Journal of Alloys and Compounds 506 (2010) 526–529

Contents lists available at ScienceDirect

Journal of Alloys and Compounds

journal homepage: www.elsevier.com/locate/jallcom

Study of obliquely deposited thin cobalt films

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ARTICLE INFO

Article history:
Received 6 April 2010
Received in revised form 12 July 2010
Accepted 12 July 2010
Available online 17 July 2010

PACS:
75.60.Ch, 68.55.Jk, 68.37.Ps, 68.37.Lp, 68.37.Hk

Keywords:
Thin cobalt films
Bitter pattern technique
Atomic force microscopy
Transmission electron microscopy
Scanning electron microscopy

ABSTRACT

Thin cobalt films, 40 nm and 100 nm in thickness, were deposited by thermal evaporation at an incidence angle of 45° in a system with a base pressure of approximately 10^{-5} mbar, simultaneously on unheated glass substrates and NaCl crystals. The magnetic microstructure of the films was investigated with the conventional Bitter pattern technique and the Fresnel mode of transmission electron microscopy (TEM), while the morphological structure was observed using TEM, atomic force microscopy (AFM) and scanning electron microscopy (SEM). The films were found to have uniaxial in-plane magnetic anisotropy. The magnetic microstructure of the films 40 nm and 100 nm thick consisted of domains running (and magnetized) predominantly in the direction perpendicular and parallel to the incidence plane, respectively. For the films studied, some correlation between the magnetic microstructure and the morphological structure was detected.

1. Introduction

Obliquely deposited thin films attract much attention from both fundamental and technological points of view. The shadowing effect, which occurs in such films, induces anisotropy in their macroscopic properties. The induced anisotropy can be magnetic, electrical, mechanical or optical in character. The properties of obliquely deposited films depend on the angle of deposition, film thickness, film composition and preparation conditions. Depending on the type of application, oblique deposition is successfully used to tailor the film properties. The range of applications is very wide and includes, for example, metal evaporated video tapes (which consist of partially oxidized $Co_{80}Ni_{20}$ or Co) [1,2], giant magnetostrictive films [3], optical filters, field emitters, solar cells, thermal barrier coatings, high-speed gas sensors and chromatography based microfluidic devices [4].

The present paper reports an investigation of thin cobalt films prepared by oblique-incidence deposition. The thicknesses of the studied cobalt films were chosen as 40 nm and 100 nm because for the mentioned film thicknesses the change in the direction of magnetic anisotropy and the transition from Néel to Bloch type walls were expected. The magnetic microstructure was observed using Bitter pattern method and transmission electron microscopy (TEM). To obtain an insight into the physical origin of the magnetic anisotropy of the films, the morphological structure was examined with TEM, atomic force microscopy (AFM) and scanning electron microscopy (SEM).

Obliquely deposited thin cobalt films were widely investigated in the past, using mainly TEM, the torque measurements, hysteresis loops and X-ray diffraction (XRD). Nevertheless, studies which report on correlation of the magnetic domain structure of these films with their morphological structure are in fact rare. Frankly speaking, we know only a few such studies [5–10]. Moreover, to our knowledge, observations of the morphological structure of obliquely deposited thin cobalt films have not yet been carried out simultaneously by TEM and AFM.

2. Experimental

The specimens under study were cobalt films 40 nm and 100 nm in thickness. They were produced by the technique of thermal evaporation at an incidence angle of 45° in a system maintained at a base pressure of approximately 10^{-5} mbar. A heater composed of two Al_2O_3 tubes threaded with tungsten wire was used. The film thickness was determined by a quartz crystal microbalance. The films were deposited simultaneously on unheated glass substrates and NaCl crystals (the latter films were prepared for the purpose of studying by TEM).

The magnetic microstructure of the films was made visible using the conventional Bitter pattern technique supported by a digital image processing system and the Fresnel (or defocus) mode of TEM. The Bitter patterns were observed under a conventional optical microscope PZO BIOLAR using transmitted unpolarized light, while magnetic studies by TEM were carried out using a Tesla BS 540 instrument.

