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INVERSE SPINEL COBALT FERRITE ULTRA-THIN FILMS AS EXPLAINED BY SECOND ORDER PERTURBED HEISENBERG HAMILTONIAN

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Abstract
The second order perturbed Heisenberg Hamiltonian has been employed to investigate the magnetic properties of ultra-thin cobalt ferrite films with three unit cells. The variation of classical magnetic energy with second order magnetic anisotropy, spin exchange interaction and angle has been plotted in order to observe the easy and hard directions of cobalt ferrite ultra-thin films. According to our 3-D and 2-D plots, the ultra-thin cobalt ferrite film can be easily aligned along some particular directions at some certain values of second order magnetic anisotropy and spin exchange interaction. In addition, the energy maximums can be also observed at some certain values of second order magnetic anisotropy and spin exchange interaction indicating that it is difficult to align the film at these values of energy parameters. However, the easy and hard directions described at this manuscript are valid only for the special values of the energy parameters used for this simulation.

Keywords: *Cobalt ferrite, ultra-thin films, Heisenberg Hamiltonian, perturbation*

1. Introduction:

The cobalt ferrite films are vastly applied in the products of microwave devices and magnetic memory devices. Cobalt ferrite magnetic thin films have been synthesized using electrophoresis [1], pulsed laser deposition [2, 3, 4], electrostatic spray method [5]. However, theoretical model of cobalt ferrite films haven't been developed using any theoretical model. The thin and thick films of Nickel ferrite have been theoretically investigated using non-perturbed [6], 2nd order [7, 8] and 3rd order [9, 10] perturbed Heisenberg Hamiltonian by us previously. The variation of magnetic energy with number of layers & stress induced anisotropy were plotted and the easy & hard directions were found using non-perturbed modified Heisenberg Hamiltonian [6]. The magnetic energy versus angle of spins & stress induced anisotropy were plotted and the easy & hard directions were calculated using the 2nd order perturbed modified Heisenberg Hamiltonian [7, 8]. Furthermore, the variation of magnetic energy with number of layers & stress induced anisotropy were plotted in order to find the easy & hard directions using the 3rd order perturbed modified Heisenberg Hamiltonian [9, 10].

In this manuscript, the variation of easy and hard directions with second order magnetic anisotropy has been investigated using second order perturbed classical Heisenberg Hamiltonian model modified by adding stress induced anisotropy. MATLAB software package was employed to plot all the 2-D and 3-D graphs. Only the effect of few parameters in Heisenberg Hamiltonian was taken into account to avoid some tedious derivations. Although it is difficult to prepare ultra-thin films with three layers in practice, theoretical studies of ultra-thin films were found to be really interesting.

2. Model:

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

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$$H= - J\sum_{m,n}\vec{S}_m.\vec{S}_n + \omega\sum_{m\neq n}\left(\frac{\vec{S}_m.\vec{S}_n}{r_{mn}^{\ 3}} - \frac{3(\vec{S}_m.\vec{r}_{mn})(\vec{r}_{mn}.\vec{S}_n)}{r_{mn}^{\ 5}}\right) - \sum_m D_{\lambda_m}^{\ (2)}(S_m^{\ z})^2 - \sum_m D_{\lambda_m}^{\ (4)}(S_m^{\ z})^4$$
$$-\sum_m \vec{H}.\vec{S}_m - \sum_m K_s Sin2\theta_m \qquad \textbf{(1)}$$

Here J, ω, θ, $D_m^{(2)}, D_m^{(4)}, H_{in}, H_{out}, K_s$, 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 m^{th} spin and the stress is θ_m.

