INFLUENCE OF EXTERNAL FIELD APPLIED DURING SPUTTERING ON THE MAGNETIC PROPERTIES OF CoCr FILMS

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This work investigates the influence of perpendicular magnetic field and negative substrate bias, applied during sputtering, on crystallographic structures and magnetic properties of CoCr films sputtered on polyimide substrates of two different temperatures; 27°C and above 150°C. The sputtering rate at the target increases with magnetic field $H_s$ and/or bias voltage $V_B$, while the deposition rate at the substrate decreases markedly with a bias above -100 V owing to the resputtering of Co and Cr adhered on the film surface, depending strongly on substrate temperature $T_s$. The film composition, thereby, changes depending on $T_s$, $H_s$ and $V_B$. The resputtering effect notably influences the film morphology and the magnetic properties. The films, having the good orientation of c-axis and thus having the high perpendicular anisotropy, can be obtained with perpendicular magnetic fields from 35 to 61 Oe.

INTRODUCTION

CoCr films are of interest for perpendicular magnetic recording [1] and have been investigated vigorously. Magnetic and crystallographic properties of CoCr sputtered films vary considerably depending on sputtering conditions: substrate temperature $T_s$[2], impurity gases in a sputtering chamber[3,4], magnetic field $H_s$ applied normal to substrates[5] and negative substrate bias voltage $V_B$[6-8]. The hcp c-axis orientation normal to the film plane is degraded by impurity gases, especially nitrogen or oxygen[4]. On the other hand, the orientation is improved by a magnetic field $H_s$[5] and by a moderate bias voltage $V_B$[7,8].

In a previous paper [7] we reported that the bias voltage $V_B$ gives only little change in Cr content, $C_{Cr}$, in the films sputtered on glass substrates. Recent experiments by Werner et al [9], however, showed that $C_{Cr}$ increases notably with $V_B$ higher than -100 V. Furthermore, Mapps et al[8] indicated that $C_{Cr}$ decreases with increasing $N_2$ partial pressure in a sputtering chamber under the negative bias of -100 V. These experiments suggest that the sputtering rate at the target and the resputtering rate at the film surface vary considerably depending on the sputtering conditions. For this reason, we examined the sputtering and resputtering rates as a function of $V_B$ with parameters of $H_s$ and $T_s$, and also studied the magnetic and crystallographic properties of CoCr films.

EXPERIMENTS

CoCr films were prepared on polyimide films of 50 x 50 mm area and 50 μm thick by diode RF sputtering. The target-to-substrate distance was fixed at 41 mm and the chamber was evacuated to lower than 1 x10$^{-6}$ Torr. Sputter deposition was carried out for 60 minutes with 100 W of RF power at 10 mTorr Ar pressure using high quality grade Ar (99.9995%). Here, the film thickness changed with $V_B$ from 1 μm at 0 V to about 0.5 μm at -200 V. The target was 100 mm dia cobalt, on which 9 pieces of Cr plates 10x10 mm square and several pieces of 5x5 mm Co plates were arranged uniformly to obtain 12 area% of Cr. Sputtering rates of Co and Cr, $S_{Co}$ and $S_{Cr}$, were evaluated from the change in weight per unit surface area of Co and Cr plates due to sputtering.

A half area of the polyimide sub-
strate was contacted thermally with the water-cooled Cu substrate holder through indium. Thus, the half area was held at a constant temperature of 27°C independent of perpendicular magnetic field \( H_n \) or of negative dc bias voltage \( V_B \). The other half area, however, was heated up above 150°C depending on \( H_n \) and \( V_B \) because of significant bombardment of electrons and ions or radiation from the hot target; the temperature changed from 150°C at \( H_n = 0 \) and \( V_B = 0 \) up to about 180°C at \( H_n = 20 \) Oe for \( V_B = 0 \), or at \( V_B = -150 \) V for \( H_n = 0 \). In this paper, we call the films deposited on the lower and higher temperature substrates the LT\(_n\)- and HT\(_n\)-films, respectively.

