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Microwave ferromagnetic properties of as-deposited Co_2FeSi Heusler alloy films prepared by oblique sputtering*

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The Co_2FeSi films are deposited on Si (100) substrates by an oblique sputtering method at ambient temperature. It is revealed that the microwave ferromagnetic properties of Co_2FeSi films are sensitive to sample position and sputtering power. It is exciting that the as-deposited films without any magnetic annealing exhibit high in-plane uniaxial anisotropy fields in a range of 200 Oe–330 Oe (1 Oe = 79.5775 A·m^{-1}), and low coercivities in a range of 5 Oe–28 Oe. As a result, high self-biased ferromagnetic resonance frequency up to 4.75 GHz is achieved in as-deposited oblique sputtered films. These results indicate that Co_2FeSi Heusler alloy films are promising in practical applications of RF/microwave devices.

Keywords: microwave ferromagnetic performances, oblique sputtering, ferromagnetic resonance, Heusler alloy

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1. Introduction

With the continuous development of information science and technology, new materials are urgently needed. The miniaturization of electronic devices, the increase of the data transmission rate and the high-capacity of circuit chips encourage researchers to constantly explore new functional materials. At present, the operating frequencies of electromagnetic devices (e.g. mobile and bluetooth) have reached the GHz band. Microwave soft magnetic films working in the GHz band require a high ferromagnetic resonance frequency f_{FMR}, which can be expressed as follows:

$$f_{FMR} = \gamma \cdot \sqrt{H_K \cdot (4\pi M_S + H_K)}, \quad (1)$$

where γ is the gyro-magnetic ratio close to 2.8 MHz/Oe, H_K the in-plane uniaxial anisotropy field, and M_S the saturation magnetization.[1,2] From Eq. (1), it is found that high M_S and appropriate H_K are required to achieve a high ferromagnetic resonance frequency of the magnetic film. It is obvious that the magnetic material working in the GHz band should have a high M_S and an appropriate H_K.[3,4] The researches on microwave performance have mostly focused on soft magnetic films, especially FeCo-based alloy.[5–7] Although FeCo-based alloy material has the largest M_S in ferromagnetic 3d transition metal alloys, its large magnetocrystalline anisotropy and magnetostriction restrict its microwave applications.[7] Recently X_2YZ-type Heusler alloys, such as Co_2FeSi, Co_2MnSi, etc., have attracted widespread attention because they are promising candidates as half-metallic ferromagnets.[8,9] These alloys are of an unusual combination of metallic and insulating properties,[10] which is attributed to the coexistence band structure of metal and insulation. Co_2FeSi Heusler alloy has a high Curie temperature ($T_C = 1100$ K) and high magnetic moment (i.e., 6 μ_B/f.u.).[11] At the same time, Co_2FeSi Heusler alloy has itinerant-electron characteristics, resulting in a small magnetic damping constant α, and a high magnetic anisotropy field H_K.[11,12]

The Co_2FeSi Heusler alloy exhibits such advantages that it is promising in microwave application. However, reports on the microwave ferromagnetic properties of Co_2FeSi Heusler alloy are very rare.[11] In the present study, an oblique sputtering method is employed to prepare Co_2FeSi Heusler alloy films at room temperature. Good microwave ferromagnetic performances are achieved in as-deposited Co_2FeSi Heusler alloy films without any magnetic annealing.

2. Experimental procedure

Figure 1(a) shows the schematic drawing of a radio frequency (RF) oblique sputtering device. Silicon (100) single crystal substrates (4 mm×84 mm) were placed on the sample table with the length direction along the radial direction of the target. The Co_2FeSi target was rotated at a certain angle with respect to the sample table. The central axis of the symme-

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try of the Co_2FeSi target is directed to one terminal of sample substrate (named A), and the opposite terminal (named B) is far from the target. This geometry configuration will give rise to a film thickness distribution decreasing from A to B. As expected, the measured thickness values of the films decrease from 195 nm to 43 nm for the test position from A to B (see Fig. 2(a)). A similar description of the oblique sputtering device was reported elsewhere.[2,13,14]

