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Fabrication and micromagnetic modeling of barium hexaferrite thin films by RF magnetron sputtering

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The synthesis and characterization of thin M-type barium hexaferrite ($BaFe_{12}O_{19}$ or BaM) films on silicon are reported. Multilayer *in situ* technique was employed to anneal the films at 850–900 °C for 10 min. The thickness dependence of the magnetic properties of the BaM films has been investigated using VSM. For the BaM 150 nm thickness film, acicular BaM grains were present having their c-axis randomly oriented. For the BaM films thicker than 150 nm, lattice relaxation favors the c-axis to be aligned in the film plane. The micromagnetic simulation was used to model the out-of-plane and the in-plane hysteresis loops. We have achieved good matching between the experimental data and the model. Using the micromagnetic model, we have estimated the deflection angle of c-axis from the normal plane $\theta = 25°$ for the 150 nm thick film.

Introduction

Because of their large uniaxial anisotropy, coupled with their good chemical stability and high mechanical hardness, perpendicular c-axis oriented M-type barium hexaferrite ($BaFe_{12}O_{19}$ or BaM) thin films are considered to be an attractive candidate for ultra-high-density magnetic recording [1,2]. To achieve higher signal-to-noise (S/N) ratio in perpendicular recording media, highly oriented perpendicular magnetic recording layers with relatively high perpendicular coercivity $H_{c\perp}$, small-grained films and large perpendicular squareness ($S_\perp = M / M_s$) are required [3].

The difficulty of growing BaM films raises from the deterioration of c-axis orientation as the film thickness increase [3], thickness dependence of crystallization is related to the substrate role. Silicon substrates are widely used for their low cost and high compatibility in semiconductor industrial integration [4,5]. The lattice mismatch between the BaM film and the substrate determines the state of the strain at the film-substrate interfacial region. Silicon has lattice parameter ($a = 5.43$ Å) while for BaM is ($a = 5.89$ Å) [6,7], therefore an epitaxial growth of BaM on a silicon substrate will induce an in plane compressive stress. Moreover, upon cooling the film down from the synthesizing temperature, a significant tensile stress is expected in the film due to the mismatch in the coefficients of thermal expansion between BaM film and the silicon substrate [4], also the diffusion and reaction between BaM and silicon substrate that occur at the synthesizing temperature, may have an effect on the magnetic properties of the film [8].

Many efforts have been made to address these issues, different under layers or buffer layers such as amorphous Ba-Fe-O layer [5], ZnO [9], Pt [10,11], and AlN [12], have been deposited to enhance the growth of the perpendicular c-axis orientation in the BaM films. BaM film's structure is usually amorphous in the as-grown state, external or *in situ* annealing is necessary to crystallize the films.

We have developed a multilayer technique, which utilizes an *in situ* annealing process, where the substrate is heated in Argon and Oxygen atmosphere, and the substrate's temperature was raised and cooled slowly to enervate the tensile stress in the film. We have studied the thickness dependence of the magnetic, the surface and the microstructural properties of the BaM films by using Vibrating Sample Magnetometer (VSM), SEM and X-ray diffraction. Since the magnetic properties of the BaM films are greatly influenced by the alignment of the c-axis relative to the normal of the film plane, micromagnetic modeling has been used to model both the hysteresis loops for perpendicular and in-plane directions and to estimate the angle of the c-axis relative to the normal of the film plane.

Experiment

BaM thin films were deposited on a clean 10 mm × 10 mm silicon (1 0 0) substrate using RF magnetron sputtering system. A turbo molecular pump was used to reach a base pressure of 3×10^{-6} torr

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or lower. The gas pressure during deposition was fixed to about 8.0 militorr using 20% pure Oxygen and 80% pure Argon. The target was 5.0 cm diameter disk and placed at 6.3 cm from the substrate. The RF power was fixed at 50 W, the deposition rate was estimated to be about 100 nm/h according to the VB-250 Vase Elliposemeter thickness measurement system. During the deposition, the substrate temperature was kept at 550 °C. Multilayer technique was used to reach the desired thickness, after each thin layer (about 150 nm), *in situ* annealing take place, where the films are heated inside the chamber without breaking the vacuum, typically after the deposition, 140 torr of Argon (80%) and Oxygen (20%) was introduced into the chamber, then increasing the substrate temperature up to 850–900 °C for 10 min. This procedure continued after each thin layer of BaM until the final thickness is achieved. We have fabricated 150 nm, 250 nm, 300 nm, 460 nm, 600 nm, 760 nm and 900 nm thickness films all by using the multilayer *in situ* technique.

