

Article

In Situ Lorentz TEM Observation of Dynamic Domain Evolution in FeCoNi Thin Films for GHz Applications

Xiufang Zhong, Yuze Ge , Zelei Feng, Ke Chen, Guohui Jin and Lianze Ji * 

Zhejiang Key Laboratory of Energy Conversion Materials for Advanced Motor, The College of Materials and Environmental Engineering, Hangzhou Dianzi University, Hangzhou 310018, China; 13593498342@163.com (X.Z.); g1509126696@163.com (Y.G.); fzl2042559855@163.com (Z.F.); 17867963819@163.com (K.C.); 15716746025@163.com (G.J.)

* Correspondence: jlz@hdu.edu.cn

Abstract

This study explores the effects of sputtering pressure and power on FeCoNi high-entropy alloy films prepared by DC magnetron sputtering, focusing on microstructure, surface morphology, and static/high-frequency magnetic properties. In situ Lorentz TEM (LZ-TEM) was used to directly observe magnetic domain evolution. Results show that low sputtering pressure (1 mTorr) promotes strong FCC (111) crystallization, and smooth and dense surfaces. Increasing pressure leads to amorphization, higher roughness, and degraded magnetic performance. Under optimized pressure, 100 W sputtering power yields the best crystallinity, the smoothest surface, and optimal soft magnetic properties, including high remanence ratio, low coercivity, and clear ferromagnetic resonance in the 2–7.5 GHz range. The optimal parameters are confirmed as 1 mTorr and 100 W, producing uniform nanocrystalline FeCoNi films. In situ LZ-TEM reveals river-like domain walls, vortex–antivortex structures, and uniform magnetic moment precession, indicating weak domain pinning and excellent high-frequency magnetization consistency. This study provides experimental and theoretical support for the controllable fabrication of high-performance FeCoNi soft magnetic films for high-frequency devices.

Keywords: medium-entropy alloys; magnetic structure; magnetron sputtering; high-frequency property

1. Introduction

Driven by the rapid development of fifth-generation mobile communications (5G) [1,2], millimeter-wave radar [3], and high-density magnetic storage [4], modern electronic devices are rapidly evolving toward miniaturization, integration, and high-frequency operation [5]. As key functional layers in microwave passive devices, including microsensors, filters, and circulators, magnetic thin films directly determine signal transmission efficiency and energy conversion quality [6] through their high-frequency magnetodynamic characteristics. For GHz-band applications, ideal soft magnetic thin films must satisfy multiple stringent requirements: high saturation magnetization (M_s) to overcome the limitations imposed by the Snoek-type trade-off in high-frequency performance [7], high resistivity (ρ) to suppress eddy-current loss, and low coercivity (H_c) with narrow ferromagnetic resonance linewidth (ΔH) for fast magnetic response and low signal attenuation [8]. However, conventional soft magnetic materials are increasingly facing performance bottlenecks [9]. Ferrites show high resistivity but low saturation magnetization (below 0.5 T), failing to meet high magnetic



Academic Editor: Jeff Rao

Received: 26 February 2026

Revised: 17 March 2026

Accepted: 20 March 2026

Published: 25 March 2026

Copyright: © 2026 by the authors.

Licensee MDPI, Basel, Switzerland.

This article is an open access article distributed under the terms and conditions of the [Creative Commons Attribution \(CC BY\)](https://creativecommons.org/licenses/by/4.0/) license.

flux density demands. Permalloy (Ni₈₀Fe₂₀) exhibits favorable soft magnetic properties but is limited by moderate saturation magnetization (≈ 1.0 T) [10,11], restricting further device miniaturization. Fe–Co binary alloys achieve high M_s up to 2.4 T yet suffer from high coercivity and large magnetostriction [12,13]. To address these drawbacks, ternary Fe–Co–Ni alloys have attracted extensive attention [14]. As a representative medium-entropy alloy (MEA) [15,16], the FeCoNi system [17–19] integrates the high saturation magnetization of Fe–Co alloys and the low coercivity and superior corrosion resistance of Ni-based alloys via compositional tuning [20,21]. Recent studies demonstrate that this system can effectively reduce magnetic anisotropy via nanocrystalline [22] or amorphous structure design [15] while retaining excellent magnetic properties, making it a promising candidate for next-generation high-performance high-frequency soft magnetic films [23–25].

Electrodeposition is a common method for FeCoNi films due to its low cost, yet the organic additives it uses inevitably introduce non-magnetic impurities [26–28] (S, C, H, O) at grain boundaries. These impurities act as magnetic pinning centers, increasing coercivity and degrading film density. In contrast, magnetron sputtering (a physical vapor deposition, PVD, technique) enables high-purity film growth in a vacuum and is fully CMOS-compatible, making it ideal for the on-chip integration [29] of magnetic devices with semiconductor circuits. Despite existing research on sputtered FeCoNi films, most studies focus only on deposition rate or annealing effects on static magnetic properties. A systematic understanding of how key kinetic parameters—sputtering pressure and power—modulate the high-frequency magnetic response and microscopic domain evolution remains elusive [30–32]. Recently, sputtered FeCoNi and structurally related high-entropy alloy (HEA) or medium-entropy alloy (MEA) films have emerged as highly promising candidates for advanced soft magnetic applications. For instance, Kunwar et al. comprehensively investigated the structural and magnetic properties of sputtered ternary FeCoNi thin films [14]. Similarly, recent explorations into HEA/MEA magnetic films, such as the FeCoNiZr_x system by Li et al. [17] and the FeCoNiAlSi system by Tan et al. [23], have shown that severe lattice distortion and complex compositional design can yield excellent static soft magnetic properties. Furthermore, to optimize high-frequency properties, Song et al. achieved a ferromagnetic resonance frequency of approximately 4.5 GHz by constructing amorphous FeCoZr composition gradient films via Zr doping [33,34].

