Effect of Al-addition on C-axis Orientation of Ba-ferrite Film Deposited on Amorphous Aluminum Oxide (*a*-AlO) Layer.

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In this study amorphous aluminum oxide (*a*-AlO) was used as an underlayer to crystallize c-axis oriented Ba-ferrite (BaM) film. But coercivity of these films was going to decrease sharply below 20 nm. A small amount of Al is added with BaM film with *a*-AlO underlayer. The substrate temperature (T_s) was kept at 550°C. Al content was varied from 0-4.5 atomic percent. Perfect c-axis orientation of BaM thin film is possible with a small addition of Al, even for the film of 10 nm with a $\Delta\theta_{50}$ of 2.5° -2.8°. On the other hand, it was difficult to crystallize BaM/*a*-AlO film thinner than 25 nm. Coercivity of Al-BaM/*a*-AlO films in perpendicular direction was higher than that of BaM/*a*-AlO films, even for the thickness below 20 nm.

Key words: Ba-ferrite, perpendicular magnetic recording, thin film

1. Introduction

The combination of high anisotropy field, excellent mechanical durability and better chemical stability makes hexagonal barium ferrite (BaM) an excellent candidate for perpendicular magnetic recording medium ⁽¹⁾. Much effort has been concentrated on c-axis orientation ⁽²⁾ as well as the decrease of substrate temperature (T_s) during deposition ⁽³⁾ of BaM thin films for the high density perpendicular magnetic recording application. In order to prepare BaM thin films with c-axis orientation on thermally oxidized silicon wafers (SiO₂/Si), the substrate temperature have to be elevated to 620°C ⁽⁴⁾. But it is strongly recommended to reduce the crystallization temperature during deposition for the practical fabrication of the recording medium. In addition Morisako *et al.* ⁽⁵⁾ found that both Ba and Fe of BaM film deposited on SiO₂/Si wafer substrate, diffused into the substrate. This may be due to the substrate heating at high temperature.

It has been found that structural and magnetic properties of thin films can be improved by using an appropriate substrate or seed layer ⁽⁶⁾. According to Hylton *et al.* ⁽¹⁾, a buffer layer between the film and substrate is necessary to minimize the interdiffusion between film and substrate.

In our previous study, an amorphous aluminum oxide (*a*-AlO) was used as an under-layer for BaM layer ⁽⁷⁾. A high coercivity, H_c of about 4.3 kOe in perpendicular direction was obtained for film thickness (δ_{BaM}) of 35 nm and grain size was reduced to about 20 nm for δ_{BaM} of 20 nm. But coercivity of these BaM/*a*-AlO films was going to decrease sharply with decrease of film thickness especially below 20 nm. In this study, *a*-AlO was also

used as an under-layer to crystallize c-axis oriented BaM and in addition a small amount of Al is introduced into the BaM layer to increase the coercivity.

2. Experimental Procedure

The BaM/*a*-AlO thin films were deposited on thermally oxidized silicon wafers by using facing targets sputtering (FTS) system. After evacuating the chamber to a pressure below 1×10^{-7} Torr, argon (Ar) and oxygen (O₂) gases were introduced and sputtering pressure was set at 2 mTorr. A pure Al metal disk was used to deposit the underlayer (*a*-AlO) by reactive sputtering in presence of oxygen gas without any substrate heating. The thickness of *a*-AlO layer was kept constant at about of 10 nm. Then BaM layer was deposited successively onto this *a*-AlO underlayer at the substrate temperature (T_s) of 550°C. The target for the deposition of BaM layer was sintered ferrite disks with the stoichiometric composition of BaM (i.e., BaFe₁₂O_x). For the deposition of BaM film with Al (Al-BaM), a few Al plates with 2 mm × 2mm square were used along with the BaM target. Thickness of BaM layer (δ_{BaM}) was varied from 10 -35 nm.

The crystallographic properties of the BaM films were characterized by x-ray diffractometry (XRD). X-ray photoemission spectroscopy (XPS) was used to analyze the amount of Al, Ba and Fe in the BaM/*a*-AlO thin film after etching for 1 minute. Both the BaM film and *a*-AlO underlayer as well as silicon wafer substrate contain a large amount of oxygen (O), which is difficult to analyze without any chemical analysis method. Due to this problem relative percentage of Al has been used in this study. The authors use following relation to estimate the Al content after analyzing the relative amount of Al, Ba and Fe from the XPS data.

$$\text{Al (at.\%)} = [\text{Al} / (\text{Al} + \text{Ba} + \text{Fe})] \times 100$$

This estimated values of Al(at.%) are relative values not the exact one. It is difficult to measure the exact value of Al content as some part of this Al may come from the *a*-AlO underlayer for very thin BaM layer. Magnetic properties of thin films were measured by vibrating sample magnetometer (VSM). Microstructure and morphological characteristics of the BaM films were investigated by scanning electron microscope (SEM).

3. Results and Discussions

At first, Al added BaM and BaM without any Al were deposited at a substrate temperature (T_s) of 600°C. From the XRD diagrams (not shown here), it was found that the intensity of BaM(001) plane was much stronger in case of Al-BaM film than that of BaM film without Al. This led

Fig. 1 X-ray diffraction diagram (XRD) of the BaM and Al-BaM film deposited at a T_s of 550 °C.

the authors to reduce the substrate temperature to 550 °C.

