Determination of Texture Orientation Related Magnetic Properties of Nickel-Cobalt Films

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The determination of texture effects in nickel-cobalt (Ni-Co) films with different thickness, which were obtained by electrodeposition, has been investigated by the measurement of hysteresis loops at different angles. Easy-axis distribution measurements were performed as a function of the squareness $M_\mathrm{p}(\beta)$ and the correlations were established among the different thicknesses. The composition of Ni-Co films was determined by energy dispersive X-ray spectroscopy. The structural analysis made by X-ray diffraction revealed that all films have a polycrystalline face-centered cubic structure but their texture degrees vary depending on the film thickness. The determination of the easy-axis orientation in 2-D films from the $M_\mathrm{p}(\beta)$ obtained by the hystersis loops was studied using Fourier series analyses. The coefficient A_0 have a value of less then unity while A_2 is inversely proportional to the width of the distribution function which may cause the change in the texture preferential orientations. Therefore, the differences observed in the magnetic easy-axis distributions were attributed to the changes in texture orientations caused by the compositional differences at different thicknesses of the polycrystalline films.

Key words: Texture Orientation; Magnetic Easy-Axis Distribution; Ni-Co Films; Fourier Series; Magnetic Properties.

1. Introduction

Magnetic thin films are important for read/write heads and micro electromechanical systems due to their capability, quality, and low cost [1]. Nickel-cobalt (Ni-Co) films have interesting magnetic properties and thus are used in magnetic recording applications [2]. The determination of the easy-axis distribution of a magnetic recording medium is essential to understand the switching process within the materials [3]. Knowledge of this distribution is also required for the interpretation of the magnetic measurements of the hysteresis loops. This is especially true for the systems of single domain particles. Therefore, the easy-axis distribution is referred to be an effective tool, which determines the other magnetic properties of the material [3 – 6].

El-Hilo et al. [4] developed a method to measure the magnetic easy-axis distribution in two-dimensional systems that is applicable to commercial tapes. The method provides a useful numerical quantity analysis of magnetic texture orientation. This study was based on an original three-dimensional technique by Shtrikman and Treves [5]. In this method, the distribution of easy axis is determined by measuring the hysteresis loop squareness M_p $(= M_\mathrm{r}/M_\mathrm{s})$ in the field distribution as function of angle β between the applied field and the texture direction. The angle β is varied by rotating the sample. The angle between the axis of a given particle and the overall texture direction is α, then El-Hilo et al. [4] give the following expression of the coefficients A_0 and A_{2n} for $f(\cos\alpha)$, the distribution of the easy-axis directions:

$$A_0 = \frac{1}{2\pi}\int_0^{\pi/2} M_\mathrm{p}(\beta)\mathrm{d}\beta, \tag{1}$$

$$A_{2n} = \sum_{n=1}^{\infty}\frac{(2n-1)(2n+1)}{\pi(1)^{n+1}}\int_0^{\pi/2} M_\mathrm{p}(\beta)\cos(2n\beta)\mathrm{d}\beta, \tag{2}$$

where $M_\mathrm{p}(\beta)$ is the squareness.

This expression can be written in the simplified form as:

$$f(\cos\alpha) = A_0 + \sum_{n=1}^{\infty} A_{2n}\cos(2n\alpha). \tag{3}$$

The coefficient A_0 for a non-interacting system should equal unity and is independent of the degree in the texturing whereas the series A_{2n} determine the degree of texture in the system. While $A_0 = 1$ is valid for all $n \geq 1, A_{2n} = 0$ stands for a random oriented system, whereas $A_{2n} = 2$ for the completed aligned system [3, 4, 6].

The magnetic properties of a film are influenced by the crystalline texture and also by the crystal orientations. The investigations on the electrodeposited Ni-Co films [7, 8] showed that their microstructure and properties strongly depend on the Ni : Co ratio in the films and hence the crystalline texture which can be controlled by the experimental parameters. It is seen that the film thickness among the parameters has also an effect on the microstructure and hence the magnetic properties of the films.

