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Letter

Dependence of soft magnetic properties and high frequency characteristics on film thickness for $Ni_{75}Fe_{25}$–ZnO nano-granular films

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ABSTRACT

$(Ni_{75}Fe_{25})_{69}(ZnO)_{31}$ granular films with various thicknesses were deposited by magnetron co-sputtering. The dependence of soft-magnetic properties and high frequency characteristics on film thickness was investigated systematically. Static magnetic research exhibits that thin films showed evident in-plane uniaxial magnetic anisotropy. And an interesting phenomenon was found that the magnetic anisotropy field H_k increases with increasing film thickness t, which is not found in previous reports. However, no obvious variation with film thickness t was found for the coercivity (H_{ce}) in easy axis and the coercivity (H_{ch}) in hard axis. The measurements of high frequency characteristics show that the ferromagnetic resonance frequency f_r increases with increasing film thickness t, which is ascribed to the increase of the magnetic anisotropy field H_k. The films fabricated with appropriate thickness exhibits good high frequency properties and can be applied in the sensors operated in high frequency range.

1. Introduction

Soft-magnetic thin film materials have attracted much attention [1–8] and are extensively incorporated in many high frequency applications, such as soft-magnetic underlayers in recording media, micro-magnetic devices and magnetic sensors. The basic requirements for these films operated in the GHz range are high permeability μ, high saturation magnetization M_S, appropriate large anisotropy field H_k, and high electric resistivity so as to effectively suppress eddy current loss and to possess high ferromagnetic resonance (FMR) frequency f_r (which determines the cut-off frequency for high frequency application). Metal insulator granular films (MIGF) consisting of magnetic metal nanogranules uniformly distributed in an insulator matrix are one of the best candidates for satisfying above demands. In the past decade, soft magnetic properties of K–M–N, K–M–O and K-fluoride films have been reported, where K are magnetic metals (Fe, Co, Ni, and their alloys) and M are nonmagnetic elements, such as Al, Si, Zr, Hf, etc. [4–8].

Comparing with that a lot of study performed in the MIGF systems, little definite work has been conducted to optimize the soft-magnetic properties and the high frequency characteristics of another systems fabricated by making use of the semiconductor materials and ferromagnetic metal. Because of the high resistivity and special transport properties of semiconductors such as ZnO, SnO_2, TiO_2, metal-semiconductor granular films (MSCGF) consisting of magnetic metal nano-granules uniformly distributed in a semiconductor matrix also possess good soft magnetic properties and high frequency properties. In our group, some work had been done to investigate the soft magnetic and high frequency properties of the MSCGF systems, and study results show good soft magnetic properties and high frequency properties in the system $(Ni_{75}Fe_{25})_x(ZnO)_{1-x}$. For the typical sample $(Ni_{75}Fe_{25})_{69}(ZnO)_{31}$, the coercivity 1.7 Oe in the hard axis and 1.1 Oe in the easy axis are obtained, and the obvious uniaxial magnetic anisotropy is also obtained. At a frequency lower than 1.0 GHz, the real part of the complex permeability of this sample is more than 110, and the FMR frequency f_r reaches 1.3 GHz, which implies that the films is promising for high frequency applications. However, no work has been performed to study the film thickness effect on soft-magnetic and high frequency properties by tailoring the film thickness. In this work, the dependence of soft-magnetic properties and high frequency properties on film thickness is investigated systematically, which is helpful for the design of soft magnetic films used on sensors as applied in high frequency range.

