Excellent soft magnetic properties realized in FeCoN thin films*

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(Received 14 June 2011; revised manuscript received 13 October 2011)

FeCoN soft magnetic thin films are prepared by using the reactive direct-current magnetron sputtering technique. It is found that the addition of N_2 can reduce the coercivity of the FeCoN film, and excellent soft magnetic properties can be obtained when the ratio of N_2 flow to total gas flow is 10%. The influences of texture, grain size, and stress on the magnetic properties and the high-frequency behaviors of the films are also discussed.

Keywords: nanocrystalline soft magnetic material, magnetic thin films, iron alloys

PACS: 75.30.Cr, 75.30.Gw, 75.47.Np **DOI:** 10.1088/1674-1056/21/3/037502

1. Introduction

Soft magnetic materials with high saturation magnetic flux density B_s are required in the writer head of high-density magnetic recording systems. It is also one of the key requirements in many high-frequency applications, such as the planar inductors and transformers used in integrated circuits.

It is well known that the highest stable saturation magnetization occurs in the bcc $Fe_{100-x}Co_x$ alloy with $4\pi M_s > 2.4$ T in the composition range of $30 \leq x \leq 50$.[1] However, it is difficult to achieve good soft magnetic properties in he FeCo-based alloy film, because the FeCo alloy shows a large saturation magnetostriction coefficient (around (40–65) $\times 10^{-6}$) and a relatively high anisotropy constant ($K_1 \sim 10$ kJ/m^3).[1] Many efforts have been made to obtain good soft magnetic properties in FeCo films.[2–4] For example, suitable underlayers (such as NiFe, Cu, and Co,[5–10] as Cu and Co have the lowest coercivities of 10 Oe (0.8 kA/m) and 3 Oe (0.24 kA/m) along the easy and the hard axes, respectively.) and the third element added into the FeCo alloy film[11–13] are used to improve the soft magnetic properties of the FeCo based alloy films. However, the frequency property is not acceptable when using underlayers or adding the third element (Al_2O_3) into the FeCo alloy films. The roll-off frequencies of the films by using the Co underlayer or adding Al_2O_3 are less than 1 GHz.

We prepare FeCoN films with good soft magnetic properties by using reactive sputtering in a mixed gas of Ar and N_2. The microstructures of the films and their effects on the magnetic properties are discussed in this article.

2. Experiment

The FeCoN thin films were deposited on the glass substrates by using reactive sputtering on an $Fe_{65}Co_{35}$ target in a CMS-18 magneto sputter system. The sputtering gas was a mixture of Ar and N_2, and the N_2 flow ratio $f_N = N_2/(Ar + N_2) \times 100\%$ was varied from 0% to 22.5%. The background pressure in the system was pumped down to 1.33×10^{-5} Pa by using a cyropump. During the sputtering process, the ambient gas pressure was maintained at 0.67 Pa, and a static magnetic field about 20 kA/m was applied in the direction parallel to the film surface to induce an in-plane magnetic anisotropy in the film. The film thickness was fixed at 50 nm.

The magnetic properties of the FeCoN films were measured with a vibrating sample magnetometer (VSM) at room temperature. The crystal structures of the films were determined by using an X-ray diffractometer (XRD) and a transmission electron mi-

*Project supported by the National Natural Science Foundation of China (Grant Nos. 10874060 and 60803035).
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croscope (TEM). The magnetostriction λ_s was evaluated by using a cantilever-beam magnetostriction tester, and the stress σ was calculated by using the Stoney equation. The film resistivity at room temperature was measured by using the standard four-point measurement technique, and the characteristic of the high frequency response was analyzed by using a vector network analyzer.

