Bounds on the dynamic properties of magnetic materials

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(Received 24 November 1999; revised manuscript received 1 June 2000)

We derive a sum rule on the dynamic magnetic dissipation in magnetic materials. We obtain a bound on the weighted sum of the gyromagnetic losses over the whole spectral range. Bounds are established for a fairly large class of bulk and composite magnetic materials, and shown to agree with experiment. It is also shown that a close approach to this bound can be experimentally attained.

I. INTRODUCTION

Years ago, Snoek established a relationship constraining the product of the initial permeability and resonance frequency of ferrites to a quantity that varied only with their saturation magnetization.[1] With a slight exception,[2] there has been no attempt to establish further bounds on the microwave properties of magnetic materials. The Kramers-Kronig relations are useful and have been recently extended to a finite-frequency range,[3] but do not directly give bounds on the variation of microwave properties with the static magnetic parameters. In contrast, in the framework of effective medium theories,[4–6] efforts to bound the range of parameters allowed for a composite material as a function of the properties of its constituents have been particularly successful for either the magnetic or dielectric susceptibility constant.[7–9] In contrast, fundamental bounds on the properties of electronic networks have been known for decades.[10]

This paper establishes a bound on the high-frequency magnetic losses for a magnetic material that can be stated in terms of its static magnetic properties. The derivation is carried on a saturated ellipsoid, extended in the general framework of effective medium theory to demagnetized materials and finally to composite materials. We show that this bound can be attained for some particular magnetic composite topologies. The validity of the relation was verified by experiment on several materials. Various consequences and physical applications of this relation are then discussed.

II. THEORETICAL RESULTS

Consider an ellipsoid with saturation magnetization $4\pi M_s$ and demagnetizing coefficients N_x, N_y, and N_z in an external field H_k along the z direction large enough to saturate the ellipsoid. The microwave properties of the magnetic material are obtained by the gyromagnetic equation of motion in which magnetic relaxation is described by a Bloch-Bloembergen (BB) damping term.[11] All quantities are expressed in the cgs system. The permeability tensor $\bar{\bar{\mu}}$ relating the microwave induction to the microwave field applied to the ellipsoid at a frequency F is $\bar{\bar{\mu}}=\bar{\bar{1}}+4\pi\bar{\bar{\chi}}$, where $\bar{\bar{\chi}}$ is the magnetic susceptibility tensor. With the $\exp(+j2\pi Ft)$ time convention, the nonzero components can be written as[11]

$$\chi_{xx}(F)=\gamma\!\!\!/ M_s\frac{F_y}{-F^2+2j\dfrac{F}{T}+F_xF_y-\dfrac{1}{T^2}},$$

$$\chi_{xy}=\gamma\!\!\!/ M_s\frac{\dfrac{1}{T}+jF}{-F^2+2j\dfrac{F}{T}+F_xF_y-\dfrac{1}{T^2}}, \tag{1}$$

with $F_x=\gamma\!\!\!/(N_xM_s+H_{\rm int})$, $F_y=\gamma\!\!\!/(N_yM_s+H_{\rm int})$, $H_{\rm int}=H_k-N_zM_s$, and T a characteristic damping time. $\gamma\!\!\!/$ is related to the gyromagnetic ratio γ by $\gamma\!\!\!/=\gamma/2\pi$ and is approximately 3 MHz/Oe for most magnetic materials.

Consider the function $G(F)=F4\pi\chi_{xx}(F)$ of the complex frequency F. Here $G(F)$ is analytical over the lower half complex plane (no poles). Applying the Cauchy theorem to a closed contour consisting of the real axis $]-\infty,+\infty[$ and a semicircle $C_{-\infty}$ in the lower half of the complex frequency plane yields

$$I_x=\int_0^{+\infty}F\mu''_{xx}(F)dF=\frac{\pi}{2}(\gamma\!\!\!/4\pi M_s)^2\left(\frac{N_y}{4\pi}+\frac{H_{\rm int}}{4\pi M_s}\right). \tag{2}$$

