

Barium Ferrite Thin Films Without a Dead Layer

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Abstract — A barium ferrite layer with low-Ba content was used as an underlayer for a high-Ba content barium ferrite thin film. During the post-deposition annealing, the underlayer which has a lower nucleation rate than the top layer prevented grains from nucleating in the interdiffused region at the interface with the substrate. As a result, the magnetic properties were dramatically improved as indicated by much higher coercivity squareness S^* and narrower switching field distribution.

Index Terms — Barium ferrite, thin film media, underlayer.

I. INTRODUCTION

Barium ferrite thin films deposited on various types of substrates followed by high temperature annealing suffer from significant interdiffusion of the heterogeneous atoms of the substrates into the films [1], [2]. Within or even near the diffused layer (traditionally named the “dead layer”), grains of undesirable *c*-axis orientation, size, or composition may be formed. In a sense, the “dead layer” is not truly “dead,” but rather contains some magnetically active grains. Consequently, the magnetic properties, particularly the coercivity squareness S^* , deteriorate.

Our previous studies showed that higher Ba content in films dramatically enhances the nucleation rate and suppresses the growth rate during the crystallization process [3]. It is also possible to achieve full crystallization in the Ba rich films at much shorter annealing times than in the low-Ba content films at sufficiently high annealing temperatures [3].

In order to overcome the “dead layer” problem, various underlayers have been proposed by others to prevent diffusion from the substrates [4], [5]. Some researchers used oxides, e.g., sputtered SiO_2 on a carbon substrate [4]. Another underlayer is sputtered SiN on a carbon substrate [4], [5]. All these underlayers introduce new diffused layers which contain equally undesirable magnetic grains. Noble metals such as Pt were also investigated. However, a Pt underlayer was shown to promote *c*-axis perpendicular orientation in barium ferrite thin films [6]. Therefore, it cannot be used solely as a diffusion barrier if *c*-axis random orientation is desired for longitudinal recording purposes.

The low-Ba content barium ferrite underlayer reported here allows a diffused layer at its interface with the substrate, but ensures that grains are prevented from nucleating in the interdiffused layer. In the meantime, it does not introduce a new diffused layer at its interface with the top layer.

II. EXPERIMENTS

Both the low-Ba content underlayer (slightly Ba deficient with ~12 wt% BaO) and the high-Ba content barium ferrite

layer (~23 wt% BaO) were deposited at room temperature onto oxidized silicon substrates using rf diode sputtering. A sputtering power of 30 W at a rate of about 7.5 Å/min, and Ar/O₂ gas pressure of 5 mT were used. The thickness of the low-Ba content underlayer studied here was fixed at 700 Å. Data of the films with a 2000 Å thick Pt underlayer [7], [8] were also shown for comparison. The as-deposited amorphous films were crystallized by rapid thermal annealing at 790 °C for 60 s in a N₂/O₂ gas mixture.

The in-plane hysteresis loops were measured using an alternating gradient force magnetometer (AGFM). The microstructures were studied using an atomic force microscope (AFM) and a transmission electron microscope (TEM). The film compositions were analyzed by depth profiling using X-ray photoelectron spectroscopy (XPS).

III. RESULTS

An XPS depth profile of a 500 Å thick Ba rich (23 wt% BaO) film deposited on an oxidized silicon substrate without an underlayer is shown in Fig. 1. After annealing, an interdiffused layer as thick as 300 Å which contains Ba, Fe, O, and Si atoms can be observed. A similar interdiffused layer thickness was observed at different film thicknesses.

As shown in Fig. 2 (a), a 700 Å thick low-Ba content underlayer remains largely amorphous after annealing at 790 °C for 60 s. Large grains are seen in Fig. 2 (b) when a 50 Å thick high-Ba content top layer was deposited on the underlayer and after the same annealing process. The grain size becomes much smaller when the top layer is 200 Å thick as shown in Fig. 2 (c). The grain size becomes even smaller in the film which has a 1400 Å top layer as shown in Fig. 2 (d).

The films with a top layer thickness of more than 200 Å have a small grain size on the order of 400 Å ~ 500 Å, which is similar to the grain size in the thick high-Ba content films without the underlayer. The plan-view TEM micrograph of a film with a 700 Å thick top layer is shown in Fig. 3.

