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EASY AXIS ORIENTATION OF BARIUM HEXA-FERRITE FILMS AS EXPLAINED BY SPIN REORIENTATION

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Abstract

The classical model of Heisenberg Hamiltonian was successfully employed to investigate the easy axis orientation of barium ferrite with hexagonal structure. First the magnetic dipole interaction energy between spins of Fe^{+3} *ions was calculated for the barium hexaferrite film. Then the average values of in plane and out of plane spin components were determined using the energy of the ferrite film, which was obtained by means of Heisenberg Hamiltonian. Finally, the variation of these average spin components with temperature was plotted in order to investigate the temperature at which these average spin components approach zero. The experimental value of the temperature (1000 °C), at which the easy axis of barium hexa-ferrite films deposited on MgO and* Al_2O_3 *substrates by thermal spray technique orients in the plane and out of plane of the film, could be explained using this model. The Heisenberg Hamiltonian without perturbation terms was considered in this manuscript.*

Keywords: *Barium ferrite, thin films, spins, easy axis orientation*

1. Introduction:

Barium ferrite belonging to the M-type ferrites category has a hexagonal structure. It is a uniaxial hard ferrite material with easy axis oriented along the C-axis of the hexagonal cell. Because of these unique properties of hexagonal ferrite materials, they are prime candidates in the applications of magnetic memory devices and microwave devices. Thin films of hexagonal ferrites have been fabricated using rf sputtering [1, 2], isothermal dipping method [3], targets facing type sputtering [4] and thermal spray [6]. In plane [1] and out of plane [4] easy axis oriented hexagonal ferrite films have been synthesized [6]. The orientation of easy axis vastly depends on the substrate temperature [1]. However, the orientation of easy axis depends on the orientation of substrate [5, 6], type of sputtering gas, sputtering pressure, separation between the substrate and target, and annealing conditions too. The easy axis orientation of cubic structured ferromagnetic thin films such as $CoPt_3$ grown on WSe_2 substrates and Fe-rich germanide films synthesized on Ge(001) substrates has been described using the classical Heisenberg Hamiltonian and the same theoretical approach by us previously [7].

Although the easy axis oriented hexaferrites find potential applications in magnetic memory and microwave devices, it is difficult to find detailed theoretical explanation of easy axis orientation of hexaferrites. For the first time, the orientation of easy axis of strontium ferrite thin films has been theoretically explained using Heisenberg Hamiltonian by us. First the matrix related to the dipole interaction energy between spins in hexagonal ferrite lattice was determined. Only the magnetic dipole interactions between Fe^{+3} ions were taken into account, because the magnetic moments of oxygen and Ba ions are zero due to the unavailability of unpaired electrons. As well as the interactions between spins within the same cell and the interactions between adjacent cells were considered in order to extend the investigation of dipole interactions for the whole ferrite film. Because the orientation depends on the type of the substrate, the simulations were carried out to explain the easy axis orientation of barium ferrite films experimentally grown on Al_2O_3 and MgO substrates [6]. In addition, the magnetic energy of Ni ferrite films belonging to nonuniaxial soft magnetic category was determined for oriented [8], 2nd order perturbed [9] and 3rd order perturbed cases [10].

2. Model:

The coordinates (x, y, z) of Fe $^{+3}$ ions in the cell of barium ferrite can be given as below [11].

Spin	x	y	z
up	0	0	1/4
up	0	0	1/2
down	2/3	1/3	0.525
down	1/3	2/3	0.300
up	1/3	1/6	0.400

Table 1: Coordinates and orientations of spins Fe^{+3} ions in Barium ferrite lattice

Then the magnetic dipole interaction energy between two spins can be found using following equation.

$$E_{dipole} = \omega \vec{S}_i . W(r_{ij}) . \vec{S}_j \qquad \textbf{(1)}$$

Here $$W(r) = \frac{1}{r^3} \begin{pmatrix} 1-3\hat{r}_x^{\,2} & -3|\hat{r}_y||\hat{r}_x| & -3|\hat{r}_z||\hat{r}_x| \\ -3|\hat{r}_x||\hat{r}_y| & 1-3\hat{r}_y^{\,2} & -3|\hat{r}_z||\hat{r}_y| \\ -3|\hat{r}_x||\hat{r}_z| & -3|\hat{r}_y||\hat{r}_z| & 1-3\hat{r}_z^{\,2} \end{pmatrix} \qquad \textbf{(2)}$$

and $\omega = \frac{\mu_0 \mu^2}{4\pi a^3}$.

