Structural, static and dynamic magnetic studies of evaporated Co$_x$Cr$_{1-x}$/Si (100) and Co$_x$Cr$_{1-x}$/glass thin films.

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Abstract.

We have evaporated a series of Co$_x$Cr$_{1-x}$ thin films under vacuum onto Si (100) and glass substrates, with a perpendicular incidence. The thickness of the magnetic layer ranged from 17 to 220 nm, and the content chromium, from 0.12 to 0.20, values determined by means of Rutherford Backscattering Spectrometry (R.B.S.) spectra using SIMNRA programme. Microscopic characterizations of the films were done with X-ray diffraction (XRD) measurements and infer that all the samples were polycrystalline, with an hcp structure and show a <0001> preferred orientation, and with the grain size increasing with the chromium content decrease. Atomic force microscopy (A.F.M.) observations reveal very smooth film surfaces. The static and dynamic magnetic properties have been investigated by means of Alternating Gradient Field Magnetometer (A.G.F.M.), and Brillouin Light Scattering (B.L.S.) measurements. The saturation magnetization $M_s$ was found to decrease from 1200 emu/cm$^3$ to 220 emu/cm$^3$ as the chromium content increases from 12% at. to 20% at., whatever the thickness is.

From the fit of the B.L.S. spectra, we have computed effective magnetic anisotropy factors, as well. All the results are discussed and correlated.

Keywords: CoCr system; thin films; Hysteresis; Magnetic anisotropy; Brillouin Light Scattering (B.L.S.).

P.A.C.S.: 81.30.Dz; 75.70.-i; 75.60.-d; 75.30.Gw; 78.35.+c.

1. Introduction

Co and CoCr thin films have been intensively studied. They are the dominant materials media designed for magnetic longitudinal recording. As promising candidates for the perpendicular recording media, the CoCr thin films have got much attention [1-10]. To achieve the perpendicular magnetic anisotropy, the anisotropy constant $\textbf{K}$ has to be greater than the demagnetizing energy. This depends, of course, on the comprehension of the microstructural and the magnetic properties, as well as on the performance of the fabrication techniques of the thin films media.

In this contribution, we study the effect of thickness of the magnetic layer on the magnetic properties of CoCr films, deposited onto Si and glass substrates.

The thickness measurements and atomic composition of the samples were performed by Rutherford Backscattering Spectrometry (RBS) technique (Section 3.1). The structural properties and film morphologies were investigated by x-ray diffraction (XRD) and atomic force microscopy (AFM). The static magnetic properties have been performed by alternating gradient field magnetometer (AGFM) (Section 3.2.1) experimental technique. The dynamic magnetic properties have been performed by Brillouin Light Scattering (B.L.S.) technique (Section 3.2.2.).

2. Experimental procedures

Two series of Co$_x$Cr$_{1-x}$ thin films were grown onto Si (100) and Corning glass substrates, under the same conditions. The CoCr samples have been prepared, at room temperature, by thermal evaporation,
under vacuum, from a CoCr (70%at.-30%at) powder to which we added a quantity of Co powder of purity 99.99%. The pressure was $10^{-7}$ mbar before deposition; during the evaporation the vacuum achieved was better than $2 \times 10^{-6}$ mbar.

The thickness measurements and the atomic composition of the samples were achieved by means of Rutherford Backscattering Spectrometry (R.B.S.) spectra using SIMNRA programme.

Each series consists of several samples with thickness ranging from 17 to 220 nm, and chromium content ranging from 12 at. % to 20 at. %.

The structural properties were monitored by x-ray diffraction, using a Siemens D-500 diffractometer with $\lambda = 1.789$ Å.

To study the macroscopic magnetic properties of the Co$_x$Cr$_{1-x}$ thin films, hysteresis loops were recorded by an Alternating Gradient Field Magnetometer (A.G.F.M.), with the external magnetic field $H$ applied both perpendicular (polar configuration) and parallel to the film plane (longitudinal configuration). For the longitudinal configuration, the measures have been done applying the magnetic field in two different directions in order to study the plane magnetic anisotropy.

