THE INFLUENCE OF THE SEGREGATED MICROSTRUCTURE ON THE MAGNETIZATION REVERSAL IN CoCr FILMS

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The angular dependences of the $H_c$, $H_{an}$, orientation ratio (OR), hysteresis loss ($W_h$) as a parameter of the applied field and the dependence of the distribution of hysteresis loss ($\Delta W_h$) on the applied field have been systematically investigated with respect to the degree of Cr segregation in CoCr films. Starting with synthetic investigation of the microscopic phenomena and macroscopic measurement results, we attempt to explain the correlation of the segregated microstructure to the magnetization reversal mechanism in CoCr films. As the substrate temperature ($T_s$) increases from RT to 300°C, leading to an increase in the degree of the separation in CoCr films, all the $M_s$, $H_c$, OR, anisotropy constant and nucleation field ($H_n$) obviously increase. On the contrary, as $T_s$ increases the stripe-domain configuration will gradually disappear. There is a continuous transition between the domain-wall motion and rotational reversal, depending on the $T_s$ and the heat-treatment. Combining the TEM observation of segregated microstructure with the measured results of magnetic behaviour, the magnetization reversal for a partly segregated film is considered as the superposition of the domain-wall with perpendicular and in-plane orientation of the magnetization. However, in the case of a fully segregated film, it is thought to be the superposition of a cos type of perpendicular incoherent rotation reversal with in-plane domain-wall motion. Based on the above assumption, it was found that the measured angular dependences of the coercivity are in good agreement with the calculated ones for partly and fully segregated CoCr films. Experimental data show that the annealing treatment is favourable for improving the perpendicular recording properties in CoCr films.

1. Introduction

Recently much attention has been devoted to investigating the compositional inhomogeneity and to correlating the magnetic and microstructural properties to the deposition conditions in CoCr films [1–12]. The existence of Cr segregated columnar boundaries has been confirmed by TEM observation of the surface replica, using a selective chemical etching technique, by the compositional analysis of the etchant after etching [1–3] and by an AES (Auger Electron Spectroscopy) compositional analysis [4] in several CoCr films. It was found that such a segregated microstructure will appear if the Cr content of the composition in sputtered and evaporated CoCr films is higher than a certain critical concentration (about 12–17 at% [5,8]). The degree of segregation strongly depends on the deposition conditions (for example, the substrate temperature and annealing treatment, Ar pressure and thickness of the film etc.), except for the Cr content. Therefore, a variety of CoCr films with different degrees of the segregation from a homogeneously to a fully segregated state can be prepared if the preparation conditions are appropriately controlled.

On the basis of the above results, a segregation model has been proposed [1–4]. The magnetic Co-rich phase inside a column is surrounded by a less-magnetic Cr-rich layer at the grain boundaries. This Cr layer at the column boundary will greatly decrease the exchange interaction between the columns, so that the media no longer become continuous if the CoCr film is in a fully segregated state. In this case the magnetization will only be switched by the rotational reversal in the columns.

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without any domain-wall motion. Hence, it is reasonable to infer that both rotational and domain-wall reversals can possibly exist together in CoCr films and there is a continuous transition between the two models, depending on the segregated state. This hypothesis of the columns with a Cr segregated layer at the boundaries in CoCr films is also supported by the following arguments;

1) Both the saturation magnetization $M_s$ and the Curie temperature $T_C$ of sputtered CoCr films were found to exceed those for a bulk alloy with the same nominal composition; i.e. $M_s$ obviously deviates from the $M_s$ vs. Cr content curve (so-called S-P curve) [5,13].

2) As the degree of Cr segregation increases, the increase of $H_C$ is far larger than the corresponding increase of the anisotropy constant $K_1$ [13].

3) The increase in the degree of Cr segregation causes the demagnetizing factor $N$ to be much lower than 1 [6].

The TEM observation of the thin foil samples by combining selective chemical etching with a successive ion-beam milling technique was also used to investigate the segregated microstructure in CoCr films. A so-called “segregated CP-structure” has been proposed [7–12]. This CP-structure exhibits the following features:

a) The Co-rich ferromagnetic phase, which is separated by a Cr-rich (less-magnetic) phase, exists in each crystallite;

b) These phase stripes are almost perpendicular to the grain boundaries, existing with a wall-like structure perpendicular to the substrate plane;

c) Ferromagnetic Co-rich regions are formed coherently in each single-phase hcp crystallite.

According to this new segregation model, the origin of the segregation with a wall-like Co structure is due to the magnetic and non-magnetic phase separation in a column. Therefore, the size of the smallest ferromagnetic region will be much smaller than that of the column. Of course, this kind of microstructure is favourable for rotational reversal.

