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Observation of rotatable stripe domain in permalloy films with oblique sputtering

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Stripe domain (SD) in obliquely sputtered permalloy films were investigated by comparing with normally sputtered ones. The critical thickness for SD formation of obliquely sputtered films was about 100 nm thinner than that of normally sputtered films. The hysteresis loops of obliquely sputtered films showed a peculiar shape. A rotation of SD towards easy axis was observed in the obliquely sputtered films, which was confirmed by permeability spectra under a bias field. The origin of the rotation could result from in-plane uniaxial anisotropy, which is induced by the shape effect of the oblique columnar growth of permalloy grains. © 2012 American Institute of Physics. [http://dx.doi.org/10.1063/1.4764311]

I. INTRODUCTION

During the past 60 years, permalloy films have attracted great attention as a consequence of their highly sought-after magnetic characteristics for application in magnetic devices such as magnetic transducers, magneto resistive random access memory, and inductor magnetic cores.^{1–3} Typically, the magnetic moments of permalloy sample dominantly lie in the plane of the thin-film due to relatively strong demagnetization energy. However, above a critical thickness, an out-of-plane magnetization component could arise. Consequently, an array of oscillating "up and down" magnetic domain, known as stripe domain (SD), often forms.⁴ SD possesses the so-called "rotatable magnetic anisotropy" property, 5-7 which means the stripe direction can be changed by the application of a sufficiently large external field. Thus, the microwave absorption along any in-plane direction can be simply tuned by the application of an external field. This property may facilitate the integration of magnetic films into magnetic devices. Therefore, SD has been a popular research topic for its potential applications. It is desirable for both of fundamental research and application that the occurrence and properties of SD can be controllable.^{8–10}

Generally, SD develops in films having a weak uniaxial anisotropy K_p whose easy axis is oriented perpendicular to the film plane. An important parameter that characterizes the magnetization distribution inside stripe domain is the Q factor,^{11,12} which is defined as the ratio between K_p and the demagnetization field energy $2\pi M_s^2$, $Q = K_p/2\pi M_s^2$. Q reveals the origin of the formation of SD, which is the competition between perpendicular anisotropy energy and demagnetization energy.¹³ According to the Murayama theory,¹⁴ the critical thickness t_c for SD formation can be expressed

$$t_c = 2\pi \sqrt{\frac{A}{K_p}},\tag{1}$$

where A is the exchange stiffness constant. It can be seen from Eq. (1) that K_p plays a very important role in the formation of SD. Therefore, previous studies have mainly focused on the effect of deposition condition, such as sputtering rate, the substrate temperature, and film thickness on the formation and properties of SD.¹⁵ Oblique sputtering, which means depositing films by sputtering at an oblique incidence angle with the film normal, has raised a sustained interest for decades.^{16,17} Oblique sputtering would induce in-plane uniaxial magnetic anisotropy (IPUMA) K_u , which is essential for high-frequency application.¹⁸ However, influence of oblique sputtering on the formation and properties of SD has rarely been discussed. In addition to perpendicular anisotropy and demagnetization energy, an extra in-plane uniaxial magnetic anisotropy term would exist in obliquely sputtered films. How the competition among these energy influences the formation and properties of SD could be intriguing.

In this paper, we investigated obliquely sputtered permalloy films. We found that oblique sputtering had a profound influence on the formation and properties of SD. The critical thickness for SD formation significantly decreased with oblique sputtering. For films deposited at an oblique angle of incidence, the in-plane easy magnetization direction (easy axis) which resulted from in-plane uniaxial magnetic anisotropy was typically perpendicular to the deposition direction. A peculiar hysteresis loop which has never been reported was found. A rotation of stripe domain towards in-plane easy axis was observed in the obliquely sputtered films. Permeability spectra with an external bias field confirmed the rotation of SD.

II. EXPERIMENTAL PROCEDURE

Permalloy films with various thickness were prepared by radio frequency (RF) sputtering on $10 \text{ mm} \times 10 \text{ mm} \times 0.42 \text{ mm}$ (111)-oriented Si substrates, which were attached to a water-cooling system with background pressure lower than 4×10^{-5} Pa. During sputtering, an oblique incidence angle of 50° was used to introduce IPUMA,¹⁸ as shown in

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FIG. 1. (a) Schematic illustration of the sputtering arrangement. The substrate is mounted on a titled holder during sputtering. The oblique angle is marked as θ . The inset shows the easy axis direction of the samples. (b) and (c) are SEM cross-section images of normally with $\theta = 0^{\circ}$ and obliquely with $\theta = 50^{\circ}$ sputtered permalloy films, respectively. The solid arrows represent the growth direction of the columnar grains and dashed arrows the incidence beam direction.

