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High-frequency magnetic characteristics of Fe-Co-based nanocrystalline alloy films

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Magnetically soft Fe-Co-based nanocrystalline alloy films were produced by two preparation methods: One using a new energetic cluster deposition technique and another using a conventional magnetron sputtering technique. Their structural, static magnetic properties and high-frequency magnetic characteristics were investigated. In the energetic cluster deposition method, by applying a high-bias voltage to a substrate, positively charged clusters in a cluster beam were accelerated electrically and deposited onto a negatively biased substrate together with neutral clusters from the same cluster source, to form a high-density Fe-Co alloy cluster-assembled film with good high-frequency magnetic characteristics. In the conventional magnetron sputtering method, only by rotating substrate holder and without applying a static inducing magnetic field on the substrates, we produced Fe-Co-based nanocrystalline alloy films with a remarkable in-plane uniaxial magnetic anisotropy and a good soft magnetic property. The obtained Fe-Co-O, Fe-Co-Ti-N, and Fe-Co-Cr-N films all revealed a high real permeability exceeding 500 at a frequency up to 1.2 GHz. This makes Fe-Co-based nanocrystalline alloy films potential candidates as soft magnetic thin film materials for the high-frequency applications.

Fe-Co-based nanocrystalline alloy films, high-frequency characteristics, energetic cluster deposition, magnetron sputtering, substrate rotation

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1 Introduction

Soft magnetic films are widely investigated [1–6] and strongly demanded for high-frequency applications, such as soft magnetic underlayers in recording media [7], high-density recording head cores [8, 9], planar inductors and film transformers in integrated circuits[1,10]. The basic demands for the operation in GHz range include high saturation magnetization (M_s), low coercivity (H_c), high permeability (μ), high electrical resistivity (ρ) and appropriate anisotropy field (H_k).

Traditional ferrite films are inapplicable in the GHz range due to the low M_s and high-temperature processing, although their ρ and μ are relatively high. Important progresses in high-frequency soft magnetic materials have also been attained in composite granular films, in which Ferich crystalline nanoparticles are embedded in a nonmagnetic insulator matrix, such as Al_2O_3 [8], ZrO_2 [11], HfO_2 [12, 13], SiO_2 [14] and Si_3N_4 [15]. However, large magnetization and high resistivity can not be simultaneously achieved because high electrical resistivity always comes out in these films at the expense of the saturation magnetization.

$Fe_{1-x}Co_x$ ($0.3 \leqslant x \leqslant 0.4$) alloy materials have the largest saturation magnetization (M_s) among ferromagnetic 3d tran-

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sition metal alloys, whereas the large magnetocrystalline anisotropy and magnetostriction limit their high-frequency applications. Therefore, to explore specific soft magnetic films with excellent high-frequency characteristics, a number of approaches on Fe-Co-based nanocrystalline alloy films and multilayer films have been developed. For example, soft magnetic Fe-Co-N, Fe-Co-Ta-N and Fe-Co-Al-N thin films [16, 17], and $Ni_{0.81}Fe_{0.19}/(Fe_{0.7}Co_{0.3})_{0.95}N_{0.05}/Ni_{0.81}Fe_{0.19}$ thin films [18] with a sandwich structure have been studied. Multilayer consisting of discontinuous metal layers with native oxide surfaces has been fabricated using Co_xFe_{100-x} alloys [19].

In this study, we pay attention to high-frequency magnetic characteristics of Fe-Co-based nanocrystalline alloy films produced by two preparation methods: One using a new energetic cluster deposition technique and another using a conventional dc magnetron sputtering method. The aim is to achieve soft magnetic films with both large magnetization and high resistivity, which are expected to have both good magnetic softness and excellent high-frequency magnetic characteristics.

