Soft Magnetic Properties and Microstructures of As-sputtered Fe-M-O (M=Hf, Al) Films

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Fe-M-O (M=Hf, Al) films are fabricated by Ar+O₂ reactive sputtering and their magnetic properties are investigated. Excellent soft magnetic properties at high frequency are obtained in Fe-Hf-O and Fe-Al-O thin films even in an as-deposited state; for example, Fe₈₂Hf_{3.4}O_{14.6} and Fe_{90.7}Al₂O_{7.3} films exhibit the values of 17.7 and 18.5 kG for saturation magnetization, 1.0 and 0.65 Oe for coercivity, and 2,200 and 3,000 for the effective permeability at 100 MHz, respectively. Microstructures, magnetic anisotropy and electrical resistivity are investigated to understand the magnetic properties of the thin films.

Key words: soft magnetic thin films, Fe-Hf-O alloy, Fe-Al-O alloy, nanocrystalline grain

1. Introduction

There is an increased demand for micro-magnetic devices and high density recording heads, operating in the high frequency range (> 10 MHz). There has been some improvement of high frequency characteristics by the formation of multilayered structure and the increase of magnetic anisotropy and electrical resistivity in soft magnetic thin films¹⁾. The previous improvement is not sufficient to meet the required properties for applications. Thus it is strongly required to develop new soft magnetic films.

Makino et. al found that nanocrystalline Fe-M-O (M=Hf, Zr)²⁾ films possessed good high frequency characteristics with reasonable soft magnetic properties. However, the Fe-M-O films required a post-annealing treatment to form nanocrystalline structures. Song et al. reported that as-deposited Fe-Hf-C-N nanocrystalline films³⁾ exhibit excellent high frequency soft magnetic properties without post-annealing, which causes several problems during the fabrication of integrated electromagnetic devices.

In this work, we fabricate as-deposited nanocrystalline Fe-M-O (M=Hf, Al) films and study the soft magnetic properties and high frequency characteristics of the as-deposited films.

2. Experimental Procedure

Fe-M-O films with a thickness of 1 μm were deposited on Si wafers by RF magnetron sputtering under Ar+O₂ atmosphere. Total gas pressure was maintained at 2 mTorr and input power fixed at 3.8 W/cm². The composition of the films was controlled by O₂ partial pressure (up to 15 %) and the number of pellets of M elements, placed on pure Fe discs.

The composition of Fe-M-O films was analyzed by Rutherford backscattering spectroscopy (RBS) and secondary ion mass spectroscopy (SIMS). Saturation magnetization ($4\pi M_s$) and coercivity (H_c) were measured using a vibrating sample magnetometer (VSM). Magnetic anisotropy field (H_k) and the dispersion angle of magnetic anisotropy (α_{90}) were measured by a M-H loop tracer and electrical resistivity (ρ) by the four point probe method. Effective permeability (μ_{eff}) was measured in the frequency range of 0.5~100 MHz by a figure-8 coil method. Transmission electron microscopy (TEM) was used to analyze the microstructures of the films.

3. Results and Discussion

Fig. 1 shows the bright and the dark field TEM images and the diffraction patterns of an

Fig. 1 The bright and the dark field TEM images and the diffraction patterns of as-deposited Fe-M-O films

(a) Fe₈₂Hf_{3.4}O_{14.6} (b) Fe_{90.7}Al₂O_{7.3}

as-deposited $\text{Fe}_{82}\text{Hf}_{3.4}\text{O}_{14.6}$ and an as-deposited $\text{Fe}_{90.7}\text{Al}_2\text{O}_{7.3}$ film, which are fabricated under the sputtering conditions described in experimental procedure. TEM images prove the formation of nanocrystalline structures in as-deposited Fe-M-O films. The diffraction patterns show the existence of two phases, α -Fe and M-oxide in both films. According to the analysis of dark field images, the average sizes of α -Fe grains and oxide precipitates in the $\text{Fe}_{82}\text{Hf}_{3.4}\text{O}_{14.6}$ are 8 nm and 5 nm and those of α -Fe grains and oxide precipitates 12 nm and 6 nm in the $\text{Fe}_{90.7}\text{Al}_2\text{O}_{7.3}$ film, respectively. It is found that the $\text{Fe}_{82}\text{Hf}_{3.4}\text{O}_{14.6}$ and the $\text{Fe}_{90.7}\text{Al}_2\text{O}_{7.3}$ film show the best soft magnetic properties with $4\pi M_s$ of 17.7 and 18.5 kG, H_c of 1.0 and 0.65 Oe, respectively.

