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Improvement of perpendicular coercivity of Hexagonal M-type ferrite thin films on Pt underlayer prepared by flash-annealing

Keisuke Matsuno, Keisuke Mizuno and Shigeki Nakagawa
Department of Physical Electronics, Tokyo Institute of Technology, 2-12-1 Ookayama,
Meguro-ku, Tokyo 152-8552, Japan
Fax: 81-3-5734-2513, e-mail: k_mat@spin.pe.titech.ac.jp

Thin films of various kinds of hexagonal Magneto-plumbite type (M-type) ferrites as magnetic layer with Pt underlayer were prepared using flash annealing process. M-type strontium ferrite with substitution of Sr^{2+} and Fe^{3+} by La^{3+} and Co^{2+} thin films (LaCo-SrM) and M-type barium ferrite thin films (BaM) on Pt underlayer were prepared by the Facing Targets Sputtering system. Since the grain size became huge enough because the high substrate temperature, e.g. around 500 °C, is maintained particles by excess heating during the deposition. Flash-annealing technique, that is to anneal the specimen at high temperature above 900 °C for a few moments after the deposition of the films at low temperature of 250 °C enough for preventing crystallization. Annealing at 900 °C for 3 minutes was an appropriate processing condition as a flash annealing process to attain high perpendicular coercivity H_c and perpendicular squareness ratio in case of BaM with maintaining the grain size as low as several tens nanometers. 30 nm-thick LaCo-SrM films prepared by flash-annealing at 900 °C and 5 minutes exhibited very smooth surface and small particle size of about 22 nm. The film revealed large H_c and squareness of 4.5 kOe and 0.75, respectively. Flash-annealing is very useful when preparing the superior magnetic recording media.

Key words: Hexagonal M-type ferrite, Flash-annealing, Pt underlayer

1. INTRODUCTION

The steep increase of recording density of hard-disc drive (HDD) in recent years expands the usage of HDD not only to personal computers but also to consumer audio visual electronic facilities, such as video recorders, personal digital assistant and so on. However, as the recording density increases, the size of magnetic grains in the recording media, that is considered to restrict a resolution of recorded bits, becomes so small. Then, the thermal fluctuation effect cannot be disregarded. In order to overcome this problem, it is necessary to choose materials which have high magnetic anisotropy for the recording layer and to maintain superior magnetic characteristics when the thickness of the films is dozens of nm.

In this study, hexagonal magneto-plumbite (M-type) barium ferrite thin films (BaM) and hexagonal M-type strontium ferrite with substitution of Sr^{2+} and Fe^{3+} by La^{3+} and Co^{2+} thin films (LaCo-SrM) were prepared as high density recording layer for perpendicular magnetic recording media. Pt underlayer is also prepared to promote c-axis orientation of hexagonal M-type ferrite thin films[1,2]. It was reported that BaM thin films with excellent c-axis orientation were able to deposit on the substrate at relatively high substrate temperature, e.g. around 500 °C, in as-deposited state by Facing Targets Sputtering technique in our previous works[1,2]. However, the crystallite particles grew so large that the film could not apply to the HDD media. In this study, the substrate temperature during the sputter-deposition was set at as low as 250 °C, then the thin films were crystallized by short time annealing at high temperature, so called "flash annealing" in order to attain fine particle size and improvement of magnetic properties of hexagonal ferrite thin films.

2. EXPERIMENTAL PROCEDURE

Multilayers composed of 3 periods of BaM(10nm)/Pt(14nm) bilayers ($[BaM/Pt]_3$) were prepared by Facing Targets Sputtering system [3,4,5]. DC power supply was used for the sputtering using BaM ferrite targets which were slightly deoxidized surface to attain DC conductivity. $BaFe_{11}O_x$ sintered disks were used as BaM targets. BaM and Pt layers were deposited on the SiO_2/Si substrates at Ar gas pressure P_{Ar} of 2 mTorr. Oxygen gas of 0.1 mTorr was introduced during the ferrite deposition. Substrate temperature during the depositions was set at 250 °C. $[BaM/Pt]_3$ thin films prepared by the above method were annealed in air circumstance in a furnace. Optimum annealing condition was investigated by changing annealing temperature and time. LaCo-SrM/Pt bilayers were prepared by the method as mentioned above. $La_{0.3}Co_{0.3}Sr_{0.7}Fe_{10.7}O_x$ sintered ferrite disks were used as the LaCo-SrM targets.

