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Oblique evaporation of $Co_{80}Ni_{20}$ part I: Fixed angle of vapour incidence

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Abstract--- This paper discusses the effect of an increase in layer thickness (from 40 nm to 1.7 μm) and an increase in deposition rate (from 0.5 to 20 nms^{-1}) on the structure and magnetic properties of $Co_{80}Ni_{20}$ thin films deposited at incidence angles between 70° and 80° (with respect to the substrate normal). The coercivity and anisotropy increase and the anisotropy in electric resistance decreases with increasing layer thickness up to a thickness of 150 nm. In all samples strong stripe domain structures were observed. The equilibrium domain period increases with increasing layer thickness. An increase in rate of evaporation from 0.5 to 20 nms^{-1} increases coercivity and anisotropy.

INTRODUCTION

The process of oblique evaporation offers unique possibilities to modify the properties of thin films. One of the applications of the oblique deposition method is the production of Metal Evaporated Tape. In part II of this paper [1] the first results with laboratory scale preparation of Metal Evaporated Tape using the method of Continuous Vapour Incidence are discussed. In this paper thin $Co_{80}Ni_{20}$ films deposited at fixed angle of incidence and without any addition of process gasses will be treated.

Surface diffusion is the key parameter governing the nucleation and growth process of thin films. The mean diffusion length of the adatoms depends on the film surface temperature, deposition rate and contamination with background gasses. For the oblique deposition process a directional component is added to the random movement of the adatoms due to the conservation of parallel momentum [2]. According to Hara *c.s.* the balance between the directional and random component determines the columnar inclination [3]. If the directional component is negligible in comparison with the random component, for instance due to substrate heating, the columns grow in the direction of the vapour source. When the random component decreases, the directional component deviates the columnar growth towards the film normal.

The amount of directional surface diffusion is determined by the kinetic energy of the arriving vapour atoms. Since the escape energy for atoms leaving the source is practically constant over the range of source temperatures used for evaporation, the directional component of surface diffusion is expected to be independent on the deposition conditions. Consequently only the random component of surface diffusion needs to be considered. The model suggested by Hara *c.s.* therefore predicts that with decreasing random surface diffusion the columns grow more towards the film normal. This rule is in agreement with a large number of observations (e.g. [4]) of the relation between columnar inclination and process parameters such as background pressure, rate and film temperature. It is found that especially the role of contamination with background gasses such as H_2O, N_2 and O_2 is of importance. H_2O molecules, with a small desorption energy, seem to have an increasing effect on surface diffusion whereas O_2 molecules, with a high desorption energy, seem to decrease surface diffusion.

The effect of layer thickness on structure and magnetic properties of obliquely evaporated thin films has not yet been studied in detail. Since there is a tendency for a decreasing layer thickness of recording media, the effect of layer thickness is of importance. Therefore a series of $Co_{80}Ni_{20}$ films evaporated at grazing incidence and with thickness ranging from 40 nm to 1.7 μm was prepared and investigated (§ II). In preparation of the evaporation of $Co_{80}Ni_{20}$ on tape using the Continuous Vapour Incidence method the effect of an increase in rate was shortly investigated for our evaporation set-up. The results will be discussed in § III of this paper.

I EXPERIMENTAL

The $Co_{80}Ni_{20(at\%)}$ films were prepared on 1x1 cm^2 Si and glass substrates mounted under angles of 70°, 75° and 80° (angle measured between the substrate normal and vapour incidence direction). Two evaporators were used. The 70° and 80° series were prepared in a 19 litre Seacom evaporator equipped with a oil-diffusion pump and Al_2O_3 Sylvania Joule-sources. The distance between source and substrates was 17 cm. The background pressure was typically 0.03 mPa increasing to 0.3-1 mPa during evaporation. The vacuum system used for the 75° samples was a 200 litre LH560 equipped with a turbo-molecular pump and cold finger featuring a background pressure of typically 0.01 mPa increasing to 0.1-0.2 mPa during evaporation. The samples were mounted at a distance of 23 cm from the 6 kW e-beam evaporation source with 30 mm carbon crucible. No sample heating or cooling was applied. The increase in temperature of the sample holder during evaporation is negligible. All samples were cleaned using an acid cleaning procedure. The angle of incidence was checked by measuring the shadowing region on a sample with an artificial ramp. The layer thickness was determined by a surface profiler.

