



# Controlling of magnetic domain structure by sputtering films on tilted substrates



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## ABSTRACT

In this paper, we investigated the magnetic domain structure evolution in nickel (Ni) and permalloy ( $\text{Fe}_{19}\text{Ni}_{81}$ ) films deposited on tilted substrates. It is found that the magnetic domain structure could be controlled from large in-plane domains to stripe-like domains, and then bubble-like domains in the films at the same thickness by just changing the oblique angle of the substrate. The angle dependence of ferromagnetic resonance (FMR) measurements was used to quantitatively analyze the changing of magnetic properties with different tilted angle. We demonstrated that the perpendicular magnetic anisotropy of the film was enhanced by increasing the tilted angle of the substrate. The origin of the enhancement of perpendicular anisotropy was also discussed.

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## 1. Introduction

Controlling the magnetic properties, such as magnetization direction and domain structure, is essential for both of fundamental physics and applications in future devices, e.g., hard disk drives, spintronics components, data storage devices, magnetic random access memory and sensors [1–6]. The domain structure in thin films is closely related to perpendicular magnetic anisotropy (PMA). The ratio between the perpendicular anisotropy energy  $K_{\perp}$  and the demagnetizing energy term  $2\pi M_s^2$  defines the quality factor  $Q$  ( $Q = K_{\perp}/2\pi M_s^2$ ), which characterizes the competition between perpendicular anisotropy and demagnetizing energy. For film with strong PMA ( $Q > 1$ ), the magnetization of the film is essentially perpendicular to the film surface and bubble domain could be observed [7]. For film with low or intermediate PMA ( $Q < 1$ ), the magnetization tends to be in the film plane with an alternating net perpendicular component and the so-called stripe domain often forms beyond a critical thickness  $t_c$  [8,9]:

$$t_c/\Lambda = 2\pi\sqrt{\frac{1}{Q}}, \quad (1)$$

where  $\Lambda$  is the exchange length ( $\Lambda = (A/2\pi M_s^2)^{1/2}$  with  $A$  the exchange constant). Many efforts have been done to explore the evolution of magnetic domains with different  $Q$  factor [9,10]. From the pioneer works, thickness dependence of domain structure evolution was investigated in permalloy [11], Co [12], FePt [13], FePd [14], CoFeZr [15], FeCoNd [16], FeSiB [17], and FeTaN films [18].

However, in all these literatures [11–18], the evolution of magnetic domains was driven mainly by changing the film thickness and could not be tuned effectively in one specific material at the same thickness. Especially, the domain structure evolution was phenomenologically described without quantitatively relating to the change of magnetic parameters with different tilted angle of substrates [19,20].

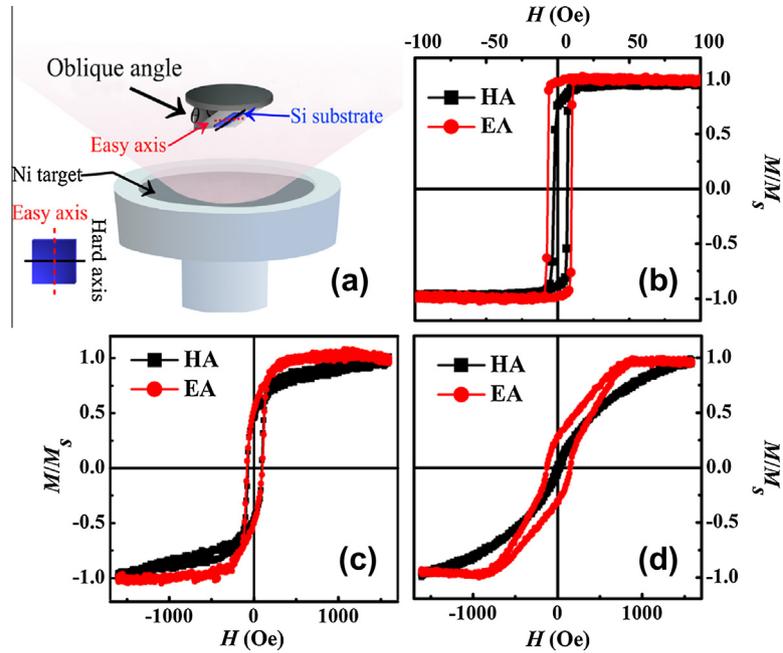
In this paper, we investigated the magnetic domain structure evolution in Ni and permalloy films deposited on tilted substrates. We demonstrated that the domain structure in magnetic films, at the same thickness, can be controlled effectively from large in-plane domains to stripe-like domains and then bubble-like domains by just increasing the tilted angle of the substrate. Angle dependence of FMR and static vibrating sample magnetometer (VSM) measurements were used to obtain the magnetic parameters of the films. By fitting FMR data, we found that the PMA of the film increases with larger tilted angle of substrate, which resulted in the domain structure evolution. The tuning of domain structure can be achieved in both of negative magnetostriction material (Ni) and near-zero magnetostriction material permalloy ( $\text{Fe}_{19}\text{Ni}_{81}$ ), which implies the magnetoelastic anisotropy is not the key factor to the enhancement of PMA. The origin of perpendicular anisotropy was ascribed to the shape effect which is induced by columnar structure, as confirmed by multilayers films which hinder the growth of columnar structure.

