

Article

Liquid-Film Temperature Regulates (222) Texture and Permeability–Frequency Response in Spin-Sprayed NiZn Ferrite Thin Films

Hai Liu ^{1,2,3,*}, Jinhua Zhu ¹, Xinglian Song ³, Wenju Liao ³, Yu Liu ⁴ and Ke Sun ² 

¹ Hubei Key Laboratory of Low Dimensional Optoelectronic Materials and Devices, Hubei University of Arts and Science, Xiangyang 441053, China

² School of Materials and Energy, University of Electronic Science and Technology of China, Chengdu 610054, China; ksun@uestc.edu.cn

³ Shandong Chunguang Magnetolectric Technology Co., Ltd., Linyi 276017, China; songxinglian@ktong.com (X.S.); liaowenju@ktong.com (W.L.)

⁴ School of Electronic Information and Artificial Intelligence, West Anhui University, Lu'an 237012, China

* Correspondence: xiaomingever@gmail.com

Abstract

Spin-spray deposition is a low-temperature route for preparing crystalline ferrite films, but the actual liquid-film temperature has not been isolated from the nominal heater temperature. NiZn ferrite films were deposited at liquid-film temperatures of 87.6, 90.1, 92.7, and 95.0 °C. As the temperature increased, the (222) Lotgering factor f_L decreased from 0.32 to 0.01, the triangular morphology weakened, and the growth rate declined. M_s remained nearly constant at 429–442 kA m^{−1}, whereas μ'_{max} increased from 44 to 83 and f_r decreased from 465 to 260 MHz. The structural and magnetic trends are consistent with a shift from surface-confined (222)-oriented growth toward less-oriented growth and a corresponding permeability–frequency trade-off. Unlike our previous studies of substrate and oxidant effects, this work isolates the measured liquid-film temperature and establishes its quantitative relationship with texture and dynamic magnetic response. This parameter provides a practical means of selecting the operating window of spin-sprayed NiZn ferrite cores for integrated high-frequency inductors.

Keywords: NiZn ferrite thin film; spin-spray deposition; liquid-film temperature; Lotgering factor; (222) texture; magnetic permeability; cutoff frequency



Academic Editor: Carlos J. Gómez García

Received: 30 June 2026

Revised: 26 July 2026

Accepted: 29 July 2026

Published: 3 August 2026

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1. Introduction

The scaling of radio frequency and power management circuits continues to put pressure on integrated passive components, especially on-chip inductors. In spiral and solenoid inductors, inductance is tied to magnetic-flux storage, so it cannot be reduced indefinitely by lithographic scaling alone. Air-core inductors are process-compatible but have limited inductance density. Metallic magnetic cores can increase inductance, but their low resistivity often leads to eddy current loss and skin-effect-related degradation at high frequency. Ferrite thin films provide a more balanced option because they combine magnetic permeability with high electrical resistivity, allowing a magnetic core to increase inductance density while keeping high-frequency loss relatively low [1–3]. Ferrite films are therefore attractive for miniaturized, high-frequency devices.

Several device-level studies have made this motivation clear. Sai et al. deposited CMOS-compatible nanocrystalline zinc ferrite films to improve the inductance density

of integrated RF inductors [4]. Wang et al. used epitaxial yttrium iron garnet films for high-frequency on-chip inductors, showing the value of low-loss ferrite films in GHz passive components [5]. More directly related to the present work, Wang et al. integrated spin-sprayed NiZn ferrite films into magnetic solenoid inductors and obtained improved inductance and quality factor over the 0.7–6 GHz range compared with air-core structures [6]. These reports also indicate that a high permeability value alone is not sufficient for an inductor core. The magnetic spectrum, including the permeability, magnetic loss and cutoff or resonance frequency, must match the intended operating band. Therefore, a method that can tune the frequency response of ferrite films is useful for device design.

Ferrite thin films have been prepared by RF sputtering, oblique sputtering, RF magnetron sputtering, pulsed laser deposition, chemical vapor deposition, chemical synthesis, and spray-based routes. These studies show that thickness, substrate temperature, strain, composition, cation substitution, and texture can alter structural and electromagnetic properties [7–19]. Recent studies have examined lithium ferrite, cobalt ferrite, NiFe_2O_4 , and Mg-doped nickel ferrite films, with their properties tuned through deposition method, substrate temperature, or cation substitution [15,17,20–22]. However, many vacuum-based routes require high processing temperatures or post-deposition annealing to crystallize the ferrite phase. Such thermal treatment may cause dopant redistribution, interfacial reactions, or degradation of interconnect and dielectric layers, and it limits compatibility with polymer substrates and low-cost, large-area processing.