However, it is rather a pity that at present the related magnetic behaviours and segregated microstructure seem to be discussed separately. No strong arguments and satisfactory models can be presented to interpret the relation between the magnetization reversal mechanism and the segregated microstructure.

Trying to clarify the above problem in this report we will concentrate our attention on synthetically investigating the angular dependence of the $H_C$, remanent coercivity $H_{cr}$, OR, hysteresis loss $W_h$ as a parameter of the applied field $H_a$ and the dependence of the distribution of hysteresis loss $\Delta W_h$ with respect to $H_a$ for three typical CoCr films, which have quite different degrees of the segregation state. The angle $\theta$ is that between the applied field and film normal. Combining the TEM observation of the segregation state by using a selective chemical etching technique [7,8] with magnetic measurement results, different models of magnetization reversal will be proposed to interpret the angular dependence of the coercivity for partly and fully segregated films. This dependence will also be calculated, according to a similar procedure [18].

2. Experimental procedure

The CoCr films used were RF sputtered on glass and quartz substrates. These samples were provided by Yasushi Maeda (NTT, Japan). The substrate temperature ranged from room temperature (RT) to 300°C. The sputtering Ar pressure was 6 mTorr. The composition of the CoCr films was measured by Auger Electron Spectroscopy. The degree of the $c$-axis orientation was evaluated by X-ray. With the coercivities varying from 14 to 151 kA/m. The sputtering conditions and film properties are summarized in table 1.

It is known that the segregated state is closely related to the composition and deposition condition of sputtered CoCr films. In order to focus our attention on the main problem, we are only interested in discussing the effect of the substrate temperature and annealing treatment on the degree of segregation and attempting to correlate the magnetization reversal mechanism to the segregated microstructure. Therefore, three samples have been chosen having almost the same thickness and composition as seen in table 1. A 17.9
Table 1
Parameters and deposition conditions in CoCr films

<table>
<thead>
<tr>
<th>No.</th>
<th>( T_s ) (°C)</th>
<th>Thickness (nm)</th>
<th>Cr content (at%)</th>
<th>Deposition rate (nm/min.)</th>
<th>( \Delta \delta_{so} )</th>
<th>Substrate</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>(v23) RT</td>
<td>240</td>
<td>18.6</td>
<td>9.8</td>
<td>3.2</td>
<td>glass</td>
</tr>
<tr>
<td>B</td>
<td>(v26) 300</td>
<td>230</td>
<td>18.2</td>
<td>11.5</td>
<td>7.6</td>
<td>glass</td>
</tr>
<tr>
<td>C</td>
<td>(v28) *</td>
<td>250</td>
<td>18.3</td>
<td>5.3</td>
<td></td>
<td>quartz</td>
</tr>
</tbody>
</table>

* Sample C was first sputtered at 300 °C, then annealed at 650 °C for 1 h at 10⁻⁷ Torr.

at% Cr alloy target was used for sputtering the samples A, B and C. As shown in table 1, the Cr content of sample C did not change after annealing treatment. This fact shows that the Cr evaporation effect can be negligible during the annealing treatment.

According to the segregated microstructure observed by TEM using an accelerating voltage of 200 kV, the above three samples can be clarified as follows [9]:
1) Type A (the initial stage of the segregation): the compositional fluctuation is very small. No CP-structure can be clearly observed.
2) Type B (the final stage of the segregation): The compositional fluctuation becomes so large that non-magnetic regions are fully formed. A CP-structure can be clearly observed.
3) Type C (annealed sample): the annealing treatment at 650 °C causes the interdiffusion of Co and Cr atoms in a segregated column, resulting in forming a more or less homogeneous film.

For the sake of convenience, the A, B and C samples will afterwards be referred to as the partly, the fully segregated and the annealed CoCr film. According to the magnetic behaviour, sample C can be considered to be in the intermediate state between the fully segregated and the homogeneous state. Recently many more homogeneous films than type C has been successfully prepared. These films show lower \( H_c \) and \( K_1 \) than those of type C [11].

The most relevant properties obtained from the VSM and torque magnetometer are summarized in table 2. The maximum applied field for VSM measurements is 840 kA/m.

The distribution of the hysteresis loss \( \Delta W_h \) as function of the applied field is defined as

\[
\int_{H_{H_{1}}}^{H_{H_{2}}} dH / \sqrt{H_{H_{1}}} \cdot d(M_1 - M_2) dH,
\]

where \( M_1 \) and \( M_2 \) represent the descending and ascending loops respectively. \( H_2 \) and \( H_3 \) are defined as the fields at which the high and low field peak appear in the \( \Delta W_h \) vs. applied field curve, respectively, but \( H_1 \) as the field at which the hysteresis loss just becomes zero [18]. The definitions with regard to the other parameters, for example, orientation OR, slope \( T \) etc., are described elsewhere [16–18].