Results and discussion

To investigate the microstructure of the BaM film, XRD was taken using the Siemens D5000 Diffractometer equipped with a solid-state detector using Cu Kα radiation. Fig. 1 shows the XRD measurements of the 150 nm, 300 nm, 450 nm thick *in situ* annealed films, and the standard XRD of the BaM. We see the out-of-plane peaks (0 0 *l*) are present in all films, but with different intensities, though it is still hard to distinguish (0 0 1 0) and (0 0 1 2) ones. Both (1 0 7) and (1 1 4) in-plane peaks are also present. As the film thickness increases, the intensities of the in-plane peaks becomes more intense than the out-of-plane ones, which indicates that thicker films favor their c-axis orientation aligned in the in-plane direction. The in-plane lattice parameter a and the out-of-plane lattice parameter c of the BaM film were calculated from formula 1 [13].

$$\frac{1}{d_{hkl}^2} = \frac{4}{3}\left(\frac{h^2 + hk + k^2}{a^2}\right) + \frac{l^2}{c^2} \quad (1)$$

where *d* is the interplanar distance and *h*, *k* and *l* are Miller indices.

The in-plane lattice parameter a = 5.85 Å of the 150 nm thick film is less than the bulk value (a = 5.89 Å) [6,7], which indicates a compressive basal strain ratio of (−0.68%). The out-of-plane lattice c = 23.07 Å is less than the bulk value (c = 23.20 Å) [6,7], which gives a strain ratio of (−0.56%). When the thickness of the film increases to 450 nm, the equivalent values of the lattice parameters a and c are: (a = 5.92 Å) and (c = 22.98 Å), which indicates a strain ratio of (+0.51%) and (−0.95%), respectively. This shows that as the film thickness increases, the strain caused by the substrate relaxes.

SEM has been used to explore the surface morphology of the 150 nm and 450 nm thick BaM films, which is shown in Fig. 2. The surface of the 150 nm thick film is totally covered with the grains, which is believed to have their c-axis in-plane oriented. In Fig. 2a, the average length of the acicular grains is between 200 and 300 nm. For the 450 nm thick film however, the surface show different surface morphology with more elongated acicular grains that start to grow on top of spherical like type of larger grains, the acicular grains cover larger area on the surface with average length between 400 and 500 nm. The XRD pattern of the 450 nm thick film show tendency of the grains to grow in the in-plane direction, the SEM data supports this, evidenced in the formation of the elongated acicular grains, which was not visible in the 150 nm thick film. We attribute this effect to the strain induced by the substrate. Our XRD result of the 150 nm thick film agrees with other research group's work [4,5].

To study the magnetic properties of the BaM thin film, the magnetization M was measured as a function of the externally static

