Effects of the Hf content on the microstructure and magnetic properties of Co-Hf-Ta thin films

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Abstract. Effects of the Hf content in Co-Hf-Ta thin films on the microstructure and magnetic properties were investigated in this study. It was found that appropriate Hf addition can effectively refine the Co grain size. Co grain sizes sharply decreased from 50 nm down to 2.3 nm with increasing the Hf content from 1.02 at.% to 2.81 at.%, leading to the reduced magneto-crystalline anisotropy. The Co-Hf-Ta thin films with small Co grains reveal low anisotropy field, low coercivity, and high resistivity. By optimizing the Hf content, the film with Hf concentration of 2.81 at.% exhibits excellent soft magnetic properties: high saturation magnetization ($4\pi M_S \sim 13.6$ kG), and low coercivity ($H_C \sim 0.6$ Oe). The effective permeability of the film reaches 800 and remains constant up to 1 GHz.

1. Introduction

Recently, lots of attention has been focused on the size-reduction of dc-dc converters for the application in portable electronic products. In these devices, passive components, especially inductors, occupy the largest space. Thus miniaturizing the inductors is strongly desired [1-5]. High permeability ferromagnetic thin films, performing the crucial task of concentrating and shaping magnetic flux, could efficiently enhance the inductance of power inductors in dc-dc converters [6]. With the increasing demand of small inductors operated at several megahertz, the development of high permeability soft magnetic thin films with large saturation magnetization ($4\pi M_S$), low coercivity ($H_C$) and suitable anisotropy field ($H_A$) is required. Co-M-Ta (M = nonmagnetic metal) is an attractive class of high permeability materials [7-9]. To obtain superior permeability and optimal magnetic properties, doping a suitable quantity of other elements (M = Hf, Zr, Ce...) is necessary. The amount of added M element is minor in comparison with the Co content. However, it is critical to microstructure control [10]. Appropriate addition of the M element could effectively prevent Co grain growth and improve soft magnetic properties. In this study, soft magnetic Co-Hf-Ta thin films were fabricated for applications as power inductors. The effects of the Hf content on the microstructure and magnetic properties of Co-Hf-Ta thin films would be probed and discussed.

2. Experimental procedures

Soft magnetic films with composition of $\text{Co}_{100-x-y}\text{Hf}_x\text{Ta}_y$ ($1.02 \leq x \leq 8.22$, $y \sim 5.4$) were
fabricated onto n-type Si (100) substrates at ambient temperature by dc reactive magnetron co-sputtering with the base pressure better than $3 \times 10^{-6}$ torr. The targets Co, Hf and Ta were utilized to deposit these films. The Hf content was varied by adjusting the gun power on the Hf target from 5 W to 30 W. The film thickness was about 750 nm for all samples. Magnetization versus field and the complex permeability were measured by vibrating-sample magnetometer (VSM) and permeameter (Ryowa PMF-3000), respectively. The domain structure in Co-Hf-Ta films was investigated by MFM (Dimension 3100, Veeco). The resistivity was obtained from standard four-point measurement. The microstructure was examined using x-ray diffractometer (XRD) and transmission electron microscopes (TEM, JEOL JEM-2010). The composition of the films was evaluated by field emission electron probe X-ray microanalyzer (FE-EPMA, JEOL JXA-8500F).

3. Results and discussion

Figure 1 reveals the x-ray diffraction (XRD) patterns of Co-Hf-Ta thin films with various Hf contents. A significant change in microstructure is revealed as the Hf content increases from 1.02 at.% to 2.81 at.%. However, further Hf addition from 2.81 at.% to 8.22 at.% has little impact on the microstructure. The sharp (002) peak in the XRD patterns of films with 1.02 at.% Hf shows obviously the preferred orientation in Co grains. When the Hf content increases to 2.81 at.% or more, a broad peak appears in the XRD patterns, implying the nanocrystalline structure with relatively random-oriented Co grains. Besides, according to Scherer's equation, Co grain size sharply decreases from 50 nm (1.02 at.% Hf) down to 2.3 nm (2.81 at.% Hf) and then remains almost constant, around 2 nm for 2.81 ~ 8.22 at.% Hf. Therefore, appropriate Hf addition to the Co-Hf-Ta films can effectively reduce the Co grain size.

![X-ray diffraction patterns for the Co-Hf-Ta films with a Hf content variation of 1.02-8.22 at.%](image)

Fig. 1. X-ray diffraction patterns for the Co-Hf-Ta films with a Hf content variation of 1.02-8.22 at.%.
The dependence of anisotropy field ($H_K$) and coercivity ($H_C$) of Co-Hf-Ta films on the Hf content is shown in Fig. 2. $H_K$ drops drastically as the Hf content increases from 1.02 at.% to 2.81 at.% and then remains almost constant. $H_C$ shows similar trends with $H_K$. The change in $H_K$ and $H_C$ for films with different Hf contents is suggested to originate from the microstructure variations.