Brillouin Light Scattering (B.L.S.) experiments were used to investigate the magnetic anisotropy in Co$_x$Cr$_{1-x}$/Si samples. The BLS measurements were done using a (2x3)–pass tandem Fabry-Perot interferometer. The samples were illuminated by a single-mode Ar$^+$ ion laser at the wavelength of $\lambda = 5145$ Å, with an incident power of 100 mW. The value of the transferred in-plane wave vector $q_{||}$ is connected to $\theta$ by the relation $q_{||} = (4\pi/\lambda) \sin \theta$. The experiments were done with different applied magnetic values in the 0 to 5 kOe range.

Finally the state of the surface of these Co$_x$Cr$_{1-x}$ films has been investigated by tapping mode atomic force microscopy (A.F.M.), using a Veeco 3100 apparatus.

3. Results and discussion

3.1 Structural and morphological characterizations

Many results concerning these Co$_x$Cr$_{1-x}$ films have been published elsewhere [10, 11] and are just summarized here. Table 1 gives the thickness and the composition of the samples, values determined by Rutherford backscattering (R.B.S.) spectrometry, using a 2MeV monoenergetic He$^+$ incident beam, and a backscattering angle equal to 160°. Figure 1 shows a specimen of the experimental and the simulated spectra; for the latter we used SIMNRA program [12, 13].

| Thickness (nm) | 17  | 19  | 44  | 56  | 70  | 215 | 220 |
|---------------|-----|-----|-----|-----|-----|-----|-----|
| Cr (at. %)    | 18  | 18  | 12  | 20  | 12  | 15  | 20  |
| Co (at. %)    | 82  | 82  | 88  | 80  | 88  | 85  | 80  |

Figure 1a shows the experimental (dotted curve) and simulated (solid curve) RBS spectra for a sample of thickness 17 nm.

The big peak corresponds to Co atoms and the small peak to Cr ones. The difference in the height between the two peaks is due, of course, essentially to the small amount of the Cr atoms in the composition of the CoCr sample.

In Figure 1b we observe a small overlapping of the chromium’s peak with the cobalt one, for this film of 44 nm thickness.
Figure 1: Example of RBS spectra for samples of thickness 17 nm and 44 nm.

The x-ray diffraction patterns of CoCr/glass (not shown here) are very noisy, the reflections being weak and very broad. Nevertheless, the Bragg hexagonal close packed (hcp) peak (00.2) is clearly identified but its intensity is drastically reduced, the grain sizes being very small and their orientation random.

Examples of x-ray diffraction spectra are presented in figure 2 for CoCr/Si for t = 17 nm (the thinnest film), t = 44 nm (intermediate thickness) and t = 215 nm (among the thickest ones). All the spectra are characterized by three Bragg peaks, corresponding to the diffraction angles \( \theta \) equal to 24.4°, 26.3° and 27.9° (i.e.2\( \theta \) = 48.8, 52.5, and 55.8°) and the diffraction planes with “Bravais-Miller” indices (10.0), (00.2) and (10.1), respectively.

It is worth noting the great intensity of the hcp Co (00.2) Bragg peak, for all samples, which infer that the films crystallize in the hcp structure, are polycrystalline and with the (0001) texture. This undoubtedly indicates the preferential orientation of the crystal plane in parallel with the substrate surface.

The values of the lattice constants \( c \) and \( a \), derived from the x-ray spectra and computed for different thicknesses and crystallographic orientations, range from 2.491 Å (for t = 70 nm) to 2.498 Å (for t = 17 nm) for \( a \), and from 4.059Å (for t = 17nm) to 4.069Å (for t= 70 nm) for \( c \). Compared to the bulk value parameters (\( a = 2.51 \) Å and \( c = 4.07 \) Å) these lattice parameters infer that the thinnest films are under a compressive stress.