3. Experimental results and discussion

3.1. A partly segregated film

It was found that the so-called CP-structure universally appears in a limited Cr content range (15–24.3 at%) [8]. The experimental data clearly show that the degree of Cr segregation increases by increasing the substrate temperature \( T_s \) up to about 300 °C. It was also confirmed [8–12] that the so-called CP-structure will become more and more easily observable if \( T_s \) approaches 300 °C.

The results listed in table 2 show that as \( T_s \) increases, all the \( M_s \), \( H_{c, \perp} \), \( H_{c, \perp} / H_{k} \), \( K_1 \), and \( Q \) values increase.

Table 2
The relevant magnetic properties (\( H_s = 840 \text{ kA/m} \))

<table>
<thead>
<tr>
<th>No.</th>
<th>( M_s ) (kA/m)</th>
<th>( H_{c, \perp} ) (kA/m)</th>
<th>( H_{c, \perp} / H_{k} )</th>
<th>( H_{c, \perp} / H_{d} )</th>
<th>OR,( \perp )</th>
<th>( K_1 ) (10⁴ J/m²)</th>
<th>( T )</th>
<th>( Q )</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>362</td>
<td>14</td>
<td>0.033</td>
<td>1.25</td>
<td>0.38</td>
<td>95</td>
<td>1.128</td>
<td>1.15</td>
</tr>
<tr>
<td>B</td>
<td>519</td>
<td>151</td>
<td>0.23</td>
<td>4.75</td>
<td>1.59</td>
<td>247</td>
<td>1.167</td>
<td>1.46</td>
</tr>
<tr>
<td>C</td>
<td>325</td>
<td>64</td>
<td>0.10</td>
<td>6.15</td>
<td>4.1</td>
<td>132</td>
<td>1.116</td>
<td>1.98</td>
</tr>
</tbody>
</table>
monotonically, except for the slope $T$ of the perpendicular loop. According to the $H_{c\perp}/H_K$ values, by experience the partly (type A) and fully (type B) segregated samples can be considered as a low and a high coercivity film [16]. With increasing $H_{c\perp}/H_K$, the orientation direction of the magnetization gradually changes from the in-plane direction (OR = 0.38) to the perpendicular direction (OR = 1.59), because whether the OR value is greater or smaller than 1 is a simple and easy criterion for judging if the orientation of the magnetization lies in the perpendicular or in-plane direction [16,17].

The segregated microstructure in sputtered CoCr films can be evaluated by the film's $M_s$ vs. Cr content curve with an S–P curve for a binary system. It was found that the $M_s$ value of 519 kA/m for a fully segregated sample is much larger than the corresponding value of 405 kA/m for a bulk CoCr alloy having the same composition. However, the $M_s$ value of 362 kA/m for a partly segregated sample is not larger than the corresponding value of 395 kA/m. This fact may suggest that the microstructure of the sample sputtered at RT approaches the homogeneous state instead of the segregation state. As expected, owing to the annealing treatment, all the $M_s$, $H_{c\perp}$, $H_{c\perp}/H_K$ and $K_1$ values become much smaller than those of a fully segregated sample. It is generally agreed that the annealing treatment will reduce the degree of segregation and induce CoCr films to approach a homogeneous microstructure. It is however, strange that the annealing treatment causes a large increase of the OR value. Perhaps the defects of the crystallites in the annealed CoCr film are reduced by the annealing treatment, resulting in the reduction of the disturbance of the local stray field to the orientation of the magnetization.

Another possibility is the reformation and perfection of the CP-structure through the coalescence of ferromagnetic regions inside a column during the annealing treatment. This fact suggest that the annealing process must on no account be simply a homogeneous process, i.e., the metallurgically different dynamics exist in the samples sputtered at low substrate temperature and annealed at 600°C, respectively.

It is clearly seen from samples A and B (table 2) that as $T$ increases from RT to 300°C, the $H_{c\perp}$ value drastically increases from 14 to 151 kA/m. The rate of increase in $H_{c\perp}$ is as high as a factor of 10.8, but the rate of increase in $K_1$ is only a factor of 2.6. Therefore, the main contribution to the increase of the coercivity for a fully segregated sample is attributed to the segregated microstructure, if at same time the films sputtered at RT and 300°C having more or less the same size of the columns are considered.