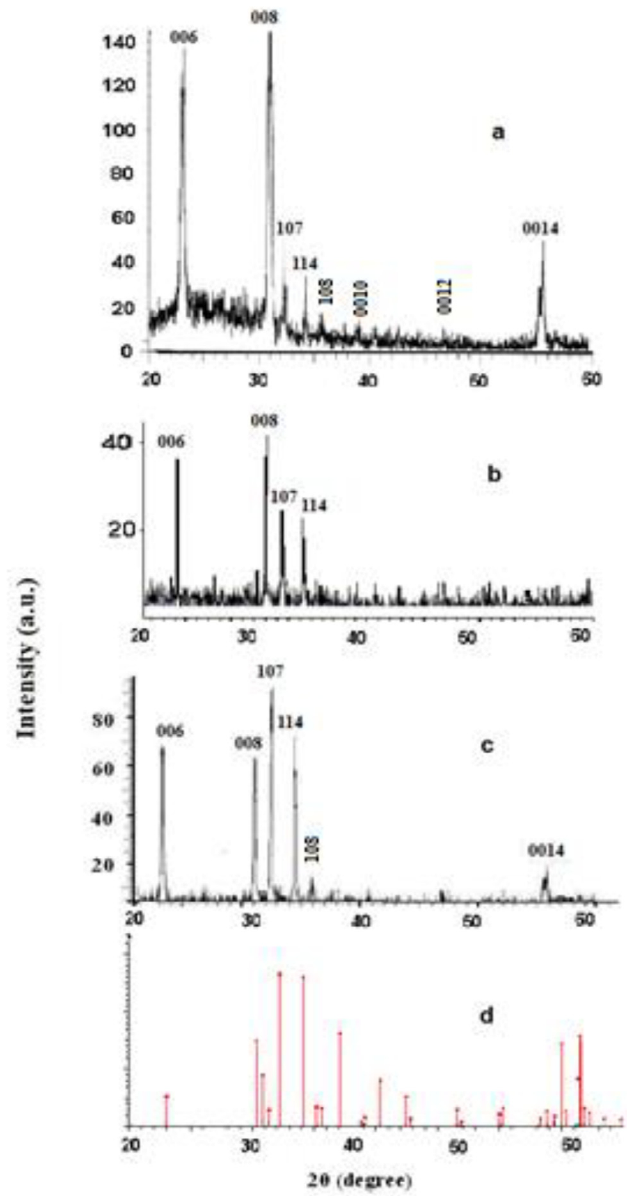

Fig. 1. (a–d) X-ray diffraction diagram of the 150 nm, 300 nm, 450 nm thick *in situ* annealed films, and the standard XRD of the BaM respectively.

applied magnetic field H with a vibrating sample magnetometer (VSM) at room temperature. In Fig. 3, we show the hysteresis loops for both out-of-plane and in-plane directions of the applied field of the 150 nm thin film. The magnetization M was calculated by dividing the magnetic moment μ by the volume of the 0.5 cm × 0.5 cm cut film. The average saturation magnetization M_s of the 150 nm thick film for the out-of-plane direction was about M_s = 230 emu/c.c., which is about 60% of the barium ferrite saturation magnetization bulk value [6,7]. This relatively low value is attributed to the formation of the acicular grains, that are occupying most of the film surface and tend to reduce the perpendicular saturation magnetization [3,5].

A commercial LLG Micromagnetic simulator has been used to model the hysteresis loop in both directions [14]. We have generated a 100 Voronoi cells (grains) media of a 10 nm length each, on a N_x = 64 x N_y = 64 x N_z = 1 slice with a total structure of 640 nm × 640 nm × 10 nm. The anisotropy constant is $K_u = 2 \times 10^6$ erg/c.c., and the exchange constant is A = 0.6 μerg/cm. The exchange

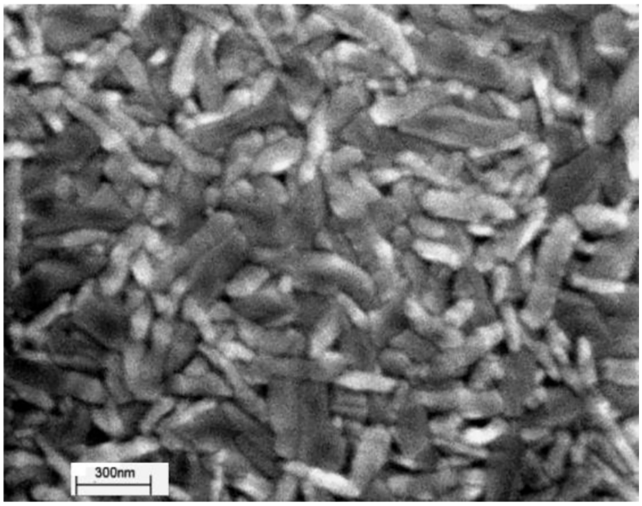

Fig. 2a. SEM observation of the *in situ* annealed BaM thin 150 nm film surface on Si (100).

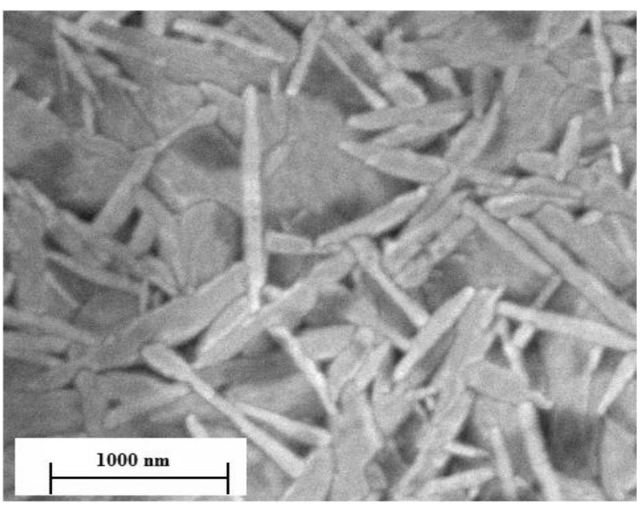

Fig. 2b. SEM observation of the in situ annealed 450 nm BaM thin film surface on Si (1 0 0).

