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## SOFT MAGNETISM OF Co-Zr AND Fe-Co FILMS WITH LARGE SATURATION MAGNETIZATION

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### ABSTRACT

Large saturation magnetization  $4\pi M_s$  is essentially required for soft magnetic thin layers used in magnetic recording devices. Amorphous Co-based alloys and Fe-Co alloys may be regarded as one of the candidates for soft magnetic materials which possess large  $4\pi M_s$ . Some preparation process to improve soft magnetism of these films were performed in this study. Addition of Ta seemed to be effective to change the magnetostriction constant  $\lambda$  from positive value to negative one. The magnetoelastic energy  $K_e$  is strongly dependent on  $\lambda$ .  $(\text{Co}_{95.7}\text{Zr}_{4.3})_{100-x}\text{Ta}_x$  films with  $K_e$  of negative value have sufficiently soft magnetic characteristics.  $\text{Fe}_{90}\text{Co}_{10}$  alloy exhibits extremely large  $4\pi M_s$  of about 24 kG. Addition of N and Ta to  $\text{Fe}_{90}\text{Co}_{10}$  films improved the soft magnetism of them.

The  $\text{Fe}_{82.0}\text{Co}_{7.6}\text{Ta}_{10.4}:\text{N/Ti}$  multilayered films exhibit better soft magnetic properties and better thermal stability than Fe-Co-Ta:N singlelayer films.

### INTRODUCTION

Magnetic thin films with soft magnetic properties are used for core layers in magnetic recording heads and the backlayers in doublelayered perpendicular magnetic recording media.<sup>[1]</sup> Since soft magnetic films have to pass sufficient magnetic flux, the soft magnetic films with large saturation magnetization are desired. Amorphous Co-based alloys and Fe-Co alloys may be regarded as one of the candidates for soft magnetic materials which possess large  $4\pi M_s$ . Some preparation processes to improve soft magnetism of these films were performed in this study. By changing Ta content  $\chi$  of  $(\text{Co}_{95.7}\text{Zr}_{4.3})_{100-x}\text{Ta}_x$  films, the relationship among  $\chi$ , magneto-

striction constant  $\lambda$  and coercivity  $H_c$  were clearly observed. The magnetostriction constant  $\lambda$  of Co-based alloy films should be minimized to attain soft magnetism. Fe-Co alloy has the largest saturation magnetization  $4\pi M_s$  of all the magnetic alloys. The addition of N and Ta to  $\text{Fe}_{90}\text{Co}_{10}$  sputtered thin films prepared by facing targets sputtering apparatus were effective to improve soft magnetic properties.<sup>[2]</sup> These soft magnetism seemed to be caused by the fine granulation in the films. The as-deposited films of  $\text{Fe}_{82.0}\text{Co}_{7.6}\text{Ta}_{10.4}:\text{N}$  possessed relatively large  $4\pi M_s$  and high relative permeability  $m_r$  of 17kG and 400, respectively. Moreover,  $\text{Fe}_{82.0}\text{Co}_{7.6}\text{Ta}_{10.4}:\text{N/Ti}$  multilayered films possessed not only high permeability  $\mu_r$  without annealing process but also excellent thermal stability.

## EXPERIMENTAL PROCEDURE

All the specimen films were prepared by the facing targets sputtering (FTS) apparatus as shown Fig.1, where two targets were located face-to-face. Permanent magnets were installed behind the targets to confine the plasma in the space between the targets. This FTS apparatus has four kinds of target materials and can deposit thin layers of them continuously. The substrates were polyethylene naphthalate tape or silicon. They were positioned on a can roll, which was located out of the plasma. Therefore the substrates were almost free from plasma. The substrate temperature  $T_s$  was kept at room temperature (R.T.). Kr was used in place of Ar as

