C-AXIS PERPENDICULARLY ORIENTED BARIUM FERRITE THIN FILM MEDIA ON SILICON SUBSTRATE

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ABSTRACT

Barium ferrite thin films with excellent perpendicular c-axis orientation were successfully fabricated on Si substrate without any buffer layer. To compensate for possible barium deficiency due to the inter-diffusion between films and substrate, a barium-rich target was used. For a 900 Å-thick film, the perpendicular remanent squareness is about 0.9, while the in-plane remanent squareness is about 0.3. The saturation magnetization (Ms) is about 220 emu/cc, while the coercivity is around 3500 Oe. X-ray diffraction (XRD) results show the (001) perpendicular c-axis texture in the films. It was also found that the rapid thermal annealing conditions greatly affect the magnetic properties of barium ferrite films. With a certain flow rate of oxygen gas in the rapid thermal annealer (RTA), barium ferrite films generally crystallize with good perpendicular c-axis texture. Without oxygen gas, the hexagonal barium ferrite phase fails to develop; instead spinel Fe_3O_4 phase forms. The reason for the collapsing of hexagonal barium ferrite texture is thought to be an oxygen deficiency in the barium ferrite films due to the reduction of oxygen in the films during the high temperature annealing.

INTRODUCTION

Hexagonal barium ferrite thin films are promising candidates as future high-density magnetic recording media due to their excellent magnetic properties and mechanical and chemical stability. The as-deposited barium ferrite thin films without substrate heating are amorphous in nature. Crystalline barium ferrite thin films have to be obtained by either applying *in situ* substrate heating during deposition or through a post-deposition annealing. As the magnetic easy axis in barium ferrite thin films is parallel to the c-axis, much effort has been made to develop suitable substrates and underlayers for obtaining thin films with a high degree in c-axis perpendicular orientation. Many different kinds of underlayers and substrates have been used including ZnO, Pt, AlN, Si_3N_4, Si and thermally oxidized silicon(SiO_2)[1-5]. For the *in situ* heating during deposition case, barium ferrite thin films with excellent perpendicular c-axis orientation were achieved on the underlayer ZnO, thermally oxidized silicon either by target-facing type of sputtering or by conventional DC magnetron type of sputtering. For the post-deposition annealing case, barium ferrite thin films with perpendicular c-axis orientation were successfully grown on the Pt and AlN underlayers.

In this paper, we study the magnetic and structural properties of post-deposition annealed barium ferrite thin films deposited on Si substrate without any buffer layer. The lack of buffer layer between the films and substrate may cause the deficiency of barium content due to the inter-diffusion during high temperature annealing. Thus, a barium-rich barium ferrite target was used to compensate for barium deficiency in the thin films. The effect of rapid thermal annealing conditions on the magnetic and structural properties of barium ferrite thin films will be also explored.

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EXPERIMENTAL

Thin films were deposited by RF diode sputtering in a Leybold-Haereus Z-400 sputtering system. The sputtering power density and Ar gas pressure were set to 2.25W/cm^2 and 5.7mTorr, respectively. The base vacuum pressure was below 1×10^{-7} Torr. Silicon (100) substrates were used for all samples. The as-deposited barium ferrite thin films are amorphous and nonmagnetic. After deposition, all the samples were annealed at 800 °C in either the RTA oven for 60s or annealed at 800 °C in a thermal oven for 20 min. Argon gas was used to purge the RTA oven before annealing, and the oxygen gas was added into the oven during the RTA oven annealing. The flow rate of oxygen gas was varied as specified later.

The magnetic properties of the films were studied using either an alternating gradient magnetometer (AGM) or a vibrating sample magnetometer (VSM). The crystalline structures of the films were characterized by a Cu K_α X-ray diffractometer. An energy dispersive X-ray fluorescence system (EDXRF) was used to determine the chemical composition of the barium ferrite targets as well as of the deposited films

RESULTS AND DISCUSSION

Magnetic and structural properties of barium ferrite thin films

The magnetic properties of stoichiometric and barium rich barium ferrite thin films deposited on Si substrate are listed in Table I. All of the samples are post-deposition annealed at 800 °C in a thermal oven for 20 min. The 900 Å–thick stoichiometric barium ferrite thin film (sample A) shows a perpendicular c-axis orientation, as indicated by a perpendicular remanent squareness of 0.75 and in-plane remanent squareness of 0.44. The perpendicular Hc is around 1460 Oe, while the in-plane coercivity is around 1900 Oe. The MH loop of sample A is shown in Fig.1 Although sample A shows a perpendicular c-axis orientation, it is not suitable for use as a perpendicular recording medium, due to its smaller Hc, and lower perpendicular remanent squareness. The barium deficiency in the film is thought to be reason for the poor magnetic properties.