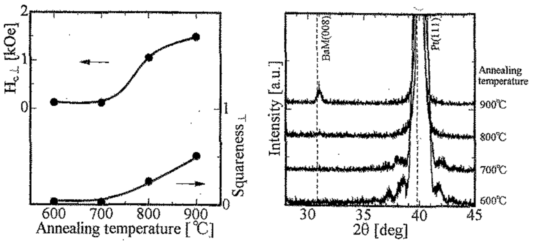
The following measurements were performed to evaluate the properties of films. The size and orientation of crystallites were evaluated by X-Ray Diffractometry (XRD). The magnetic characteristics, such as coercivity and squareness were estimated by Vibrating Sample Magnetometer (VSM). The particle size and surface roughness of films were evaluated by Atomic Force Microscope (AFM).

3. RESULTS AND DISCUSSION

3.1 [BaM/Pt]$_3$ multilayer films

Figure 1 shows (a) the change of perpendicular coercivity $Hc_{\perp}$ and perpendicular squareness $S_{\perp}$ of [BaM/Pt]$_3$ thin films as a function of annealing temperature and (b) XRD diagrams of [BaM/Pt]$_3$ thin films annealed at different annealing temperatures. The specimen annealed at 600 °C and 700 °C cannot be crystallized and their $Hc_{\perp}$ and $S_{\perp}$ were so small. The specimen annealed at 800 °C exhibited crystallization, but $Hc_{\perp}$ and $S_{\perp}$ were 1.0 kOe and 0.25, respectively. The specimen annealed at 900 °C exhibited better crystallization and c-axis orientation. Coercivity and squareness of the film annealed at 900 °C were 1.5 kOe and 0.5, respectively. It was confirmed that the best annealing temperature for [BaM/Pt]$_{*3}$ thin films was about 900 °C.

Figure 2 shows the change of $Hc_{\perp}$ and $S_{\perp}$ of [BaM/Pt]$_3$ thin films annealed at 900 °C as a function of annealing time. The specimens annealed at 1 or less minutes were not crystallized. The specimens annealed at 900 °C for 3 minutes or more were crystallized and $Hc_{\perp}$ and $S_{\perp}$ of them were abruptly increased. Then $Hc_{\perp}$ and $S_{\perp}$ decreased gradually as annealing time became long. Figure 3 shows AFM images of the specimens annealed at (a) 3 minutes and (b) 60 minutes. The particle size of the specimen annealed at 3 minutes was about 28 nm, though the particle size of the specimen annealed for 60 minutes was 250 nm.



(a) (b)

Fig.1. (a) Changes of $Hc_{\perp}$ and $S_{\perp}$ of [BaM/Pt]$_3$ multilayers as a function of annealing temperature and (b) XRD diagrams of the multilayers annealed at different annealing temperatures.

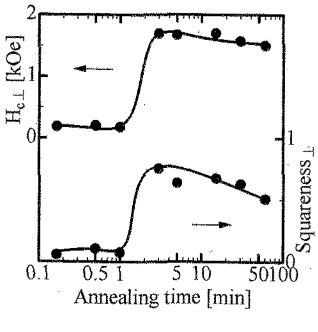


Fig.2. Changes of $Hc_{\perp}$ and $S_{\perp}$ of [BaM/Pt]$_3$ multilayers as a function of annealing temperature as a function of annealing time. Annealing temperature was 900 °C.

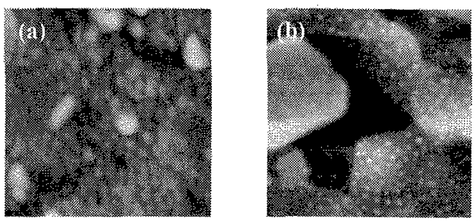


Fig.3. AFM images (500 nm x 500 nm) of [BaM/Pt]$_3$ thin films annealed at (a) 3 minutes and (b)60 minutes at 900 °C.

It was considered that the optimum annealing condition for maintaining the particle size as small as several tens nanometer was annealing at 900 °C for 5 minutes. Such short time annealing at high temperature is called as "flash-annealing".

3.2 LaCo-SrM/Pt bilayer thin films

LaCo-SrM films were deposited on Pt underlayers to improve perpendicular magnetic anisotropy in thin layer region of recording layer. LaCo-SrM magnetic layer thickness of the specimens was set at 30 nm, and Pt underlayer thickness was changed from 0 nm (i.e. LaCo-SrM single layer film) to 70 nm. Figure 4 (a) shows the changes of grain size <*D*> and c-axis dispersion angle $\Delta\theta_{50}$ of LaCo-SrM/Pt films estimated from XRD as a function of Pt underlayer thickness. Grain size of LaCo-SrM layer reduced as Pt underlayer became thin, but c-axis dispersion angle $\Delta\theta_{50}$ became large which means worse c-axis orientation. Figure 4 (b) shows the changes of $Hc_{\perp}$ and $S_{\perp}$ of LaCo-SrM/Pt films as a function of Pt underlayer thickness.

