



Letter

Soft magnetic and high-frequency properties of FeCoB–SiO₂ granular films deposited on flexible substratesF.F. Yang^{a,b}, S.S. Yan^{a,*}, M.X. Yu^b, S.S. Kang^a, Y.Y. Dai^a, Y.X. Chen^a, S.B. Pan^b, J.L. Zhang^b, H.L. Bai^a, D.P. Zhu^a, S.Z. Qiao^a, W.W. Pan^a, G.L. Liu^a, L.M. Mei^a^aSchool of Physics, National Key Laboratory of Crystal Materials, Shandong University, Jinan, Shandong 250100, PR China^bShandong Non-metallic Materials Institute, Jinan, Shandong 250031, PR China

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ABSTRACT

A series of FeCoB–SiO₂ granular films were deposited on Kapton flexible substrates by magnetron co-sputtering technique at room temperature. All films are amorphous with granular morphology. The coercivity in the easy axis (H_{ce}) and the resistivity (ρ) decrease with the decrease of the Ar pressure, but they decrease with the increase of the film thickness. The granular films deposited at low Ar pressure exhibit obvious in-plane uniaxial magnetic anisotropy. Excellent soft magnetic properties with the H_{ce} as low as 1.57 Oe was obtained in the 500 nm films deposited at 0.3 Pa. In addition, high ρ (~ 3.5 m Ω cm), relatively high complex permeability ($\mu' = 152.8$ at low frequency and $\mu''_{max} = 221.2$) and ferromagnetic resonance frequency ($f_r \sim 3.61$ GHz) were simultaneously obtained.

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1. Introduction

As one of the best candidates applied to high-frequency magnetic devices and electromagnetic wave absorbers, metal–insulator composite films have received considerable attention [1–8] due to high complex permeability ($\tilde{\mu} = \mu' - j\mu''$), high resistivity (ρ), high saturation magnetization (M_s) and appropriate anisotropy field (H_k). Fe-based [9], Co-based [10], and FeCo-based [11–15] granular films with good soft magnetic properties and high permeability deposited on rigid substrates such as silicon and glass have been reported. Compared with the rigid substrates, the flexible substrates have the advantages such as mechanical flexibility, enhanced durability and light weight. Therefore, the films deposited on flexible substrates have great potential for applications [16]. Some important progress of granular films deposited on flexible substrates has been made in recent years. Zuo et al. have obtained that the resistivity ρ of the Fe–Co–Si/native oxide films deposited on flexible substrates is larger than that of the films deposited on glass [16]. Xu et al. have observed ultra-wide frequency linewidth of the permeability spectra in the FeCoN films deposited on Kapton [17]. However, the flexible substrates usually deteriorate the soft magnetic and high-frequency properties. Liu et al. have reported that the CoAlO films deposited on

the flexible films have relatively higher coercivity ($H_{ce} = 20$ – 40 Oe) and lower resonance frequency (f_r) [18]. The ferromagnetic resonance character in the permeability spectra of the FeTaN films deposited on flexible substrates was deteriorated compared with the films deposited on silicon [19]. Thus, the soft magnetic and high-frequency properties of the granular films deposited on the flexible substrates should be modified exigently for application.

Compared with the magnetic alloy nitride and oxide granular films deposited on flexible substrates, FeCo alloy has large M_s and SiO₂ is one of the best insulators due to high resistivity and low cost [20]. In addition, appropriate B addition is very effective in increasing resistivity and decreasing coercivity [21], so FeCoB–SiO₂ granular films should be one of the best candidates for realizing soft magnetic and high-frequency properties.

In the present work, FeCoB–SiO₂ granular films were deposited on flexible substrates by magnetron co-sputtering technology. The soft magnetic and high-frequency properties were tuned by the Ar pressure and film thickness. Low coercivity (~ 1.57 Oe) and good high-frequency properties were obtained at low Ar pressure and appropriate film thickness.