2 Experimental

In the energetic cluster deposition method, Fe-Co alloy clusters were generated by the plasma-gas-condensation (PGC)-type cluster beam deposition apparatus, whose details were described elsewhere [20]. Since it was a combination of a hollow cathode discharge mode and a magnetron mode, we could obtain a high ionization efficiency of Ar and a high sputtering rate owing to the magnetic field applied perpendicular to the surfaces of Fe-Co target. Fe-Co alloy clusters thus generated were partially charged and no additional ionization process was necessary for ionizing clusters because they resided in the plasma region where the electron and ion densities were high [21, 22]. The clusters were ejected from the small nozzle by differential pumping systems and a part of the cluster beam was intercepted by a skimmer (5 mm in diameter), and then deposited onto a substrate that was fixed on a metallic substrate holder. This metallic substrate holder could be kept at a bias voltage V_a up to ±20 kV in the deposition chamber (7×10^{-3}–10×10^{-3} Pa), so that electrically charged clusters were accelerated by a large electric field and directed onto the substrate together with neutral clusters from the same cluster source, to form dense $Fe_{65}Co_{35}$ cluster-assembled films. We prepared $Fe_{65}Co_{35}$ cluster-assembled films without and with a static inducing magnetic field during deposition to obtain in-plane uniaxial magnetic anisotropy.

In the conventional dc magnetron sputtering method, $Fe_{65}Co_{35}$-based alloy films were deposited at room temperature on glass slides and single-crystal (110) Si wafers, using an $Fe_{65}Co_{35}$ alloy target with a diameter of 3 inches and a Ti (or Cr) target with a diameter of 2 inches. The chamber was evacuated to a base pressure of 6.5×10^{-4} Pa. The O_2 or N_2 concentration was adjusted by changing the O_2 (or N_2) flow ratio, R (O_2 or N_2) = [O_2 or N_2 flow rate]/ [Ar flow rate + O_2 or N_2 flow rate]. The $Fe_{65}Co_{35}$ alloy and Ti (or Cr) targets were connected to dc source and rf source and kept at 100 and 40 W, respectively. The rotation speed of substrates was kept at 30 r/min. The film thickness t was controlled by the sputtering time. After deposition, the film thicknesses were measured by a surface profiler (Dektak-III). The crystalline structures of the films were analyzed by an X-ray diffractometer (Panalytical X'pert-PRO).

For morphological observation, a transmission electron microscope (TEM) and a scanning electron microscope (SEM: LEO-1530FE) were used. In addition, the electrical resistivity at room temperature was measured using the conventional four-probe method. The magnetic properties were investigated by a superconducting quantum interference device (SQUID) and a vibrating sample magnetometer (VSM). The high-frequency characteristics at room temperature were evaluated using a high-frequency permeameter (Ryowa PMF-3000).

3 Results and discussion

3.1 High-density Fe-Co alloy cluster-assembled films

The samples were prepared using a new PGC-type cluster beam deposition apparatus with a facing-target-type dc sputtering source. We obtained $Fe_{65}Co_{35}$ alloy clusters with the mean cluster diameter from d = 7 to 12 nm with R_{Ar} increasing from 300 to 500 sccm and R_{He} decreasing from 500 to 0 sccm (Figures 1 and 2).

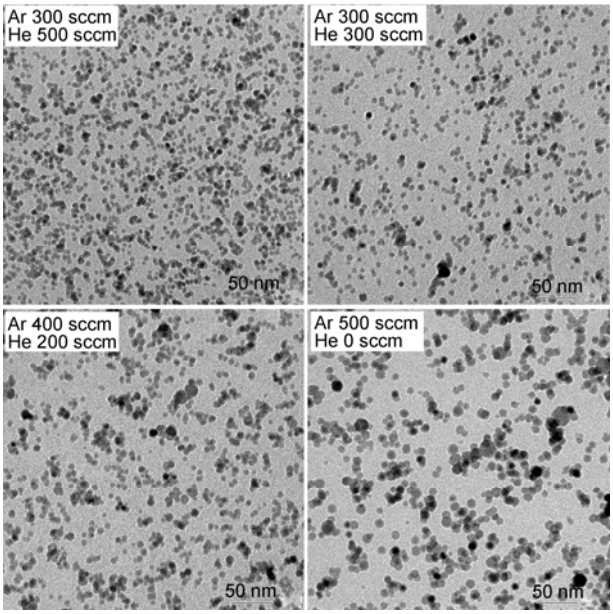

Figure 1 Bright-field TEM images of $Fe_{65}Co_{35}$ alloy clusters with effective thickness t = 1.5–2.0 nm, prepared on a carbon microgrid by varying the Ar gas flow rate R_{Ar} and the He gas flow rate R_{He} at acceleration voltage V_a = 0 kV.