However, a critical comparison reveals the limitations of these state-of-the-art approaches: They often rely on intricate doping processes or high-temperature post-annealing. Moreover, they predominantly focus on static magnetic properties or lower-frequency responses, struggling to simultaneously balance ultra-low coercivity with broadband high-frequency responses (e.g., >5 GHz) in an as-deposited single-layer film. Furthermore, direct experimental evidence elucidating the high-frequency domain wall pinning mechanisms via in situ microscopy [35] is significantly lacking in the current literature. To fill this critical gap, this study fabricated pure-phase FeCoNi medium-entropy alloy (MEA) films via DC magnetron sputtering without introducing any complex doping or post-annealing, systematically investigating the effects of sputtering pressure and power on the microstructure, morphology, and high-frequency performance. By precisely optimizing these fundamental kinetic parameters (1 mTorr and 100 W), we successfully achieved a significantly broad and clear ferromagnetic resonance in the 2–7.5 GHz range with an extremely low coercivity directly in the as-deposited state. Compared to the aforementioned state-of-the-art films, our work achieves highly competitive high-frequency dynamic responses through a much simpler fabrication process. Uniquely, by employing in situ Lorentz TEM, we directly visualized the dynamic domain evolution under an external field. The observation of river-like domain walls and vortex–antivortex structures explicitly revealed the weak domain pinning effect, establishing a direct link between the dense nanocrystalline structure and the

highly coherent magnetic moment precession. This work provides a critical experimental and theoretical basis for the controllable fabrication of superior FeCoNi soft magnetic films for high-frequency devices.

2. Experimental Procedure

Sample preparation: The substrate was pretreated via reactive ion etching (RIE, Samco Inc., Kyoto, Japan, model RIE-10NR) under the following parameters: oxygen pressure of 20 Pa, power of 70 W, and etching duration of 25 s. Two types of substrates were used depending on the subsequent characterization requirements: single-crystalline Si (100) wafers with a 300 nm thermally grown SiO₂ layer (p-type, resistivity 1–10 Ω·cm, polished surface, Fangdao Semiconductor, Hangzhou, China) for general film deposition and magnetic property tests, and 100 nm thick low-stress SiN_x (Ai Experiment, Shanghai, China) membranes with a square window of 50 × 50 μm² for in situ Lorentz TEM (LZ-TEM) characterization. After pretreatment, a 30 nm thick FeCoNi film was deposited using direct current magnetron sputtering with a Fe₃₃Co₃₃Ni₃₄ (at.%) alloy target (99.99% purity, Zhongnuo Advanced Materials, Beijing, China) at room temperature without intentional heating or post-deposition annealing. To ensure the experimental reproducibility, three independent samples were prepared for each sputtering condition (each combination of sputtering pressure and power). High-frequency magnetic performance was consistently characterized for the three parallel samples, and excellent repeatability was observed with negligible deviations in key parameters. In situ Lorentz TEM (Talos F200S, Thermo Fisher Scientific, Brno, Czech Republic) characterization was only performed on the samples prepared under the optimal sputtering conditions (1 mTorr, 100 W), while the reproducibility of magnetic properties was verified for all other conditions. All parameters of the samples can be found in Table 1.

LZ-TEM characterization: To observe magnetic structure evolution, FeCoNi films were deposited onto 100 nm thick SiN_x layers using identical procedures. All samples were characterized by LZ-TEM characterization. The magnetic field was applied along the Z-axis, with the in-plane field introduced by tilting the sample 10°. The perpendicular magnetic field component exerted negligible influence on magnetic structure evolution.

FMR characterization: To obtain ferromagnetic resonance profiles, 5 × 5 mm² samples were fabricated on silicon substrates using the same deposition method. FMR spectra were obtained using the short-circuited microstrip line method. Samples were placed within a constructed cavity, with microwave signals recorded using a vector network analyzer. The in-plane magnetic field was generated by externally applied Helmholtz coils, whose magnetic field direction was parallel to the microwave propagation direction.

3. Results

3.1. Sputtering Pressure Analysis

FeCoNi films with a fixed thickness of 30 nm were deposited by precisely controlling the sputtering time. Figure 1 presents XRD patterns and atomic force microscopy (AFM) topography images of the films fabricated at sputtering pressures ranging from 1 to 15 mTorr. XRD analysis (Figure 1a) reveals a distinct (111) diffraction peak near 2θ ≈ 44° under low-pressure conditions, corresponding to the FCC structural characteristic and indicating a polycrystalline film. To further characterize the crystal structure, the crystallite size of the polycrystalline FeCoNi films under low-pressure conditions was estimated using the Scherrer equation, and the average crystallite size was determined to be 17.3 nm. This value provides a quantitative reference for understanding the grain growth behavior of the films under low sputtering pressure. As sputtering pressure increases, peak intensity progressively diminishes and peak width broadens; at high pressures, the diffraction peak

essentially vanishes. This demonstrates a significant reduction in long-range order and a gradual transition towards an amorphous state. The underlying mechanism is as follows: At high pressures, the molecular number density of the working gas increases. Sputtered particles undergo frequent collisions during transport, significantly shortening their mean free path. Consequently, the kinetic energy upon reaching the substrate is substantially reduced. Atoms struggle to diffuse and rearrange adequately on the surface, leading to random stacking and ultimately forming a structure with low crystallinity or even near-amorphous characteristics. AFM results (Figure 1b) show that the film surface is relatively flat and dense at low pressure, with uniform grain distribution. As sputtering pressure increases, surface flatness gradually deteriorates and surface roughness increases significantly. The root-mean-square (RMS) roughness increases from 3.7 nm at 1 mTorr to 11.91 nm at 15 mTorr, indicating a clear pressure-dependent evolution of surface morphology. At low pressures, sputtered atoms have a longer mean free path and higher kinetic energy for surface migration, favoring uniform nucleation and ordered grain growth. Therefore, the film exhibits a dense structure and smooth surface. Conversely, at high pressures, particle collision and scattering effects are significantly enhanced. Insufficient deposition energy and limited atomic diffusion lead to more random nucleation and growth processes, ultimately resulting in lower film density and deteriorated surface morphology.