3. Results and discussion

The saturation magnetization $4\pi M_s$ and the coercivity H_c of the FeCoN films versus f_N are plotted in Fig. 1. As f_N increases, $4\pi M_s$ decreases monotonically, coercivities H_{ce} and H_{ch}, which are along the easy and the hard axes, decrease drastically to less than 0.4 kA/m and 0.16 kA/m, respectively, and then become stable until f_N is increased to beyond 18%. The excellent soft magnetic properties are obtained when $f_N = 10\%$, $4\pi M_s \sim 2.02$ T, $H_{ce} \sim 0.358$ kA/m, and $H_{ch} \sim 0.071$ kA/m.

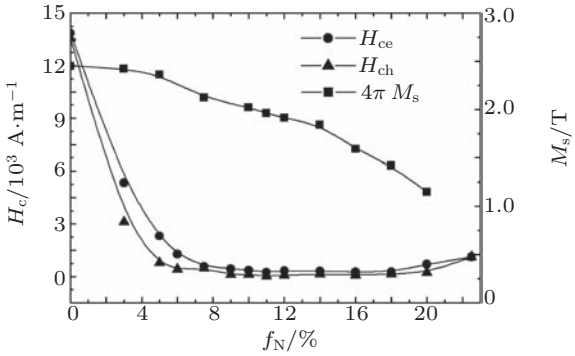


Fig. 1. Dependences of magnetic properties of FeCoN films on N_2 flow ratio.

The magnetic properties of the thin films are closely related to their microstructures. The XRD patterns of the FeCoN films with varying f_N show that as f_N increases, the (110) peak of FeCoN shifts toward the lower angle side. It is also noticed that when $f_N \geq 7\%$, the XRD diffraction peak is a mixture of the sharp (110) peak and an amorphous package, which means that the N atoms have entered into the FeCo lattices and act as the interstitial atoms, thereby expanding the crystalline lattices instrumentally and refining the grain sizes. We believe that the decrease of the saturation magnetization $4\pi M_s$ of the film can be caused by the increase of the volume fraction of the amorphous phase, since the Fe-based amorphous material has a lower saturation magnetization. This inference is also confirmed by the TEM observation (Fig. 2). The TEM (110) selected area diffraction rings of the FeCoN film are broadened obviously, indicating that there are some amorphous phases in the film. The TEM bright-field image shows that the average grain size is less than 10 nm.

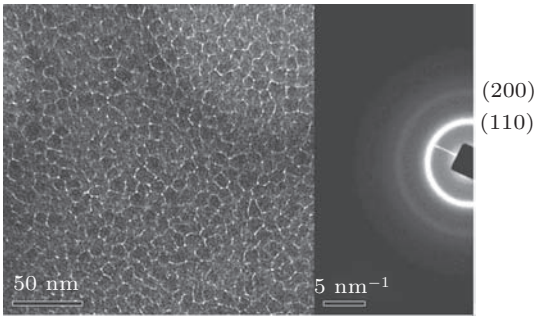


Fig. 2. TEM bright-field image and selected area diffraction pattern of FeCoN film prepared at $f_N = 10\%$.

It is well known that the basic condition for good soft magnetic properties generally requires that the magnetic anisotropy is low (a few J/m^3) or zero. The relevant anisotropy contribution mainly includes (in descending order of magnitude) the magnetocrystalline and the magnetoelastic anisotropies, the latter results from the mechanical stress and the magnetostrictive coupling.

It is demonstrated that the magnetocrystalline anisotropy constant K_1 of a material can be deduced by measuring the rotational magnetization.[14] Based on this method, the magnetocrystalline anisotropy constant K_1 of the FeCoN film prepared at $f_N = 10\%$ is about 2.8 kJ/m^3. It means that compared to the pure FeCo with a magnetic anisotropy of 10 kJ/m^3, the N element added into the FeCo alloy reduces the magnetocrystalline anisotropy.