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doi:10.1016/j.jallcom.2010.07.096

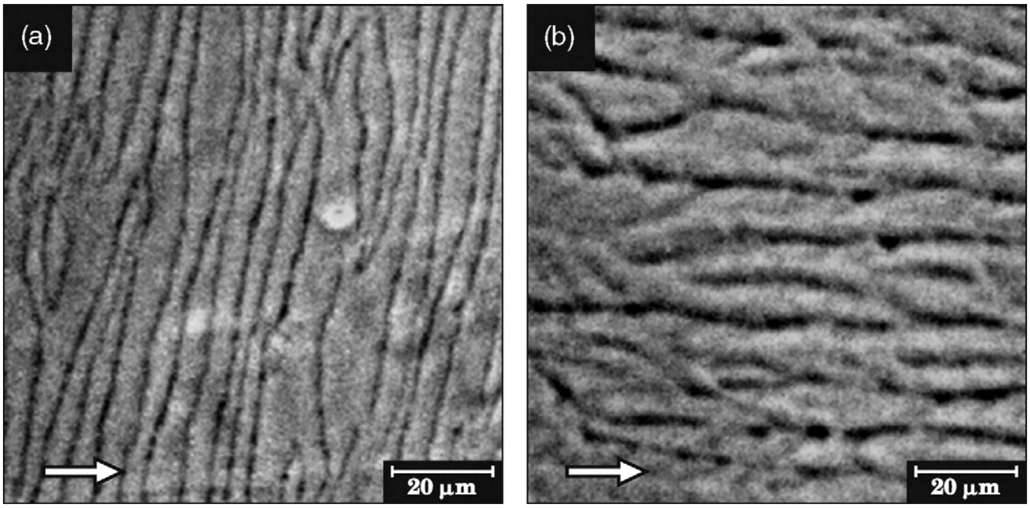

Fig. 1. Images of the magnetic microstructure of cobalt films (a) 40 nm and (b) 100 nm thick recorded with the conventional Bitter pattern technique. The arrow in each image indicates the projection of the vapor beam into the film plane.

For both of the methods applied, experimental conditions concerning observation of the magnetic microstructure were optimized to improve the magnetic contrast in the images (see Refs. [11,12] for details). The morphological structure of the films was investigated by TEM using a Tesla BS 540 instrument, by AFM using an Omicron instrument and by SEM using a Tesla BS 340 instrument.

3. Results and discussion

Fig. 1a and b presents images of the magnetic domain structure of the films 40 nm and 100 nm in thickness, respectively, recorded by the conventional Bitter pattern method. It is seen that the magnetic microstructure of the films 40 nm and 100 nm thick is composed of domains running predominantly in the direction perpendicular and parallel to the incidence plane, respectively. The magnetization of these films was found to be oriented predominantly perpendicular and parallel to the incidence plane, respectively, and to lie essentially in the film plane (of course except for small regions near the domain walls in which the magnetization exhibits an out-of-plane component), as indicated by the behavior of the Bitter patterns under the influence of external magnetic fields [13,14].

The type of the domain walls present in the investigated films could be determined more precisely with the Fresnel mode of TEM. In general, the occurrence of both cross-tie and Néel type walls was observed in the films 40 nm thick [13,14]. The cross-tie walls represent a transition between Néel and Bloch type walls and occur only in a certain range of the film thickness, from approximately 30–40 nm to 90–100 nm [15,16]. The domain walls in the films 100 nm in thickness were found to be of Bloch type, and most likely they were asymmetric Bloch walls. Moreover, a ripple structure of the magnetization could be seen in TEM images taken for the films of both thicknesses [13,14].

The films 40 nm and 100 nm thick were found to have uniaxial in-plane magnetic anisotropy in the direction perpendicular and parallel to the incidence plane, respectively. In the case of polycrystalline cobalt films, the contributions to the magnetic anisotropy originate from the geometric alignment of the grains through the shape anisotropy (the anisotropy of the demagnetizing field) as well as from the crystallographic alignment (texture) of the grains through the magnetocrystalline anisotropy [17,18].