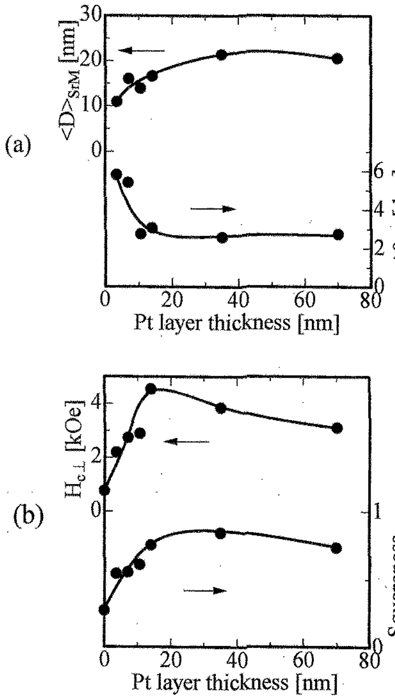


Fig.4. Dependence of (a) $\Delta\theta_{50}$ and grain size <*D*> of LaCo-SrM and (b) $Hc_{\perp}$ and $S_{\perp}$ of LaCo-SrM/Pt thin films on Pt underlayer thickness.

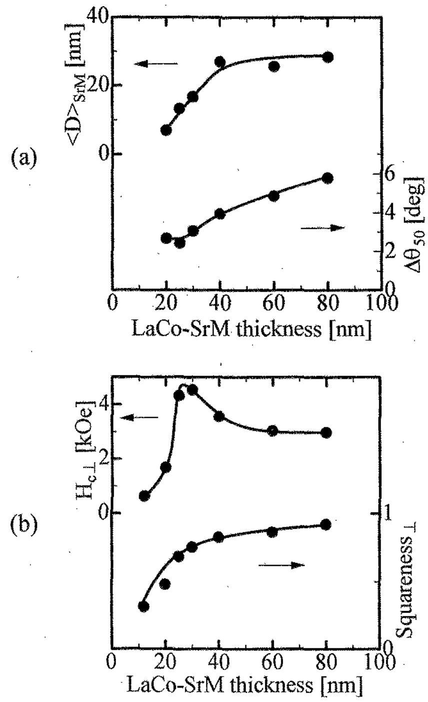


Fig.5. Dependences of (a) $\Delta\theta_{50}$ and grain size of LaCo-SrM and (b) coercivity and squareness of LaCo-SrM/Pt thin films on LaCo-SrM layer thickness.

$Hc_\perp$ and $S_\perp$ of the films increased rapidly as the thickness of Pt underlayer increased from 0 nm to 15 nm. Then, $Hc_\perp$ and $S_\perp$ became the maximum at Pt underlayer of about 15 nm. $Hc_\perp$ and $S_\perp$ decreased gradually for above Pt layer thickness. Therefore, it was considered that optimum Pt underlayer thickness of LaCo-SrM/Pt bilayer thin films was around 14 nm.

Figure 5(a) shows changes of grain size <*D*> and c-axis dispersion angle $\Delta\theta_{50}$ of LaCo-SrM/Pt(14nm) bilayer films estimated from XRD as a function of LaCo-SrM layer thickness. Thickness of LaCo-SrM ferrite layer was changed from 12 nm to 80 nm. The specimen with 12 nm-thick LaCo-SrM layer didn't crystallized. The grain size reduced and also $\Delta\theta_{50}$ was improved as the LaCo-SrM layer became thin. Figure 5(b) shows the changes of $Hc_\perp$ and $S_\perp$ of LaCo-SrM/Pt(14nm) bilayer films estimated from XRD as a function of LaCo-SrM layer thickness.

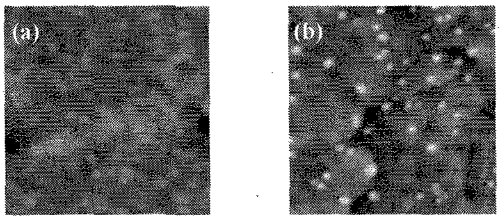


Fig.6. AFM images whose one side is 500 nm of LaCo-SrM/Pt thin films (a) with 25 nm magnetic layer and (b) with 80 nm magnetic layer.

