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Fabrication of $Co_{90}Zr_{10}$ thin films with adjustable resonance frequency from 1.8 to 7.1 GHz

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A B S T R A C T

A series of $Co_{90}Zr_{10}$ thin films deposited at different oblique angles from 10° to 70° was prepared by radio frequency magnetron sputtering. The static magnetic properties and high frequency characteristics were investigated by measuring the hysteresis loops and microwave permeability spectra. The static magnetic results revealed that $Co_{90}Zr_{10}$ thin films prepared by oblique sputtering possessed a well-defined in-plane uniaxial magnetic anisotropy. It is found that the in-plane uniaxial magnetic anisotropy increased with the increasing oblique angle with a peak at 55° and then dropped rapidly above this angle. This behavior was qualitatively explained to be a result of the microstructure due to self-shadowing effect. Continuous modification of the resonance frequency in the range of 1.82–7.13 GHz has been achieved, which is essential in high frequency applications of soft magnetic thin films.

1. Introduction

Recently, there has been an increasing demand for soft magnetic films in high frequency applications such as soft under layers for perpendicular media, magnetic recording write heads and thin film wireless inductor cores [1–7]. For these kinds of application, high initial permeability (μ_i) and large cut-off frequency of the material is important. The in-plane uniaxial magnetic anisotropy (IPUMA) plays a key role in the physics of magnetic thin films since it affects the high frequency magnetization precession and consequently the high frequency behavior. Previous theoretical and experimental studies have shown that both μ_i (the real component of the permeability at zero frequency) and resonance frequency (f_r) are determined by the IPUMA field (H_k), when the saturation magnetization (M_s) is fixed [8]. Thus, one can optimize μ_i and f_r by tailoring the magnitude of IPUMA.

Several methods that have been applied to induce the IPUMA of ferromagnetic thin films [11–18]. The methods frequently employed are applying DC magnetic field during film deposition and/or anneal [9,10], and deposition on pre-stressed substrates [11,12]. However, it is found that the amplitude of the induced IPUMA is independent on the intensity of the applied magnetic field. On the other hand, deposition on pre-stressed substrates can adjust the intensity of IPUMA, but it is difficult to employ in those films deposited on hard substrates. Oblique sputtering deposition method is an effective way to alter the IPUMA and thus the resonance frequency in a wide range [13–18]. Moreover, introduction of transition element can grade the soft magnetic properties by enhancing the exchange coupling between the grains [13,18]. In our previous work, based on oblique deposition and heavy transition element doping, we obtained soft magnetic films with continuous adjustable IPUMA and excellent high frequency respond in a wide frequency range [13]. In this work, we would concentrate on the films deposited at larger oblique angle in order to see how large the amplitude can be obtained and to understand the origin of the IPUMA. In this work, we have fabricated a series of Zr doped Co thin films by oblique deposition with the oblique angle from 10° to 70°. The static magnetic properties and high frequency characteristics of $Co_{90}Zr_{10}$ films prepared by oblique sputtering method have been investigated.

2. Experimental

$Co_{90}Zr_{10}$ soft magnetic thin films with thickness ~50 nm were prepared by radio frequency sputtering onto $10 \times 20\ mm^2$ Si (111) substrates attached to a water-cooling system. Deposition rate at different oblique angles was verified by a thickness profile meter (Vecco Dektak 8). Deposited time was adjusted to make sure all the samples had the same thickness at different oblique angles. Deposition background pressure was lower than 2×10^{-5} Pa. A Co target on which Zr chips were placed in a regular manner was used to deposit $Co_{90}Zr_{10}$ films, as shown in Fig. 1. The composition of the films was adjusted by controlling the number of Zr chips. The radio frequency power density was 1.7 W/cm^2. Films were deposited at oblique angle ranging from 10° to 70° without applied magnetic field, as shown in Fig. 1. The static magnetic measurements were performed by a vibrating sample

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magnetometer (Lakeshore model 7304). The value and direction of H_k was verified by calculating the measured easy axis and hard axis loops of the reduced magnetization [19],

$$H_k = 2\int_0^{H_{up}} [m_{ea}(H) - m_{ha}(H)]\mathrm{d}H$$

where H is applied field, m_{ea} and m_{ha} means the reduced magnetization ($m = M/M_s$) taken from the measured easy and hard axis loops. The upper integration boundary H_{up} was chosen higher than the saturation field. This is an effective method to determine H_k for our $Co_{90}Zr_{10}$ thin films with IPUMA. Surface and cross-sectional images were obtained with the help of scanning electron microscope(S-4800). The composition of the films was determined by energy dispersive X-ray spectroscope (EDS). The microwave permeability measurements of the films were carried out with a vector network analyzer (E8363B) using the shorted microstrip method [20] from 1.0 to 9.0 GHz.