TABLE I FILM STRUCTURA AND MAGNETIC PROPERTIES OF BARIUM FERRITE THIN FILMS

Sample	Film Structure	Hc(⊥) (Oe)	Hc(//) (Oe)	Perpendicular Squareness	In-plane Squareness	δM	Ms (emu/cc)
A	Stoichiometric barium ferrite 900 Å/Si	1460	1400	0.78	0.32	-0.35	170
B	Stoichiometric barium ferrite 1500 Å/Si	1900	200	0.74	0.63	-0.40	
C	Ba-rich barium ferrite 900 Å/Si	3300	2600	0.88	0.28	-0.65	220
D	Ba-rich barium ferrite 1500Å/Si	4970	4940	0.70	0.66	-0.35	

To improve the magnetic properties, 10wt% barium rich barium ferrite thin films were deposited on Si substrate to compensate for the barium deficiency due to the inter-diffusion. The magnetic properties of a 900 Å–thick barium rich thin film (sample C) are shown in Table I, and the MH loop of sample C is shown in Fig. 2. Significant changes in magnetic properties were observed. Compared to the properties of sample A, sample C has a higher perpendicular remanent squareness of 0.88 and smaller in-plane remanent squareness of 0.28. The perpendicular Hc of sample C is around 3300 Oe, which is much larger than that of sample A. The increase of coercivity may be due to a decrease of grain size for barium rich thin films[6]. The much lower Hc observed in the stoichiometric barium ferrite films may be caused by the incoherent rotation of magnetization, which is more pronounced in larger grains. The Ms for barium-rich thin film samples C and D is around 220 emu/cc, while the Ms is only around 170 emu/cc for stoichiometric thin film samples A and B. It means that enriching barium content is effective to compensate for the barium loss, which tend to decrease of the saturation magnetization of barium ferrite thin films

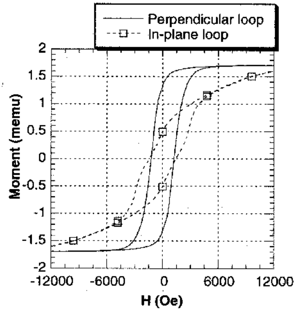

Fig. 1 In-plane and perpendicular MH loops for sample A

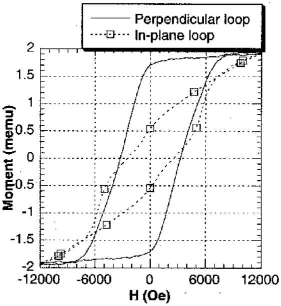

Fig. 2 In-plane and perpendicular MH loops for sample C

The magnetic properties of sample B and D, which have thickness of 1500 Å, are shown in Table I. There is no preferred c-axis orientation for sample D, while sample B shows a very weak perpendicular orientation. The similar values in the perpendicular and in-plane remanent squareness of sample D indicate random c-axis orientation of the film. The deterioration of perpendicular c-axis orientation with the increase in thickness is due to the increase of bulk nucleation sites, which favors to crystallize into random oriented barium ferrite grains[7]. δM curves were also measured for all samples and are also shown in Table I. There are only negative peaks for all barium ferrite films, which indicates magnetostatic interactions among grains. δM value is roughly the same for sample A and B. A significant difference in the δM values was observed for sample C and D. The smaller δM value for sample D with respect to sample C suggests a weaker magnetostatic interaction among barium ferrite grains for sample D. The coercivity Hc of sample D is much bigger than that of sample C. The increase in Hc is consistent with a reduction in magnetostatic interaction among barium ferrite grains, as shown by a decrease in absolute value of δM from a value of 0.65 to 0.35. Due to the directional nature of magnetic interaction, the magnetostatic interactions in the easy-axis randomly oriented films are weaker than in the easy-axis well-aligned films.

XRD was also used to characterize the structural properties for all the samples. XRD curves for sample A and sample C are shown in Fig 3. The dominant reflection peaks are (006) (008) and (0014), which indicate excellent c-axis orientation. The XRD results are consistent with the magnetic measurement results shown above. With the same thickness of 900 Å, the (001) peaks of sample C are bigger than those of sample A, which indicates a better c-axis perpendicular orientation for barium rich film sample C. Sample B and D have weak (006) (008) and (0014) peaks, as shown in Fig. 4. In addition to the (001) peaks, there are also weak peaks of (107) and (114) peaks. The existence of all (001), (107) and (114) diffraction peaks indicates a random orientation of the c-axis for sample D. Sample B has relatively bigger (001) peaks over (107) and (104) peaks, as indicated in Fig.4. So sample B has a weak perpendicular c-axis orientation, which is consistent with magnetic properties shown in Table I.