## 2. Material and methods

Ni and permalloy films were prepared by radio frequency (RF) sputtering on  $20\text{ mm} \times 10\text{ mm} \times 0.42\text{ mm}$  (111)-oriented silicon substrates with background pressure lower than  $4 \times 10^{-5}\text{ Pa}$ . A Ni target and a permalloy target were used to

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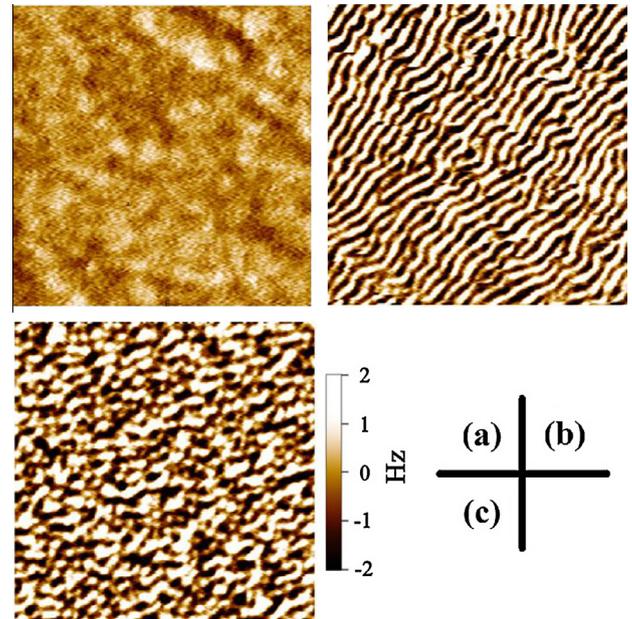
**Fig. 1.** (a) Schematic illustration of the sputtering arrangement. The substrate is mounted on a tilted holder during sputtering. The tilted angle of the substrate is marked as  $\theta$ . The inset shows the easy axis direction of the samples. (b), (c) and (d) are in-plane hysteresis loops of Ni films deposited with  $\theta = 10^\circ$ ,  $30^\circ$ , and  $60^\circ$ , respectively, at the thicknesses of 100 nm. Circle line are hysteresis loops along easy axis (EA) and square line along hard axis (HA).

deposited the films, respectively. During sputtering, the substrate was tilted with an angle changing from  $10^\circ$  to  $60^\circ$ , as shown in Fig. 1a. The working Ar pressure was 1 Pa with Ar flow rate of 10 SCCM (SCCM denotes cubic centimeter per minute at STP), the radio frequency power density was 2.5 W/sc. The thickness of the film, which was controlled by deposition time, was 100 nm for Ni films and 220 nm for permalloy films. The static magnetic properties were determined by vibrating sample magnetometer (VSM, Lakeshore model 7304). The magnetic domain images were captured at room temperature by MFM with soft magnetic tips magnetized perpendicular to the sample plane. The ferromagnetic resonance (FMR) measurements were performed in a JEOL, JES-FA 300 (X-band at 8.969 GHz) spectrometer.