It seems inconceivable that the slope $T$ of the perpendicular hysteresis loop is independent of the $T$ and heat-treatment, even though the three samples are in quite different segregated states. According to the particulate model [20], the slope is closely related to the size and the separation distance of the columns. The reason for this discrepancy of CoCr films is due to the fact that the slope is mainly determined by the characteristic of the initial layer instead of the "top layer". It was known [17] that the domain-wall dominates in the initial layer. According to the amplitude of the discontinuous magnetization jump in the in-plane loops, the thickness of the initial layer can be roughly estimated as about 250 Å for three samples. Based on the above results, it was found that the thickness of the initial layer for three samples is about 10% of the total thickness, independent of the substrate temperature.

It is clearly shown from fig. 1 that no matter how the applied field changes, the OR vs. $\theta$ curves are always below the straight line (OR = 1) for a partly segregated sample. This fact suggests that the magnetization preferentially lies in the in-plane direction. The angular dependence of $H_{c\perp}/H_{c\parallel}$ with respect to the applied field are shown in fig. 2. As can be seen from this figure, the effect of the applied field on the angular dependence of the coercivity is quite remarkable. If $H_a$ is higher than about 400 kA/m, there is a critical angle $\theta_c$ (about 45°) in the $H_{c\perp}/H_{c\parallel}$ vs. $\theta$ curve, above which the change tendency is almost identical with the inverse $\cos(90 - \theta)$. It is well known [17] that the change tendency will follow the inverse $\cos(90 - \theta)$ law if its magnetization is reversed by the in-plane domain-wall motion. However, below this angle $\theta = H_{c\perp}/H_{c\parallel}$ will drastically decrease with
decreasing angle. This kind of change pattern can be qualitatively explained by measuring the normalized demagnetizing factor $N$ vs. $\theta$ curve [17]. If $H_a$ is further reduced, $H_c/H_{c\|}$ will decrease monotonically with decreasing angle.

In comparison, the $H_c/H_{c\|}$ vs. $\theta$ curve for a bulk CoCr sample (Cr content: 20 at%, $H_{c\perp} + 1.37$ kA/m) is also plotted with a dashed line in fig. 2. It is clear that the angular dependence of the coercivity for a partly segregated CoCr film is strikingly similar to the behaviour of the bulk CoCr sample. It was known from our measurement results [18] that the magnetization reversal mechanism of a bulk CoCr sample belongs to domain-wall motion.

Similarly, the behaviour of the angular dependence of hysteresis loss $W_{h}/W_{h\perp}$ is also very similar to that of the low coercivity films which has previously been discussed in detail [17]. As shown in fig. 3, when $H_a$ is higher than about 400 kA/m, the $W_{h}/W_{h\perp}$ vs. $\theta$ curves decrease monotonically with increasing angle. However, if $H_a$ is decreased to about 80 kA/m, the $W_{h}/W_{h\perp}$ vs. $\theta$ curve will gradually change from a monotonically decreasing one to a more or less straight line, which is thought to be one of the important indications of the domain-wall motion [21].

The distribution of the hysteresis loss ($\Delta W_h$) with respect to $H_a$ for a partly segregated film was measured. It is clearly shown in fig. 4 that the distribution curve exhibits a typical double-peak characteristic (in fig. 4, $H_1$, $H_2$ and $H_3$ are marked on the curve only corresponding to the angle $\theta = 0$). The low and high field peaks on the distribution curves have the following distinguishing features: 1) As the direction of $H_a$ is changed from the perpendicular to the in-plane direction, the position of the high field peak $H_2$ will gradually move to the high field region, while the position of the low field peak $H_3$ is found to be independent of the field direction. Its peaks always appear at about zero field.

2) As the direction of $H_a$ is changed from 0 to 90°, the amplitudes of a high field peak $H_2$ de-
increase gradually, while those of a low field peak $H_3$ increase.

It has been previously expounded and proven [18] that the high and low field peaks exhibit the behaviour of the domain-wall motion with perpendicular and in-plane orientation of the magnetization, respectively. The magnetization reversal for this kind of CoCr film can be thought to be the superposition of the domain-wall motion with perpendicular and in-plane orientation of the magnetization. On the other hand, the proportion function $P(\theta)$ of the domain-wall motion with perpendicular orientation to the in-plane orientation strongly depends on the direction of $H_a$. If the heights of the field peak $H_2$ and low field peak $H_3$ for a partly segregated sample are assumed to represent the proportions of domain-wall motion with perpendicular and in-plane orientation of the magnetization then the proportion function $P(\theta)$ can be determined. According to the respective height of the distribution curve in fig. 4, the proportion of domain-wall motion with perpendicular to in-plane orientation of the magnetization reversal can be calculated for each angle ranging from 0 to 90°. (Notes: in order to make the graph clear, only the distribution curves for 0, 40°, 60° and 90° have been plotted in fig.