To our knowledge, there have not been significant studies on the film texturing, especially Ni-Co films, using Fourier analysis except the reports mentioned above [1 – 6]. Therefore, the aim of this study is to determine the effects of the film thickness on the texture of Ni-Co films electrodeposited on titanium (Ti) substrate by means of Fourier series analyses. The magnetic easy-axis distributions are discussed in terms of the variations in the crystal orientations occurring at different thicknesses, and the results of the Ni-Co films are compared with the cobalt-phosphorous (Co-P) [4] and barium-ferrite (Ba-Fe) media [6]. Systematic variations in the Fourier coefficients correspond to the magnetic properties that may be ascribed to associate with variations in the texture orientation with the change of the thicknesses of polycrystalline Ni-Co film.

2. Experimental

Ni-Co films were electrodeposited from the sulphate bath containing 0.4 M $NiSO_4 \cdot 7H_2O$, 0.1 M $CoSO_4 \cdot 7H_2O$, and 0.2 M H_3BO_3. The electrodeposition system consists of a potentiostat/galvanostat (EGG model 362) with three electrodes, a computer, and an electrochemical cell. Before deposition, the Ti substrate was first mechanically polished and then activated in 10 % H_2SO_4 and finally rinsed with distilled water. The polished Ti substrate was used as cathode whereas the anode was a platinum foil. The saturated calomel electrode (SCE) was used as a reference electrode. The Ni-Co films were deposited on the Ti substrate at -1.2 V vs. SCE and the electrolyte pH was 2.1 ± 0.1 for all films deposited during this investigation. After the deposition is completed, the films were mechanically peeled from their substrates and stored in proper conditions for characterizations.

The composition of electrolyte was analysed by a Perkin-Elmer Optima, 3100 XL inductively coupled plasma atomic emission spectrometry (ICP-AES). The film compositions were determined by energy dispersive X-ray spectroscopy (EDX) in the Zeiss Supra 50 Vp model scanning electron microscopy. The crystal structure of the deposits was determined by a Rigaku-rint 2200 X-ray diffraction (XRD) with Cu-K_α radiation. For a general pattern the range recorded was $2\theta = 40° - 80°$. Hysteresis loops of the films were measured at a saturated field of ± 10 kOe in the film plane using an ADE EV 9 model vibrating sample magnetometer (VSM). The sample rotation facility of the VSM was used to measure the variation of the loops with the angles between the applied field and the arbitrary chosen texture axis. All depositions and measurements were carried out at room temperature.

3. Results and Discussion

Ni-Co films were produced with the electrodeposition technique at 2 µm, 4 µm, and 10 µm thicknesses, respectively. It is found that the composition of the prepared electrolyte is consistent with the composition of the electrolyte analysed using ICP-AES. The film compositions, measured by EDX, are listed in Table 1. As seen in this table, the Co content in the films increases since Co is a more noble metal although the concentration of the electrolyte was kept constant. Consequently, the ratio of Ni to Co in the film slightly decreased as the film thickness increased.

The crystal textures of the films analysed by XRD measurement are shown in Figure 1. In the XRD patterns, all films have the (111), (200), and (220) reflections of face-centered cubic (fcc) crystalline structure clearly appearing at $2\theta \cong 44°$, 51°, and 76°, respectively. To assess the texture formation of films, the relative peak intensities of the reflections were considered and the normalized peak intensities are listed in Table 1. The strongest peaks in the pattern of the sample grown at low (2 µm) and high (10 µm) thickness are the (111) and (220) peak, respectively. As the film thickness increase, the (111) peak weakens and the (220) peak becomes preferred. Therefore, the film at low thickness has the (111) preferential orientation

Table 1. Compositional, structural, and magnetic properties, and Fourier coefficients of Ni-Co films.