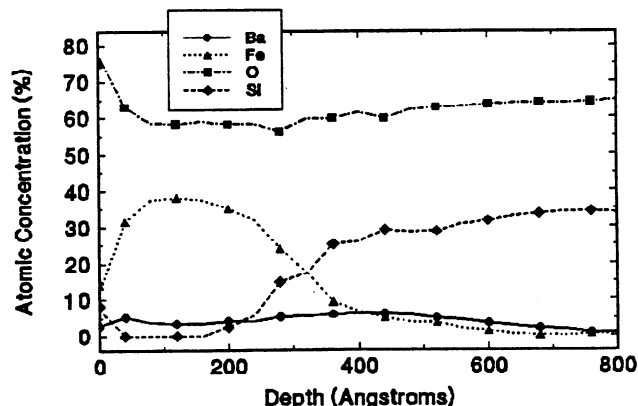


Fig. 1. XPS depth profile of a 500 Å barium ferrite film with 23 wt% BaO on SiO_2 substrate. Si atoms are shown to diffuse ~340 Å into the film.

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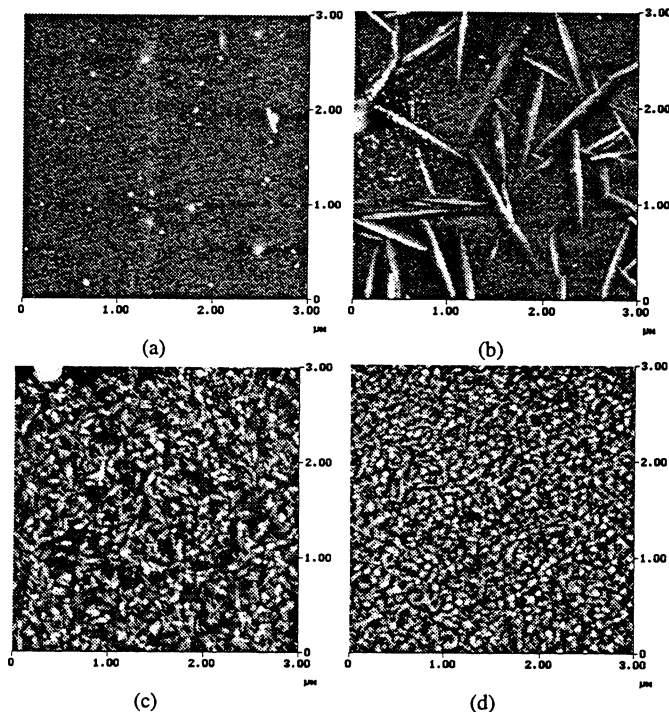


Fig. 2. (a) A 700 Å low-Ba content underlayer without high-Ba content top layers, and with the top layers thickness of (b) 50 Å, (c) 200 Å, and (d) 1400 Å, respectively.

The TEM cross-sectional micrographs of a 1200 Å high-Ba content film deposited directly on an SiO₂ substrate, and a 200 Å high-Ba content top layer on a 700 Å low-Ba content underlayer are shown in Fig. 4. It was observed in the film without the underlayer that the grains at the vicinity of the SiO₂ substrate appear to be larger than the grains near the film surface as seen in Fig. 4 (a). In the film with a low-Ba content underlayer, however, nearly all grains seem to be nucleated in the 200 Å top layer. Some grains have their c-axes perpendicularly oriented as indicated by the crystal lattice planes parallel to the film surface. These grains, which range from 300 Å ~ 800 Å in size, tend to grow laterally within the top layer. Other grains with in-plane or tilted c-axis orientations tend to grow partially into the underlayer.

As compared to the films without an underlayer or with the Pt underlayers, the values of S^* are greatly increased by using the low-Ba content underlayer as seen in Fig. 5 (a), particularly in the films with 200 - 300 Å top layers. Further

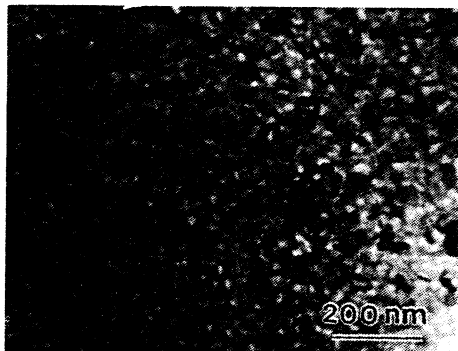


Fig. 3. TEM plan-view micrograph of a film with 700 Å high-Ba content top layer on 700 Å low Ba content underlayer.