Fig. 5. The measured angular dependence of the $P(\theta)$ function for a partly and a fully segregated CoCr film.

Fig. 6. The calculated and measured angular dependence of the coercivity for a partly segregated CoCr film.
Table 3
The measured $H_n$ and $H_{cw}$ values for several CoCr films

<table>
<thead>
<tr>
<th>No.</th>
<th>A</th>
<th>B</th>
<th>C</th>
<th>D</th>
<th>E</th>
</tr>
</thead>
<tbody>
<tr>
<td>$H_n$ (kA/m)</td>
<td>342</td>
<td>844</td>
<td>509</td>
<td>320</td>
<td>448</td>
</tr>
<tr>
<td>$H_{cw}$ (kA/m)</td>
<td>262</td>
<td>*</td>
<td>192</td>
<td>260</td>
<td>*</td>
</tr>
</tbody>
</table>

Note: samples D and E are typical low and high coercivity CoCr films.
* $H_2$ is not observable.

4.) The measured dependence of the proportion function $P(\theta)$ is shown in fig. 5. Hence, the coercivity as function of the angle can be calculated by formula:

$$
H(\theta)/H_{c\perp} = (1 - \alpha \sin^2\theta)^{-1/2} P(\theta) \\
+ \left( H_{c0}/H_{c\perp} \right) (1 - \alpha \cos^2\theta)^{-1/2} (1 - P(\theta)),
$$

where $\alpha$ is an adjustable parameter. With $P(\theta)$ obtained from fig. 5, the calculated curves of the coercivity for $H_{c\parallel}/H_{c\perp} = 0.95$ are shown in fig. 6 if the $\alpha$ values are taken as 0.7 and 0.8. It can be seen from fig. 6 that the calculated angular dependences of the coercivity are quite consistent with the measured one drawn with a dashed line ($H_n = 840$ kA/m). It is also seen from fig. 4 that the in-plane magnetization component has a tremendous influence on the magnetic behaviour. Even in the perpendicular direction the influence can still not be neglected.

According to a similar procedure [18], the dot-like domain nucleation $H_n$ and stripe-domain nucleation $H_{cw}$ can be measured. The $H_n$ value for low $H_{c\parallel}/H_k$ CoCr films was obtained by extrapolating the dot-like domain density to zero according to the curve of the domain density as a function of the applied field, while the $H_{cw}$ can be determined by observing the appearance of the first stripe domain in the Kerr-microscopic photograph [22]. It has been discovered [18] that when the applied field is applied along the film normal, the position of $H_2$, where the high field peak of the distribution curve $\Delta W_h$ appears, corresponds to the stripe-domain nucleation field $H_{cw}$, while the position of $H_1$, where the hysteresis loss just become zero, corresponds to the dot-like domain nucleation field. The $H_n$ and $H_{cw}$ values for a partly segregated sample are 342 and 262 kA/m, respectively. Comparing these values with those of a typical low coercivity film D (table 3), it can also be seen that both the $H_n$ of 342 kA/m and $H_{cw}$ of 262 kA/m for a partly segregated sample are very close to those values of 320 and 260 kA/m for low coercivity films.

3.2. A fully segregated film

The angular dependences of OR as a parameter of $H_n$ for a fully segregated sample are shown in fig. 7. It is very interesting to see that if the applied field is higher than $H_{c\perp \text{max}}$ (151 kA/m), the behaviour of the OR vs. $\theta$ curves is usually seen for perpendicular oriented media, but in the low $H_n$ range (about < 60 kA/m) the OR vs. $\theta$ curves will be changed from larger than 1 to less than 1. This fact clearly suggests that the orientation direction of the magnetization strongly depends on the applied field. In the high $H_n$ the magnetization lies in the perpendicular direction, but in the low $H_n$ the orientation of the magnetization will be in the in-plane direction.

The angular dependences of $H_{c\parallel}/H_{c\perp}$ and $H_{c\parallel}/H_{c\perp}$ on the applied field are shown in figs. 8, 9. It can be seen from fig. 8 that when $H_n$ is higher than $3H_{c\perp \text{max}}$ (151 kA/m), the angular depen-

![Fig. 7. The angular dependence of OR for a fully segregated CoCr film.](image-url)
Fig. 8. The angular dependence of $H_c / H_{c\perp}$ for a fully segregated CoCr film and the theoretical curves for the cos. and W-W rotation reversal models.