Vacuum techniques such as sputtering and pulsed-laser deposition can produce dense films with accurate thickness control and, under suitable conditions, epitaxial or highly oriented microstructures. Spin-spray deposition is not claimed to be universally superior in crystallinity or magnetic performance. Its principal advantage is that crystalline ferrite can be formed from aqueous precursors near 100 °C without vacuum equipment or a separate high-temperature crystallization treatment [7–23]. This low thermal budget broadens compatibility with completed interconnects, dielectric layers, glass, silicon-based substrates, polymers, and functional oxides [23–25].

Spin-spray deposition evolved from aqueous ferrite plating, first demonstrated by Abe and Tamaura [23]. Its permeability was later described phenomenologically by Abe et al. [24], and its low-temperature process compatibility has been demonstrated in nickel manganite thermistor films and ferrite/piezoelectric magnetoelectric heterostructures [25,26].

For spin-sprayed NiZn ferrite films, links between liquid-phase processing and high-frequency magnetic response have been reported. Obi et al. prepared spin-sprayed $\text{Ni}_{0.27}\text{Zn}_x\text{Fe}_{2.73-x}\text{O}_4$ films under a 360 Oe magnetic field and obtained a real permeability above 50 in the GHz range, with a ferromagnetic-resonance frequency of approximately 2.7 GHz [27]. Wang et al. showed that precursor and oxidizer pH strongly affect growth and RF/microwave properties; under optimized conditions, the real permeability exceeded 200 at 0.5 GHz with low loss [28]. Ray proposed a μ -droplet regime with a higher Snoek product than continuous liquid-layer deposition. In related work, Lorentz TEM revealed multi-grain domains in spin-sprayed NiZnCo ferrite films [29]. Guzman et al. subsequently examined the growth, magnetic, and electronic properties of NiZn ferrite films [30].

The structural origin of the unusual morphology in spin-sprayed NiZn ferrite films has also been investigated. Liu et al. reported triangular grains in spin-sprayed NiZn ferrite films and related them to a preferred (222) orientation [31]. They proposed that the orientation originates from anisotropic growth along the close-packed $\{hhh\}$ plane composed of oxygen anions and B-site cations in the spinel lattice; in cubic indexing, this plane corresponds to the (222) plane. The strength of this orientation is evaluated by the Lotgering factor f_L , calculated from XRD peak intensities. The triangular morphology is

therefore a surface manifestation of the same anisotropic growth process, not the source of f_L itself.

Subsequent work showed that this (222)-oriented growth is sensitive to the reaction environment. Ran et al. found that aqueous ethanol solutions changed the structure and magnetic properties of spin-sprayed NiZn ferrite films, suggesting that the liquid medium itself affects film formation [32]. In the substrate study, Liu et al. showed that substrate thermal conductivity, rather than wettability alone, strongly affects film growth; excessive heat conduction can shift part of the reaction from the substrate surface into droplets, giving larger grains, higher apparent growth rate and degraded magnetic properties [33]. In a later oxidant-concentration study, excessive NaNO_2 disrupted the (222) preferred orientation, changed the morphology from triangular plate-like grains to agglomerated or spherical grains and shifted the magnetization mechanism toward a stronger domain wall contribution [34].

These studies suggest that the (222) texture is not a secondary structural detail. It is linked to how the ferrite phase forms during spin-spray deposition and may also affect the dynamic magnetic response. In epitaxial ferrite films, magnetic anisotropy can be tuned through strain, texture and atomic replacement [10,35]. In spin-sprayed polycrystalline NiZn ferrite films, however, the effect of orientation-induced anisotropy on the magnetic spectrum has not been systematically examined. In particular, it is still unclear how a controlled decrease in the XRD-derived Lotgering factor f_L affects the apparent anisotropy field, the balance between magnetization rotation and domain-wall displacement, and finally the permeability spectrum.

In the present work, the liquid-film temperature is used as the regulating parameter. The liquid-film is a transient reaction layer generated by the coalescence of sprayed droplets on the heated substrate. In an appropriate temperature window, ferrite crystallization is confined near the liquid-film/growing-film interface, producing compact layer-by-layer deposition. At insufficient temperature, the interfacial oxidation–dehydration reaction is too slow to sustain film growth, whereas excessive liquid-film temperature may promote nucleation away from the interface or even inside the liquid film, which would disturb compact surface-confined growth and favor particle attachment or less-oriented grain growth. This parameter is different from the nominal heating-plate temperature and is closer to the actual reaction environment where Fe^{2+} adsorption, oxidation by NO_2^- , dehydration, evaporation, nucleation and surface incorporation occur. NiZn ferrite thin films were prepared by spin-spray deposition at liquid-film temperatures of 87.6, 90.1, 92.7 and 95.0 °C. The evolution of the (222) Lotgering factor, XRD peak intensity, triangular grain morphology, growth rate, static magnetic properties and magnetic spectrum parameters was examined. The purpose is to clarify whether weakening the (222) preferred orientation can tune the anisotropy-related dynamic magnetic response of spin-sprayed NiZn ferrite films.