2. Experimental details

FeCoB–SiO₂ granular films were deposited on Kapton by magnetron co-sputtering technique at room temperature. Two-inch Fe₄₀Co₄₀B₂₀ alloy disc and SiO₂ disc were used as the sputtering targets. The base pressure was better than 1×10^{-5} Pa. The SiO₂ volume fraction of the films was 20%. One group of the films were deposited at different Ar pressures (0.3, 0.5, 0.75, 1.0, and 1.5 Pa) altered by

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changing the gas flow rate, while the thickness was fixed at 250 nm. The other group of the films with different thicknesses (25, 100, 175, 250, 325, and 500 nm) were deposited by controlling the sputtering time, while the Ar pressure was maintained at 0.3 Pa. SiO₂ layer with 2 nm thickness was used as a capping layer for all films. During the deposition process, a magnetic field of 500 Oe was applied to induce an in-plane uniaxial anisotropy. The substrate rotary speed was 2 r/min.

The structures of the films were detected by X-ray diffraction (XRD) and transmission electron microscopy (TEM). The surface morphology was investigated by atomic force microscope (AFM). The magnetic hysteresis loops were measured by an alternating gradient magnetometer (AGM). The complex permeability was characterized by a vector network analyzer using the shorted microstrip transmission-line perturbation method [22]. The resistivity was measured by the standard four-point measurements. All above experiments were done at room temperature.

3. Results and discussion

Fig. 1 shows the XRD patterns of the FeCoB–SiO₂ granular films with different thicknesses. Obvious diffraction peak was not detected except the substrate signals, indicating all granular films are amorphous phase. Similarly, obvious diffraction peak was not detected for the films with Ar pressure between 0.3 Pa and 1.5 Pa (not shown here), suggesting they are also amorphous phase. Typical three-dimensional AFM surface morphology of the 250 nm FeCoB–SiO₂ granular films deposited at 0.3 Pa is shown in Fig. 2. The average roughness (R_a) is about 1.50 nm. Such relatively low R_a will benefit to obtain low coercivity [23]. The average particle size is estimated around 7.2 nm.

A typical cross section bright-field TEM image of FeCoB–SiO₂ granular films deposited at 0.3 Pa and corresponding selected area electron diffraction (SAED) are shown in Fig. 3. The granular structure with metal particles (shown in dark contrast) separated by the insulating SiO₂ matrix (shown in bright contrast) can be clearly seen. The metal particle size is not uniform and ranges from 5 nm to 15 nm, which is similar to the AFM results. The average thickness of insulating SiO₂ between metal particles is about 1 nm. The exchange coupling between the magnetic metal particles can occur at the appropriate insulating layer thickness. The continuous and broad diffraction ring of the SAED pattern in the Fig. 3 inset indicates the films are amorphous phase, which is in agreement with the result of XRD measurements.

Fig. 4 shows the magnetic hysteresis loops of the FeCoB–SiO₂ granular films deposited at different Ar pressure. The coercivities in the easy axis direction (H_{ce}) of the films deposited at 0.3 Pa, 0.75 Pa and 1.5 Pa are 3.58 Oe, 9.31 Oe and 42.93 Oe, respectively, indicating the H_{ce} increases with the increase of Ar pressure. In addition, the coercivity in the hard axis direction (H_{cd}) also increases obviously. The results show that the low H_c can be obtained at low Ar pressure. The good soft magnetic properties are due to

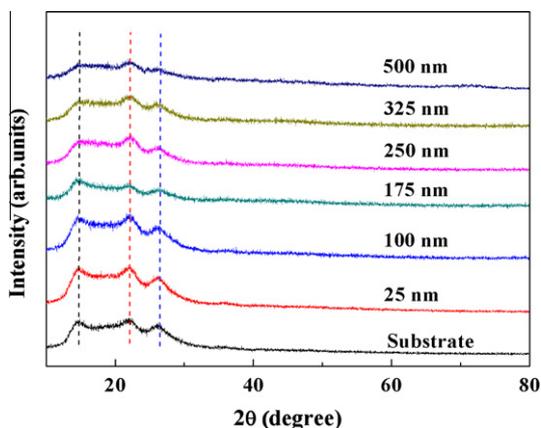


Fig. 1. XRD patterns of the FeCoB–SiO₂ granular films with different thicknesses.