Magnetic field \( H_n \) was produced by a solenoid coil set under the target. The field intensity was uniform in the target area and decreased by a factor of 0.25 with going from the target to the substrate; for example it changed from 76 Oe at the target surface to 61 Oe at the substrate surface. Here, \( H_n \) is the value at the center of the substrate surface.

The film thickness was measured using Talystep method and SEM. The deposition rate, \( D \), was evaluated as the film thickness per unit sputtering time. The magnetic properties were studied with VSM. The perpendicular anisotropy field \( H_k \) was calculated using the relation \( H_k = 2K_u/M_s \), where \( M_s \) is the magnetization at 15 kOe and \( K_u \) the perpendicular anisotropy energy estimated from the in-plane and perpendicular hysteresis loops. The film composition was analysed by particle-induced X-ray emission (PIXE). The crystallographic structure was observed using X-ray diffractometer with Cu Ka radiation.

RESULTS AND DISCUSSION

Fig.1(a) shows that the low levels of \( H_n = 35 \) Oe and/or \( V_B = -50 \) V cause the abrupt increase in the net sputtering rate at the target, \( S = (S_{Co} \cdot A_{Co} + S_{Cr} \cdot A_{Cr}) / (A_{Co} + A_{Cr}) \), where \( A_{Co} \) and \( A_{Cr} \) are the surface area of Co and Cr targets, respectively, and also that the atomic ratio of sput-
tered Cr to total sputtered atoms, \( SR_{Cr} \), increases with \( H_s \) and \( V_B \). On the other hand, the deposition rate, \( D \), decreases steeply with \( V_B \) higher than \(-50 \text{ V}\) and thereby the ratio of \( D/S \) decreases monotonically with increasing \( V_B \) as shown in Fig.1(b). The decrease in \( D/S \) indicates that the atoms accumulated onto the substrate are resputtered owing to the bombardment of energetic \( \text{Ar}^+ \) ions accelerated by negative bias.

According to Cuomo et al [10], the fluxes of atoms leaving the Co and Cr targets, \( F_{Co} \) and \( F_{Cr} \), are given by

\[
F_i = \frac{1}{4} n^+ v_i^* \eta_i^{+} A_i, \quad (i = \text{Co, Cr}) \quad (1)
\]

where \( n^+ \) is the number density of \( \text{Ar}^+ \) ions in the plasma, \( v_i^* \) is their average random velocity at the target, and \( \eta_{Co}^{+} \) and \( \eta_{Cr}^{+} \) are the sputtering yield of Co and Cr at the target. Similar expressions are given for the resputtering rates, \( R_{Co} \) and \( R_{Cr} \), at the substrate,

\[
R_i = \frac{1}{4} n^+ v_s^* \eta_i^{-} A_i, \quad (i = \text{Co, Cr}) \quad (2)
\]

where \( \eta_i^{-} \) is the sputtering yield at the substrate at \( V_B \). The deposition rate, \( D \), then becomes

\[
D = F_{Co} R_{Co} + F_{Cr} R_{Cr} - R_{Co} - R_{Cr}
\]

\[
= \frac{1}{4} n^+ [v_s^* (F_{Co} \eta_{Co}^{+} A_{Co} + F_{Cr} \eta_{Cr}^{+} A_{Cr}) - v_s^* (\eta_{Co}^{-} + \eta_{Cr}^{-})],
\]

where \( F_{Co} \) and \( F_{Cr} \) are the fractions of the incoming fluxes of Co and Cr atoms which reach the substrate. The steep fall of \( D/S \) with \( V_B \) indicates that the resputtering rate \( (R_{Co} + R_{Cr}) \) increases drastically with \( V_B \), reaching a half value of \( (F_{Co} R_{Co} + F_{Cr} R_{Cr}) \) at \( V_B = -200 \text{ V} \).