### 3. Results and discussions

Fig. 1b–d shows the in-plane hysteresis loops of Ni films deposited at the tilted substrate angle of  $10^\circ$ ,  $30^\circ$ , and  $60^\circ$ , respectively. The slight difference between easy axis (EA) and hard axis (HA) indicates the existence of small in-plane uniaxial magnetic anisotropy [21]. Films deposited at a small tilted angle exhibit a square hysteresis loop while for larger tilted angle the hysteresis loop deviates significantly from the square one and is typical for films having perpendicular magnetic anisotropy, 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 [11].

Fig. 2 shows the corresponding MFM images of films deposited at the tilted substrate angle of  $10^\circ$ ,  $30^\circ$ , and  $60^\circ$ . The magnetic domain structure of Ni films is found to be significantly affected by the deposition angle. Films deposited at a small tilted angle, as shown in Fig. 2a, show typical magnetic domain structure with large in-plane domains, indicating the magnetization mainly oriented in the film plane. As expected from the hysteresis loop in Fig. 1c, a stripe-like domain structure is observed in Ni film deposited at  $30^\circ$ . The brightness contrasts are due to magnetization canted upward or downward out of the film's plane, which results from the competition between perpendicular magnetic anisotropy and demagnetization energy. For films deposited at a larger oblique angle, as shown in Fig. 2c, the films show bubble-like domain structure, which has been reported in film with a relatively large perpendicular magnetic anisotropy.



**Fig. 2.** MFM images ( $4 \times 4 \mu\text{m}^2$ ) of Ni films deposited at the tilted substrate angle of  $\theta = 10^\circ$  (a),  $30^\circ$  (b), and  $60^\circ$  (c), respectively. The colored vertical bar represents the shift of resonant frequency of the cantilever. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Ferromagnetic resonance (FMR) was used to determine the value of PMA. The total free energy density for uniformly magnetized film can be expressed as:

$$F = -HM_S[\cos \theta_M \cos \theta_H + \sin \theta_M \sin \theta_H \cos(\varphi_M - \varphi_H)] + 2\pi M_S^2 \cos^2 \theta_M - K_u \sin^2 \theta_M \cos^2 \varphi_M + K_\perp \sin^2 \theta_M. \quad (2)$$

It includes Zeeman energy as the first term, the second term is the demagnetizing field energy, the third term is the energy of in-plane uniaxial anisotropy  $K_u$ , and the last term is the energy of

perpendicular anisotropy  $K_{\perp}$ .  $\gamma$  is the gyromagnetic ratio,  $\theta_H$  is the angle between external field and film normal,  $\theta_M$  the angle between magnetization vector and film normal,  $\varphi_M$  and  $\varphi_H$  are azimuthal angle of  $\mathbf{M}$  and  $\mathbf{H}$ , respectively. Substituting the free energy density  $F$  into FMR frequency derived by Suhl and Smit [22,23]:

$$\left(\frac{\omega}{\gamma}\right)^2 = \frac{1}{M_S^2 \sin^2 \theta_M} \left[ \frac{\partial^2 F}{\partial \theta_M^2} \frac{\partial^2 F}{\partial \varphi_M^2} - \left( \frac{\partial^2 F}{\partial \theta_M \partial \varphi_M} \right)^2 \right], \quad (3)$$

then we get the ferromagnetic resonance equation for out-of-plane measurement configuration:

$$\left(\frac{\omega}{\gamma}\right)^2 = \left\{ H \cos(\theta_M - \theta_H) - \left( 4\pi M_S - \frac{2K_{\perp}}{M_S} \right) \cos 2\theta_M - \frac{2K_{\parallel}}{M_S} \cos 2\theta_M \right\} \times \left\{ H \cos(\theta_M - \theta_H) - \left( 4\pi M_S - \frac{2K_{\perp}}{M_S} \right) \cos^2 \theta_M - \frac{2K_{\parallel}}{M_S} (\cos^2 \theta_M - 1) \right\}, \quad (4)$$

the measurement configuration is shown in the inset of Fig. 4. Taking into account the equilibrium equation of magnetization  $H \sin(\theta_M - \theta_H) - (4\pi M_S - 2K_{\perp}/M_S) \sin \theta_M \cos \theta_M = 0$  and the saturation magnetization  $4\pi M_S$  obtained by static VSM measurement, the perpendicular magnetic anisotropy constant  $K_{\perp}$  can be acquired by fitting the experimental data with Eq. (4).