Shown in Fig. 2a and b are TEM and AFM images of the morphological structure of the cobalt film 40 nm thick, respectively. As expected, the morphological structure consists of grains. The columnar structure of grains is not pronounced, but the alignment of columnar grains in the direction perpendicular to the

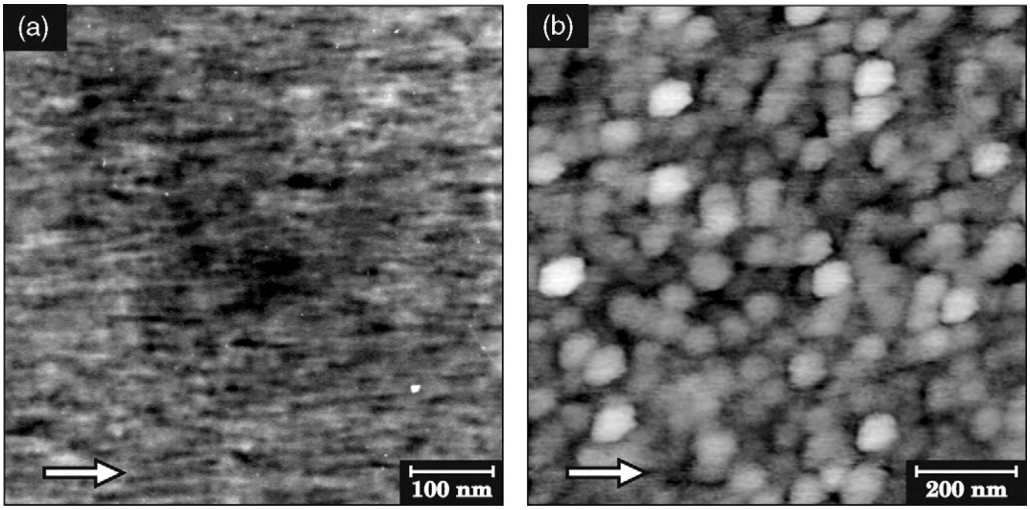

Fig. 2. (a) TEM and (b) AFM images of the morphological structure of a cobalt film 40 nm thick. The arrow in each image indicates the projection of the vapor beam into the film plane.

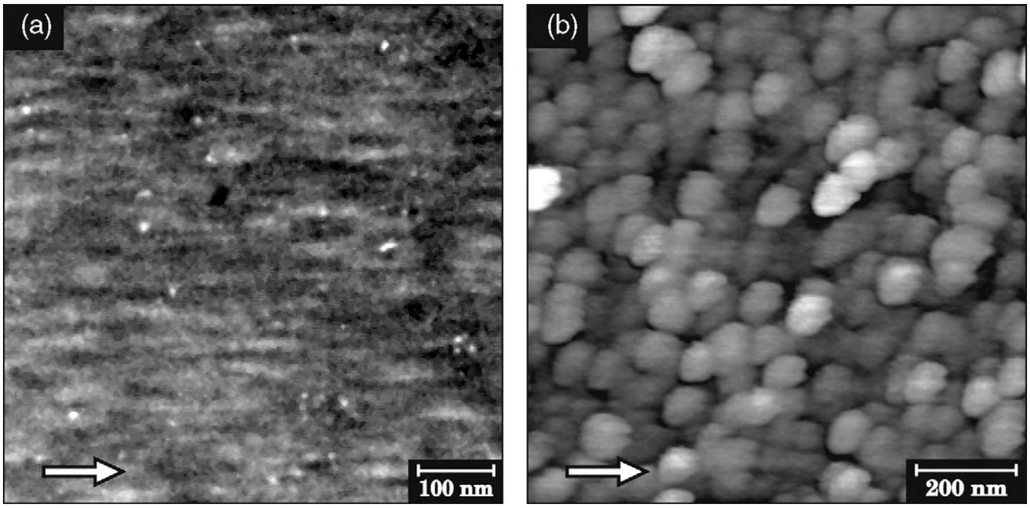

Fig. 3. (a) TEM and (b) AFM images of the morphological structure of a cobalt film 100 nm thick. The arrow in each image indicates the projection of the vapor beam into the film plane.