Fig. 2. Anisotropy field ($H_K$) and coercivity ($H_C$) for the Co-Hf-Ta films with various Hf contents.

At 1.02 at.% Hf content, the film reveals columnar structure with large Co grains of 50 nm, as shown in Figs. 1 and 2. For large grains, magnetization follows the easy magnetic directions in the single grains [11]. Domains can be formed within the grains. Therefore, the magneto-crystalline anisotropy $K_1$ of the crystallites determines the magnetization process.

When the Hf content increases to 2.81 at.%, the film exhibits nanocrystalline structure with small Co grain size of 2.3 nm. According to the random anisotropy model [11-12], the effective anisotropy $\langle K \rangle$ affecting the magnetization process results from averaging the $N = (l_{\text{ex}} / D)^3$ grains of size D within the volume $V = L_{\text{ex}}^3$, where $L_{\text{ex}}$ is the exchange length. Thus, $\langle K \rangle$ is determined by the mean fluctuation amplitude of anisotropy energy in the N grains, i.e. $\langle K \rangle \approx K_1 / \sqrt{N}$. In other words, in the films with small grain sizes, several grains are magnetically coupled to each other by ferromagnetic exchange interaction. As a result, the magneto-crystalline anisotropy is averaged out and thus reduced.

Based on the theory of ferromagnetism, anisotropy field ($H_K$) is proportional to the effective anisotropy $\langle K \rangle$, i.e. $H_K \approx 2 \langle K \rangle / M_S$ [13]. Besides, coercivity ($H_C$) is also related to $\langle K \rangle$, i.e. $H_C = p_c \langle K \rangle / M_S$, where $p_c$ is a material parameter [11,14]. Thus, for the 1.02 at.% Hf film with large grain size of 50 nm, $\langle K \rangle$ approaches $K_1$, resulting in high $H_K$ and $H_C$. For the films with Hf concentration of 2.81 at.% or more, $\langle K \rangle$ approximates to $K_1 / \sqrt{N}$, leading to low $H_K$ and $H_C$. The
arguments addressed above clarify the significant decrease in $H_K$ and $H_C$ as the Hf content increases from 1.02 at.% to 2.81 at.% in Fig. 3. Furthermore, $H_K$ and $H_C$ for the films with higher Hf content ($\geq$ 2.81 at.%) remain almost constant due to the slight difference in grain sizes.

The dramatic decrease in $H_K$ and $H_C$ as the Hf content increases from 1.02 to 2.81 at.% leads to two entirely different permeability spectra. The corresponding hysteresis loop and MFM image reveal that the film with 1.02 at.% Hf content has characteristic stripe-domain behavior with high $H_C$ and $H_K$. Stripe domains lead to high coercivity and low permeability, both of which correspond to the frequency dependence of complex permeability spectra. The real part permeability ($\mu'$) is very low (~20-30) due to the high $H_K$. Besides the imaginary part permeability ($\mu''$) rises up at low frequencies because of large hysteresis loss originating from the high $H_C$. On the other hand, the film with 2.81 at.% Hf content exhibits excellent soft magnetic properties with large $4\pi M_S$ and low $H_K$. According to Stoner-Wohlfarth theory, permeability ($\mu'$) is inversely proportional to $H_K$ ($\mu' \approx 4\pi M_S/H_K$) [15]. Thus, this film is expected to have relatively high $\mu'$. Fig. 3 shows the frequency dependence of complex permeability spectra for the film with 2.81 at.% Hf content. The experimental results fit well with the calculated values. $\mu'$ is up to 800 and flattens almost to 1 GHz. Besides, the cut-off frequency ($f_{\text{cut-off}}$), the ferromagnetic resonance frequency ($f_{\text{FMR}}$) and the half width in FMR ($\Delta f_{1/2}$) are 1.1 GHz, 1.6 GHz and 1.02 GHz, respectively.

![Fig. 3. Frequency dependence of complex permeability spectra for the Co-Hf-Ta films with 2.81 at.% Hf content.](image-url)
To sum up, appropriate Hf addition to the Co-Hf-Ta magnetic thin films can effectively reduce grain sizes, enhance soft magnetic properties, and achieve a high permeability.

4. Conclusions

In this study, effects of the Hf content on the microstructure and magnetic properties of the Co-Hf-Ta thin film were investigated. Appropriate Hf addition can effectively reduce Co grain sizes and improve magnetic properties. By optimizing the Hf content, the film with Hf concentration of 2.81 at.% exhibits excellent soft magnetic properties: high saturation magnetization ($4\pi M_S \sim 13.6$ kG), and low coercivity ($H_C \sim 0.6$ Oe). The effective permeability of the film reaches 800 and remains constant up to 1 GHz. The high value of permeability and the soft magnetic properties reveal that the Co-Hf-Ta thin films possess potential for applications as power inductors.

References

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