Spin-spray deposition is an established synthesis route [23,24]. Here, the measured liquid-film temperature—not the heater set point, substrate, solution composition, or oxidant concentration—is treated as the independent variable. Earlier work established the triangular/(222) morphology [31], the effect of substrate thermal conductivity [33], and the influence of oxidant concentration [34], but did not isolate the temperature of the transient liquid layer or relate a controlled change in f_L to the simultaneous evolution of μ'_{max} and f_r . The present ranges of 87.6–95.0 °C, $f_L = 0.32$ –0.01, $\mu'_{\text{max}} = 44$ –83, and $f_r = 465$ –260 MHz establish this process–texture–dynamic-property relationship.

Across this 7.4 °C interval, all films remain crystalline and soft ferrimagnetic, with $M_s = 429$ –442 kA m^{−1} and $H_c = 1.59$ –1.83 kA m^{−1} (20–23 Oe). Thus, liquid-film temperature provides a low-thermal-budget means of tuning the permeability–frequency window without changing the nominal composition or adding a post-deposition annealing step.

2. Materials and Methods

2.1. Film Preparation

NiZn ferrite thin films were prepared by spin-spray deposition using a Sono-Tek ultrasonic atomizing system. An oxidant solution containing 5 mmol L^{−1} NaNO₂ and 20 mmol L^{−1} CH₃COONa was prepared using deionized water, and the metal-ion precursor solution was prepared by dissolving NiCl₂, ZnCl₂ and FeCl₂ separately in deionized water. The oxidant and precursor solutions were simultaneously sprayed onto 0.2 mm glass substrates fixed on a rotating heating plate by a vacuum pneumatic sucker. The nominal composition of the NiZn ferrite was Ni_{0.2}Zn_{0.1}Fe_{2.70}O₄.

In this work, the heating condition was adjusted to obtain different liquid-film temperatures during deposition. The actual liquid-film temperatures, measured from the liquid-covered central region by infrared thermography, were 87.6, 90.1, 92.7 and 95.0 °C. The corresponding samples are denoted as S1, S2, S3 and S4, respectively. The deposition time was kept constant, and the growth rate was calculated by dividing the film thickness by the deposition time.

For each liquid-film-temperature condition, three specimens from the same deposition batch were characterized for the magnetic measurements. The values of M_s , H_c , $\mu'_{\max}$, and f_r reported in this work are arithmetic means of these three specimens ($n = 3$). The specimen-to-specimen differences within a batch were small and substantially lower than the differences among the four processing conditions. This protocol therefore evaluates within-batch repeatability; it should not be interpreted as a full assessment of batch-to-batch reproducibility.

2.2. Characterization

The liquid-film temperature was monitored by infrared thermography using a FOTRIC 220-series thermal camera. The crystal structure and preferred orientation were characterized by X-ray diffraction (XRD, Shimadzu Maxima-X XRD-7000, Kyoto, Japan) using Cu K_α radiation. Surface morphology was observed by field-emission scanning electron microscopy (FESEM, JEOL JSM-7800F, Tokyo, Japan). FESEM was used to compare surface morphology qualitatively; no SEM-derived grain-diameter statistics are reported. The average crystallite size L listed was calculated using JADE software (V6.2) from XRD peak broadening according to the Scherrer equation. Room-temperature hysteresis loops were measured using a vibrating sample magnetometer (VSM, Lake Shore 8604, Westmont, IL, USA). The complex permeability spectra were measured using a vector network analyzer (VNA, Agilent N5227A PNA network analyzer, Santa Clara, CA, USA) with a shorted microstrip-line fixture [36]. The maximum real permeability $\mu'_{\max}$ and cutoff frequency f_r were extracted from the measured spectra.

The Scherrer equation used to calculate the XRD-derived average crystallite size L is:

$$L = \frac{0.9\lambda}{\beta \cos \theta} \quad (1)$$

where L is the XRD-derived average crystallite size, λ is the wavelength of Cu K_α radiation, β is the corrected full width at half maximum, and θ is the Bragg angle. Thus, L is an XRD coherence-length estimate rather than a direct SEM grain diameter. For a cubic spinel lattice, the interplanar spacing is given by:

$$d = \frac{a}{\sqrt{h^2 + k^2 + l^2}} \quad (2)$$

where a is the lattice constant. The close-packed $\{hhh\}$ plane composed of O^{2-} and B-site cations has an interplanar spacing of $\sqrt{3}a/6$; substituting this value into Equation (2) gives $h = k = l = 2$, corresponding to the (222) plane [31].

The degree of (222) preferred orientation was evaluated using the Lotgering factor:

$$f_L = \frac{P - P_0}{1 - P_0}, P = \frac{\sum I_{(222)}}{\sum I_{(hkl)}}, P_0 = \frac{\sum I_{0(222)}}{\sum I_{0(hkl)}} \quad (3)$$

Here P is the intensity fraction of the (222) reflection in the measured film, and P_0 is the corresponding fraction for a randomly oriented spinel ferrite reference. The integrated intensities of the spinel diffraction peaks in the measured 2θ range were used for the calculation. In this manuscript, f_L refers only to the XRD-derived Lotgering factor of the (222) orientation.

