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Mössbauer study of the field induced uniaxial anisotropy in electro-deposited FeCo alloy films*

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Thin ferromagnetic films with in-plane magnetic anisotropy are promising materials for obtaining high microwave permeability. The paper reports a Mössbauer study of the field induced in-plane uniaxial anisotropy in electro-deposited FeCo alloy films. The FeCo alloy films were prepared by the electro-deposition method with and without an external magnetic field applied parallel to the film plane during deposition. Vibrating sample magnetometry and Mössbauer spectroscopy measurements at room temperature indicate that the film deposited in external field shows an in-plane uniaxial anisotropy with an easy direction coinciding with the external field direction and a hard direction perpendicular to the field direction, whereas the film deposited without external field does not show any in-plane anisotropy. Mössbauer spectra taken in three geometric arrangements show that the magnetic moments are almost constrained in the film plane for the film deposited with applied magnetic field. Also, the magnetic moments tend to align in the direction of the applied external magnetic field during deposition, indicating that the observed anisotropy should be attributed to directional ordering of atomic pairs.

Keywords: electro-deposition, FeCo film, uniaxial anisotropy, Mössbauer spectra

PACC: 3340, 7570, 7530G

1. Introduction

Ferromagnetic films have been investigated in a variety of systems due to their wide range of applications[1−5] and fundamental theoretical interests.[6−8] Applications such as inductors, transformers and other magnetic devices require magnetic films with high cut-off frequency and high permeability.[1,3] Thin magnetic films with in-plane uniaxial anisotropy are essential to achieve better high frequency properties in realizing the target of increasing static permeability and resonance frequency simultaneously.[6] As is well known, in-plane uniaxial anisotropy can be obtained with oblique incidence of the vapor deposition beams,[9,10] and an easy direction of magnetization can also be established parallel to the magnetic field applied during the deposition[11,12] or during magnetic annealing of the specimens.[13,14] Uniaxial anisotropy can also be obtained by other means such as sputtering,[15] electro-deposition,[16−19] etc. Among these methods, electro-deposition is an important cost-effective method for preparing pure nanocrystalline metal and alloy films.

The characteristics of induced magnetic anisotropy were studied comprehensively in the 1960s (for a review, see Refs.[9], [20] and [21] and references therein). It has been suggested that the anisotropy constant, K_u, is strongly dependent upon the film composition and deposition conditions. Different mechanisms have also been proposed mainly considering the anisotropy of atomic pair ordering,[22,23] the strain anisotropy[24] resulting from the constraint of the magnetostriction strain imposed on the film by the substrate and the shape anisotropy.[11,25] Recently, Jin Han-Min *et al* reported a combination model[21] of pair–strain–shape anisotropy to reasonably explain the major characteristics of experiments in a broad range. But none of these models can explain all the experimental data and describe the origin of the anisotropy satisfactorily.

For electro-deposited films, the anisotropy derived from directional ordering of atomic pairs has been mostly considered.[16,18] However, direct experimental evidence for directional ordering of atomic pairs has not yet been reported, and prevents clear understanding of the origin of field induced uniaxial anisotropy. In the present work, using the electro-

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deposition method, we prepared FeCo alloy film with in-plane uniaxial anisotropy by applying an external magnetic field during deposition, and film deposited without an external magnetic field for comparison. The films are studied by Mössbauer spectroscopy in three geometric arrangements in order to probe the configuration of the magnetic moments in the film on a more microscopic scale.

2. Experiment

The FeCo alloy films were electro-deposited from the bath containing $FeSO_4{\cdot}7H_2O$(28 g/l), $CoSO_4 \cdot 7H_2O$ (28 g/l), H_3BO_3 (25 g/l), and L-Ascorbic acid (0.5 g/l) at a current density of 20 mA/cm^2, and the temperature of the bath was kept at room temperature. The magnetic field applied parallel to the film plane during deposition was 1200 Oe (1 Oe = 79.5775 A/m).