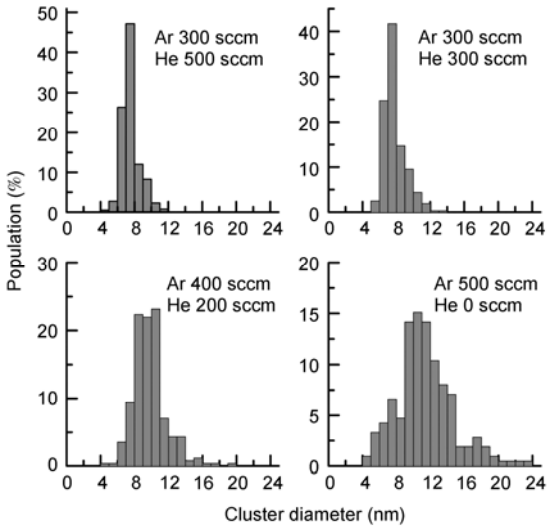

Figure 2 Size distributions of $Fe_{65}Co_{35}$ alloy clusters prepared on a carbon microgrid by varying the Ar gas flow rate R_{Ar} and the He gas flow rate R_{He} at acceleration voltage V_a = 0 kV.

In cluster-assembled films where spherical or polyhedral clusters are generated in gas phases and randomly deposited on substrates to form their porous stacks (low density, about 30%), M_s is low, and the magnetic coercivity (H_c) is also rather large (a few hundreds Oe) due to magnetic anisotropy of single domain particles and interparticle dipole interactions. However, density of the cluster-assembled films can be improved by energetic clusters deposition, where electrically charged clusters impinge on electrically biased substrates [20].

Figure 3 shows the in-plane magnetization curves at room temperature for the $Fe_{65}Co_{35}$ alloy cluster-assembled films with d = 7.8 nm and $t \approx$ 1000 nm prepared on room temperature substrates at V_a = 0 and −20 kV. The application of V_a led to a magnetically soft behavior. At V_a = 0 kV, the magnetization was saturated very slowly with a magnetic coercivity (H_c) = 110 Oe. At V_a = −20 kV, on the contrary, the magnetization was saturated very rapidly and H_c decreased to 35 Oe. Here, it is important to mention that the saturation magnetization M_s for the $Fe_{65}Co_{35}$ alloy cluster-assembled film prepared at V_a = −20 kV reached about 2.0 T (namely, about 81% of M_s (=2.45 T) of bulk bcc-$Fe_{65}Co_{35}$ alloy). That is, if we do not consider the oxidation effect of cluster-assembled film, its packing fraction is about 81%, which exceeds the fcc or hcp packing fraction (74%) in the hard ball model. In these high-density ferromagnetic cluster-assembled films, M_s is large and H_c becomes negligibly small because many ferromagnetic clusters are ferromagnetically coupled by exchange interactions. Such an exchange coupling can suppress both the magnetoelastic and crystalline anisotropy energies, and thus enhance the magnetic softness [23].

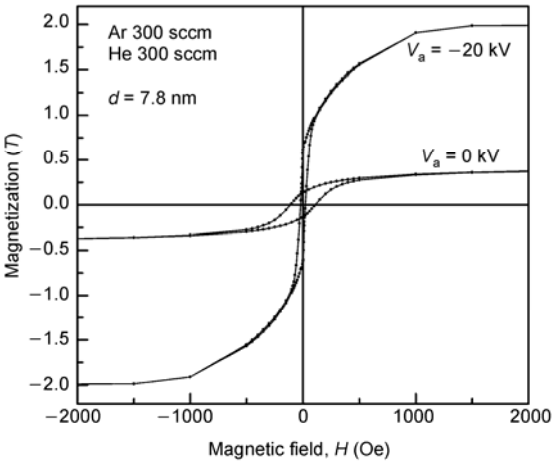

Figure 3 In-plane magnetization curves at room temperature for $Fe_{65}Co_{35}$ alloy cluster-assembled films with d = 7.8 nm and $t \approx$ 1000 nm prepared at room temperature and acceleration voltage V_a = 0 and −20 kV.