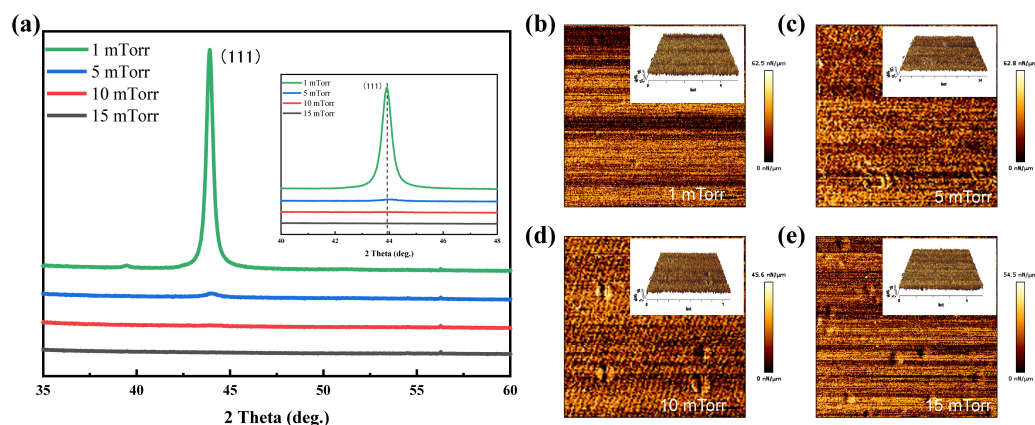


Figure 1. (a) XRD patterns of Fe-Co-Ni films grown at different sputtering pressures; (b–e) AFM images of Fe-Co-Ni films grown at different sputtering pressures (with the 3D image corresponding to the respective pressure displayed in the top right corner).

Table 1. Deposition parameters of the thin films prepared under different conditions.

Working Pressure	Sputtering Power	Deposition Time	Thickness
1 mTorr	100 W	15 min	30 nm
5 mTorr	100 W	13 min	30 nm
10 mTorr	100 W	16 min	30 nm
15 mTorr	100 W	20 min	30 nm
1 mTorr	50 W	25 min	30 nm
1 mTorr	100 W	15 min	30 nm
1 mTorr	150 W	10 min	30 nm

To investigate the effect of sputtering pressure on the static magnetic properties of thin films, room-temperature hysteresis loops were measured using SQUID. All samples exhibited similar, narrow and elongated, and approximately rectangular hysteresis loops (Figure 2a), displaying pronounced isotropic characteristics consistent with the typical behavior of soft magnetic materials. As sputtering pressure increased, the remanence of the films gradually decreased. This phenomenon is closely associated with the microstructural

and morphological evolution induced by sputtering pressure, which is well-confirmed by our XRD and AFM results for pressure-dependent samples (Figure 1a,b). Specifically, with increasing pressure, the kinetic energy of sputtered Fe/Co/Ni atoms decreases significantly, which further leads to increased structural disorder, reduced crystallinity (XRD results in Figure 1a), lower film density, and higher surface roughness (AFM results in Figure 1b). These microstructural and morphological inhomogeneities, including lattice distortions, grain boundary irregularities, and surface defects, act as enhanced pinning centers for magnetic domain walls, thereby restricting domain wall motion and hindering the coherent rotation of magnetic moments during demagnetization. To further verify this domain pinning mechanism, we performed in situ Lorentz transmission electron microscopy (LZ-TEM) characterization on the film prepared under the optimal sputtering pressure (1 mTorr), which directly revealed weak domain wall pinning and ordered magnetic moment motion in this sample. When combined with the pressure-dependent trends of increasing structural disorder, defects, and surface roughness from Figure 1, this TEM observation indirectly confirms that high-pressure films, with more microstructural and morphological inhomogeneities, exhibit stronger domain wall pinning. Consequently, magnetic moments struggle to maintain their oriented alignment after the removal of the external field, which manifests macroscopically as reduced remanence. Based on this, the high-frequency magnetic properties of films under varying pressures were characterized using a short-circuited microstrip line method coupled with a vector network analyzer. Within the 1–5 mTorr range, the film exhibited pronounced ferromagnetic resonance behavior (Figure 2b), with the resonance frequency progressively increasing from 2 GHz to 7.8 GHz as the applied magnetic field strengthened [36]. Conversely, no distinct resonance peaks were observed within the tested frequency range for samples at 10 mTorr and 15 mTorr high pressure. This behavior closely aligns with the structural evolution of the films: At elevated pressures, insufficient kinetic energy during particle deposition leads to an amorphous structure. Weakened exchange coupling reduces the effective magnetization and enhances magnetic relaxation processes, ultimately weakening the high-frequency magnetic response.

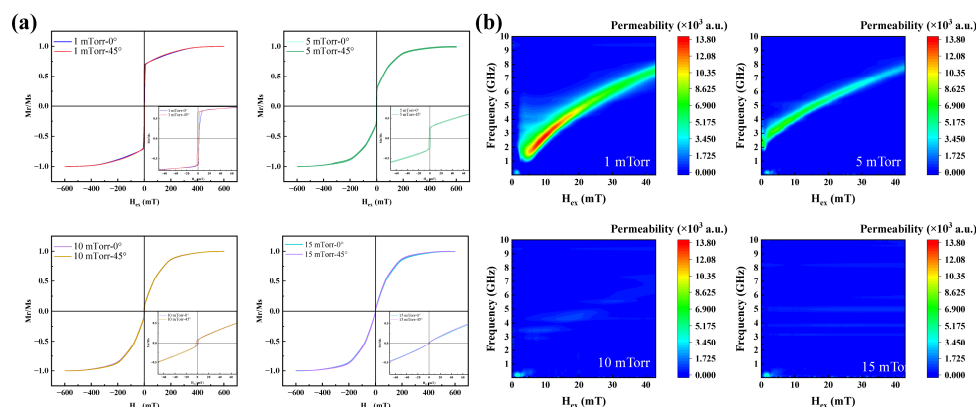


Figure 2. (a) Hysteresis loops of Fe-Co-Ni films grown under different sputtering gas pressures; (b) ferromagnetic resonance spectra of Fe-Co-Ni films grown under different sputtering gas pressures.