The samples were investigated by cross-section TEM, Vibrating Sample Magnetometer measurements in three directions: perpendicular to the film plane (perpendicular) and in-plane both parallel (parallel) and perpendicular to the vapour incidence direction (transverse), torque magnetometer measurements in three orthogonal planes, resistance measurements both in parallel and transverse direction and domain observations by Magnetic Force, Kerr, Bitter and TEM-Lorentz microscopy.

II EFFECT OF LAYER THICKNESS

Films with increasing layer thickness were prepared at 70° and 80° in the Seacom evaporator and at 75° in the LH560. The vapour flux was kept constant resulting in a decrease in deposition rate from 0.2 down to 0.1 nms^{-1} with increasing angle of deposition.

Morphology

A 1.1 μm 70° film was investigated by cross section TEM (see for instance Figure 4). Clearly a tilted columnar (or fibre) structure could be observed with an average column diameter of less than 5 nm and an inclination of 57° (±3°) with respect to the substrate normal. Comparison between samples prepared at normal and oblique incidence leads to the conclusion that the film density is about 70%. The ratio between the apparent saturation magnetisations (M_s^*) of the oblique and normal incidence samples is slightly lower, about 60%. This might indicate oxidation of the obliquely deposited layer.

Diffraction studies on the cross section revealed a hcp structure with a weak preferred orientation. The average c-axis direction is inclined at an angle of 44° ± 20° with respect to the film normal. Next to the hcp phase a fcc phase was found with the lattice parameter of fcc Co or Ni (which are equal). This fcc phase could however also be caused by stacking faults in the hcp structure. The oxide in the film is cubic Co oxide (Co_3O_4).

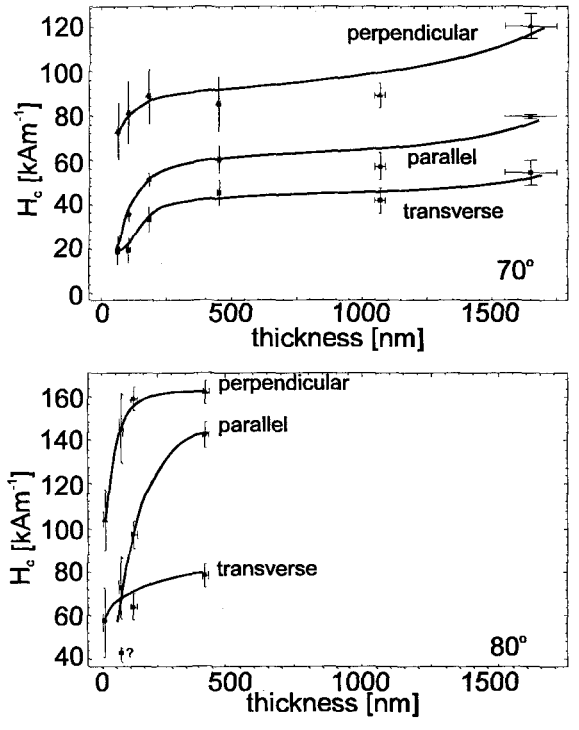

Figure 1 Coercivity H_c versus layer thickness for $Co_{80}Ni_{20}$ films evaporated at 70° (top) and 80°.