A similar relation holds for the permeability along y and the permeability along z is unity. Therefore, defining $I=I_x+I_y+I_z=\int_0^{+\infty}F\,\mathrm{Tr}\{\bar{\bar{\mu}}''(F)\}dF$,

$$I=\frac{\pi}{2}(\gamma\!\!\!/4\pi M_s)^2\left(1-\frac{N_z}{4\pi}+\frac{2H_{\rm int}}{4\pi M_s}\right). \tag{3}$$

This result can be extended to unsaturated materials, such as bulk ferrites, using either of the two approaches that have been proposed to describe their permeability.[12–14] In one approach, with the magnetization along z and a domain configuration isotropic in the xy plane, the permeability along the x and y axes has been shown to be given by $\overline{\mu}$ with $\overline{\mu}^2=(1+4\pi\chi_{xx})^2+(4\pi\chi_{xy})^2$.[13,14] Unlike Refs. 13 and 14, a Bloch-Bloembergen dissipation term is used here. Then it can be shown that Eq. (3) also holds, with $N_z=0$, $H_{\rm int}$ being the anisotropy field.

Consider now a composite material. Homogenization laws are widely used to describe the effective properties of composite materials as a function of the properties of their constituents.[4–9] The predicted properties are very dependent on the shape of the inclusions and on the particular formulation used (such as Bruggeman or Maxwell-Garnett). However, in the quasistatic limit, it has been established that the effective susceptibility $\hat{\chi}$ of a two-component composite with a known volume fraction should lie within known bounds that are independent of the particular formulation and of the shape of the inclusions.[7,8] The bounds are given by the Maxwell-Garnett equation with either constituent as the matrix.[6–9] For a matrix of material a with volume fraction f, if the susceptibility of the constituents a, b is noted χ_a,χ_b, respectively it can be written

$$\hat{\chi}^a = f\chi_a + (1-f)\chi_b - q\,\frac{f(1-f)(\chi_a-\chi_b)^2}{(1/4\pi)+\chi_a(1-qf)+\chi_b qf}, \tag{4}$$

where the shape factor q varies from 0 to 1. Let us suppose that χ_a and χ_b are given by the BB equation, possibly with $4\pi M_s = 0$ if one constituent is nonmagnetic, and that their eigenaxes are parallel. Then, applying the Cauchy theorem on the same contour as for Eq. (2) gives

$$I_x = \int_0^{+\infty} F\hat{\mu}''^{a}_{xx}(F)\,dF = fI_{x,a} + (1-f)I_{x,b}, \tag{5}$$

with $I_{x,a}$ and $I_{x,b}$ defined by Eq. (2). This equation can be generalized to any number of constituents, provided the material can be described as a mixture of composites. This important result (to our view) states that the weighted sum I_x for a composite is simply the volume average of this quantity applied to each constituent. Another important result is obtained for the case of soft materials so that $H_{\mathrm{int}} \ll 4\pi M_s$. In the absence of a strong external field, most microwave materials, such as soft spinel ferrites and soft ferromagnetic films, meet this condition. Taking $I_{\max} = (\pi/2)\langle(\gamma 4\pi M_s)^2\rangle$ where the angular brackets indicate a volume averaging, then from Eqs. (3) and (5) we obtain

$$I_x/I_{\max} \leqslant I/I_{\max} \leqslant 1. \tag{6}$$

This inequality yields a bound on the frequency-dependent properties of the material I_x and I, as a function of a quantity $I_{\max}$ that is dependent on the saturation magnetization only. This relation can be viewed as a generalization of Snoek's law, with a much broader range of validity since it also holds for inhomogeneous materials. If the material is isotropic, $I = 3I_x$ and a more severe bound can be established by Eq. (6): $I_x/I_{\max} < 1/3$. If the material is a composite with only one magnetic constituent, $I_{\max} = f(\pi/2)(\gamma 4\pi M_s)^2$, where f is the volume fraction of the magnetic constituent and $4\pi M_s$ is its saturation magnetization.