Furthermore, table 2 displays the grain sizes computed using the x-ray diffraction patterns and the Scherer formula [14]:

\[
L = \frac{\lambda}{\Delta \theta \cos \theta}
\]

where \( L \) is the grain size for a grain with a particular orientation, \( \lambda \) the x-ray wavelength, \( \theta \) the diffraction angle and \( \Delta \theta \) the width at half height of the Bragg peak corresponding to this particular
orientation. Table 2 gives the evolution of the grain size vs. the thickness $t$ of the magnetic layer, for the three Bragg peaks present in the x-ray spectra. For the three Bragg peaks present, namely (10.0), (00.2) and (10.1), respectively, the most intense Bragg peak, which corresponds to hcp (00.2) peak of cobalt, shows the greater grain size (49, 70 and 70 nm) while the other two peaks display grain sizes with an average value of 20 nm.

Table 2. Grain size vs. thickness.

| $t$ (nm) | 17 | 44 | 215 |
|----------|----|----|-----|
| (hk.l)   | (10.0) | (00.2) | (10.0) | (00.2) | (10.0) | (00.2) | (10.0) |
| L (nm)   | 26  | 49  | 20   | 19   | 70   | 13    | 16    |

For the most intense peak, a slightly increase of the grain size is noticed between the thinnest film, with $t=17$ nm, and the other films where the grain size is almost constant. Other researchers have reported an increase of the grain size with thickness [9, 15].

Moreover, topographic images given by the atomic force microscope (AFM) in the tapping mode reveal very smooth surfaces for most of the films, the root mean square (rms) roughness amplitude ranging from 0 (within the measures uncertainty) to 18 Å, the roughest film being the 70 nm thick film, and the 17 nm thick (the thinnest one) film displaying a very smooth surface (see table 3). So, no clear dependence of the roughness vs. the thickness of the magnetic layer is noticed. The AFM scans of some samples (among the thinnest ($t = 17$ nm) and the thickest ($t = 215$ nm) films) are presented in figure 3. Scan area is $5 \times 5 \mu m^2$ in all cases.

![Figure 2 Examples of x-ray diffraction spectra for CoCr/Si for $t = 17$ nm (the thinnest film), $t = 44$ nm (intermediate thickness) and $t = 215$ nm (among the thickest ones).](image.png)
Table 3. Root-mean-square (rms) values of surface roughness as a function of thickness for Co$_x$Cr$_{1-x}$ thin films.

| t (nm) | 17  | 18  | 44  | 56  | 70  | 215 | 220 |
|-------|-----|-----|-----|-----|-----|-----|-----|
| rms (Å)| ----| 15  | ----| 14  | 18  | ----| 11  |

Figure 3: 2D ((a) and (c)) and 3D ((b) and (d)) surface topography of CoCr thin films as observed by tapping mode AFM. Thicknesses are 18 nm (a,b) and 215 nm(c,d). Scan area is 5 x 5 µm$^2$ in all cases.

3.2. Magnetic characterizations

3.2.1 Static magnetic characterizations. In order to study the macromagnetic properties of the films, magnetic field dependence of magnetization curves are recorded, at room-temperature, using an alternating gradient field magnetometer (AGFM). The hysteresis loops depicted in figure 4 for two representative CoCr films, 18 nm thick, either on Si (100) and Corning glass substrates, are plotted both in the longitudinal as well as in the polar configurations.

From this loops it can be noticed clearly that the easy axis lies in the film plane: the longitudinal configuration is characterized by a quite square (rectangular) loops implying an easy axis lying in this direction, whereas the polar configuration, on the other hand, shows a small hysteresis and an extremely reduced remanent magnetization. Moreover, the loops in the polar configuration, representing the hard direction, is characterized at intermediate fields by a wide linear portion,
followed at greater fields by a curvature going on up to the saturation. We may deduce at once that the demagnetizing field exceeds the anisotropy constant $K_a$.

![Graph showing hysteresis loops for 17 nm CoCr/Si and CoCr/glass thick films.](image)

**Figure 4.** Hysteresis loops for 17 nm CoCr/Si and CoCr/glass thick films. The external magnetic field is applied both in the longitudinal configuration ($H_{||}$) and the polar configuration $H_{\perp}$.