The cubic cell was divided into 8 spin layers with alternative A and Fe spins layers [6]. 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[6] 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.
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 and the interface of the cell, respectively.
Then the total energy is given by

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

Here the anisotropy energy term and the last term have been explained in our previous report for oriented spinel ferrite [6]. If the angle is given by $\theta_m=\theta+\varepsilon_m$ with perturbation ε_m, after taking the terms up to second order perturbation of ε only,
The total energy can be given as $E(\theta)=E_0+E(\varepsilon)+E(\varepsilon^2)$
The non-perturbed term is given by

$$E_0= -10JN+72pNJ-22Jp^2N+8Jp(N-1)-48.415\omega N-145.245\omega N\cos(2\theta)$$
$$+20.41\omega p[(N-1)+3(N-1)\cos(2\theta)]$$
$$-\cos^2\theta\sum_{m=1}^{N}D_m^{\ (2)} - \cos^4\theta\sum_{m=1}^{N}D_m^{\ (4)} - 4(1-p)N(H_{in}\sin\theta + H_{out}\cos\theta + K_s\sin 2\theta) \quad \textbf{(3)}$$

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$$E(\varepsilon) = 290.5\omega \sin(2\theta)\sum_{m=1}^{N}\varepsilon_m - 61.23\omega p \sin(2\theta)\sum_{m=1}^{N-1}(\varepsilon_m + \varepsilon_n)$$
$$+\sin 2\theta \sum_{m=1}^{N} D_m^{(2)}\varepsilon_m + 2\cos^2\theta \sin 2\theta \sum_{m=1}^{N} D_m^{(4)}\varepsilon_m$$
$$+4(1-p)[-H_{in}\cos\theta\sum_{m=1}^{N}\varepsilon_m + H_{out}\sin\theta\sum_{m=1}^{N}\varepsilon_m - 2K_s\cos 2\theta\sum_{m=1}^{N}\varepsilon_m] \quad \textbf{(4)}$$

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

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(ε)=$\vec{\alpha}.\vec{\varepsilon}$

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

$$B_\lambda(\theta) = -122.46\omega p + D_\lambda^{(2)} + 2D_\lambda^{(4)}\cos^2\theta \quad \textbf{(6)}$$

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+20.4ωp-61.2pωcos(2θ)

For m=1 and N,

C_{mm}= -8Jp-20.4ωp-61.2pωcos(2θ)+581ωcos(2θ) $-2(\sin^2\theta - \cos^2\theta)\ D_m^{(2)}$

$$+4\cos^2\theta(\cos^2\theta - 3\sin^2\theta)\ D_m^{(4)} + 4(1-p)[H_{in}\sin\theta + H_{out}\cos\theta + 4K_s\sin(2\theta)] \quad \textbf{(7)}$$

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

C_{mm}= -16Jp-40.8ωp-122.4pωcos(2θ)+581ωcos(2θ) $-2(\sin^2\theta - \cos^2\theta)\ D_m^{(2)}$

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

Otherwise, C_{mn}=0

Therefore, the total energy can be given as

E(θ)=E_0+$\vec{\alpha}.\vec{\varepsilon} + \frac{1}{2}\vec{\varepsilon}.C.\vec{\varepsilon}$ =$E_0 - \frac{1}{2}\vec{\alpha}.C^{+}.\vec{\alpha}$ **(8)**

Here C^+ is the pseudo-inverse given by

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

Here E is the matrix with all elements E_{mn}=1.

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3. Results and discussion:

In the inverse spinel structure of $Fe^{3+(}Co^{2+}Fe^{3+)}O_4$, the spins of Co^{2+} and Fe^{3+} are $3\mu_B$ and $5\mu_B$, respectively. So the value of p for cobalt ferrite is 1.67. When N=3, the each C^+_{nm} element found using equation 9 is contained more than 20 terms. To avoid this problem, matrix elements were found using $C.C^+=1$. Then C^+_{mn} is given by $C^+{}_{mn} = \frac{cofactor C_{nm}}{\det C}$. 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 an invariant for the convenience.

Then $C_{11}=C_{33}$, $C_{12}=C_{21}=C_{23}=C_{32}$, $C_{13}=C_{31}=0$, $\alpha_1=\alpha_2=\alpha_3$.