3.2. Sputtering Power Analysis

Building upon optimized sputtering conditions, the influence of sputtering power (50 W–150 W) on the structure, morphology, and magnetic properties of FeCoNi thin films was further investigated. Figure 3 presents the XRD patterns and AFM topography images of continuous FeCoNi films deposited at different powers levels. All samples exhibited a broadened diffraction peak near $2\theta \approx 44^\circ$ (Figure 3a), indicating a nanocrystalline structure. At a sputtering power of 100 W, the (111) diffraction peak intensity was highest, significantly exceeding that of the 50 W and 150 W samples. The diffraction peak intensity directly

reflects the crystallinity and grain-preferential grain orientation of the film, indicating that 100 W is more conducive to the crystallization and ordered grain growth of FeCoNi films. At low power (50 W), the lower energy of sputtered particles results in insufficient migration and diffusion capabilities of atoms on the substrate surface, hindering the formation of well-ordered grains and leading to lower crystallinity. Conversely, excessively high power (150 W) results in overly energetic particles, readily inducing strong bombardment and secondary sputtering effects on the substrate and film surface. This introduces additional defects and internal stresses, which in turn reduce crystallinity and weaken the diffraction peak intensity. Based on the earlier analysis of the influence of sputtering gas pressure on film surface morphology, we further examined the role of sputtering power. AFM results for films deposited at different power levels (Figure 3b) show that surface roughness decreases initially and then increases with rising power, reaching a minimum at 100 W. The corresponding RMS roughness values show a trend of initial decrease and subsequent increase, ranging from 4.2 nm to 3.7 nm and then rising to 5.72 nm. At low power (50 W), insufficient particle kinetic energy limits atomic surface diffusion, resulting in lower film density and relatively pronounced micro-undulations. When power increases to 100 W, moderate particle energy facilitates more complete atomic migration and arrangement, yielding a dense, uniform film with minimal roughness. However, when power is further increased to 150 W, excessively high particle energy intensifies bombardment effects and secondary sputtering. This disrupts surface uniformity, leading to uneven grain growth and increased roughness. Therefore, an optimal power range exists for sputtering; both excessively high and low power levels degrade the surface morphology and structural uniformity of the film.

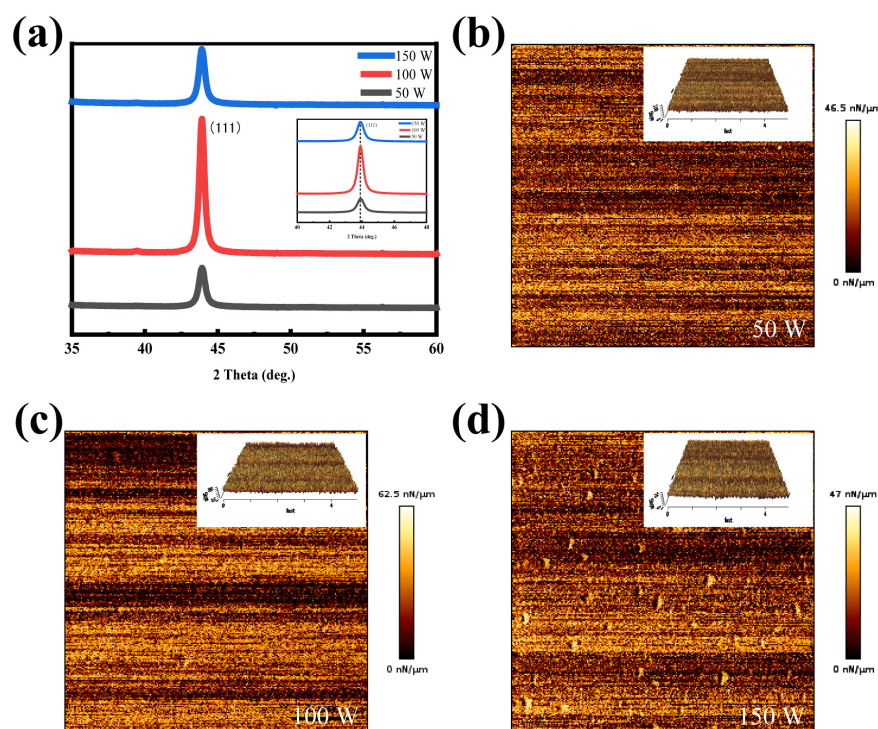


Figure 3. (a) XRD patterns of Fe-Co-Ni films grown at different sputtering powers. (b–d) AFM images of Fe-Co-Ni films grown at different sputtering powers (with corresponding 3D images at the top right corner).

Figure 4 displays the normalized in-plane hysteresis loops of FeCoNi films measured at different angles under various sputtering powers. All samples exhibit typical characteristics of soft magnetic materials, featuring narrow hysteresis loops and low coercivity.

With increasing sputtering power, the film's remanence and remanence ratio initially rise before decreasing, reaching maximum values at 100 W and significantly outperforming samples processed at 50 W and 150 W. This trend agrees well with the XRD and AFM findings: At 100 W, the films exhibit higher crystallinity and a smoother, denser surface, facilitating uniform domain alignment and efficient magnetic moment reversal, thus yielding superior soft magnetic properties. Building upon the static magnetic characterization, the high-frequency magnetic properties of samples fabricated at different power levels were evaluated under identical testing conditions. Figure 4b depicts the variation in the imaginary part of the film's magnetic permeability as a function of the applied magnetic field. The ferromagnetic resonance frequency of all samples shifted towards higher frequencies with increasing applied magnetic field, progressively rising from approximately 2 GHz to 7.5 GHz [37]. Notably, the 100 W sample exhibited the strongest permeability signal and the most pronounced resonance peak, directly attributable to its higher crystallinity, lower defect density, and superior microstructure.