dence of $H_c / H_{c\perp}$ basically follows the change pattern of the incoherent rotation. However, comparing this change tendency with some classical models of rotational reversal, it was found that the $H_c / H_{c\perp}$ vs. $\theta$ curve obviously follows neither the S-W model nor the cos model. The reason for giving rise to this discrepancy will be explained in the ensuring paragraph about the calculation of the angular dependence of the coercivity at saturation field. As seen in fig. 9, if $H_a$ is gradually reduced to approach the $H_{c\perp,max}$, the $H_c / H_a$ vs. $\theta$ curve will basically follow the inverse $\cos(90 - \theta)$ law (see broken line), which is a strong indication of the in-plane domain-wall motion. If $H_a$ is continuously decreased to become lower than $H_{c\perp,max}$, then $H_c / H_{c\parallel}$ increases with decreasing angle, starting from the in-plane direction, a peak of this curve will appear at a critical angle $\theta_{cr}$. Then the $H_c / H_{c\parallel}$ will drastically decrease with decreasing angle if it is lower than $\theta_{cr}$. The reason for causing the $H_c / H_{c\parallel}$ vs. $\theta$ curve to have such a change tendency has already been described elsewhere [17]. In view of the above results about the behaviour of OR and the coercivity with respect to the applied field, it can be assumed that the $H_{a} > H_{c\perp,max}$ is an essential condition to ensure this kind of CoCr film exhibiting perpendicular recording properties, because OR $> 1$ and $H_{c\perp} / H_{c\parallel} > 1$ are considered as indispensable conditions for perpendicular recording. The angular dependence of remanent coercivity $H_{c\perp}$ for a fully segregated CoCr film and the critical field of the rotational reversal for a single-domain particle with a uniaxial anisotropy [19], which are drawn with a solid and a dashed line respectively, are shown in fig. 10. It was unexpected found that there is a strikingly similar change tendency between them both. When $\theta = 45^\circ$, both the $H_{cr}$ for a fully segregated film and the critical field for a single-domain particle are minimum. This means that for a fully segregated film sputtered at 300°C the remanent coercivity starts exhibiting a more or less typical behaviour of single-domain particles. This fact also suggests that as the substrate temperature is increased from RT to 300°C the de-
degree of Cr segregation is greatly improved. It is proven from magnetic behaviour that the wall like Co-rich ferromagnetic part inside the columns is perfectly surrounded by the non-magnetic Cr-rich part in a CoCr film sputtered at 300 °C.

Obviously, this kind of segregated microstructure is very favourable for a CoCr film exhibiting magnetic behaviour associated with the particles. However, it should be noticed that the microstructure of the segregated CoCr films is quite different from a single-domain particle, because the magnetic interaction between the columns in CoCr films has a tremendous influence on the magnetic behaviour. Therefore, an attempt to directly compare the measured magnetic behaviours of CoCr films with classical ones of single-domain particles without any modification will be unsuccessful.

For a fully segregated sample, the angular dependence of the measured hysteresis loss \( W_b/W_{hi} \) and \( W_b/W_{hil} \), which are drawn in fig. 11 with a solid and a dashed line, respectively, are plotted against different \( H_b \), representing the applied field higher and lower than the \( H_{c,\perp \max} \) (151 kA/m). It can be seen that the \( W_b/W_{h,\perp} \) vs. \( \theta \) curves for three different \( H_b \) without any exception monotonically decrease with increasing angle, and qualitatively follow the same change pattern expected by incoherent rotation models in fields higher than a \( H_{c,\perp \max} \) of 151 kA/m. However, if \( H_b \) is lower than \( H_{c,\perp \max} \), the hysteresis loss \( W_b/W_{hil} \) vs. \( \theta \) curves follow quite different change patterns. If the angle increases, the hysteresis loss first increases, reaching a maximum value at an appropriate critical angle, further increase of the angular results in a diminished hysteresis loss.

This fact suggests that at present the magnetization reversal mechanism of a CoCr film must differ from that of the rotation reversal model. It is noted here that the magnetic behaviours (the angular dependence of the coercivity, orientation ratio and hysteresis loss with respect to \( H_b \)) for a fully segregated CoCr film are very similar to

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**Fig. 11.** The angular dependence of \( W_b/W_{hil} \) on the applied field for a fully segregated CoCr film.

