Correlation between crystalline structure and soft magnetic properties in sputtered sendust films

P. M. Dodd, R. Atkinson, and P. Papakonstantinou
Department of Pure and Applied Physics, Queen’s University of Belfast, Belfast BT7 1NN, Northern Ireland, United Kingdom

M. S. Aragh and H. S. Gamble
Electronic and Electrical Engineering Department, Queen’s University of Belfast, Belfast BT7 1NN, Northern Ireland, United Kingdom

We have investigated the change in crystallographic and magnetic properties of sendust thin films before/after annealing. Films were deposited by both rf diode and rf magnetron sputtering. rf diode-sputtered films did not possess soft magnetic properties in the as-deposited state. Films were subsequently annealed in both a conventional oven and a rapid thermal annealing (RTA) system. The coercivity of the films decreased from over 10 Oe to a minimum value of 0.6 Oe after annealing and a systematic shift in the (110) x-ray diffraction (XRD) peak position towards that of bulk sendust was observed. No additional XRD peaks were observed after annealing. The optimum annealing conditions to minimize coercivity was found to be 1–2 h at 550 °C. By comparison, films processed by RTA resulted in minimum coercivity of 0.9 Oe obtained after annealing for 2 min at 550 °C. XRD for films annealed by both RTA and oven annealing showed a similar decrease in the d spacing of the (110) plane, indicating a release of tensile strain in the films that had been introduced during deposition. rf magnetron-deposited samples possessed coercivities as low as 1.4 Oe as deposited and showed in-plane anisotropy. XRD again showed a (110) texture, however, the peak intensity of magnetron-sputtered samples was a factor of 20 less than the diode-sputtered films of the same thickness. The position of the (110) peak, however, was close to that of rf diode deposited films after annealing indicating that the magnetron-sputtered films were relatively stress-free. © 1997 American Institute of Physics. [S0021-8979(97)24708-0]

I. INTRODUCTION

Sendust thin films have applications in both inductive and magnetoresistive recording heads due to their soft magnetic properties and high degree of mechanical hardness. Both magnetotstrictive and magnetocrystalline anisotropy approach zero for an approximate composition of Al 5.4%, Si 9.6%, Fe; balance (by weight) resulting in soft magnetic properties. Generally, as-deposited sendust films do not possess very soft magnetic properties and a high-temperature anneal is usually required. Many authors have suggested that the attainment of soft magnetic properties in sendust is linked with the formation of material having the ordered DO3 superlattice structure. Although DO3 structured material has been detected in postannealed films by both x-ray diffraction (XRD) and Mossbauer spectroscopy, it remains unclear if the formation of DO3-phase material is essential for producing low coercivity films. To investigate this point further, we have deposited a series of films by rf diode sputtering and measured the magnetic and structural properties pre- and postannealing.

Conventional oven annealing has been used for many years, however, recently the rapid thermal annealing (RTA) technique has been shown to be very efficient in realizing soft magnetic properties in very short times. We determine the optimum annealing conditions to minimize coercivity in 2 μm thick diode-sputtered films for both of these methods and compare the structural and magnetic properties of the films. Films were also prepared by rf magnetron sputtering and we compare the properties of these films with those of the rf diode samples.

II. EXPERIMENTAL PROCEDURES

The 2 μm thick sendust films were deposited by both rf diode and rf magnetron sputtering. The substrate material used was Al2O3/TiC ceramic previously coated with a 10 μm thick base layer of Al2O3. Conventional oven annealing was performed in a N2 atmosphere over the temperature range 400–600 °C for 0.5–6 h. Rapid thermal annealing was performed in a tungsten-halogen lamp RTA unit again in N2 over the same temperature range for times of 15–600 s. The crystalline structure of films pre- and postannealing was investigated by XRD (Cu Kα) operated in the standard θ–2θ geometry. Hysteresis loops of the films were measured by a BH looper operating at 900 Hz. Grain structure was investigated by atomic force microscopy (AFM).