3. Results

3.1. Liquid-Film Temperature and Structural Parameters

Figure 1 shows the infrared thermal images of the liquid-film during spin-spray deposition. The measured liquid-film temperature increases from 87.6 to 95.0 °C for S1–S4. These values are lower than the temperature of the heating plate, which confirms that the liquid-film temperature is a local reaction parameter rather than a simple reading of the heater set point.

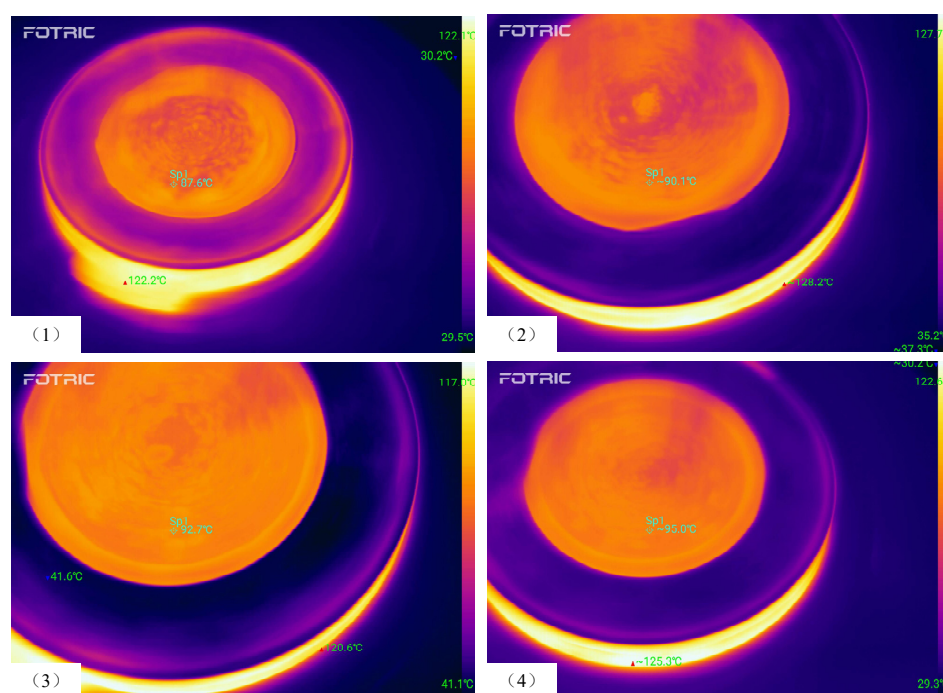


Figure 1. Infrared thermal images of the spin-spray deposition process for (1) sample S1, (2) sample S2, (3) sample S3, and (4) sample S4. The corresponding measured liquid-film temperatures are 87.6, 90.1, 92.7, and 95.0 °C, respectively.

Table 1 summarizes the structural parameters of the films. The Lotgering factor f_L decreases continuously from 0.32 to 0.01 as the liquid-film temperature increases. At the same time, the growth rate decreases from 40 to 31 nm min^{−1}. The XRD-derived average crystallite size L does not change monotonically: it decreases from 148.3 nm for S1 to approximately 93–97 nm for S2 and S3 and then increases to 137.1 nm for S4. The L values

and their reported uncertainties were obtained from the JADE analysis. The lattice constant increases slightly from 8.4082 to 8.4169 Å.

Table 1. Measured liquid-film temperature, XRD-derived (222) Lotgering factor f_L , JADE-calculated average crystallite size L , lattice constant, and growth rate of spin-sprayed NiZn ferrite films. L was obtained from XRD peak broadening using the Scherrer equation and is not an SEM grain diameter.

Sample	Liquid-Film Temperature (°C)	f_L of (222)	Average Crystallite Size L (nm)	Lattice Constant (Å)	Growth Rate (nm min ^{−1})
S1	87.6	0.32	148.3 ± 22.3	8.4082	40
S2	90.1	0.23	93.2 ± 15.2	8.4103	38
S3	92.7	0.06	97.1 ± 36.7	8.4155	33
S4	95.0	0.01	137.1 ± 32.9	8.4169	31

3.2. XRD Texture Evolution

Figure 2 presents the XRD patterns of the four films. All samples show characteristic reflections of the spinel ferrite phase, indicating that crystalline NiZn ferrite films were formed under all liquid-film-temperature conditions. The main difference lies in the relative peak intensities. With increasing liquid-film temperature, the (222) reflection becomes weaker, while the contribution of non-(222) reflections, particularly the (440) reflection, becomes more pronounced. The (400) reflection is weak or not clearly resolved in the lower-temperature samples and becomes more visible at higher liquid-film temperature. This evolution agrees with the calculated f_L values in Table 1.