In order to examine the magnetic power loss of $Fe_{65}Co_{35}$ alloy cluster-assembled films at a high operation frequency, we measured their initial permeability. Figure 4 shows the frequency (f) dependence of permeability for $Fe_{65}Co_{35}$ alloy cluster-assembled films with d = 7.8 nm and $t \approx$ 1000 nm prepared at V_a = 0 and −20 kV. Clearly, the f dependence of permeability for low-density (packing fraction of about 30%) $Fe_{65}Co_{35}$ alloy cluster-assembled films prepared at V_a = 0 kV showed a heavy noise and was incapable of measurement. When V_a = −20 kV, high-density $Fe_{65}Co_{35}$ alloy cluster-assembled films revealed obvious f dependence of permeability. The real part (μ') of permeability had a value of 135 and was flat up to 1.5 GHz for the films. It was also notable that the permeability of the $Fe_{65}Co_{35}$ alloy cluster-assembled films prepared at V_a = −20 kV still maintained such a large value of μ' = 135 at f = 1.5 GHz, and imaginary part (μ'') of permeability had a maximum ($\mu'' \approx$

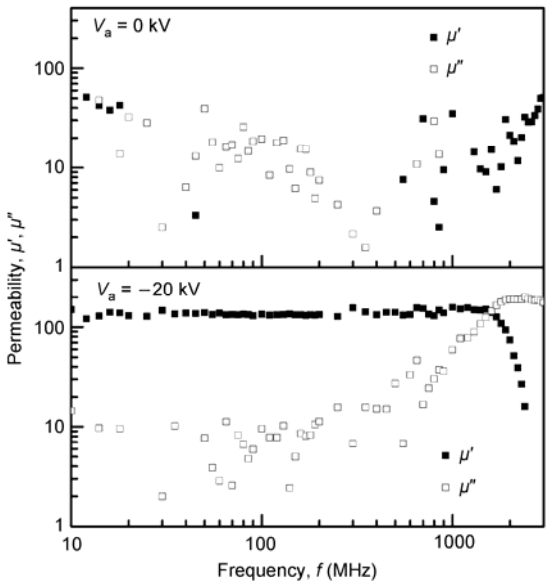

Figure 4 Frequency (f) dependence of permeability for $Fe_{65}Co_{35}$ alloy cluster-assembled films with d = 7.8 nm and $t \approx$ 1000 nm prepared at V_a = 0 and −20 kV.

190) at about 2.5 GHz, being corresponding to ferromagnetic resonance frequency (f_r). Such cluster-assembled film materials can become potential candidates for microwave absorbers.

3.2 Fe-Co-based nanocrystalline alloy films

In this study, though we prepared Fe-Co-based nanocrystalline alloy films without a static magnetic field on the substrates, we observed a remarkable in-plane uniaxial magnetic anisotropy. The easy axis, which was at an angle of 90° with respect to the hard axis, was parallel to the rotation direction of the substrate. Such an in-plane uniaxial magnetic anisotropy can mainly be attributed to the preferential orientation of crystallite growth, originated from slantwise atom incidence caused by the substrate rotation.

3.2.1 $Fe_{65}Co_{35}$-O nanocrystalline alloy films

Figure 5 shows the hysteresis loops (a) and permeability spectra (b) of the $Fe_{65}Co_{35}$-O nanocrystalline films with t = 105 nm obtained at an optimized condition of $R(O_2) = 1.0\%$. Clearly, the magnetic hysteresis loops revealed that the $Fe_{65}Co_{35}$-O thin film had evident uniaxial magnetic anisotropy at a condition without a static inducing magnetic field (Figure 5(a)). The as-deposited film has a large saturation magnetization of 2.15 T, a high resistivity of 2215 μΩ·cm and low coercivities of 2.8 Oe and 2.1 Oe in easy and hard axes, respectively. It was notable that the real permeability of the $Fe_{65}Co_{35}$-O alloy film deposited at R (O_2) = 1.0% was as high as 525 at a frequency up to 1.2 GHz and still maintained 200 at 3 GHz (Figure 5(b)). Such an excellent magnetic softness could be attributed to grain refinement caused by the addition of very low dose of oxygen, which basically did not lead to the full formation of Fe and/or Co oxide phases with low saturation magnetizations. It was worth mentioning that the film had a high resistivity of up to 2215 μΩ·cm. This is one order of magnitude larger than that of some other FeCo-based films [4, 5, 24], which may result from the slight surface oxidization of $Fe_{65}Co_{35}$ alloy crystallites. In order to verify this point, we measured the temperature dependence of resistance ρ. The film displayed a monotonic and very slow decrease in resistivity with temperature increasing, conforming to negative temperature coefficients of resistivity (TCR) below room temperature. However, we found that the TCR value was much smaller than that of semiconductors which are characterized by an exponential decrease of resistivity with temperature. Therefore, it was probable that the FeCo grain surface got slightly oxidized to form a small amount (very thin layer) of semiconductive $(Fe, Co)_3O_4$.

