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Reversal Mechanism in Perpendicular Media with Columnar Microstructure

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A contribution is given to the discussion concerning the nature of the magnetization reversal process in sputtered CoCr recording media with columnar morphology. The paper is restricted to the additional effects on the total perpendicular anisotropy by studying the columnar shape anisotropy and the properties of the perpendicular hysteresis loop (shearing and coercivity). On the basis of experiments and calculated results the conclusion is made that the columnar shape anisotropy has no significant influence and a final remark is that a strict particle model cannot account for the magnetic properties of CoCr layers and that a domain wall model offers a better alternative.

Es wird ein Beitrag über die Natur des Ummagnetisierungsprozesses in gesputterten CoCr-Speicherschichten mit säulenförmiger Morphologie gegeben, der auf Effekte beschränkt ist, die zur gesamten magnetischen Senkrechtanisotropie beitragen, indem die Formanisotropie der säulenförmigen Körner und Eigenschaften der Senkrecht-Hystereseschleife (Scherung und Koerzitivfeldstärke) untersucht werden. Auf der Grundlage von Experimenten und Berechnungen wird geschlossen, daß die Formanisotropie der Säulen keinen signifikanten Einfluß hat. Mit einem strengen Teilchen-Modell können die magnetischen Eigenschaften der CoCr-Schichten nicht erklärt werden, während ein Domänenwand-Modell eine bessere Alternative darstellt.

1. Introduction

Perpendicular magnetic recording has been proposed as a method to achieve very high densities [1, 2]. A CoCr film with perpendicular anisotropy is the most commonly proposed medium for this mode of recording.

Extensive studies on the structural properties and magnetic behaviour have been presented in the literature. It appears that films with about 19 at% Cr possess a relatively large saturation magnetization and large perpendicular anisotropy. This has been observed in layers, which are prepared by various sputter deposition methods, such as triode dc [3], dc magnetron [4], rf magnetron [5], target facing method [6], and ion-beam [7] as well as electron-beam evaporation [8]. The published data clearly show the relationship between magnetic properties, microstructure, and morphology as well as the deposition process.

The prepared CoCr layers exhibited more or less a columnar-like morphology with the *hcp c*-axis orientation perpendicular to the substrate. Such a columnar microstructure is shown by fractured cross-sectional SEM observation [9] and TEM studies of microtome slices [10].

A very important problem, in the case of media having such a morphology, is the nature of the magnetization reversal process [2, 11]. Our contribution to this question is the discussion of the additional effects caused by the columnar morphology to the total perpendicular anisotropy. In the case of CoCr the perpendicular magnetization

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Fig. 1. Fractional cross-section of rf sputtered CoCr layer

is mainly established by crystalline anisotropy, e.g. [9]. Our results are based on rf sputtered CoCr layers which have been prepared using optimal deposition parameters [12]. A typical fractional cross-section observed with SEM is shown in Fig. 1. Independent of the layer thickness our samples exhibit a hep structure with the [001] axis perpendicular to the substrate. This has been measured by the X-ray rocking curve method [9] and electron diffraction [12]. Even the so-called initial layer (up to ≈ 100 nm) already shows highly oriented columns and a very low in-plane remanence [9]. The morphology strongly suggests that the CoCr medium may be considered as particulate, supposing that the exchange coupling is broken at the crystallites boundaries. This type of magnetic microstructure is proposed by some authors for the rf sputtered CoCr medium [13, 14].

In this paper we will present the following contributions:

- (i) the study of the shape anisotropy of a columnar structure additional to the crystal anisotropy of the CoCr layers.
- (ii) The magnetic reversal mechanism of a columnar structure.

Fig. 2. Simplified geometry of the columnar structure

These results will be compared with data obtained from the domain-wall model and experiments. The discussion will be focussed on three items: (i) the columnar shape anisotropy, (ii) shearing of the easy-axis loop, and (iii) coercivity.

2. Columnar Shape Anisotropy

First we will consider the shape anisotropy of this columnar structure, which is different from that of a uniformly magnetized layer. Fig. 2 shows a simplified geometry as proposed by Iwata et al. [15] for the elucidation of perpendicular anisotropy in NiFe layers. This simplified geometrical model has already been discussed in literature [16, 17, 18].