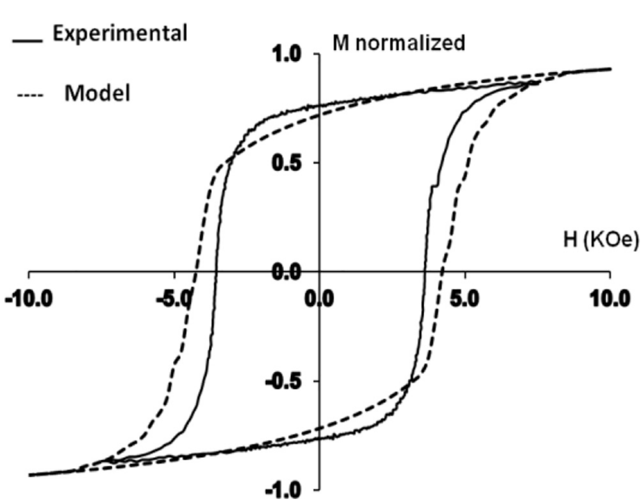

Fig. 3a. Experimental and modeled out-of-plane hysteresis loops for the 150 nm annealed BaM thin film.

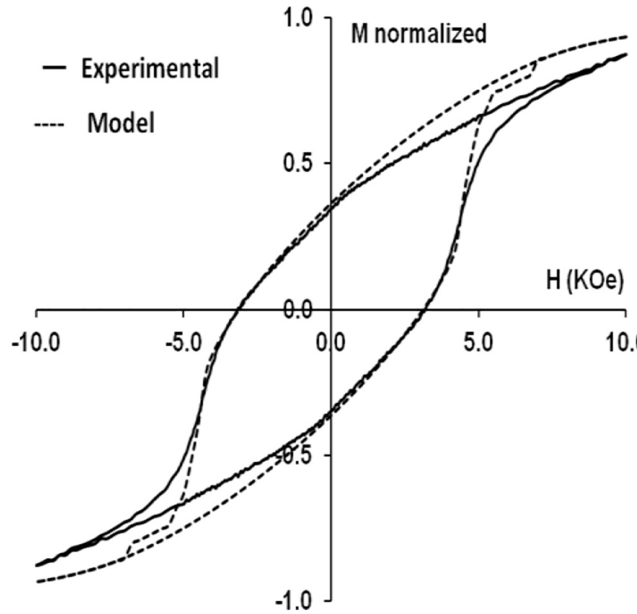

Fig. 3b. Experimental and modeled in-plane hysteresis loops for the 150 nm annealed BaM thin film.

coupling across the grain boundaries was set to zero. The damping coefficient $\alpha = 1.0$ and a time step of 0.7 ps were used.

Table 1 shows the magnetic parameters of the experimental and modeled films. The agreement between the model and the experimental data is good in the in-plane and out-of-plane loops of the 150 nm thick film as shown in Fig. 3a, 3b, and Table 1. The deflection angle of the c-axis from the out-of-plane direction is $\theta = 25°$ for the 150 nm thick film, and $\theta = 35°$ and $\theta = 40°$ for the 300 nm and 450 nm thick films respectively. This shows that the c-axis orientation is relatively far from the normal direction and deteriorates more as the thickness of the films increases. For a well-oriented c-axis film, the angle from the normal plane should not exceed few degrees. We found that both the in-plane coercivity and squareness are sensitive to the deflection angle, while the out-of-plane coercivity is sensitive to the anisotropy constant K_u. Mainly, the deflection angle depends on the difference between the out-of-plane and in-plane coercivities and squareness, as these difference increases, the deflection angle decreases, which gives rise to a more out-of-plane anisotropy.

