SOFT MAGNETISM AND MICRO-STRUCTURE OF IRON FILMS
BY ION BEAM SPUTTERING

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Abstract --- Soft magnetic iron films have been prepared under various deposition conditions by using dual ion beam sputtering (DIBS) apparatus, and the relation between their magnetic properties and micro-structure have been investigated in detail.

In this study, the both accelerating voltages of sputtering ions [V_{mg}] for the target and bombarding those [V_{sg}] for the growing films surface were varied individually. Argon, hydrogen and nitrogen ions were used for bombarding the films. As a result, the minimum H_c below 5 Oe was obtained in the films bombarded by mixed ions of argon and hydrogen, or argon and nitrogen accelerated at proper energy.

It has been found that the moderate mobility of sputtered iron atoms on film surface and the proper incorporation of hydrogen or nitrogen into interstices of iron lattice are very essential for decrease of H_c resulting from an improvement of microscopic uniformity and atomic ordering in the films.

INTRODUCTION

It is well known that the bulk iron with purity above 99.99% shows excellent soft magnetic properties. Especially, its saturation magnetization $4\pi M_s$ is 21.5 kG, being largest among all elementary substances. Recently, the preparation of films with such a high $4\pi M_s$ has become of major interest for the magnetic layer of thin film heads or the soft magnetic under layer in perpendicular recording medium.[1] However, the films deposited by sputtering from iron targets do not show always the magnetic properties same as bulk iron.[2,3] Especially, the coercivity H_c of films is apt to be affected remarkably by preparation conditions. Therefore, the preparation techniques must have the excellent controllability of deposition parameters, in order to attain the best soft magnetic properties. In this regard, the DIBS method is very useful and effective technique because of independent control of the ions when they sputter the target and bombard the film surface.

In this study, the effects of ion bombardment during deposition on soft magnetism of iron films have been examined. Then, hydrogen and nitrogen were

selected for bombarding ions besides argon. These elements can enter into the lattice of iron crystallites, and therefore, it is very interesting to bombard their ions for the growing surface layer of iron.

EXPERIMENTAL PROCEDURE

Figure 1 shows the schematic diagram of a dual ion beam sputtering apparatus used in this study. Both of the ion sources are Kaufman type. The lower one sputters the target. The upper one is the bombarding ion source.

The base pressure in chamber was 2×10^{-7} Torr. As a sputtering gas, pure argon was introduced through the sputtering ion source up to about 0.1 mTorr. The acceleration voltage of sputtering ions (V_{mg}) was set at 500V or 1000V. The total current of ions sputtering the target was fixed at about 4mA.

For bombarding ion source, the acceleration voltage (V_{sg}) was varied in the range of 100 to 500V. Pure argon or either mixture of argon and hydrogen ($Ar+H_2$) or argon and nitrogen ($Ar+N_2$) was introduced to 0.18 mTorr for the

Fig. 1 Schematic diagram of the double ion beam sputtering apparatus.

gases of bombarding ions. The ratio in percentage of hydrogen or nitrogen partial pressure to total pressure of each gas mixture will be represented by RH_2 and RN_2 , respectively. RH_2 was set at 50%, and RN_2 was varied from 10 to 100%.

The specimen iron films were deposited on water-cooled glass slides by sputtering a iron target of 99.99% purity. Film morphology was observed by means of scanning electron microscopy (SEM) and crystal structure was analyzed by X-ray diffractometry. Saturation magnetization, $4\pi M_s$, and coercivity, H_c , were measured by a vibrating sample magnetometer.

RESULTS AND DISCUSSION

Effects of low-energy argon ion assist

Figure 2 shows cross sectional SEM micrographs of iron films deposited under different conditions. The total argon

pressure in chamber, P_{Ar} , was varied up to 1.6 mTorr by adjusting a gas inlet valve. Film thickness of these films was in the region of 600 - 1000 nm. Specimen films of a), b) and c) in figure were not ion-bombarded.

All specimen films had the very smooth surface. The cross sections of films deposited at P_{Ar} below 0.3 mTorr did not show an obvious texture at both V_{mg} of 500 and 1000V. Especially, ones at V_{mg} of 1000V showed a dense and columnless structure. However, as P_{Ar} increased above 1 mTorr, the cross sectional morphology of the films at V_{mg} of 500V changed to a columnar structure as seen in Fig. 2, b). A similar structure was observed in the films at V_{mg} of 1000V. On the other hand, the films bombarded by argon ions at V_{sg} of 100V and a current density of $20 \mu A/cm^2$ did not show such texture even at P_{Ar} of 1.6 mTorr seen in Fig. 2, c).