Finally the total magnetic dipole energy of a Barium hexaferrite thin film with N cells is given by following equations after considering the magnetic dipole interactions within one cell and between two adjacent cells.

$$E_{dipole} = \omega S^2 [N(88.3197\sin^2\theta + 11.3541\sin\theta\cos\theta - 127.9435\cos^2\theta) + (N-1)(93.0605\sin^2\theta + 25.3002\sin\theta\cos\theta - 15.423\cos^2\theta)] \qquad \textbf{(3)}$$

Here the magnetic dipole interactions between spins in adjacent cells were considered only when the separation between two spins is less than 0.7945, which is the maximum separation between two spins within one cell. S denotes the value of spin in one Fe^{+3} ion.

Then the total magnetic energy can be found by applying following Heisenberg Hamiltonian for the whole barium ferrite film.

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

Within a single domain, M is parallel to the spin. If stress is applied normal to the film plane, then θ_m is the angle between the normal to the film plane and the local spin. So θ is the angle between local magnetization (M) and the stress. Here the last term indicates the change of magnetic energy under the influence of a stress. K_s depends on the product of magnetostriction coefficient (λ_s) and the stress (σ). K_s can be positive or negative depending on the type of stress whether it is compressive or tensile. Integer m and n denote the indices of planes, and they vary from 1 to N for a film with N number of layers. First, second, third and fourth terms represent the spin exchange interaction, magnetic dipole interaction, second order anisotropy and fourth order anisotropy, respectively. $\vec{H}$ is the net internal magnetic field due to the surrounding spins.

The second term in equation (4) is given by equation (3). So finally the total magnetic energy given in equation (4) can be deduced to

$$E(\theta) = 3NJ + 5(N-1)J + \omega[N(88.3197\sin^2\theta + 11.3541\sin\theta\cos\theta - 127.9435\cos^2\theta)$$

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$$+(N-1)\,(93.0605\sin^2\theta + 25.3002\sin\theta\cos\theta - 15.423\cos^2\theta)]$$

$$-\cos^2\theta\sum_{m=1}^{N} D_m^{(2)} - \cos^4\theta\sum_{m=1}^{N} D_m^{(4)} + 3N(H_{in}\sin\theta + H_{out}\cos\theta + K_s\sin^2\theta) \qquad \textbf{(5)}$$

This is the total magnetic energy per unit spin. Here $\sum_{m=1}^{N} D_m^{(2)}$ and $\sum_{m=1}^{N} D_m^{(4)}$ are the total second and fourth order anisotropy constants in the whole film.

3. Results and discussion:

The spin is assumed to be in the plane of y-z, and x component of the spin is assumed to be zero. The average value of out of plane spin component is given by

$$\bar{S}_z = \frac{\int_0^{\pi} e^{-\frac{E}{kT}}\cos\theta d\theta}{\int_0^{\pi} e^{-\frac{E}{kT}} d\theta} \qquad \textbf{(6)}$$

By substituting the energy given in equation (5) into the equation (6), $\bar{S}_z$ can be found as a function of the temperature. According to our model, $\bar{S}_z$ is really sensitive to H_{in}, H_{out}, J, ω and K_s, and it is not sensitive to $\sum_{m=1}^{N} D_m^{(2)}$ and $\sum_{m=1}^{N} D_m^{(4)}$. Because it is difficult to find the experimental values of J, ω, $\sum_{m=1}^{N} D_m^{(2)}$, $\sum_{m=1}^{N} D_m^{(4)}$, H_{in}, H_{out}, and K_s for this Barium ferrite film, a reasonable set of values have been plugged into equation. This simulation was performed for J = 10^{-56} Joules, ω=10^{-36} Joules, $\sum_{m=1}^{N} D_m^{(2)}$ =10^{-34} Joules, $\sum_{m=1}^{N} D_m^{(4)}$ =10^{-42} Joules, K_s= 10^{-30} Joules, H_{in}=10^{-40} Am^{-1} and H_{out}=10^{-34} Am^{-1}. $\bar{S}_z$ approaches zero at 1273 K as given in figure 1. Value of N used for all these simulations was about 1.2×10^5. This implies that barium hexaferrite film is not oriented below 1273 K, and it's easy axis is oriented in the plane of the film above 1273 K. So this model can explain the experimental data of barium ferrite synthesized on MgO substrate by some other researchers [6].