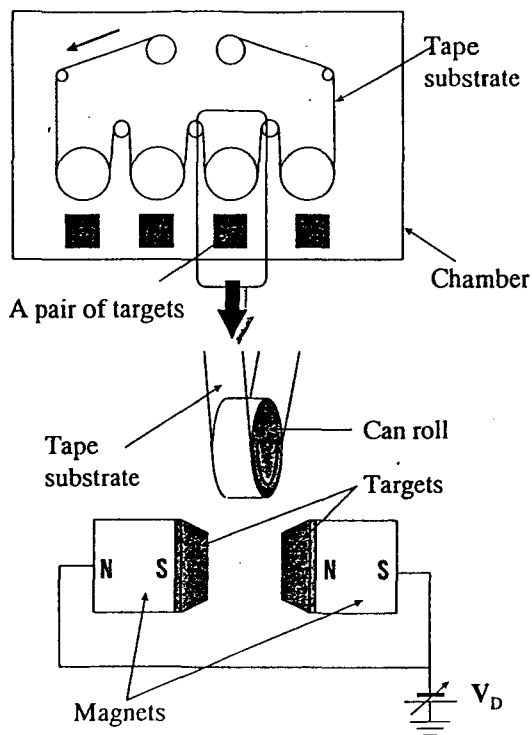


Fig. 1 Schematic figure of facing targets sputtering (FTS) apparatus

sputtering gas in depositing  $(\text{Co}_{95.7}\text{Zr}_{4.3})_{100-x}\text{Ta}_x$  and Ti films to minimize damage of films by recoiled particles.  $\text{Fe}_{82.0}\text{Co}_{7.6}\text{Ta}_{10.4} : \text{N}$  films were deposited by reactive sputtering method using mixture gas of  $\text{N}_2$  and Kr. Total pressure of the mixture gas was set at 1 mTorr.  $\text{N}_2$  partial pressure  $P_{\text{N}_2}$  was adjusted to control the N content from 0.02 mTorr to 0.34 mTorr.

Crystallographic characteristics of the films were analyzed by X-ray diffractometry (XRD), and magnetic properties were measured by vibrating sample magnetometer (VSM). Internal stress  $s$  was evaluated from the degree in curling of the sheet substrates with the deposited films. Films composition was analyzed by inductively coupled plasma (ICP) analysis. Relative permeability of magnetic films were measured by shunt core method at 1MHz. Magnetostriction constant  $l$  of  $(\text{Co}_{95.7}\text{Zr}_{4.3})_{100-x}\text{Ta}_x$  films were evaluated by the method which will be described below.

### Measurement of magnetostriction constant $\lambda$ of $(\text{Co}_{95.7}\text{Zr}_{4.3})_{100-x}\text{Ta}_x$ films

Since the magnetostriction constant  $\lambda$  of Co-Zr alloy thin films are so small below  $10^{-6}$ , it is difficult to measure  $l$  of the films with small thickness around  $0.1\mu\text{m}$ . Schematic configuration of the measurement system used in this study were shown in Fig. 2. Two pairs of coils were installed to measure M-H loop. Outer coils were used to apply AC magnetic field  $H$  to  $(\text{Co}_{95.7}\text{Zr}_{4.3})_{100-x}\text{Ta}_x$  films, and inner coils were used to detect magnetization  $M$  of  $(\text{Co}_{95.7}\text{Zr}_{4.3})_{100-x}\text{Ta}_x$  films. When the tensile stress  $F$  was applied to the films, the anisotropic tensile stress was induced in the soft magnetic films. By applying the tensile stress  $F$  to the films, anisotropy field  $H_k$  in the films