The columnar crystallites with length L (i.e. the film thickness h) have a square cross-section (length of the sides S) and have their side faces parallel to each other. The intrinsic saturation magnetization of the columns is equal to M_s and uniformly perpendicular. They are separated by a nonmagnetic medium (i.e. the crystallite boundary layer) of thickness d . This morphology gives rise to shape anisotropy, defined by the energy expression

$$E = K_s \cos^2 \theta, \quad (1)$$

where θ is the angle between the magnetization and the normal of the film plane, and K_s the perpendicular shape anisotropy constant. The shape anisotropy of this columnar structure will differ from that expected for a layer with a measured "uniform" magnetization $\bar{M}_s$, given by

$$\bar{M}_s = M_s \left\{ \frac{S}{S+d} \right\}^2 = M_s p. \quad (2)$$

Here M_s is the intrinsic column magnetization and p is called the "packing fraction" defined as the volume fraction of magnetic particles in the medium.

K_s can be expressed by

$$K_s = \frac{1}{2} \mu_0 M_s^2 (N_{\perp} - N_{\parallel}) p. \quad (3)$$

Because the sum of the demagnetizing factors is given by $N_{\perp} + 2N_{\parallel} = 1$ it follows from (3)

$$K_s = \frac{1}{4} \mu_0 M_s^2 (3N_{\perp} - 1) p. \quad (4)$$

Iwata et al. [15] calculated K_s , including the effect of a finite length L . Combining

their results with (4) yields $N_{\perp}$ for a finite length L of the columns,

$$N_{\perp} = p + \frac{2(S+d)}{\pi^2 L} \sum_{m=1}^{\infty} \frac{1}{m^2} \sin^2(m\pi\sqrt{p}) \left[1 - \exp\left(-\frac{2m\pi L}{S+d}\right) \right] + \\ + \frac{2(S+d)}{\pi^2 L p} \sum_{m=1}^{\infty} \sum_{n=1}^{\infty} \frac{1}{m^2 n^2 (m^2 + n^2)^{1/2}} \sin^2(m\pi\sqrt{p}) \sin^2(n\pi\sqrt{p}) \times \\ \times \left\{ 1 - \exp\left(-\frac{2\pi(m^2 + n^2)^{1/2} L}{S+d}\right) \right\}. \quad (5)$$

$N_{\perp}$ appears to be quite independent of L/S , only changing significantly for relatively small values of L/S . In the limit $L/S = \infty$, $N_{\perp}$ equals p and in the limit $L/S = 0$, $N_{\perp} = 1$, being the demagnetizing factor of a thin film with infinite in-plane dimensions.

3. Shearing Easy-Axis Loop

Next we shall discuss some aspects of the easy-axis loop, which may be expected for this columnar particle structure.

It can be shown for $L/S = \infty$ that the demagnetizing field, acting on the columns, is uniform and just equals $-\bar{M}_s$. If the layer is not saturated, the demagnetizing field will be $-\bar{M}$ because of the same arguments. This means that the perpendicular hysteresis loop will not show any anomalous shearing for $L/S = \infty$. Some anomaly of the loop must therefore be caused by the finite length of the columns. In the general case L/S is finite, the demagnetizing fields H_{ds} which is effective in switching can be calculated by considering the switching of one single domain column with intrinsic coercive field H_{ci} , which does not depend on magnetostatic interactions.

The column will switch at H_{cr} , a certain critical value of the external field, and we can write

$$H_{cr} - N_x M_s = -H_{ci}. \quad (6)$$

The field $-N_x M_s$ represents the effective demagnetizing field and N_x is the unknown demagnetizing factor which has to be solved. Switching of the column requires a energy E_{cr} , which is dissipated because of the coercivity of the medium,

$$E_{cr} = 2\mu_0 H_{ci} M_s V, \quad (7)$$

where V denotes the volume of the column. Switching may be expected, if the required energy equals the difference in magnetostatic energies between the states of the layer before and after switching. According to Cape and Lehman [19] this energy difference ΔE is given by

$$\Delta E = 2\mu_0 M_s^2 V (N_{\perp} - N_p) - 2\mu_0 H_{cr} M_s V, \quad (8)$$

where N_p denotes the demagnetizing factor of the individual column parallel to its axis. By stating that $E_{cr} = \Delta E$ and by means of (6) it follows that

$$N_x = N_{\perp} - N_p. \quad (9)$$

The demagnetizing field H_{ds} , which is effective in column switching, is thus given by

$$H_{ds} = - (N_{\perp} - N_p) M_s = - \frac{(N_{\perp} - N_p) \bar{M}_s}{p}. \quad (10)$$

