

Law Governing the Intrinsic High-Frequency Permeability of Magnetic Composites: Uncertainty Principle

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Abstract

The intrinsic high frequency permeability spectra of ferromagnetic composites containing different volume fractions of iron and cobalt have been simulated. A law (called Mghan's law) is proposed to explain the simulated results by assuming that there are plenty of LLG (Landau-Lifshitz-Gilbert) type natural resonances contributing to the intrinsic permeability spectra. The results clearly show that the spectra strongly depend on the distribution of local effective magnetic field, the interaction between the magnetic particles, the inhomogeneous damping constant of LLG precession, and the initial equilibrium states. Especially, the effect of particles shape distribution in each sampling on the local effective magnetic field. In view of this fact: it is absolutely impossible to have the same effect from these factors when someone prepares several measurement samples, an uncertainty principle is believed to hold for measuring the intrinsic permeability of an electromagnetic (EM) composite. Therefore, this law tells us that it should be cautious when comparing or evaluating the EM properties of composites (for instance, EM wave absorbing composites). Memory effect can be used to restore the intrinsic high frequency permeability for a specific defunct composite sample.

Full Text

Preamble

Law Governing the Intrinsic High Frequency Permeability of Magnetic Composites: Uncertainty Principle

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Author Contribution Statements: HAN has proposed the research ideas, designed the simulation details, analyzed the data, and prepared the whole manuscript.

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Abstract

The intrinsic high frequency permeability spectra of ferromagnetic composites containing different volume fractions of iron and cobalt have been simulated. A law (called Mghan's law) is proposed to explain the simulated results by assuming that numerous LLG (Landau-Lifshitz-Gilbert) type natural resonances contribute to the intrinsic permeability spectra. The results clearly demonstrate that the spectra strongly depend on the distribution of the local effective magnetic field, the interaction between magnetic particles, the inhomogeneous damping constant of LLG precession, and the initial equilibrium states. Particularly noteworthy is the effect of particle shape distribution within each sampling on the local effective magnetic field. Given this reality—it is absolutely impossible to achieve identical effects from these factors when preparing multiple measurement samples—an uncertainty principle is believed to hold for measuring the intrinsic permeability of electromagnetic (EM) composites. Therefore, this law cautions that care must be exercised when comparing or evaluating the EM properties of composites (for instance, EM wave absorbing composites). The memory effect can be used to restore the intrinsic high frequency permeability for a specific defunct composite sample.

Keywords: permeability; micromagnetics simulation; damping constant; natural resonance; permeability memory effect; Mghan's law

1. Introduction

Understanding magnetic permeability (μ) at high frequencies is critical for developing magnetic devices and materials that operate at high frequencies. Electromagnetic attenuation requires larger imaginary parts (μ'') of complex permeability to dissipate electromagnetic energy via substantial magnetic losses [1]. Conversely, planar magnetic inductors and microwave devices demand smaller μ'' values to minimize power losses [2]. It has also been proposed that the imaginary parts of permeability can become negative ($\mu'' < 0$) under the influence of spin transfer torque [3].

Two well-known laws govern the relationships between permeability and ferromagnetic resonance frequency. The first is “Snoek's law,” which describes the inverse relationship between initial permeability and the loss peak of the permeability spectrum [4]. The second is Acher's law [5], a revised version of Snoek's law expressed as an integral of the product ($\mu'' \times f$), stating that the integration of a magnetic loss spectrum must be smaller than or equal to a constant ($4\pi M_s$),

where M is saturation magnetization). However, neither law explains the relationship between the frequency dependence of intrinsic permeability and the microstructures (including magnetic domain structures) of magnetic materials, nor do they reveal the underlying physics governing the intrinsic permeability of magnetic composites.

This limitation is particularly problematic for electromagnetic composites, which contain numerous ferromagnetic particles (or ferrite particles) in unsaturated states, with irregular shapes randomly distributed throughout the matrix. These two laws cannot adequately explain the variation in intrinsic permeability dispersion. Furthermore, many published papers claim excellent electromagnetic properties for various composites, such as superior electromagnetic wave absorption characteristics. Can these “excellent” properties be experimentally reproduced by other researchers? In other words, what is the probability of replicating these results? Neither Snoek’s law nor Acher’s law can address these questions. This paper delves into these critical issues.