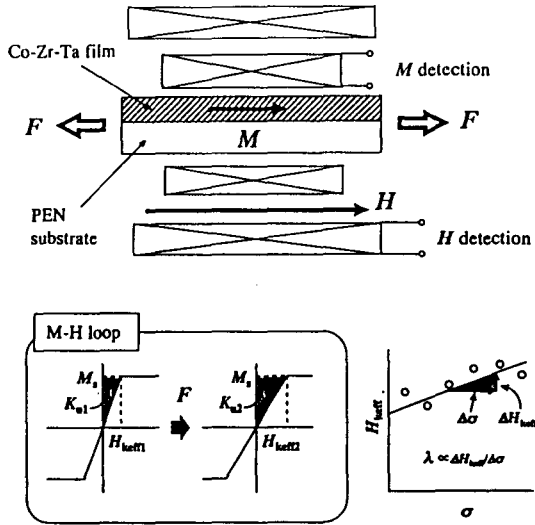


Fig. 2 Schematic configuration of the system to measure magnetization constant  $\lambda$

changed due to the reverse magnetostrictive effect. The change of anisotropic field  $\Delta H_{\text{keff}}$  by applying anisotropic stress  $\sigma$  to the films is

$$\Delta H_{\text{keff}} = 2\Delta K_u' / M_s \tag{1}$$

where  $\Delta K_u'$  and  $M_s$  are the change of anisotropy energy  $K_u$  and saturation magnetization, respectively. Anisotropy energy  $K_u$  induced due to reverse magnetostrictive effect caused by the applied anisotropic stress  $s$  is

$$K_u^* = (-3/2)\lambda\sigma \tag{2}$$

$\Delta K_u^*$ , the change of anisotropy energy  $K_u$  to anisotropic stress  $\Delta\sigma$ , is

$$\Delta K_u^* = (-3/2)\lambda\Delta\sigma \tag{3}$$

Assuming that the change of anisotropic field is induced due to reverse magnetostrictive effect,  $\Delta K_u' = \Delta K_u^*$ . Then 1 is calculated as follows;

$$\lambda = (-1/3)M_s\Delta H_{\text{keff}}/\Delta\sigma \tag{4}$$

where  $\Delta H_{\text{keff}}/\Delta\sigma$  is determined by the gradient of  $H_{\text{keff}}$  vs.  $\sigma$  characteristic curve.

## RESULT AND DISCUSSION

### Co-Zr-Ta films

Fig. 3 shows the dependence of magnetostriction constant  $\lambda$  on the Ta contents  $\chi$  for the  $(\text{Co}_{96.7}\text{Zr}_{4.3})_{100-x}\text{Ta}_x$  films with various thickness and prepared at various  $P_{\text{Kr}}$ .  $\lambda$  decreased monotonously from positive value to negative one as  $x$  increased.  $\lambda$  seems to take the value of nearly zero at  $\chi$  around 6 at.%. Fig. 4 shows the change of coercivity  $H_c$  as a function of magne-toelastic energy  $K_e$ . Since  $D_s$  depended weakly on  $x$  and were in the range from  $-2$  to  $-6 \times 10^8$  (dyne/cm<sup>2</sup>) for films deposited at  $P_{\text{Kr}}$  of 1mTorr,  $K_e$  seemed to be strongly dependent on  $\lambda$ . It was confirmed that lower  $H_c$  was observed at  $K_e$  of negative value.

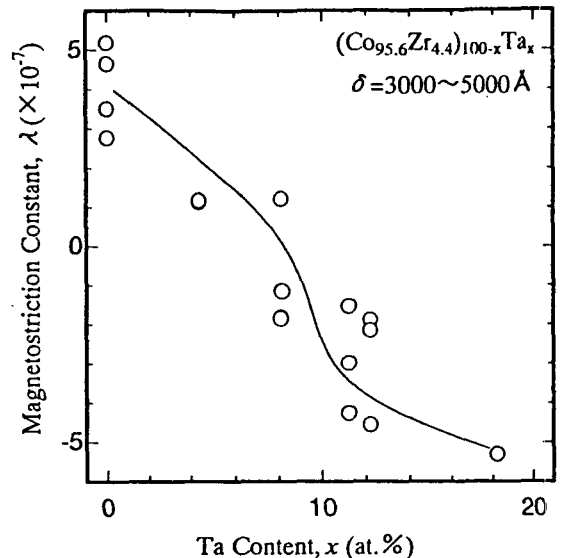


Fig. 3. Dependence of  $\lambda$  of  $(\text{Co}_{96.7}\text{Zr}_{4.3})_{100-x}\text{Ta}_x$  films on Ta content  $\chi$ .