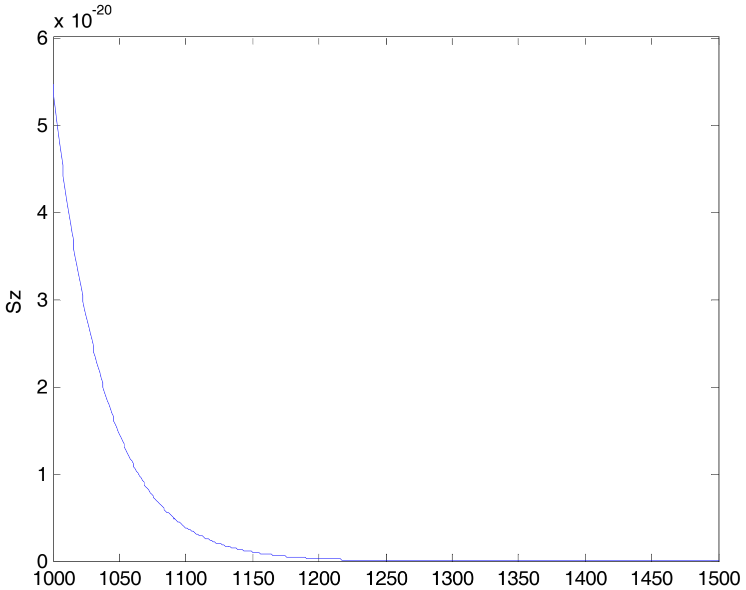

Figure 1: Graph of $\bar{S}_z$ versus temperature at K_s= 10^{-30} Joules.

Spin reorientation temperature can be varied by changing energy parameters as given in figure 2. When J = 10^{-56} Joules, $\omega=10^{-40}$ Joules, $\sum_{m=1}^{N} D_m^{(2)}=10^{-34}$ Joules, $\sum_{m=1}^{N} D_m^{(4)}=10^{-42}$ Joules, $K_s=10^{-29}$ Joules, $H_{in}=10^{-40}$ Am^{-1} and $H_{out}=10^{-34}$ Am^{-1}, $\overline{S}_z$ approaches zero at about 725 K. According to our model, spin reorientation temperature can be reduced by increasing K_s and by decreasing ω. K_s depends on the thermal expansion coefficients of film and substrate, and the Young's modules of film [12, 13].

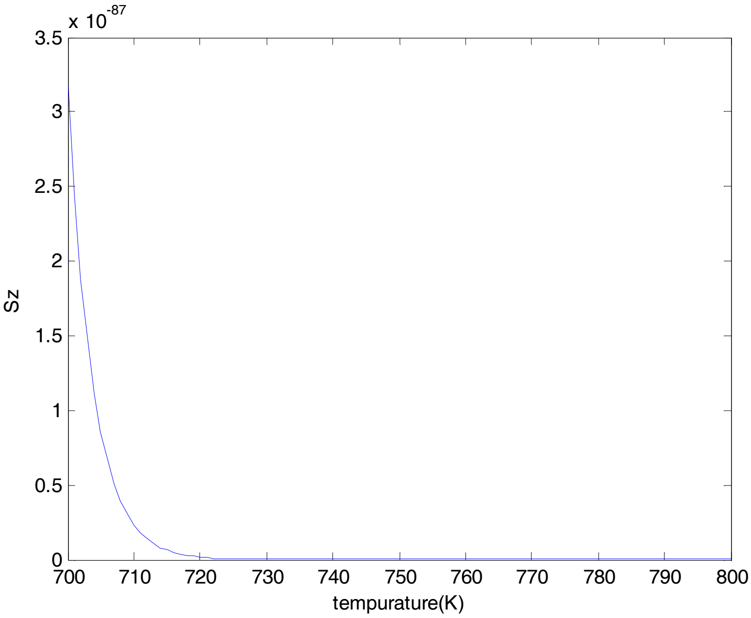

Figure 2: Graph of $\overline{S}_z$ versus temperature at $K_s=10^{-29}$ Joules.