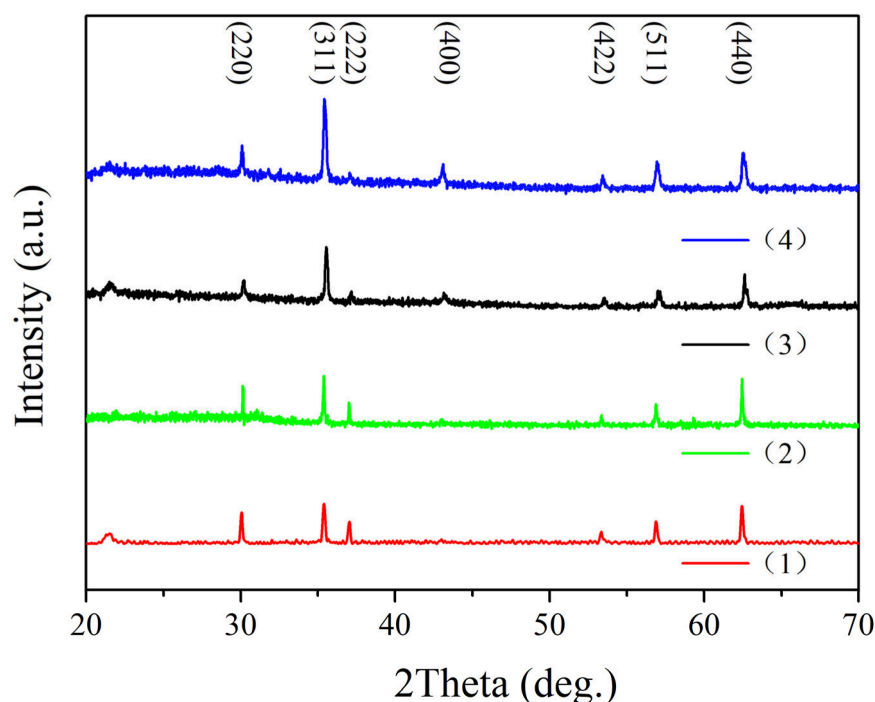


Figure 2. XRD patterns of spin-sprayed NiZn ferrite thin films prepared at different liquid-film temperatures: (1) 87.6 °C, (2) 90.1 °C, (3) 92.7 °C and (4) 95.0 °C.

The gradual decrease in f_L indicates that the liquid-film temperature strongly affects the (222)-oriented growth. At 87.6 °C, the film still contains a clear (222) texture. At 95.0 °C, f_L is only 0.01, meaning that the preferred orientation is almost suppressed. Because f_L is calculated from XRD intensities, this conclusion is independent of the visual judgment of SEM morphology.

3.3. Surface Morphology

The FESEM images in Figure 3 provide morphological evidence for the texture change. S1 exhibits well-defined triangular grains. S2 still contains many faceted triangular grains, although the surface features appear finer. In S3, the triangular feature is much less distinct and the surface becomes more irregular. In S4, the grains are coarser and more agglomerated, and the triangular morphology is largely lost. This sequence follows the decrease in f_L and supports the view that triangular grains are a surface expression of (222)-oriented anisotropic growth. These FESEM observations are qualitative; the numerical L values in Table 1 are XRD-derived crystallite sizes rather than SEM grain diameters.

