High Permeability Soft Magnetic Multilayer Films with Fe-Ni Alloy for GHz Range

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Frequency characteristics of complex permeability (µ-f properties) for the soft magnetic FeNi multilayer films were investigated. The Ni-Fe/Ni film was composed of an Ni-Fe plating layer around 0.2μ m-thick, an Ni layer made by evaporation and a PET base film of 13 µm thick. The films were stacked in the form of multilayer with adhesive, and the multilayer films showed excellent μ -f properties as well as the single layer film. The complex permeability of films were affected by the thickness ratio of Ni-Fe layer to Ni under-layer. The films with higher thickness ratio over 6 showed higher permeability and resonance frequency in GHz area. And also heat treatment at 110°C improved complex permeability in GHz area. It was considered that the stress generated by shrinking of PET film with the heat treatment introduced the higher magnetic anisotropy into the magnetostrictive Ni-Fe layer. The large permeability of 260 at 1.8GHz and resonance frequency of 2.2GHz were obtained. The small tan δ of 0.28 and the permeability of 89.5 were also obtained at 1.8GHz. The latter showed the high resonance frequency over 3GHz. The dependence of Fe content(40 to 65wt%) on μ -f properties of the Ni-Fe layer was also investigated. The films with Fe content ranging from 40 to 60wt% showed the similar μ -f profile and showed resonance characteristics showed by the heat treatment at 110° C. The films with the thinner Ni-Fe layer showed higher resonance frequency, and this tendency was weakened by the heat treatment.

Key words: electroplating, anisotropy field, FMR frequency, annealing, soft magnetic film

1. INTRODUCTION

Flexible soft magnetic sheets with high permeability and low loss in the range up to 2GHz have been demanded as the electromagnetic reflector or the electromagnetic noise absorber in mobile communication devices[1-2]. The large anisotropy field H_k and saturation magnetization $4\pi M_s$ are important for increasing ferromagnetic resonance (FMR) frequency. Recently the soft magnetic film with large H_k of 550 Oe and low FMR loss was reported [3].

In this study, Fe-Ni alloys were electroplated on the Ni under-layer evaporated on the polyethylene terephthalate (PET) film. The PET film is a good candidate for flexible substrate having high elastic modulus of around 4500 MPa and can be used to easily control the stress in the magnetic layers in the sheets by annealing, which leads higher anisotropy field, H_{k} , and FMR frequency. Frequency dependences of the complex permeability were investigated on the thickness ratio of Fe-Ni plated layer to evaporated Ni under-layer. Dependence of the thickness ratio and annealing condition on the high frequency permeability are also studied.

2. EXPERIMENTS

Iron-nickel alloy films with the thickness of 0.2 to 0.4 μ m-thick which contain 60wt% Fe were made by electroplating on the evaporated Ni under-layer in Watts bath with current density of 0.8A/dm² at 30°C. Ni under-layers of 0.019 to 0.078 μ m thick were prepared by evaporation on the PET film of 13.1 μ m thick with winding speed of 6.0 m/min., tension of 34 MPa and incident beam angle of 30 degree to the PET surface.

After electroplating the Fe-Ni alloys, the magnetic sheets were annealed at the temperature above the glass transition temperature of PET film, $69 \,^{\circ}\text{C}$. In the annealing process, the magnetic sheets were annealed by two different methods. One was annealing in the press machine (MP-SNH made by Toyo-Seiki co. Ltd) for 60 seconds, after inserting the magnetic sheets between a pair of stainless steel plates of 2 mm thick preheated to the annealing temperature. The other was annealing by inserting into the twin roller heated up the annealing temperature with the roller surface speed of 0.23 m/min.

Multilayers were prepared by stacking the soft magnetic sheets after spreading epoxy adhesive diluted with xylene on the surface of the sheets.

Alloy composition and layer thickness were measured



Fig. 1 Frequency dependence of the permeability on the thickness of Fe-Ni layer.

by using the X-ray fluorescence analysis. Magnetic properties were measured using a vibrating sample magnetometer (VSM) and the complex permeability to 3GHz was measured using a thin film permeameter PMF-3000 (made by Ryowa electron co. Ltd). The structure was investigated by X-ray diffraction (XRD).