In practice, it is frequently observed that the permeability spectrum ($\mu''(f)$) exhibits a broad peak, especially for magnetic composites with inhomogeneous magnetic and material microstructures [6,7]. Numerous effective medium laws have been proposed to retrieve the intrinsic permeability spectra from composite data consisting of magnetic particles (commonly flakes) and non-magnetic matrices (such as polymers) [6]. These approaches typically yield broad intrinsic permeability spectra. Here, “intrinsic” permeability refers to the permeability wholly attributed to magnetic materials, excluding dilution effects from the non-magnetic matrix and eddy current effects. Several important questions regarding this intrinsic permeability remain unanswered: What factors determine the dispersion behaviors of permeability—why do permeability values vary significantly with frequency? What governs the shape of the spectrum? How do interactions between magnetic particles impact the obtained intrinsic permeability? We believe that the intrinsic permeability of magnetic components significantly influences the measured effective permeability of composites. Therefore, understanding the frequency dependence of intrinsic permeability is critical and helpful for developing high-frequency magnetic devices and composites. To address these questions, this paper employs a micromagnetics simulation tool widely used in the magnetism community to study the intrinsic high frequency permeability of a system containing two magnetic phases.

2. Simulation Backgrounds and Details

Micromagnetics is a continuum theory for studying both static magnetization behaviors (hysteresis, magnetic domains) and dynamic responses of magnetization. It describes magnetization on a length scale large enough to replace atomic magnetic moments with a continuous function of position, yet small enough to reveal transitions between magnetic domains. The micromagnetics simulations are performed using a three-dimensional object-oriented micromagnetics framework (OOMMF) by solving the Landau-Lifshitz-Gilbert (LLG) equation as a

function of time [8-9]:

$$= -\gamma(\vec{M} \times \vec{H}_{eff}) + (\vec{M} \times$$

—(1)

where M is the saturation magnetization and $H_{\{eff\}}$ is the effective field accounting for exchange, demagnetization, anisotropy, and applied field terms. γ is the Gilbert gyromagnetic ratio (2.21×10^5 m/A · s), and α is the damping constant for magnetization precession.

In this work, the frequency dependence of permeability for a ferromagnetic plate ($400 \text{ nm} \times 200 \text{ nm} \times 20 \text{ nm}$, i.e., Length(L) \times Width(w) \times Thickness(T)) is simulated following these procedures. First, the equilibrium configuration of magnetization is acquired in the absence of an external applied magnetic field. Second, a weak pulse magnetic field with the form $h(t) = 100\exp(-10^9 t)$ (t in s, h in A/m) is applied along the “z” axis (see Fig. 2 [Figure 2: see original paper]). This pulse field is too weak to drive magnetic domain walls. The temporal response of magnetization can be expressed as:

$$\delta\vec{M}(r_i, t) = \vec{M}(r_i, t) - \vec{M}_0(r_i, t) = \chi(r_i, t)\delta\vec{h}(t)$$

—(2)

The dynamic response of magnetization over the entire sample volume is recorded. Both the pulse field and excited magnetization are then processed using a fast Fourier transform (FFT) approach, after which the relative permeability spectrum is calculated as follows:

$$\mu'(f) - j\mu''(f) = 1 + \chi'(f) - j\chi''(f)$$

—(3)

$$\mu'(f) = 1 + \chi'(f)$$

—(4)

$$\mu''(f) = \chi''(f)$$

—(5)

where f is frequency, $m(f)$ and $h(f)$ are the frequency-domain expressions for M and the pulse field (h) after FFT treatment, respectively; ' refers to the real parts of susceptibility and '' denotes the imaginary parts.

The default parameters in OOMMF for the Co phase are used in our simulations: saturation magnetization (M) is 14×10^5 A/m, exchange stiffness constant (A) is 30×10^{-12} J/m, and anisotropy constant (K_1) is 5.2×10^5 J/m³. For the Fe phase, M is 17×10^5 A/m, A is 21×10^{-12} J/m, and K_1 is 4.7×10^4 J/m³. For all simulations, the object is discretized into many tetrahedrons with a cell size of $2.5 \text{ nm} \times 2.5 \text{ nm} \times 2.5 \text{ nm}$.