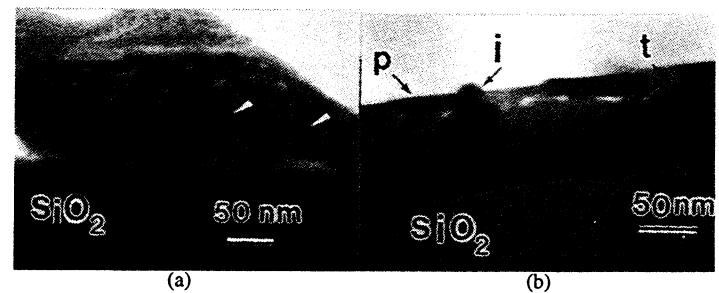


Fig. 4 Cross-section TEM micrograph of (a) a 1200 Å high-Ba content film deposited directly on SiO₂ substrate, which shows larger grains near the substrate interface, and (b) a 200 Å high-Ba content top layer on 700 Å low-Ba content underlayer. The grains with perpendicular (p), in-plane (i), and tilted (t) orientations are noted.

increase of the top layer thickness actually decreases the values of S^* , which eventually becomes the same as those of the films without the underlayer.

The values of M_{t} of films with low-Ba content underlayers, which are shown in Fig. 5 (b), are higher than one would expect from the top layer alone.

The values of H_c reach the asymptotic value of 4500 Oe when the top layer thickness is greater than 200 Å as shown in Fig. 5 (c). These values are similar to those found in high-Ba content films of more than 500 Å thickness, and deposited on SiO₂ substrates or on Pt underlayers. Larger grain size in the films with top layer thickness less than 200 Å results in lower H_c values as shown in Fig. 5 (c), which is due to incoherent rotation [8]. In films deposited on SiO₂ substrates, very low values of M_{t} and H_c at film thicknesses less than 500 Å suggested that the films were partially crystallized and had larger grain sizes. The high nucleation rate in the high-Ba content films studied here generally resulted in a small grain size as well as random easy axis orientation even when the Pt underlayer was used [7], which was previously shown to promote perpendicular c-axis orientation in stoichiometric films [6]. This would explain the high in-plane H_c values in the films deposited on a Pt underlayer. At a thickness of less than 500 Å, the films were nevertheless slightly perpendicularly oriented when a Pt underlayer was used, which resulted in small in-plane M_{t} and H_c values.

Remanence squareness, S_q of films with thickness more than 200 Å are in the range of 0.5-0.7, which is expected for the random easy axis orientation of the films. The film with a 100 Å top layer showed preferred perpendicular orientation as indicated by the higher S_q in the perpendicular direction.

The hysteresis loop of a film with a 300 Å thick top layer on a low-Ba content underlayer is shown in Fig. 6 (a). The value of S^* is at a maximum value of 0.83. Sharp slopes in the remanent magnetization curves with a narrow switching field distribution (SFD) of 0.12 were observed as seen in Fig 6 (b), which shows a film with a 200 Å top layer.

IV. DISCUSSION AND CONCLUSIONS

During the annealing process of the films with the low-Ba content underlayer, a temperature of 790 °C and time of 60 s was chosen, the combination of which is just sufficient to