3. Results and discussion

A typical surface morphology of the $Co_{90}Zr_{10}$ films is shown in Fig. 2(a). Due to the instrument limit, it is difficult to observe the cross-sectional imaging of the tilted column structure of the films with thickness ∼50 nm. Thus we carried out cross-sectional images of $Co_{90}Zr_{10}$ thin films with thickness of ∼200 nm deposited at 60° are given in Fig. 2, which has the similar static magnetic and high frequency properties. It is indicated from Fig. 2(a) that $Co_{90}Zr_{10}$ films are nanocrystalline with its particle size about 10 nanometers. Thin films fabricated by oblique deposition are consist of an array of parallel columns of higher density separated by a network of pores due to limited mobility of impinging atoms and to local self-shadowing [21]. Fig. 2(b) shows the tilted column crystal of the incidence plane. The magnetic properties are strongly related to the special structure. The composition of the films versus different oblique angles is shown in Fig. 2(c). It is demonstrated that the composition of Co and Zr elements basically remains the same.

The results of the static magnetic measurements revealed that the $Co_{90}Zr_{10}$ thin films possess a well-defined IPUMA. Fig. 3(a) and (b) shows in-plane hysteresis loops along easy and hard axis in films deposited at the oblique angle 10° and 55°, which have typical hysteresis loops for all our $Co_{90}Zr_{10}$ thin films. The easy axis hysteresis loop shows a good rectangle, and the hard axis hysteresis loop is basically a closed curve without hysteresis. The difference between the hysteresis loop along easy axis and hard axis indicates an IPUMA. The values of H_k were verified by calculating the measured easy axis and hard axis hysteresis loops of the reduced magnetization as shown above. Residual magnetization (M_r) was obtained by reading the value of magnetic moment at zero magnetic field from the hysteresis loops. Fig. 3(c) and (d) gives residual magnetization ratio (M_r/M_s) and coercivity (H_c) values as a function of the oblique angle. The values of M_r/M_s along the easy axis are close to 1.0, and that of M_r/M_s along the hard axis are close

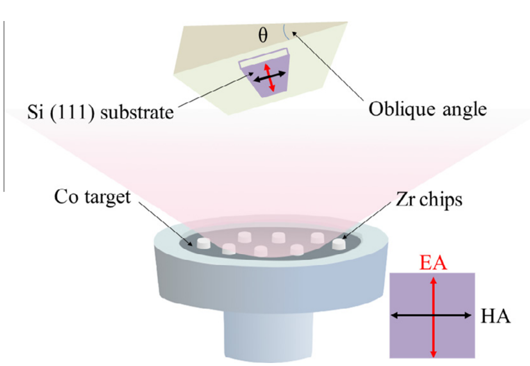

Fig. 1. Schematic drawing of the sputtering arrangement. A Co target, on which several Zr chips were placed in a regular manner, was used. Films were deposited with an oblique angle from 10° to 70°.

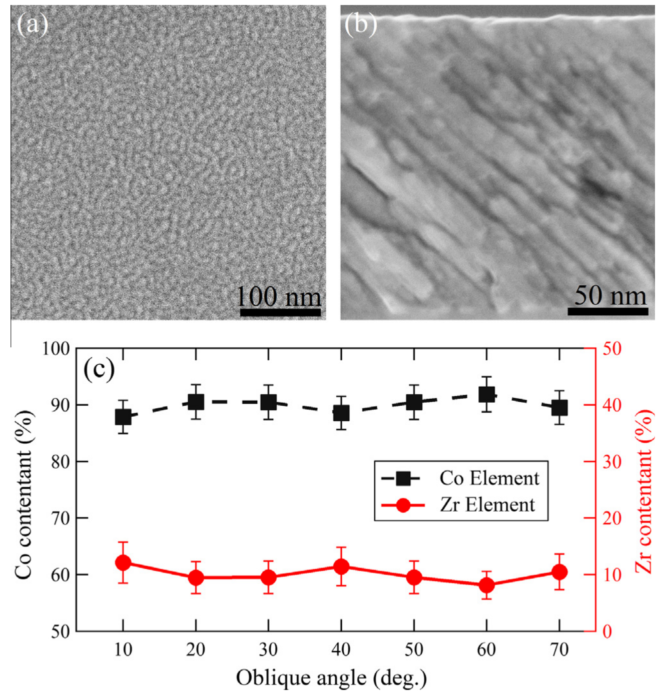

Fig. 2. (a) The SEM surface morphology and (b) cross-sectional images of $Co_{90}Zr_{10}$ thin films with total thickness of 200 nm deposited at 60°. (c) Co and Zr composition of the samples deposited at different oblique angles.