Hysteresis

The shape of the hysteresis loops does not change with increasing layer thickness, apart from an increase in coercivity which is shown in Figure 1 for the 70° and 80° series. The coercivity in all directions increases with increasing layer thickness, the changes are more pronounced for the 80° series. The measured magnetisation M_s^* was independent on film thickness and varied from 0.66, to 0.60 and 0.45 (±0.08) MAm^{-1} for the 70°, 75° and 80° samples respectively.

To investigate the origin of the increase in coercivity, torque measurements were performed.

Anisotropy

The torque measurements were corrected for the finite field error by extrapolation to infinite field of Fourier coefficients measured at different field values. The three measurements result in six coefficients from which two anisotropy constants (K_{mo}, K_{mt}) and an angle (ψ_{mo}) are extracted. K_{mo} is the energy difference when rotating the magnetisation in the vapour incidence plane, the minimum energy (easy axis) is obtained at ψ_{mo} (measured from the film normal) and K_{mt} is the energy difference obtained when rotating the magnetisation in the plane perpendicular to K_{mo} (see inset Figure 2). Correction for the thin film demagnetisation was made by using a mean field theory giving the intrinsic anisotropy constants K_o, K_t and easy axis inclination ψ_o:

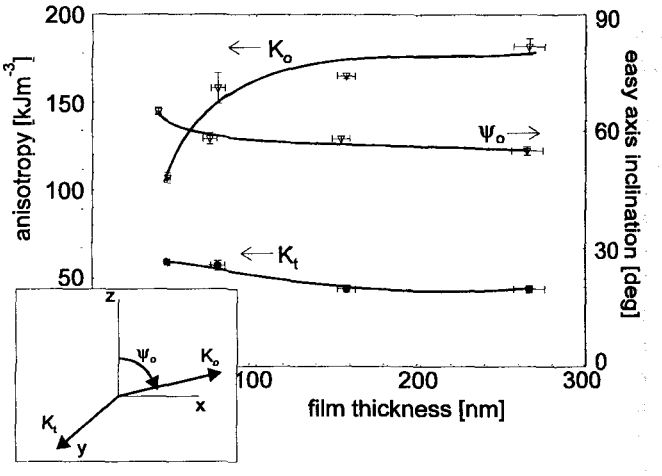

Figure 2 Intrinsic anisotropy constants K_o, K_t and easy axis inclination ψ_o. (Inset: Main axes orientations, plane xy represents the film plane and xz the vapour incidence plane).

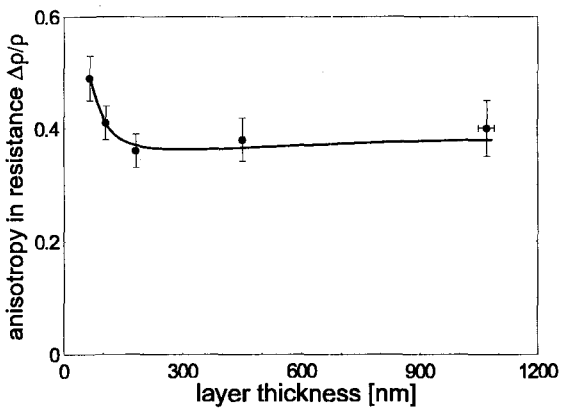

Figure 3 Anisotropy in resistance for $Co_{80}Ni_{20}$ films evaporated at 70°.

$$\psi_o = \tfrac{1}{2}\arctan\left(\frac{K_{mo}\sin(2\psi_m)}{K_f + K_{mo}\cos(2\psi_m)}\right) \quad [^\circ]$$

$$K_o = K_{mo}\frac{\sin(2\psi_m)}{\sin(2\psi_o)} \quad [Jm^{-3}] \quad (1)$$

$$K_t = K_{mt} - \tfrac{1}{2}\left(K_f + K_{mo} - K_o\right) \quad [Jm^{-3}]$$

where K_f is the shape anisotropy of a thin film with apparent saturation magnetisation M_s^*, $^1/_2\ \mu_o(M_s^*)^2$. All values are given with respect to the geometric volume of the film. (Note that the index m indicates the anisotropy constants as measured, when index m is omitted the anisotropy corrected for the thin film demagnetisation is indicated.) Figure 2 shows thickness dependence of the intrinsic anisotropy on the 75° series. With increasing film thickness the out-of-plane anisotropy increases and turns slightly out of the film plane. The increase in anisotropy is consistent with the observed increase in coercivity (Figure 1).

