Control of Microstructure of Co-Cr Thin Films by Adjusting Sputtering Condition

Masahiko NAOE

Department of Physical Electronics, Tokyo Institute of Technology
2-12-1 O-okayama, Meguro, Tokyo 152, JAPAN

Abstract- The Co-Cr films deposited at low Ar pressure, low substrate temperature and moderate biasing voltage on the plasma-free substrates by using the facing targets sputtering apparatus revealed almost perfectly homogeneous microstructure in the surface and interior of the films and were composed of the fine grains with excellent crystallinity. It was found that the addition of third element such as Ta and Re, the exchange of Ar for Kr as sputtering gas and the preparation of the Co-Cr/C, Al multilayers were significantly effective to make the Co-Cr grains finer and more uniform in terms of size and shape.

I. INTRODUCTION

The Co-Cr thin films have been regarded as one of the most hopeful candidates for the recording layer in the perpendicular magnetic recording media with ultra-high density[1-2]. The strict control of the microstructure of the Co-Cr films as well as the well c-axis orientation of the Co-Cr crystallites are necessary to clear its high potential of recording density and to attain the sufficient perpendicular magnetic anisotropy.

The facing targets sputtering (FTS) system[3] can strictly control their microstructure by adjusting the sputtering conditions such as the Ar pressure PAr, the substrate temperature Ts[5] and the substrate biasing voltage Vb, since this system can deposit the ferromagnetic alloy thin films on the plasma-free substrates over a wide range of PAr, Ts and Vb under the state of very stable glow discharge.

The microstructure of the Co-Cr films with different composition and thickness deposited by the FTS system at various PAr, Ts and Vb will be presented in this paper. Furthermore, it will be shown that the addition of the third element such as Ta, Mo, V and Re[7], the exchange of Ar for Kr as sputtering gas[5,8] and the preparation of the Co-Cr/C, Al multilayer[9] should be significantly effective to make the Co-Cr grains finer and more uniform in terms to size and shape.

II. EXPERIMENTAL PROCEDURE

The specimen Co-Cr films were deposited on the substrates such as glass, Si, SiO2/Si, polyimide, PET and PEN by using the facing targets sputtering (FTS) apparatus. The substrate temperature Ts and the substrate biasing voltage Vb were adjusted in the range of 30 ~ 300 °C and 0 ~ -200 V, respectively. A pair of alloy plates with composition of Co83Cr17, Co81Cr19 and Co79Cr21 were used as the targets. Ar and Kr were used as the working gas of which the pressure was varied in the range of 0.1 ~ 30 mTorr.

The deposition rate R and the film thickness tF (or δ) were changed in the range of 100 ~ 5,000 Å/min. and 50Å ~ 2 μm, respectively. The microstructure such as the surface appearance and the cleaved cross-sectional morphology of the films were observed by the conventional SEM, the ultra high resolution SEM (UHR-SEM) with LaB₆ filament and the TEM. At the same time, the crystal structures of the same specimen films also were analyzed by X-ray and electron beam diffractometry and the mean size of crystallites <D> and the dispersion of c-axis orientation Δθ₅₀ were estimated to investigate the relationship between crystallinity and microstructure of the films.

III. RESULTS AND DISCUSSION

A. Argon Pressure PAr

The microstructure as well as the crystal structure of the Co-Cr films changed remarkably with the Ar pressure PAr. Figure 1 shows the typical SEM photographs observed in perspective of the films deposited at PAr of (a) 15.0, (b)4.0 and (c) 0.15 mTorr. Regarding the surface texture, the films at high PAr of 15 mTorr have the rough surface based on the very distinct columnar structure. Many small bumps are seen over the whole surface of the films at middle PAr of 4 mTorr, resulting in less uniform surface as compared with one of the films at low PAr of 0.15 mTorr. The more detailed observation on the various films by UHR-SEM also indicated that the surface texture changed apparently with PAr.

The morphology of the cleaved cross section was observed by UHR-SEM. It was found that the morphology also changed drastically with PAr. For example, the distinct columnar grains are observed in the films at higher PAr. Such morphology becomes less distinctive with decrease of PAr and finally disappear in the films at low PAr. The films at PAr as low as 0.15 mTorr are composed of the fine polyhedral micrograins packed
Fig. 1: SEM photographs observed in perspective of films deposited at various $P_{Ar}$.

densely from the initial layer to the surface layer of the films as seen in these SEM photographs.