III. RESULTS AND DISCUSSION

A. Films deposited by rf diode sputtering

Figure 1(a) shows an XRD scan from the sendust target. The target scan shows three peaks characteristic of a bcc structure. Figure 1(b) shows the XRD diagram for a 2 μm thick diode-sputtered film. It shows a single peak of very high counts indicating a bcc structure with strong alignment along the (110) bcc direction. This peak may also be assigned to reflections from the (220) plane from a DO3 structure. The position of the film’s (110) peak, however, is...
shifted to a lower $2\theta$ value than the corresponding peak from the bulk sendust target [$2\theta$ (bulk) = 45.09°, $2\theta$ (film) = 44.80°], despite the fact that the composition of the film was determined by electron microprobe analysis to be nominally the same as the target material. The coercivity of the film in the as-deposited state was measured to be 10.5 Oe and the magnetic properties were isotropic in the film plane. Grain sizes of 40–60 nm were measured by AFM.

Films were then annealed in a conventional oven over the temperature range 400–600 °C for 2 h. The coercivity decreased to a minimum value of 0.6 Oe, which was attained at 550 °C. No additional peaks were detected after annealing at which time both the coercivity and (110) peak position remain constant. The peak shifts as measured from the position of the unannealed film correspond with the reduction in grain size with annealing time. This was later confirmed by AFM observations, which indicated an increase in grain size from an average diameter of about 50 nm as-deposited to 70–80 nm after annealing.

A series of films were then annealed at 550 °C in the time range 0.5–6 h. Again no additional XRD peaks appeared after annealing and the (110) peak position shifted towards the bulk (110) position with increasing anneal duration.

Figure 2 shows the variation of coercivity and (110) peak position with annealing time. A minimum coercivity is attained after 1–2 h annealing after which time both the coercivity and (110) peak position are constant. The peak shift as measured from the position of the unannealed film correspond with the reduction in grain size with annealing time. This was later confirmed by AFM observations, which indicated an increase in grain size from an average diameter of about 50 nm as-deposited to 70–80 nm after annealing.

Films were then annealed in the RTA processor over the same temperature range. The optimum RTA temperature was found to be 500–550 °C. The coercivity values are not quite as low as those obtained by conventional oven annealing, however, this method has the obvious advantage of much shorter cycle times. A similar shift in the (110) XRD peak for DO$_3$ ordering to occur. Therefore, we conclude that in the case of our samples, the most important factor in minimizing coercivity appears to be the relaxation of stress introduced during deposition. Residual film stress has been reported to have a considerable effect on soft magnetic properties in several other Fe-based alloy films such as FeTaN.

A series of films were then annealed at 550 °C in the time range 0.5–6 h. Again no additional XRD peaks appeared after annealing and the (110) peak position shifted towards the bulk (110) position with increasing anneal duration.

Figure 2 shows the variation of coercivity and (110) peak position with annealing time. A minimum coercivity is attained after 1–2 h annealing after which time both the coercivity and (110) peak position remain constant. The peak shifts as measured from the position of the unannealed film correspond with the reduction in the films coercive force. The full width at half-maximum (FWHM) of the (110) peak also decreases with annealing time indicating an increase in grain size with annealing time. This was later confirmed by AFM observations, which indicated an increase in grain size from an average diameter of about 50 nm as-deposited to 70–80 nm after annealing.

Films were then annealed in the RTA processor over the same temperature range. The optimum RTA temperature was found to be 500–550 °C. The coercivity values are not quite as low as those obtained by conventional oven annealing, however, this method has the obvious advantage of much shorter cycle times. A similar shift in the (110) XRD peak for DO$_3$ ordering to occur. Therefore, we conclude that in the case of our samples, the most important factor in minimizing coercivity appears to be the relaxation of stress introduced during deposition. Residual film stress has been reported to have a considerable effect on soft magnetic properties in several other Fe-based alloy films such as FeTaN.

A series of films were then annealed at 550 °C in the time range 0.5–6 h. Again no additional XRD peaks appeared after annealing and the (110) peak position shifted towards the bulk (110) position with increasing anneal duration.

