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A generalization of Snoek's law to ferromagnetic films and composites

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Abstract:

The integral $\int_0^{\infty} \mu''(f) \cdot f \cdot df$ of the imaginary permeability times frequency associated to magnetic materials has very remarkable properties. A generalization of Snoek's law is that this quantity is bounded by the square of saturation magnetization times a constant. While previous results have been obtained in the case of non conductive materials, this work is a generalization to ferromagnetic materials and ferromagnetic-based composites with significant skin effect. The effect of truncating the summation to finite upper frequencies is investigated, and estimates associated to the finite summation are given. It is established that in practice, the integral does not depend of the damping model under consideration. Numerical experiments are performed in the exactly solvable case of ferromagnetic thin films with uniform magnetization. The validity of our derivations is confirmed with a good precision. Microwave permeability measurements on soft amorphous films are reported. The relation between $\int_0^F \mu''(f) \cdot f \cdot df$ and $4\pi M_s$ is verified experimentally. The integral is useful to quantify the magnitude of the magnetization normal to the microwave measurement field. It provides an unprecedented "see-through" ability to investigate the orientation of the magnetization in materials with complex magnetization configuration. It may also be used to demonstrate the accuracy of microwave measurements systems. For some applications, such as electromagnetic compatibility or radar absorbing materials, the relations established here provide useful indications for the design of efficient materials, and simple figure of merits to compare the properties measured on different materials.

I. INTRODUCTION

The microwave permeability μ of magnetic materials is a quantity of interest on both applied and fundamental points of views. High frequency inductors,¹ magnetic recording write heads, broadband skin antenna,² microwave filters,^{3,4} noise suppressors⁵ and Radar Absorbing materials^{6,7} require high broadband permeability levels at high frequencies. However, it has been known since Snoek⁸ that there are tradeoffs between high permeability levels and high frequency operation. In a bulk polycrystalline material, Snoek's law writes

$$(\mu'_{F=0} - 1)F_0 = \frac{2}{3} \varphi 4\pi M_s . \quad (1)$$

where $\mu'_{F=0}$ is the low frequency permeability, F_0 the resonance frequency, $4\pi M_s$ the saturation magnetization, and $\varphi = \gamma / 2\pi \approx 3\text{MHz}/\text{Oe}$ the gyromagnetic factor. However, the tradeoff depends on the shape of the magnetic domains or particles.^{9,10} For soft thin films with uniform uniaxial in-plane anisotropy, it writes

$$(\mu'_{F=0} - 1)F_0^2 = (\gamma 4\pi M_s)^2. \quad (2)$$

These relations are easily established from the gyromagnetic permeability of a saturated ellipsoid. However, they no longer hold if the magnetic material is heterogeneous, or in the case of composite materials. In addition, they provide no clue on the linewidth of the permeability.

Recently,^{10,11} another expression of the tradeoff between permeability levels and frequency has been found. It can be written¹²

$$\int_0^{\infty} \mu''(f) \cdot f \cdot df = k_A \frac{\pi}{2} (\gamma 4\pi M_s)^2, \quad (3)$$

where k_A is a numerical constant that has a simple expression. For uniform soft thin films, $k_A=1$. For bulk sintered ferrites, $k_A=1/3$. For isotropic composite materials with a volume fraction τ of magnetic filler, $k_A \leq \tau/3$. In any cases, $k_A \leq 1$.

The ratio k_A is easily determined from experimental data. It can be used to quantify the quality of thin films for microwave applications¹² and to guide their design.¹³ Eq. (3) has also been found useful as an indication for the conception of microwave absorbers.¹⁴ In this case μ'' is a quantity of direct interest.