III. COMPARISON WITH EXPERIMENTAL RESULTS

The permeabilities of different magnetic materials have been measured, using a conventional coaxial transmission line technique.[15] The partial integral $I_x(F) = \int_0^F F'\mu''_{xx}(F')dF'$ has been calculated using the experimental permeability spectra. The saturation magnetizations of each magnetic constituent were measured using a vibrating sample magnetometer and used to determine $I_{\max}$. Figure 1 shows the spectra of the imaginary permeability and the ratio $I_x(F)/I_{\max}$ for several materials. The data in Fig. 1(a) are for (a, A) a bulk-sintered $Ni_{0.5}Zn_{0.5}Fe_2O_4$ ferrite and (b, B) a dispersion of ferrite powder with the same composition in a binder. The imaginary permeabilities of the two materials are seen to be very different. The sintered material exhibits a large response at low frequencies, while that of the composite material occurs at much higher frequency, but is 100 times smaller. For the sintered ferrite, $I_x(F)/I_{\max}$ asymptotically approaches a value of 1/3, which is the maximum value allowed for an isotropic material. For the composite material, this quantity approaches a smaller, but comparable value of 0.2, which also agrees with Eq. (6). The results in Fig. 1(b) were obtained for laminated insulator ferromagnetic on the edge (LIFE) composites[15] that consist of alternating ferromagnetic and insulating films. The LIFE composites were measured with the microwave H field parallel to the hard axis. One LIFE composite was made of soft, amorphous CoZrPt films that have gyromagnetic resonance loss peaks at ~2 GHz. As expected, for this composite,

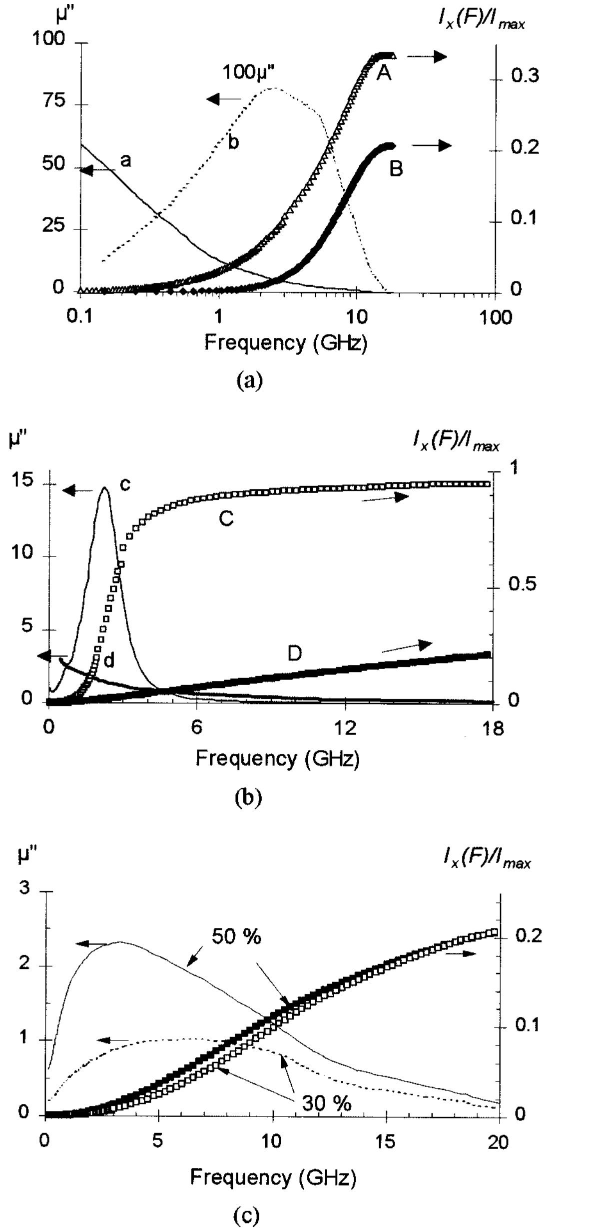

FIG. 1. Spectra of imaginary permeability and normalized $I_x(F)/I_{\max}$: (a) (a, A) sintered $Ni_{0.5}Zn_{0.5}Fe_2O_4$ ferrite and (b, B) dispersion of a $Ni_{0.5}Zn_{0.5}Fe_2O_4$ ferrite powder. (b) Composites made of ferromagnetic laminations (c, C) CoZrPt and (d, D) Fe. (c) Dispersion of Permalloy powder with 30% and 50% volume fraction.