3. RESULTS AND DISCUSSION

3.1 Structure and Configuration

Since most of the loss in the high frequency area originated from eddy current and FMR losses, the thickness of the Fe-Ni alloy layer was designed less than 0.4 μ m for suppressing eddy current loss. The resistivity ρ of 60%Fe-Ni film was measured by four-terminal- method around $40 \times 10^{-8} \Omega m$. Supposing that the permeability, μ , of the magnetic film is 200, skin depth δ is calculated to 0.5 μ m at 2GHz by using the equation,

$$\delta = \sqrt{\frac{2\rho}{\omega\mu}}$$

where ω is angular frequency. The thickness dependence on the frequency permeability characteristics is shown in Fig.1, where thickness of 60%Fe-Ni layer, d, varies from 0.2 to 1.1 µm. Every sample has Ni under-layer of $0.02 \,\mu\text{m}$ -thick and was annealed at 85°C for 60 seconds. In case of the thickness $d = 1.1 \mu m$, fairly large permeability of 1200 was obtained at 10 MHz and its attenuation starts around 60MHz. In case of $d = 0.6 \mu m$, permeability showed 600 at 10 MHz and its attenuation starts at around 400 MHz. In comparison with the calculated frequencies from skin depth, 70MHz assuming μ '=1200 with skin depth of 1.1 μ m, and 160MHz assuming μ '=600 with skin depth of 0.6 um, it is considered that the attenuation in permeability for thick samples of 0.6 µm less than was dominated by eddy current. On the other hand, frequency permeability characteristics of the samples of 0.2 µm or less exhibit big change around 1GHz. It is assumed that FMR loss is dominant for the sample of 0.2 µm or less.

The relationship between the Fe content of Fe-Ni alloy



Fig.2 The relation between μ -f profile and Fe content in Fe-Ni layer.



Fig. 3 XRD patterns of Fe-Ni platings.

and the frequency permeability characteristics at room temperature is shown in Fig.2. All the μ -f profiles of Fe-Ni alloys containing 44wt%, 52wt%, and 60wt% Fe showed the similar profiles but slightly lower attenuation frequency of the 60wt% Fe-Ni alloy.

Figure 3 shows the effect of layer configuration on the XRD patterns of the Fe-Ni plating on the Ni plating, Fe-Ni plating on Ni layer evaporated in vacuum, and Fe-Ni plating without under-layer. XRD peaks of the film with Ni plating especially assigned as (220) and (311) were observed broader than that with evaporated Ni under-layer. It seems that the Ni as a under-layer induce larger isotropic stress in the Fe-Ni plating than the Ni under-layer evaporated in vacuum, which will decrease the anisotropy field. And the frequency permeability characteristics of the Fe-Ni layer on Ni layer evaporated in vacuum shifted the attenuation frequency higher than the film on Ni plating. Thus, the Ni layer evaporated in vacuum was preferred.



Fig. 4. TEM image of cross section of the film.



Fig. 5 The dependence of frequency characteristics on thickness ratio: (a) thickness ratio 5.4, (b) thickness ratio 9.8, (c) tan δ vs. thickness ratio.

Figure 4 shows the TEM image of a cross section of the film and the distribution of the Fe content in the Fe-Ni layer from the surface of the Ni layer. Fe content was observed maximum around the interface between Fe-Ni layer and Ni layer, which leads strong magnetic coupling of magnetic layers around the interface.

Effect of thickness ratio, the thickness of Fe-Ni plating divided by the thickness of Ni layer, on the frequency permeability characteristics is shown in Fig. 5. Frequency independent μ^{2} profile up to 2GHz and small tanð were observed for the films with the thickness ratio larger than 6. Furthermore, the frequency permeability characteristics also depend on the under layer thickness. The coercivity of 0.019 µm-thick Ni layer was 41 Oe, and that of 0.037 µm thick Ni layer was 76 Oe. This indicates that the frequency permeability characteristics can be effected by the magnetic properties of Ni under layer.