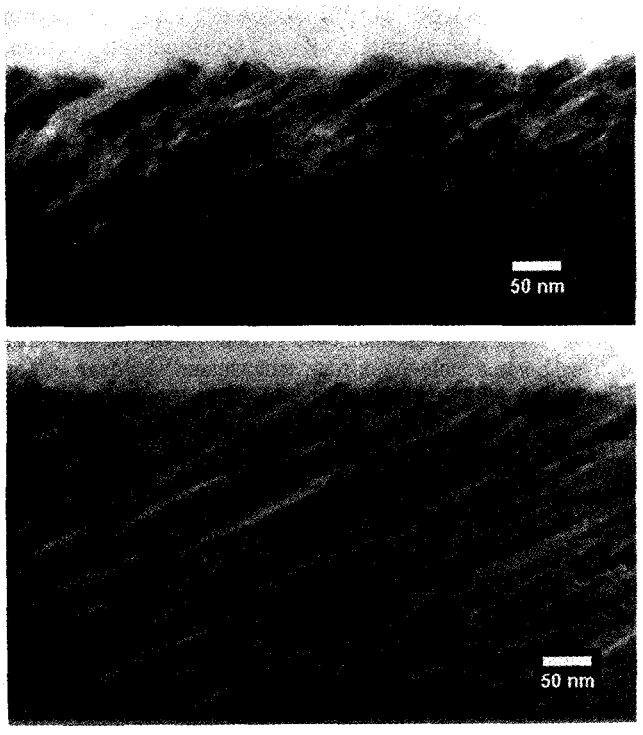

Figure 4 Cross section TEM of a $Co_{80}Ni_{20}$ layer evaporated at 75° at 0.5 nms^{-1} (top) and 20 nms^{-1} (bottom).

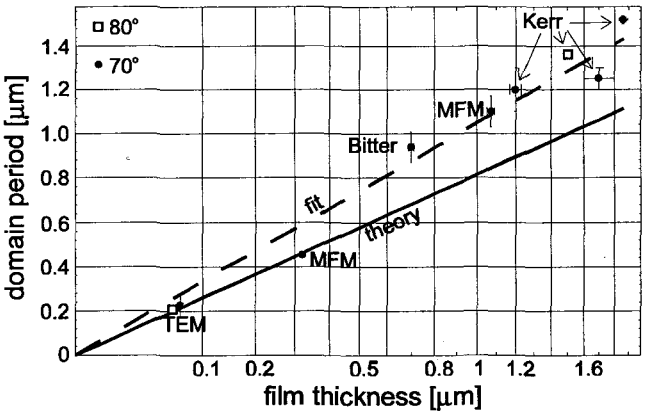

Figure 5 Equilibrium domain period versus film thickness.

Resistance

In search for the origin of the increase in anisotropy the resistance of the 70° samples was measured in parallel and transverse direction. The resistance measured in parallel direction is always larger. Figure 3 shows the difference between the specific resistance in parallel and transverse direction divided by the average specific resistance. If the increase in coercivity and anisotropy is caused by an increase in columnar separation in parallel direction, one would expect to find an increase in anisotropy of resistance too. The opposite observation however is made, from which one might conclude that the increase in anisotropy could have another origin.