Fig. 3 shows the original data out of plane FMR signal with different polar angle  $\theta_H$ . It can be clearly seen that the resonance field changes with different  $\theta_H$ . The out-of-plane resonance field versus  $\theta_H$  for films deposited at the tilted angle of  $10^\circ$ ,  $30^\circ$ , and  $60^\circ$  is summarized in Fig. 4. The experimental data can be very well fitted by the theoretical result in Eq. (3), which implies the validity of the method. The resonance fields tend to decrease monotonically for each film with increasing  $\theta_H$  (in range of  $0$ – $90^\circ$ ), which is caused by the demagnetization energy when the external field  $\mathbf{H}$  is parallel to film normal. Moreover, the maximum of resonance field decreases with increasing deposition oblique angle, which is closely related to the perpendicular anisotropy field  $2K_{\perp}/M_S$ .

Fig. 5 shows the perpendicular magnetic anisotropy as a function of oblique angle obtained by the aforementioned fitting procedure. It can be clearly seen that  $K_{\perp}$  monotonically increases from  $0.5 \times 10^5$  to  $7 \times 10^5$  erg/cm<sup>3</sup> with increasing tilted angle from  $10^\circ$  to  $60^\circ$ . The demagnetization energy is given by  $2\pi M_S^2$  for a continuous film, which favors in-plane magnetization and keeps unchanged for the as-prepared films. The effect of perpendicular magnetic anisotropy gradually increases in competition with demagnetization energy, which results in the magnetic domain structure evolution from in-plane domains to stripe-like domains, and then bubble-like domains, as shown in Fig. 2. The perpendicular anisotropy could come from magnetocrystalline anisotropy, mag-

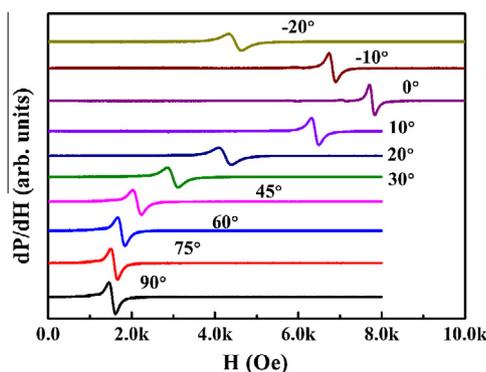


Fig. 3. Original data of FMR with different polar angle  $\theta_H$ . The Ni film was prepared on tilted angle of substrate of  $10^\circ$ .

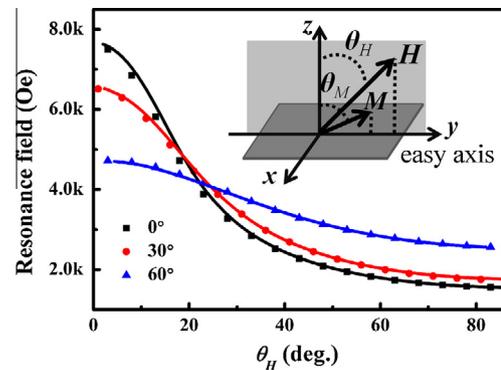


Fig. 4. Out of plane angle dependence of resonance field. The square, circle and triangle dots represent resonance fields for Ni films deposited at the tilted substrate angle of  $10^\circ$  (a),  $30^\circ$  (b), and  $60^\circ$  (c), respectively. The solid lines are theoretical lines obtained by Eq. (4). The inset shows the field direction during FMR measurement.

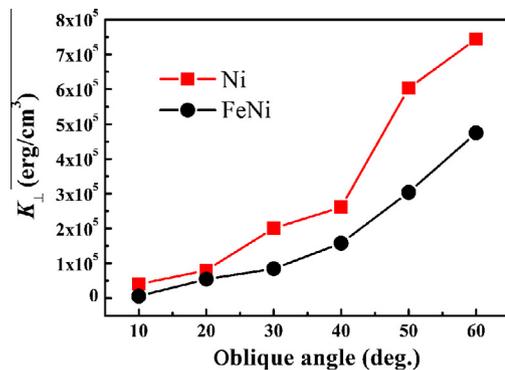


Fig. 5. The perpendicular anisotropy  $K_{\perp}$  of Ni and permalloy films versus titled substrate angle. The  $K_{\perp}$  increases with larger substrate angle.

