E()=E₀+
$$\vec{\alpha}.\vec{\varepsilon}$$
+ $\frac{1}{2}\vec{\varepsilon}.C.\vec{\varepsilon}$

Using a suitable constraint in above equation⁸, it is possible to show that $\vec{\varepsilon} = -C^+ \cdot \cdot \cdot \vec{\alpha}$ Here C⁺ is the pseudo-inverse given by

$$C.C^+ = 1 - \frac{E}{N}.$$
 (10)

E is the matrix with all elements given by $E_{mn}=1$.

After using in equation 9, E()=E₀ - $\frac{1}{2}\vec{\alpha}.C^+.\vec{\alpha} - (C^+\alpha)^2\vec{\beta}(C^+\alpha)$ (11)

3. Results and discussion:

The energy given in above equation 11 will be calculated for film with two layers (N=2). The equations will be proven under the assumption of $D_1^{(2)}=D_2^{(2)}$ and $D_1^{(4)}=D_2^{(4)}$. According to above equations, $C_{11}=C_{22}$ and $C_{12}=C_{21}$.

Therefore from equation 10,
$$C_{12}^{+} = C_{21}^{+} = \frac{1}{2(C_{21} - C_{22})} = -C_{11}^{+} = -C_{22}^{+}$$

Using above results, $\vec{\alpha}.C^+.\vec{\alpha} = (\alpha_1 - \alpha_2)^2 C^+_{11}$

But from equation 6, for a film with two layers
$$\[\] \]$$
 Therefore, $\vec{\alpha}.C^+.\vec{\alpha} = 0$.
Also $(C^+\alpha)^2 \vec{\beta}(C^+\alpha) = (C_{11}^+ + C_{12}^+ + 2)^2 [-11(C_{11}^+ + C_{12}^+ + 2) + (C_{21}^+ + C_{22}^+ + 2)] + (C_{21}^+ + C_{22}^+ + 2)^2 [-21(C_{11}^+ + C_{12}^+ + 2) + (C_{21}^+ + C_{22}^+ + 2)] + (C_{21}^+ + C_{22}^+ + 2)]$

$$(12)$$

If $C_{12}^{+} = -C_{11}^{+}$ and $C_{21}^{+} = -C_{22}^{+}$, and then $(C^{+}\alpha)^{2} \vec{\beta}(C^{+}\alpha) = 0$. Therefore, when anisotropy constants do not vary, the energy given in equation 11 is deduced to the energy of a perfectly oriented film. But when anisotropy constant varies within the film, $C_{12}=C_{21}$ and $C_{22} \neq C_{11}$.

Therefore,
$$C^{+}_{11} = -C^{+}_{12} = \frac{C_{22} + C_{21}}{2(C_{11}C_{22} - C_{21}^{2})}$$
 and $C^{+}_{21} = -C^{+}_{22} = \frac{C_{21} + C_{11}}{2(C_{21}^{2} - C_{11}C_{22})}$.

Hence, $\vec{\alpha}.C^+.\vec{\alpha} = (\alpha_1 - \alpha_2) (C^+{}_{21}\alpha_2 - C^+{}_{12}\alpha_1)$ If all the terms are considered, the C₁₁C₂₂ product will cor

If all the terms are considered, the $C_{11}C_{22}$ product will consist of 80 terms. Therefore, only the magnetic exchange energy, second order anisotropy, and the stress induced anisotropy terms will be considered for this simulation. For Ni ferrite, p=2.5.