The analytical solutions of $\mu'(f)$ and $\mu''(f)$ for natural resonance from the LLG equation can be expressed as follows [10], where “ ω ” is the angular frequency ($\omega = 2\pi f$):

$$\mu' = 1 + \frac{[M\gamma^2 H_0(\gamma^2 H_0^2 - \omega^2) + 2\omega^2 \alpha^2]}{\alpha\omega(\gamma^2 H_0^2 + \omega^2)}$$

— (6)

$$D = (\gamma^2 H_0^2 - \omega^2)^2 + 4\omega^2 \alpha^2$$

— (7)

In these equations, H_0 can be expressed as follows, which approximately equals H_{eff} since α (0.5 used in this paper) is much smaller than (γM) . f_r is the resonance frequency of one LLG natural resonance:

$$2\pi f_r / \gamma = H_{\text{eff}}(1 + \gamma^2 M^2)^{1/2} \approx H_{\text{eff}}$$

— (8)

3. Results and Discussions

3.1. Intrinsic High Frequency Permeability of Two Magnetic Phases

In our simulations, a nanostructured magnetic system with two magnetic phases of different volume fractions is constructed using the “Voronoi diagram” method [11]. As shown in Fig. 1 [Figure 1: see original paper], the Fe and Co nanoparticles inside a ferromagnetic plate (simulation object) are randomly oriented and irregularly shaped, resembling the microstructures of actual magnetic composites such as thin films prepared by co-deposition techniques. The volume fractions of the iron phase are 90.37% and 62.49% in Fig. 1a and 1b, respectively.

The initial spontaneously magnetized states for cases with both phases coexisting and for cases with only one Fe phase (or only Co phase) are compared, as shown in Figs. 2, 3, and 5. In Fig. 2, the magnetization distributions in each composite sample are inhomogeneous, clearly indicated by the colors and directions of arrows. The equilibrium direction of a magnetization vector depends

on the local effective magnetic field (H_{eff}), suggesting that H_{eff} values within the sample are different and exhibit a wide distribution.

This inhomogeneity is believed to influence the frequency dependence of relative permeability (referred to as “permeability dispersion” in this work). For some applications (e.g., planar magnetic inductors) [12], magnetic materials are magnetized to saturation to align almost all magnetization in the same direction. In this case, the permeability dispersion spectrum shows a sharp shape with high μ' values (real parts of complex permeability), and particularly low μ'' values (ac magnetic losses), which is one of the technically essential requirements. Narrow magnetic loss spectra $\mu''(f)$ with loss peaks far from the working frequency are necessary for many electromagnetic devices. Conversely, some applications require large high-frequency magnetic losses, such as electromagnetic noise suppressors [13,14]. In these applications, magnetic composites contain numerous magnetic particles that are usually not magnetized to saturation, existing instead in their spontaneously magnetized states—similar to the conditions shown in Fig. 2. The random orientation of local magnetizations implies a wide distribution of local effective fields, resulting in spread-out high-frequency permeability spectra, as will be demonstrated.

To elucidate the law governing the intrinsic high frequency permeability of magnetic materials—especially those with two ferromagnetic phases and many irregularly shaped particles—we decompose the composite plate into two parts in subsequent separate simulations without changing the original particle positions. This decomposition resembles jigsaw puzzles and is crucial for eliminating effects from altered particle positions and size distributions. It is critical that this prerequisite be followed; otherwise, the composite would not be identical, rendering the discussions meaningless. The equilibrium magnetization configurations of only the Fe phase in composites with different volume fractions are illustrated in Fig. 3, where empty zones represent the original locations of cobalt particles (where Co phase: $A = 0$; $M = 0$; $K = 0$). Subsequently, the permeability spectra of each phase are simulated separately for composites with different volume fractions, as shown in Fig. 4 [Figure 4: see original paper] (Fe phase only) and Fig. 6 [Figure 6: see original paper] (Co phase only). In both cases, the permeability spectra exhibit spreading behavior. For most electromagnetic applications (including this work), the microwave magnetic field is too weak to drive domain walls, eliminating any contribution from domain wall resonance mechanisms (typically with $f_{\text{r}_{\text{DW}}}$ in MHz). Natural resonance is the sole mechanism, as shown in the LLG equation (1). However, the LLG equation describes the behavior of natural resonance for only a single magnetization vector. LLG natural resonance also applies to magnetic materials in a magnetized saturation state (single domain), where saturated magnetization can be treated as one “macrospin” [12]. Nevertheless, electromagnetic composites contain many magnetic fillers that are not in a saturated state, and their permeability dispersion spectra generally resemble those shown in Fig. 4 [7].