The morphology and composition for both films were investigated by scanning electron microscopy (HitachiS-4800, Japan) with energy dispersive x-ray spectroscopy (SEM/EDS). The magnetic properties of the films were measured by a vibrating sample magnetometer (Lake Shore 7304, USA) (VSM). ^{57}Fe conversion electron Mössbauer spectra (CEMS) of the samples were recorded by a conventional Mössbauer spectrometer (Wissel, Germany) working in constant acceleration mode at room temperature. The γ-ray source of the Mössbauer spectrometer is 25 mCi ^{57}Co in a palladium matrix. α-Fe was used to calibrate the isomer shifts and the driver velocity.

3. Results and discussion

Figure 1 shows SEM images for film A deposited without the applied external magnetic field and film B deposited with the external magnetic field. It can be seen that the films are composed of small grains, and the surface of film B is much smoother, which means that the grains are smaller and composed much more densely in film B than film A. But, no macroscopic structure such as nonspherical grains responsible for uniaxial anisotropy was observed. This is consistent with the result reported by Fujita *et al*[18] in their electro-deposited FeB films, and suggests that the anisotropy should be attributed to other origins of a more microscopic scale such as short-range atomic pair ordering.[22] Cross-sectional images obtained from SEM revealed that the thicknesses of both film A and film B are about 1 μm. EDS analysis shows that the composition is $Fe_{50}Co_{50}$ for both film A and film B, and this is a very good composition for attaining the largest uniaxial magnetic anisotropy constant K_u[16] in electro-deposited FeCo alloy films.

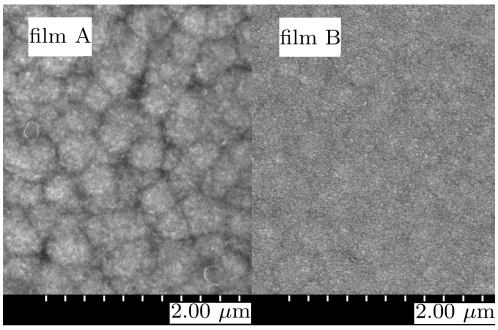

Fig.1. SEM images for film A deposited without external field and B deposited with external field.

Figure 2 shows the typical in-plane hysteresis loops of the film deposited in the absence of external magnetic field (film A) and the film deposited in the presence of external magnetic field of H_{ex} = 1200 Oe (film B). Here, "$H_{\parallel}$" and "$H_{\perp}$" denote that the measurement field is parallel and perpendicular to the direction of the applied external magnetic field during deposition respectively. It can be seen clearly from the inset figure in the right-lower corner of (film A) and (film B) that the film deposited without external field exhibits isotropy while a distinct uniaxial anisotropy is established in the film deposited with applied external field, with its easy direction being parallel to the applied field direction during deposition. The anisotropy field, H_k, obtained from the VSM result is about 105 Oe.

In order to study the anisotropy deeply on a microscopic scale, CEMS of the FeCo alloy films were recorded at room temperature in three geometric arrangements to probe the configuration of the magnetic moments in the film. The CEMS of the alloy films are shown in Fig.3. As can be seen, the spectra are broad sextets because different iron sites coexist due to random substitution of solid solution,[26,27] and therefore were fitted with the hyperfine field distribution of a slightly modified version of the Hesse and Rubartsch method using the Mosswinn programme.[28,29] Spectrum A and spectrum B were measured with the γ-ray beam perpendicular to the film plane for film A and film B respectively. Spectrum B-e60 and spectrum B-h60 were also measured for film B, but with the γ-ray

beam forming an angle of 60° with the easy direction (while perpendicular to the hard direction) and the hard direction (while perpendicular to the easy direction) respectively. The fitted parameters are given in Table 1. The hyperfine field and isomer shift observed for both film A and film B are in agreement with those reported by Sorescu and Grabias[30] and Cohen *et al.*[26] But the hyperfine field of film B is slightly smaller than that of film A; this may be caused by the differences of grain size and density of the two films.