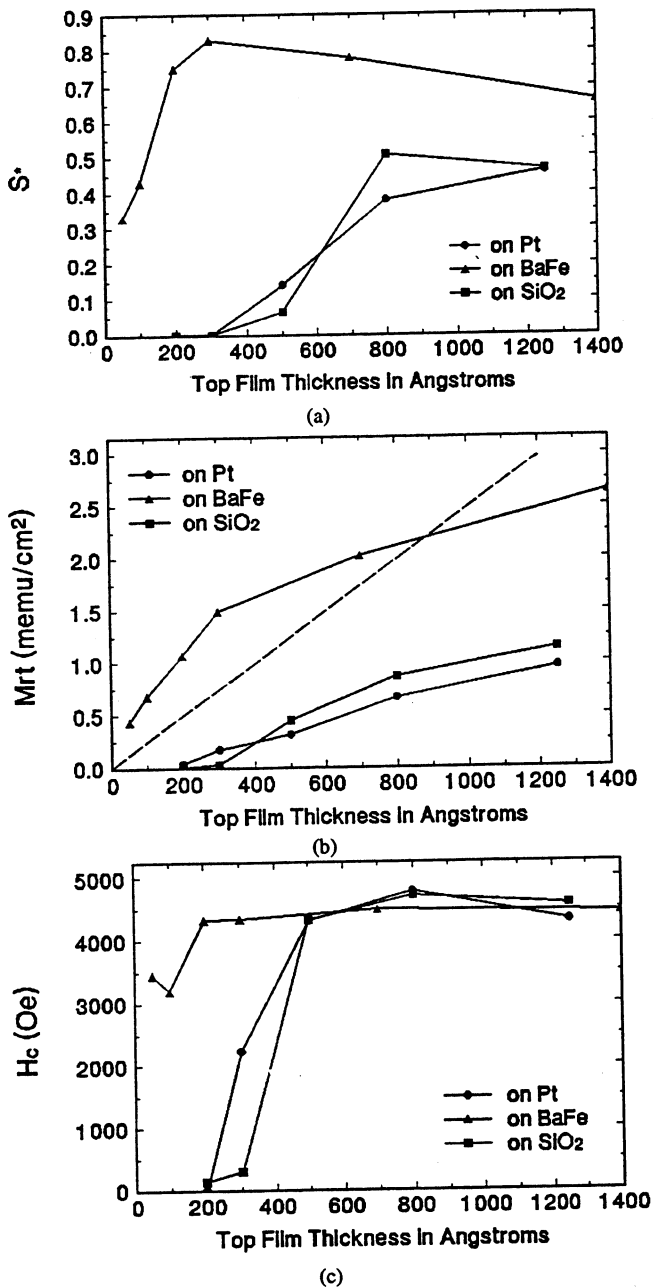


Fig. 5. Thickness dependence of (a) S^* , (b) M_{rt} , (c) H_c of high-Ba content top layers deposited on 1) 700 Å low-Ba content underlayer; 2) oxidized silicon substrate; 3) 2000 Å thick Pt underlayer. In (b) the long dashed line is the predicted thickness dependence of M_{rt} , with the following assumptions: 1) without "dead layer"; 2) the underlayers are nonmagnetic; 3) M_{rt} increases linearly with the film thickness.

fully crystallize the high-Ba content film. Such annealing conditions were, however, insufficient to crystallize the underlayer alone as suggested by Fig. 2 (a). It was found, however, that grains nucleated within the high-Ba content top layer may grow partially into the low-Ba content underlayer, as observed in the TEM cross-section micrographs. The partial grain growth into the underlayer is also consistent with the higher than expected M_{rt} values as shown in Fig. 5 (b). However, nucleation in the low-Ba content underlayer which contains the interdiffused layer was suppressed. The

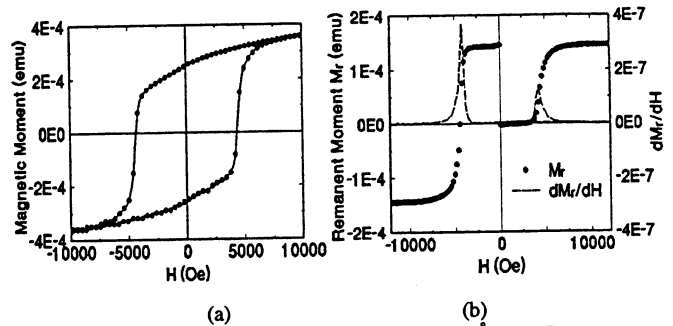


Fig. 6. (a) The hysteresis loop of a film with 300 Å high-Ba content top layer, and (b) the remanent magnetization curves M_r and the irreversible susceptibilities dM_r/dH of a film with 200 Å high-Ba content top layer.

suppression of nucleation in the interdiffused layer is believed to have resulted in more uniform magnetic properties of grains, and thereby higher S^* and narrower SFD.

The thickness of the low-Ba content underlayer must be sufficient to prevent diffusion of the substrate atoms into the top layer. The thickness of the interdiffused layer as suggested by XPS depth profile is in agreement with the values reported by Hylton et al [1]. This value is conceivably the minimum thickness required for the low Ba content underlayer. The underlayer thickness may be adjusted to accommodate different annealing conditions as well as various atoms in the substrates with different diffusivity within the barium ferrite films. It was also found that an additional 300 Å ~ 400 Å underlayer thickness beyond the interdiffusion thickness is necessary to accommodate the growth of grains nucleated within the top layer, but which partially grow into the underlayer.

It is concluded that the top layer should be thick enough to provide a significantly higher nucleation rate relative to the underlayer. A 50 Å top layer, which contains only a small number of nucleation sites, results in large grain size and a partially crystallized film as seen in Fig. 2 (b). Further increases in the top layer thickness to 200 Å ~ 300 Å resulted in significant improvements in S^* and reductions in grain size. In films with much thicker top layers, due to the relatively insignificant effect of the "dead layer", the enhancement of S^* is no longer discernible.

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