Magnetic properties of Co nanocolumns fabricated by oblique-angle deposition

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The magnetic properties of columnar Co films on SiO_2 substrates fabricated by oblique-angle incident thermal evaporation at room temperature were systematically examined by multiple techniques, including magnetic force microscopy (MFM), magneto-optical Kerr effect (MOKE), and scanning electron microscopy (SEM). Films with thickness ranging from 50 to 500 nm were deposited at the incident angles (with respect to substrate normal) from 0° to 85°. For films with thickness of ~500 nm, the SEM shows the column tilt angle increases as the angle increases and becomes almost constant for all angles. The MFM images show that for 55°<θ<75°, stripe domains are formed and are nearly parallel to the direction of incident vapor beam. The hysteresis loops obtained from MOKE show that along the direction perpendicular to the incident vapor beam the coercivity H_c stays almost constant for all angles and the squareness decreases as the angle increases. This is in contrast to the increase of H_c and the increase of squareness in the direction parallel to the incident vapor beam for θ>60°. The result implies that for θ>60°, the axis parallel to the incident beam behaves more like the in-plane easy axis. These magnetic anisotropies are correlated to the angular-dependent columnar structure of Co films. © 2003 American Institute of Physics. [DOI: 10.1063/1.1558209]

I. INTRODUCTION

Magnetic properties of ferromagnetic thin films are closely related to their morphology and microstructures. A thin film prepared by a vapor beam with oblique-angle incidence forms a nanocolumn structure, which is very different from a continuous film grown under normal incidence. The magnetic films prepared by this oblique incident vapor in the presence of a considerable amount of oxygen are used as high-density and audio recording media, where the columns are considered as exchange-isolated. The structures and their magnetic anisotropy, especially the magnetic stripe domains of the magnetic films made by oblique incident vapor have been studied by various groups for wide ranges of thickness and temperature. For example, the microstructure of CoNi columns has been characterized by scanning electron microscopy (SEM), transmission electron microscopy, and x-ray diffraction. The magnetic properties, including anisotropy and coercivity of GdCo_2, have been studied by investigating the hysteresis loops obtained by using magneto-optical Kerr effect (MOKE). The magnetic domains of Co thin films have been characterized by magnetic force microscopy (MFM). Examples of previous major findings are that oblique-angle deposition introduces magnetic anisotropy, change of easy-axis direction, and change of the value of coercivity in the columnar films. In our present work we grew batches of 13 Co films on SiO_2 substrates at room temperature under the same deposition condition with an oblique-angle incident vapor beam at 0°, 15°, 30°, and in 5° increments from 45° to 85°. Five batches of 13 films per batch with thickness of ~500, ~300, ~200, ~100, and ~50 nm were grown. SEM top and cross-sectional images showing column structures were obtained. The in-plane magnetic anisotropy of columnar films was obtained by MOKE. The domain structures of the as-grown films versus vapor incident angle and thickness were investigated by MFM. The combination use of MOKE and MFM measurements on various incident-angle-deposited films under the same growth condition provide us a systematic and clear correlation between magnetic properties and column structures that has not been well characterized previously. In particular, by knowing the column orientations and column spacings, we are able to argue that the appearance of magnetic stripe domains above a certain incident deposition angle along ϕ=0° results from the difference of magnetic dipole–dipole interactions among columns along the incident vapor beam direction (azimuthal angle ϕ=0°) and that perpendicular to the incident vapor beam direction (ϕ=90°). The disappearance of magnetic stripe domains at large deposition angle is due to the increased spacing between columns. The sample preparation and experimental setup are described in Sec. II. The data collection and analysis are presented in Sec. III. The discussion of the correlation is in Sec. IV.

II. EXPERIMENTS

The Co nanocolumns were grown using the oblique-angle-incidence deposition technique as shown in Fig. 1. In
order to grow multiple samples under the same deposition environment with different vapor incident angles a special sample holder was made. It consisted of an aluminum base supported by three metal prongs used for mounting the base inside the e-beam evaporation chamber. Twelve metallic blocks with their top surfaces chiseled at various inclined angles from 15° to 85° were screwed onto this plate, as shown in Fig. 1(a). Cleaned silicon substrates, approximately 1-cm-rectangular in size, were mounted onto these inclined surfaces. As markers for later characterization, the silicon substrates had straight lines scribed on them parallel to the direction of incident beam. The distance between the evaporation source and substrates was about 40 cm, so that uncertainty in the exact angle of incidence caused by the divergence of the vapor beam was insignificant for our present study. The base pressure of the chamber was \(5 \times 10^{-6}\) Torr. Samples of varying thickness from 50 to 500 nm were prepared. Most depositions were carried out at a rate of 0.15–0.2 nm/s, determined by a quartz crystal monitor, while for \(~500\)-nm-thick films, the rate is about 1 nm/s. After deposition, the samples were cooled under vacuum. Each sample was cut into three pieces for later SEM, MFM, and MOKE measurements.