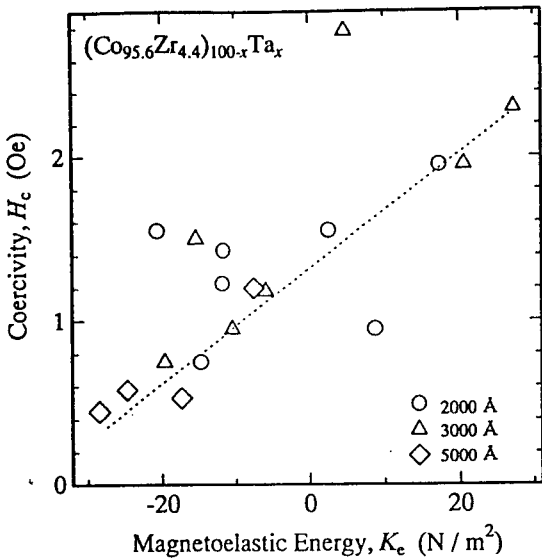


Fig. 4 Change of  $H_c$  as a function of  $K_e$

#### Fe-Co-Ta:N films

Fe-Co alloy has the largest saturation magnetization  $4\pi M_s$  among all the magnetic alloys.

The addition of N and Ta to  $Fe_{90}Co_{10}$  reduced the grain size  $\langle D \rangle$ . These soft magnetism seemed to be caused by the fine granulation in the films. Fig. 5 shows the dependence of saturation magnetization  $4\pi M_s$  and relative permeability  $\mu_r$  of  $Fe_{82.0}Co_{7.6}Ta_{10.4}:N$  films on  $P_{N_2}$ .  $Fe_{82.0}Co_{7.6}Ta_{10.4}:N$  films prepared of  $P_{N_2}$  at 0.06 mTorr exhibited relatively large  $4\pi M_s$  and high relative permeability  $\mu_r$  of 17kG and 400, respectively.

$Fe_{82.0}Co_{7.6}Ta_{10.4}:N$  films thicker than 2000, however, did not have soft magnetic properties. So it was attempted to improve their soft magnetism by depositing  $Fe_{82.0}Co_{7.6}Ta_{10.4}:N$  layers thinner than 1000 Å. In this study,  $Fe_{82.0}Co_{7.5}Ta_{10.4}:N$  singlelayer film (specimen A) and  $Fe_{82.0}Co_{7.6}Ta_{10.4}:N/Ti$  multilayered films (specimens B and C) were prepared as shown in Fig. 6. Total thickness of magnetic

layers in these specimens were 2000 Å.  $P_{Kr}$  and  $P_{N_2}$  in depositing  $Fe_{82.0}Co_{7.6}Ta_{10.4}:N$  layers are 0.84mTorr and 0.16mTorr, respectively.  $P_{Kr}$  in depositing Ti layers was set at 1mTorr. These films were annealed for 2 hours in vacuum at temperature  $T_A$  of 150, 220°C and 300°C.

Fig. 7 shows the dependence of relative permeability  $\mu_r$  of  $Fe_{82.0}Co_{7.6}Ta_{10.4}$  singlelayer film and  $Fe_{82.0}Co_{7.6}Ta_{10.4}:N/Ti$  multilayered films on annealing temperature  $T_A$ . Specimen A exhibited low relative permeability  $\mu_r$  of about 50 in as-deposited state. After annealing at high  $T_A$  of 220°C,  $\mu_r$  of it could be higher than 400°C.