netoelastic anisotropy, shape anisotropy or surface/interface effect. It is noteworthy that the value of  $K_{\perp}$  in the as-prepared Ni films is significantly larger than the magnetocrystalline anisotropy of bulk Ni of  $4.5 \times 10^4$  erg/cm<sup>3</sup>, thereby the magnetocrystalline anisotropy does not play a major role to the enhanced perpendicular anisotropy. Meanwhile, the surface/interface effect can be neglected for thin polycrystalline films of 100 nm. The magnetoelastic anisotropy  $K_{\sigma}$  can be expressed as  $K_{\sigma} = 3/2\lambda\sigma$ , where  $\lambda$  is the magnetostriction constant and  $\sigma$  is the stress in the polycrystalline films. As  $\lambda$  is near-zero for permalloy films [24], the contribution of magnetoelastic anisotropy can also be ruled out. The shape anisotropy induced by columnar structure is most likely the main reason for the enhancement of perpendicular anisotropy. Moreover, the shape anisotropy increases with increasing deposition oblique angle. The enhancement of shape anisotropy could result from self-shading effect which favors the growth of columnar structure with larger deposition angle [25,26]. In order to verify that the perpendicular anisotropy originates from shape anisotropy, we deposited  $[\text{Ni}(10 \text{ nm})/\text{Ta}(10 \text{ nm})]_{10}$  multilayer at the deposition oblique angle of  $60^\circ$ . The inserting of Ta layers hinders the growth of columnar structure [27–29], so perpendicular anisotropy is expected to be vanished in the multilayer films, thus, the magnetic moments will lie in the film plane due to relatively large demagnetization energy. The MFM image of the as-sputtered multilayer (not shown here) shows in-plane magnetization distribution, which confirms the perpendicular anisotropy is induced by shape anisotropy.

Fig. 6 shows the position of our samples in the phase diagram of  $(Q, t/\lambda)$ . The dash line is the reduced critical thickness for the stripe

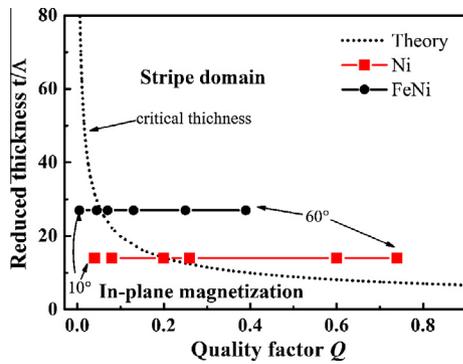


Fig. 6. The position of our samples in the phase diagram of  $(Q, t/\lambda)$ .

domain nucleation given by Eq. (1) [30]. Taking the exchange constant  $A$  for Ni ( $0.6 \times 10^6$  erg/cm) and permalloy ( $1 \times 10^6$  erg/cm) into Eq. (1), the samples locate on horizontal lines in the phase diagram as the film thickness keeps unchanged with Ni films 100 nm and permalloy films 220 nm. The  $Q$  factor of the films increases with increasing tilted angle of substrates, which originates from increased perpendicular anisotropy with larger deposition angle, as shown in Fig. 5. The experimental data agree well with theory critical thickness, for small deposition angles, the films locate below the critical line and show in-plane magnetization domains; for larger deposition angles, the films locate above the critical line and stripe domains are observed, which is in consistent with MFM measurement as shown in Fig. 2. Thereby, tilted substrates sputtering is an effective way to changing the domain structure in thin films. Comparing with the former works in Refs. [9–16] which controlled the domain structure by just changing the film thickness, we realized the controlling the domain structure in sputtered thin films at the same thickness by simply changing the oblique angle of substrates.

#### 4. Conclusions

In summary, we investigated the magnetic domain structure of films deposited on a tilted substrate. We demonstrated that the films showed domain structure evolution from in-plane domains to stripe-like domains, and then bubble-like domains with increasing the tilted angle of the substrates. Out of plane FMR was used to extract the value of perpendicular anisotropy. We found that the perpendicular magnetic anisotropy is responsible for the domain structure evolution. The tuning of domain structure was achieved in both of negative magnetostriction constant Ni films and near-zero magnetostriction constant permalloy films. Therefore, we concluded that the shape anisotropy played a major role to the

enhancement of perpendicular anisotropy, which was confirmed by multilayers films that hinder the growth of columnar structure.

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