Acher's constraint on the high-frequency magnetic performance of composites

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Abstract

The paper generalizes the results available from the literature on the constraint on the high-frequency permeability of magnetic composites, with the stress made on the opportunities for obtaining of materials with high microwave permeability. The rigorous derivation is presented of the integral constraint on the permeability. A simple estimation of the effect of eddy currents is given. The applicability limits and opportunities to overcome the constraint are discussed. That the constraint is believed to be invalid in flake particles with hard magnetic axis perpendicular to the flake plane and in tiny magnetic particles possessing exchange resonance modes.

1. Introduction

Application of magnetic composites in microwave devices often requires multi-scale modeling of the performance. In the course of this, the effective material parameters of the composite are estimated with the use of mixing rules; then, an estimate is made of parameters of the device based on these composites. In such studies, application of constraints on the microwave performance materials is often helpful. Examples are the analysis of the ultimate bandwidth of magnetic radar absorbers [1] and the study patch antennas with magneto-dielectric substrates [2].

The microwave magnetic behavior of materials is determined by the ferromagnetic resonance. In the first approximation, the complex permeability, $\mu=\mu'+i\mu''$, of a material is close to the static permeability, μ_s, at frequencies bellow the ferromagnetic resonance frequency, f_r, and is close to unity at frequencies above the resonance. The microwave permeability is large if both μ_s and f_r are high, which, however, is constrained by well-known Snoek's law. Snoek's law follows readily from the Landau–Lifshits (LL) equation for the susceptibility of a uniformly magnetized spherical particle and is known to hold for most isotropic polycrystalline magnetic materials.

For non-spherical particles, Snoek's law may be not valid. From the standpoint of obtaining of high microwave permeability, of the most interest is the case when two of three effective demagnetization fields involved in the LL equation are close to zero. Then, one of these fields determines the direction of permanent magnetic moment in the particle and the permeability along the other direction is given by [3]:

$$(\mu_s - 1)\cdot f_r^2 = (\gamma 4\pi M_0)^2, \tag{1}$$

with the saturation magnetization $4\pi M_0$ and $\gamma\approx 3$ GHz/kOe. The permeability along two other principal axes of the ellipsoid is close to unity. Equation (1) may yield permeability values significantly larger than those permissible by Snoek's law. Therefore, Eq. (1) is considered as an ultimate constraint for the microwave permeability of magnetic materials. The same, the equation limits the opportunities for obtaining of materials with high microwave magnetic loss.

Most microwave applications require bulk non-conducting materials. This is mostly because the high-frequency microwave magnetic performance of bulk ferromagnetic material is deteriorated by the effect of eddy currents, since most ferromagnets are conductors. Therefore, magnetic composites are of interest comprising inclusions that obey Eq. (1). Two classes of such inclusions are known, namely, flake inclusions and microwires with circumferential magnetization, both attracting a great attention recently, see [4] for the review of experimental data.

The paper generalizes the results available from the literature on the constraint on the high-frequency permeability of magnetic composites. This problem has also been considered in [5], which contains a discussion aimed mostly at the use of measured data on microwave permeability for obtaining of data on the structure and properties of magnetic constituents. In the present paper, the stress is made on the opportunities for obtaining of materials with high microwave permeability.

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2. Integral constraint on microwave magnetic performance of composites

Equation (1) is derived under assumption of Lorentzian frequency dependence of permeability. A generalization of Eq. (1) to the cases of arbitrary dispersion laws and inhomogeneous materials has been introduced by Acher and coworkers based on heuristic consideration [6]. It is frequently referred to as Acher's constraint and is given by:

$$\frac{2}{\pi}\int_0^\infty \mu''(f) f\, df \leq pk(\gamma 4\pi M_0)^2 \tag{2}$$

where f is the frequency, p is the volume fraction of the magnetic constituent, $0<p<1$, and k is a factor accounting for the orientation of magnetic moment relatively to the microwave magnetic field. For composites with random orientation of inclusions, k=1/3; for uniformly orientated flake particles with randomly orientated in-plane anisotropy, k=1/2.