X-ray diffraction diagram (XRD) of the BaM film deposited at T_s of 550 °C is shown in the Fig.1. The presence of diffraction lines from BaM (00 l) planes reveals that all the films prepared by adding 2.8at. % Al shows perfect c-axis orientation even for the film thickness of 10 nm. The intensity of these diffraction lines is also very strong. On the other hand, BaM films deposited at 550°C without any Al-addition exhibit weaker diffraction lines for a film thickness of 30 nm and no peak is found when thickness is about 20 nm. The c-axis dispersion angle, $\Delta\theta_{50}$, which was measured as the full width at half maximum of the rocking curve of BaM (008) reflection, is as small as about a range of 2.5- 2.8 degree for all the Al-BaM (Al =2.8at.%) films. On the other hand, $\Delta\theta_{50}$ of BaM films without any Al is about

Fig. 2 Dependence of coercivity, H_c of BaM/a-AlO thin film as a function of Al content.

Fig. 3 Dependence of remanent squareness, S of BaM/a-AlO thin film as a function of Al content.

5.7-6 degree. This implies that an addition of Al enhances the c-axis orientation of BaM. Though the reason is not clear, Al atom may play a role to promote the nucleation of BaM crystals in the c-axis orientation.

Fig.2 shows the dependence of coercivity as a function of Al content. It is found that coercivity in perpendicular direction ($H_{c\perp}$) increases with a small increase of Al content (2.8 at.% Al) then starts to decrease with increasing Al content whereas no significant change has been noticed in the in-plane coercivity ($H_{c\parallel}$). The highest coercivity, $H_{c\perp}$ is found when only 2.8 at. % Al is added to the BaM layer. Similar trend is also found from the remanent squareness values, S , as shown in the Fig.3. Perpendicular squareness ratio ($S_{\perp}$) increases with a small increase of Al content (2.8 at.% Al) then starts to decrease with increasing Al contents while in-plane

Fig. 4 Dependence of coercivity, H_c of BaM and Al-BaM films as a function of the thickness of BaM layer.

Fig. 5 Dependence saturation magnetization, M_s of BaM and Al-BaM film as a function of the thickness of BaM layer.

squareness ($S_{//}$) remains almost constant for any amount of Al. From these figures, it is clear that BaM film with 2.8 at.% Al shows the highest coercivity and squareness in perpendicular direction among the Al-added BaM films. This led the authors to concentrate further interest on BaM film with 2.8 at.% Al.

BaM film with 2.8 at.% Al shows better magnetic properties than that of BaM film without any Al. Fig.4 shows that the $H_{c\perp}$ of BaM film with 2.8 at.% Al is greater than that of BaM film without any Al for any value of δ_{BaM} . For Al-BaM film the maximum $H_{c\perp}$ is about 3.5 kOe, which is greater than that of BaM film without Al (2.2 kOe) for a film thickness (δ_{BaM}) of 30 nm. Al can substitute the Fe atoms of BaM ($BaFe_{12}O_{19}$) and hence it may increase the coercivity⁽⁸⁾. However coercivity decreases with decreasing film thickness of BaM layer even for Al-added BaM films. Though it is not shown here, an inter-diffused layer in between the BaM layer and α -AlO underlayer was found from the XPS depth profile. Al diffuses to the BaM layer from the α -AlO underlayer. This increases the relative amount of Al in the BaM film especially in the BaM/ α -AlO interface region and reduces the coercivity when film thickness decreases.

Fig.5 shows the dependence of saturation magnetization on thickness for both BaM and Al-added BaM films. BaM film without Al possesses M_s of about 225-250 emu/cm³ but the BaM film with 2.8 at.% Al shows higher M_s values within a range of 300-335 emu/cm³. Though it is not confirmed but it can be assumed that Al may occupy the preferential Fe sites of the BaM crystal and hence causes the increase of M_s values.

Again the remanent squareness ratio, S of BaM film with 2.8 at.% Al is also higher than that of the BaM film without any Al. $S_{\perp}$ is found about 0.7 and 0.4 for

Fig. 6 SEM images of the BaM and Al-BaM film of $\delta_{BaM} = 30$ nm deposited at a T_s of 550 °C

Al-BaM and BaM film respectively. All these magnetic data shows the better perpendicular magnetic anisotropy for the BaM film can be prepared with very few addition of Al.

Figure 6 shows the surface images of both BaM and Al-BaM film. From these images it is found that grains are coarser and grain size is larger in case of Al added BaM than those of BaM without Al. It is also found from these images grains are agglomerated instead of isolated grains. This kind of microstructure is not so desired but addition of Al improves the magnetic properties as well as crystallographic texture. Further reduction of T_s may improve the microstructure.

4. Conclusion

Ba-ferrite films with a small addition of Al and without any Al are deposited at 550°C onto α -AlO under-layer, which is deposited without substrate heating on SiO₂/Si wafer. It is found that a small addition of Al can improve magnetic and crystallographic properties remarkably. Addition of Al produces the perfect c-axis orientation even for very thin film about 10 nm with a c-axis dispersion angle of 2.5 degree. It also increases the coercivity to 3.5 kOe in perpendicular direction with a M_s of 335 emu/cm³ and a remanent squareness of 0.7 in perpendicular direction.

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