$I_x(F)/I_{max}$ tends toward unity.[2] Another LIFE composite was fabricated with iron thin films. The iron film laminates exhibited a much smaller permeability over a lower and much wider frequency range. Because of the relatively high anisotropy of the iron films, the integral $I_x(F)/I_{max}$ does not reach an asymptote for $F=18\,\text{GHz}$, but it is lower than unity as predicted. Figure 1(c) shows the spectra of two composites made by dispersing different volume fractions of Permalloy powder in an insulating matrix. Although the permeabilities of these composites differ significantly, the ratio $I_x(F)/I_{max}$ approaches the same asymptotic value for both composites and is lower than 1/3, in agreement with Eq. (6).

IV. DISCUSSION

A. Range of validity

The bound on the dynamic responses of magnetic materials we have derived is well supported by the experimental evidence presented. Nevertheless, it is of consequence to examine the basic assumptions that were made to obtain this result. A Bloch-Bloembergen-type damping parameter was used, rather than either a Landau-Lifschitz (LL) or a Landau-Gilbert (LG) type. A common objection to the BB term is that it does not conserve the magnetization when dealing with large excitations. However, to first order, in the low-field linear susceptibility regime considered here, the susceptibilities obtained from either a BB, LL, or LG parameter conserve the magnetization. If an LL or LG parameter were used, the damping far above spin resonance would cause $I_x(F)$ to diverge as $\ln F$. However, since the divergence is slow, the permeability of the soft material of Fig. 1(b) was fitted using a LG damping term, and the quantity $I_x(F)$ was computed from the integration of the LG permeability up to 18 GHz. The result differs by less than 5% from the quantity $I_x(F)$ obtained by integrating the experimental spectrum. This suggests that the choice of the damping term is not a major issue for the evaluation of $I_x(F)$ up to $\sim$18 GHz. Indeed, a common criticism to all these phenomenological models is that while they are generally well suited to describe the permeability near spin resonance, they do not yield a very good estimate of it at much higher frequencies. It is necessary then to introduce a dispersive damping parameter, as shown experimentally.[16] The introduction of a frequency-dependent LL or LG damping term can then lead to convergence of the integral $I_x(F)$.

It has been assumed that the bounds of value allowed for the effective properties of composite materials[7–9] hold at finite frequencies, whereas they have been derived at zero frequency. In fact, it has been shown that these bounds do not hold when characteristic dimensions of inclusion are not negligible compared to the wavelength.[17,18] In particular, it is known that composites that contain conducting nonmagnetic inclusions can exhibit a nonzero imaginary permeability,[19] clearly showing that the sum rules on the dynamic permeability established here are not valid when there is skin effect. The bound for materials with conducting particles is $I(F_{max})/I_{max}<1$, where $I(F_{max})$ is the partial integral up to the frequency F_{max} where the skin effect becomes significant. The experimental results reported in Figs. 1(b) and 1(c) were obtained on composites made with thin ferromagnetic films and small ferromagnetic particles to minimize the contribution of eddy currents to the permeability at frequencies below 20 GHz.

The model presented considers only the gyromagnetic permeability. It is known that the domain wall permeability may also contribute to the overall permeability at lower frequencies[13] and is expected to yield an additional contribution to I_x. However, the imaginary permeability is weighted by the frequency in the integral I_x. The impact of additional contributions to the permeability at frequencies much below 1 GHz is expected to be small compared to the gyromagnetic contribution that extends over a much higher-frequency range. Bulk ferrites are known to exhibit a significant domain wall contribution at low frequencies, but the result in Fig. 1(a) indicates that Eq. (6) is valid for the ferrites investigated in this work.