The derivation is analogous to that of the Kramers–Kronig relations and is based on the treatment of complex permeability as an analytic function of complex frequency. It involves application of the Cauchy theorem; representation of an arbitrary magnetic dispersion law as a sum of Lorentzian terms; and determination of high-frequency asymptotes of these terms from the LL equation. For composites, validity of Eq. (2) has been deduced in [6] from the equality of its left part for higher and lower Hashin–Shtrikman limits. A rigorous derivation of Eq. (2) for any form of magnetic dispersion law has been given in [7] and, independently, in [5]. Introducing the volume fraction into Eq. (2) is based on the high-frequency asymptote for composites follows from the Landau–Lifshitz–Looyenga mixing rule,

$$\mu = \left(1 + p\left(\mu_i^{1/3} - 1\right)\right)^3 \approx 1 + p(\mu_i - 1)\,, \tag{3}$$

which is a rigorous result for the case when the permeability of inclusions in a composite is close to that of the host matrix. Since $\mu \to 1$ at $f \to \infty$, this is the law governing the high-frequency asymptote of permeability for any composite. Hence, the high-frequency susceptibility of a composite is just proportional to the susceptibility of inclusions with a factor of p.

In materials with inhomogeneous crystalline structure, a broad magnetic peak is frequently observed. In terms of the Lorentzian dispersion law, the inhomogeneity of the material increases the Gilbert damping parameter α and, therefore, broadens the magnetic loss peak. Formally, Eqs. (1) and (2) hold in this case as well. However, the frequency involved in the left part of Eq. (1) is the true resonance frequency, i. e., the frequency, above which the real permeability takes values below unity. From the standpoint of technical applications

This frequency is of minor importance for technical applications, since the magnetic absorption peak is located at lower frequencies for the case of large damping. Therefore, the frequency of maximal magnetic absorption, f_a, must be considered as the high-frequency cutoff frequency of magnetic behavior. For this frequency, it readily follows that:

$$\mu_s f_a = \frac{\sqrt{1+\alpha^2}}{\alpha}\left(\mu_s\ f_r\right). \tag{4}$$

The value in the brackets in the right part of (4) is Snoek's constant. Hence, Eq. (1) does not yield an accurate estimate for the microwave permeability in the case of large damping. The properties of the material obey Snoek's law rather than Acher's law.

3. The effect of eddy currents

In this section, distortions of the magnetic dispersion law due to the effect of eddy currents are considered under assumption that the intrinsic permeability of inclusions, μ_i, is governed by the Lorentzian law. The skin effect is conventionally accounted for by the renormaliztion of μ_i to apparent permeability, μ. For the sake of simplicity, the renormalization is written below for the case of flake-shaped inclusion:

$$\mu = \mu_i \frac{\tan\left((1+i)\pi d\sqrt{\mu_i \sigma f}/c\right)}{(1+i)\pi d\sqrt{\mu_i \sigma f}/c} \tag{5}$$

where c is the light velocity; σ and d are the flake conductivity and thickness, respectively. The frequency-dependent magnetic behavior is due to poles of the Lorentzian dispersion curve, $\pm f_{r,i}+if_{r,i}^2/(2f_{d,i})$. Similarly, the frequency dependence of the renormalized permeability (5) is determined by the poles of the right part of Eq. (5). It readily obtained that the effect of eddy current transforms each Lorentzian pole of the intrinsic permeability into an infinite set of poles of the apparent permeability, the Lorentzian parameters of which are given by:

$$\frac{1}{f_{d,i}^{\ *}}=\frac{1}{f_{d,i}}+8\frac{4\pi\chi_{s,i}d^2\sigma}{(2n-1)^2c^2},\ f_{r,i}^{*}=f_{r,i} \tag{6}$$

where n=1,2,…,∞ and the intrinsic permeability is assumed to possess a narrow magnetic absorption band, $f_{r,i}<<f_{d,i}$. The partial susceptibilities $\chi_{s,i}$ are derived from the residues in these poles.

Since the effect of eddy currents affects neither the resonance frequency, as it follows from Eq. (6), nor the static permeability, then, formally, it does not affect Eq. (1). However, eddy currents result in broadening and low-frequency shift of magnetic absorption peak, which may be considered in the same way as in the previous section, see Eq. (4). If the skin effect is well pronounced, then the magnetic absorption peak is formed by eddy currents solely, with negligible contribution from the ferromagnetic resonance, and is located at the frequency where the skin depth is equal to the lowest dimension of the particles. The decrease in the cutoff frequency restricts opportunities for the microwave applications. Again, the constraint on the high-frequency magnetic behavior of the material is given by Snoek's law rather than by Eq. (1).

In addition to poles (6) related to the ferromagnetic resonance, eddy currents arise another set of poles corresponding to the optical permeability of the material. The parameters of the lines of this set are also found from Eq. (5) with μ_i=1. For these lines the resonance frequency is infinity, and, therefore, the present of these results in a divergence of the integral in the left part of Eq. (2). Notice that this set of poles appears in non-magnetic conductors as well, where these are responsible for the effective permeability arising due to the effect of eddy currents.