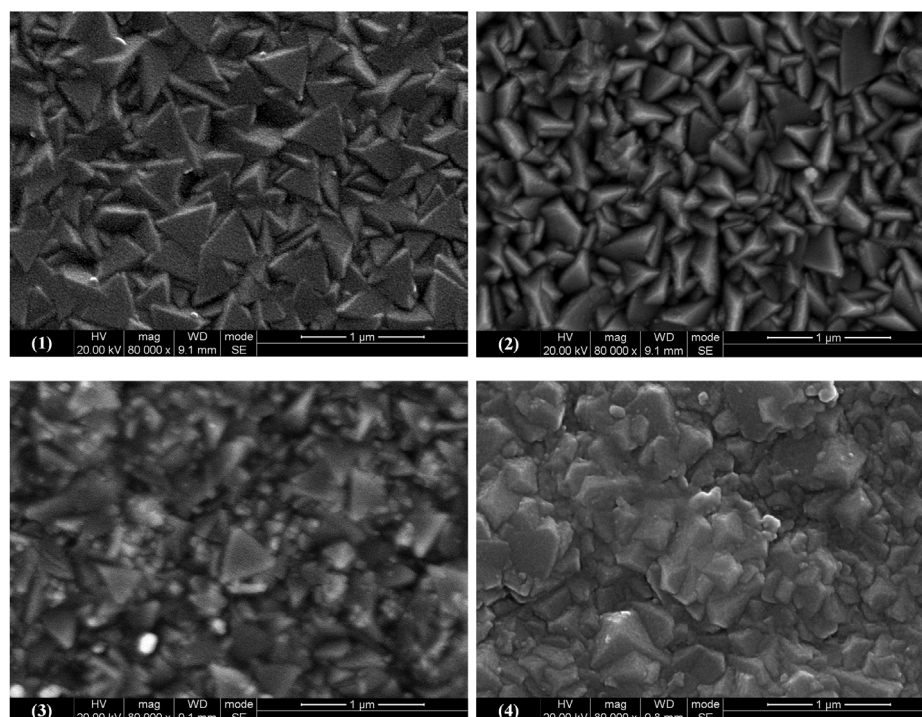


Figure 3. FESEM images of spin-sprayed NiZn ferrite films prepared at different liquid-film temperatures: (1) 87.6 °C, (2) 90.1 °C, (3) 92.7 °C and (4) 95.0 °C.

The triangular morphology can be related to the spinel structure. Oxygen anions and B-site cations form a close-packed {hhh} plane, which corresponds to (222) in cubic indexing. Growth with this plane parallel to the substrate can produce triangular projected facets, so the morphology supports (222)-related anisotropic growth. At lower liquid-film temperature, Fe^{2+} oxidation, dehydration, and cation incorporation are expected to remain concentrated at the liquid-film/growing-film interface, favoring surface-confined growth. At a higher temperature, faster evaporation and greater local supersaturation may promote nucleation within the liquid film and the attachment of less-oriented particles, thereby disrupting (222)-oriented growth. This mechanism remains a working hypothesis because the present measurements do not locate the nucleation events directly.

The XRD-derived crystallite-size change should be considered together with, but distinguished from, the texture and surface-morphology changes. S1 and S4 have comparable average crystallite sizes L , whereas their f_L values differ strongly. Similarly, S2 and S3 have comparable L values, but f_L decreases from 0.23 to 0.06. These comparisons show that liquid-film temperature does not produce a simple monotonic change in L . Instead, the data are consistent with competition among oriented surface growth, nucleation, and local coalescence. Because L is XRD-derived whereas Figure 3 describes surface morphology, the two descriptors are discussed separately.

3.4. Static Magnetic Properties and Magnetic Spectrum Parameters

Figure 4 shows the room-temperature hysteresis loops. All samples exhibit soft ferri-magnetic behavior with narrow loops. The magnetic parameters are summarized in Table 2. The saturation magnetization remains in a narrow range of $429\text{--}442\text{ kA m}^{-1}$. This suggests that the main ferrite phase and the effective magnetic composition are largely preserved as the liquid-film temperature changes. The coercivity decreases slightly from 1.83 to 1.59 kA m^{-1} (23 to 20 Oe), indicating a small reduction in the resistance to magnetization reversal. The values in Table 2 are means from three same-batch specimens; the within-batch variation was small compared with the changes caused by liquid-film temperature.

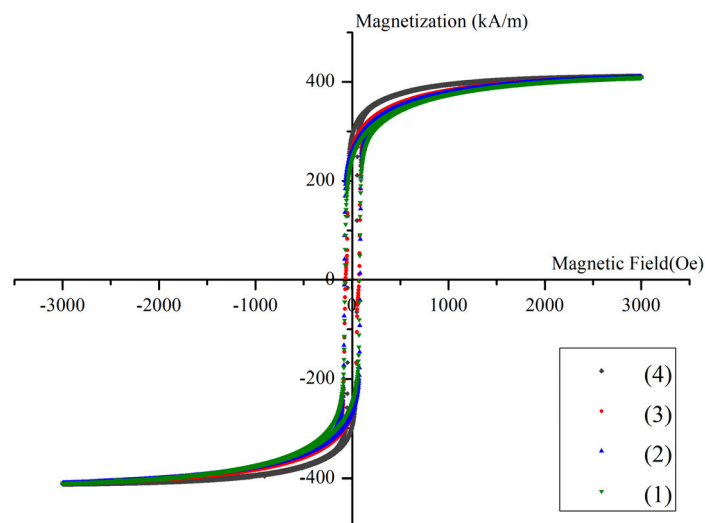


Figure 4. Magnetic hysteresis loops of spin-sprayed NiZn ferrite films prepared at different liquid-film temperatures. For SI reference, $1\text{ Oe} = 79.577\text{ A m}^{-1}$; the original field axis is retained as exported from the instrument.

Table 2. Mean static and dynamic magnetic parameters of spin-sprayed NiZn ferrite films deposited at different liquid-film temperatures. Each value is the arithmetic mean of three specimens from the same deposition batch ($n = 3$); the within-batch specimen-to-specimen variation was small compared with the differences among processing conditions.