3.2 Heat treatment of the soft magnetic film

Figure 6 shows the effect of annealing temperature of 44%Fe-Ni film of 0.15 μ m thick on the frequency permeability characteristics. Both of the attenuation frequency of χ and the peak frequency of μ " increased with the increase of annealing temperature. Figure 7 shows the relationship between the shrinkage of the PET film by heating and the permeability of μ ' at 10MHz. The $\mu'(\parallel)$ and $\mu'(\perp)$ mean the permeability when magnetic field is parallel or perpendicular to the longitudinal direction of the PET film, respectively.

The PET film dose not shrink below the temperature of glass transition point, Tg. Anisotropic characteristics of the permeability, large in width direction and small in longitudinal direction, were observed at the temperature below Tg of the PET film. The PET film started to shrink at around the Tg and, at this temperature, anisotropic characteristics of permeability was disappeared. And a larger permeability was observed in the longitudinal direction of the PET film than in width direction at the annealing temperature over the Tg. This behavior indicates the stress due to shrinkage of the PET film induced by the heat-treatment will strongly influences the magnetic characteristics of the soft magnetic film.



Fig.6 The frequency dependence of μ -f profiles on the heat treatment temperature.



Fig.7 The relationship between the permeability and heat treatment temperature and shrink rate of PET.

Figure 8 shows that the dependency of the attenuation frequency of μ ' and the peak frequency of μ '' on a square root of H_k for the film annealed as mentioned above. The increase of the H_k leads the increase of the attenuation frequency of μ ', peak frequency of μ '', and FMR frequency, according to the relation,

$$\omega_0^2 = \left(\gamma^2 / \mu_0 \right) H_k M$$

where ω_0 is FMR frequency, γ is magnetomechanical ratio and μ_0 is permeability in vacuum. In this case, the eddy current loss is ignored owing to sufficiently thin thickness of the films. And FMR loss in the range up to 2GHz is considered also small for the films having large H_k , which leads much higher FMR frequency over 2GHz.

Figure 9 shows the relationship between the frequency characteristics of the single layer and triple layer and their thickness of single layer for the 60% Fe-Ni platings on Ni layer evaporated in vacuum on PET film having the thickness of 0.2, 0.4, and 0.8 μ m. Each sample was annealed at 110 °C after stacking three layers by epoxy adhesive coating. The peak frequency of μ " and the μ ' at 2.0 GHz for triple layer are almost the same as single one. It was found that the frequency characteristics were not changed by stacking.



Fig.8 The relationship between permeability and square root $H_{k\!\cdot\!}$



Fig.9 The characteristics of the permeability of single and triple film.



Fig.10 The dependence of μ -f profiles on the heat treatment.

The permeability of the extremely low loss films at high frequency is shown in Fig. 10. By annealing the film at 150°C using twin roller, a small tan δ of 0.15 and μ ' of 74 at 1GHz, tan δ of 0.28 and μ ' of 89.5 were obtained.

4.CONCLUSION

Soft magnetic films consisted of Fe-Ni platings, Ni layers evaporated in vacuum, and PET films were fabricated. Frequency characteristics for the soft magnetic film were investigated to control the permeability in high frequency up to 3GHz.

1) The frequency dependence of the permeability of films was effected by the thickness ratio of Fe-Ni layer to Ni under layer. The films with higher ratio over 6 showed higher permeability and resonance frequency of μ -f properties in GHz range.

2) The frequency characteristic of the films at high frequency range was able to be easily controlled by utilizing the stress owing to shrinkage of the PET film generated by heat-treatment. A small tan δ of 0.28 and permeability of 89.5 were obtained at 1.8GHz by heating of the film at 150°C.

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REFERENCES

[1] K.Suezawa, Y. Takahashi, M. Yamaguchi, K.I. Arai, Y. Shimada, W.D.Li, S. Tanabe and K.Ito: *J. Magn. Soc. Jpn.*,23, 1649-1652(1999)

[2] W.D.Li, K. Kato, O. Kitakami, and Y.Shimada: J. Magn. Soc. Jpn., 22, 449-452(1998)

[3] M. Munakata, M. Namikawa, M. Motoyama, M. Yagi, Y. Shimada, M. Yamaguchi and K.I. Arai, *Trans. Magn. Soc. Jpn*, 2, 388-393 (2002)

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