For materials with a permeability that coincides with the gyromagnetic permeability of a saturated ellipsoid, the Snoek's law under the discrete form (Eqs. (1), (2)) may be a more straightforward expression of the balance between high permeability levels and high frequency operation than Eq. (3).¹⁵ But in many cases, Snoek's law under the discrete form does not apply, while Eq. (3) still holds. As a consequence it can be considered as a generalization of Snoek's law. Hexagonal ferrites used in microwave applications are not soft materials in the sense that their in-plane magnetization can be comparable or larger than the saturation magnetization. So Eq. (3) does not apply to hexagonal ferrites, but a more general integral relation has been proposed and verified experimentally.¹⁶

The purpose of this paper is to provide significant extensions for Eq. (3). It had been established for non-conducting materials materials. Since conducting ferromagnetic materials are more and more used instead of ferrites, it is important to investigate the case where skin effect due to finite conductivity has a significant impact on the permeability. An important result obtained in this paper is that the integral quantity is hardly affected by moderate skin effect, and slightly decreases when skin effect becomes larger. Another limitation of Eq. (3) was about the model used to describe the damping. It had been established assuming a Bloch-Bloembergen damping term. This paper establishes that it also holds for a Gilbert damping term.

When the integral in the left member of Eq. (3) is determined from experimental permeability measurements, it has to be truncated to a finite upper frequency within the measurement range. This paper provides estimates of upper integration frequencies that can be used with negligible error. It also provides simple estimates for the truncation error. These estimates are useful when permeability measurements are performed up to an upper frequency that is not significantly larger than the resonance frequency.

The paper is organized as follows. In part II, the mathematical approach is outlined and general results are presented. Analytical details are given in the appendix. In part III, the results are formulated for different particular cases, namely thin films, multilayers, and composite materials. In part IV, it is checked that the approximations are relevant by performing numerical experiments on an exactly solvable case. In part V, our theoretical findings are confronted to experimental values of permeabilities measured on thin films. The potential applications of our findings are discussed in part VI.

II. THEORETICAL APPROACH

The derivation of the estimates for $\int_0^F \mu''(f).f.df$ for conducting magnetic inclusions and composites is outlined in this section. Detailed calculations are presented in the appendix.

A. Permeability of an ellipsoid with uniform magnetization

The susceptibility tensor of an ellipsoid with a uniform magnetization is well known.¹⁷ Several models have been proposed for the damping term. The Landau-Gilbert expression figuring the adimensional damping parameter α is one of the most popular. The expression of the permeability of a uniformly magnetized ellipsoid writes:

$$\mu_G = 1 + \frac{F_M (F_y + j\alpha f)}{F_0^2 + j\alpha (F_x + F_y) f - f^2} \quad (4)$$

with

$$F_0 = \sqrt{F_x \cdot F_y} \quad (5a)$$

$$F_M = \gamma 4\pi M_s; F_x = \gamma (N_x M_s + H_{int}); F_y = \gamma (N_y M_s + H_{int}) \quad (5b)$$

$$H_{int} = H_k - N_s M_s; \alpha \ll 1 \quad (5c)$$

$4\pi M_s$ is the saturation magnetization of the material, and N_x, N_y, N_z are the demagnetizing coefficients of the ellipsoid, and H_k is the external field (or anisotropy field) that saturates the ellipsoid along $+z$. $\gamma = \gamma / 2\pi$ is the gyromagnetic ratio, close to 3 MHz/Oe. F_0 is called the resonance frequency. The orientation conventions are similar to that in ref [11], the permeability given by (5) being in the x direction. In the case of soft magnetic materials that are generally considered in microwaves, and for null or moderate external fields, the different contributions to the H_k field will be small compared to the saturation magnetization. The internal field H_{int} has to be positive for the magnetization to be stable in the $+z$ direction, as a consequence H_{int} is small also.