Sample	Liquid-Film Temperature ($^{\circ}\text{C}$)	M_s (kA m^{-1})	H_c (kA m^{-1} ; Oe)	f_r (MHz)	$\mu'_{\max}$
S1	87.6	441	1.83 (23)	465	44
S2	90.1	442	1.75 (22)	360	63
S3	92.7	429	1.75 (22)	280	61
S4	95.0	439	1.59 (20)	260	83

The main change occurs in the permeability spectrum. As liquid-film temperature increases, $\mu'_{\max}$ rises from 44 to 83, with a small fluctuation at S3, while f_r decreases from 465 to 260 MHz. Thus, weakening of the (222) texture is accompanied by a shift from a higher cutoff frequency toward higher permeability. All three specimens under each condition showed the same trend, and the within-batch scatter was much smaller than the process-induced shift.

4. Discussion

4.1. Effect of Liquid-Film Temperature on (222)-Oriented Growth

Measured quantities include the liquid-film temperature, XRD and FESEM results, growth rate, and magnetic parameters. Faster evaporation, local supersaturation, off-

interface nucleation, and particle attachment are mechanistic interpretations and are treated below as a working hypothesis.

The present results can be understood from the reaction sequence of spin-spray deposition. In the surface-confined growth picture, ferrous ions adsorb on the substrate or on the growing ferrite surface and are oxidized by NO_2^- . Adjacent hydroxyl groups then dehydrate, and new cations are incorporated into the spinel network. Previous studies associated this process with the close-packed stacking of oxygen anions and B-site cations, which has been proposed to favor the (222) preferred orientation and the triangular surface morphology.

At the lower liquid-film temperature of 87.6 °C, the liquid layer appears to provide sufficient residence time for ion adsorption, oxidation and incorporation at the film surface. This condition is consistent with surface-confined anisotropic growth, giving the highest f_L value and the most evident triangular grains. As the liquid-film temperature increases, evaporation becomes faster and the local supersaturation in the liquid-film may rise. The surface reaction sequence is then more easily disturbed. Part of the reaction products may form in the liquid-film or droplets rather than being incorporated through an ordered surface pathway. This proposed scenario could account for the lower effective growth rate despite the higher temperature.

The nonmonotonic XRD-derived crystallite-size change is also consistent with this working hypothesis. At intermediate liquid-film temperatures, a higher nucleation density and shorter surface residence time may yield smaller L values. At 95.0 °C, local coalescence or agglomeration may contribute to a larger L while f_L remains very low. These statements are interpretations of the correlated structural trends; the present data do not directly resolve the location or time sequence of individual nucleation events.

4.2. Texture-Related Anisotropy and Magnetic Spectrum Shift

M_s changes only slightly across the series, so the large shifts in μ'_{max} and f_r are unlikely to arise mainly from a change in magnetic phase fraction. The spectrum shift is therefore discussed in relation to texture, morphology, and magnetization dynamics rather than phase fraction alone.

Texture evolution is therefore treated as an important correlate, but not as the only possible origin of the magnetic changes. Residual stress, point and extended defects, porosity, grain-boundary pinning, and cation/oxidation-state redistribution can alter the effective anisotropy, damping, domain-wall mobility, and permeability. The nearly constant M_s values argue against a large change in the total magnetic phase fraction, but they do not exclude local changes in cation occupancy or defect structure. Because stress, porosity, and site occupancy were not directly measured, their individual contributions cannot be separated from the texture-related contribution in the present dataset.

At low field, the initial susceptibility can be written as the sum of rotational and domain-wall contributions:

$$\mu_i - 1 = \chi_{\text{rot}} + \chi_{\text{dw}} \quad (4)$$

The rotational response is controlled by an effective restoring field associated with magnetic anisotropy. In a simplified form, this field can be written as:

$$H_k^* = 2K_{\text{eff}}/(\mu_0 M_s) \quad (5)$$

where K_{eff} is an effective anisotropy term that may include crystallographic texture, grain shape effects, internal stress and local pinning. The characteristic frequency of rotational response can be expressed qualitatively by a Kittel-type relation:

$$f_r \propto [H_k^*(H_k^* + M_s)]^{1/2} \quad (6)$$

where H_k^* is used here as an apparent dynamic anisotropy field rather than a single intrinsic magnetocrystalline anisotropy constant. Within this simplified comparative model, and because M_s varies only slightly, the decrease in f_r is consistent with a decrease in H_k^* ; however, it does not constitute a direct measurement of magnetic anisotropy. For a rough comparison, H_k^* can be normalized as:

$$H_k^*(T)/H_k^*(S1) \approx \{[f_r(T)]^2/M_s(T)\}/\{[f_r(S1)]^2/M_s(S1)\} \quad (7)$$

Here, μ_i is the initial relative permeability; χ_{rot} and χ_{dw} are the rotational and domain-wall susceptibility contributions; K_{eff} is an effective anisotropy energy density; μ_0 is the permeability of free space; M_s is the saturation magnetization; f_r is the characteristic cutoff/resonance-related frequency used for comparison; and H_k^* is an apparent dynamic anisotropy field. The superscript asterisk distinguishes H_k^* from a directly measured intrinsic anisotropy field.