We have studied the thickness dependence of the coercivity H_c and squareness S of the BaM films using the *in situ* multilayer technique. Fig. 4 shows the coercivity and squareness as a function of the film thickness for the (in-plane) and (out-of-plane) directions. It seems from the magnetic data, that c-axis is not well oriented to the out-of-plane direction, due to the high in-plane coercivity and squareness. When the thickness of the film is about 200 nm or less, the tendency of the c-axis orientation is to be aligned along the (out-of-plane) of the film. As the thickness of the film increases, c-axis orientation deteriorates evidenced in the increase of the in-plane squareness and the decrease of the out-of-plane squareness in agreement with the XRD data. We believe this is due to the substrate effect, as it is known, when the film thickness increases, the lattice strain becomes smaller due to lattice relaxation [15], when the thin film is stressed, it induces anisotropy that adds to the existing large magnetocrystalline uniaxial anisotropy [16], dominating the in-plane magnetocrystalline component. Annealing and layer stacking release the stress by relaxing the film lattice, this weakens the perpendicular magnetocrystalline anisotropy, and the in-plane anisotropy becomes dominant [17].

Table 1
The magnetic data for the experimental and modeled annealed BaM films.

Thickness (nm)	Experimental Data				Modeled Data					θ (°)
	$H_{c\ Per}$ (Oe)	S_{Per}	H_{cPar} (Oe)	S_{Par}	$H_{c\ Per}$ (Oe)	S_{Per}	H_{cPar} (Oe)	S_{Par}	K_u erg/c.c.	
150	3600	0.80	3175	0.40	4267	0.77	3133	0.39	2×10^6	25
300	3450	0.66	3380	0.50	3580	0.70	2900	0.44	1.7×10^6	35
450	3300	0.65	3100	0.56	3600	0.67	3000	0.46	1.7×10^6	40

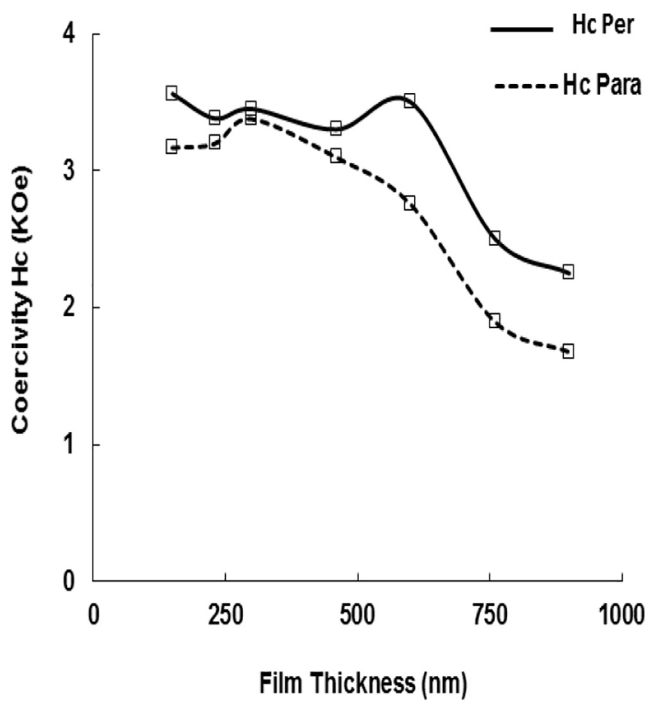

Fig. 4a. Coercivity H_c as a function of film thickness for the *in situ* annealed BaM thin films.

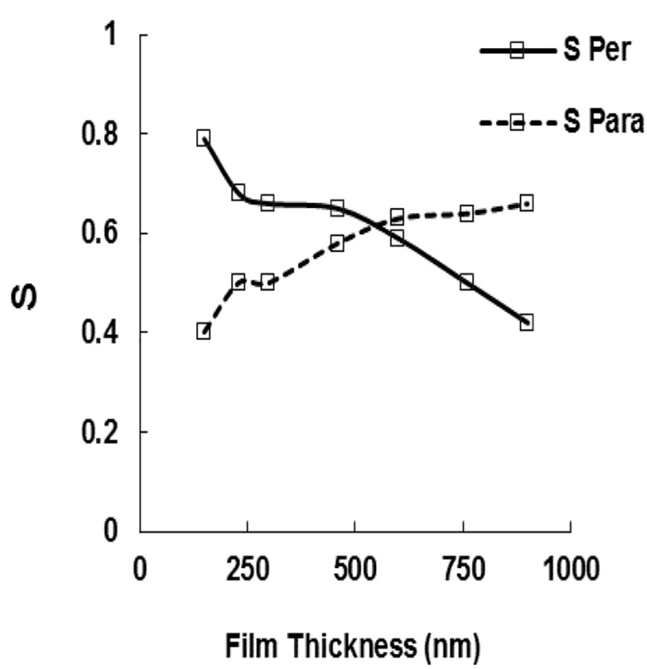

Fig. 4b. Squareness S as a function of film thickness for the *in situ* annealed BaM thin films.