incidence plane can be recognized. Nevertheless, there are differences between the images in Fig. 2a and b. The reason for this is certainly related to the fact that AFM images the surface of the film, and in the case of TEM the morphological information is probed through the whole film thickness. The elongation of grains in the direction parallel to the incidence plane is marked in the TEM image of Fig. 2a, while the AFM image of Fig. 2b shows grains larger in size and slightly elongated in the direction perpendicular rather than parallel to the incidence plane. This in turn means that surface diffusion plays an important role in the process of film growth. The result obtained appears to confirm the finding of Ref. [19] where it was shown by simulations using a three-dimensional Monte Carlo method that surface diffusion forces columns to grow towards the columnar axes. To our knowledge, the observed differences between TEM and AFM images of the morphological structure of the film are reported for the first time. It is also to be noted that the recorded SEM images of the surface of the cobalt films were of very poor quality; they were much worse than images taken by AFM.

Fig. 3a and b presents TEM and AFM images of the morphological structure of the cobalt film 100 nm thick, respectively. In general, the observation of columnar grain structure in the films of this thickness is more difficult. In the considered context, however, it is to be noted that studies made on the basis of magnetic, optical and topographic measurements, reported in the literature, show that in evaporated films the columnar grains generally align in the direction perpendicular to the incidence plane [1,17,20]. In comparison with the films 40 nm in thickness, the films 100 nm thick have grains somewhat larger in size (in agreement with the result obtained by X-ray diffraction linewidth in Ref. [21]) and morphology more continuous. Note also that the root mean square (rms) values of the surface roughness, obtained by AFM measurements, were 0.8 nm and 1.2 nm for the films 40 nm and 100 nm thick, respectively.

The obtained images of the morphological structure prove that the shadowing effect was pronounced neither for the films 40 nm thick nor for the films 100 nm thick. The reason for this is found to be related to the fact that the investigated films were deposited at an intermediate incidence angle of 45°. The finding is in general agreement with the results reported in Refs. [3,6,9,10,19,20,22], where the effect of well-defined columnar grain morphology was observed for large deposition angles, larger than approximately 60°. It is also to be noted that the origin of the columnar grain structure, as well as the origin of the elongation of grains in the columns, can be explained by taking into account only geometric causes [1].

To get information on the crystallographic structure of the films, the selected area electron diffraction (SAED) patterns of TEM were recorded. For the films both 40 nm and 100 nm thick, the presence of continuous diffraction rings was observed, which means that the films are polycrystalline with randomly oriented grains, i.e. no crystallographic texture could be detected by the technique employed. Moreover, the occurrence of the set of three strong diffraction rings corresponding to the $(0\,1\,\bar{1}\,0)$, $(0\,0\,0\,2)$ and $(0\,1\,\bar{1}\,1)$ planes shows that the films studied are mainly composed of the hexagonal close-packed (HCP) phase of cobalt. An example of the SAED pattern of TEM is presented in Fig. 4, for the film 40 nm in thickness. To obtain a better insight into the crystallographic orientation of the films, it is necessary to apply more sensitive techniques, such as XRD Θ–2Θ spectra [22,23], the X-ray Schulz pole figures [17,24] or the electron backscattered diffraction (EBSD) method [24].