Fig. 2 shows the frequency dependency of the real part (μ') and the imaginary part (μ'') of the permeability for the $\text{Fe}_{82}\text{Hf}_{3.4}\text{O}_{14.6}$ and the $\text{Fe}_{90.7}\text{Al}_2\text{O}_{7.3}$ film. The μ' values of the $\text{Fe}_{82}\text{Hf}_{3.4}\text{O}_{14.6}$ and the $\text{Fe}_{90.7}\text{Al}_2\text{O}_{7.3}$ film remain 2200 and 3000 up to 100 MHz, respectively. Conventional soft magnetic nanocrystalline films, such as Fe-Si-Al-Hf-C film⁴⁾, usually show high permeability at low frequency, but the permeability decreases rapidly as the frequency increases (>10 MHz). Though post-annealed nanocrystalline Fe-M-O films with high M and O contents exhibit excellent high frequency characteristics, soft magnetic properties such as the saturation magnetization, the coercivity and the permeability are not as good as the present Fe-M-O films with

Fig. 2 The frequency dependency of the real part (μ') and the imaginary part (μ'') of the permeabilities of an as-deposited $\text{Fe}_{82}\text{Hf}_{3.4}\text{O}_{14.6}$ film and an as-deposited $\text{Fe}_{90.7}\text{Al}_2\text{O}_{7.3}$ film

low M and O contents, which also possess high frequency characteristics simultaneously.

Fig. 3 shows magnetization curves for the $\text{Fe}_{82}\text{Hf}_{3.4}\text{O}_{14.6}$ and the $\text{Fe}_{90.7}\text{Al}_2\text{O}_{7.3}$ film. H_k and the dispersion angle (α_{90}) for the $\text{Fe}_{82}\text{Hf}_{3.4}\text{O}_{14.6}$ film are 10 Oe and 8.5° , respectively. These values are slightly less than to the values of a $\text{Fe}_{61}\text{Hf}_{13}\text{O}_{26}$ film reported by Hayakawa⁵⁾. The $\text{Fe}_{90.7}\text{Al}_2\text{O}_{7.3}$ film has the H_k value of 6.5 Oe. The electrical resistivities of the $\text{Fe}_{82}\text{Hf}_{3.4}\text{O}_{14.6}$ and the $\text{Fe}_{90.7}\text{Al}_2\text{O}_{7.3}$ film are $150 \mu\Omega\text{cm}$ and $80 \mu\Omega\text{cm}$, respectively. These electrical resistivities of the both films are very different to those of the $\text{Fe}_{61}\text{Hf}_{13}\text{O}_{26}$ film ($910 \mu\Omega\text{cm}$). Fig. 4 shows the frequency dependency of quality factors ($Q = \mu'/\mu''$) for the $\text{Fe}_{82}\text{Hf}_{3.4}\text{O}_{14.6}$ and $\text{Fe}_{90.7}\text{Al}_2\text{O}_{7.3}$ films. The as-deposited films exhibit high values of the quality factor of 15~25 at 20 MHz.

According to the Landau-Lifshitz equation and the eddy current effect, the effective permeability of a soft magnetic thin film increases in high frequency range as ρ and H_k increase and as film thickness decreases. Specially, the high frequency characteristics are strongly related to the eddy current loss. In order to decrease the eddy current loss, thin films with high electrical resistivity are used in high frequency range. In the same manner, nanocrystalline Fe-M-O films for good high frequency characteristics need to increase the amount of oxides, which should

Fig. 3 Magnetization curves for an as-deposited $\text{Fe}_{82}\text{Hf}_{3.4}\text{O}_{14.6}$ film and an $\text{Fe}_{90.7}\text{Al}_2\text{O}_{7.3}$ film

Fig. 4 The frequency dependency of quality factors for an as-deposited Fe₈₂Hf_{3.4}O_{14.6} film and an Fe_{90.7}Al₂O_{7.3} film

deteriorate the soft magnetic properties. Based on electrical resistivity and anisotropy field, the better high frequency characteristics and the lower loss of the Fe₈₂Hf_{3.4}O_{14.6} film, compared with the Fe_{90.7}Al₂O_{7.3} film, are well explained. However, the high frequency characteristics of the as-deposited films, which have excellent soft magnetic properties, are almost as good as those of the Fe₆₁Hf₁₃O₂₆ film⁵⁾ in spite of lower electrical resistivity. It is due to that the skin depth of the present as-deposited films at 100 MHz is close to 1 μm so the resistivity of 100 μΩcm is good enough to minimize the eddy current loss.

4. Summary

Nanocrystalline Fe-M-O films are fabricated in an as-deposited state by Ar+O₂ reactive sputtering. Transmission electron microscopy shows that the Fe-M-O films consist of two phases: fine α-Fe grains with the size of around 10 nm and M-oxide precipitates of around 5 nm. An as-deposited Fe₈₂Hf_{3.4}O_{14.6} and an Fe_{90.7}Al₂O_{7.3} film exhibit excellent soft magnetic properties, with 4πM_s of 17.7 and 18.5 kG, H_c of 1.0 and 0.65 Oe, Q of 25 and 15 at 20 MHz and μ_{eff} of about 2,200 and 3,000 up to 100 MHz, respectively. These excellent soft magnetic properties and high frequency characteristics and low loss result from the nanocrystalline structures, high electrical resistivity and anisotropy field. These properties give several advantages in the applications of the as-deposited films to high frequency magnetic integrated devices and high density recording heads.

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