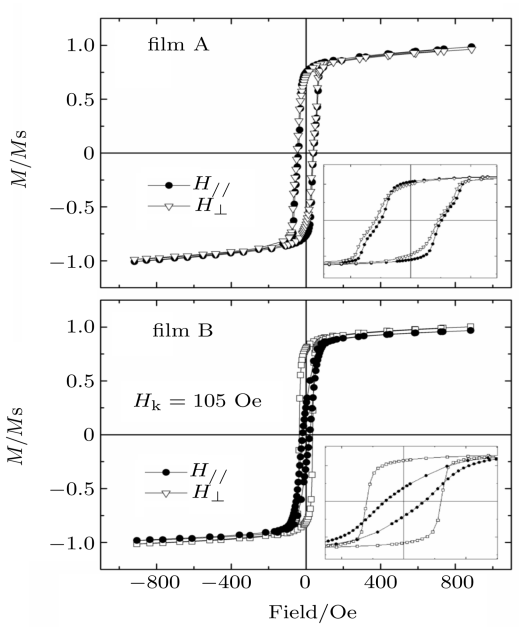

Fig.2. Typical in-plane hysteresis loops for films A and B at room temperature. "$H_\parallel$" and "$H_\perp$" denote that the measurement field is parallel and perpendicular to the direction of the applied external magnetic field during deposition, respectively.

As is well known, using the Mössbauer effect, the polar angle θ can be calculated from the relative intensities of the hyperfine splitting of the Mössbauer lines. The relation between θ and the relative intensities of ^{57}Fe lines are given by[31,32]

$$\begin{aligned} & I_{1,6} : I_{2,5} : I_{3,4} \\ &= 3(1+\cos^2\theta) : 4\sin^2\theta : (1+\cos^2\theta) \\ &= 3 : x : 1, \end{aligned} \tag{1}$$

where $(0 \le x \le 4)$ corresponds to $(0° \le \theta \le 90°)$. The polar angle θ calculated by Eq.(1) from the value x in table 1 is also listed. The polar angle θ for film A(71.38°) is much smaller than the value for film B(87.13°), which could be attributed to the rough surface of film A, indicating that the applied magnetic field tends to align the magnetic moments in the film plane. If the magnetic moments were spatially isotropic in the film plane, no differences of the polar angle should be observed between B-e60 and B-h60. However, in our experiments the θ obtained for B-e60 (65.16°) is much smaller than that for B-h60 (80.32°), suggesting that the magnetic moments of film B are anisotropic in the film plane and tend to align in the direction of the easy direction of the film. This means that the magnetic moments of film B have a directional ordering in the film plane, and the ordering direction is parallel to the external magnetic field applied during deposition. de Oliveira *et al*[27] have studied both external and internal surface sides of electro-deposited films that detached from the substrate, and no significant difference was observed. So, we may neglect the effect of substrate on the formation of the uniaxial anisotropy. Tanahashi and Maeda[16] have reported that the observed values of the uniaxial anisotropy constant K_u of their electro-deposited FeCo alloy films are considerably larger than those obtained for the vacuum evaporated FeCo alloy films, and by comparing the theoretical dependence of K_u, the average internal stress σ and $(3/2)\lambda\sigma$ on the film composition with the experimental data, they concluded that the observed anisotropy seems to depend mainly on the atomic ordering. Also, as there was no macroscopic structure such as nonspherical grains giving rise to shape anisotropy was observed from the SEM image, we suggest that the observed anisotropy in our sample should also be attributed to the directional ordering of atomic pairs,[22] a microscopic phenomenon proposed by Néel.