B. Consequences and applications

While the effective permeability of composite materials is highly dependent on the volume fraction of the inclusions and on their distribution, by using Eq. (5) it has been shown for soft magnetic composites that the integral of the gyromagnetic loss response, weighted by the frequency, is simply the volume average of this quantity for each constituent. It is independent of the topology of the composite, provided that the demagnetizing coefficients N_z of the domains in Eq. (3) are not much affected by the topology. Figure 1(c) provides a good illustration of this sum rule. Bulk ferrite and ferrite powders have a very different domain structure, and Eq. (6) yields a bound I_{max} on the quantities I_x and I that is valid for both bulk and unsaturated soft materials. I_{max} is simply related to the spatial average of the square of the saturation magnetization. Although it is intuitive that there is a relationship between dynamic magnetic loss and saturation magnetization, a quantitative relation that would generalize Snoek's law[1] had not been previously formulated. It was also well known that the dynamic permeability can be significantly altered by the magnetization distribution in bulk materials[12–14] and also by the shape and the arrangement of magnetic particles in a composite,[20] as illustrated by the permeability curves in Fig. 1. This work has shown, however, that despite the wide diversity of the dynamic magnetic responses observed experimentally, there is a quantitative limit on these responses that depends only on the saturation magnetization of the material.

Equations (2), (3), (5), and (6) allow a better understanding of the permeability spectra of magnetic materials. They should be useful in frequency-response engineering of materials,[20] in the development of new permeability measurement techniques, and in frequency-resolved ferromagnetic resonance (FMR) experiments.[21] Equation (6) provides a convenient way to check the high-frequency accuracy of permeability measurements.[22]

In addition, the quantity I_x has a direct physical significance for reflection coefficients of waves incident on magnetic media. Consider the reflection coefficient R of a plane wave of frequency F normally incident on a magnetic layer of thickness e backed by a metallic plane. The incident magnetic field is parallel to the x direction which coincides with one of the material eigenaxes. The permeability μ of the material has the properties listed above. R is expressed by

$$-R=[1-Z\tanh(X)][1+Z\tanh(X)]^{-1}, \quad (7)$$

with $Z=[\mu(F)/\varepsilon]^{1/2}$ and $X=j2\pi[\varepsilon\mu(F)]^{1/2}eF/c$.

Assume that the thickness of the material is much less than the wavelength at least up to a frequency F_m, so that $\tanh(X)\cong X$ and that $|R|\sim 1$ for $F\geqslant F_m$. A proper application of the Cauchy theorem[23] in the complex plane can be used to integrate the attenuation $\Gamma=-20\log|R|$, and Eq. (6) is then used to bound this sum:

$$\int_0^{F_m}\Gamma(F)dF\leqslant\frac{1}{d}\frac{40\pi^2}{\ln(10)c}e\langle(\gamma 4\pi M_s)^2\rangle, \quad (8)$$

where $d=3$ if the material is isotropic, $d=2$ if $\mu_{xx}=\mu_{yy}$, and $d=1$ otherwise. The experimental results in Fig. 1(b) for a 1.4-mm-thick CoZrPt-based LIFE composite can be used to verify Eq. (8). The left-hand side is found by calculating the attenuation using the experimental value of the permeability and permittivity over the available spectral range. It is found to be 3% lower than the value of the right-hand side bound computed from the saturation magnetization of the ferromagnetic film, in agreement with Eq. (8).

V. CONCLUSION

In summary, general properties have been established for the integral of the frequency-weighted gyromagnetic permeability spectrum of soft magnetic materials. The maximum values of the integral depend only on the saturation magnetization and gyromagnetic constant. The convergence of the weighted sum is not critically dependent on the underlying assumptions. The theoretical bounds on the weighted sum agree with experiments on a large variety of magnetic and composite materials, and appear to have a substantial range of physical application.

ACKNOWLEDGMENTS

We are grateful to Professor R Walser and W. Win for fruitful comments on the manuscript.

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