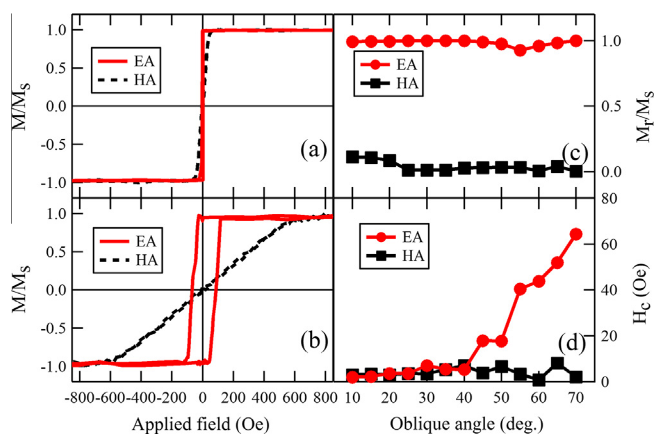

Fig. 3. Hysteresis loops of $Co_{90}Zr_{10}$ films with the oblique angle of (a) 10° and (b) 55°. Oblique angle dependence of easy axis (red) and hard (black) axis (c) remanent magnetization ratio and coercivity of $Co_{90}Zr_{10}$ films. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

to zero with a max value of 0.2. That is to say, almost all the magnetic moments in $Co_{90}Zr_{10}$ thin films are aligned along the easy axis in the demagnetized state due to the well-defined IPUMA. On the other hand, the H_c along hard axis is lower than 10 Oe for all our $Co_{90}Zr_{10}$ thin films, and the H_c along easy axis increases with the increasing oblique angle, which is a common result in oblique deposited thin films [22]. All the above static magnetic measurements results have revealed that the as-deposited $Co_{90}Zr_{10}$ thin films have good soft magnetic properties and possess a well-defined IPUMA.

Fig. 4(a) shows hard axis hysteresis loops of films deposited at various oblique angles. The saturation field first increases and then decreases with the increasing oblique angle. The H_k versus oblique angle has the same behavior with the saturation field, as the H_k of

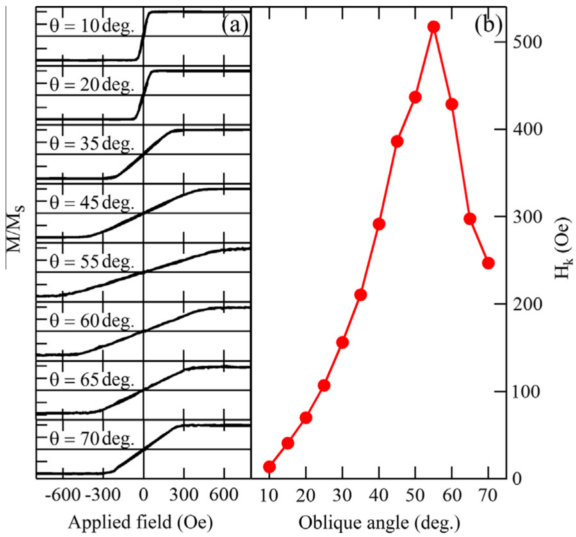

Fig. 4. (a) Hysteresis loops of hard axis of $Co_{90}Zr_{10}$ films with the various oblique angles. (b) Dependence of H_k on incidence angle.

thin films with well-defined IPUMA has the same magnitude as saturation field along the hard axis. Fig. 4(b) exhibits the H_k of $Co_{90}Zr_{10}$ films deposited at different oblique angles from 10° to 70°. The results shows that H_k of $Co_{90}Zr_{10}$ films increases from 13.6 to 517.6 Oe as the oblique angles variation from 10° to 55° and then decreases to 246.9 Oe as the oblique angle increases to 70°. It is noticed that the value of the H_k does not increase monotonically with the increasing oblique angle at higher angles, which needs further discussion.