**Fig. 12.** The dependence of the distribution of hysteresis loss \( \Delta W_b \) on the applied field as a parameter of the angle for a fully segregated CoCr film.
those of a high coercivity CoCr film, which was investigated previously [17], except for the angular dependence of the remanent coercivity on $H_a$. As expected, the behaviour of the distribution of the hysteresis loss $\Delta W_h$ with respect to $H_a$ for a fully segregated sample is also similar to that of a high coercivity film [17]. As shown in fig. 12, there is the presence of a single peak on the $\Delta W_h$ vs. $H_a$ curves and no matter how the applied field changes, these distribution curves always exhibit a monotonically decreasing form, which was previously proven to be one of a number of important indication of the rotational reversal [18]. As the angle increases from 0 to $90^\circ$ the low field peak will become higher and higher. This fact suggests that the in-plane magnetization component exerts a tremendous influence on the magnetic behaviour of this sample in the vicinity of $90^\circ$. The presence of a low field peak, which appears at about zero field, is attributed to the in-plane domain-wall motion [17,18]. Combining this with an angular dependence of $H_c$, which is similar to that of a single-domain particle, we assume that the magnetization reversal is the superposition of an S–W type of rotational reversal with the perpendicular orientation of the magnetization and the in-plane domain motion. As the angle increase, the proportion of the S–W type of rotational reversal to in-plane domain-wall motion will gradually decrease. According to a similar procedure [18], the measured proportion function $P(\theta)$ of the perpendicular rotational reversal to the in-plane domain-wall motion is plotted in fig. 5. Therefore, the angular dependence of the coercivity for a fully segregated sample can be calculated by formula:

$$H_c(\theta)/H_{c\perp} = \frac{F(\theta)P(\theta) + (H_{c\parallel}/H_{c\perp})(1 - \alpha \cos^2 \theta)^{-1/2}}{(1 - P(\theta))},$$  

(2)

where $F(\theta)$ represents the function of S–W rotational reversal. Unfortunately, as seen in fig. 13, there is a rather large discrepancy between the measured and calculated curves of the coercivity if $\alpha$ and $H_{c\parallel}/H_{c\perp}$ are taken as 0.8 and 0.2, respectively. i.e the good agreement between the calculated and measured curves only exists in the range of 60–90°, but in the range of 0–60°, there is a quite different change pattern between them both. In order to solve this problem, we again assume that the rotational reversal for a fully segregated sample follows the cos type model, because it was discovered [16,17] that for most high coercivity CoCr films the angular dependence of the coercivity is a rather better fit for a cos model than an S–W model. Hence, the angular dependence of the coercivity is again recalculated by formula:

$$H_c(\theta)/H_{c\perp} = \cos(\theta)P(\theta) + (H_{c\parallel}/H_{c\perp})(1 - \alpha \cos^2 \theta)^{-1/2} \times (1 - P(\theta)),$$

(3)

where $\alpha = 0.8$ and $H_{c\parallel}/H_{c\perp} = 0.2$. It is very interesting to see in fig. 13 that the calculated curve is almost identical with the measured one. One of the most distinct features of the perpendicular loop for partly and fully segregated CoCr films is the presence or absence of a characteristic shoulder, respectively. Whether the presence of the characteristic shoulder is generally considered as one of the criteria for judging if the domain-wall motion dominates or not [7–10]. This fact again shows that the substrate temperature certainly has a tremendous influence on the magnetization reversal mechanism in sputtered CoCr films.
3.3. An annealed CoCr film

Comparing a fully segregated sample with an annealed sample, it can be clearly shown from table 2 that for the latter all the values, including $M_t$, $H_{c\perp}$, $K_1$ and $H_{c\perp}/H_K$, are much less than those of a fully segregated sample. It is easily understood that as a result of the annealing treatment, the segregated Cr atoms will again interdiffuse into the columns. Of course, it will lead the composition of a segregated film to be in relatively homogeneous state and result in a reduction of the degree of the segregation. Experimental data shows that this sample is incompletely annealed, because other fully annealed 18 at% CoCr films exhibit much lower $H_e$ and anisotropy constant [11]. For a thoroughly annealed sample, the originally segregated microstructure of CoCr films sputtered at 300°C, will presumably be again homogenized.

As seen in table 2, the orientation ratio and $H_{c\perp}/H_{c||}$ for an annealed sample is much higher than those of non-annealed CoCr films. This means that annealing treatment is rather favourable for improving apparently perpendicular anisotropy. The reason for the increase of $OR_{\perp}$ and $H_{c\perp}/H_{c||}$ instead of them becoming less, due to the decrease of $K_1$, still requires further investigation. The angular dependences of $OR_{\perp}$, $H_{c\perp}/H_{c\perp}$ and $W_h/W_{h\perp}$ with respect to $H_a$ for an annealed sample are shown in figs. 14–17. Comparing the corresponding magnetic behaviour of non-annealed samples with those of this annealed sam-

\[ H_a = 880 \text{ kA/m} \]
\[ H_a = 64 \text{ kA/m} (H_{c\perp} \text{ max}) \]
\[ H_a = 40 \text{ kA/m} \]

Fig. 15. The dependence of $H_e/H_{c\perp}$ with respect to the applied field for an annealed CoCr film.