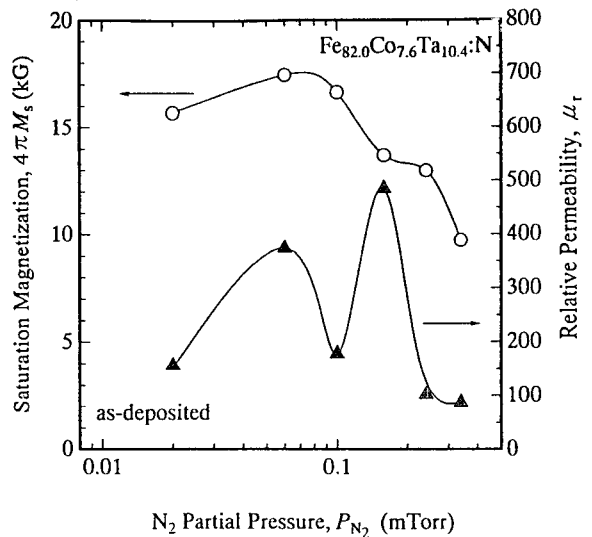


Fig. 5  $P_{N_2}$  dependence of  $4\pi M_s$  and  $\mu_r$  of Fe-Co-Ta : N films

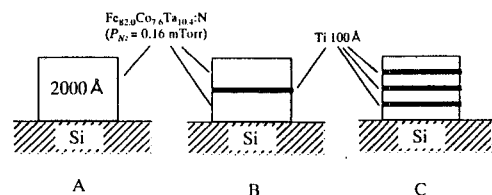


Fig. 6 Schematic configuration of  $Fe_{82.0}Co_{7.6}Ta_{10.4}:N$  singlelayer film and  $Fe_{82.0}Co_{7.6}Ta_{10.4}:N/Ti$  multilayered films.

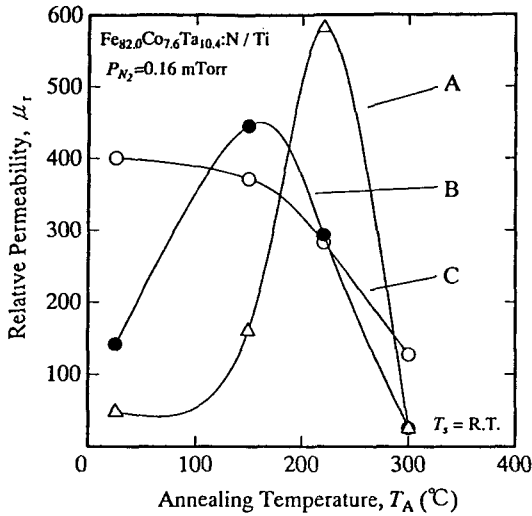


Fig. 7.  $T_A$  dependence of  $\mu_r$  of  $\text{Fe}_{82.0}\text{Co}_{7.6}\text{Ta}_{10.4}:\text{N}$  singlelayer film and  $\text{Fe}_{82.0}\text{Co}_{7.6}\text{Ta}_{10.4}:\text{N}/\text{Ti}$  multilayered films.

Specimen C exhibited high  $\mu_r$  of about 400 in as-deposited state. Deposition of  $\text{Fe}_{82.0}\text{Co}_{7.6}\text{Ta}_{10.4}:\text{N}/\text{Ti}$  multilayered films seems to be effective to improve soft magnetism. And the  $\text{Fe}_{82.0}\text{Co}_{7.6}\text{Ta}_{10.4}:\text{N}/\text{Ti}$  multilayered film divided into four magnetic layers by Ti layers maintained  $\mu_r$  of about 150 after annealing at 300°C though  $\mu_r$  of the other specimens were less than 30.