2. Experiments

$(Ni_{75}Fe_{25})_{69}(ZnO)_{31}$ granular thin films with various thicknesses were deposited on water-cooled glass substrates in a magnetron co-sputtering system at room temperature, using a pure ZnO (99.99%) target of 2 in. and $Ni_{75}Fe_{25}$ alloy target of 2 in. in diameter. During depositing, ZnO and $Ni_{75}Fe_{25}$ alloy targets were both connected to rf source. Fig. 1 gives the schematic illustration of sputtering system. During deposition, both targets face the substrate with an incident angle of approximate 30° without an external magnetic field. An in-plane uniaxial magnetic anisotropy

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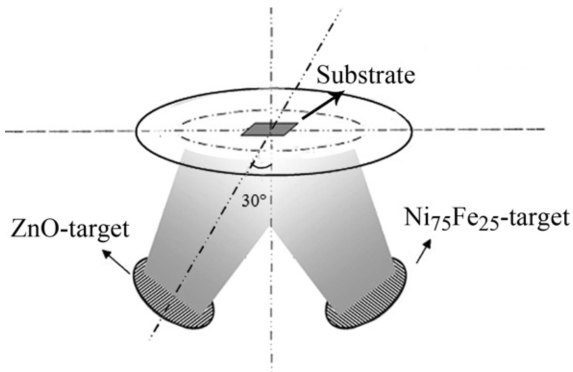

Fig. 1. The schematic illustration of sputtering system.

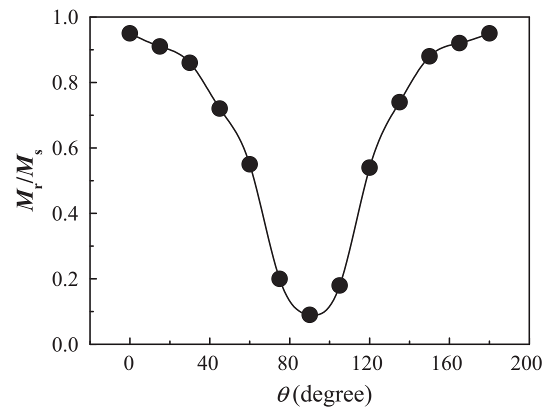

Fig. 3. The remanence ratios (M_r/M_s) variation with θ for the sample $(Ni_{75}Fe_{25})_{69}(ZnO)_{31}$.

Table 1
The sputtering conditions.

Background pressure	$<6.0 \times 10^{-4}$ Pa
The working pressure	0.12 Pa
ZnO target power	70 W
$Ni_{75}Fe_{25}$ alloy target power	100 W
Substrate	Glass slides
Deposition rate	78 Å min^{-1}

was achieved by the inclined incidence of the sputtering beams. The film thickness was altered by changing the sputtering time. The sputtering conditions are listed in Table 1.

Saturation magnetization M_S, coercivity H_c and in-plane magnetic anisotropy field H_k were measured by the vibrating sample magnetometer (VSM, Lake Shore model 7304). The thickness of the films were determined by the surface profile meter (Dektak 6M). The resistivity at room temperature was measured by standard dc four-probe method. The structure and microstructure were analysed by X-ray diffraction (XRD) with Cu Kα radiation and high resolution transmission electronic microscope (HRTEM) using a Tecnai-G2-F30 system, respectively. The microwave permeability measurements were carried out with a PNA E8363B vector network analyzer using the shorted microstrip transmission-line perturbation method, which works from 100 MHz to 5.5 GHz [9]. The measurement was performed along the hard axis in-plane of the film. All the above measurements were performed for as-deposited samples without any post heat treatment. The f_r and damping parameter α were determined by fitting the permeability spectrum with Landau–Lifshitz–Gilbert equation [10,11].

3. Results and discussion

Fig. 2 shows the bright-field HRTEM image (a) and the corresponding electron diffraction (ED) pattern (b) for a typical sample $(Ni_{75}Fe_{25})_{69}(ZnO)_{31}$. From the image (a), it can be seen that the sample consists of fcc $Ni_{75}Fe_{25}$ particles uniformly embedded in the semiconductor ZnO matrix. The average granular size as determined by HRTEM is about 4.2 nm. The gap between granules is filled with the gray ZnO medium and is about 1.0 nm. Fig. 2(b) exhibits the diffraction peaks from (1 1 1), (2 0 0), (2 2 0) and (3 1 1) planes of fcc $Ni_{75}Fe_{25}$ alloys, and there are no diffraction peaks corresponding to crystalline ZnO, which indicates that ZnO is amorphous.