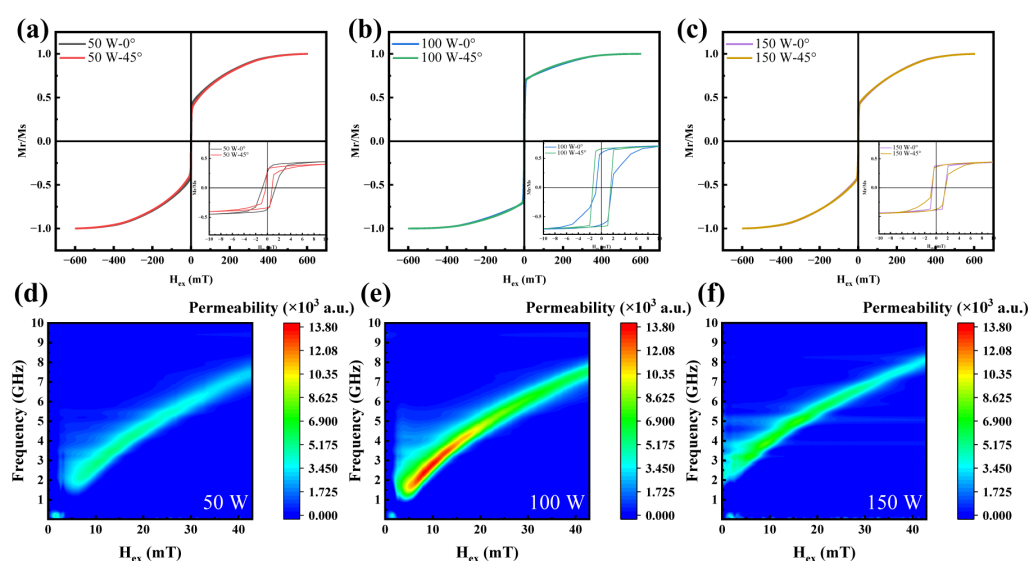


Figure 4. (a–c) Hysteresis loops of Fe-Co-Ni films grown at different sputtering powers; (d–f) ferromagnetic resonance spectra of Fe-Co-Ni films grown at different sputtering powers.

3.3. Magnetic Domain Analysis

Integrating structural, morphological and magnetic property results, this study ultimately selected 1 mTorr and 100 W as the optimal sputtering parameters for subsequent magnetic structure characterization. Under vacuum conditions, a 30 nm thick FeCoNi film was deposited onto a 100 nm SiN_x buffer layer via direct current magnetron sputtering. EDS elemental mapping (Figure 5a) and Table 2 confirm the film's composition is consistent with the FeCoNi target (Fe:Co:Ni ≈ 1:1:1) with uniform element distribution. To elucidate the intrinsic mechanism by which sputtering parameters (pressure and power) regulate the structural, morphological, and magnetic properties discussed above, this section provides a deeper fundamental understanding of co-alloying effects on the microstructural evolution of FeCoNi medium-entropy alloy (MEA) films during DC magnetron sputtering. Fe, Co, and Ni are all face-centered cubic (FCC) metals with small atomic size mismatches ($\leq 4.5\%$) and complete solid solubility. Their co-alloying forms a single-phase FCC solid solution matrix (confirmed by XRD in Figures 1a and 3a) characterized by high configurational entropy. This high-entropy stabilization effect plays a critical role in suppressing the formation of intermetallic compounds or phase segregation during the non-equilibrium sputtering deposition process, thereby ensuring the formation of a homogeneous nanocrystalline structure rather than multi-phase mixtures.

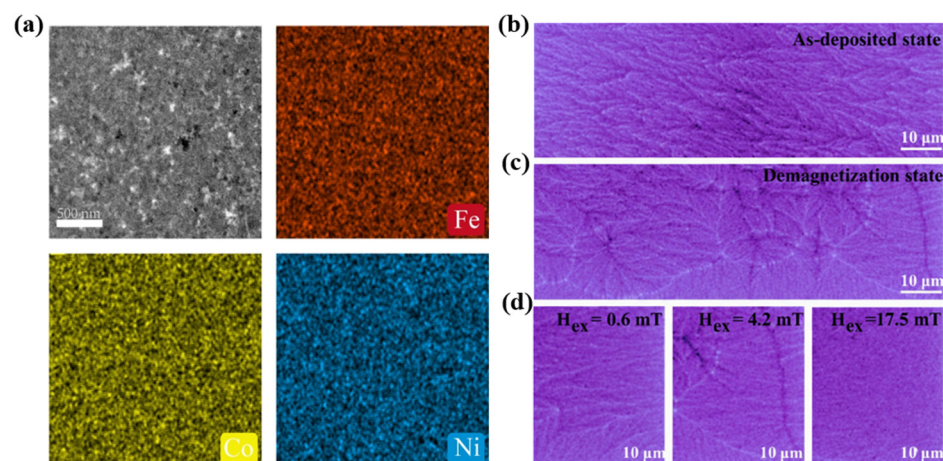


Figure 5. (a) EDS elemental mapping. (b) River-like domain walls in the as-deposited state ($H_{\text{ex}} = 0$ mT). (c) Random fine-domain state in the demagnetization state. (d) The evolution of magnetic domain structures under external in-plane magnetic fields of 0.6 mT, 4.2 mT and 17.5 mT. The scale bar is 10 μm . The defocuses are -6.8 mm for all images.

Table 2. Chemical stoichiometry of the FeCoNi ternary alloy determined by EDS analysis.

Element	Line	Mass%	Atom%
Fe	K	32.90 ± 0.14	32.90 ± 0.11
Co	K	34.25 ± 0.38	34.13 ± 0.26
Ni	K	32.85 ± 0.21	32.97 ± 0.08
Total		100.00	100.00

Notably, this work deepens the fundamental understanding of co-alloying effects by clarifying that the high-configurational-entropy FCC solid solution matrix is not merely a structural basis, but also actively mediates the microstructural evolution induced by sputtering parameters. Specifically, we reveal how co-alloying modulates the competition between crystallization and amorphization under varying pressure and power, filling the gap in understanding the intrinsic correlation between co-alloying, sputtering conditions, and microstructural formation in FeCoNi MEA films.