The magnetoelastic anisotropy energy constant can be calculated by

$$K_\sigma = \frac{3}{2}\lambda_s \sigma \cos\theta. \quad (1)$$

Figure 3 shows the dependences of the stress and the magnetostriction of the FeCoN film on f_N. The magnetoelastic anisotropy energy constant K_σ of the FeCoN film varies from 10 kJ/m^3 to 80 kJ/m^3. The K_σ is estimated to be about 30 kJ/m^3. It means that there is a bigger magnetoelastic anisotropy in the film, which is one order of magnitude higher than the magnetocrystalline anisotropy constant K_1.

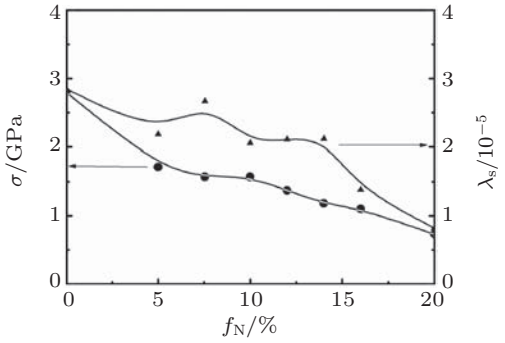


Fig. 3. Dependences of film stress σ and magnetostriction λ_s of FeCoN films on f_N.

Based on the ferromagnetism theory, the coercivity of a material, controlled by magnetocrystalline anisotropy and magnetoelastic anisotropy, may be estimated by

$$H_c = 2K_1/M_s + \rho\lambda_s\sigma/2M_s, \tag{2}$$

where ρ is the resistivity. According to the equation mentioned above, H_c of the film should be more than a few thousands A/m, and it is dominated by the magnetoelastic anisotropy, because the magnetoelastic anisotropy is higher than the magnetocrystalline anisotropy. Actually, the coercivity of the FeCoN film is much smaller than the expected value, especially for the films prepared at $f_N > 10\%$.

Because the grain size is reduced to below the critical ferromagnetic exchange length L_{ex}, according to the random anisotropy model,[15] the crystalline anisotropy might be averaged over the grains in the size of L_{ex}, and then the exchange interaction leads to a lower effective magnetocrystalline anisotropy constant. The effective magnetocrystalline anisotropy constant K_{eff} can be expressed as[16]

$$K_{eff} = \frac{K_1^4 D^6}{A^3}, \tag{3}$$

where D and A denote the grain size and the exchange stiffness constant, respectively.

From Fig. 2, it can be seen that the average grain size of the film prepared at $f_N = 10\%$ is about 10 nm, which is smaller than the critical ferromagnetic exchange length L_{ex} (for Fe based alloys, it is about 35 nm),[17] therefore, the calculated effective magnetocrystalline anisotropy constant K_{eff} is about 2.5×10^{-4} kJ/m^3, which is four orders of magnitude smaller than K_1.

As mentioned above, the magneto-elastic anisotropies dominate the magnetic anisotropy of the FeCoN film, and the film shows good soft magnetic properties. It means that the FeCoN film is insensitive to magneto-elastic anisotropy. This is a consequence of the averaging effect of the exchange interaction for nanostructural materials. The influence of the exchange coupling interaction among nanograins on the magnetoelastic anisotropy might be similar to the magnetocrystalline anisotropy, the effective magnetoelastic anisotropy is averaged over several grains, thus is considerably reduced in magnitude owing to the exchange interaction.

It is interesting that the FeCoN film shows good high frequency magnetic behaviors. We have prepared the FeCo-based alloy film by adding Al, O, and C elements.[11] Its roll-off frequency is lower than 2 GHz. However, for the FeCoN film, the roll-off frequency is a constant over 2 GHz when $f_N \leq 7.0\%$. The roll-off frequency is limited by the ferromagnetic resonance (FMR) frequency in the thin film state, which can be estimated by Kittel's equation

$$f_{FMR} = (\gamma/2\pi)\,\mu_0\sqrt{M_s H_k}, \tag{4}$$

where M_s and H_k are the saturation magnetization and the effective anisotropy field of the film, respectively.

