High Frequency Permeability of Electrodeposited CoNiFeS Film with High B_s and ρ

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Abstract--- A high frequency permeability for an electroplated CoNiFeS film having high saturation magnetic flux density(B_s) of 1.7T and high resistivity(ρ) of $51\mu\Omega\text{-cm}$ was investigated in comparing with CoNiFe film having $\rho=25\mu\Omega\text{-cm}$ value. Both films had suitable thermal stability on magnetic properties up to 300°C and magnetic anisotropy(H_k) could be controlled by annealing with magnetic field. Higher permeability of the CoNiFeS film could be attributed to decrease the eddy current loss based on twice higher resistivity compared with the CoNiFe film.

Key word: electroplating, soft magnetic CoNiFe and CoNiFeS film, high B_s and ρ

I. INTRODUCTION

Recently, a magnetic recording density has been increasing to such a level that the coercivity(H_c) of a recording medium has reached a value as high as 2500Oe. For such a high H_c medium, a write head core of a merged MR head should have a saturation magnetic flux density(B_s) higher than that of a commonly used permalloy film. For a soft magnetic core to be compatible with the high magnetic recording density, its resistivity(ρ) should also be high to avoid an eddy current loss at high frequencies.

The CoNiFe alloy films have been studied for an application as magnetic core materials of thin-film heads or MR heads due to high B_s in the low saturation magnetostriction(λ_s) region[1,2]. As the same purpose, we have tried to prepare the CoNiFe film by electroplating method, which is a currently used in the industrial process for the head core of MR magnetic heads. The electroplated CoNiFe films prepared from a bath containing saccharin, which is a conventional additive in electroplating permalloy bath, has high B_s in the low λ_s region[3], however, the ρ value is the same as the well known permalloy film [4]. To increase the ρ value, the electroplated CoNiFeS film, possessing high B_s of 1.7T and ρ of $51\mu\Omega\text{-cm}$ in the low λ_s region was developed by our research group. The film contained a few atomic percent of sulfur from a bath with adding thiourea instead of

saccharin, and the higher ρ is achieved[5].

In this study, the high frequency permeability of the CoNiFeS film is investigated in comparison with the CoNiFe film.

II. EXPERIMENTAL

The composition and plating conditions of the plating baths used are listed in Table 1. All samples of 1.0 μm thick were plated using a paddle cell system. The glass wafer, which has sputtered permalloy(110nm) with Ti(30nm) adhesion layer, was used as substrate for measuring magnetic properties. The B_s and H_c values were measured by using VSM with an earth magnetic field compensation circuit. The frequency dependence of permeability was measured by permeance meter [6] and 8 figure coil method[7]. The samples for measuring permeability were electroplated with applying the static magnetic field(600Oe). The λ_s was measured by optical cantilever method and the ρ was measured by a four-point probe method. The structure and composition of each film were examined by TEM and EDX.

Table 1 Composition and plating conditions of the electroplating baths for CoNiFe and CoNiFeS films

Film	CoNiFe	CoNiFeS
Chemicals	mol dm ⁻³	mol dm ⁻³
FeSO ₄ ·7H ₂ O	0.015	0.015
CoSO ₄ ·7H ₂ O	0.048	0.060
NiSO ₄ ·6H ₂ O	0.200	0.200
H ₃ BO ₃	0.40	0.40
NH ₄ Cl	0.28	0.28
Additive	Saccharin 2.0 g/dm ⁻³	Thiourea 0.02 g/dm ⁻³
Bath temp.	Room temp.	Room temp.
pH	2.8	2.8
Rotation speed	0.4m min ⁻¹	0.4m min ⁻¹
Current density	15 mA cm ⁻²	20 mA cm ⁻²

III. RESULTS AND DISCUSSIONS

Table 2 shows the characteristics of the CoNiFe and CoNiFeS films using in this study. The CoNiFe film has $B_s = 1.6\text{T}$ and $\rho = 25\mu\Omega\text{-cm}$ and $H_k = 20\text{Oe}$ in the as-plated conditions. The CoNiFeS film has higher $B_s = 1.7\text{T}$ and $\rho = 51\mu\Omega\text{-cm}$ and the same $H_k = 20\text{Oe}$. First, thermal stabilities on H_c and B_s were studied for the CoNiFe and CoNiFeS films. Samples were annealed at 200, 250, 300, 350, 400°C and kept for 1hr at desired temperature under the 10^{-5} torr in vacuum without applying magnetic field. H_c and B_s values were measured by using VSM before and after heat treatment. The B_s values of the both films did not change after the heat treatment. Figure 1 shows the H_c values on various heat treatment temperatures for the each film. The H_c values of both films do not change up to 300°C. Above 300°C annealing, the H_c value of the CoNiFe film increases grater than that of the CoNiFeS film. In order to clarify the reason why the both films show such a difference, the films of as-plated and annealed were analyzed by using TEM. The samples for used this analysis were prepared by ultra-microtomy method, and the cross-sectional image was observed. Figure 2 shows the TEM bright field images and their diffraction patterns of as-plated CoNiFe and CoNiFeS films. From these figures, the CoNiFeS film was consisted of fine crystal grains with approximately 5~10nm ϕ in diameter, however, the CoNiFe film was consisted of larger crystal grains with approximately 20~30nm ϕ . The annealed films under 300°C had almost the same microstructure comparing with those of as-plated films. Figure 3 shows the TEM bright field images and their TED patterns for 350°C annealed films. In Fig.3, grain sizes of the annealed films become bigger than those of as-plated films and the mean grain diameters for CoNiFeS and CoNiFe films are 10~20nm and 20~35nm, respectively. Figure 4 shows the results of 400°C annealed films, the grain size for CoNiFe film also becomes larger approximately 30~50nm ϕ , however, it doesn't great change for CoNiFeS film. The results of structural change from TED analysis give the help for understanding of the crystallite growth. As for the CoNiFe film, the grain size becomes large. On the other hand, the TED pattern of annealed CoNiFeS film has many rings except for those of fcc-Co phase. The excess rings were identified with those from FeS. It is inferred that the FeS formation by annealing plays an important role of preventing the crystallite growth. From these results, the reason of the difference of H_c for the CoNiFe and CoNiFeS films at 400°C was attributed to the crystal grain size. Therefore, the magnetic properties in the as-plated films were retained up to 300°C annealing because the films were consisted of the same

microstructure in comparison with the as-plated films.