The microstructural evolution of the co-alloyed FeCoNi films is further regulated by sputtering pressure and power, with the co-alloying effect serving as the structural basis for such controllable evolution. When modulated by sputtering pressure, the FeCoNi system exhibits a distinct structural transition from crystalline FCC to amorphous: At low pressure (1 mTorr), the high kinetic energy of sputtered Fe/Co/Ni atoms enables sufficient surface diffusion and ordered stacking, and the FCC solid solution matrix promotes the preferential growth of the (111) orientation—the lowest-energy plane for close-packed metals. With increasing pressure, the reduced atomic kinetic energy limits diffusion, and random atomic stacking (driven by high configurational entropy) accelerates the transition to an amorphous state without phase separation. This pressure-induced structural disorder and increased defect density (including lattice distortions and grain boundary irregularities) directly act as enhanced pinning centers for magnetic domain walls, which restricts domain wall motion and hinders the coherent rotation of magnetic moments during demagnetization, thereby leading to the observed decrease in remanence with increasing pressure. For sputtering power modulation, the co-alloying effect further optimizes crystallinity: At moderate power (100 W), the balanced atomic kinetic energy facilitates the ordered arrangement of Fe/Co/Ni atoms within the FCC lattice, yielding the highest (111) peak intensity; at low (50 W) or excessively high (150 W) power, the insufficient diffusion or

bombardment-induced defects are uniformly distributed within the co-alloyed matrix (rather than localized at phase boundaries), avoiding severe crystallinity degradation or phase transformation.

The microstructural evolution governed by Fe-Co-Ni co-alloying and sputtering parameters directly determines the magnetic domain structure of the FeCoNi films, which in turn underpins their soft magnetic properties. Therefore, based on the optimal sputtering parameters (1 mTorr, 100 W) identified above, we employed in situ Lorentz transmission electron microscopy (LZ-TEM) to directly visualize the magnetic domain structure and magnetization dynamics of the continuous FeCoNi film, further clarifying the correlation between co-alloying-induced microstructural evolution and magnetic domain behavior. Results indicate that, in the as-deposited continuous film, magnetic domain walls nucleate in a river-like domain wall configuration (Figure 5b). During demagnetization, these river-like domain walls gradually disappear; as an in-plane magnetic field was progressively applied, vortex–antivortex pairs began to emerge within the system, forming meandering domain wall structures between them. Upon further strengthening of the external in-plane magnetic field to $H_{\text{ex}} \geq 17.5$ mT, the network structure formed by the meandering domain walls gradually disappeared. The magnetic moment orientation progressively aligned with the external field direction, ultimately reaching a saturated magnetization state. The evolution patterns of magnetic domains reveal that, within the FeCoNi continuous film fabricated under optimal conditions, the pinning forces acting on domain walls are relatively weak. Consequently, the magnetic moments can achieve coherent precession under the influence of an external magnetic field. It is precisely this low-pinning, high-coherence magnetic moment dynamics that enables the continuous film to maintain a distinct ferromagnetic resonance response under high-frequency conditions. Moreover, the resonance frequency exhibits a significant increase with rising external field strength, providing strong corroboration for the high-frequency magnetic property test results presented earlier.

The weak domain pinning effect, coherent and uniform magnetic moment precession, and reversible evolution of river-like domain walls and vortex–antivortex pairs observed in the optimized FeCoNi films are not only the microphysical origins of their excellent high-frequency magnetic properties but also the core structural and dynamic characteristics that determine the device-level application value of the films in high-frequency electronic systems. First, weak domain pinning eliminates the magnetic energy loss caused by domain wall pinning/depinning during high-frequency magnetization reversal, directly reducing signal attenuation and improving energy conversion efficiency—two critical performance metrics for microwave passive devices. Second, coherent magnetic moment precession ensures a rapid and consistent magnetic response to the external alternating magnetic field in the 2–7.5 GHz band, which is essential for maintaining signal stability and bandwidth consistency of high-frequency devices and avoiding signal distortion caused by disordered magnetic moment motion. Third, reversible magnetic domain evolution under low external magnetic field (vortex–antivortex nucleation at H_{ex} as low as 0.6 mT) endows the films with low-power magnetic tunability, laying a microstructural foundation for the design of miniaturized, low-power-consumption magnetic functional devices that align with the development trend of modern electronic integration and energy saving. In contrast, the amorphous FeCoNi films prepared at high sputtering pressure exhibit severe domain pinning and disordered magnetic moment motion, leading to the disappearance of FMR peaks and thus completely losing practical application value in GHz-band high-frequency devices. This contrast further confirms that the microscale magnetic domain dynamic characteristics achieved by the optimized process parameters in this study are the key premise for the FeCoNi films to possess device-level application potential.

4. Conclusions

This study employed direct current magnetron sputtering to fabricate FeCoNi medium-entropy alloy films. The effects of sputtering pressure and power on microstructure, surface morphology, and static/high-frequency magnetic properties were systematically investigated. In situ Lorentz transmission electron microscopy revealed the evolution patterns of magnetic domains. Findings indicate that low sputtering pressure (1 mTorr) promotes the formation of well-crystallized face-centered cubic (FCC) structures with dense, flat surfaces and superior soft magnetic properties. Conversely, high pressure induces amorphization, increased surface roughness, and resulted in significant degradation of magnetic performance. Under optimized pressure conditions, a sputtering power of 100 W yielded the highest crystallinity and smoothest surface, demonstrating optimal static and high-frequency soft magnetic properties: high remanence ratio, low coercivity, and distinct ferroresonance behavior across 2–7.5 GHz. FeCoNi films prepared under the optimal conditions (1 mTorr, 100 W) exhibited a nanocrystalline structure with uniform elemental distribution. In situ magnetic domain observations revealed weak domain pinning effects, enabling consistent precession of magnetic moments during magnetization—a phenomenon highly consistent with the high-frequency performance. This study establishes intrinsic correlations between process parameters, microstructure, magnetic properties, and domain dynamics, providing experimental evidence and theoretical guidance for the controlled fabrication of FeCoNi soft magnetic films and their application in high-frequency devices.

Author Contributions: X.Z. and Y.G. contributed equally to this work. Conceptualization, X.Z. and Y.G.; methodology, X.Z., Y.G. and Z.F.; writing—original draft preparation, X.Z., Y.G., Z.F. and K.C.; writing—review and editing, X.Z., Y.G., Z.F., K.C., G.J. and L.J.; visualization, X.Z. and Y.G.; project administration, L.J.; funding acquisition, L.J. All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by the National Natural Science Foundation of China (12404250) and the Natural Science Foundation of Zhejiang Province (LQN25A040008).