The morphologies and structures of the Co films were imaged by a field-emission SEM (FESEM 6330F, Jeol Ltd., Tokyo, Japan). The magnetic domain structures were imaged by an atomic force microscope (AFM) (Auto CP, Park Scientific Instrument). For the MFM measurement, a magnetic tip, coated with 50–100-nm-thick cobalt and magnetized along its long axis, was used instead of a regular silicon nitride AFM tip. The macroscopic magnetic property of the samples was measured by the MOKE technique. The Kerr intensity measurement consisted of a He–Ne laser, a polarizer, an analyzer, a photodetector, and an electronic circuit. The change of the in-plane azimuthal angle in a sample was controlled by a rotational feed-through with its axis perpendicular to the film plane. The incident plane in the MOKE setup and a sample with two directional markers are shown in Fig. 2. The angles illustrated in Fig. 2 are defined as the following: \(\theta\) is the vapor incident angle with respect to the film normal, \(\beta\) is the column tilt angle with respect to the film normal, and a zero azimuthal angle \(\phi = 0^\circ\) is defined when the straight-line markers are parallel to the applied magnetic field direction and the laser beam is incident from the opposite side of vapor incident beam. The sample was rotated clockwise when the rotational axis perpendicular to the film is viewed from the top of the film.

III. RESULTS AND DATA ANALYSIS

A. SEM images

Two representative SEM images of \(~500\)-nm-thick films deposited at different vapor incident angles are shown in Fig.
The cross-sectional SEM was imaged from the plane cut parallel to the scribed line direction, that is, the incident vapor direction. For the normal incident deposition angle $\theta = 0^\circ$, the film shows no obvious column. This is further supported by the top view of SEM that is not shown here. As the incident angle increases, the columns are formed. At a high incident angle of $\theta = 85^\circ$, the column tilt angle $\beta$ is about $55^\circ$ away from the film normal. From cross-sectional SEM images, the tilt angle $\beta$, length $l$, width $w$ of the columns, and the film thickness $d$ can be obtained. The top-view SEM images of the same samples are shown in Fig. 3(b). For $\theta = 0^\circ$ the surface looks smooth, while for $\theta = 85^\circ$ the surface looks corrugated, indicating that the surface is very rough. The cross-sectional SEM images obtained from the plane cut perpendicular to the incident-beam deposition direction are shown in Fig. 3(c) for films deposited at $\theta = 60^\circ$ and $70^\circ$. The images showed ends of protrusion of columns or columns with no tilt in the direction perpendicular to the incident vapor beam direction.

Figure 4 is a plot of the column tilt angle $\beta$ obtained from the SEM images versus vapor incident angle $\theta$. The $\beta$ increases as the $\theta$ increases. The $\beta$ versus $\theta$ has been fitted by cosine and tangent rules shown in Fig. 4 as the solid and dashed curves, respectively. The fits give $\beta = \theta - \sin^{-1}(1 - \cos(\theta)/2)$ and $\tan \beta = 0.5 \tan \theta$. Both fits are consistent with the trend that as the $\theta$ increases the $\beta$ deviates more from the slope of 1 shown as the straight dotted line in Fig. 4.

The column length $l$, column width $w$, and film thickness $d$ versus incident deposition angle $\theta$ are shown in Fig. 5. The width is $\sim 30$ to $\sim 25$ nm (see the inset of Fig. 5 for a magnified view). The film thickness $d$ is defined as the vertical distance between the top film surface and the bottom interface of Co and Si substrate. The film thickness $d$ decreases with the incident angle $\theta$ and can be fitted by a cosine relationship, $d = l \cos \beta$, shown as the dashed curve in Fig. 5.