**Table 4.** Values of saturation magnetization $M_s$.  

| $t$ (nm) | 17  | 18  | 44  | 56  | 70  | 215 | 220 |
|---------|-----|-----|-----|-----|-----|-----|-----|
| Cr (% at.) | 18  | 18  | 12  | 20  | 12  | 15  | 20  |
| Si      | 700 | 700 | 1200| 220 | 1200| 870 | 220 |
| glass   | 500 | 500 | 900 | 170 | 900 | 840 | 170 |

No substantial change in the hysteresis loops was observed when rotating the substrate plane comparative to the applied field direction, indicating the absence of in-plane magnetic anisotropies.

On another hand, using the magnetization curves and the volume of the samples, we compute the saturation magnetization values. Table 4 displays value of $M_s$ as a function of thickness and chromium content, the most prevailing factor being the Cr proportion. $M_s$ values range from 220 emu/cm$^3$ for the Co$_{88}$Cr$_{20}$/Si, with $t = 220$ nm to 1200 emu/cm$^3$ for Co$_{88}$Cr$_{12}$/Si with $t = 44$ nm. For the glass substrate, the $M_s$ values are somewhat smaller, ranging from 170 emu/cm$^3$ for Co$_{88}$Cr$_{20}$/glass.
with $t = 220$ nm to 900 emu/cm$^3$ for Co$_{88}$Cr$_{12}$/glass with $t = 44$ nm. So, the effect of substrate on the saturation magnetization is significant.

Several authors found a similar dependence of $M_s$. Shun-ichi Iwasaki [2] reported values of $M_s$ ranging from 300 emu/cm$^3$ to 700 emu/cm$^3$, when the percentage of chromium varies between 17 at.% Cr and 13 at.% Cr. J. Suzuki et al. [16] reported values of $M_s$ ranging from 300 emu/cm$^3$ to 500 emu/cm$^3$, when the substrate temperature ($T_s$) rises from room temperature to 300°C, in their samples, prepared by RF magnetron sputtering. T. Takanashi et al. [17] have prepared Co$_{81}$Cr$_{19}$/Al multilayered films on glass-slide substrate at room temperature by facing targets sputtering (FTS) apparatus. They noticed a significant dependence of $M_s$ on the thickness of Co$_{81}$Cr$_{19}$ layer and reported a maximum value of 660 emu/cm$^3$ when this thickness was 84 Å.

Figures 5 and 6 display the in-plane squareness, ratio of the remanent magnetization to the saturation magnetization $S = M_r/M_s$, and the coercive field $H_c$, vs. thickness of the magnetic layer, for both the substrates.

The same dependence of the squareness $S$ on thickness is observed for both Si and glass substrates, the difference existing just on the amount of the value of this squareness. The thinnest film having the highest value, $S = 0.83$ and 0.67, for Si and glass, respectively.

As for $H_c$, it is observed from figure 6 that the in-plane coercive field for CoCr/glass films decays with increasing thickness, following a law of the type $1/t^2$, reflecting a strong dependence on the surface effects. Moreover, for the CoCr/Si films, this behaviour is noticeable for films thicker than 70 nm. For films thinner than 70 nm, the relatively smaller values of $H_c$ and their dependence on thickness may be related to the surface roughness. Indeed, the presence of roughness being directly associated to the tridimensional growth of a polycrystalline structure, possessing a great quantity of defects which work as pinning centres for domain walls, so, as the surface roughness values for the 17 nm and the 44 nm thick films are much lower than the 70 nm thick film one, a decrease of the coercive field for these films is observed comparatively to $H_c$ for thicker films.
3.2.2 Dynamic magnetic characterizations. In the last decade, Brillouin light scattering has been used accurately to investigate magnetic materials [18-21]. Brillouin light scattering (BLS) experiments were performed on the CoCr/Si series under the conditions described in Section 2. In Figure 7, we show examples of experimental and calculated Brillouin spectra obtained with $H = 1 \text{kOe}$ for CoCr(56nm)/Si sample, and with $H = 5 \text{kOe}$ for CoCr(215nm)/Si sample. The peak with the highest intensity in the Stokes (low frequency) side of the spectra corresponds to the Damon-Eshbach (DE) surface spin wave. The DE [22] mode only appears in one side of the spectra because of its non-reciprocal nature and because the film is sufficiently thick that the light cannot interact with the DE mode localized on the lower surface of the film.