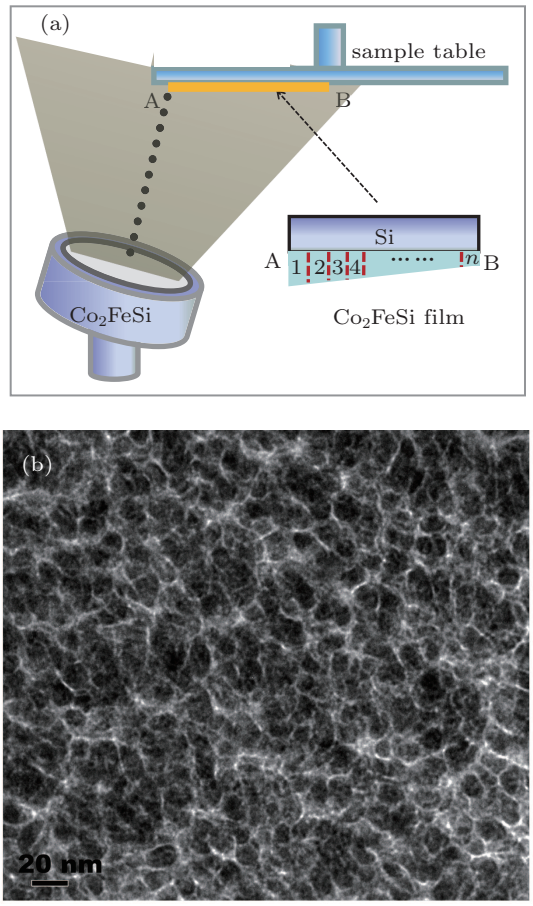


Fig. 1. (color online) (a) Schematic drawing of the oblique sputtering device, and (b) the typical TEM image of sample.

The Co_2FeSi Heusler alloy films were deposited on Si (100) substrates using an oblique sputtering method at an ambient temperature. The background pressure of the chamber was lower than 3.0×10^{-4} Pa, and the working pressure was maintained at 0.1-Pa Ar atmosphere with a flowing rate of 4.5 sccm. All the samples were deposited for 25 min at RF powers of 60 W–100 W. The sample was cut into 21 segments each with a length of 4 mm along the length direction, and the segments were sequentially numbered from $n = 1$ to 21 for the position from A to B. The magnetic properties of each segment were measured by a vibrating sample magnetometer (VSM) at room temperature. The microwave permeabilities of the films were measured by an Agilent vector network analyzer (PNA N5224A). The microstructures of the films were detected with an X-ray diffractometer (XRD), and observed by transmission electron microscopy (TEM).

3. Results and discussion

3.1. Soft magnetic properties and nanocrystalline Co_2FeSi films

The soft magnetic properties are very sensitive to the grain sizes of the ferromagnetic films. The coercivity will dramatically decrease according to a six-power relation. This phenomenon has been well explained by use of random anisotropy mode.[15,16] Based on the random anisotropy theory, good soft magnetic properties will be obtained only in the film composed of nanocrystallines. The grain size can be estimated by the Scherrer equation,

$$D = \frac{\kappa\lambda}{\beta\cos\theta}, \tag{2}$$

where D is the grain size, κ is the Scherrer constant, λ is the X-ray wavelength, β is the line broadening at full width at half maximum (FWHM) intensity, and θ is the Bragg angle. The grain sizes of Co_2FeSi films, calculated by the Scherrer equation, are around 15 nm on average. Similarly, the TEM image shown in Fig. 1(b) also demonstrates that the grain size of Co_2FeSi film is around 15 nm on average. As discussed below, the grain size of the film prepared by oblique sputtering is fine enough to achieve a good soft magnetic property.