Fig. 2, Cross sectional SEM micrographs of films deposited under different conditions.

In the case of high P_{Ar} , the arrival energy of sputtered atoms decrease through collision process to residual gas atoms, and the mobility of them on film surface is reduced. As a result, the film structure could become columnar. Then, the mean free pass λ of sputtered iron particles is a important factor to estimate the range of such a P_{Ar} . From the result of the calculation, it has been found that the λ become smaller than the distance between the target and the substrate, 8cm, at P_{Ar} above 0.8 mTorr. Therefore, the columnar structure seems to appear in the films deposited at P_{Ar} above 0.8 mTorr.

On the other hand, the bombardment of argon ions with proper energy during deposition may increase the mobility of iron atoms on film surface and eliminate columnar microstructure.

Figure 3 shows the dependence of $4\pi M_s$ and H_c of these iron films with thickness of 600-1000nm on P_{Ar} . H_c tended to increase with P_{Ar} . H_c of the films at V_{mg} of 500V abruptly changed at P_{Ar} above 0.5 mTorr, while that of the films at V_{mg} of 1000V gradually increased to more than 20 Oe at P_{Ar} above 1 mTorr.

From the results of the internal stress of these films, films deposited at V_{mg} of 1000V showed larger compressive stress than those of 500V. This may be due to the additional and irregular bombardment by argon atoms recoiled from target plane. And also, such a bombardment of recoiled argon could increase the value of H_c due to the increase of the stress irregularity in films.

$4\pi M_s$ of these films decreased from about 20 kG with increase of P_{Ar} above 1 mTorr at both values of V_{mg} . The small $4\pi M_s$ of films with columnar structure may be due to the the low packing density of iron atoms.

On the other hand, when the surface layer of growing films were bombarded by argon ions accelerated at 100V, $4\pi M_s$ did not change with P_{Ar} in the range of 0.3 -1.6 mTorr, and H_c decreased to a low value below 8 Oe.

It has been found that the bombardment by argon ions with proper energy improves the uniformity of growth orientation of the films and makes the

Fig. 3, The dependence of $4\pi M_s$ and H_c on argon gas pressure, P_{Ar} , and the effect of ion bombardment on soft magnetic properties.

microstructure dense from the view-point of morphology. As a result, the soft magnetism of iron films could maintain at the wide P_{Ar} range.

Effects of hydrogen or nitrogen ion bombardment

Figure 4 shows the dependence of $4\pi M_s$ and H_c of films bombarded by argon and hydrogen ions on V_{sg} . Then, the acceleration voltage of sputtering ions, V_{mg} , and film thickness, δt , were set at 500V and 300±50 nm, respectively. For the δt , H_c showed constant values of about 10 Oe in the region of 600 - 1000 nm as seen in Fig. 3. But, as δt decrease below 600 nm, H_c decreased gently with δt . H_c of iron films with δt of about 300 nm took

about 7 Oe. In addition, the magnetic properties of these films changed significantly with V_{sg} and current density, I_d , of argon and hydrogen ions bombarding the film surface.

$4\pi M_s$ took the maximum value of about 21 kG at I_d of $20 \mu A/cm^2$, while the films bombarded by only argon ions at V_{sg} above 100V and the films bombarded by argon and hydrogen ions at I_d of 10 and $100 \mu A/cm^2$ showed $4\pi M_s$ below 20 kG.

H_c of the films bombarded by only argon ions was about 7 Oe and did not depend apparently on V_{sg} . On the other hand, when RH_2 and I_d were 50% and $100 \mu A/cm^2$, respectively, H_c increased monotonically with V_{sg} . However, when I_d was 20 and $10 \mu A/cm^2$ at RH_2 of 50%, H_c took the minimum value below 5 Oe at V_{sg} of 100V and 300V, respectively.

Each of films having different H_c showed also different X-ray diffraction diagrams as shown in Fig. 5. H_c looked to be in proportion to intensity of diffraction peak from (110) plane of α -Fe. The films with the lower H_c showed the X-ray diffraction diagrams with the broader and smaller peaks.

The iron thin films with minimum H_c of below 5 Oe and maximum $4\pi M_s$ of 21 kG have been prepared at I_d of $20 \mu A/cm^2$ and V_{sg} of 100V when RH_2 was 50%.

On the other hand, nitrogen is one of well known active elements for iron.[4] Figure 6 shows the dependence of the interplanar spacing d of the iron-nitride crystallites on V_{sg} and RN_2 , where value of d was calculated from the position of the highest peak in X-ray diffraction diagrams. All the iron-nitride films deposited on the substrates at room temperature showed only one diffraction peak, and the strongest diffraction intensity was obtained in the films at V_{sg} of 150 - 300V.

