MAGNETIC PROPERTIES OF Fe-Ni-M (M=Si,Ce,Ta) THIN FILMS

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Abstracts--- Fe-Ni alloy films with nearly 40wt% Fe have a large saturation magnetic flux density as large as 1.48T. However, their magnetostriction constant and magnetocrystalline anisotropy have large absolute values. In this study, in order to improve the soft magnetic properties of 40wt%Fe-Ni alloy thin films, the third elements, Si and Ta which were soluble in Fe-Ni alloy and Ce which was not soluble in it, were added to Fe-Ni alloy. Experimental results show it is necessary to add the elements which are soluble in Fe-Ni and also have smaller atomic radius than that of Fe and Ni for realizing high effective permeability in these films. In case of Si addition, high effective permeability about 2700 at 1MHz is obtained.

INTRODUCTION

Much attention has been paid for Fe based soft magnetic alloy thin films with high saturation magnetization. Recently it has been reported that Fe-Ta-N-O[1], Fe-Co-Nb[2] thin films show fairly well soft magnetic properties.

However, relatively large value of magnetocrystalline anisotropy and magnetostriction of Fe degrade soft magnetic properties of Fe based alloy thin films. According to the micromagnetic ripple theory by Hoffmann[3], for obtaining high effective permeability μ_e , it is necessary to decrease local anisotropy fluctuation by decreasing grain size[4][5][6].

In the present study of Fe-N alloy films, the reduction of total magnetic anisotropy energy in each grain through the magnetostrictive anisotropy due to residual lattice strain were found to have a strong relation to the appearance of high μ_e [7].

In this study, in order to improve the soft magnetic properties of 40wt%Fe-Ni alloy films, (40wt%Fe-Ni)-M alloy films were examined. For additive elements, Si and Ta which were

soluble in Fe-Ni alloy, and Ce which was not soluble in it were selected. In case of Si, reduction of lattice spacing causes tensile stress and in case of Ta, expansion of lattice spacing causes compressive stress. Based on these results, the relations between μ_e and grain size, preferred orientation and magnetostriction anisotropy due to residual lattice strain are systematically discussed.

EXPERIMENTAL PROCEDURE

The films were prepared by RF sputtering method. The sputtering was carried out under the condition listed in Table 1.

Table 1. Sputtering condition

Ar gas pressure	9×10^{-3} Torr
Substrate temperature	320 °C
Target-substrate distance	75mm
Input power	1kW
Film thickness	3 μ m

The deposition chamber was evacuated to less than 7×10^{-7} Torr. Specimen films were deposited on crystallized glass substrates. The target used for preparing the (40wt%Fe-Ni)-M (M=Si,Ce,Ta) thin films was a composite target consisting a 40wt%Fe-Ni alloy and additive element pellets.

Specimen films were annealed at 400-600°C for 1 hour without magnetic field in a vacuum of less than 5×10^{-6} Torr.

Coercive force H_c and saturation magnetic flux density B_s were measured by vibrating sample magnetometer (VSM) and magnetostriction constant λ was determined by measuring the displacement of cantilever sample. The value of μ_e was determined by the inductive method using ferrite core.

Crystalline structure was characterized by using X-ray (Cu-K α) diffraction analysis and scanning electron microscopy (SEM).

RESULTS AND DISCUSSION

Magnetic Properties

Fig. 1 shows the dependence of the effective permeability μ_e at 1 MHz on the M content for Fe-Ni-M alloy thin films after annealing at 600°C. μ_e of Fe-Ni-Si thin film is larger than that of Fe-Ni thin film at the Si content of 1wt%~3wt%. And μ_e is sensitive to the content and the peak μ_e occurs around Si=2wt%. The maximum of μ_e is approximately 2700 at Si=2wt%. And as the Si content increases still more, μ_e decreases abruptly. On the other hand, μ_e of Fe-Ni-Ta thin film shows the broad maximum value of about 1200 at the optimum Ta content and is a little larger than that of Fe-Ni thin film in this region. μ_e of Fe-Ni-Ce thin film is decreases gradually as temperature increases.

Fig. 2 shows the annealing temperature dependence of the effective permeability μ_e at 1 MHz for Fe-Ni and Fe-Ni-M thin films. In this figure, μ_e is plotted at optimum content of Fe-Ni-M thin films. Maximum μ_e of Fe-Ni-Si thin film occurs at 600°C, and the temperature is higher than that of Fe-Ni thin film. However, maximum μ_e of Fe-Ni-Ce thin film

Fig. 1 Dependence of the effective permeability μ_e on the M content for Fe-Ni-M alloy thin films

Fig. 2 Temperature dependence of the effective permeability μ_e for Fe-Ni-M and Fe-Ni alloy thin films at optimum content; Fe-Ni-2wt%Si, Fe-Ni-5wt%Ta, Fe-Ni-0.5wt%Ce and Fe-Ni

occurs at the same temperature of Fe-Ni thin film. And, in contrast, μ_e of Fe-Ni-Ta thin film decreases monotonously as temperature increases in this region.