Fig. 1(a). Normally sputtered films under the same deposition conditions were also prepared as comparison. A Fe₁₉Ni₈₁ target, 70 mm in diameter and 3 mm in thickness, was used to deposit permalloy films. The working Ar pressure was 0.7 Pa with Ar flow rate of 80 sccm (sccm denotes cubic centimeter per minute at STP), and the radio frequency power density was 2.5 W/sc. The static magnetic properties were determined by vibrating sample magnetometer (VSM, Lakeshore model 7304). A field emission scanning electron microscope (SEM, Hitachi S-4800) was employed to observe the cross-section microstructures. The magnetic domain images were captured at room temperature by an Asylum Research (C) MFP-3D magnetic force microscopy (MFM) with soft magnetic tips magnetized perpendicular to the sample plane. Permeability spectra were carried out with a PNA E8363B vector network analyzer using the microstrip method.

III. RESULTS AND DISCUSSION

Figs. 1(b) and 1(c) show the SEM cross-section images of normally and obliquely sputtered permalloy films. One can see the obvious columnar structures in both normally and obliquely sputtered films. The columns of obliquely sputtered film have an angle of about 40° with the film normal. The angle of columnar structures is closely related to the oblique angle of sputtering. In our experiments, the angle of columnar structures is about 40° for films deposited at an oblique angle of 50° . Oblique columnar structures would induce an in-plane uniaxial anisotropy.^{19,20} Thus, comparing with normally sputtered films, an extra in-plane uniaxial anisotropy term exists in obliquely sputtered films.

Fig. 2 shows the in-plane hysteresis loops of obliquely sputtered films with different thickness. The difference



FIG. 2. In-plane hysteresis loops of obliquely sputtered permalloy films at the different thicknesses. Circle line and square line are hysteresis loops along easy axis (EA) and hard axis (HA), respectively.

between hysteresis loops along easy and hard axis indicates that in-plane uniaxial anisotropy exists in the obliquely sputtered film, which is consistent with SEM cross-section images. For films thinner than 100 nm, as shown in Fig. 2(a), the loop is typical for films with well-defined IPUMA.¹⁸ For films thicker than 140 nm, as shown in Figs. 2(b)–2(d), the loops along easy axis are representative for films with SD formation, which is indicated by the reduction of remanence, the enhancement of coercivity, a linear magnetization rotation part, and a steep switching part at small fields.^{13,21} How-ever, the loops along hard axis are really strange. They include two parts: linear magnetization rotation part from saturation magnetization and a reduction of remanence part at small fields.

Thickness dependence of normalized remanence in both normally and obliquely sputtered films is compared in Fig. 3.



FIG. 3. Thickness dependence of normalized remanence for both normally and obliquely sputtered permalloy films. The open symbols mean that stripe domain was observed in the sample and solid symbols without SD observed. The critical thickness is represented by vertical dash line.

The remanence of normally sputtered films is identical in any in-plane direction and decreases continuously with increasing films thickness. The decrease is caused by out of plane rotation of magnetic moments when SD forms. For obliquely sputtered films, the trend of remanence along easy axis is similar to that of normally sputtered films. However, the remanence along hard axis is lower than that of easy axis and exhibits an enhancement with the formation of SD. The critical thickness is obtained by directly observing the domain structure of the films with various thicknesses using MFM. The critical thickness of obliquely sputtered films is about 100 nm thinner than normally sputtered ones, which implies that K_p is increased by oblique sputtering. Increased K_p leads to a bigger Q factor and results in the reduction of critical thickness, which means oblique sputtering could be another approach to modify the occurrence of SD.

In order to get a better understanding of mechanism the peculiar hysteresis loops in Fig. 2, domain structure evolution under an applied field along easy and hard axis is captured as shown in Fig. 4. When a field of 500 Oe is applied, all the magnetic moments will align to the direction of the applied field due to Zeeman energy, so there is no brightness contrast in the MFM image.¹³ While decreasing the applied magnetic field, the magnetic moments gradually oscillate out of the film plane in a periodical manner and a clearly stripe domain is observed. The perpendicular component of magnetization increases with decreasing magnetic field; thereby, the brightness contrast of MFM images is gradually increasing.^{22,23} When the bias field is applied parallel to easy axis, as shown in Fig. 4(a), the stripe direction keeps unchanged with decreasing field. However, when the magnetic field is applied along hard axis, as shown in Fig. 4(b), the stripe direction rotates towards in-plane easy axis. The stripe direction at remnant state does not align along the direction of the previously applied bias field. With decreasing bias field, the magnetic moments turn towards easy axis due to the existence of IPUAM, resulting in the rotation of stripe direction and reduction of remanence at small fields.