Then from equation 7,
$$C_{11} = -20J + 2\cos 2\theta D_1^{(2)} - 24K_s \sin 2\theta$$

 $C_{22} = -20J + 2\cos 2\theta D_2^{(2)} - 24K_s \sin 2\theta$, and $C_{12}=C_{21}=8Jp=20J$
 $\beta_{11} = -0.67\sin 2\theta D_1^{(2)} - 8K_s \cos 2\theta$
 $\beta_{22} = -0.67\sin 2\theta D_2^{(2)} - 8K_s \cos 2\theta$, and $_{12}=_{21}=0$
 $\alpha_1 = D_1^{(2)}\sin 2\theta$, and $\alpha_2 = D_2^{(2)}\sin 2\theta$
Finally from equation 12,

$$(C^{+}\alpha)^{2}\vec{\beta}(C^{+}\alpha) = (\alpha_{1} - \alpha_{2})^{3} \frac{\beta_{11}(C_{22} + C_{21})^{3} + \beta_{22}(C_{21} + C_{11})^{3}}{8(C_{11}C_{22} - C_{21}^{2})^{3}}$$

When and second and third order perturbation terms become zero and the film behaves as an oriented film. From equation 3,

 $E_0 = 85J - \cos^2 \theta [D_1^{(2)} + D_2^{(2)}] + 12K_s \sin(2)$

The total energy can be found from equation 11.

The graph between
$$\frac{E(\theta)}{J}$$
 and is given in figure 1, for $\frac{D_1^{(2)}}{J} = \frac{K_s}{J} = 10, \frac{D_2^{(2)}}{J} = 5$.

Consecutive maximum and minimum can be observed at 45° and 135° , respectively. Two nearest maximums can be observed at 45° and 225° . The angle between easy and hard direction is 90° in this case. The energy at hard or easy directions does not vary with angle. This energy curve is smoother than that of ferromagnetic ultra-thin film with third order perturbation ⁹. Although the separation between maximum and minimum is 90° for ferromagnetic ultra-thin film with third order perturbation, the positions of maximum and minimum of this curve are different from those of ferromagnetic ultra-thin film with third order perturbation.



When
$$\frac{K_s}{J}$$
 is a variable, the 3-D plot of $\frac{E(\theta)}{J}$ versus and $\frac{K_s}{J}$ is given in figure 2. At

some stress values the energy minimums can be observed in certain directions indicating that the film can be easily oriented in that particular direction by applying a certain stress.

When N=3, the each C^+_{nm} element found using equation 10 is consist of more than 20 terms. To avoid this problem, matrix elements were found using C.C⁺=1. Then C^+_{mn} is given by $C^+_{mn} = \frac{cofactorC_{nm}}{\det C}$. Under this condition, $\vec{E}.\vec{\alpha} = 0$, and the average value of first order perturbation is zero. The second order anisotropy constant is assumed to be a constant within the film for the convenience.

Then
$$C_{12}=C_{21}=C_{23}=C_{32}=20J$$
, $C_{13}=C_{31}=0$, $_{1}=_{2}=_{3}=D_{m}^{(2)}\sin 2\theta$.
 $C_{11}=C_{33}=-20J+2(\cos 2\theta) D_{m}^{(2)}-24K_{s}\sin(2)$
 $C_{22}=-40J+2(\cos 2\theta) D_{m}^{(2)}-24K_{s}\sin(2)$ and
Therefore, $C_{11}^{+}=\frac{C_{11}C_{22}-C_{32}^{-2}}{C_{11}^{-2}C_{22}-2C_{32}^{-2}C_{11}}=C_{33}^{+}$, $C_{13}^{+}=\frac{C_{32}^{-2}}{C_{11}^{-2}C_{22}-2C_{32}^{-2}C_{11}}=C_{31}^{+}$
 $C_{12}^{+}=\frac{-C_{32}C_{11}}{C_{11}^{-2}C_{22}-2C_{32}^{-2}C_{11}}=C_{23}^{+}=C_{32}^{+}$, $C_{22}^{+}=\frac{C_{11}^{-2}}{C_{11}^{-2}C_{22}-2C_{32}^{-2}C_{11}}$





Figure 2. The 3-D plot of $\frac{E(\theta)}{J}$ versus and $\frac{K_s}{J}$ for N=2 with second order anisotropy

