Enhancement of ferromagnetic resonance in Al_2O_3-doped Co_2FeAl Heusler alloy film prepared by oblique sputtering*

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Large and variable in-plane uniaxial magnetic anisotropy in a nanocrystalline $(Co_2FeAl)_{97.8}(Al_2O_3)_{2.2}$ soft magnetic thin film is obtained by an oblique sputtering method without being induced by magnetic field or post annealing. The in-plane uniaxial magnetic anisotropy varies from 50 Oe to 180 Oe (1 Oe = 79.5775 A·m^{-1}) by adjusting the sample's position. As a result, the ferromagnetic resonance frequency of the film increases from 1.9 GHz to 3.75 GHz.

Keywords: uniaxial magnetic anisotropy, oblique sputtering, ferromagnetic resonance, nanocrystalline thin films

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1. Introduction

Recently, there has been an increasing demand for magnetic thin films in high frequency application, such as magnetic recording heads, wireless inductor cores, and microwave noise filters.[1–7] High frequency soft magnetic films working in the GHz band require high ferromagnetic resonance frequency $f_{\rm FMR}$,

$$f_{\rm FMR} = \frac{\gamma}{2\pi}\sqrt{H_{\rm k}\cdot(H_{\rm K}+4\pi M_{\rm S})}, \tag{1}$$

where γ is the gyromagnetic ratio, $M_{\rm S}$ is the saturation magnetization, and $H_{\rm K}$ is the in-plane uniaxial magnetic anisotropy (IPUMA).[8] Equation (1) demonstrates that increasing the saturation magnetization $M_{\rm S}$ and/or the in-plane uniaxial magnetic anisotropy $H_{\rm K}$ is an effective way to obtain high ferromagnetic resonance frequency. Since $H_{\rm K}$ is easy to improve and its magnitude can be enhanced by one or two orders, most studies focus on these methods to enhance the IPUMA, such as *in-situ* magnetic deposition,[9] post-annealing in a magnetic field,[10] composition gradient deposition,[11–13] face target sputtering,[14] oblique sputtering,[15,16] etc.

In this work, nanocrystalline $(Co_2FeAl)_{97.8}(Al_2O_3)_{2.2}$ soft magnetic thin films with a well-defined in-plane uniaxial magnetic anisotropy are prepared by an oblique sputtering method. The value of $H_{\rm K}$ can increase from 50 Oe to 180 Oe, depending on the position of the substrate. Consequently, the value of $f_{\rm FMR}$ is enhanced from 1.9 GHz to 3.75 GHz in the nanocrystalline $(Co_2FeAl)_{97.8}(Al_2O_3)_{2.2}$ soft magnetic thin film.

2. Experimental procedure

Nanocrystalline $(Co_2FeAl)_{97.8}(Al_2O_3)_{2.2}$ soft magnetic thin films were deposited on Si (100) substrates by an oblique sputtering method. Figure 1(a) shows the schematic drawing of the oblique sputtering device. The Si (100) single crystal substrates each with 4 mm in width and 32 mm in length were pasted on the sample table with the length direction being along the radial direction of the target. The Co_2FeAl target was rotated to a certain angle with respect to the sample table, and some Al_2O_3 chips each with dimensions of 5 mm×5 mm×0.5 mm were located on the surface of Co_2FeAl target symmetrically. The central axis of symmetry of Co_2FeAl target was directed to one terminal of the sample substrate, which is denoted as N because it is near the target, and the opposite terminal is far from the target, which is denoted as F (see Fig. 1(a)). This geometry configuration will give rise to a decrease in film thickness distribution from N to F. As expected, the measured thickness values of the films decrease from 255 nm to 195 nm for the test positions moving from N to F. The $(Co_2FeAl)_{97.8}(Al_2O_3)_{2.2}$ soft magnetic thin films were deposited at 100 W for 16 min under an Ar pressure of 0.11 Pa at an argon flow rate of 5 sccm. It should be mentioned here that the film composition of $(Co_2FeAl)_{97.8}(Al_2O_3)_{2.2}$ was obtained by comparing the compositions between the films sputtered with and without Al_2O_3 chips. The sample was cut into eight segments, each with an equal length of 4 mm along the length direction,

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and the segments were sequentially numbered from $n = 1$ to 8 for the positions from N to F. The magnetic properties of segments were measured by a vibrating sample magnetometer (VSM) at room temperature. The microwave permeability values of the films were measured in a coplanar waveguide fixture by an Agilent vector network analyzer (PNA N5224A). The microstructures of the films were detected by high resolution transmission electron microscopy (HRTEM).