FIG. 4. MFM images of obliquely sputtered permalloy film of 280 nm with a bias magnetic field applied perpendicular to in-plane easy axis (HA) (a) and hard axis (EA) (b), respectively. *H* denotes the applied external field. The arrows represents the direction of *H* and EA (or HA). The magnitude of the field is gradually decreasing from 500 Oe to 0 Oe. The scan size of all images is $10 \,\mu\text{m} \times 10 \,\mu\text{m}$. The colored vertical bar represents the shift of resonant frequency of the cantilever.



FIG. 5. A typical permeability spectra obtained by microstrip method. Where open circles represent imaginary part μ'' and solid circles represent real part μ' of permeability, respectively. f_r is the resonance frequency. The inset shows the direction of bias field and microwave field.

Magnetic permeability ($\mu = B/H$) spectra under a bias field are used to confirm the rotation of stripe direction. The permeability spectra were carried out with a PNA E8363B vector network analyzer using the microstrip method.²⁴ The spectra with different bias field are obtained by saturating the film first and then applying a magnetic field decreasing from 200 Oe to 0 Oe. The stimulating microwave field is perpendicular to the bias field, as shown in Fig. 5. The resonance frequency f_r is defined as the frequency where the maximum μ'' occurs. Fig. 6(a) shows the square of f_r as a



FIG. 6. Bias field dependence of the square of resonance frequency f_r for (a) normally sputtered SD film of 280 nm and (b) obliquely sputtered SD film of 140 nm, respectively. The square dot represents the experimental data when the bias field is parallel to easy axis, and circle dot when the bias field is parallel to hard axis. The half solid circles represent the data of the second resonance peak. Insets show the imaginary parts (μ'') of permeability spectra with different bias field.

function of the bias field for normally sputtered permalloy film with the bias field parallel to any in-plane direction. As normally sputtered SD film possesses rotatable anisotropy, the permeability spectra along any in-plane direction are exactly the same, which implies the in-plane homogeneity of normally sputtered films. The stripe direction does not change with the decreasing bias field, so the microwave field is always perpendicular to the stripe direction. The square of f_r decreases with decreasing bias field, which can be fitted very well by Kittel formula²⁵

$$f_r^2 = \left(\frac{\gamma}{2\pi}\right)^2 4\pi M_S (H_u + H_{app}),\tag{2}$$

where γ is the gyro magnetic constant, H_{app} is the applied bias field, and H_u the in-plane uniaxial magnetic anisotropy field. For obliquely sputtered SD films, as shown in Fig. 6(b), bias field dependence of the square of f_r with the field parallel to easy axis is identical to that of normally sputtered SD film and can be fitted with Eq. (2). However, the magnetic excitation becomes really complicated when the bias field is parallel to hard axis. When the bias field is 200 Oe, the magnetic moments nearly aligned along the direction of the bias field, and thereby only one resonant peak is observed in the permeability spectra. With decreasing magnetic field, the stripe direction turns towards easy axis gradually, the microwave field is not perpendicular to the stripe direction and a double resonance peaks is observed. Vukadinovic et al.^{26,27} have calculated the magnetic excitations in weak stripe domain structure with microwave field perpendicular or parallel to the stripe direction. They predicted multiple resonance peaks with different measurement configurations, which is in agreement with our experimental results. Hence, the emergence of double peaks comes from the stripe direction rotation in obliquely sputtered SD film. However, the origin of each resonance peak is still an open question.

IV. CONCLUSIONS

In summary, permalloy films sputtered at an oblique incidence angle have been investigated by comparing with normally sputtered ones. The critical thickness of obliquely sputtered films was about 100 nm thinner than that of normally sputtered films. The hysteresis loop of obliquely sputtered films showed a peculiar shape due to the existence of IPUMA. A rotatable SD was observed, which resulted in the reduction of remanence along hard axis. Permeability spectra under a bias magnetic field confirmed the rotation of stripe direction. The origin of difference between obliquely and normally sputtered SD could result from the shape effect of the oblique columnar growth of the permalloy grains. Therefore, in addition to sputtering rate, the substrate temperature, and film thickness, the occurrence and properties of SD could also be modified by changing the deposition angle. Especially, the critical thickness is reduced by increasing the deposition angle, which implies we can obtain SD structure in a relatively thin film. However, deposition angle dependence of the micro magnetization distribution and domain width is still to be studied, which is really important for applications such as data storage devices and sensors.^{28,29}

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