Based on previous reports, two possible effects may contribute to the IPUMA in oblique deposited thin films: (1) anisotropic stress in combination with the isotropic magnetostriction [23], and (2) the column chain structure developed in the film plane due to the so-called self-shadowing effect [23,24]. From the cross-sectional images above, it is a reasonable conjecture that the IPUMA is mainly a result of column chain structure developed in the film plane due to self-shadowing effect. These parallel columns developed in thin films fabricated by oblique deposition tend to elongate and join into chains perpendicular to the plane of incidence due to the self-shadowing effect, which becomes more and more apparent with the angle increasing up to 60°. At higher angles, the column chains

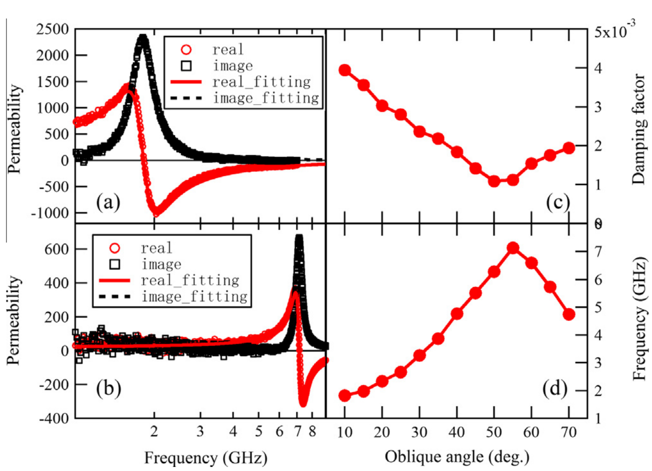

Fig. 5. Permeability spectra of $Co_{90}Zr_{10}$ films with the oblique angle of (a) 10° and (b) 55°. Dependence of (c) damping factor and (d) resonance frequency on oblique angle.

begin to disappear due to the extreme shadowing. Therefore, the H_k rising from the special column structure increases before about 60° and then shows a dramatic drop at higher angles, which is consistent with the H_k behavior in our $Co_{90}Zr_{10}$ thin films. The origin of the IPUMA is the shape anisotropy in respect with the special microstructure in the oblique deposited $Co_{90}Zr_{10}$ thin films. This model qualitatively explains the H_k behavior in $Co_{90}Zr_{10}$ thin films.

Fig. 5(a) and (b) shows the complex permeability of films with oblique angle of 10° and 55° (dots) and fitting data (solid and dashed lines). The $Co_{90}Zr_{10}$ thin films exhibit excellent high frequency properties. The oblique angle dependence of effective damping factor and resonance frequency is shown in Fig. 5(c) and (d). The f_r is proportional to the square root of M_s multiplied by H_k, $f_r \sim (M_sH_k)^{1/2}$ [25]. In this frame, resonance frequency of $Co_{90}Zr_{10}$ thin films should have the same trend with $H_k^{1/2}$, which has been observed in Fig. 5(d). Resonance frequency first increases with the increasing oblique angle with a peak at 55° and then drops rapidly above this angle. The values of f_r in $Co_{90}Zr_{10}$ thin films deposited at 10°, 55° and 70° are 1.82, 7.13 and 3.51 GHz, respectively. The effective damping factor is obtained by fitting the complex permeability equations derived from LLG equation [26]. The values of damping factor is lower than 2×10^{-3} for sample deposited at 55°. The effective damping factor of thin films first decreases and then increases with the increasing oblique angle. The behavior of the effective damping factor shows a reversal trend with the H_k. The damping factor is usually discussed in terms of intrinsic and extrinsic processes. The intrinsic ones are referred to Gilbert damping which is isotropic and depends on the local electric/magnetic environment of the films. The extrinsic damping is determined by the inhomogeneity rising from either the surface morphology or the spatial variation of magnetic moments, which lead to a superposition of slightly shifted absorption peaks. This variation can be partly suppressed by the IPUMA field H_k in $Co_{90}Zr_{10}$ films, and the magnetic moments are more inclined to arrange along the easy axis in thin films with higher IPUMA field. This interprets the negative correlation between the effective damping factor and the in-plane uniaxial magnetic anisotropy field.

4. Conclusions

In summary, $Co_{90}Zr_{10}$ soft magnetic thin films with adjustable IPUMA were fabricated by radio frequency sputtering with an oblique angle from 10° to 70°. The H_k can be varied in the range from 13.6 to 517.5 Oe; as a result, continuous modification of the resonance frequency of the films in the range of 1.82 to 7.13 GHz has been achieved. The H_k of the $Co_{90}Zr_{10}$ thin films first increases and then decreases with the increasing oblique angle, which is qualitatively explained to be a result of the special column microstructure. The effective damping factor and the in-plane uniaxial magnetic anisotropy field shows a negative correlation. Oblique sputtering method controlling over the magnitude of the in-plane uniaxial magnetic anisotropy is essential in high frequency applications of soft magnetic thin films. Our work provides a feasible approach to conveniently control the in-plane uniaxial magnetic anisotropy of thin films with certain compositions, which can further promote the applications of soft magnetic thin films in the gigahertz region.

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