ple, some quite unique features can be listed as following from these figures: 1) No matter how the applied field changes (from 40–880 kA/m), all the OR vs. $\theta$ curves always maintain monotonically increasing tendencies with increasing angle (see fig. 14). Even at the field below $H_{c\perp}$ max of 66.4 kA/m, this situation is still true. 2) No matter how the applied field changes, all the $H_{c\perp}/H_{c\perp}$ and $W_h/W_{h\perp}$ vs. $\theta$ curves exhibit monotonically decreasing forms with increasing angle (see figs. 15, 16). It is interesting to see from fig. 15 that at a saturated field of 880 kA/m the $H_{c\perp}/H_{c\perp}$ vs. $\theta$ curve is in quite good agreement with the $\cos(\theta)$ law, which is considered as one of the important indications for incoherent rotation, except in the range of 80–90°. 3) The values of

\[ H_a = 880 \text{ kA/m} \]
\[ H_a = 64 \text{ kA/m} (H_{c\perp} \text{ max}) \]
\[ H_a = 40 \text{ kA/m} \]

Fig. 16. The angular dependence of the hysteresis loss as a parameter of the applied field for an annealed CoCr film.
the orientation ratio OR for an annealed sample are much higher than those of non-annealed ones at varying \( H_a \). This means that the annealing treatment is very advantageous for the improvement of the perpendicular orientation of the magnetization.

4) As shown in fig. 17 the angular dependence of the remanent coercivity is very consistent with the inverse cos curve, which is another indication of the rotational reversal.

Based on the above results, it is perhaps reasonable to conclude that for the annealed CoCr film tested the rotational reversal governs the magnetization switch, even though a characteristic shoulder appears at the perpendicular loop.

As a result of the annealing treatment, the magnetic behaviour associated with the rotational reversal will gradually weaken. Comparing this annealed film with the non-annealed ones the values of \( H_{c_{\perp}} \), \( K_1 \) and \( H_{c_{\perp}}/H_k \) are reduced from 151 kA/m, 247 \( \times 10^4 \) J/m and 0.23 to 64 kA/m, 132 \( \times 10^4 \) J/m and 0.1, respectively. At the same time, the related behaviour associated with domain-wall motion will gradually strengthen (for example, the presence of a shoulder in the perpendicular loop). The above argument can be supported by the nucleation field for the dot-like and stripe-domain configurations in CoCr films. As seen in table 3, when the substrate temperature increases from RT to 300°C, the nucleation field \( H_n \) for a dot-like domain increases from 342 to 844 kA/m. However, \( H_n \) will decrease from 844 to 509 kA/m if the CoCr film sputtered at 300°C is again annealed at 650°C for 1 h. On the other hand, the nucleation field \( H_{cw} \) of the stripe-domain for a CoCr film sputtered at RT is 262 kA/m, but there is not any observable \( H_{cw} \) in a film sputtered at 300°C. An observable \( H_{cw} \) for this annealed film was found to be 192 kA/m. Based on the above-mentioned results, it is reasonable to infer that there is a continuous transition between the rotational reversal and domain-wall motion in CoCr films, depending on the \( T_s \) and heat-treatment.

4. Conclusions

1) The degree of segregation in CoCr film is closely related to the composition and preparation condition. As the substrate temperature is increased from RT to 300°C, all the values of \( M_s \), \( H_{c_{\perp}} \), OR \( \perp \), anisotropy constant and the nucleation field obviously increase.

2) Both the rotational reversal and domain-wall motion can possibly exist in CoCr films, depending on the segregation state. There is a continuous transition between the two models.

3) Combining the TEM observation of the segregated microstructure with the measured results of magnetic behaviour, the magnetization reversal for a CoCr film sputtered at RT is considered as the superposition of the domain-wall motion with perpendicular and in-plane orientation of the magnetization. In the case of the film sputtered at 300°C, it is thought to be the superposition of a cos type of perpendicular incoherent rotational reversal with in-plane domain wall-motion. Based on the above models, it was found that the measured angular dependence of the coercivity is very consistent with the calculated one.

4) Experimental data show that the annealing treatment is favorable for improving the perpendicular recording properties. It was discovered that the angular dependences of \( H_n \) and \( H_{cw} \) follow quite well the cos and inverse cos law. The rotational reversal governs the magnetization switch for a CoCr film annealed at 650°C, even though a characteristic shoulder appears in the perpendicular loop.
5) The degree of segregation degree and the magnetization reversal mode are strongly controlled by the $T_s$ or annealing, while the films have an almost constant value of Cr content.
6) Practice again confirms that the method, which differentiates magnetization reversal mechanism according to the $H_{c\perp}/H_k$ values, is an efficient measure.

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