Thickness (μm)	EDX (wt. %)		XRD (Relative integral peak intensities)	VSM ($\beta = 0°$)		Fourier Coefficients	
	Ni	Co	$I_{11}/I_{200}/I_{220}$	M_p	H_c(Oe)	A_0	A_{2n} ($n = 1$)
2	78.51	19.49	78/43/100	0.44	36.37	0.691	1.320
4	74.87	25.13	39/15/100	0.47	41.85	0.738	1.410
10	70.79	29.21	100/37/66	0.64	44.34	0.990	1.892

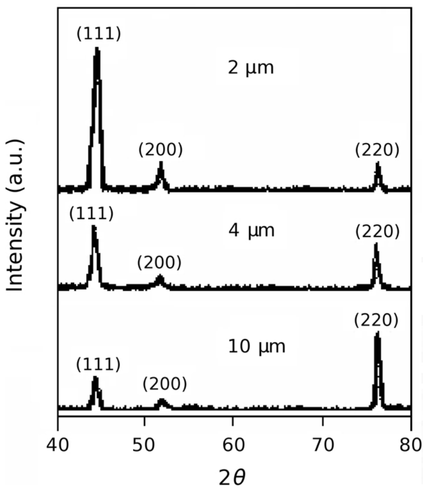

Fig. 1. XRD patterns of polycrystalline Ni-Co films.

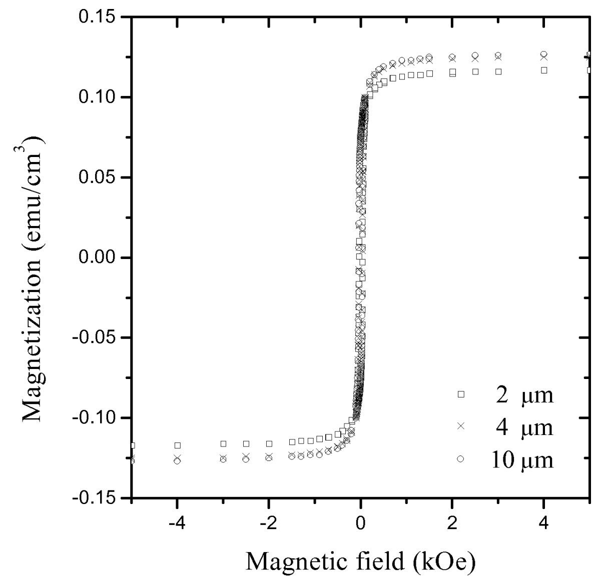

Fig. 2. Hysteresis loops of Ni-Co films measured at $\beta = 0°$.

whereas the (220) orientation was observed for the film grown at the high thickness. To the results obtained from EDX analyses, the crystalline preferential orientation turned from the (111) peak to the (220) peak with the decrease of Ni : Co ratio of the films caused by the film thickness. All samples have reflection of the fcc phase, which is thought to be most likely due to the high Ni content, and/or also the Co fcc phase occurred in the films.

The lattice parameters were found to be 0.3527 ± 0.0011 nm, 0.3530 ± 0.0012 nm, and 0.3536 ± 0.0011 nm for the samples prepared at 2 μm, 4 μm, and 10 μm thickness, respectively, using the least squares technique to fit the experimental data to a straight line [9]. These values are almost intermediate between the lattice parameters of Ni (0.3523 nm) and of Co (0.3544 nm) [9]. The errors in the lattice parameters are the standard errors, which were determined from the standard deviations from the slope. Significant differences in the lattice parameters are not observed and also near to the Ni lattice parameter. This most probably arises from the more Ni content than the Co in the samples, see EDX data in Table 1. The average grain sizes of crystallites determined using Sherrer's relation [9] sizes were calculated to be about 23 nm.

The squareness $M_p(\beta)$, obtained from the hysteresis loops, in arbitrary chosen direction was followed by the rotation of the sample through an angle β, which was incremented at 15° steps. As an example, the hysteresis loops measured at $\beta = 0°$ are illustrated in Figure 2. The $M_p(0°)$ values obtained from the loops are also listed in Table 1. The $M_p(0°)$ increased as the thickness increased, and a greater variations between the 10 μm film and others is observed. The consequences of the alignments seen in the coercivity H_c obtained from hysteresis loops at $\beta = 0°$ are also given in Table 1. The increase in coercivity may be due to differences in the intrinsic pigment coercivity with the increase of the thickness rather than changes due to the applied orientation field. Therefore, it can also be said that the variations in $M_p(\beta)$ seen in the Ni-Co films may be due to the differences in the intrinsic texturing. The original data for the variation of $M_p(\beta = 0° – 90°)$ for all films is plotted in Figure 3 indicating that all films are anisotropic with an easy-axis direction at $\beta = 90°$. The same trend for magnetic properties is also reflected at all angles $\beta = 15°, 30°, 45°, 60°, 75°$,