Fig. 3 shows the remanence ratios (M_r/M_s) variation with the direction of the measurement magnetic field for the sample $(Ni_{75}Fe_{25})_{69}(ZnO)_{31}$, where θ represents the angle between the direction of the measurement field and that of the reference. It is seen that while θ changes from 0° to 180°, M_r/M_s only exhibits a minimum that corresponds to the direction of hard axis, indicating the existence of uniaxial anisotropy in the film. It is important to notice that although no external magnetic field is applied on the substrate during deposition, good in-plane uniaxial anisotropy obviously exists in the film, which is thought to be caused by the inclined incidence of the sputtering beams [12,13].

Fig. 4 shows the magnetic hysteresis loops measured along the easy axis and hard axis of the $(Ni_{75}Fe_{25})_{69}(ZnO)_{31}$ granular thin films with various thicknesses t. The easy axis, which is at the angle of 90° to the hard axis, is parallel to the deposition direction of atoms sputtered, which can be interpreted by the anisotropic coupling model: the magnetic coupling in the deposition direction of atoms is greater than that in the orthogonal direction [14]. All the films exhibit obvious in-plane uniaxial magnetic anisotropy, and the films with $t > 100$ nm have good rectangular ratio, namely large

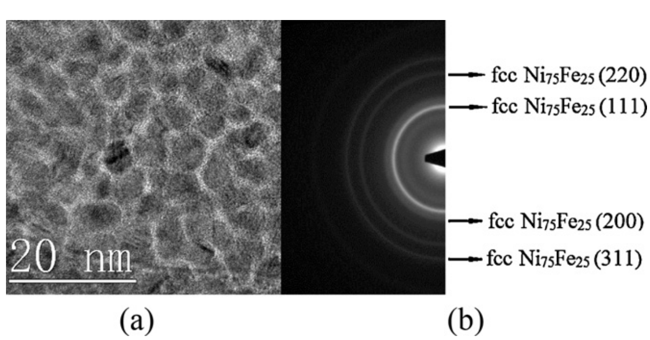

Fig. 2. HRTEM image (a) and ED pattern (b) of the $(Ni_{75}Fe_{25})_{69}(ZnO)_{31}$.

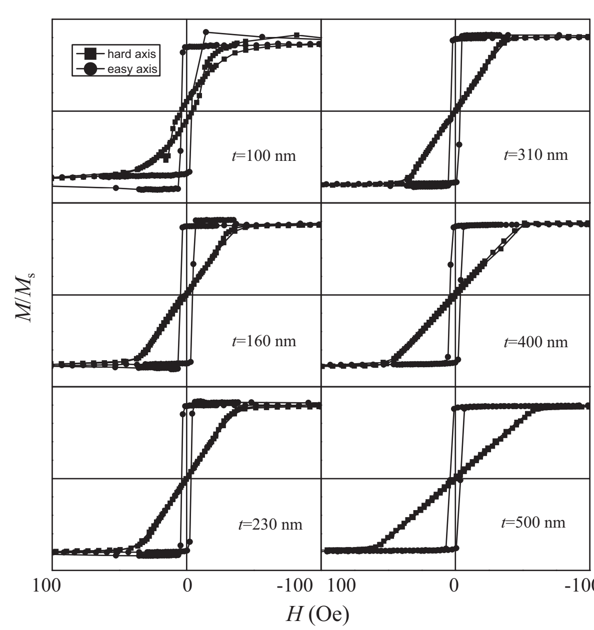

Fig. 4. M–H loops of the $(Ni_{75}Fe_{25})_{69}(ZnO)_{31}$ with various film thickness t.