3.2. Position dependence of magnetic properties

The oblique sputtered Co_2FeSi films each exhibit a strong position dependence of magnetic properties. Figure 2 shows the variations of magnetic properties and thickness with sample position n for the oblique sputtered Co_2FeSi films at an RF power of 80 W. As illustrated in Fig. 2(a), the film thickness values from A to B dramatically decrease from 195 nm to 43 nm due to the oblique sputtering. The saturation magnetization $4\pi M_S$ of the film stays about 9.6 kGs (1 Gs = 10^{-4} T), but it decreases when the film thickness is less than 60 nm ($n > 18$). Moreover, the thinner the film, the more the $4\pi M_S$ decreases. This fact demonstrates that the reduction of $4\pi M_S$ in thinner film is caused by the magnetic moment disorder at the surface and the interface of Co_2FeSi film. The reduction of $4\pi M_S$ will deteriorate the microwave ferromagnetic performance.

Each of the as-deposited nanocrystalline Co_2FeSi films, shown in Fig. 2(b), exhibits a small coercivitiy H_C in the range 5 Oe–28 Oe due to the random anisotropy interaction between the nanocrystalline grains as discussed above.[15,16] Especially, H_C is smaller than 20 Oe for the sample positions $n \leq 18$, indicating good microwave ferromagnetic properties with lower magnetic loss present in these samples. In addition, it is exciting that the values of anisotropy field H_K of the

films continuously increase from 40 Oe to 265 Oe from sample positions $n = 1$ to $n = 21$. From Fig. 2(b), one can also see that the value of H_K only slightly increases, but the H_C is rapidly enhanced after $n > 18$. These facts imply that the microwave ferromagnetic performance will be deteriorated when $n > 18$.

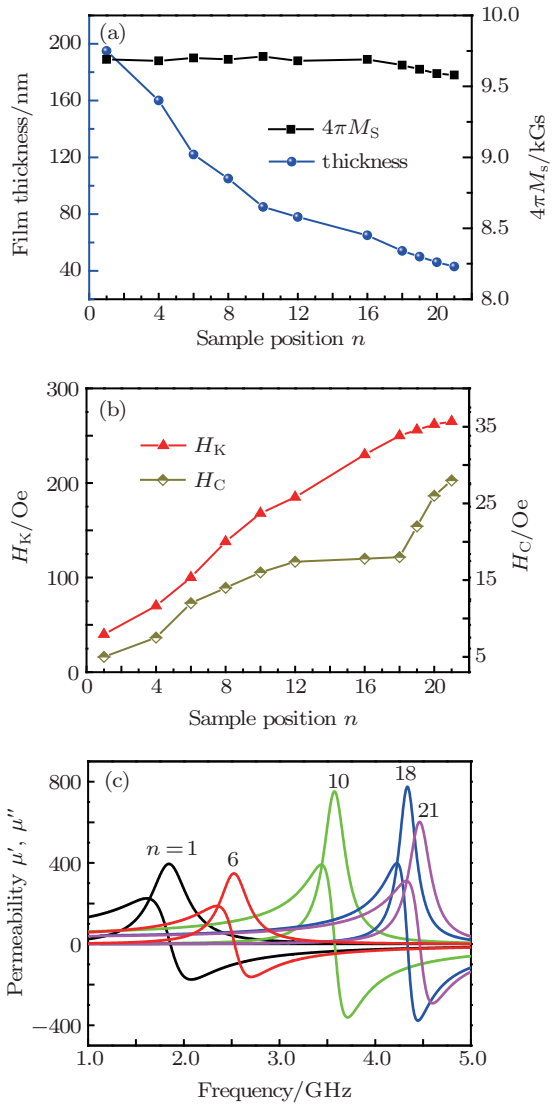


Fig. 2. (color online) Sample position dependences of (a) film thickness and $4\pi M_S$, (b) H_C and H_K, and (c) the permeability spectra.