This result is in principle only valid for the saturated layer. Nevertheless, we will assume that (10) is also applicable to the non-saturated case. This seems reasonable because application results in hysteresis loops with a shearing, which depends on the relative dimensions of the columns. For $L/S = 0$, we obtain $N_{\perp} = N_p = 1$ and $H_{ds} = 0$, which implies that the loop does not shear, as can be expected in this limit

Fig. 3

Fig. 3. The relative columnar shape anisotropy K_s versus the film thickness h for various columnar separations d . CoCr at 15 and 19 at% Cr

Fig. 4

Fig. 4. The first-order anisotropy constant K_1 versus the film thickness h , determined by means of torque ($\circ$, $\bullet$) as well as VSM (Δ , $\blacktriangle$) measurements. $\bullet$, $\blacktriangle$ 15 at% Cr; $\circ$, Δ 19 at% Cr

because there is no further interaction between the particles. If $L/S = \infty$, we obtain $N_{\perp} = p$ and $N_p = 0$, so that $H_{ds} = -\bar{M}$, resulting in the "normal" shearing of a perpendicular hysteresis loop.

4. Contribution of the Columnar Shape Anisotropy

We shall now use the results, previously given, to calculate the possible contribution of the columnar shape anisotropy to the total anisotropy of CoCr layers. The measured crystallite diameter D [9] is taken as the sum of the columnar diameter S and the boundary layer thickness d . The measured averaged diameter D , however, depends on the film thickness, while in the model $S + d$ is assumed to be constant across the entire film thickness. As an approximation we assume D , as measured at a certain film thickness, to be constant across that film thickness. A practical estimate of d , the columnar separation, is difficult.

Only the thinnest films (25 nm) yield clear TEM pictures of the crystallites [9]. In fact these pictures do not show any distinct boundary layer between the crystallites. Therefore we assumed d values of 0.5, 1, 2, and 3 nm. Fig. 3 shows the columnar shape anisotropy K_s of the layers, as calculated according to [15]. A significant deviation of the perpendicular anisotropy from the "homogeneous" shape anisotropy $0.5\mu_0\bar{M}_s^2$ is only observed for the thinnest films and is in the order of 10 to 50%, strongly dependent on the film thickness and the assumed columnar separation. This predicted behaviour, however, is not obvious from the measurements.

The intrinsic anisotropy constant K_1 , which is predominantly crystalline [20] and is depicted in Fig. 4, was obtained from torque measurements by correcting the

Fig. 5

Fig. 5. The measured hysteresis loop shearing T at $M = 0$ versus the film thickness h for two different CoCr layer compositions: $\square$ 15, $\circ$ 19 at% Cr

Fig. 6

Fig. 6. The perpendicular hysteresis shearing T at $M = 0$ calculated according to the columnar model with demagnetizing N_p for one column

measured, effective anisotropy with the "homogeneous" shape anisotropy $0.5\mu_0\bar{M}_s^2$. A significant contribution from the columnar structure would have resulted in a relatively larger K_1 for the thinnest layers, according to Fig. 3. However, Fig. 4 clearly shows that K_1 of 19 at% Cr layers is practically independent of the film thickness and for 15 at% Cr layers K_1 of the thinnest films even decreases. This counters a significant contribution of columnar shape anisotropy.