B. Influence of skin effect on the permeability

The permeability of conductive inclusions depends on its conductivity, shape and dimensions, and of course of the permeability of the constitutive materials. It has been derived by many independent works, and for many shapes. The conductivity of the inclusion can be written^{6,18,19,20,21}

$$\mu = \mu_G \cdot A(ka), \quad (6)$$

where μ_G is the intrinsic permeability of the material, a is its characteristic size, and k is the wavevector inside the inclusion. The function $A(ka)$ acts as a renormalisation quantity. It depends implicitly of the permeability μ_G and of the conductivity σ of the material. The expression of $A(ka)$ for different inclusion shapes are listed on Fig. 1. Though they may appear dissimilar at first sight, their first order development in ka has the same form

$$A(ka) = 1 + \frac{(k.a)^2}{p} + \text{higher order terms} \quad (7)$$

where p is a number that depends on the inclusion shape. It is seen on Fig. 1 that p is larger for a sphere ($p=10$) than for a plate ($p=3$). This supports the intuitive evidence that skin effect is less pronounced on a sphere, where the electromagnetic field can penetrate from all sides, than on a plate, where the electromagnetic field can penetrate only from the top and bottom.

The cases for cylinders are in-between. This suggests that Eq. (7) can be extended to many regular shapes, and is fairly general.

C. Derivation of the integral bound

The permeability $\mu(f)$ is analytical in the lower complex half plane. This is due to causality principle. As a consequence, the Cauchy theorem can be applied to the quantity $f\mu(f)$: the circulation of this function on a closed contour in the lower half plane is zero. The integration contour is sketched on Fig. 2. It goes from the frequencies $-F$ to $+F$ on the real axis, and then follows a half circle $F \cdot \exp(j\theta)$, θ ranging from 0 to π .

$$\int_{-F}^F \mu(f) \cdot f \cdot df + \int_{C^-} \mu(f) \cdot f \cdot df = 0. \quad (8)$$

The first term can be transformed using the general properties $\mu(-\bar{f}) = \bar{\mu}(f)$, where the bar corresponds to the conjugate. The second term can be transformed into an integral on the angular coordinate θ on the semicircle C^- . One finds

$$\int_0^F \mu''(f) \cdot f \cdot df = \frac{1}{2} \int_0^\pi \mu(F e^{j\theta}) \cdot (F e^{j\theta})^2 \cdot d\theta. \quad (9)$$

The left side of the above equation is the quantity of interest. The right side is a quantity that is relatively easy to calculate provided F is large enough, but not too large. This is because good approximations on μ are available at high frequencies F . Detailed calculations are shown in the appendix. One finds

$$\int_0^F \mu''(f) \cdot f \cdot df \approx \frac{\pi}{2} N_y F_M^2 \cdot [1 - t - s \pm e], \quad (10)$$

where t and s are small corrective terms, with positive sign. t corresponds to the finite truncation; s is related to skin effect. The term e is the error induced by the measurement errors and uncertainties $\Delta\mu$ on μ when the integration is performed.

$$t = \frac{2}{\pi} \alpha (2N_x + N_y) \frac{F_M}{F} \quad (11)$$

$$s = \frac{4\mu_0 a^2 \sigma}{p} N_y \frac{F_M^2}{F} \quad (12)$$

$$|e| \leq \frac{\Delta\mu}{\pi N_y} \left(\frac{F}{F_M} \right)^2. \quad (13)$$

The validity of Eqs. (10) to (13) requires that the upper summation frequencies verifies the conditions in the first 3 lines of Table I. These are very reasonable conditions, as will be evidenced in sections IV and V. Table I also indicates the conditions for the corrective terms t , s or e to be negligible. In case they are small but not negligible, they can be determined from the analytical expression above.

D. Generalization to magnetic materials with complex magnetization state and composites

Let us consider magnetic matter made of a collection of magnetic domains, with different shapes. Each magnetic domain is described as a saturated ellipsoid, with possibly different demagnetizing coefficients and internal fields. Let us also allow some non magnetic matter. The permeability of this complex matter can be determined through an appropriate

homogenization law. In practice, the difficulty is that the homogenization law depends not only on the permeability of each domain, but also on the details of their geometries and arrangement. If only the permeabilities of the constituents are known, but not the exact topology, it is not possible to know precisely the permeability μ_{eff} of the homogenized medium, but it is nevertheless possible to know some bounds on the complex values of μ_{eff} . These bounds are called Hashin-Shtrikman and Milton-Bergman bounds.^{22,23,24} If the frequency F is large enough, the permeability of each constituent shall be rather close to unity, and in this case, the bounds converge to a single value of μ_{eff} that is independent on the geometry:

$$\mu_{eff} \approx \sum_i \tau_i \cdot \mu_i = \langle \mu_i \rangle, \quad (14)$$

where τ_i designates the volume fraction of each domain labelled i , and $\langle \rangle$ corresponds to a volume average. Using this result in Eq. (10), one finds:

$$\int_0^F \mu_{eff}''(f) \cdot f \cdot df = \sum_i \tau_i \frac{1}{2} \int_0^\pi \mu_i (Fe^{j\theta}) \cdot (Fe^{j\theta})^2 \cdot d\theta = \sum_i \tau_i \int_0^F \mu_i''(f) \cdot f \cdot df. \quad (15)$$

This result is similar to eq. (20.6) in ref [25]. This is a very important result in the homogenization process the integral quantity averages linearly according to the volume fraction of the constituents.

E. Isotropic composites made of ferromagnetic loads in a dielectric matrix

In an isotropic material, one third of the magnetization is along the microwave exciting field, and therefore has a unit permeability which does not contribute to the integral ($\mu_z=1$ is indeed a companion Equation of Eq. (4)). The volume fraction of magnetic particles is noted τ , and the average of the demagnetizing coefficients of the domains along their magnetization is noted $\langle N_{//} \rangle$. $\langle N_{//} \rangle$ is expected to be small compared to unity, since domains elongated along the magnetization are pose stable in soft materials. since the magnetization tends to align the elongated in soft materials. The demagnetizing coefficient in the elongated direction of an ellipsoid is less than 1/3, and is close to zero if the aspect ratio is large. Therefore it will be treated as a first order correction. We obtain from Eqs. (10), (15):

$$\int_0^F \mu''(f) \cdot f \cdot df \approx \frac{\pi}{6} \tau (\neq 4\pi M_s)^2 (1 - \langle N_{//} \rangle - \langle t \rangle - \langle s \rangle \pm e). \quad (16)$$

One finds

$$\langle t \rangle = \frac{2}{\pi} \langle \alpha \rangle (1 + \eta) \frac{F_M}{F} \quad (17)$$

where $\eta = 2 \langle N_x \cdot N_x \rangle$ is the average of the product of the demagnetizing coefficients normal to the magnetization. It is comprised between 0 and 1/2.

It is important to note that s does not depend on the internal fields. This suggests that for a multi-domain particle with half radius a and shape parameter p , Eq. (12) is still the appropriate expression for s , provided an averaging on N_y is performed. In practical cases, there may be a significant dispersion in radius a within the magnetic filler, while its conductivity σ remains constant. It follows

$$\langle s \rangle = \frac{4\mu_0 \sigma \langle a^2 \rangle}{p} (1 - \eta) \frac{(F_M)^2}{F}. \quad (18)$$

The absolute error due to measurement uncertainty is bounded by $\frac{\Delta\mu}{2}F^2$, and a majoration of the relative error e is :

$$|e| \leq \frac{3}{\pi} \frac{\Delta\mu}{\tau} \left(\frac{F}{F_M} \right)^2. \quad (19)$$

III. RESULTS

The above equations associated to validity conditions summarized in the three upper lines of Table I are very general results. However, it is useful to write them for a few cases of particular interest, in order to provide ready-to-use equations.

A. Application to uniform soft thin films

In the case of a uniaxial thin film magnetized along the y direction with in-plane orientation, the demagnetizing coefficient N_y is unity. The hard axis permeability has the following properties:

$$\int_0^F \mu''(f) \cdot f \cdot df \approx \frac{\pi}{2} (\gamma 4\pi M_s)^2 [1 - t - s \pm e], \quad (20)$$

with

$$t = \frac{2}{\pi} \alpha \frac{\gamma 4\pi M_s}{F}. \quad (21)$$

$$s = \frac{4\mu_0 a^2 \sigma}{p} \cdot \frac{(\gamma 4\pi M_s)^2}{F}. \quad (22)$$