The total energy can be found using equation 11. The graph between $\frac{E(\theta)}{J}$ and is given in

figure 3, for $\frac{D_m^{(2)}}{J} = \frac{K_s}{J} = 10$. The first nearest maximum and minimum are observed at 47^o and 137^o, respectively. Two nearest maximums are at 47^o and 227^o. The angle between easy and hard directions is 90^o. The energy at easy or hard directions does not vary with angle. Because some sudden overshooting can be observed for ferromagnetic ultra-thin film with third order perturbation, this energy curve is smoother than that ⁹. Also the positions of maximum and minimum of this curve are different from those of ferromagnetic ultra-thin film with third order perturbation. Energy of this film is smaller than that of ferromagnetic ultra-thin film with third order perturbation. When both second and fourth order anisotropies are taken into account,

 $C_{12}=C_{21}=C_{23}=C_{32}=20J, C_{13}=C_{31}=0$

$$I = 2 = 3 = (2D_m^{(4)} \cos^2 \theta + D_m^{(2)}) \sin 2\theta.$$

$$C_{11} = C_{33} = -20J + 2(\cos 2\theta) D_m^{(2)} - 24K_s \sin(2) + 4\cos^2 \theta(\cos^2 \theta - 3\sin^2 \theta) D_m^{(4)}$$

$$C_{22} = -40J + 2(\cos 2\theta) D_m^{(2)} - 24K_s \sin(2) + 4\cos^2 \theta(\cos^2 \theta - 3\sin^2 \theta) D_m^{(4)}$$

$$E_0 = 137.5J - 3\cos^2 \theta D_m^{(2)} - 3\cos^4 \theta D_m^{(4)} + 18K_s \sin(2)$$

$$\beta_{11} = \beta_{22} = \beta_{33} = -0.67\sin 2\theta D_m^{(2)} - 8K_s \cos 2\theta - 4\cos \theta \sin \theta(\frac{5}{3}\cos^2 \theta - \sin^2 \theta) D_m^{(4)}$$

$$I_{2} = 2I = I_{3} = 3I = 32 = 23 = 0$$



Figure 3. Graph between $\frac{E(\theta)}{J}$ and for N=3 with second order anisotropy only

Similarly, total energy can be found from equation 11. The graph between $\frac{E(\theta)}{J}$ and is given in figure 4, for $\frac{D_m^{(2)}}{J} = \frac{K_s}{J} = 10$ and $\frac{D_m^{(4)}}{J} = 5$. Although the angles at maximum and minimum remain same as those of figure 1, some sudden change of energy can be observed at the center part of the curve. Therefore, introducing the fourth order anisotropy slightly destroys the smoothness of the curve. But this curve is smoother than that of ferromagnetic ultra-thin film with third order perturbation⁹.



Figure 4. Graph between $\frac{E(\theta)}{J}$ and for N=3 with second and fourth order anisotropies

4. Conclusion:

Film of N=2 and N=3 behave as an oriented and non-oriented film for invariant second order anisotropy, respectively. Also the second and third order perturbations become zero in $\int_{-\infty}^{\infty}$ and directions, and films behave as oriented films. Nearest maximum and minimum of N=2 ultra-thin film can be observed at 45[°] and 135[°], respectively. The first nearest maximum and minimum of N=3 ultrathin film are observed at 47[°] and 137[°], respectively. In both cases, the angle between easy and hard direction is 90[°], and the energy at hard or easy directions does not vary with angle. The 3-D plot of total energy versus angle and stress induced anisotropy shows some energy minimums implying that the ultra-thin ferrite film can be easily oriented in certain direction by applying particular stresses. Although this simulation was performed for $\frac{D_1^{(2)}}{J} = \frac{K_s}{J} = 10, \frac{D_2^{(2)}}{J} = 5$ values of Nickel ferrite only,

this simulation can be carried out for any value of any spinel ferrite.

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