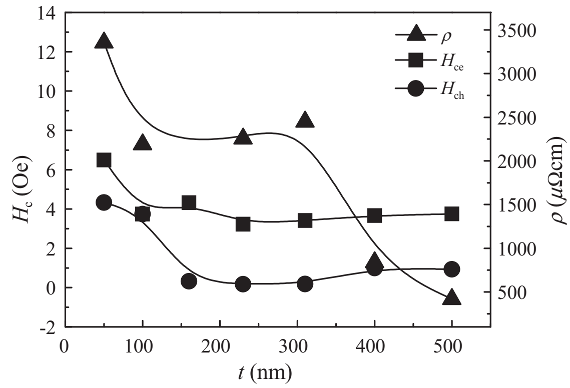

Fig. 5. The dependence of ρ, H_{ce}, H_{ch} on film thickness t.

ratio of M_r/M_s, which is important for realizing good high frequency properties [4].

Fig. 5 depicts the dependence of the electric resistivity ρ, the coercivity (H_{ce}) in easy axis and the coercivity (H_{ch}) in hard axis on film thickness t. It is can be seen that the ρ increases from 400 μΩ cm to more than 2500 μΩ cm with reducing t. This high ρ can effectively depress the eddy current loss as applied in the high frequency range. However, no obvious variation was found for H_{ce} and H_{ch} in the range of $160 < t < 500$ nm, only for the films with $t < 100$ nm, the H_{ce} and H_{ch} increases largely and the loop shape deteriorates. For the films with very small thickness, the interface effect between film and substrate is strong enough to impede the magnetization process, bringing out a large coercivity. And the increasing film thickness gradually makes the interface effect on magnetization process unimportant. Furthermore, samples are fabricated in the same sputtering conditions, the difference of the microstructure is not obvious and the mean granular size D is almost same for the samples with $160 < t < 500$ nm. Based on the random anisotropy model originally proposed by Alben et al. [15], the coercivity (H_c) is determined by the grain size (D):

$$H_c = 0.64\frac{K_1^4 D^6}{J_s A^3} \quad (1)$$

where K_1, J_s and A denote the magneto-crystalline anisotropy constant, saturation magnetization and exchange stiffness, respectively. Eq. (1) holds for ferromagnetic particles oriented at random as long as the grain size (D) is smaller than the exchange length (L_{ex}). For our samples, the mean granular size D is almost same, so, there is no obvious variation for coercivity force H_c in the range $t > 100$ nm according to Eq. (1). But for samples with $t < 100$ nm, the effect of glass substrates on magnetic properties begins to take on with reducing the film thickness t, resulting in the increase of coercivity H_c at some extent. A very interesting phenomena is found that the uniaxial magnetic anisotropy field H_k gradually increases with increasing t. As reported in some systems, films fabricated by oblique deposition often take on uniaxial magnetic anisotropy H_k and the formation mechanism of H_k was discussed [12,13,16,17]. For our system, the formation mechanism of H_k is ascribed to the thickness gradient effect caused by the inclined incidence of the sputtering beams. And with the increase of t, the thickness gradient effect is more obvious, so, the H_k increases accordingly.

The behaviour of magnetization in high frequency magnetic field can be described by the Landau–Lifshitz [11], and the dependence of complex permeability $\mu = \mu' + i\mu''$ on frequency is shown in Fig. 6. It can be seen that for samples larger than 160 nm, the real component μ' and the imaginary component μ'' of the permeability are segregative clearly, especially for the samples with $t > 300$ nm, good high frequency characteristics are obtained. For the typical sample with $t = 500$ nm, it is seen that μ' is more than