The nitrogen content in films, it was estimated by X-ray micro-analysis(XMA), increased monotonically to about 21 at.% with increase of RN_2 . For RN_2 of 100 and 50%, the orthorhombic phase ζ - Fe_2N and the hexagonal phase Fe_3N , respectively, were synthesized in the films. And these films had semi-hard magnetism. Namely, $4\pi M_s$ of these films took the minimum values 1 kG and 10 kG at RN_2 of 100 and 50%,

Fig. 4, The dependence of $4\pi M_s$ and H_c on accelerating voltage V_{sg} for films bombarded by argon and hydrogen ions.

Fig. 5, X-ray diffraction diagrams of films bombarded by argon and hydrogen ions.

respectively, at V_{sg} of 150V. Also, the H_c of these higher nitriding films increased drastically with V_{sg} in the region of 0 - 150V. These results can be attributed to the well-crystallization of ζ - Fe_2N and Fe_3N grains, because ζ - Fe_2N is paramagnetic at room temperature and Fe_3N has much smaller $4\pi M_s$ than α -Fe.

For RN_2 of 20 and 10%, the small amount of nitrogen atoms may enter into the interstices among iron atoms, expanding and distorting the lattice of iron crystallites, as shown in Fig. 6.

For the films bombarded by argon and hydrogen ions, the change of d was less pronounced. This may be due to the difference of atomic size between hydrogen and nitrogen.

Soft magnetism of these films with low nitrogen content was improved by ion bombardment at V_{sg} of 150 - 250V. Figure 7 shows the dependence of $4\pi M_s$ and H_c of films deposited at RN_2 of 10%. $4\pi M_s$ and H_c took a maximum value of above 22kG and a minimum value below 5 Oe, respectively, at V_{sg} of 250V.

The post annealing for these films did not significantly change $4\pi M_s$ which was about 21kG, but it noticeably decreased H_c which took a minimum as low as 1.5 Oe at V_{sg} of 250V as shown in Fig. 7. Then, the expansion of d spacing was removed by annealing. The decrease of d spacing may indicate significant reduction of internal stress of films. Therefore, the decrease of H_c may be attributed to the growth of Fe crystallites with low nitrogen content and the stress reduction in films by annealing at 300°C.

These films with best soft magnetism bombarded by argon plus hydrogen or nitrogen ions showed also a uniaxial magnetic anisotropy in plane. The direction of easy axis corresponded to the incident direction of ion beam from the bombarding ion source.

These results suggest that the bombardment of proper amount of hydrogen or nitrogen ions with moderate energy to the growing film surface improve the microscopic uniformity and atomic ordering in the films, and so, enhance the soft magnetic properties of iron.

Fig. 6, The dependence of interplanar spacing d on V_{sg} .

Fig. 7, The dependence of $4\pi M_s$ and H_c on V_{sg} for films bombarded by argon and nitrogen ions at RN_2 of 10%.

CONCLUSION

Iron thin films have been deposited by means of the double ion beam sputtering apparatus with iron target of 99.99% purity. The effects of bombardment of argon, hydrogen and nitrogen ions to the surface layer of growing film on their crystal structure and magnetic properties has been investigated. The summary is as follows;

- 1) The films deposited at lower P_{Ar} revealed dense and columnless structure, which changed to columnar structure at P_{Ar} above 1 mTorr with a decrease in arrival energy of sputtered iron atoms.
- 2) The films bombarded by argon ions with proper energy during deposition revealed dense structure even at P_{Ar} of 1.6 mTorr, and had H_c of 8 Oe and $4\pi M_s$ above 20 kG.
- 3) The films bombarded by argon and hydrogen ions with proper kinetic energy took $4\pi M_s$ of 21 kG and H_c below 5 Oe at V_{sg} of 100V and I_d of 20 $\mu A/cm^2$.
- 4) As for the films bombarded by argon and nitrogen ions, the semi-hard compounds of $\zeta-Fe_2N$ and Fe_3N were synthesized in the films with higher nitrogen content. On the other hand, for lower nitrogen content, the films exhibited large $4\pi M_s$ above 22 kG and low H_c of 5 Oe by adjusting the nitrogen percentage to 10% and the accelerating voltage to about 250V.
- 5) It has been found that the bombardment of hydrogen or nitrogen ions is very useful technique for enhancement of soft magnetism of iron films, giving rise to an improvement of microscopic uniformity and atomic ordering.

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