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: Data underlying the results presented in this paper are not publicly available at this time but may be obtained from the authors upon reasonable request.

Conflicts of Interest: The authors declare no conflicts of interest.

References

1. Silveyra, J.M.; Ferrara, E.; Huber, D.L.; Monson, T.C. Soft magnetic materials for a sustainable and electrified world. *Science* **2018**, *362*, eaao0195. [[CrossRef](#)] [[PubMed](#)]
2. Lv, H.; Yang, Z.; Liu, B.; Wu, G.; Lou, Z.; Fei, B.; Wu, R. A flexible electromagnetic wave-electricity harvester. *Nat. Commun.* **2021**, *12*, 834. [[CrossRef](#)]
3. Cai, Y.; Bai, J.; Shen, H.-L.; Huang, L.; Rao, B.; Wang, H. Development of Low-Cost Single-Chip Automotive 4D Millimeter-Wave Radar. *Sensors* **2025**, *25*, 4640. [[CrossRef](#)] [[PubMed](#)]
4. Hollenbach, M.; Kasper, C.; Erb, D.; Bischoff, L.; Hlawacek, G.; Kraus, H.; Kada, W.; Ohshima, T.; Helm, M.; Facsko, S.; et al. Ultralong-Term High-Density Data Storage with Atomic Defects in SiC. *Adv. Funct. Mater.* **2024**, *34*, 2313413. [[CrossRef](#)]
5. Ghatge, M.; Walters, G.; Nishida, T.; Tabrizian, R. An ultrathin integrated nanoelectromechanical transducer based on hafnium zirconium oxide. *Nat. Electron.* **2019**, *2*, 506–512. [[CrossRef](#)]
6. Yang, W.; Liu, J.; Yu, X.; Wang, G.; Zheng, Z.; Guo, J.; Chen, D.; Qiu, Z.; Zeng, D. The Preparation of High Saturation Magnetization and Low Coercivity Feco Soft Magnetic Thin Films via Controlling the Thickness and Deposition Temperature. *Materials* **2022**, *15*, 7191. [[CrossRef](#)]
7. Acher, O.; Dubourg, S. Generalization of Snoek's law to ferromagnetic films and composites. *Phys. Rev. B* **2008**, *77*, 104440. [[CrossRef](#)]