**B. MFM images**

MFM images of size $10 \mu m \times 10 \mu m$ were collected for all as-grown films with deposition angles $\theta$ ranging from $0^\circ$ to $85^\circ$. Visual inspection of all the MFM images show that the magnetic domain structure changes with $\theta$. For the small and large $\theta$ angles, the domain structures are not regular, and random distributed spots are seen. In contrast, regular stripes with distinct contrasts (bright and dark) nearly parallel to the line marker direction, that is, incident-vapor beam direction,
occur within a certain angular range depending on the thickness of films. Figure 6 shows the stripe domains at \( \theta = 60^\circ \) with the thickness of \( \sim 100, \sim 200, \sim 300, \) and \( \sim 500 \text{ nm} \). The stripe structures are denser and clearer for thicker films and the width of stripe domains for \( \sim 500 \text{-nm-thick} \) film reaches a maximum. For \( \sim 500 \text{-nm-thick} \) film, the stripe domains appear between \( \theta = 55^\circ \) and \( \theta = 75^\circ \). MFM images for selected \( \theta \) angles for films \( \sim 500 \text{-nm-thick} \) are shown in Fig. 7. The stripe direction is nearly parallel to the incident vapor direction. The domain length increases as the deposition angle \( \theta \) increases from \( 50^\circ \), and reaches a maximum at \( \theta = 70^\circ \) and then decreases as the \( \theta \) angle continues to increase. The domain width change has a similar trend and also reaches its maximum at \( \theta = 70^\circ \). The corresponding power spectra for \( \sim 500 \text{-nm-thick} \) films are shown as insets in Fig. 7. The power spectrum changes with the deposition angle \( \theta \). For small angles and large angles, each power spectrum is like an ellipse. For the MFM image showing apparent stripes, the power spectrum has two separated peaks, due to the regular spacing between stripe domains. For a thinner film \( \sim 100 \text{ nm thick} \), the incident angle where the maximum domain length and the maximum domain width occur shifts around \( 60^\circ \).

C. MOKE loops

The in-plane longitudinal loops versus vapor incident angle \( \theta \) for magnetic field applied in the directions parallel \( H_{0^\circ} \) (\( \phi = 0^\circ \)) and perpendicular \( H_{90^\circ} \) (\( \phi = 90^\circ \)) to the incident plane of deposition for \( \sim 500 \text{-nm films} \) are shown in Fig. 8. The laser incident angle and reflected angle were fixed at \( \sim 35^\circ \). For the normal incidence deposited film, the measured loops are alike and are isotropic in two perpendicular directions. As \( \theta \) increases, the loops in these two directions become different. The loops obtained with applied \( H_{0^\circ} \) have larger areas compared with the loops measured with applied \( H_{90^\circ} \). Moreover, for \( \theta = 45^\circ \) and \( 55^\circ \), both loops under applied \( H_{0^\circ} \) and \( H_{90^\circ} \) exhibit kinks.

The coercivity \( H_c \) and approximate squareness versus \( \theta \) extracted from the loops shown in Fig. 9 are plotted in Figs. 9(a) and 9(b), respectively, for two azimuthal directions, \( \phi = 0^\circ \) and \( \phi = 90^\circ \). The \( H_c \) is \( \sim 20 \text{ Oe} \) for \( \theta = 0^\circ \) but the
value increases after \( \theta = 60^\circ \) and reaches \( \sim 600 \) Oe for \( H_\parallel \). In contrast, the \( H_c \) only increases mildly from \( \sim 20 \) to \( \sim 80 \) Oe for \( H_{90^\circ} \) as the \( \theta \) increases. The order of magnitude difference in \( H_c \) and the distinct loop shapes along two azimuthal directions imply magnetic anisotropy exists for columns grown at large oblique-angle deposition. The approximate squareness in our case is the ratio of remanence Kerr intensity over Kerr intensity at the maximum applied magnetic field. Since some loops were not fully saturated, the values of the saturation Kerr intensity were underestimated and the approximate squareness was overestimated. For \( H_{90^\circ} \), this approximate squareness decreases as the \( \theta \) increases but for \( H_\parallel \), the approximate squareness first decreases as the \( \theta \) increases and then increases after \( \theta \geq 60^\circ \).