When the magnetic field \( H_s \) is applied, the electrons in the plasma will occur the cyclotron motion and thereby frequently collide with Ar atoms causing the ionization of Ar, namely the increase of \( n^+ \). On the other hand, the negative bias causes \( \text{Ar}^+ \) ions to bombard the substrate emitting a large number of secondary electrons, which collide with Ar atoms and increase \( n^+ \), again. We can confirm the increase of the numbers of collisions between electrons and Ar atoms by observing the intensity of Ar luminescence in the plasma during sputtering. As shown in Fig.2, the intensity \( I_{Ar} \) increases with \( H_s \) and \( V_B \) on the empirical function:

\[
I_{Ar} = I_{Ar}^{0} (1 + m_1 H_s) (1 + m_2 V_B^{0.3}). \quad (4)
\]

This suggests that \( n^+ \) increases with \( H_s \) and \( V_B \) on a function similar to Eq.(4), which is consistent with the empirical relation given by Cuomo et al [10]. We can understand, thus, that the flux \( F \), namely the sputtering rate \( S \), increases with \( H_s \) and \( V_B \).

Fig.3 shows the \( V_B \)-dependence of the Cr contents in HTs- and LTs-films, \( C_{Cr}(\text{HTs}) \) and \( C_{Cr}(\text{LTs}) \). For \( V_B = 0 \) where the resputtering effect can be neglected, \( C_{Cr}(\text{HTs}) \) and \( C_{Cr}(\text{LTs}) \) show the same values, which decrease with \( H_s \) contrary to the \( H_s \)-dependence of \( SR_{Cr} \) in Fig.1(a). This discrepancy between the \( H_s \)-dependences of \( C_{Cr} \) and \( SR_{Cr} \) is mysterious at present. The decrease in \( C_{Cr} \) indicates that \( f_{Cr}/f_{Co} \) decreases with \( H_s \), plausibly relating to the ionization of Cr due to the collision with electrons of cyclotron motion. For \(-V_B > 50 \text{ V}\) where the resputtering effect is sensitive,

\[
\text{Fig.2 Intensity of Ar luminescence at } \lambda = 556 \text{ nm in plasma during sputtering.}
\]
The above $V_B$-dependence of $C_{Cr}$ predicts that the saturation magnetization $M_s$ increases with $V_B$ for the $LT_s$-films, while it decreases for the $HT_s$-films. However, $M_s$ of the $LT_s$-films decreases monotonically with $V_B$ for all the magnetic fields $H_s$, as shown in Fig.4(a), contrary to the prediction. Furthermore, the decrease in $M_s$ for the $HT_s$-films is steeper than the prediction as shown in Fig.4(b). The experimental values of $M_s$ are replotted in Fig.4(c) for $H_s = 0$ as a function of $Cr$ content by open (○) and closed circles (●) for the $LT_s$- and $HT_s$-films, respectively. Here, the dash-dotted line and the broken line
represent the values of $M_s$ for the LT$_n$- and HT$_n$-films at $V_B = 0$ and $H_s = 0$ [11]. When $V_B$ increases, the magnetization of both the LT$_n$- and HT$_n$-films approaches the values of bulk materials, which are indicated by the solid line [12]. Thus, $M_s$ decreases to approach the value of bulk material with negative bias for both the substrate temperatures $T_s$ and for all the magnetic fields $H_s$. As discussed by several authors, the decrease in $M_s$ arises from the homogenization of Cr distribution, which is induced by $V_B$ through several mechanisms: i) the break of Cr-Cr clusters due to the ion-bombardment [6], ii) the suppression of the growth of inter-columnar voids [8], iii) the decrease in the oxygen gettering effect related to the resputtering of oxygen [7, 13], and iv) the promotion in the surface diffusion of Cr due to the ion-assist effect [13].