As shown above, our electro-deposited FeCo alloy films have a distinct in-plane uniaxial anisotropy. This is a perpetual requirement for magnetic films in high-frequency applications.[6,15] It is well known that for thin magnetic films with in-plane anisotropy, the natural resonance frequency can be calculated by the following equation[33,34]

$$f_r = \frac{\gamma}{2\pi}(4\pi M_s H_k)^{1/2}, \tag{2}$$

where γ is the gyro magnetic constant, M_s is the saturation magnetization and H_k is the in-plane anisotropy field. If we take the parameters $\gamma/2\pi$ = 2.8 GHz/kOe, $4\pi M_s = 1.74$ T and use the anisotropic field H_k = 105 Oe, we can obtain a resonance frequency of 3.8 GHz which is in good agreement with the measured data of 3.9 GHz. Detailed experimental results and a model for the complex permeability of the film will be reported soon.

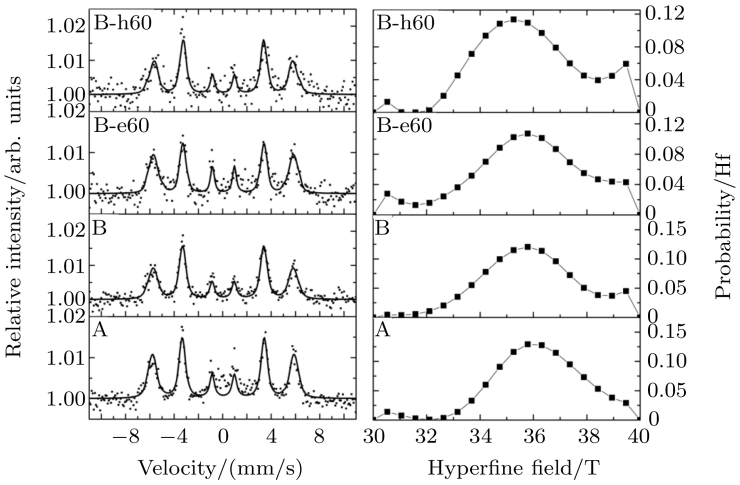

Fig.3. Mössbauer spectra taken at room temperature with the γ-rays perpendicular to the film plane for (A) and (B), and with the γ-rays forming an angle of 60 ° with the easy direction (while perpendicular to the hard direction) of film B for (B-e60) and hard direction (while perpendicular to the easy direction) of film B for (B-h60).

Table 1. Mössbauer parameters for FeCo alloy films. δ is the isomer shift relative to α-Fe, $B_{\rm hf}$ is the hyperfine field, $\Gamma_{\rm exp}$ is the line width of the spectra, x denotes the intensity ratio of $I_{2,5}/I_{3,4}$. θ is the polar angle calculated from Eq.(1).

sample	δ /(mm/s)	$B_{\rm hf}$ /T	$\Gamma_{\rm exp}$ /(mm/s)	x/arb. units	$\theta/(^\circ)$	normalized Chisquare χ
A	0.047	36.167	0.439	3.26	71.38	1.84
B	0.042	35.886	0.445	3.98	87.13	1.21
B-e60	0.061	35.700	0.329	2.80	65.16	1.14
B-h60	0.066	35.692	0.402	3.78	80.32	1.18

4. Conclusion

In summary, FeCo alloy film with a distinct in-plane uniaxial anisotropy has been prepared by the simple electro-deposition method. The film is a good candidate for high frequency applications. The uniaxial anisotropy was studied by means of a vibrating sample magnetometer and conversion electron Mössbauer spectroscopy. VSM results show that the film deposited with an magnetic field applied parallel to the film plane during deposition has an easy direction coinciding with that of the magnetic field, while the film deposited without external magnetic field does not show any easy direction in the film plane. Mössbauer spectra taken with the γ-ray beam perpendicular to the film plane show that the magnetic moments are almost constrained in the plane for the film deposited with external magnetic field. The spectra taken with the γ-ray beam forming an angle of 60 ° with the easy direction (while perpendicular to the hard direction) and hard direction (while perpendicular to the easy direction) of the film give different polar angles, indicating that the anisotropy of the film depends mainly on the directional ordering of atomic pairs.

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