The average value of in plane spin component is given by

$$\overline{S}_y=\frac{\int_0^{\pi} e^{-\frac{E}{kT}}\sin\theta d\theta}{\int_0^{\pi} e^{-\frac{E}{kT}} d\theta} \tag{7}$$

Similarly by putting the energy in above equation, $\overline{S}_y$ can be found as a function of temperature. $\overline{S}_y$ is also really sensitive to J, H_{in}, H_{out} and K_s. This simulation was performed for $J=10^{-31}$ Joules, $\omega=10^{-32}$ Joules, $\sum_{m=1}^{N} D_m^{(2)}=10^{-25}$ Joules, $\sum_{m=1}^{N} D_m^{(4)}=10^{-56}$ Joules, $K_s=10^{-41}$ Joules, $H_{in}=10^{-30}$ Am^{-1} and $H_{out}=10^{-33}Am^{-1}$. $\overline{S}_y$ becomes zero at 1273 K as given in figure 3. This means that barium ferrite film is not oriented below 1273 K, and magnetic easy axis is oriented perpendicular to the plane of the film above 1273 K. So this model can be used to understand the experimental data of barium ferrite film synthesized on Al_2O_3 substrate [6]. The spin reorientation temperature varies with energy parameters in this case too. When $J=10^{-33}$ Joules, $\omega=10^{-32}$ Joules, $\sum_{m=1}^{N} D_m^{(2)}=10^{-25}$ Joules, $\sum_{m=1}^{N} D_m^{(4)}=10^{-56}$ Joules, $K_s=10^{-29}$ Joules, $H_{in}=10^{-30}$ Am^{-1} and $H_{out}=10^{-33}Am^{-1}$, $\overline{S}_y$ approaches zero at 1023 K as given in figure 4. Decreasing J and increasing K_s reduce the spin reorientation temperature.

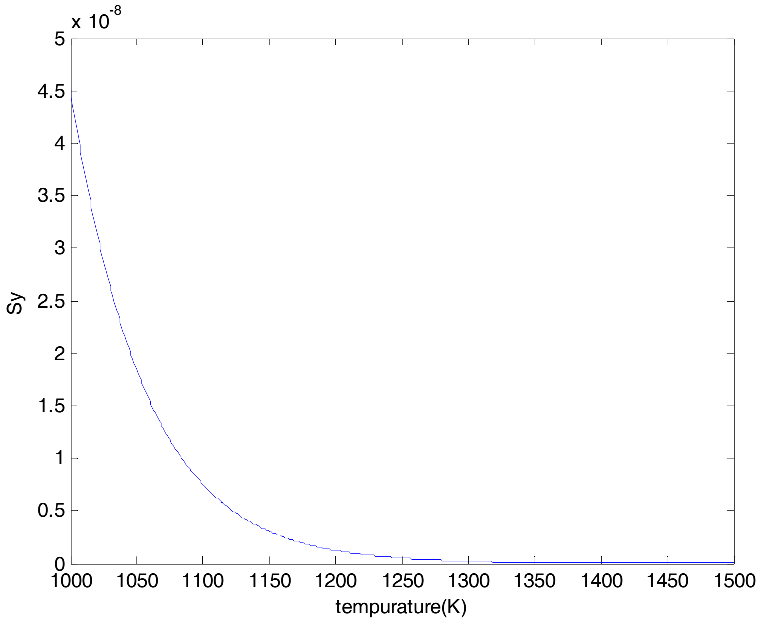

Figure 3: Graph of $\overline{S}_y$ versus temperature at J=10^{-31} Joules and K_s= 10^{-41} Joules.

According to our theoretical model, in plane and out of plane easy axis orientations occurs at 1273 K for K_s= 10^{-30} and 10^{-41} Joules, respectively. Barium ferrite films with in plane and out of plane easy axis orientation can be fabricated at 1273 K on MgO and Al_2O_3 substrates, respectively [6]. According to some of our previous experimental data, the stress induced anisotropy (K_s) depends on the type of substrate [12, 13].