The changes in the sputtering and resputtering rates due to $H_s$ and $V_B$ cause the notable variations in the crystallographic structure and the magnetic properties as well as $M_s$. Fig.5 shows the data for the HT$_n$-films: the grain size $D_{(002)}$, evaluated from the half width of (002) XRD peak, the hcp c-axis dispersion $\Delta \theta_{50}$, the in-plane remanence ratio $M_r ||/M_s$, the perpendicular coercivity $H_{c\perp}$ and the perpendicular anisotropy field $H_k$. Similar data were obtained for the LT$_n$-films, except that the perpendicular coercivity is smaller by one order than that of the HT$_n$-films (not shown here).

Fig.5(a) shows the considerable improvement in the c-axis orientation due to $H_s$ or a moderate bias around $-50$ V. From the previous reports on the impurity gas effects on the orientation [3,4], we suggest that the improvement of the orientation is related to the oxygen- and nitrogen-resputtering at the film surface, which is bombarded by electrons, ions, and/or sputtered atoms accelerated with

![Fig.5 Crystallographic structures (a), in-plane remanence ratio $M_r ||/M_s$ (b), perpendicular coercivity $H_{c\perp}$ (c) and perpendicular anisotropy field $H_k$ (d) for HT$_n$-films.](image-url)
the magnetic field \( H_s \) [5] as well as by lower energetic \( Ar^+ \) ions under the lower bias \( V_b \) [14,15]. The oxygen-release suppresses the Cr segregation, i.e. the gettering effect at the grain boundaries[3,4,16] and then promotes the grain growth, sometimes the growth of very large grains at around \(-50\) V[17]. The bias higher than \(-100\) V, however, prevents the growth of the grain with the preferential orientation by resputtering Co and Cr atoms adhered on the film surface. Thereby, the grain size becomes smaller and the crystal orientation becomes isotropic with increasing \( V_b \) as shown in Fig.5(a).

The above change in \( \Delta \theta_{50} \) affects the perpendicular magnetic anisotropy. The in-plane magnetic component decreases with the magnetic field or at \( V_b = -50\) V(Fig.5b), thus sharp shoulders being obtained in perpendicular hysteresis loops. However, the perpendicular coercivity \( H_{c,\perp} \) decreases fairly at \( V_b = -50\) V(Fig.5c). The reduction due to \( V_b \) arises from the uniform distribution of Cr, which results the wall motion in the flux reversal[11]. On the other hand, the variation is only little for the application of the magnetic field \( H_s \). Thus, we can obtain a good quality film having the higher values of \( H_{c,\perp} \) and \( M_s \) for the perpendicular magnetic recording by applying the magnetic field \( 35 \sim 61\) Oe rather than the negative bias.

Further increase in the negative bias degrades the c-axis orientation, resulting the in-plane magnetization: the large value of \( M_s/\mu_0 \) (Fig.5b) and the negative anisotropy field \( H_K \) (Fig.5d).

**CONCLUSION**

We examined the sputtering rate, the deposition rate and the film composition as a function of \( V_b \), with a parameter of \( H_s \) for the LTs- and HTs-films. The external fields \( H_s \) and \( V_b \) increase the number of \( Ar^+ \) ions and thus promote the sputtering rate. Furthermore, the negative bias \( V_b \) causes \( Ar^+ \) ions to bombard the substrate and to resputter the Co and Cr atoms deposited on the film surface, thus resulting the decrease in the deposition rate.

The resputtering rates of Co and Cr vary depending on the substrate temperature \( T_s \), and thus the film composition changes depending on \( T_s, H_s \) and \( V_b \).

The ion bombardment to the substrate influences the film morphology and the magnetic properties. The negative bias homogenizes the Cr distribution, thereby notably reducing the saturation magnetization. The good quality films, with high perpendicular anisotropy, can be obtained with the perpendicular magnetic field of \( 35 \sim 61\) Oe or the bias around \(-50\) V.

**REFERENCES**