Table 2 Characteristics of as-plated CoNiFe and CoNiFeS films

Film	CoNiFe	CoNiFeS
Composition	$(\text{Co}_{75}\text{Ni}_{16}\text{Fe}_9)_{99.65}\text{S}_{0.35}$	$(\text{Co}_{73}\text{Ni}_{12}\text{Fe}_{15})_{96.50}\text{S}_{3.50}$
B_s / T	1.6	1.7
H_c / Oe	4.0	1.0
λ_s	1.6×10^{-6}	4.4×10^{-6}
H_k / Oe	20	20
ρ / $\mu\Omega\text{-cm}$	25	51

Fig.1 Relation between H_c and heat treatment temperature.

(a) CoNiFe film

(b) CoNiFeS film

50nm

Fig.2 TEM bright field images and TED patterns for as-plated (a) CoNiFe and (b) CoNiFeS films.

(a) CoNiFe film

(b) CoNiFeS film

50nm

Fig.3 TEM bright field images and TED patterns for 350°C annealed CoNiFe(a) and CoNiFeS(b) films.

Next, permeability at the high frequency region was investigated. First, the H_k change with heat treatment temperature was studied. The H_k value was calculated from the M-H loops of easy and hard axis directions. All the samples were annealed with applying 500Oe magnetic field to the hard axis direction for 3hr at 200, 230 and 250°C. Table 3 indicates the values of H_k of as-plated and after annealed films. The H_k values do not change at 200°C annealing. After 230°C annealing, the direction of hard axis was reversed to the easy axis direction and the H_k values became 14Oe and 5Oe for the CoNiFeS and CoNiFe films, respectively. After 250°C annealing, the H_k values reach to 20Oe for the CoNiFeS, 14Oe for the CoNiFe films. The result shows that the H_k can be controlled by the method of annealing with applying magnetic field. Prior to the study of high frequency permeability, the permeability value at 10MHz was measured by 8-figure coil method for hard axis direction. The results are also indicated in Table 3. Each film has almost equal to that of calculated value from the equation of $\mu = B_s/H_k$, which is based on the assumption as the uniaxial anisotropy model. Next, high frequency dependence of the permeability, up to 700MHz, was measured using the films having the same $H_k=14$ Oe value. It was measured by using microstrip pickup coil. The results are shown in Fig.5. In comparison with these two films, the relative permeability(μ') are approximately 1000-1100 up to

(a) CoNiFe film

(b) CoNiFeS film

50nm

Fig.4 TEM bright field images and TED patterns for 400°C annealed CoNiFe(a) and CoNiFeS(b).

30MHz. Above 30MHz, their values decrease slightly and then decrease drastically around 100MHz. In addition, the value of μ'' for the CoNiFeS film is smaller than that of the CoNiFe film. Therefore, the frequency of crossing of μ' and μ'' curves for the CoNiFeS film is shifted to high frequency region approximately 150MHz. It is suggested that the film with high ρ achieves higher permeability in high frequency region due to the small eddy current loss. In the same figure, the calculated permeability based on the eddy current and ferromagnetic resonance[8] are also indicates as a solid and a dotted lines. By comparing with the values obtained by calculation and experiments, the measured permeability(μ') in low frequency is the same as calculated one, while, the large decreasing over 30MHz are observed in the measured one. In the calculated permeability in Fig.4, the dispersion of magnetic anisotropy is not taken into account. In addition, it is reported that granular structured highly resistive films have the large decrease of permeability in high frequency region could come from the microstructure of the films[9]. Therefore, future investigation on mechanism of the decrease in permeability of the electroplated films in high frequency region is necessary in order to obtain high permeability.

Table 3 H_k and μ' (at 10MHz) values of the CoNiFe and CoNiFeS films in as-plated and annealed conditions

	CoNiFe		CoNiFeS	
	H_k	μ'	H_k	μ'
as-plated	20	600	20	650
200°C	20	600	20	650
230°C	5	1300	14	1050
250°C	14	1000	20	700

Fig.5 Relative permeability of CoNiFeS(a) and CoNiFe(b) films.

IV. SUMMARY

The permeability at the high frequency for electroplated CoNiFeS film was studied by comparing with electroplated CoNiFe film having a different resistivity value. Each film has a thermal stability up to 300°C on magnetic properties, and H_k value can be controlled by annealing with applying magnetic field. The CoNiFeS film with $H_k=14\text{Oe}$ shows higher permeability as compared with that of CoNiFe film, especially, in high frequency region. Higher permeability of the CoNiFeS film could be attributed to the decrease of the eddy current loss based on twice larger resistivity, compared with the CoNiFe film. Furthermore, it was found that the large difference in permeability of the CoNiFeS film between the measured value and the calculated one in high frequency region. It is necessary that mechanism of decrease in permeability of the film in high frequency region should be investigated.

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