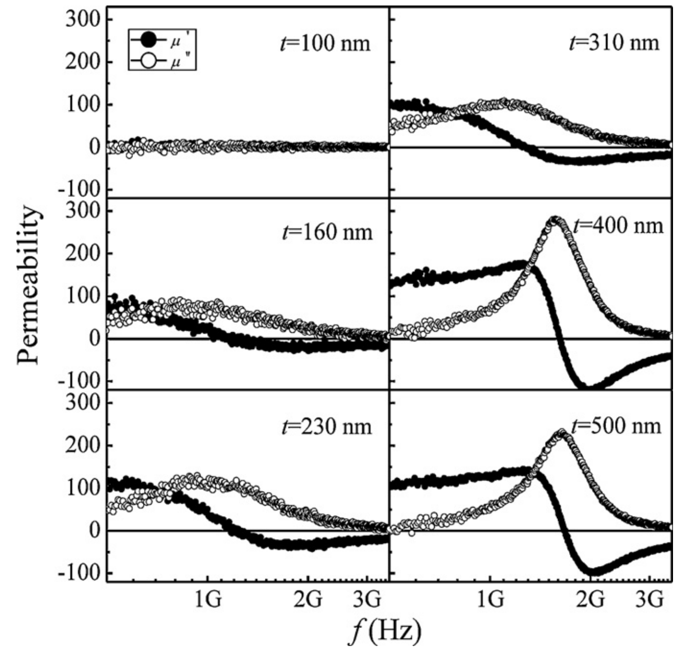

Fig. 6. The permeability spectrum of samples with different film thickness t.

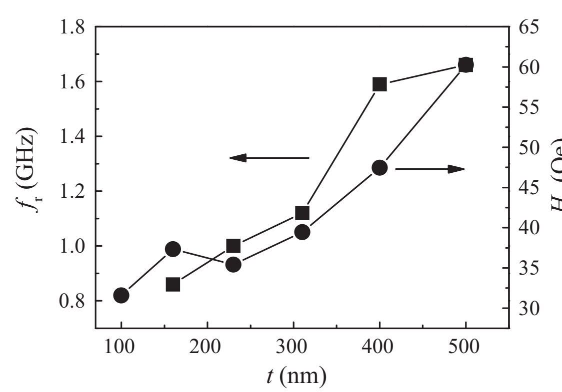

Fig. 7. The dependence of f_r, H_k on film thickness t.

110 below 1.0 GHz, while the imaginary part μ'' is much smaller than μ', which indicates that the film can be the potential materials applied on sensors operated in high frequency range. The μ'' increases to a maximum at $f = 1.65$ GHz, which can be ascribed to the ferromagnetic resonance FMR, the FMR frequency is stated as [11,18]:

$$f_r = \frac{\gamma}{2\pi}\sqrt{4\pi M_s H_k} \quad (2)$$

where γ, M_s and H_k is the gyromagnetic factor, the saturation magnetization and in-plane anisotropy field, respectively. For our samples, $4\pi M_s$ is equal for every sample, however, the H_k shown in Fig. 7 increases with increasing t, so, according to formula (2), the f_r increases gradually with the increase of film thickness t, which is shown in Fig. 7.

4. Conclusions

$(Ni_{75}Fe_{25})_{69}(ZnO)_{31}$ granular films with various thicknesses were deposited by magnetron co-sputtering. Static magnetic research exhibits that the thin films with thickness from 50 nm to 500 nm showed evident in-plane uniaxial magnetic anisotropy H_k and the H_k increases with increasing film thickness, while no obvious variation with the increase of film thickness t was found for the coercivity (H_{ce}) in easy axis and the coercivity (H_{ch}) in hard axis. For the typical sample with t = 500 nm, the coercivity 0.9 Oe in the hard axis and 3.7 Oe in the easy axis are obtained, and the resistivity reaches 418 $\mu\Omega$ cm. The measurements of high frequency characteristics show that the ferromagnetic resonance frequency f_r increases with increasing film thickness t. For the sample with t = 500 nm, at a frequency lower than 1 GHz, the real part of the complex permeability is more than 110, and the FMR frequency reaches 1.65 GHz, which implies that the films is promising for high frequency applications.

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