Using Table 2, the normalized H_k^* values for S1–S4 are approximately 1.00, 0.60, 0.37, and 0.31. The trend follows the decrease in f_L and is consistent with a weaker apparent restoring field. These values are comparative indices, not absolute anisotropy fields, and do not separate magnetocrystalline, stress, shape, defect, and pinning contributions.

The permeability–frequency trade-off is clear: S1 has the strongest (222) texture and highest f_r but lowest μ'_{max} , whereas S4 has almost no (222) texture, the highest μ'_{max} , and the lowest f_r . S1 and S4 have similar L values, so L alone cannot explain the spectrum shift. The decrease in H_c and the higher μ'_{max} at lower f_r are consistent with a larger domain-wall contribution, although this interpretation remains indirect [37,38].

4.3. Implications for Magnetic-Core Thin-Film Inductors

For thin-film inductors, permeability and cutoff frequency must be considered together. Higher μ'_{max} can increase magnetic-flux linkage and inductance density, whereas f_r sets the usable frequency range. S4 ($\mu'_{\text{max}} = 83$, $f_r = 260$ MHz) favors higher permeability in a lower-frequency window; S1 ($\mu'_{\text{max}} = 44$, $f_r = 465$ MHz) provides a wider high-frequency margin. The nearly constant M_s and low H_c show that this tuning retains soft-ferrimagnetic behavior. Device-level gains in inductance, quality factor, and core loss must be verified in fabricated inductors.

Spin-spray deposition provides aqueous, vacuum-free crystallization near 100 °C without post-deposition annealing [23–26], which is advantageous for low-thermal-budget integration. The present μ'_{max} (44–83) and f_r (260–465 MHz) ranges are comparable to those reported for spin-sprayed NiZn ferrite films [27,28,33,34]. S4 exceeds the maximum μ'_{max} reported in Refs. [33,34], but at a lower f_r . Because composition, thickness, processing, and measurement conditions differ, this comparison is contextual rather than a direct performance ranking.

The slight increase in lattice constant may reflect residual stress, cation redistribution, or oxidation-state changes, but these factors were not measured directly. Likewise, H_k^* is a phenomenological index inferred from the permeability–frequency response, not a direct anisotropy measurement, and may include contributions from domain-wall motion, stress, defects, porosity, cation distribution, shape, and pinning. Cross-sectional TEM/SAED, XPS, Raman spectroscopy, ferromagnetic resonance, angular VSM, and independent-batch

replication are planned to test the proposed growth mechanism, quantify anisotropy and damping, and assess batch-to-batch reproducibility.

5. Conclusions

NiZn ferrite films were prepared by spin-spray deposition at measured liquid-film temperatures of 87.6, 90.1, 92.7, and 95.0 °C. The XRD-derived Lotgering factor f_L decreased from 0.32 to 0.01 with increasing temperature.

This decrease was accompanied by the loss of triangular morphology and a reduction in growth rate from 40 to 31 nm min^{−1}. The average crystallite size L changed nonmonotonically, consistent with, but not proof of, competition among nucleation, oriented growth, and coalescence.

M_s remained nearly constant, whereas $\mu'_{\max}$ increased from 44 to 83 and f_r decreased from 465 to 260 MHz. The normalized apparent dynamic anisotropy field decreased to about one-third of its initial value. Within the simplified model, this trend is consistent with a weaker effective restoring field and a larger domain-wall contribution, but direct ferromagnetic-resonance and domain-sensitive measurements are required.

Liquid-film temperature is therefore a useful low-temperature control parameter for tuning the texture and permeability–frequency window of spin-sprayed NiZn ferrite films. The results support subsequent integration into magnetic-core inductors, although device-level improvements in inductance, quality factor, core loss, and power efficiency remain to be verified.

Author Contributions: Conceptualization, H.L.; Data Curation, H.L. and Y.L.; Investigation, J.Z.; Methodology, J.Z. and W.L.; Project Administration, X.S.; Supervision, K.S.; Writing—Original Draft, H.L.; Writing—Review and Editing, X.S. and K.S. All authors have read and agreed to the published version of the manuscript.

Funding: This research was supported by the Key R&D Program of Shandong Province, China (project number 2023CXGC010306), the Hubei Provincial Natural Science Foundation—Innovation and Development Joint Fund (project number 2023AFD052) and the High-level Talent Start-up Project of West Anhui University (WGKQ2021052).

Data Availability Statement: The original contributions presented in this study are included in the article. Further inquiries can be directed to the corresponding author.

Conflicts of Interest: Authors Xinglian Song and Wenju Liao are affiliated with Shandong Chunguang Magnetoelectric Technology Co., Ltd. The authors declare that this affiliation does not constitute a conflict of interest affecting the reported results.

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