Domain structure

In all samples strong stripe domain structures were observed, even in the high coercivity 80° samples. Figure 5 shows the change in equilibrium domain period (measured in transverse direction) versus layer thickness. Also shown is a calculation of the equilibrium domain period W_{eq} which uses the stray field free model proposed by Aitlamine [5]:

$$W_{eq} \approx 2\sqrt{\frac{2\sigma_B}{(K_o - K_t)\cos(\psi_o)}}\sqrt{d} \quad [m] \quad (2)$$

where σ_B is the Bloch wall energy $4\sqrt{AK_O}$ [Jm^{-2}] (A being the (bulk) exchange constant [$Jrad^{-2}m^{-1}$]).

The measured domain period lies 30-40% above the prediction. For an accurate prediction stray fields should be included, for instance in a micromagnetic model [6].

III EFFECT OF RATE

In preparation of the Continuous Vapour Incidence set-up [1] the rate of evaporation was increased. Therefore a short investigation on the effect of rate on film structure and magnetic properties was made. Two samples were prepared at 0.5 nms^{-1} (thickness 245 nm) and 20 nms^{-1} (thickness 310 nm) at an incidence angle of 75°.

Morphology

Figure 4 shows the cross section TEM images of the low and high rate sample. The columnar inclination β (Table 1) is approximately equal for both rates. If one identifies the width of the regions with identical contrast as the width of one single column (or fibre) relatively thick columnar diameters are found: 18±4 nm for the low rate and 11±3 nm for the high rate sample.

Hysteresis

Table 2 lists the coercivity H_c, squarness S and saturation magnetisation M_s^* of both samples. The increase in rate of deposition strongly increases the coercivity in all directions but does not affect the saturation magnetisation. The squarness in parallel and perpendicular direction increase whereas the squarness in transverse direction decreases.

Anisotropy

The intrinsic anisotropy constant K_o strongly increases with increasing rate which is accompanied by a slight increase in intrinsic easy axis inclination (Table 1). This probably is the reason for the observed changes in squarness (Table 2). Since the value of M_s^* and columnar inclination β do not change considerably, it is unlikely that the increase in K_o is caused by an increase in shape anisotropy. It is more likely that the increase in intrinsic anisotropy is caused by an increase in crystal anisotropy. This however remains to be verified.

Surface diffusion calculations [7] show that the increase in rate mainly decreases the contamination with H_2O molecules during film growth. At 20 nms^{-1} the contamination with background gasses is negligible. How the decrease in H_2O contamination could influence texture formation is unknown.

Table 1

Rate [nms^{-1}]	β (±3°)	K_o [kJm^{-3}] (±10)	K_t [kJm^{-3}] (±5)	ψ_o (±2°)
0.5	58°	147	42	51°
20	62°	213	26	56°

Table 2

Rate [nms^{-1}]	M_s^* [Mam^{-1}] (±0.05)	$H_{c,per}$ [kAm^{-1}] (±8)	$H_{c,par}$ [kAm^{-1}] (±8)	$H_{c,tra}$ [kAm^{-1}] (±8)	S_{per} (±0.01)	S_{par} (±0.01)	S_{tra} (±0.01)
0.5	0.53	121	76	43	0.21	0.74	0.43
20	0.55	199	118	60	0.31	0.94	0.33

CONCLUSION

For the observed films the coercivity and anisotropy increase with layer thickness and the anisotropy in resistance decreases up to a layer thickness of about 150 nm. Above this thickness these values remain constant. It is unlikely that this effect is caused by a change in deposition conditions during film growth. No direct proof of an initial layer has however been found. The origin of the change in film properties with film structure therefore remains uncertain.

The equilibrium domain period of the strong stripe domains that have been observed in all films increases with increasing film thickness. Stray-field free domain calculations underestimate the domain period by 30-40%. For an accurate prediction of the equilibrium domain period (micromagnetic) models including stray fields should be used.

An increase in rate of evaporation from 0.5 to 20 nms^{-1} increases coercivity and anisotropy. Since the columnar inclination is not affected, the observed increase in coercivity and anisotropy is probably not caused by a change in morphology but by a change in film texture.

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