Conclusion

We have developed a multilayer *in situ* technique to synthesize BaM thin films on silicon substrates using RF magnetron sputtering. The surface morphology of the 150 nm and 450 nm thick BaM films show acicular grains growth with their c-axis oriented in-plane direction. The thickness dependence of the magnetic properties of the BaM films thicker than 150 nm, can be related to the effect of the substrate. However, we expect that for BaM films thinner than 150 nm, an interfacial strain is present, and the stress anisotropy is dominant, which tends to align the easy c-axis orientation in the out-of-plane of the film. We have developed a micromagnetic model that agrees well with the experimental data, from the model we have estimated the deflection angle of the c-axis from the out-of-plane direction, and found that the angle depends on the difference between the out-of-plane and in-plane coercivities and squareness. We believe that the micromagnetic model complements the experimental data, and allows for determination of the magnetic parameters of the BaM film such as the coercivity, squareness, and anisotropy constant K_u. Furthermore, to be compatible with high-density recording media applications, we suggest that using a buffer layer would promote the growth of small-sized grains that have their easy magnetic c-axis aligned in the out-of-plane direction.

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References

[1] Matsuoka M, Naoe M. Ba-ferrite thin-film disk for perpendicular magnetic recording. J Appl Phys 1985;57:4040–2.
[2] Sui X, Scherge M, Kryder MH, Snyder JE, Harris VG, Koon NC. Barium ferrite thin-film recording media. J Magn Magn Mater 1996;155:132–9.
[3] Cho TS, Doh SJ, Je JH. Thickness dependence of the crystallization of Ba-ferrite films. J Appl Phys 1999;86:1958–64.
[4] Papakonstantinou P, O'Neill M, Atkinson R, Salter IW, Gerber R. Oriented barium, strontium ferrite films pulsed laser deposited on YSZ (1 0 0) and Si (1 0 0) substrates. J Magn Soc Jpn 1996;20:333–6.
[5] Morisako Akimitsu, Matsumoto Mitsunori, Naoe Masahiko. Properties of c-axis oriented Ba-ferrite sputtered films. J Magn Magn Mater 1999;193:110–3.
[6] von Aulock WH. Handbooks of Microwave Ferrite Materials. New York: Academic Press; 1965.
[7] Kojima H. Ferromagnetic Materials, vol. 3. New York: E.P. Wohlfarth North Holland; 1982.
[8] Xu Z, Lan Z, Sun K, Yu Z, Guo R, Bai F. Effects of BaM interfacial layer on the c-axis orientation of BaM thin films deposited on SiO_2/Si substrates. IEEE Trans Magn 2013;49(7):4226–9.
[9] Hoshi Y, Kubota Y, Naoe M. Control of orientation and crystallite size of barium ferrite thin films in sputter deposition. IEEE Trans Magn 1995;31:2782–4.
[10] Zhuang Z, Rao M, Laughlin DE, Kryder MH. The effect of Pt interlayers on the magnetic and structural properties of perpendicularly oriented barium ferrite media. J Appl Phys 1999;85:6142–4.
[11] Zhuang Z, Rao M, White RM, Laughlin DE, Kryder MH. Barium ferrite thin film media with perpendicular c-axis orientation and small grain size. J Appl Phys 2000;87:6370–2.
[12] Morisakoa A, Nakaa T, Itoa K, Takizawaa A, Matsumotoa M, Hongb YK. Properties of Ba-ferrite/AlN double layered films for perpendicular magnetic recording media. J Magn Magn Mater 2002;242–245:304–10.

[13] Warren BE. X-ray Diffraction, Reading, Massachusetts. Addison-Wesley; 1969.
[14] LLG simulation package information is available at http://llgmicro-home.mindspring.com.
[15] Matthews JW, Blakeslee AE. Defects in epitaxial multilayers. J Cryst Growth 1974;27:118–25.
[16] Cullity BD, Graham CD. Introduction to Magnetic Materials. second ed. New Jersey: Wiley; 2009. p. 264.
[17] Lisfi A, Nguyen LT, Lodder JC, Williams CM, Corcoran H, Chang P, et al. Analysis and contribution of stress anisotropy in epitaxial hard ferrite thin films. J Magn Magn Mater 2005;290:219–22.