We assume that the observed broad spectra result from the coexistence of many

LLG natural resonances. For any given initial equilibrium magnetized state (1, 2, 3...), a generalized law (called Mghan's law) is proposed: summing the permeability dispersion spectrum of each LLG resonance yields the observed spectra. Suppose the magnetic material is in one magnetic equilibrium state "s":

$$\mu(f) = \left(\sum w_i \times \mu(f_r, \alpha)_i \right) + \varepsilon$$

—(10)

$$f_r(i) = \gamma H_e(i)$$

—(11)

$$\vec{H}_e(i) = \vec{H}_k(i) + \vec{H}_s(i)$$

—(12)

$$H_s(i) = 2K_s(i)$$

—(13)

$$K_s(i) = \mu_0 \times \Delta N_i \times M_s^2$$

—(14)

where w_i is the weight of one LLG natural resonance (i) among all resonances (n). ΔN is the demagnetization factor difference between the hard axis and easy axis. Each LLG resonance has a specific permeability dispersion $\mu(f, \alpha)_i$ with a resonance frequency $f_r(i)$ and a specific damping constant α_i . $H_e(i)$ is the local effective anisotropic field. Since composites contain many irregular particles (such as flakes), ΔN follows a lognormal distribution, and consequently, $H_e(i)$ and $f_r(i)$ values also follow this distribution according to the above equations.

is an error term due to interactions between magnetic phases and nonuniform damping constants, which will be discussed shortly. From an engineering perspective with initial spontaneously magnetized states, special attention should be paid to the distributions of H_k and H_s . H_k strongly depends on material selection (i.e., compositions), while H_s strongly depends on fabrication techniques (such as ball milling, thin film deposition, microwire drawing). The distribution of H_s (and thus (f) spectra) can be controlled by using sieves to regulate the lognormal distribution parameters (such as "mode," "median," "mean," or "standard deviation") of ΔN for a batch of magnetic powder. If attempting to control permeability spectra by controlling remanence states (discussed later), the distribution of H_{ex} also plays an important role and should

be included in Eq. (12). The mathematical expression for the H_{ex} distribution is very complex but can be mapped by FFT techniques, as evidenced in our published paper [15].

For a single LLG natural resonance, the resonance frequency (f_r) of the Fe phase is calculated to be 1.58 GHz if only the magnetocrystalline anisotropic field (H_k) is considered ($H_k = 2k/(0M)$). Taking into account the aspect ratio (L/T) of Fe particles, the shape anisotropic constant (K_s) can be expressed as: $K_s = 0.5 \times 0 \times \Delta N \times M^2$. ΔN is approximately 0.5 for Fe particles in the composites. The f_r value becomes 31.0 GHz when simultaneously considering both magnetocrystalline and shape anisotropic fields (H_s): ($H_e = H_k + H_s$). This value of 31.0 GHz corresponds approximately to the peak frequencies simulated and shown in Fig. 4.

The results for composites containing only cobalt particles are presented next. The equilibrium magnetization states of the Co phase are illustrated in Fig. 5, where empty areas (with Fe phase set to: $A = 0$; $M = 0$; $K = 0$) represent the locations of Fe particles in the original nanocomposites from Fig. 2. A similar inhomogeneous distribution of magnetization vectors indicates an inhomogeneous distribution of local effective magnetic field. The permeability spectra are also expected to spread out, as simulated and shown in Fig. 6. The shape of these permeability spectra can be explained using the proposed Mghan's law in Equations 10-14. If only H_k is considered, a typical LLG natural resonance would have an f_r value of 20.8 GHz. If both H_k and H_s are considered, f_r should be 45.4 GHz, which does not appear in Fig. 6a. According to Mghan's law, both H_s and H_{ex} have wide distributions that result in a broad distribution of f_r values [15]. When Fe particles are absent and no longer interact with Co particles (Fig. 5), the initial magnetized states of Co particles change significantly. Consequently, the distributions of H_s and H_{ex} values are greatly affected, which critically determines the resulting spectra. A simulated intrinsic permeability spectrum in this work represents a weighted average of many LLG-type natural resonances. Furthermore, by controlling remanence states or particle shape distribution (ΔN), we can easily control the distribution of H_e and thus the permeability spectra [15].