It should be noted that in the case of obliquely deposited cobalt films, the *c*-axis of texture was observed to be always perpendicular to the preferred direction of columnar grain structure independent of the preparation method, and moreover crystallographic textures were observed conclusively only for larger film thicknesses (above approximately 50 nm) [17,18,25]. As a consequence, the degree of

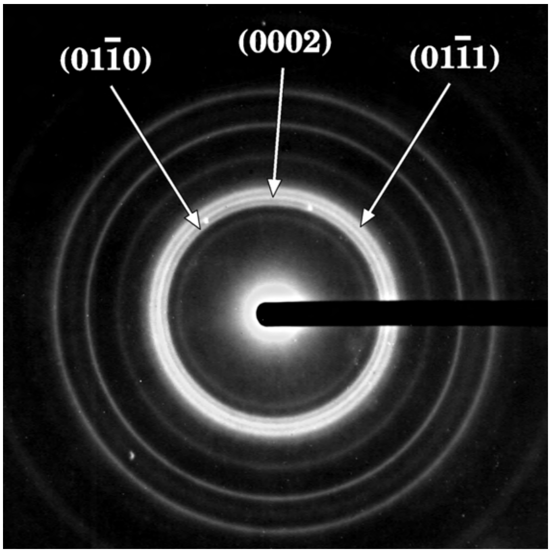

Fig. 4. TEM diffraction pattern of a cobalt film 40 nm thick.

crystallographic orientation in the films 40 nm thick is found to be low in comparison with that in the films 100 nm thick.

The change of the direction of magnetic anisotropy from perpendicular to parallel with respect to the incidence plane of the vapor beam, reported for obliquely deposited films as the film thickness increases, is of considerable interest, as discussed in several recent papers (e.g. Refs. [8,17,18,25]). In these papers the magnetic anisotropy of obliquely deposited films with different thickness was investigated using the torque measurements, hysteresis loops and transverse biased initial susceptibility (which provide information of the macroscopic character), but no magnetic domain studies were conducted. In this context, the present paper, which demonstrates by domain visualization (i.e. at the microscopic level) the reorientation of magnetic anisotropy from the direction perpendicular to parallel with respect to the incidence plane of the vapor beam, observed for cobalt films evaporated at an incidence angle of 45° as the film thickness is changed from 40 nm to 100 nm, appears to be a valuable contribution.

4. Conclusions

The paper presents an investigation of cobalt films 40 nm and 100 nm thick, thermally evaporated at an incidence angle of 45° in a system with a base pressure of approximately 10^{-5} mbar (simultaneously on unheated glass substrates and NaCl crystals). The films of both thicknesses were found to be magnetized in the plane of the film, except for small regions near the domain walls in which the magnetization possessed an out-of-plane component. The magnetic microstructure of the films 40 nm in thickness was composed of domains running and magnetized predominantly in the direction perpendicular to the incidence plane of the vapor beam. As the film thickness was changed from 40 nm to 100 nm, the magnetic anisotropy was observed to change from the direction perpendicular to parallel with respect to the incidence plane.

To get an insight into the physical origin of the magnetic anisotropy of the films, their morphological structure was studied for the first time simultaneously with TEM and AFM. Because TEM provides information originating from the whole film thickness and AFM substantially senses only the film surface, consequently complementary information on the morphological structure of the films could be obtained. In comparison with TEM images, AFM images revealed grains larger in size and slightly elongated in the direction perpendicular rather than parallel to the incidence plane. These experimental findings clearly show that surface diffusion plays an important role in the process of film growth.

For the films 40 nm thick, the alignment of columnar grains in the direction perpendicular to the incidence plane was observed. This correlates well with the magnetic domain structure of these films. For the films 100 nm thick, the perpendicular alignment of columnar grains could also be found, although in fact with larger difficulty. With the SAED patterns of TEM, it was obtained that the films consisted mainly of the HCP crystalline structure, but no preferred crystallographic orientation of the grains could be detected for the films of both thicknesses. For the films 100 nm in thickness, the alignment of magnetic domains in the direction parallel to the incidence plane indicates that the crystallographic contribution to the magnetic anisotropy is dominant over the shape anisotropy, however, experimental studies are needed to verify this hypothesis.

Acknowledgements

Witold Szmaja dedicates the paper to the memory of his mother. The work was supported by the Łódź University.