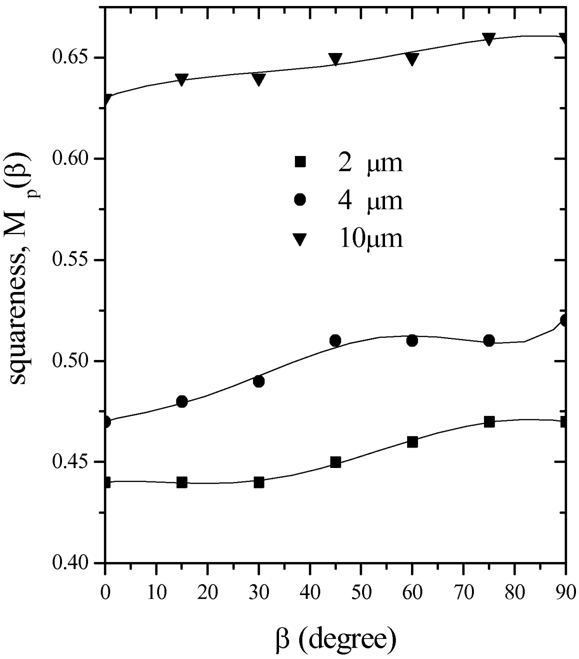

Fig. 3. Angular dependence of $M_p(\beta)$ of the films.

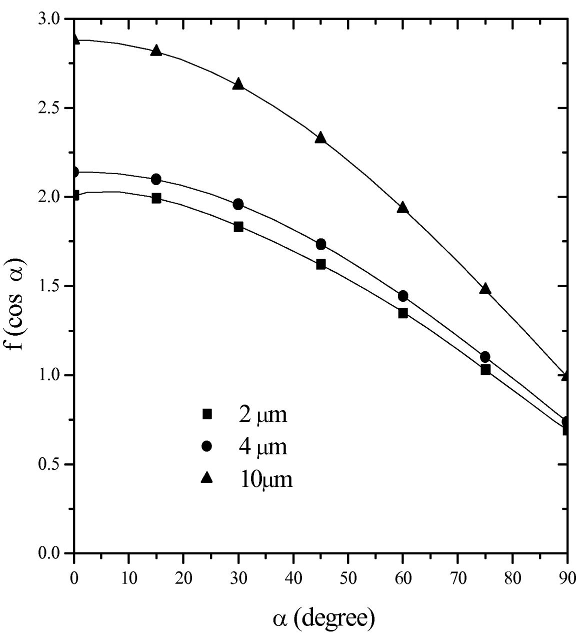

Fig. 4. Easy-axes distributions of the film textures.

90°. The results were fitted to a five-degree polynomial and consequently the corresponding distributions of easy axis $f(\cos\alpha)$ as a function of angle α between the magnetization and the texture direction was computed and plotted in Figure 4, following the method

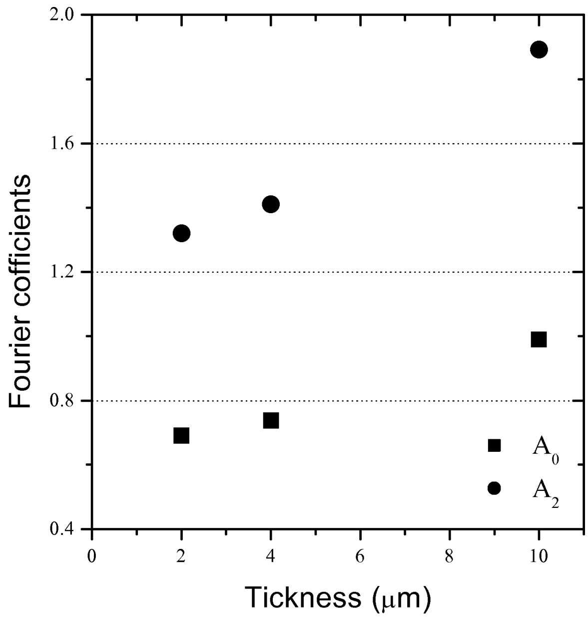

Fig. 5. Variations of the A_0 and A_2 coefficients.

described by El-Hilo et al. [4]. It is obvious that the Ni-Co films with higher $M_p(\beta)$ have a larger easy-axis distribution, which implies that the alignment of texture at high film thickness is improved for polycrystalline Ni-Co films.