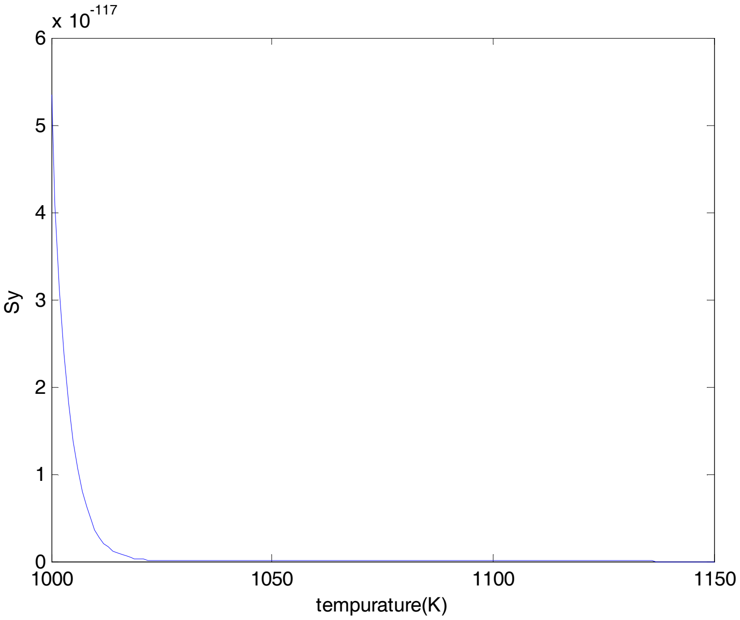

Figure 4: Graph of $\overline{S}_y$ versus temperature at J=10^{-33} Joules and K_s= 10^{-29} Joules.

Graph of spin reorientation temperature (T_s) versus K_s determined by investigating the variation of $\overline{S}_y$ is given in figure 5. Here K_s-axis is given on a logarithmic scale in order to observe the variation of T_s clearly. When K_s varies from 10^{-28} to 10^{-41} Joules, T_s changes from 0 to 1273 K. Similarly, the spin reorientation temperature changes with J, H_{in} and H_{out}. According to our previous theoretical studies, the spin reorientation temperature of ferromagnetic thin films of $CoPt_3$ and Fe-rich germanide is also really sensitive to H_{out} and K_s [7]. When K_s varies from 10^{-28} to 10^{-29}, T_s abruptly changes. However, in the range of K_s from 10^{-29} to 10^{-31}, T_s slightly changes.

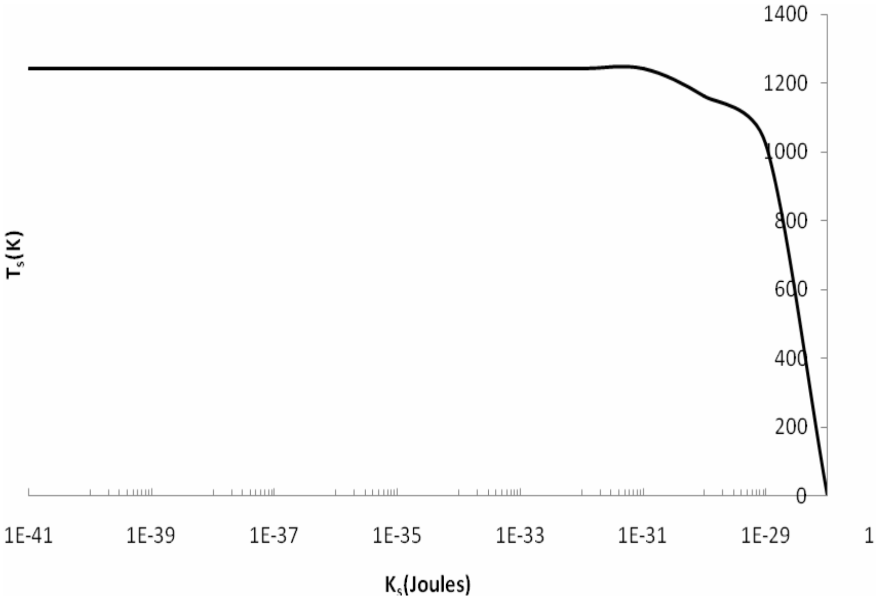

Figure 5: Graph of spin reorientation temperature versus K_s for $\overline{S}_y$.

4. Conclusion:

The in plane and out of plane easy axis orientation of Barium ferrite films deposited on MgO and Al_2O_3 substrates could be explained using our modified Heisenberg Hamiltonian model. Average values of in plane and out of plane spin components in 2-D model were plotted in order to determine spin reorientation temperature. Energy parameters in Heisenberg Hamiltonian were varied to investigate the change of spin reorientation temperature of Barium ferrite films. In plane orientation was observed at 1273 K for $K_s = 10^{-30}$ Joules. Out of plane orientation occurred at 1273 K for $J=10^{-31}$ Joules and $K_s = 10^{-41}$ Joules. According to our model, spin reorientation temperature solely depends on J, ω, H_{in}, H_{out} and K_s. The spin reorientation temperature significantly varies from about 725 to 1273 K, as above energy parameters were changed. Because K_s depends on the type of substrates, the in plane and out of plane orientations could be observed at 1273 K for different values of K_s.

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