$$|e| \leq \frac{\Delta\mu}{\pi} \left(\frac{F}{\gamma 4\pi M_s} \right)^2. \quad (23)$$

where $2a$ is the thickness of the film. It follows in a straightforward manner that :

$$\int_0^F \mu''(f) \cdot f \cdot df \leq \frac{\pi}{2} (\gamma 4\pi M_s)^2. \quad (24)$$

in the case measurements errors are neglected. This generalizes previous results^{10,11} to the case where significant skin effect is present, provided the summation is performed up to a reasonably high frequency. This demonstrates that skin effect cannot increase the factor of merit¹² k_A defined in Eq. (3). Since the contribution of skin effect is a first order correction, it suggests that some skin effect can be tolerated without significant degradation of k_A . This confirms that k_A is a good figure of merit for the microwave properties of thin films. The closer the value of k_A to unity, the better the magnetization of the material is used to obtain microwave properties. An alternate quantity may be useful in order to exploit experimental permeability spectra for thin films.

$$M_\mu(F) = \frac{1}{\gamma} \sqrt{\frac{2}{\pi} \int_0^F \mu''(f) \cdot f \cdot df} \quad (25)$$

The quantity $M_\mu(F)$ may be described as the “efficient dynamic magnetization” participating to the permeability up to frequency F , for the direction of the permeability measurement. For a “perfect film” with $k_A=1$, $M_\mu(F)=4\pi M_s$. In a more general case :

$$M_{\mu}(F) = \frac{1}{\gamma} \sqrt{\frac{2}{\pi} \int_0^F \mu''(f) \cdot f \cdot df} \leq 4\pi M_s \quad (26)$$

It is also possible to find an estimate of the saturation magnetization from the integral of the imaginary permeability

$$4\pi M_s = M_{\mu}(F) \left(1 + \frac{t+s}{2} \pm \frac{e}{2} \right) \quad (27)$$

where s , t and e can be easily computed from Eqs. (21)-(23).

B. Application to soft films with magnetization dispersion and multilayers

Let us consider a multilayer made of thin films with in-plane magnetization, but with possible non uniformities along its thickness. This non uniformity may arise from difference of anisotropy between the layers,²⁶ from antiferromagnetic coupling,²⁷ or exchange coupling, provided these fields are much smaller than the saturation magnetization. The non uniformity may also arise from unwanted phenomena²⁸, or interfacial anisotropies. Let us also allow some non uniformity within the film plane, provided the demagnetization coefficient normal to the film plane remains close to unity. The angle between a reference direction in the film plane and the magnetization is noted ϕ . Then, neglecting measurement errors, the in-plane permeability $\mu_{\phi=0}$ along this reference direction has the following properties:

$$\int_0^F \mu_{\phi=0}''(f) \cdot f \cdot df = \frac{\pi}{2} \left\langle \sin^2 \phi (\gamma 4\pi M_s)^2 \right\rangle [1 - t - s]. \quad (28)$$

It follows that the integral of the imaginary permeability can be a very useful tool to have insights on the orientation of the magnetization within multilayers.

In the case $4\pi M_s$ is uniform within the film but the orientation fluctuates, it is possible to get some information on the orientation from permeability measurements along the $\phi=0$ and $\phi=\pi/2$ directions:

$$\frac{\langle \sin^2 \phi \rangle}{\langle \cos^2 \phi \rangle} = \frac{\int_0^F \mu_{\phi=0}''(f) \cdot f \cdot df}{\int_0^F \mu_{\phi=\pi/2}''(f) \cdot f \cdot df}. \quad (29)$$

Though the permeability spectra along different directions are expected to be much influenced by the detailed topology of the magnetization dispersion, the ratio of integrals provides a robust evaluation of the angular dispersion. This quantity has already been used in order to assess the effect of field annealing on the orientation of the magnetization.²⁹ The result presented here extends the validity of the method to thicker films.