The adjustment of the theoretical and experimental spectra have been performed (taking $4\pi M_{\text{eff}} = 1.02 \ T$, for CoCr (56nm) and $4\pi M_{\text{eff}} = 1.05 \ T$ for CoCr (215nm), and the published values of the splitting factor and the spin wave exchange constant ($g = 2.16$, and $D = 2.6 \times 10^{-9} \text{erg.cm}^2$) and a negligible magnetocrystalline anisotropy.

Figure 8 displays the evolution of the frequency shift versus the applied magnetic field (for the Damon-Eshbach mode). This figure represents the experimental data ($\bullet$, $\Delta$) and the theoretical fit (—) computed for the dipolar mode using the dispersion relation:

$$\frac{\omega}{2\pi} = \frac{\gamma}{2\pi} \left\{ H(H + 4\pi M) + (2\pi M)^2 (1 - e^{-2q_\parallel t}) \right\}^{1/2}$$

where $f$ is the frequency in GHz, $\gamma$ is the magnetogyric ratio, related to the g factor (a value of 2.16 was taken for the splitting factor), $q_\parallel$ is the transferred in-plane wave vector $q_\parallel = 1.73 \times 10^5 \text{cm}^{-1}$ (for $\theta = 45^\circ$, in this experiment).

Finally, the study of the variation of the Damon-Eshbach mode frequency versus applied magnetic field intensity (Figure 8) infers that the magnetic parameters determined for the Co$_x$Cr$_{1-x}$/Si(100) investigated samples are satisfactory for a large range of the applied magnetic field variation.
Figure 7: Examples of BLS spectra for CoCr(56nm)/Si sample, for applied field \( H = 1 \text{kOe} \), and CoCr(215nm)/Si sample for applied field \( H = 5 \text{kOe} \).

Figure 8: Damon-Eshbach mode frequency versus applied magnetic field intensity for the Co\(_x\)Cr\(_{1-x}\)/Si samples. Indications are inside the figure.
From the fit of the BLS spectra, we extracted values of the anisotropy field $H_a$ from which we computed the effective anisotropy magnetic factors $K_{\text{eff}}$, these constants being connected to the anisotropy fields by the relation

$$K_{\text{eff}} = \frac{1}{2} M_s H_a$$

where $M_s$ is the saturation magnetization of the film.

The $K_{\text{eff}}$ values range from $-0.7 \times 10^6$ to $-3.2 \times 10^6$ erg/cm$^3$. These negative values, in the whole range of the chromium content, infer that the easy magnetization axis lies in the surface of the film for all samples and corroborate the conclusion found by means of the magnetization curves study (see section 3.2.1).

Unfortunately, due to the low thermal conductivity of glass, it was not possible to perform BLS measurements on CoCr/glass samples in order to point out the substrate effect. The experiments has to be performed using very low illuminating powers (less than 40 mW instead of the 100 mW used for CoCr/Si) in order to avoid undesirable heating. So, due to this experimental difficulty, no conclusive spectra were obtained about CoCr/glass samples.

### 4. Conclusion

The structural and magnetic properties of the Co$_x$Cr$_{1-x}$ series prepared by thermal evaporation under vacuum onto Si(100) and glass substrates have been studied as a function of the magnetic layer thickness, ranging from 17 nm to 220 nm. In this range, it has been observed that $M_s$ decreases from 1200 emu/cm$^3$ to 220 emu/cm$^3$, for the CoCr/Si sample. The value of $M_s$ was smaller for the CoCr/glass correspondent samples. It was also observed that the coercive field decreases with the thickness increase for CoCr/glass films, following a $1/t^n$ law, and hence, reflecting a clear dependence on the surface effects, whereas this behaviour is available only for CoCr/Si films thicker than 70 nm. For thinner films, the in-plane coercivity may be correlated to the surface roughness of the films. The anisotropy field values deduced from the fit of the BLS spectra give negative values for the effective anisotropy factors which infer that the easy magnetization axis lies in the film plane for all the samples, result corroborated by the magnetization curves study.

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