5. Hysteretic Properties

The shearing of the perpendicular hysteresis loop is considered by taking the slope T at $M = 0$: $T = (dM/dH)_{M=0}$. A value of T that is different from 1 indicates some anomaly in the magnetic behaviour, as may be caused by domain walls or a columnar structure. Fig. 5 shows T , as determined from the perpendicular hysteresis loops of films of both compositions in the thickness range of 25 to 600 nm. Especially the thinnest films exhibit a large anomaly of the hysteresis loop slope, T being much larger than unity. The shearing of the hysteresis slope at $M = 0$ in the columnar model is determined by the demagnetizing field H_{ds} , as given by (10). The slope T of the magnetization curve is then calculated to be $T = p/(N_\perp - N_p)$. We have determined T for $d = 0.5, 1, 2$, and 3 nm, respectively. The results, as depicted in Fig. 6, show that the calculated T cannot be fitted to the experimentally found T . The calculated slope T appears to be quite independent of the parameter d for the actual value of d/S and L/S . The calculated T cannot be fitted to the measured T by adjusting the columnar separation d .

Another important parameter of the perpendicular hysteresis loop is its coercivity. It was found that the coercivity of the CoCr medium is much less than its intrinsic anisotropy field H_{k1} [11]. If we assume the crystalline particles to be single domain, which especially for the thinnest films should be true, this reduction of the crystalline coercivity can only be explained by possible thermal perturbation of the magnetization.

This temperature influence on the coercivity is given by [21]

$$H_{cl} = H_{k1} \left\{ 1 - \left(\frac{T}{T_b} \right)^{1/2} \right\}, \quad (11)$$

Fig. 7. The relative magnetization m at the relative field $H/M_s = 1$, both calculated for 19 at% Cr films. --- measured m , — calculated according to Langevin theory

where T_b denotes the so-called blocking temperature for superparamagnetic behaviour. Straightforward calculation according to (11) yields a coercivity in the order of 100 kA/m, which appears to be in reasonable agreement with the measured coercivity of the thinnest films [11]. Nevertheless, the supposed thermal perturbation of the magnetization should be visible in the magnetization curves of the thinnest films, as measured by a VSM. We shall apply the Langevin theory of paramagnetism on ferromagnetic particles with intrinsic anisotropy K_1 [22]. Consider an assembly of columnar particles in a thin film, as shown in Fig. 2. The magnetization of all particles has the same polarity as the applied perpendicular field H . This situation is easy to realize experimentally by saturating the film first in a large field and then keeping $H > (M_s - H_c)$ to avoid switching of the particles. As a result of thermal activation, the magnetization of each particle may deviate from the film normal, i.e. the easy axis. It is assumed that the magnetization within each particle is uniform by exchange interactions. The energy E of a particle with volume V , of which the magnetization M_s makes an angle θ with the easy axis is given by

$$E = V \{ K_1 \sin^2 \theta - \mu_0 M_s H_t \cos \theta \}. \quad (12)$$

The total field H_t is given by $H_t = H + H_d = H - \bar{M}$, where H_d denotes the perpendicular demagnetizing field acting on the particles. Because the diameter is small compared with the length of the crystallites, H_d can be approximated by $-\bar{M}$, where $\bar{M}$ is the measured, average magnetization of the layer. In the case of ferromagnetic particles with many atoms, the angle may have any value and applying Boltzmann statistics the relative, perpendicular magnetization m is given by

$$m = \frac{M}{M_s} = \frac{\int_0^\pi \exp(-E/kT) \cos \theta \sin \theta d\theta}{\int_0^\pi \exp(-E/kT) \sin \theta d\theta}. \quad (13)$$

Straightforward calculation yields the equation

$$\int_{-1}^{+1} (m - x) \exp \{ \alpha(u - m)x - \beta(1 - x^2) \} dx = 0, \quad (14)$$

where

$$x = \cos \theta, \quad \alpha = \frac{\mu_0 M_s^2 V}{kT}, \quad u = \frac{H}{M_s}, \quad \text{and} \quad \beta = \frac{K_1 V}{kT}.$$

We have calculated the relative magnetization m at $u = 1$ for 19 at% Cr films with $10 < h < 60$ nm. The result is shown in Fig. 7 together with the measured m .

The theory predicts a significant deviation of m from 1. This is in disagreement with the experimental results, which exhibit no significant decrease of M_s at this field.

6. Discussion and Conclusions

6.1 Loop shearing

The experimentally observed hysteresis loops exhibit anomalous shearing which is different from what is expected from the demagnetizing field of thin, magnetically uniform layers. This anomaly can, in principle, arise from a domain structure including walls or from the particle character of the columnar morphology.