IV. DISCUSSION

A. Magnetic anisotropy

The observed magnetic anisotropy is directly related to the tilt column structure produced by the oblique-angle deposition. The SEM images show that the column’s major axis tilts toward the direction of the incident beam in the plane of incidence (\( \phi = 0^\circ \)) and do not have an obvious tilt in the direction perpendicular (\( \phi = 90^\circ \)) to the incident beam direction. The tilted columns break the structural symmetry with respect to the plane perpendicular to the incident vapor direction, but not in the plane parallel to the incident deposition plane direction. A previous x-ray pole figure experiment of Co film grown on a glass at \( \theta = 60^\circ \) by Okamoto et al. showed that the \( (0002) \) pole intensity localizes in the vicinity of \( 60^\circ \) with a few degrees of distribution and that the \( c \)-axis is along the column’s long axis direction.\(^4\)

The tilt crystalline structures of Co columns induce magnetocrystalline anisotropy while the elongated columns will induce the shape anisotropy. If we assume that the column long axis is close to the \( c \)-axis and then two kinds of anisotropic energy could be defined according to an angle \( \varphi_{MC} \) which is the magnetization with respect to the \( c \)-axis or column long axis. We will first examine a single column and then discuss the interactions among columns. The total magnetostatic energy for a column is the sum of the magnetocrystalline anisotropy energy and the shape anisotropy energy. The magnetocrystalline anisotropy energy per unit volume can be expressed as

\[
E_c = K_1 \sin^2 \varphi_{MC} + K_2 \sin^4 \varphi_{MC},
\]

where \( K_1 \) and \( K_2 \) are the first- and second-order anisotropy constants, respectively. If we approximate the column as a prolate spheroid, the shape anisotropy energy is

\[
E_s = \frac{1}{2}(N_a - N_c)M_h^2 \sin^2 \varphi_{MC},
\]

where \( N_c \) and \( N_a \) are the demagnetizing factors along the \( c \)-axis and in the plane perpendicular to \( c \)-axis, respectively, and are related to the aspect ratio \( \gamma \) of the columns. (The \( \gamma \) is the ratio of column length \( l \) over the column width \( w \).) For a prolate spheroid with \( \gamma = 10 \), \( N_c = 0.256 \) and \( N_a = N_b = 6.16 \). The shape anisotropy is not considered in the following discussion and this will not affect our conclusion.

Following Okamoto et al.,\(^4\) assuming only domain rotational mechanism for the columnar films and \( c \)-axis coincident with vapor incident angle \( \theta \), we calculated the in-plane anisotropy energy due to magnetocrystalline anisotropy with \( K_1 = 4.53 \times 10^5 \) J/m\(^3\) and \( K_2 = 1.44 \times 10^5 \) J/m\(^3\) for \( \theta = 0^\circ, 40^\circ, \) and \( 80^\circ \), respectively. The results are shown in Fig. 10. \( \varphi_{MC} \), \( \theta \) and \( \phi \) are connected by spatial angle relation \( \cos \varphi_{MC} = \sin \theta \cos \phi \). For \( \theta = 0^\circ \), the anisotropy energy is isotropic. For \( \theta = 40^\circ \) or \( 80^\circ \), the anisotropy energy has a two-fold symmetry with a maximum value at a direction perpendicular to the incident-vapor-beam direction. The value of anisotropy energy \( E_{10^\circ} (\theta) \) along the \( \phi = 0^\circ \) direction decreases as the \( \theta \) increases, while the value of anisotropy energy \( E_{90^\circ} (\theta) \) along \( \phi = 90^\circ \) keeps the same maximum value. If the magnetization rotation is assumed, the hard axis will have the maximum anisotropy energy, while the easy axis will have the minimum anisotropy energy. The in-plane anisotropy field \( H_k \) due to the magnetocrystalline anisotropy is

\[
H_k = 2[E_{90^\circ}(\theta) - E_{10^\circ}(\theta)]/M_s, \quad \text{where } M_s \text{ is the saturation magnetization.}
\]

From calculation, we can see that as \( \theta \) increases, anisotropy field \( H_k \) increases, with \( \theta = 0^\circ \) becoming more like easy axis. These are qualitatively consistent with our experiments: as \( \theta \) increases, the anisotropy becomes more obvious as seen from differences in loop shapes, \( H_c \), and squareness.

B. Stripe domains

The MFM images show stripe magnetic domains developed for \( 55^\circ < \theta < 70^\circ \). The stripe domains were observed...
for magnetic films grown under various conditions.\textsuperscript{11–15} The stripes are formed nearly parallel to the plane of incident vapor beam or along the stripes are formed nearly parallel to the plane of incident beam plane as a mirror plane. The stripe domains gradually disappear for $\theta>75^\circ$. The average of stripe width is about 500 nm comparable to the film thickness. For a stripe width of about 500 nm, it contains many columns with an average diameter of ~30 nm.