3.2.2 Fe-Co-Ti-N nanocrystalline alloy films

Figure 6 shows the hysteresis loops (a) and permeability spectra (b) of the Fe-Co-Ti-N nanocrystalline films obtained at an optimized condition of R (N_2) = 15% and t = 100 nm. Similar to the $Fe_{65}Co_{35}$-O nanocrystalline films, the mag-

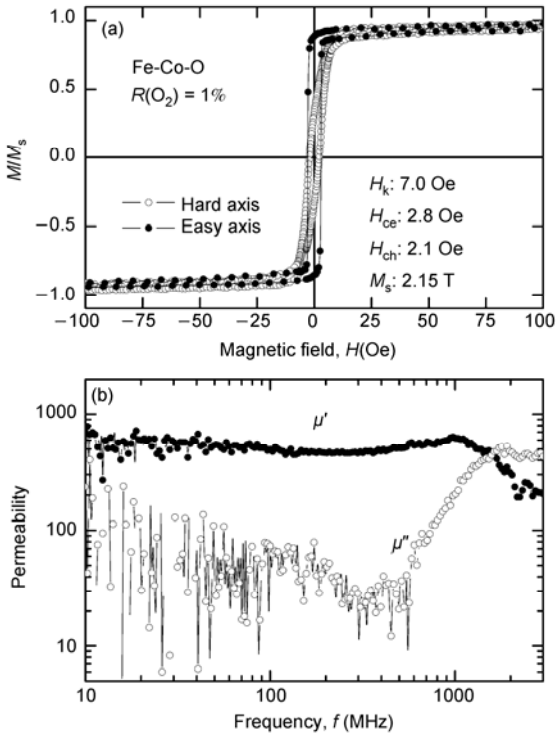

Figure 5 Hysteresis loops (a) and permeability spectra (b) of the $Fe_{65}Co_{35}$-O nanocrystalline films obtained at an optimized condition of $R(O_2)$ = 1.0% and t = 105 nm.

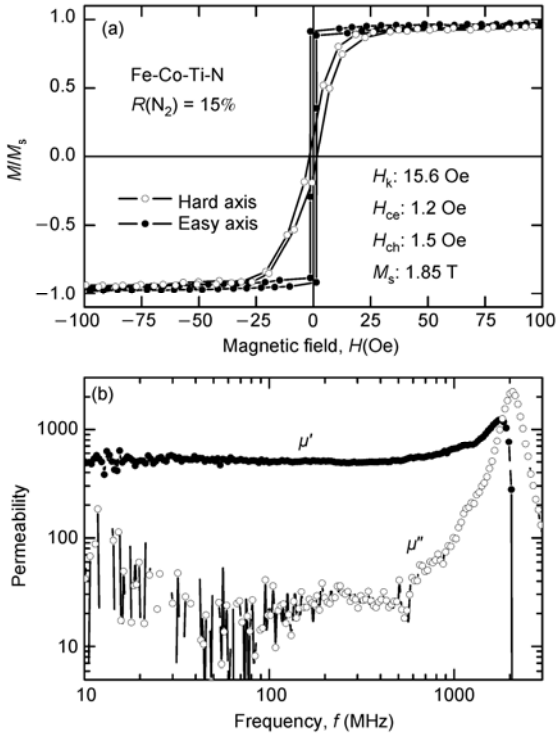

Figure 6 Hysteresis loops (a) and permeability spectra (b) of the Fe-Co-Ti-N nanocrystalline films obtained at an optimized condition of $R(N_2)$ = 15% and t = 100 nm.

netic hysteresis loops revealed that the Fe-Co-Ti-N nanocrystalline film also had evident uniaxial magnetic anisotropy at a condition without a static inducing magnetic field (Figure 6(a)). The as-deposited film had coercivity values of 1.2 Oe and 1.5 Oe along easy and hard axes, respectively, saturation magnetization of 1.85 T, anisotropy field of 15.5 Oe, and resisitivity of 240 μΩ·cm. The microwave permeability revealed a real permeability above 500 at a frequency of up to 2 GHz and a ferromagnetic resonance frequency about 2.1 GHz (Figure 6(b)).