References

[1] L. Abelmann, C. Lodder, Thin Solid Films 305 (1997) 1.
[2] D. Shindo, Y. Murakami, J. Phys. D: Appl. Phys. 41 (2008) 183002.
[3] K. Muramatsu, T. Yamaki, N. Matsuoka, M. Takeuchi, Y. Matsumura, H. Uchida, J. Alloys Compd. 408–412 (2006) 335.
[4] J.J. Steele, M.J. Brett, J. Mater. Sci.: Mater. Electron. 18 (2007) 367.
[5] S.L. Zeder, J.-F. Silvain, M.E. Re, M.H. Kryder, C.L. Bauer, J. Appl. Phys. 61 (1987) 3804.
[6] A. Lisfi, J.C. Lodder, Phys. Rev. B 63 (2001) 174441.
[7] A. Lisfi, J.C. Lodder, J. Magn. Magn. Mater. 242–245 (2002) 370.
[8] A. Lisfi, J.C. Lodder, H. Wormeester, B. Poelsema, Phys. Rev. B 66 (2002) 174420.
[9] F. Tang, D.-L. Liu, D.-X. Ye, Y.-P. Zhao, T.-M. Lu, G.-C. Wang, A. Vijayaraghavan, J. Appl. Phys. 93 (2003) 4194.
[10] F. Tang, D.-L. Liu, D.-X. Ye, T.-M. Lu, G.-C. Wang, J. Magn. Magn. Mater. 283 (2004) 65.
[11] W. Szmaja, J. Magn. Magn. Mater. 219 (2000) 281.
[12] J.N. Chapman, M.R. Scheinfein, J. Magn. Magn. Mater. 200 (1999) 729.
[13] W. Szmaja, J. Balcerski, Czech. J. Phys. 54 (Suppl. (D)) (2004) D245.
[14] W. Szmaja, in: P.W. Hawkes (Ed.), Advances in Imaging and Electron Physics, vol. 141, Elsevier, Amsterdam, 2006, p. 175.
[15] A. Hubert, R. Schäfer, Magnetic Domains: The Analysis of Magnetic Microstructures, Springer, Berlin, 1998, p. 447.
[16] H.-N. Lin, Y.H. Chiou, B.-M. Chen, H.-P.D. Shieh, C.-R. Chang, J. Appl. Phys. 83 (1998) 4997.
[17] K. Itoh, K. Okamoto, T. Hashimoto, J. Magn. Magn. Mater. 190 (1998) 176.
[18] K. Hara, K. Itoh, K. Okamoto, T. Hashimoto, J. Magn. Magn. Mater. 162 (1996) 177.
[19] T. Karabacak, J.P. Singh, Y.-P. Zhao, G.-C. Wang, T.-M. Lu, Phys. Rev. B 68 (2003) 125408.
[20] J.L. Bubendorff, S. Zabrocki, G. Garreau, S. Hajjar, R. Jaafar, D. Berling, A. Mehdaoui, C. Pirri, G. Gewinner, Europhys. Lett. 75 (2006) 119.
[21] O. Kohmoto, Y. Yoshitomi, H. Murakami, IEEE Trans. Magn. 41 (2005) 3434.
[22] F. Liu, C. Yu, L. Shen, J. Barnard, G.J. Mankey, IEEE Trans. Magn. 36 (2000) 2939.
[23] A. Kharmouche, S.-M. Chérif, A. Bourzami, A. Layadi, G. Schmerber, J. Phys. D: Appl. Phys. 37 (2004) 2583.
[24] M.F. de Campos, T. Yonamine, M. Fukuhara, R. Machado, S.A. Romero, F.J.G. Landgraf, D. Rodrigues, F.P. Missell, J. Appl. Phys. 101 (2007) 09K516.
[25] J.M. Alameda, F. Carmona, F.H. Salas, L.M. Alvarez-Prado, R. Morales, G.T. Pérez, J. Magn. Magn. Mater. 154 (1996) 249.