When both Co and Fe nanoparticles coexist (see Fig. 2) and their microwave permeability spectra are simulated, the results are shown in Fig. 7 [Figure 7: see original paper] and marked as ($'(AB)$, $''(AB)$). Additionally, spectra marked “(A+B)” indicate simple addition of the permeability spectra for Fe and Co phases from Fig. 4 and Fig. 6. When the Co volume fraction is much smaller, the difference between the “(AB)” spectra and “(A+B)” spectra is not significant. However, when the volume fractions of Fe and Co are comparable, the discrepancy between the “(AB)” and “(A+B)” spectra becomes obvious, as shown in Fig. 7b. This is believed to result from stronger interactions among Fe and Co particles in Fig. 1b.

Such interaction scenarios universally exist in electromagnetic composites and can strongly affect static magnetic parameters such as coercivity and rema-

nence. In our previous work on Fe nanowire array permeability spectra, we showed that when the spacing between nanowires is sufficiently small, localized nanowires exhibit different coercive fields, indicating strong interactions between nano-entities [16]. What more can we learn from this finding? Suppose we have an unchanged particle distribution (exactly identical particles) and a fixed volume fraction for either Fe or Co particles in their spontaneously magnetized states (i.e., unsaturated). Each time, we simply mix these same particles differently and measure the permeability spectra of the composites. Since it is absolutely impossible to ensure that the surroundings of each particle remain unchanged across these composites, the magnetic particle interactions will inevitably differ. Moreover, variable particle distributions in each sampling from a batch of powder will lead to unexpected spectral shapes. Therefore, we can claim that it is impossible to experimentally reproduce permeability spectra. A typical application of electromagnetic materials involves developing electromagnetic wave absorbing composites containing randomly oriented particles. For this application, absorbing properties critically depend on permeability spectra. Based on the above discussion of unexpected permeability spectra, it is reasonable to believe that the measured performance of electromagnetic wave absorbing composites is uncertain. We propose naming this the “Uncertainty Principle.” It should be noted that when evaluating whether a specific magnetic material is suitable for electromagnetic wave absorption, judgments cannot be based on just a few measurements. Instead, numerous measurements (i.e., statistical sampling) are needed to determine the probability of obtaining “good” properties.

Furthermore, if we also consider variations in particle distribution across different measurement samples containing the same magnetic fillers—even when these fillers were prepared under identical conditions (typically ball milling)—the above viewpoint regarding the uncertainty of permeability spectra becomes even more compelling. Although many mixing laws exist for studying composite permeability spectra, they ignore the fact that the “uncertainty” demonstrated in this paper exists from sample to sample, including statistical distributions of particle shapes (average, standard deviation, median, mode), particle agglomeration, and initial magnetization configuration patterns.

3.2. Effect of Damping Constants

For a single LLG natural resonance, the damping constant (α) determines how quickly magnetization (M) returns to its equilibrium position during precession and is positively related to the magnitude of high-frequency magnetic loss. However, when numerous LLG natural resonances exist in a material, the term “damping constant” becomes misleading. The intrinsic origins of α include spin-orbit coupling and scattering by magnetic impurities. The intrinsic α is NOT constant; it is space-dependent and strongly related to nonuniform precessions (as demonstrated in this work) [17-20]. Additionally, microstructural inhomogeneity gives rise to a distribution of damping constants (known as extrinsic α)

[21-23]. However, only a single damping constant is used in the LLG equation, which does not reflect reality. Here, simulations demonstrate the impact of different damping constants on high-frequency permeability spectra, as shown in Fig. 8 [Figure 8: see original paper]. First, the composite with two phases (Fe: volume fraction = 90.37% and Co: volume fraction = 9.63%) is assumed to have a homogeneous damping constant ($\alpha = 0.5$). The composite spectra are simulated and marked with “(AB).” Second, each phase’s spectrum is separately simulated with different damping constants (for example, Fe: $\alpha = 0.3$; Co: $\alpha = 0.7$) and added to obtain the composite permeability spectra (marked with “A+B”). The poor agreement between the “AB” and “A+B” spectra indicates that inhomogeneous damping constants matter significantly.