Therefore, the integral (2) is divergent in conductive magnets due to the effect of eddy currents, even if the intrinsic permeability is of the Lorentzian type, since the spectrum includes a Debye line. For this line,

$$\frac{2}{\pi}\int_0^f \mu'' f\, df \approx \frac{c^2 f}{4\pi\mu_s d^2\sigma} \tag{7}$$

This contribution to the integral (2) increases with frequency and becomes equal to that from ferromagnetic resonance at $f\approx 4\pi\mu_s^2 f_r^2 d^2\sigma/c^2$, which is typically about hundreds of GHzs. Below this frequency, the left part of Eq. (7) does not depend on frequency and is related to the magnetostatic properties of the magnetic material. Therefore, this integral is suitable for estimating of microwave magnetic performance of the material.

4. Discussion

The above derivations reveal the validity conditions of Eq. (2). First, the application of the Cauchi theorem implies that the permeability is proportional to $1+f^{-2}$ at $f\to\infty$. As is shown above, this is not true in the case of pronounced skin effect, when the high-frequency asymptote of the permeability is proportional to $1+if^{-1}$ and the left part of Eq. (2) diverges. However, the divergence appears at very high frequencies, much higher than typical ferromagnetic resonance frequencies. The physical reason for the divergence may be that the conductivity of the material is assumed to be constant in Eq. (5) and, therefore, material properties do not disappear at infinite frequency, which is necessary for causal behavior providing the validity of the Cauchi theorem. Therefore, this divergence is due to simplistic model of the magnetic material.

The same can be said on another possible source of divergence of integral in Eq. (2). This is because the dispersion law following from the conventional Landau–Lifshitz–Gilbert equation differs from the Lorentzian dispersion. The Lorentzian dispersion corresponds to the Bloch-Blombergen damping term, which, however, does not keep the magnetic moment constant and is therefore considered as unsuitable for ferromagnetic materials. The application of the Gilbert damping term also produces the permeability proportional to $1+i/f$ at $f\to\infty$ and, therefore, results in the divergence.

In both the cases, the divergence appears at very high frequencies, much higher than typical natural resonance frequencies of magnetic materials and is not observable by microwave measurements. Therefore, it is not observable by microwave measurements and has no impact on conclusions that can be made from Acher's law on either estimating of ultimate microwave performance of magnetic and crystalline structure of the sample.

The only class of the materials where Eq. (2) is found experimentally to be invalid includes hexagonal ferrites. For these, the difference from the above consideration is that in-plane location of the magnetic moment is provided by large out-of-plane anisotropy field, H_θ, rather than by the demagnetization field, as in ferromagnetic flakes. Under assumption of two anisotropy fields, the out-of-plane anisotropy field H_θ and in-plane anisotropy field H_ϕ, Eq. (1) is rewritten as [8]:

$$(\mu_s - 1) f_r^2 = \frac{1}{3} (\gamma' 4\pi M_0)^2 \left(1 + (H_\theta + H_\phi)/(4\pi M_0)\right), \tag{8}$$

In contrast to Eq. (1), the right-hand part of Eq. (8) depends on the anisotropy fields, which are dependent on the method of production and features of technology of the ferrite. This does not allow Eq. (8) to be used as an ultimate constraint for microwave magnetic performance in hexagonal ferrites. However, useful estimates can still be obtained with Eq. (8). It is worth noting that $H_\theta > 4\pi M_0$ in most hexagonal ferrites, hence the last parenthesis may introduce a factor of 3 to 4 into the right part of Eq. (8), thus significantly enhancing the performance of the material.

Another promising approach to overcome Acher's constraint employs exchange resonances. The conventional derivation of Eqs. (1) and (2) assumes uniform magnetization of magnetic particle. However, resonance modes related to non-uniform magnetization may appear in tiny magnetic particles at frequencies higher than the ferromagnetic resonance and contribute to Acher's constraint. The corresponding theory is developed in [9], but the experimental data available do not allow a conclusion to be made on omitting Acher's law.

Therefore, rare evidences for invalidity of Eqs. (1) and (2) are found in the literature. In most cases, Acher's law provides an upper limit for the microwave permeability of magnetic materials and can be used for corresponding estimates. The estimates must be made with care in cases of wide magnetic absorption peak, where Snoek's law is shown to provide adequate estimations instead of Eq. (1).

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