**B. Substrate Biasing Voltage $V_b$**

Figure 2 shows the typical SEM photographs of the surface texture and the cross sectional morphology of the films deposited on the polyimide at the substrate biasing voltage $V_b$ of 0, -75, -100 and -200 V. The films at $V_b$ of -75 V had smaller grains and revealed more homogeneous microstructure than ones at $V_b$ of 0, -100 and -200 V. As $V_b$ increased, the large round grains were seen in the films at $V_b$ around -100 V and the clear columnar structure was observed for the films at $V_b$ in the range of -130 ~ -170 V. The films at $V_b$ of -200 V revealed the irregular columnar structure with the microcracks between the grains. Such $V_b$ dependences of the microstructure were the same for all kinds of substrates used in this study and for D.C. and R.F. voltage.

**C. Substrate Temperature $T_s$**

The microstructure of the films at high and middle $P_{Ar}$ changed drastically with the substrate temperature $T_s$. Figure 3 shows the SEM photographs of the cross section and the surface of the films deposited at $P_{Ar}$ of 1.2 and 0.1 mTorr and $T_s$ of room temperature, 120, 160 and 200 °C. Both of the films at 1.2 and 0.1 mTorr showed the same trend of increase in grain size for the surface texture with elevation of $T_s$. However, when $T_s$ was 120, 160 and 200 °C, the distinct columnar grains were observed in the SEM photographs of the cross section of the films at middle $P_{Ar}$ 1.2 mTorr, while they were not seen in the photographs at low $P_{Ar}$ of 0.1 mTorr. This difference of the cross section morphology between these films may be attributed to the difference in the mobility of the adatoms, that is, that in the kinetic energy of the atoms arriving on the substrate at various $P_{Ar}$, since the collision number of the sputtered atoms to Ar atoms before arrival onto the substrates may be different at various $P_{Ar}$.

**D. Deposition Rate $R_d$**

The surface texture became rougher with increase of the deposition rate $R_d$, as seen in Fig. 3. This may be based on the higher ratio in number of the arriving Co and Cr atoms to the bombarding Ar atoms to the bombarding Ar atoms. But it attributed to the elevation of $T_s$ due to the enhancement of heat radiation from the high energy plasma and the very hot target planes, since the higher input power was supplied.
Fig. 3: SEM photographs of cross section and surface of films deposited at various $T_s$.

(a) $R_d = 800$ Å/min.
(b) $R_d = 1900$ Å/min.

Fig. 4: SEM Photographs of surface of films deposited at $R_d$ of 800 and 1,900 Å/min.

for the sputtering apparatus to increase $R_d$.

E. Film Thickness $t_F$ (or $\delta$)

The size and shape of grains changed significantly with increase of the film thickness $t_F$ as seen in Fig. 5. This figure shows the SEM photographs of the surface texture for various $t_F$ of 210, 650, and 1,000 Å. The grain size $<D>$ increased monotonically with increase of $t_F$. For example, $<D>$ of the films with $t_F$ of 1,000 Å was about 200 Å, while $<D>$ of the films with $t_F$ of 210 Å was as small as 20 Å. These continuous increase of $<D>$ with $t_F$ may be attributed to the model for the continuous film growth in the sputter-deposition as illustrated in Fig. 5.

F. Film Composition $Co_{100-x}Cr_x$

The microstructure depended significantly on the film composition $x$ in $Co_{100-x}Cr_x$ system as seen in Fig. 6. This

Fig. 5: SEM photographs of surface of films with various thickness.

Fig. 6: TEM photographs of Co$_{79}$Cr$_{21}$ and Co$_{83}$Cr$_{17}$ ultra-thin films with various thickness.

- 481 -
The TEM photographs of Co<sub>x</sub>Cr<sub>21</sub> (x=21) and Co<sub>83</sub>Cr<sub>17</sub> (x=17) ultra-thin film with t<sub>f</sub> of 50, 100 and 200 A. The ultra-thin films with larger x of 21 revealed evidently better crystallinity than ones with smaller x of 17.

IV. CONTROL OF MICROSTRUCTURE

A. Addition of Third Element

V, Ta, Mo and Re as the third element M were added to the Co-Cr films, so that the film composition should be Co<sub>x</sub>Cr<sub>y</sub>M<sub>z</sub> (M: V, Ta, Mo and Re). As seen in Fig. 7, these additions of the third element were effective to decrease the grain size in order of Re, Mo, Ta and V.

B. Normal and Oblique Incidence

Figure 8 shows the TEM images of the ultra-thin films deposited at oblique and normal incidences, where the oblique incidence angle φ was 60°. The grain growth and the crystallite formation with good c-axis orientation were seen even in the films with t<sub>f</sub> as very small as 50A deposited at normal incidence, while they were not seen in ones deposited at oblique incidence.