Additionally, with advances in modern magnetism, particularly spintronics, it has been found that the damping behavior of LLG natural resonance can be affected by spin transfer torque (STT). We have reported that the imaginary parts of permeability can become negative ($\mu'' < 0$) under STT effects [3]. Moreover, microwave magnetic losses can be suppressed or enhanced by STT through its influence on the effective damping constant. Accordingly, permeability spectra can also be actively controlled by STT effects [24].

3.3. Effect of Remanent States (Memory Effect)

In practice, the high-frequency properties of electromagnetic composites containing magnetic fillers often deteriorate due to decay of their magnetic properties. The equilibrium magnetized states (such as those discussed above) are easily affected by external magnetic fields or temperature fluctuations, making permeability spectra unstable. Can we recover these spectra? Technically, this is feasible if we remagnetize the materials to reestablish the original remanent state. Here, we demonstrate the dependence of permeability spectra on remanent states. First, we magnetize the composite (Fe: vol. 62.49%; Co: vol. 37.51%) along the “x” axis with different maximum magnetic fields (see points A, B, and C where A = 50 mT, B = 80 mT, and C = 20 mT) starting from the initial fully demagnetized state (“O” point in Fig. 9a [Figure 9: see original paper]). Second, the magnetizing field is slowly reduced to zero, leaving the sample in different remanent states (see points A’ and B’). The composite is identical to that in Fig. 2b but with slightly different initial magnetization states. The dynamic permeability spectra are simulated as before. We find that identical remanent states produce identical permeability spectra. For instance, different maximum magnetizing fields (A and C) yielding the same remanent state (A’) produce identical dynamic permeability spectra. We term this phenomenon the “Memory Effect.” The term “memory” signifies that reestablishing the remanent state causes the permeability spectra to reappear as if from the magnetic material’s memory. The vectorial magnetization distributions of these remanent states are obviously different, as shown in Fig. 9(b, c, d). Different remanent states (O, A’, and B’) give rise to different permeability spectra, as shown in Fig. 10 [Figure 10: see original paper]. Since the difference between

remanent states “O” and A’ is not dramatic, their permeability spectra are similar. However, the spectra for state B’ and state O are distinct. These results imply that any factor strongly influencing a sample’s remanent state will produce different dynamic magnetic responses. Therefore, when comparing results for identical materials from different research groups, we must consider that their remanent states might differ. Note that the frequency axis uses a logarithmic scale in Fig. 10. The differences in permeability spectra between Fig. 10a and Fig. 7b (see spectra labeled “AB”) arise from their different initial magnetization configurations, despite being the exact same composites.

Interestingly, the well-known Snoek’s law does not hold in Fig. 10a and 10c. Snoek’s law states that the loss peak frequency (f_r) is inversely proportional to initial permeability (μ_i), meaning the loss peak frequency can be increased only at the expense of permeability, and vice versa. By controlling remanent states, the distribution of H_e components (i.e., H_d , H_{ex}) can be changed [15], thereby significantly altering the permeability spectra.

4. Conclusions

The intrinsic high frequency permeability spectra of composites containing different volume fractions of Fe and Co particles have been simulated. A law (called “Mghan’s law”) is proposed to explain the spreading of permeability spectra. It is believed that the local effective magnetic field, magnetic particle interactions, inhomogeneous damping constants, and initial remanent states strongly impact the spectra. The numerous LLG natural resonances arise from inhomogeneous local magnetic fields. An uncertainty principle is believed to hold for the electromagnetic properties (such as EM wave absorption) of composites using magnetic particles as functional fillers. The memory effect of permeability is also reported.

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