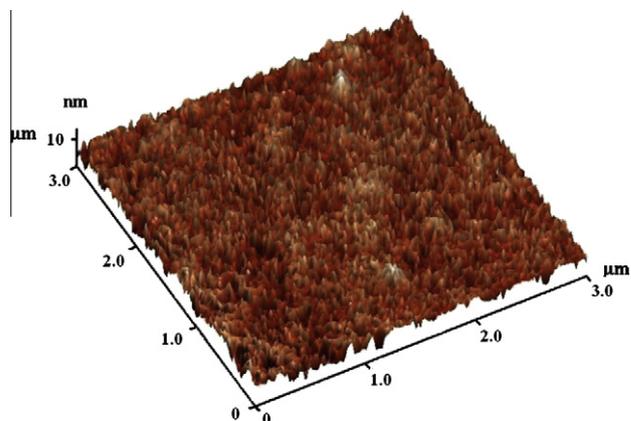


Fig. 2. Typical three-dimensional AFM surface morphology of the 250 nm FeCoB–SiO₂ granular films deposited at 0.3 Pa. (Scan area: 3 μm × 3 μm).

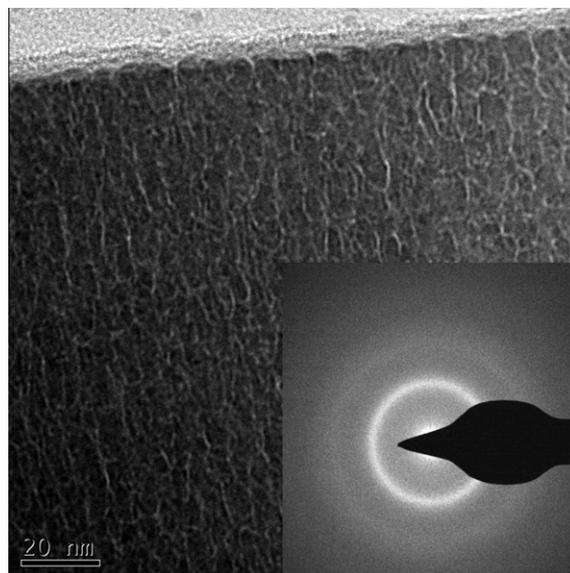


Fig. 3. A typical cross section bright-field TEM image of FeCoB–SiO₂ granular films deposited at 0.3 Pa and corresponding selected area electron diffraction (SAED).

the existence of certain compressive stress in the films deposited at low Ar pressure [24].

In order to further decrease H_c , the granular films with different thicknesses were deposited at 0.3 Pa. Fig. 5 shows the thickness dependence of magnetic hysteresis loops of the FeCoB–SiO₂ granular films. The H_{ce} decreases obviously from 40.11 Oe to 1.57 Oe with the increase of film thickness from 25 nm to 500 nm, which are shown in Fig. 4a. According to the XRD results in Fig. 1, all the films with different thicknesses are amorphous, so the influence of grain size on the H_{ce} can be ignored. The big H_{ce} of the 25 nm films is due to big stress between the films and the substrates [25]. The stress is strong enough to impede the magnetization process in the thinner films [26]. Compared with the granular films deposited on the rigid substrates [27], the change in H_{ce} of the films deposited on flexible substrates is larger with increasing thickness, indicating the larger stress exists between the films and the flexible substrates. The flexibility and relatively big surface roughness of the flexible substrates may be responsible for this behavior. It is worthy to note that the H_{ce} of the films with the thickness bigger than 250 nm is lower than 4 Oe, and the H_{ce} of the 325 nm and 500 nm films are as low as 1.59 Oe and 1.57 Oe,

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