Figure 2 shows the variation of coercivity and (110) peak position with annealing time. A minimum coercivity is attained after 1–2 h annealing after which time both the coercivity and (110) peak position remain constant. The peak shifts as measured from the position of the unannealed film correspond with the reduction in the films coercive force. The full width at half-maximum (FWHM) of the (110) peak also decreases with annealing time indicating an increase in grain size with annealing time. This was later confirmed by AFM observations, which indicated an increase in grain size from an average diameter of about 50 nm as-deposited to 70–80 nm after annealing.

Films were then annealed in the RTA processor over the same temperature range. The optimum RTA temperature was found to be 500–550 °C. The coercivity values are not quite as low as those obtained by conventional oven annealing, however, this method has the obvious advantage of much shorter cycle times. A similar shift in the (110) XRD peak

<table>
<thead>
<tr>
<th>$T_{\text{anneal}}$ (°C)</th>
<th>$H_c$ (Oe)</th>
<th>$2\theta_{(110)}$ (degrees)</th>
</tr>
</thead>
<tbody>
<tr>
<td>No anneal</td>
<td>10.5</td>
<td>44.80</td>
</tr>
<tr>
<td>400</td>
<td>2.2</td>
<td>44.92</td>
</tr>
<tr>
<td>450</td>
<td>1.4</td>
<td>45.96</td>
</tr>
<tr>
<td>500</td>
<td>1.0</td>
<td>45.00</td>
</tr>
<tr>
<td>550</td>
<td>0.6</td>
<td>45.04</td>
</tr>
<tr>
<td>600</td>
<td>0.7</td>
<td>45.04</td>
</tr>
</tbody>
</table>
with annealing time was observed and Fig. 3 shows the variation of coercivity and $(110)$ peak position with RTA cycle time at 550 °C.

Significantly, one can see by comparing Figs. 2 and 3 that not only is the coercivity slightly higher for RTA samples but also the $(110)$ peak position does not shift quite as large an amount as was observed for the oven annealed samples. The lowest coercivity value obtained by RTA was 0.85 Oe, which was produced by annealing at 550 °C for 2 min.

B. Films produced by rf magnetron sputtering

By comparison, the rf magnetron-deposited films showed much lower coercivity than rf diode-sputtered films in the as-deposited state. The magnetron-sputtered films also showed significant in-plane anisotropy, which is a desirable property for many device applications. This anisotropy is a result of stray fields from the magnetron interacting with the growing film during deposition. Figure 4(a) shows the easy and hard axis loops measured from a 2 μm thick magnetron-sputtered film without any annealing. The hard and easy axis coercivities are approximately equal at 1.4 Oe with an anisotropy field of about 12 Oe. The XRD pattern for the magnetron-deposited films shows a single $(110)$ peak. The FWHM of this peak is considerably larger than obtained from diode-sputtered samples [FWHM (diode) =0.25°, FWHM (magnetron)=0.65°] and the peak intensity is a factor of 20 less than diode-sputtered samples of the same thickness. Interestingly, the $(110)$ peak is positioned close to the $2\theta$ value of the rf diode-samples after annealing (i.e., closer to the bulk value).

These observations would tend to suggest that the rf magnetron-sputtered films have lower levels of internal stress than diode-sputtered films of similar thickness and reinforces the suggestion that residual stress is the dominant factor affecting the coercivity.

The different crystallographic and magnetic properties of the magnetron-sputtered samples may be due to the greatly reduced bombardment of the growing film by ions and electrons produced in the Ar plasma. The plasma discharge during magnetron sputtering is confined to within 2–3 cm of the sputter target and does not extend to the substrate position, while during diode sputtering the growing film can be seen to be engulfed in the plasma discharge. A more detailed study of the factors affecting the magnetic properties of magnetron-sputtered films is currently underway.

ACKNOWLEDGMENTS

The authors would like to acknowledge funding from Seagate Technology (Ireland) and the Industrial Research and Technology Unit for Northern Ireland under the START program.