Fig. 3 Dependence of the magnetostriction constant λ on the M content for Fe-Ni-M alloy thin films

Fig. 3 shows the dependence of the magnetostriction constant on the M content for Fe-Ni-M thin films. Fe-Ni thin film has large magnetostriction constant λ , 1.8×10^{-5} , and λ of Fe-Ni-M thin films decrease with increasing M content. Especially λ of Fe-Ni-Si thin film is down to 1×10^{-5} . Optimum contents of μ_s disagree with optimum content of λ , so it seems that μ_s should be influenced by not only magnetostriction but also crystal structure[8] and that is clarified in next step.

Crystal Structure

Fig. 4 shows the X-ray diffraction patterns for Fe-Ni and Fe-Ni-M thin films after annealing at 600°C. The diffraction lines corresponding to (111), (200) and (220) planes of fcc Fe-Ni thin film are detected. Especially (111) peak is very strong, so this means that the (111) plane of this thin film is oriented parallel with the substrate. The diffraction pattern for Fe-Ni-Si thin film is similar to that for Fe-Ni thin film and shows fcc (111) preferred orientation. But addition of Ta in Fe-Ni thin film makes the peak intensity weaken and it shows random preferred orient-

Fig. 4 X-ray diffraction patterns for Fe-Ni and Fe-Ni-M alloy thin films

ation. The diffraction lines of Fe-Ni-Ce thin film corresponding to these planes of Fe-Ni thin film are detected and the lines corresponding to (101), (002), (110) and (103) planes of Ce are detected slightly. And the preferred orientation changes from (111) plane to (220) plane in Fe-Ni-0.5wt%Ce thin film.

Fig. 5 shows the changes in cross-sectional SEM micrograph of Fe-Ni and Fe-Ni-M thin films. SEM micrograph of Fe-Ni, Fe-Ni-2wt%Si

Fig. 5 Changes in SEM cross-section of Fe-Ni-M alloy films; a) Fe-Ni b) Fe-Ni-2wt%Si c) Fe-Ni-0.5wt%Ce d) Fe-Ni-5wt%Ta

and Fe-Ni-0.5wt%Ce thin films show highly-oriented structures. On the other hand, the oriented structure is vanished and grain size changes smaller in the Fe-Ni-5wt%Ta thin film.

Fig. 6 shows the dependence of the lattice constant on the M content for Fe-Ni and Fe-Ni-M thin films. Lattice constant of Fe-Ni-Si

Fig. 6 Dependence of the Lattice Constant a on the M content for Fe-Ni-M alloy thin films

Fig. 7 Temperature dependence of the average grain size D for Fe-Ni, Fe-Ni-2wt%Si, Fe-Ni-5wt%Ta and Fe-Ni-0.5wt%Ce alloy thin films

thin film decreases and that of Fe-Ni-Ta thin film increases continuously with increasing M content due to the different size of atomic radius and it shows that Si and Ta are soluble in Fe-Ni. On the other hand, lattice constant of Fe-Ni-Ce thin film increases up to 3wt% content. And then lattice constant is fixed about 3.593Å. This result agrees with the fact that diffraction lines of Ce are detected over 3wt% in X-ray diffraction.

Fig. 7 shows the temperature dependence of the average grain size which is evaluated from main peak in X-ray diffraction patterns of Fe-Ni and Fe-Ni-M thin films. The average grain size of Fe-Ni-M thin film is smaller than that of Fe-Ni thin film. And the grain of Fe-Ni thin film grows up over 500°C, while the grains of Fe-Ni-M thin film remain in the microcrystallized state at 600°C. μ^* of Fe-Ni-Ta thin film is smaller than that of Fe-Ni-Si thin film after annealing at 600°C, however grain size of Fe-Ni-Ta thin film is the smallest at 600°C. These results suggest that μ^* depends on not only grain size but also another origin.

Fig. 8 shows the temperature dependence of

Fig. 8 Temperature dependence of the diffraction intensity ratio $I_{(111)} / I_{\Sigma}$ for Fe-Ni and Fe-Ni-M alloy thin films Here, $I_{\Sigma} = I_{(111)} + I_{(200)} + I_{(220)}$

the (111) diffraction intensity ratio $I_{(111)}/I_{\Sigma}$ for Fe-Ni and Fe-Ni-M alloy thin films. $I_{(111)}/I_{\Sigma}$ of Fe-Ni-Si thin film increases slightly as temperature increases, but $I_{(111)}/I_{\Sigma}$ of Fe-Ni-Ta and Fe-Ni-Ce thin films decrease at more than 500°C. Especially $I_{(111)}/I_{\Sigma}$ of Fe-Ni-Ce thin film at 600°C is about one third as small as the value at 500°C because of changing to (220) orientation. In case of Fe-Ni-Si thin film, $I_{(111)}/I_{\Sigma}$ is as large as that of Fe-Ni thin film, but μ_e of Fe-Ni-Si thin film is superior to Fe-Ni thin film. This means that μ_e depends on not only preferred orientation but also another origin.

The origin of soft magnetism

According to the micro magnetic ripple theory by Hoffmann, the soft magnetic properties are dependent on the structure constant S , corresponding to short period local anisotropy fluctuation[3]. And it's known that as S decreases, permeability increases nearly proportional to $1/S^2$ [7]. So S should be decreased to realize high permeability.