3.2.3 Fe-Co-Cr-N nanocrystalline alloy films

In the same way, we found that at a preparation condition without a static inducing magnetic field, the as-deposited Fe-Co-Cr-N nanocrystalline films also had evident uniaxial magnetic anisotropy. In order to further increase anisotropy field and improve the high frequency characteristics of the Fe-Co-Cr-N nanocrystalline films, we investigated Fe-Co-Cr-N nanocrystalline film sample deposited in the same sputtering condition except that a static inducing magnetic field of about 200 Oe was applied to the substrate surface along the rotation direction during deposition. Figure 7 shows the hysteresis loops (a) and permeability spectra (b) of the Fe-Co-Cr-N nanocrystalline films obtained at an optimized condition of R (N_2) = 20% and t = 110 nm. The H_{ce}, H_{ch}, H_k, M_s, and ρ values of this film sample were 3.5 Oe, 3.9 Oe, 21.5 Oe, 1.73 T, and 195 μΩ·cm, respectively (Figure 7(a)). As shown in Figure 7(b), the substrate rotation and introduction of a static magnetic field during deposition led the real permeability (μ') of the film to become above 550 at a frequency of up to 1.8 GHz and a high ferromagnetic resonance frequency of 2.0 GHz.

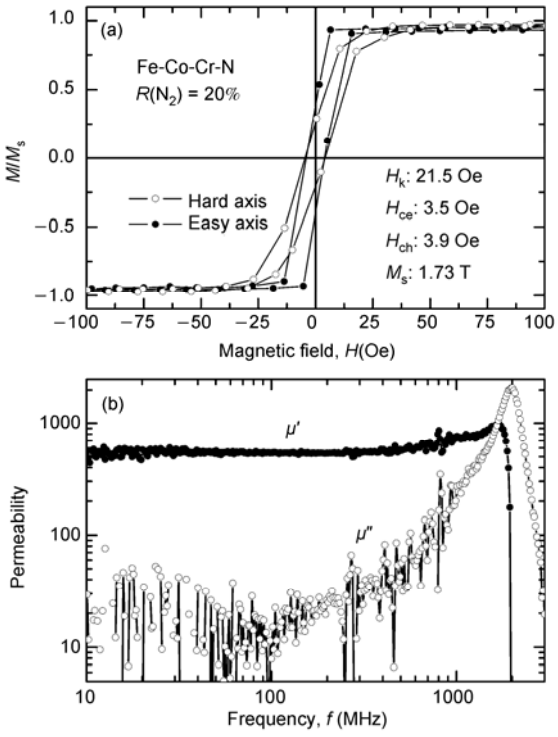

Figure 7 Hysteresis loops (a) and permeability spectra (b) of the Fe-Co-Cr-N nanocrystalline films obtained at an optimized condition of R(N_2) = 20% and t = 110 nm.

4 Conclusion

We have used two routes (a new energetic cluster deposition method and a conventional magnetron sputtering method) for preparing nanostructured Fe-Co-based soft magnetic film materials. We have obtained Fe-Co-based nanocrystalline films with good soft magnetic properties and high-frequency magnetic characteristics though bulk $Fe_{1-x}Co_x$ ($0.3 \leqslant x \leqslant 0.4$) alloy materials have the large magnetocrystalline anisotropy and magnetostriction. In the energetic cluster deposition method, by applying a high-bias voltage to a substrate, positively charged clusters in a cluster beam were accelerated electrically and deposited onto a negatively biased substrate together with neutral clusters from the same cluster source, to form a high-density Fe-Co alloy cluster-assembled film with good high- frequency magnetic characteristics. In the conventional magnetron sputtering method, the oxygen or nitrogen incorporation changed thin films from polycrystalline to nanocrystalline or amorphous-like phase, leading to an increased resistivity and enhanced magnetic softness. Only by rotating the substrate and without applying a static inducing magnetic field on the substrates, can we produce Fe-Co-based nanocrystalline alloy films with a remarkable in-plane uniaxial magnetic anisotropy and good soft magnetic properties. The obtained Fe-Co-O, Fe-Co-Ti-N, and Fe-Co-Cr-N films all have revealed a high real permeability of >500 at a frequency up to 1.2 GHz. This makes Fe-Co-based nanocrystalline alloy films potential candidates as soft magnetic thin film materials for the high-frequency applications.

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