$Hc_\perp$ took the maximum value of about 4.5 kOe at around 25 nm. Squareness of the film increased gradually as the layer thickness increased, then squareness achieved high value of 0.92 at magnetic layer thickness was 80 nm.

AFM images of the films with LaCo-SrM thickness of 25 nm and 80 nm were shown in Fig.6. The particles were isolated in the sample with 25 nm LaCo-SrM layer, but had enlarged and become hexagonal-block like particle in the sample with 80nm LaCo-SrM layer. Surface roughness of the sample with 80 nm LaCo-SrM layer was rougher than that of the sample with 25 nm magnetic layer. Therefore, it was considered that the optimum thickness of LaCo-SrM layer was about 30nm.

3.3 Thickness dependence of various kind of hexagonal M-type ferrite thin films

Four kinds of the following samples were prepared to compare the magnetic properties at thin film region.

1. LaCo-SrM/Pt prepared by flash-annealing after sputtering in Ar and Kr mixture gas
2. SrM/Pt prepared by substrate heating in Ar and Kr mixture gas
3. SrM/Pt prepared by substrate heating in Ar gas
4. BaM/Pt prepared by substrate heating in Ar gas

Ar and Kr mixture gas was used as sputtering gas for above (1) and (2) cases, since the atomic weight of Kr is almost equal to that of the atomic weight of Sr and momentum transfer might be effective in a sputtering phenomenon [6].

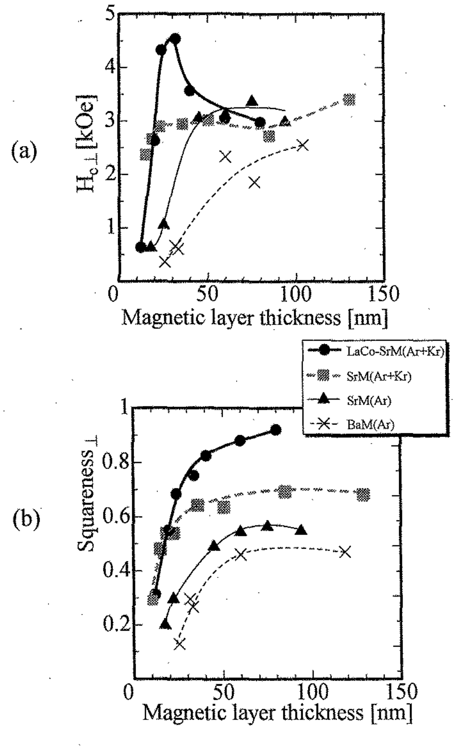


Fig.7. Dependence of (a) coercivity and (b) squareness of bilayer on Magnetic layer thickness.

Figure 7 shows the changes of (a) $H_{c\perp}$ and (b) $S_{\perp}$ of the various kinds of hexagonal ferrite thin films prepared by the methods mentioned above as a function of magnetic layer thickness. Thickness of Pt underlayer was set at 14 nm in all cases. $H_{c\perp}$ of the films crystallized by substrate heating methods (2, 3 and 4) increased as the magnetic layer thickness increased. Usage of Ar and Kr mixture gas (1 and 2) seems to increase $H_{c\perp}$ in thin film region below 50 nm. On the other hand, $H_{c\perp}$ of LaCo-SrM prepared by flash-annealing was abruptly increased in ultra thin film region. Although squareness $S_{\perp}$ of all samples increased as magnetic layer thickness increased, the maximum values are different for each film. Usage of Ar and Kr mixture gas increased squareness, and using LaCo-SrM as magnetic layer also increased squareness. Therefore, LaCo-SrM prepared by flash-annealing method is very effective for perpendicular magnetic recording media.

4. SUMMARY

Thin films of various kinds of hexagonal M-type ferrites as magnetic layer with Pt underlayer were prepared using flash annealing process. It was clarified that flash-annealing at 900 °C for about 3~5 min was effective to attain higher $H_{c\perp}$ and $S_{\perp}$ at very thin film region below 50 nm and to reduce the grain size in the films. LaCo-SrM is suitable to attain high $H_{c\perp}$ and $S_{\perp}$ which are proper for the properties of perpendicular magnetic recording media. LaCo-SrM(30nm)/Pt(14nm) bilayered thin film exhibited the largest coercivity of 4.5 kOe and small grain size of 28 nm. LaCo-SrM ferrite thin films prepared by flash-annealing method seems to be suitable for perpendicular magnetic recording media.

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