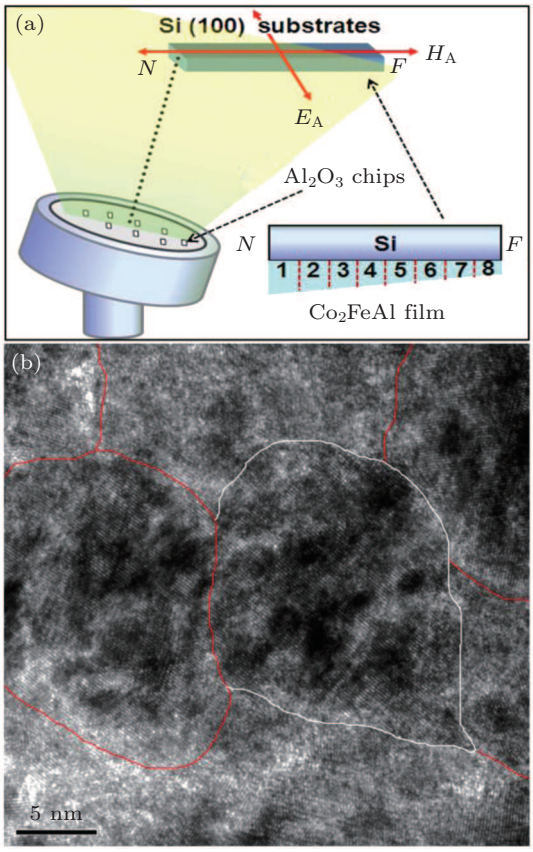


Fig. 1. (color online) (a) Schematic drawing of the oblique sputtering device and films (in the inset), and (b) HRTEM image of Al_2O_3-doped Co_2FeAl film (the inserted lines in panel (b) are for guiding the eyes, showing the profile of grains).

3. Results and discussion

The soft magnetic properties are very sensitive to the grain size of the ferromagnetic film. According to the random anisotropy model,[17,18] the coercivity will dramatically decrease according to a sixth-power relation, that is, $H_C \propto D^6$, where D is the grain size of ferromagnetic phase. Therefore, good soft magnetic properties will only be obtained in the film composed of nanocrystallines. In this study, the doping of Al_2O_3 not only increases the resistance, leading to a lower eddy current loss, but also refines the grain size to the nanoscale, resulting in a lower coercivity and lower magnetic hysteresis loss. The grain sizes of the $(Co_2FeAl)_{97.8}(Al_2O_3)_{2.2}$ films were observed by HRTEM, and the results are shown in Fig. 1(b). As illustrated, the grain size of the films is around 20 nm, which is sufficiently fine to obtain good soft magnetic properties.

Figure 2 shows the normalized hysteresis loops of the nanocrystalline $(Co_2FeAl)_{97.8}(Al_2O_3)_{2.2}$ soft magnetic thin films at various test positions. As illustrated, the films exhibit the similar values of M_S (around 10 kGs, 1 Gs = 10^{-4} T), and an obvious uniaxial magnetic anisotropy field with the hard axis (H_A) along the radial (length) direction and the easy axis (E_A) perpendicular to the radial direction (width direction). Moreover, the H_K is very sensitive to the test position n. The H_K dramatically increases from 50, 100, 130 to 180 Oe for the test positions changing from $n = 1$, 3, 5, to 7, respectively. For the variation of the H_K in oblique sputtered films, it can be attributed to the residual stress-induced magnetic anisotropy.[11–13] Since the Si substrates and the metal films have very different expansion coefficients, residual stress will appear in the ferromagnetic film after the sample is cooled from the deposition temperature (about 250 °C) to room temperature. In this study, a compressive stress is formed because the expansion coefficient of Si substrate is less than that of $(Co_2FeAl)_{97.8}(Al_2O_3)_{2.2}$ metal alloy. On the other hand, the oblique sputtering gives rise to a gradient distribution of film thickness. Thus, a position-dependent compressive stress will be achieved, which increases from N to F. According to the theory of ferromagnetism, magnetostriction energy is expressed as

$$E = -\frac{3}{2}\lambda_s \sigma \cos^2 \alpha, \tag{2}$$

where λ_S is the saturation magnetostriction coefficient, σ is the stress, whose positive and negative signs are taken for tensile and compressive stresses, respectively, and α is the angle between stress and magnetization. Based on Eq. (2), for a film with a positive λ_S, the compressive stress results in the arrangement of magnetic moments perpendicular to the compressive stress direction, and *vice versa*.[18,19] Generally, the intrinsic stress is randomly dispersed in the sample, giving rise to a high damping constant, which impedes the enhancement of the resonance frequency. However, if the intrinsic stress orientation is arranged along one direction, then a uniaxial magnetic anisotropy will be formed, which is accompanied with an enhancement of high-frequency ferromagnetic performance. For the investigated films, the uniaxial stress increasing from N to F along radial (length) direction is induced by the oblique sputtering, owing to the film thickness decreasing from N to F. The film with a positive λ_S and compressive stress direction along the radial direction has a magnetic moment that is preferentially arranged in the direction perpendicular to the radial direction, leading to an H_A along radial direction and an E_A perpendicular to the radial direction, which are shown in Figs. 1(a) and 2.