And S is given by

$$S = K_s \cdot \sigma \cdot D \cdot n^{-1/2}$$

Here, K_s , σ , D , n are the magneto-crystalline anisotropy, the crystal orientation intensity, grain size and the number of grains, respectively. So permeability is determined by K_s , σ and D .

Fig.9 shows the change of μ_e after annealing at optimum temperature of Fe-Ni-M thin films versus the (111) diffraction intensity ratio and grain size. In the figure, equi-value zone of grain size experimentally obtained are shown. As seen in this figure, in Fe-Ni-M thin films, the value of μ_e increases gradually with increasing $I_{(111)}/I_{\Sigma}$. And the value of μ_e increases with decreasing grain size on equal $I_{(111)}/I_{\Sigma}$. The experimental results suggest that the value of μ_e more than 3500 in 40wt%Fe-Ni based alloy films is expected under the condition of grain size less than 250Å and also (111) diffraction intensity ratio more than 0.9.

Fig. 9 Average grain size D versus the diffraction intensity ratio $I_{(111)}/I_{\Sigma}$ and μ_e for Fe-Ni and Fe-Ni-M alloy thin films

Fig. 10 μ_e versus $\{I_{(111)}/I_{\Sigma}\}/D$ and the ratio of the lattice spacing of the (111) crystal plane $\Delta d_{(111)}/d_{(111)}$ for Fe-Ni and Fe-Ni-M alloy thin films

As the next step, in order to clarify the relation between μ_e and magnetic anisotropy caused by lattice strain, the ratio of lattice spacing of the (111) plane $\Delta d_{(111)}/d_{(111)}$ caused by additive elements is examined.

Fig.10 shows the relation between the μ_e

and the ratio of the lattice spacing of the (111) plane and the ratio $\{I_{(111)}/I_z\}/D$ which shows effect of grain size and the (111) diffraction intensity ratio on μ_e . From this figure, it is understood that the high μ_e is realized by enlarging $\{I_{(111)}/I_z\}/D$ and reducing the lattice spacing of the (111) plane. Realizing the high μ_e by reducing the lattice spacing shows that the magnetic anisotropy is decreased by internal stress due to residual lattice strain[10].

Consequently, in case of 40wt%Fe-Ni based alloy thin films with large magnetostriction constant, it is necessary to reduce the grain size and enlarge the (111) preferred orientation and reduce the magnetic anisotropy simultaneously. Based on the results, it is considered that additive elements which have smaller atomic radius than that of Fe and Ni, and also are soluble in Fe-Ni alloy are effective for realizing higher μ_e .

CONCLUSION

The soft magnetic properties and especially the origin of high effective permeability are examined in case of 40wt%Fe-Ni based alloy films with large magnetostriction constant.

- (1) High effective permeability is determined by microcrystallized state and (111) highly-oriented structure, and it is necessary to realize not only one term but also both of them.
- (2) It is effective to add the elements which have smaller atomic radius than that of Fe and Ni and also are soluble in Fe-Ni alloy.
- (3) In case of 37wt%Fe-61wt%Ni-2wt%Si with magnetostriction constant 14×10^{-6} , the maximum value of effective permeability about 2700 and the value of saturation magnetic flux density about 1.3T are obtained.
- (4) The experimental results show that high effective permeability more than 3500 in 40

wt%Fe-Ni based alloy thin films is expected under the condition of grain size less than 250Å and (111) highly-oriented structure and reduction of lattice spacing of the (111) plane.

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REFERENCES

- [1] Y.Watanabe, H.Oura and N.Onozato, IEEE Trans. Magn., Vol.26, No.5, pp.1500-1502, 1990.
- [2] F.Sato and T.Miyazaki, J. Magn. Soc. Jpn., Vol.15, pp.307-310, 1991.
- [3] H.Hoffmann, Phys. Stat. Sol., Vol.33, pp.175, 1969.
- [4] M.Takahashi, T.Sato, T.Miyazaki, H.Uwazumi, M.Takahashi and T.Wakiyama, J. Magn. Soc. Jpn., Vol.13, No.S1, pp.743-748, 1989.
- [5] T.Shimatsu, M.Takahashi and T.Wakiyama, J. Magn. Soc. Jpn., Vol.13, No.S1, pp.577-583, 1989.
- [6] G.Herzer, IEEE Trans. Magn., Vol.25, No.5, pp.3327-3329, 1989.
- [7] M.Takahashi and T.Shimatsu, IEEE Trans. Magn., Vol.26, No.5, pp.1485-1490, 1990.
- [8] O.Shimizu, K.Nakamichi and S.Yoshida, J. Magn. Soc. Jpn., Vol.14, pp.293-295, 1990.
- [9] N.Hasegawa, S.Sato, A.Kojima, A.Makino, Y.Kenzaki and T.Watanabe, J. Magn. Soc. Jpn., Vol.14, pp.319-322, 1990.
- [10] K.Sato, T.Imai, K.Kondo and T.Mizoguchi, J. Magn. Soc. Jpn., Vol.13, pp.273-276, 1989.