$C_{11}=C_{33}= -13.36J-34.07\omega+478.8\omega\cos(2\theta)+2(\cos 2\theta)\ D_m^{(2)}-10.72K_s\sin(2\theta)$

$C_{22}= -26.72J-68.14\omega+376.6\omega\cos(2\theta)+2(\cos 2\theta)\ D_m^{(2)}-10.72K_s\sin(2\theta)$

$C_{32}=13.36J+34.07\omega-102.2\omega\cos(2\theta)$

$\alpha_1=[-204.51\omega+D_1^{(2)}]\sin(2\theta)$

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

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

The total energy can be found using following equation.

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

Here $E_0= 173.37J-145.245\omega-435.735\omega\cos(2\theta)+68.17\omega[1+3\cos(2\theta)]$

$\quad -3D_1^{(2)}\cos^2\theta\ +2.68K_s\sin(2\theta)$

Figure 1 shows the 3-D plot of $\frac{E(\theta)}{\omega}$ versus $\frac{D^{(2)}}{\omega}$ and angle, when $\frac{J}{\omega}$ and $\frac{K_s}{\omega}$ are kept at 10. The graph indicates many energy maximums and minimums at certain values of angle and second order magnetic anisotropy. Energy maximums can be observed at $\frac{D^{(2)}}{\omega}$=2, 5, 9, ---etc, indicting that it is difficult to align the magnetization of cobalt ferrite film along hard direction at these particular values of $\frac{D^{(2)}}{\omega}$. However, energy minimums can be seen at $\frac{D^{(2)}}{\omega}$=3, 8, ---etc, implying that the film can be easily aligned along magnetically easy direction at these values of second order magnetic anisotropy.

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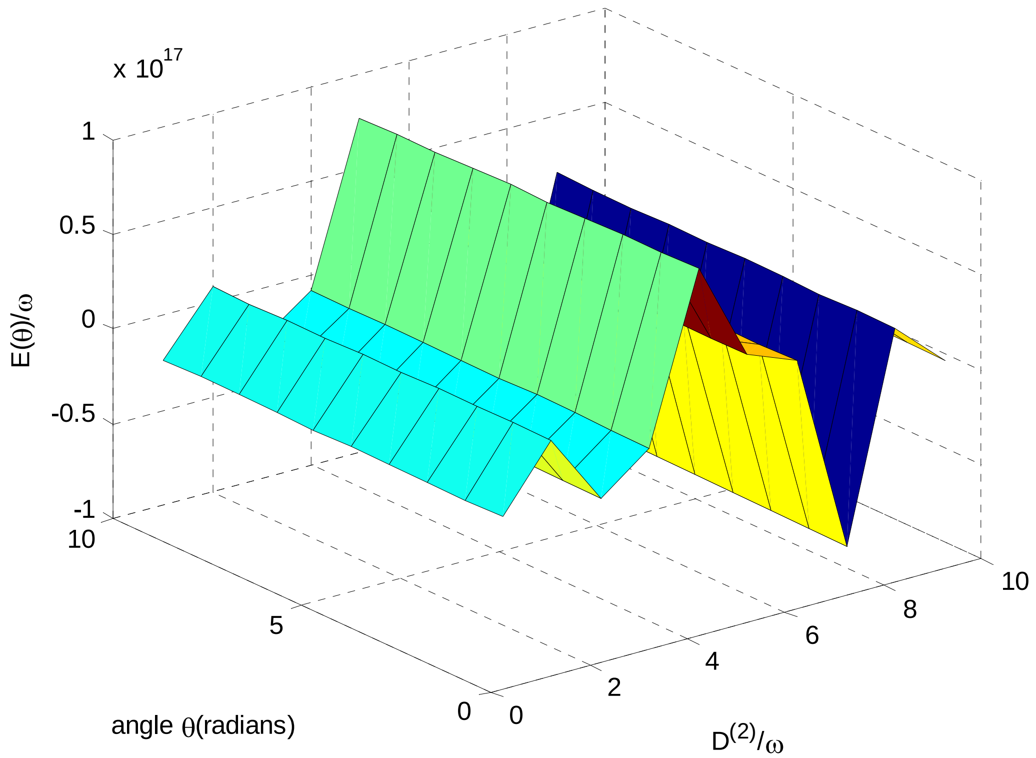

Figure 1: 3-D plot of $\frac{E(\theta)}{\omega}$ versus $\frac{D^{(2)}}{\omega}$ and angle for N=3.