8. Heyderman, L.J.; Kläui, M.; Schäublin, R.; Rüdiger, U.; Vaz, C.A.F.; Bland, J.A.C. David Fabrication of magnetic ring structures for Lorentz electron microscopy. *J. Magn. Magn. Mater.* **2005**, *290–291*, 86–89. [[CrossRef](#)]
9. Herzer, G. Modern soft magnets: Amorphous and nanocrystalline materials. *Acta Mater.* **2013**, *61*, 718. [[CrossRef](#)]
10. Shukla, V. Review of electromagnetic interference shielding materials fabricated by iron ingredients. *Nanoscale Adv.* **2019**, *1*, 1640. [[CrossRef](#)] [[PubMed](#)]
11. Kockar, H.; Demirbas, O.; Kuru, H.; Alper, M. Differences observed in properties of ternary NiCoFe films electrodeposited at low and high pH. *J. Mater. Sci. Mater. Electron.* **2012**, *24*, 1961. [[CrossRef](#)]
12. Scheunert, G.; Heinonen, O.; Hardeman, R.; Lapicki, A.; Gubbins, M.; Bowman, R.M. A review of high magnetic moment thin films for microscale and nanotechnology applications. *Appl. Phys. Rev.* **2016**, *3*, 011301. [[CrossRef](#)]
13. Tabakovic, I.; Venkatasamy, V. Preparation of metastable CoFeNi alloys with ultra-high magnetic saturation ($B_s = 2.4\text{--}2.59\text{ T}$) by reverse pulse electrodeposition. *J. Magn. Magn. Mater.* **2018**, *452*, 306–314. [[CrossRef](#)]
14. Kunwar, S.; Chelvane, J.A.; Raja, M.M. Structural, magnetic and Magnetic-Microstructural properties of sputtered FeCoNi thin films. *J. Magn. Magn. Mater.* **2023**, *572*, 170597.
15. Jiao, M.; Lei, Z.; Wu, Y.; Du, J.; Zhou, X.-Y.; Li, W.; Yuan, X.; Liu, X.; Zhu, X.; Wang, S.; et al. Manipulating the ordered oxygen complexes to achieve high strength and ductility in medium-entropy alloys. *Nat. Commun.* **2023**, *14*, 806. [[CrossRef](#)]
16. Zhang, Y.; Zuo, T.T.; Tang, Z.; Gao, M.C.; Dahmen, K.A.; Liaw, P.K.; Lu, Z.P. Microstructures and properties of high-entropy alloys. *Prog. Mater. Sci.* **2014**, *61*, 1–93. [[CrossRef](#)]
17. Li, Z.; Wang, F.; Zhao, C.; Liao, Y.; Gao, M.; Zhang, H. Microstructural evolution and enhanced magnetic properties of FeCoNiZrx medium entropy alloy films. *J. Alloys Compd.* **2024**, *971*, 172649.
18. Cheng, Z.; Zhao, Q.; Tao, M.; Du, J.; Huang, X.; Liu, C. Preparation of FeCoNi medium entropy alloy from $\text{Fe}^{3+}\text{--Co}^{2+}\text{--Ni}^{2+}$ solution system. *Int. J. Miner. Metall. Mater.* **2024**, *32*, 92.
19. Peng, W.; Xia, Y.; Xu, H.; Tan, X. Tuning Corrosion Resistance and AC Soft Magnetic Properties of Fe-Co-Ni-Al Medium-Entropy Alloy via Ni Content. *Entropy* **2024**, *26*, 1038.
20. Arab, A.; Mardaneh, M.R.; Yousefi, M.H. Investigation of magnetic properties of MnZn-substituted strontium ferrite nanopowders prepared via conventional ceramic technique followed by a high energy ball milling. *J. Magn. Magn. Mater.* **2015**, *374*, 80.
21. Zhang, Y.; Zhang, M.; Li, D.; Zuo, T.; Zhou, K.; Gao, M.C.; Sun, B.; Shen, T. Compositional Design of Soft Magnetic High Entropy Alloys by Minimizing Magnetostriction Coefficient in $(\text{Fe}_{0.3}\text{Co}_{0.5}\text{Ni}_{0.2})_{100-x}(\text{Al}_1/3\text{Si}_2/3)_x$ System. *Metals* **2019**, *9*, 382. [[CrossRef](#)]
22. Toparli, C.; Ebin, B.; Gürmen, S. Synthesis, structural and magnetic characterization of soft magnetic nanocrystalline ternary FeNiCo particles. *J. Magn. Magn. Mater.* **2017**, *423*, 133. [[CrossRef](#)]
23. Tan, X.; Li, J.; Zhang, S.; Xu, H. Enhanced DC and AC Soft Magnetic Properties of Fe-Co-Ni-Al-Si High-Entropy Alloys via Texture and Iron Segregation. *Metals* **2024**, *14*, 1113. [[CrossRef](#)]
24. Jiang, D.; Yuan, Z.; Zhu, Z.; Yao, M. NiCoCrFeY High Entropy Alloy Nanopowders and Their Soft Magnetic Properties. *Materials* **2024**, *17*, 534. [[CrossRef](#)]
25. Kitagawa, J.; Shintaku, D. Magnetic Properties of High-Entropy Alloy FeCoNiTi. *ACS Omega* **2024**, *9*, 37197. [[CrossRef](#)]
26. Nadzri, N.I.M.; Halin, D.S.C.; Al Bakri Abdullah, M.M.; Joseph, S.; Salleh, M.A.A.M.; Vizureanu, P.; Burduhos-Nergis, D.-P.; Sandu, A.V. High-Entropy Alloy for Thin Film Application: A Review. *Coatings* **2022**, *12*, 1842. [[CrossRef](#)]
27. Rodwihok, C.; Choopun, S.; Ruankham, P.; Gardchareon, A.; Phadungdhithidhada, S.; Wongratanaphisan, D. UV sensing properties of ZnO nanowires/nanorods. *Appl. Surf. Sci.* **2019**, *477*, 159. [[CrossRef](#)]
28. Wickramaarachchi, K.; Minakshi, M. Status on electrodeposited manganese dioxide and biowaste carbon for hybrid capacitors: The case of high-quality oxide composites, mechanisms, and prospects. *J. Energy Storage* **2022**, *56*, 106099. [[CrossRef](#)]
29. Mehr, A.K.; Mehr, A.K. Magnetron sputtering issues concerning growth of magnetic films: A technical approach to background, solutions, and outlook. *Appl. Phys. A* **2023**, *129*, 662. [[CrossRef](#)]
30. Ma, Z.; Wu, T.; Li, Q.; Sun, K.; Wu, C.; Jiang, X.; Yu, Z.; Lan, Z. Dynamic magnetization resonance enhancement of Co-based soft magnetic thin films induced by two-step oblique angle deposition. *J. Alloys Compd.* **2025**, *1043*, 184227. [[CrossRef](#)]
31. Khaydukov, Y.; McCafferty, G.; Dobrynin, A.; Devishvili, A.; Vorobiev, A.; Bencok, P.; Fan, R.; Steadman, P.; McNeill, K.; Ormston, M. Improved performance of polycrystalline antiferromagnet/ferromagnet stack by nitrogen-assisted deposition. *Appl. Phys. Lett.* **2025**, *126*, 222408.
32. Tkadletz, M.; Hofer, C.; Wüstefeld, C.; Schalk, N.; Motylenko, M.; Rafaja, D.; Holzschuh, H.; Bürgin, W.; Sartory, B.; Mitterer, C.; et al. Thermal stability of nanolamellar fcc-Ti1-xAlxN grown by chemical vapor deposition. *Acta Mater.* **2019**, *174*, 195.
33. Song, C.; Han, Z.; Zhou, J.; Wang, X.; Zhang, L.; Ma, Z.; Ma, L.; Zheng, F. Regulation of static and dynamic magnetic properties of amorphous FeCoZr composition gradient films by Zr doping. *AIP Adv.* **2023**, *13*, 125109.
34. He, Z.; Ma, Z.; Li, Z.; Du, Y.; Yang, J.; Wu, C.; Li, Q.; Jiang, X.; Wang, C.; Yu, Z.; et al. Strain Modulation of Microstructure, Magnetic Domains, and Magnetic Properties of Ti/Fe/Ni₈₁Fe₁₉/Fe/Ti Multilayer Thin Films. *Coatings* **2023**, *13*, 363.

35. Vashisht, G.; Shashank, U.; Gupta, S.; Medwal, R.; Dong, C.L.; Chen, C.L.; Asokan, K.; Fukuma, Y.; Annapoorni, S. Pinning-assisted out-of-plane anisotropy in reverse stack FeCo/FePt intermetallic bilayers for controlled switching in spintronics. *J. Alloys Compd.* **2021**, *877*, 160249.
36. Ji, L.; Zhao, R.; Hu, X.; Hu, C.; Shen, X.; Liu, X.; Zhao, X.; Zhang, J.; Chen, W.; Zhang, X. Reconfigurable Ferromagnetic Resonances by Engineering Inhomogeneous Magnetic Textures in Artificial Magnonic Crystals. *Adv. Funct. Mater.* **2022**, *32*, 2112956. [[CrossRef](#)]
37. Shen, X.; Bo, L.; Zhao, R.; Hu, C.; Ji, L.; Zhang, J.; Zhang, X.; Dong, X. Spontaneous nucleation of vortex–antivortex pairs in confined magnetic microstructures. *J. Phys. D Appl. Phys.* **2023**, *57*, 105003. [[CrossRef](#)]

Disclaimer/Publisher’s Note: The statements, opinions and data contained in all publications are solely those of the individual author(s) and contributor(s) and not of MDPI and/or the editor(s). MDPI and/or the editor(s) disclaim responsibility for any injury to people or property resulting from any ideas, methods, instructions or products referred to in the content.