In Fourier series expansion, the coefficients A_0 and A_{2n} are expected to provide information about the texturing properties of the films. As stated in the reports [4, 6], A_0 is sensitive to interaction effects and should be unity for a non-interacting system whereas A_{2n} is an indicative of magnetic texture in the sample. The A_0 and A_{2n} coefficients of the expression for $f(\cos\alpha)$ in the Fourier series computed from the fitted distribution function of the original data of $M_p(\beta)$ for all samples are illustrated in Figure 5. A list of the Fourier series coefficients is also shown in Table 1. A systematic and small increase in A_0 towards the unity exists and the coefficients A_{2n} $(n = 1)$ increases strongly with increasing film thickness. The consequences of the alignment of $M_p(\beta)$ can also be seen in the coefficients. From these values, a systematic and a wider increase from 1.320 to 1.892 in A_2 values may result in determining the preferred orientation of the Ni-Co films and provide a good correlation with the easy axis at $\beta = 90°$ in all $M_p(\beta)$ distributions of the films. However, it can be revealed that A_0 may not be a sensitive measure of demagnetising interaction effects due to the small range of variations. Besides, A_0 is less than unity and increase regularly towards very closed to unity in which implies the decrease of the range of demagnetising interaction strengths and hence the increase of the non-interaction in these samples as the film thickness increased.

For all $n \geq 1$, the coefficients $A_0 = 1$ and $A_2 = 2$ and the texture is completely oriented as reported by El-Hilo et al. [4]. In our polycrystalline Ni-Co films, the increase of A_0 from 0.691 towards almost unity (0.990) gives the indication of a small existence of demagnetising interactions to a non-interacting system. A_2 increased from 1.320 to 1.892 with the change of the oriented texture. Therefore, there exist a close correlation between A_2, which is obtained from the distribution of the easy-axis data, and the preferential texture orientation as turned from (111) to (220) with the decrease of Ni : Co ratio of the films caused by the increase of the film thickness. El-Hilo et al. [4] also predicted that the values of A_0 should be unity for non-interacting particles and showed that for particulate media with demagnetising interaction the value $A_0 < 1$, whereas for Co-P thin films in which the interactions are magnetising, the values $A_0 > 1$. The investigation of Ba-Fe media reported by Morales and O'Grady [6] showed that A_0 is always less than unity and generally increases towards unity as the magnetising interactions gain over the demagnetising magnetostatic interactions seen in the randomly oriented media. In our polycrystalline Ni-Co films, the values of A_0 increase towards unity as the demagnetising magnetostatic interactions decrease caused by the increasing magnetising interactions seen in the films when the orientation changed from (111) to (220). The results may be explained through a combination of magneto static interaction in which $A_0 < 1$ and an increasing contribution from magnetising interacting in which $A_0 > 1$ as the texture orientation of the polycrystalline Ni-Co films changed. This is consistent with the interpretation that the texture orientations may be responsible for the $M_p(\beta)$ values.

4. Conclusions

The texture orientations in electrodeposited Ni-Co films with various thicknesses were examined. Fourier analysis was used for the determination of the coefficients A_0 and A_{2n} from the easy-axis orientation in 2-D films. The structural analysis of the films exhibited that the films have fcc crystalline structure with the variations of the texture orientations as the film thickness changes. According to the XRD data, at 2 µm the (111) orientation is developed preferentially while at 10 µm the (220) orientation is more intense. It is revealed that the squareness of the films were altered by the changes occurred in crystalline orientation with the changes in composition of the film caused by the film thickness. The two lower-order coefficients in the Fourier series expansion reflect the variations in the film texturing and highlight the different origins of texture at the different film thicknesses of polycrystalline Ni-Co films.

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