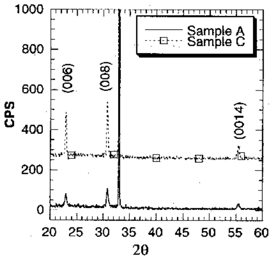

Fig. 3 The XRD curves for sample A and C

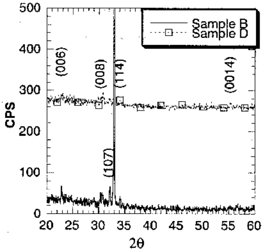

Fig. 4 The XRD curves for sample B and D

RAPID THERMAL ANNEALING EFFECT

Several samples with the same film structural as sample C were used to study the rapid thermal annealing effect on the c-axis texture and the magnetic properties of barium ferrite films. Sample E and F were annealed with an oxygen gas flow rate of 2 standard liter per minute (SLPM) and 5 SLPM respectively in a RTA at 800 °C for 60 s, while sample G was annealed in RTA without supplying oxygen gas at 800 °C for about 20 min.

Both samples E and F have a perpendicular c-axis orientation. The MH loops for sample F is shown in Fig.5. With the oxygen gas in the annealing process, barium ferrite films show perpendicular c-axis texture after annealing. The XRD curve for the sample E and F are shown in Fig. 7. The perpendicular c-axis texture of sample F is better than that of sample E, indicated by the larger (001) peak intensity for sample F, as shown in Fig. 7. So a larger oxygen gas flow rate during annealing is beneficial to get better c-axis texture for barium ferrite thin films.

When annealing barium ferrite thin films in RTA without supplying oxygen gas, both film magnetic properties and film texture display a dramatic change. Sample G has an in-plane easy axis orientation, with a much higher Ms of 320 emu/cc, and a much lower coercivity of 600 Oe, as shown in Fig. 6. The XRD curve for sample G, as shown in Fig. 8, indicates spinel Fe_3O_4 (111), (222), and (511) reflections, and no hexagonal barium ferrite (006) (008) and (0014) reflections are observed. So the hexagonal barium ferrite crystallographic texture has collapsed with the formation of the spinel Fe_3O_4 (111) texture during annealing without oxygen gas. The reason for failing to form hexagonal barium ferrite texture is thought to be due to an oxygen deficiency in barium ferrite thin films. Without an oxygen atmosphere, oxygen atoms in the films will more likely diffuse to the Si substrate and react with Si during the high temperature annealing process, which makes the films oxygen deficient. Fe^{2+} ions may be produced by the reduction from Fe^{3+} ions due to the deficiency of oxygen in the film. So the spinel Fe_3O_4 phase is formed with the reduction of oxygen in the films during annealing.

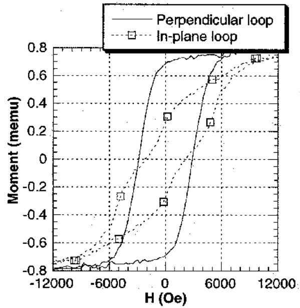

Fig.5 In-plane and perpendicular MH loops for sample E

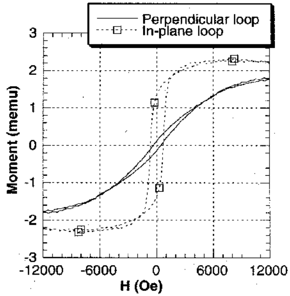

Fig. 6 In-plane and perpendicular MH loops for sample G

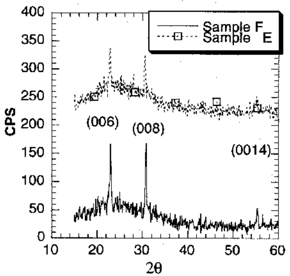

Fig. 7 The XRD curves for sample E and F

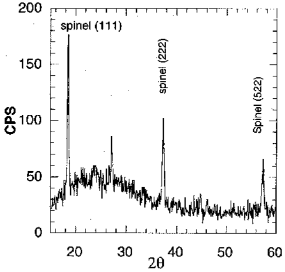

Fig. 8 The XRD curve for sample G

CONCLUSIONS

In this work we have found that barium ferrite thin films with excellent perpendicular c-axis orientation can be fabricated on Si substrate without using any buffer layer. Barium-rich thin films were deposited to compensate for the deficiency of barium content due to the inter-diffusion. For a 900 Å -thick film, the perpendicular remanent squareness for barium ferrite thin films is about 0.9, while the in-plane remanent squareness is about 0.3. It was also found that the rapid thermal annealing conditions greatly affect the magnetic properties of barium ferrite films. The perpendicular c-axis structure tends to collapse with the formation of the spinel Fe_3O_4 phase during annealing without oxygen gas flowing during annealing. The collapse of the hexagonal barium ferrite phase can be attributed to an oxygen deficiency in barium ferrite films due to the oxygen reduction in high temperature annealing. To avoid the reduction of oxygen during the annealing, an oxygen-rich annealing atmosphere is necessary.

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