Fig. 8 shows the dependence of  $4\pi M_{s(\text{annealed})}/4\pi M_{s(\text{as-deposited})}$  of  $\text{Fe}_{82.0}\text{Co}_{7.6}\text{Ta}_{10.4}:\text{N}$  singlelayer film and  $\text{Fe}_{82.0}\text{Co}_{7.6}\text{Ta}_{10.4}:\text{N}/\text{Ti}$  multilayered films on annealing temperature  $T_A$ . As  $T_A$  became high,  $4\pi M_s$  gradually decreased.  $4\pi M_s$  of singlelayer film annealed at 300°C were about 40% of that of as-deposited film. But  $4\pi M_s$  of  $\text{Fe}_{82.0}\text{Co}_{7.6}\text{Ta}_{10.4}:\text{N}/\text{Ti}$  multilayered films was not so small as that of singlelayer film annealed at 300°C. It seemed to be due to that ferromagnetic  $\epsilon\text{-Fe}_2\text{N}$  and  $\gamma\text{-Fe}_4\text{N}$  crystallites were formed in multilayered films by annealed at 300°C and that nonma-

gnetic  $\zeta\text{-Fe}_2\text{N}$  crystallite were formed mainly in singlelayer film since N atoms in magnetic layers of multilayered films were diffused into Ti layers. Deposition of  $\text{Fe}_{82.0}\text{Co}_{7.6}\text{Ta}_{10.4}:\text{N}/\text{Ti}$  multilayered films seems to be effective to prevent  $4\pi M_s$  from decreasing by annealing at high  $T_A$ .

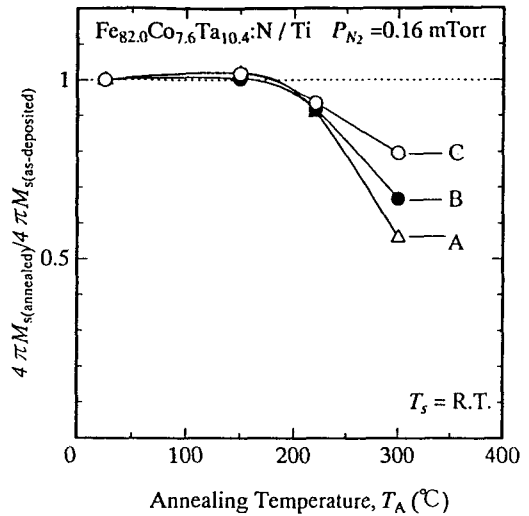


Fig. 8.  $T_A$  dependence of  $4\pi M_s$  of  $\text{Fe}_{82.0}\text{Co}_{7.6}\text{Ta}_{10.4}:\text{N}$  singlelayer film and  $\text{Fe}_{82.0}\text{Co}_{7.6}\text{Ta}_{10.4}:\text{N}/\text{Ti}$  multilayered films

## CONCLUSION

Ta was added to  $\text{Co}_{96.7}\text{Zr}_{4.3}$  to minimize magnetostriction constant  $\lambda$ .  $\lambda$  of  $(\text{Co}_{96.7}\text{Zr}_{4.3})_{100-x}\text{Ta}_x$  films decreased monotonously from positive value to negative one as  $x$  increased.  $\lambda$  seems to take the value of nearly zero at  $x$  around 6 at.%.  $K_e$  seemed to be strongly dependent on  $\lambda$ . It was confirmed that  $H_c$  was lower at  $K_e$  of negative value. Soft magnetic properties of Fe-Co alloy films were improved by adding both Ta and N to  $\text{Fe}_{90}\text{Co}_{10}$  films. These soft magnetism of Fe-Co alloy films seemed to be caused by the fine granu-

lation in the films.  $\text{Fe}_{82.0}\text{Co}_{7.6}\text{Ta}_{10.4} : \text{N}$  films possessed relatively large  $4\pi M_s$  and high relative permeability  $\mu_r$  of 17kG and 400, respectively.

Deposition of  $\text{Fe}_{82.0}\text{Co}_{7.6}\text{Ta}_{10.4} : \text{N/Ti}$  multilayered films seems to be effective to improve soft magnetism and to prevent  $4\pi M_s$  from decreasing by annealing at high  $T_A$ .

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