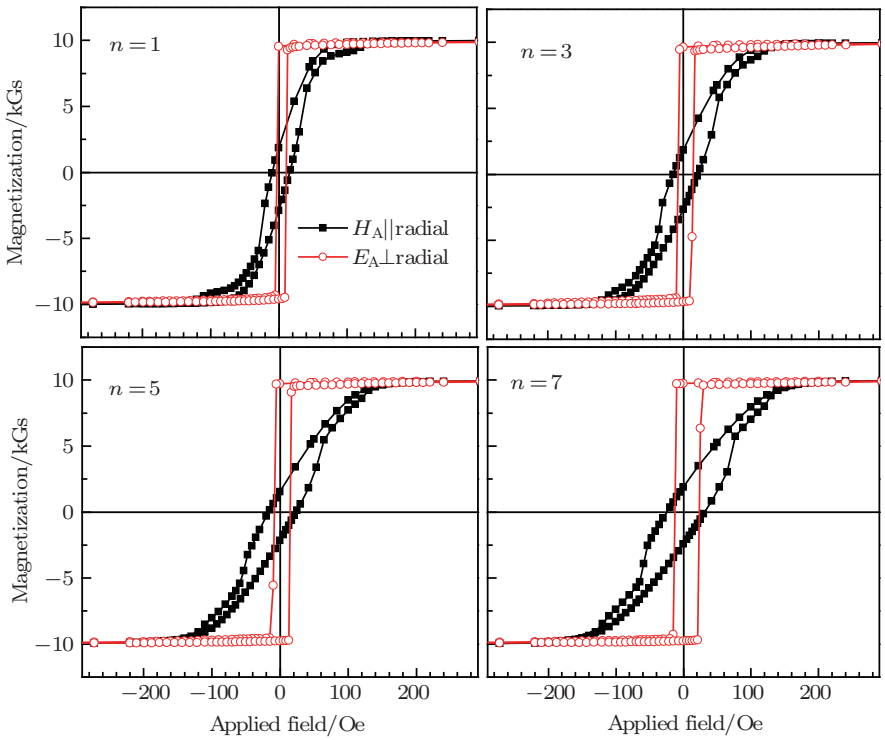


Fig. 2. (color online) Representative hysteresis loops of the nanocrystalline $(Co_2FeAl)_{97.8}(Al_2O_3)_{2.2}$ soft magnetic thin films at different positions (i.e., different values of n).

According to Eq. (1), the increase of H_K will result in an enhancement in the ferromagnetic resonance frequency f_{FMR} of the film. As expected, the f_{FMR} is dramatically enhanced from 1.9 GHz to 3.75 GHz for the test positions from N to F (see Fig. 3). Therefore, the *in-situ* manipulation of the high frequency performance of the nanocrystalline $(Co_2FeAl)_{97.8}(Al_2O_3)_{2.2}$ soft magnetic thin film can be realized by choosing the different positions. The relationship between H_K and f_{FMR} is shown in Fig. 4. As illustrated, H_K and f_{FMR} undergo the same trend, further demonstrating that the higher f_{FMR} is caused by the higher H_K.

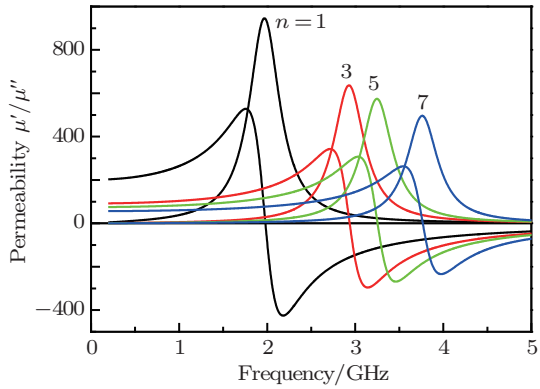


Fig. 3. (color online) Position dependences of the permeability for the nanocrystalline $(Co_2FeAl)_{97.8}(Al_2O_3)_{2.2}$ soft magnetic thin films at different positions (different values of n).

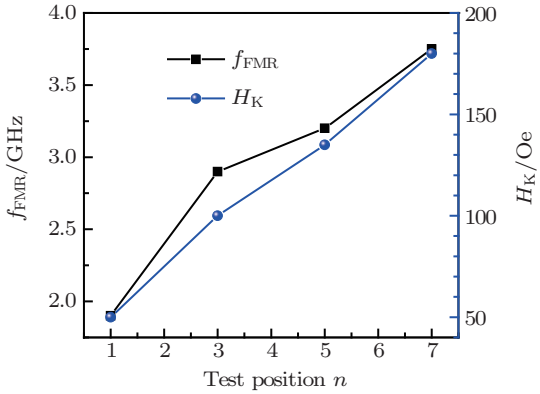


Fig. 4. (color online) Relationships between in-plane uniaxial magnetic anisotropy H_K and ferromagnetic resonance frequency f_{FMR}.

4. Conclusions

Nanocrystalline $(Co_2FeAl)_{97.8}(Al_2O_3)_{2.2}$ soft magnetic thin films with adjustable in-plane uniaxial magnetic anisotropy are *in-situ* fabricated by an oblique sputtering method. The magnetic anisotropy field of the as-deposited film, without being induced by the magnetic field or post magnetic annealing, can be enhanced from 50 Oe to 180 Oe, owing to the variation of film thickness in oblique sputtered film. As a result, f_{FMR} continuously increases from 1.9 GHz to 3.75 GHz.

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