Figure 9 shows the SEM photographs of cross section of the films deposited at φ of 60° for P<sub>Ar</sub> of (a) 15, (b) 2.0 and (c) 0.2 mTorr. A distinct columnar structure was seen

Fig. 7: SEM photographs of Co-Cr and Co-Cr-M films with M of V, Ta, Mo, and Re as third element.

Fig. 8: TEM images of ultra-thin films deposited at oblique (60°) and normal incidences.

Fig. 9: SEM photographs of films deposited at various P<sub>Ar</sub> for oblique incidence angle of 60°.
in the films at high $P_{Ar}$ of 15 mTorr, but it became very vague with decrease of $P_{Ar}$ and finally it disappeared at low $P_{Ar}$ below 0.5 mTorr.

C. Sputtering by Kr Ions

When Kr was used as working gas in place of Ar, the grain size was rather smaller in the films at the working gas pressure in the range of 0.1 ~ 5.0 mTorr, as seen in Fig. 10. This seemed to be due to that the kinetic energy of the Kr atoms recoiled from the target planes and bombarding the growing films is much lower than that of Ar atoms, since Kr and Ar atoms are heavier and lighter,

---

**Fig. 10.** SEM photographs of surface of films deposited at various $P_{Ar}$ when working gas is Ar and Kr.

**Fig. 11.** Illustration of grain growth process with increase of film thickness.

**Fig. 12.** Illustration of suppressing grain growth by alternately depositing ultra-thin Co-Cr and carbon layers.

**Fig. 13.** SEM photographs observed in perspective of Co-Cr single layer and Co-Cr/Al multilayer films.
respectively, than Co and Cr atoms.

D. Preparation of Multilayer

The process of grain growth in increase of the film thickness $t_F$ can be schematically illustrated as shown in Fig. 11. It was found that the ultra-thin films with $t_F$ in the insert of the range of $50 \sim 500$ Å were composed of the ultra-fine grains with the size $<D>$ almost equal to $t_F$. The Co-Cr/Al multilayers have been prepared in order to decrease $<D>$ in the recording layer as schematically illustrated in Fig. 12.

Figure 13 shows the surface appearance and the cross section morphology of the $100$ Å-Co$_{81}$Cr$_{19}$/7 Å-Al multilayers deposited at $P_{Ar}$ of 0.5 mTorr, compared with the Co$_{81}$Cr$_{19}$ single layer deposited at $P_{Ar}$ of 15 mTorr and 0.15 mTorr. The grain size in the multilayers was about 100 Å and almost equal to the thickness of Co-Cr layers. The grain size can be sufficiently controlled by adjusting the thickness of Co-Cr ultra-thin layer in the multilayers.

V. CONCLUSION

The Co-Cr films have been deposited by using the facing targets sputtering (FTS) apparatus in which the substrates are completely free from high-energy plasma. It has been confirmed that their microstructure has strong relationship with the sputtering conditions such as the Ar pressure $P_{Ar}$, the substrate temperature $T_s$, the substrate biasing voltage $V_b$, the film thickness $t_F$ and the target composition Co$_{100-x}$Cr$_x$.

The films deposited at lower $P_{Ar}$, lower $T_s$ and moderate $V_b$ revealed the perfectly homogeneous microstructure and were composed of very fine grains with excellent crystallinity. It has been found that the addition of third element such as Ta and Re, the normal incidence of the arriving atoms to the substrate plane, the exchange of Ar for Kr as the sputtering gas and the preparation of the Co-Cr/C, Al multilayer were significantly effective to make the grains finer and more uniform in terms of size and shape.

Consequently, the FTS system may be very useful to deposit the Co-Cr recording layers suitable for the perpendicular magnetic media with ultra-high density.

ACKNOWLEDGEMENTS

The author appreciate the agreement to cite the SEM and TEM photographs and the results from the doctoral thesis by Dr. Y. Hoshi of Tokyo Institute of Polytechnics, Dr. M. Matsuoka of NTT, Dr. A. Morisako of Shinshu Univ., Dr. M. Sagoi of Toshiba Corp., Dr. Y. Niimura of Sumitomo 3M, Dr. S. Kadokura of Teijin Ltd., Dr. S. Akiyama of Bridgestone Corp., Dr. T. Kawanabe of IBM Japan Ltd. and Dr. S. Nakagawa of Tokyo Institute of Technology.

REFERENCES