The MFM images show alternate bright and dark stripes in the $\phi=90^\circ$ direction, implying different magnetostatic interactions along $\phi=0^\circ$ and $\phi=90^\circ$. Assuming each column has a magnetic dipole\textsuperscript{10} and magnetostatic interactions among magnetic columns can be approximated as dipole–dipole interactions, we may consider two configurations along $\phi=0^\circ$ and $\phi=90^\circ$ in Fig. 11. Along the $\phi=0^\circ$ direction the dipoles tilt $\beta$ angle away from the normal of a film with components parallel to the $\phi=0^\circ$ direction and along the $\hat{n}$ direction [see Fig. 11(a)], in which the parallel components also lie in the same line. The dipole–dipole interaction energy $E_d$ is proportional to $\cos^2 \beta M^2 R_0^2 - 2 \sin^2 \beta M^2 R_1^2$, where $R$ is dipole spacing.\textsuperscript{10} The first term comes from the interaction between vertical components of magnetization and the second term comes from the parallel components’ interaction. For the normal incidence, only the vertical component exists, and the dipole–dipole interaction energy will be lowered when a dipole flips and points opposite to an adjacent dipole. As $\beta$ increases ($\theta$ increases), the parallel component increases. At $\beta=38^\circ$, interaction energies for two components are equal, or $\cos^2 \beta M^2 R_0^2 = 2 \sin^2 \beta M^2 R_1^2$. For $\beta$ above $38^\circ$, the interaction energy for parallel components plays a more important role. The magnetizations of dipoles will point to a close direction to minimize the magnetostatic energy and form stripe domains. Experimentally stripe domains form for $\theta>55^\circ$ (or $\beta=43^\circ$, see Fig. 4), which is higher than $38^\circ$, probably because the close distance between columns makes the simple long-range dipole–dipole interaction approximation not suitable.

For the $\phi=90^\circ$ direction the dipoles tilt away from the normal of the film plane, but the components parallel to $\phi = 0^\circ$ do not align in the same line. Figure 11(b) shows components along the surface normal direction $\hat{n}$, in the plane perpendicular to the incident vapor beam. In this configuration dipoles tend to gradually change their direction to minimize the dipole–dipole interaction.\textsuperscript{16} Because the magnetization direction tilts out of film plane with a small angle when stripe domains are observed, the magnetostatic energy relating to dipole–dipole interaction may not be large enough\textsuperscript{17} to flip magnetization $180^\circ$ opposite to each other. The magnetization between two adjacent domains may just form a certain angle that deviates from the average magnetization in order to balance the magnetostatic energy, the exchange energy, and the magnetocrystalline anisotropy energy. This deviation gives rise to the bright and dark contrast variations in MFM images.

Due to the shadowing effect, the distance between columns becomes larger\textsuperscript{9,18} and magnetic interaction becomes weaker as $\theta$ increases. For $\theta>75^\circ$, the magnetic interaction may become too weak to form a long stripe. In addition, the magnetization will be almost in-plane at higher vapor incident angle, which will reduce the MFM contrast between stripe domains. For $\theta<55^\circ$, either well-defined columns are not formed or the formed columns are so closely packed that the magnetostatic interactions are about isotropic, causing no stripes to appear on the as-grown film.

V. CONCLUSIONS

In this article, we systematically studied the magnetic domain structure and the magnetic anisotropy of Co column films prepared by the oblique-angle vapor deposition under the same growth conditions. MFM images show regular stripe domains nearly parallel to the vapor deposition direc-
tion \((\phi=0^\circ)\) between \(\theta=55^\circ\) and \(\theta=75^\circ\). The domain widths, lengths, and power spectra of MFM images were analyzed. The stripe domains result from different magnetic interactions along \(\phi=0^\circ\) and \(\phi=90^\circ\) directions. The hysteresis loops measured along two azimuthal directions also show anisotropy. The hysteresis loops along \(\phi=0^\circ\) behave more like easy-axis loops as the deposition angle increases. The \(H_c\) at \(\phi=0^\circ\) increases rapidly after \(\theta=60^\circ\), in contrast to the \(H_c\) in the \(\phi=90^\circ\) direction, that only increases mildly as the \(\theta\) increases. The magnetic anisotropy can be qualitatively correlated to the anisotropic structures of tilted columns formed by the oblique-angle deposition.

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