The 3-D plot of $\frac{E(\theta)}{\omega}$ versus $\frac{J}{\omega}$ and angle is shown in figure 2 for $\frac{D^{(2)}}{\omega}=\frac{K_s}{\omega}$=10. According to this graph, several energy minimums and maximums can be observed at certain values of spin exchange spin interaction (J). Energy minimums of the graph can be obtained at $\frac{J}{\omega}$=3.9, 7, ---etc, by implying that the film can be easily oriented along easy direction at these values of spin exchange interaction. Maximums can be seen at $\frac{J}{\omega}$=9.9, 5.9, ---etc, by indicating that it is hard to magnetize the film along the magnetically hard direction at these values of $\frac{J}{\omega}$.

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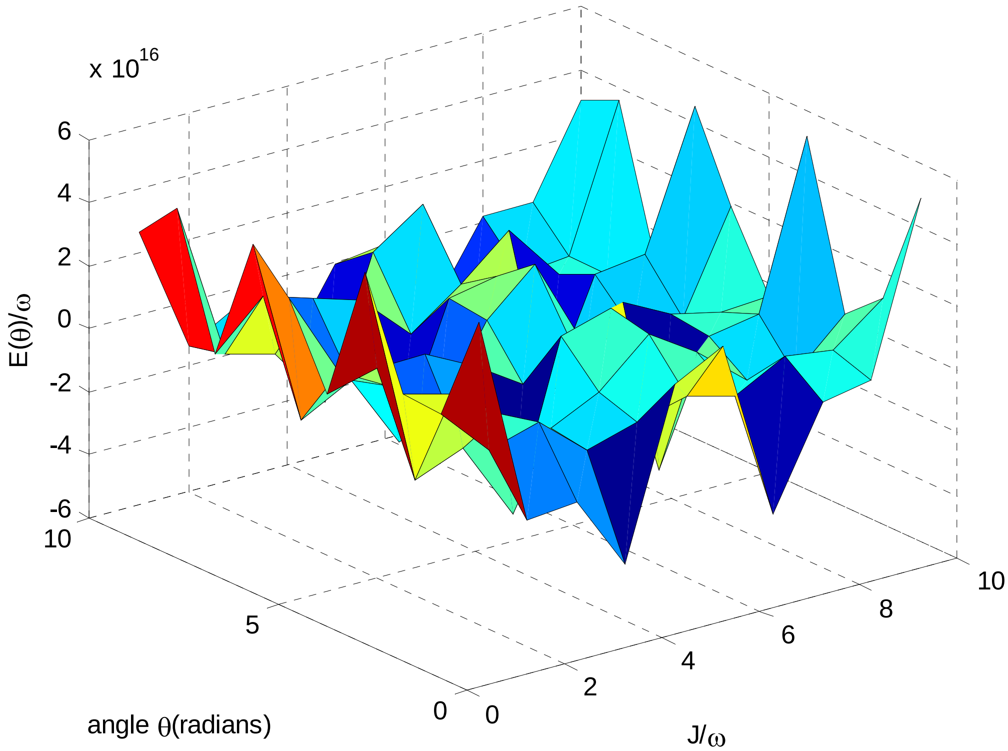

Figure 2: 3-D plot of $\frac{E(\theta)}{\omega}$ versus $\frac{J}{\omega}$ and angle for N=3.

Graph of energy versus angle has been plotted in figure 3. Because energy is minimum at $\frac{D^{(2)}}{\omega}$=3 and $\frac{J}{\omega}$=7, this graph has been plotted for $\frac{D^{(2)}}{\omega}$=3 and $\frac{J}{\omega}$=7 in order to determine the magnetically easy direction. According to this plot, the film can be easily oriented in directions given by θ=2.2 and 5.34 radians. But it is hard to magnetize film along the directions of θ=0.6 and 3.77 radians even at these values of $\frac{D^{(2)}}{\omega}$ and $\frac{J}{\omega}$ corresponding to the lowest energy points in previous 3-D plots given in figure 1 and 2.