Figure 2(c) shows the sample position dependences of permeability spectra. As illustrated, the ferromagnetic resonance frequency f_{FMR} of obliquely sputtered Co_2FeSi film increases monotonically with the increase of n. As expected, the sample $n = 18$ exhibits the best comprehensive microwave ferromagnetic properties, such as relatively large $4\pi M_S$ (9.65 kGs), larger H_K (250 Oe), lower H_C (18 Oe), higher f_{FMR} (4.34 GHz), the lowest ferromagnetic linewidth, and lower ferromagnetic resonance loss. For the samples with $n > 18$, although they have slightly higher values of f_{FMR} than that of sample $n = 18$, they have wider ferromagnetic linewidths and magnetic losses, which will restrict their applications in microwave devices. Thus, the sample position $n = 18$ is the optimal one where the optimal comprehensive microwave ferromagnetic performances are achieved.

3.3. Effect of RF power on the microwave ferromagnetic properties

The magnetic properties and permeability spectra of the obliquely sputtered Co_2FeSi films are very sensitive to the sputtering power. The magnetic properties of the sample at position $n = 18$ are chosen to compare the magnetic properties for each of Co_2FeSi films sputtered at various powers. The RF power dependences of H_C, H_K, and the permeability spectra are summarized in Fig. 3. As shown in Fig. 3(a), the H_C is around 20 Oe for different films, while the H_K increases from 200 Oe to 330 Oe with the increase of RF power from 60 W to 100 W, indicating that the microwave ferromagnetic properties can be improved by increasing the sputtering power. The permeability spectra shown in Fig. 3(b) verify that the microwave ferromagnetic properties are enhanced by increasing the RF power. The ferromagnetic resonance frequency monotonically increases from 3.78 GHz to 4.75 GHz with the increase of RF power from 60 W to 100 W.

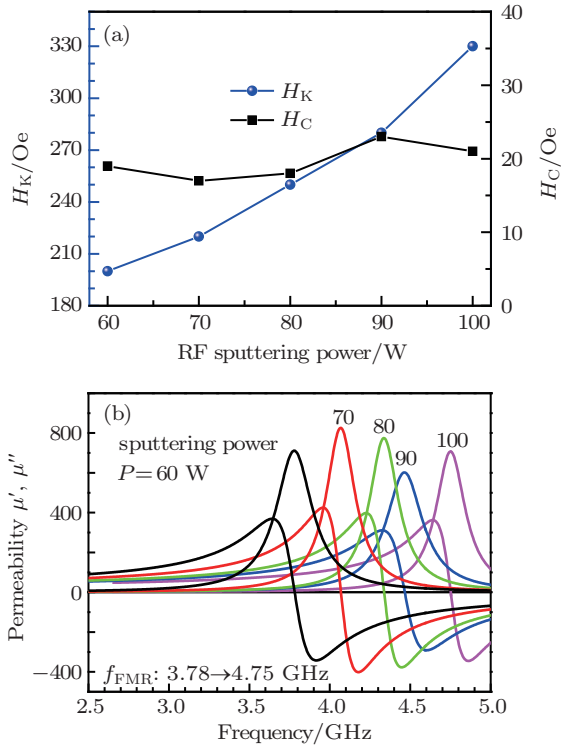


Fig. 3. (color online) (a) RF power dependences of H_C and H_K and (b) permeability spectra for samples at position $n = 18$.

4. Conclusions

Nanocrystalline Co_2FeSi Heusler alloy thin films with good microwave ferromagnetic performances are prepared by an obliquely sputtering method at an ambient temperature. It is revealed that the magnetic properties and microwave performances of the obliquely sputtered Co_2FeSi thin films are

very sensitive to the position and sputtering power. The as-deposited Co_2FeSi film without post-annealing exhibits lower coercivity (less than 28 Oe), and higher in-plane uniaxial anisotropy field (as high as 200 Oe–330 Oe). As a result, good microwave ferromagnetic properties with the ferromagnetic resonance frequency up to 4.75 GHz are obtained. These beneficial features of obliquely sputtered Co_2FeSi thin films make it possible to use such films in RF/microwave devices.

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