As can be seen above one of the basic assumptions of our model is the quasi-homogeneous magnetization $\bar{M}_s$ of the layer in all stages of the hysteresis loop, i.e. during the total process of magnetization reversal.

Andrä et al. [18, 23, 24] have investigated a model where the existence of "domains" in the form of rows or a chain of columns is permitted. In this domain model the magnetization of the columns is simultaneously switched within each chain. They have shown by means of numerical calculations that successive magnetization reversals of rows of columns is likely to occur.

On the basis of neutron depolarisation studies Hemmes et al. [25] also agreed that a "chain of columns" model can be applicable to our rf sputtered CoCr layers. After comparison of the volume coercivity (measured with VSM) with the surface coercivity (M. O. Kerr measurement) we proposed a preliminary model on how reversals in CoCr can be nucleated [26].

Using a digital imaging processing method the group of Hubert [27, 28] have demonstrated the process of nucleation and growth of domains in sputtered CoCr as a function of the applied perpendicular field. The observed images resemble those already known from bubble materials.

One consequence of the difference between Andrä's approach and our work is a different result with respect to the shearing of the hysteresis curve. He replaces our demagnetizing factor of one column (N_p) by (N_r) which is the demagnetizing factor of a chain of columns. In the case of a long straight chain $N_r = S/S + L$ where S and L are approximately the grain size (D) and the film thickness (h), respectively (see Fig. 2). Andrä [29] has calculated the slope T as a function of h . He used our data [9] for the relation of the crystal size as a function of the film thickness. His calculations are given in Fig. 8 for two values of the columnar separation ($d = 0.5$ nm and 2.0 nm).

Fig. 8. The slope of the perpendicular hysteresis loop (T) versus the layer thickness (h) for a 19 at% Cr layer. The squares are measured values from the hysteresis loop shearing. Open circles and triangles are calculated according to the columnar model of the authors for $d = 0.5$ and 2.0 nm, respectively. The closed circles and triangles are calculated from Andrä's chain of columns model. The solid line is the calculated domain-wall model.

It can be concluded that the behaviour $T(h)$ gives a better fit to the experimental values (squares). In comparison the calculated dependence (solid line) for a pure stripe-domain structure, according to the Kooy and Enz model [30], is also presented in Fig. 8. It is shown that these values are close to the calculated chain of columns values and both are in good agreement with our experimental data.

Despite the consideration that the quantitative results from the stripe-domain theory are controversial in our case because CoCr layers have a high coercivity, we reported [11, 16] calculations concerning the domain-wall energy, the exchange constant, and the domain-wall thickness. We found values [11, 16] with those usually found for common materials. On the basis of domain-wall calculations Hubert [31] confirms the domain model for CoCr layers.

6.2 Coercivity

The other point of study is the coercivity. We have measured the perpendicular coercivity field as a function of the thickness [11, 26]. H_c exhibits a maximum which depends on the chemical composition [11] and the deposition method [26]. The experimental values of H_c for our films are small as compared with the intrinsic anisotropy field H_{kt} and cannot be explained by thermal perturbations as shown above.

As a consequence of the difference between the volume and surface coercivity [26] the magnetization cannot be homogeneous throughout the film during all stages of the hysteresis curve. The H_c of the initial layer (substrate side) is found to be lower (approximately by a factor 4) regardless of the film thickness. In the case of rf sputtered films we found a critical thickness of around 120 nm. Above this thickness the surface volume coercivity ratio is smaller and below it is larger than one. If we combine this result with the experimental data measured by neutron depolarisation [25] we arrive at a model of branched domain structure of reversed domains within the main ones.

Honda et al. [32] have shown that the angular dependence of the coercivity can be well explained on the basis of the domain-wall model.

Finally, on the basis of the experimental and calculated results we can conclude that there is no significant influence of the columnar shape anisotropy on the total anisotropy. The other two arguments (shearing and coercivity behaviour) are more complicated and are not at present completely understood. Further experimental work will have to be carried out to verify our preliminary results.

In accordance with Andrä and co-workers we can summarize our conclusion that a strict particulate model cannot account for the magnetic properties of our CoCr layers and that a domain-wall model offers a better alternative for the interpretation of the magnetic behaviour of CoCr as proposed in [11].

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