Figure 4 shows the dependences of the roll-off frequency and the effective anisotropy field of the FeCoN films on f_N. It is shown that with f_N increasing, the $f_{roll-off}$ first keeps unchanged and then decreases monotonically. We notice that when $f_N < 5\%$, the roll-off frequency shows the highest value, which is beyond 2.8 GHz. We can seen from Fig. 1 that M_s of the FeCoN film has a small decrease first with f_N increasing, so the good high frequency response of the FeCoN films prepared at lower f_N should be mainly related to high anisotropy field H_k, because H_k of the film prepared at lower f_N is over 4 kA/m, which is twice that of the film prepared at high f_N.

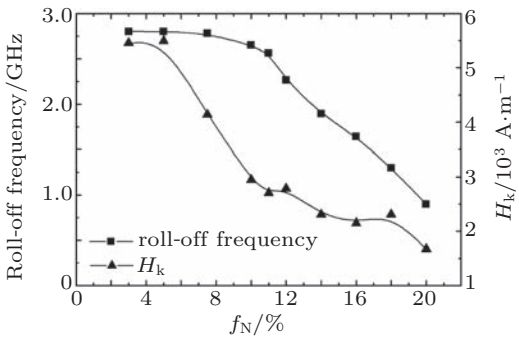


Fig. 4. Dependences of roll-off frequency and H_k of FeCoN films on f_N.

The effective anisotropy field is induced by an external field generated by a static magnet. The induced anisotropy has its original anisotropic ordering of defects (vacancies), pair ordering of atoms, and anisotropy ordering of the interstitial atoms.[18] The classic studies of the interstitial N atoms in bulk Fe indicate that some N atoms fill in the interstitial sites in the (110) plane to increase the anisotropy.[19] Therefore, it is reasonable to believe that the uniaxial anisotropy of the FeCoN film is the consequence of the anisotropic distribution of the N atoms in the α-Fe lattice and the pair ordering of the FeCo atoms. When $f_{\rm N} < 5\%$, the FeCoN film mainly is in a polycrystalline phase, the film has a high anisotropic field $H_{\rm k}$ and a high roll-off frequency. When $f_{\rm N} > 5\%$, the film is in a mixed phase consisted of nanocrystalline and amorphous phases. As $f_{\rm N}$ increases, the decrease of the volume fraction of the nanocrystalline phase leads to lower anisotropy field $H_{\rm k}$, thereby lowering the roll-off frequency.

4. Conclusion

The FeCoN films are prepared at varying N_2 flow ratio $f_{\rm N}$. The influences of $f_{\rm N}$ on the structures and the magnetic properties are investigated. As $f_{\rm N}$ increases, the film microstructures change from a polycrystalline phase to a mixed phase consisted of nanocrystalline and amorphous phases, and the volume fraction of the amorphous phase increases. The film prepared at $f_{\rm N} = 10\%$ shows excellent soft magnetic properties: $4\pi M_{\rm s} \sim 2.02$ T, easy axis coercivity $H_{\rm ce} \sim 0.358$ kA/m, hard axis coercivity $H_{\rm ch} \sim 0.071$ kA/m, and $H_{\rm k} \sim 4$ kA/m. Although the experimental measurements indicate that there is large magnetostriction and stress in the film, the film shows good soft magnetic properties. It means that due to the strong exchange coupling interaction among nanoscale grains, the magnetoelastic anisotropy can be averaged, which leads to the insensitivity of the anisotropy. The film prepared at lower $f_{\rm N}$ also shows good high-frequency response, its roll-off frequency is up to 2.8 GHz. It is the anisotropic distribution of the N atom in the α-Fe lattice that results in a larger induced uniaxial anisotropy field $H_{\rm k}$, because some N atoms fill in the interstitial sites in the (110) plane, thereby increasing the anisotropy.

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