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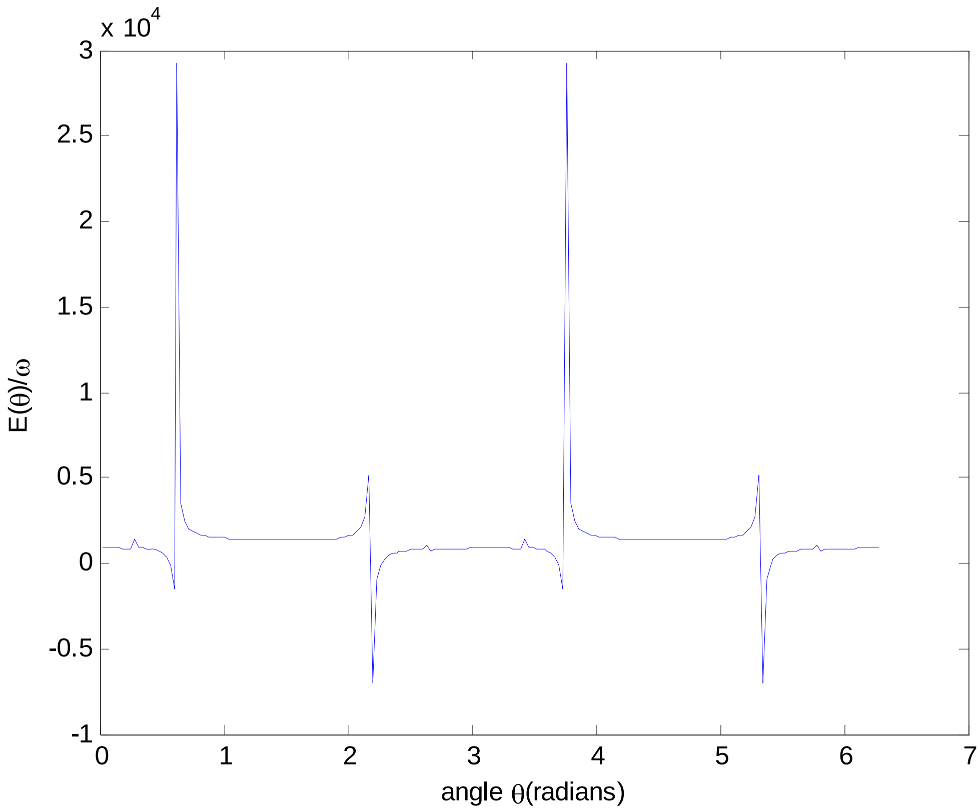

Figure 3: Graph of energy versus angle at $\frac{D^{(2)}}{\omega}$=3 and $\frac{J}{\omega}$=7.

4. Conclusion:

For $\frac{J}{\omega}=\frac{K_s}{\omega}$=10, the ultra-thin cobalt ferrite film with a thickness of three unit cells can be easily oriented along some particular directions at the values of second order magnetic anisotropy given by $\frac{D^{(2)}}{\omega}$=3, 8, ---etc. For $\frac{D^{(2)}}{\omega}=\frac{K_s}{\omega}$=10, film can be easily oriented along magnetically easy direction at $\frac{J}{\omega}$=3.9, 7, ---etc. At $\frac{D^{(2)}}{\omega}$=3 and $\frac{J}{\omega}$=7, the easy directions of film is given by θ=126 and 306 degrees. The angle between these two easy directions is 180 degrees as expected. According to 2-D plot, angle for first hard direction is 34.4 degrees. The angle between first easy and hard directions is approximately 90 degrees as usual. So the theoretical data obtained in this manuscript well agree with the normal behavior of magnetic easy and hard directions.

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