Multilayers have also been designed to provide both magnetic softness and high saturation magnetization, alternating materials with different saturation magnetization but with the same uniaxial in-plane orientation.²⁶ Then

$$\int_0^F \mu_{\phi=0}''(f) \cdot f \cdot df = \frac{\pi}{2} \sum_n \tau_n (\gamma 4\pi M_{s,n})^2 [1 - \langle t \rangle - \langle s \rangle \pm e]. \quad (30)$$

where τ_n is the volume fraction of material with saturation magnetization $4\pi M_{s,n}$. The corrective terms can be easily expressed from Eqs. (11)-(13), or neglected in the case the summation is performed up to sufficiently high frequency.

C. Application to isotropic composite made of a magnetic load in a dielectric matrix

Microwave composites made of magnetic spherical powder dispersed in a dielectric matrix are widely used as microwave materials^{30,31,32} and magnetic absorber.^{6,7,14} In most cases, the radius of the particles is significant compared to skin depth. Eq. (16) establishes that

$$\int_0^F \mu''(f) \cdot f \cdot df \leq \frac{\pi}{6} \tau (\gamma 4\pi M_s)^2 . \quad (31)$$

This result had already been obtained theoretically for composites made of insulating materials such as ferrite powders, and checked experimentally.¹¹ Present work further establishes that this result is remains largely unaffected by finite frequency summation, and by skin effect.

Eq. (18) (using $p=10$ for a sphere) provides useful guidelines to choose appropriate granulometry $\langle a^2 \rangle$ of the particles in order to obtain small or negligible loss of dynamic permeability in the range of interest. The quantity

$$k_{A,3D} = \int_0^F \mu''(f) \cdot f \cdot df / \left[\frac{\pi}{6} \tau (\gamma 4\pi M_s)^2 \right] \quad (32)$$

is an adimensional figure of merit for isotropic composites. The closer to unity, the better is the material. Alternatively, it may be preferred to use

$$M_{\mu,3D}(F) = \frac{1}{\gamma} \sqrt{\frac{6}{\pi \cdot \tau} \int_0^F \mu''(f) \cdot f \cdot df} . \quad (33)$$

The quantity $M_{\mu,3D}(F)$ is expressed in Oe. It may be described as the “efficient dynamic magnetization” participating to the permeability up to F .

It is also possible to find an estimate of the saturation magnetization from the integral of the imaginary permeability

$$4\pi M_s = M_{\mu,3D}(F) \left(1 + \frac{N_{//} + t + s}{2} \pm \frac{e}{2} \right) \quad (34)$$

IV. NUMERICAL VALIDATION

The aim of this numerical validation is to provide good confidence that the estimates of the corrective terms $-t$, $-s$, are correct. It should also confirm that the terms $+S'$ and $+g$ that in Eq. (A17) can be neglected. It will be performed on conducting thin magnetic films with uniform in-plane magnetization. This case is numerically simple, and also quite representative of typical measurements on soft ferromagnetic films. The numerical experiments will be carried with a precision that can not be attained in real experiments.

The thin film is described by the following parameters: $4\pi M_s=10$ kG, $H_k=16$ Oe, $\gamma=3$ GHz/kOe, $\alpha=2\%$, $\rho=130$ μ Ohm.cm, $N_y=1$. These are common values for thin soft ferromagnetic films. The resonance frequency is $F_0=1.2$ GHz. The calculated imaginary permeability spectrum is presented on Fig. 3 for thicknesses $2a$ ranging from 0.1 μ m up to 2 μ m. For the smallest thickness, μ'' is not affected by skin effect, but at 1 and 2 μ m, μ'' is strongly affected by skin effect.

Table II provides relevant lower and upper frequency bounds on the upper integration frequency in the case of the 2 μ m thick film. It appears that $F=6$ GHz is in the adequate range.

The values of $\int_0^F \mu''(f) \cdot f \cdot df$ obtained both numerically and analytically are displayed. The corrective terms are evaluated, both numerically and analytically. It appears that the corrections g and s' are extremely small. They will be neglected in the following. The effect

of the truncation $-t$ is to underestimate the integral by about 6%, and our analytical estimate agrees with the numerical determination. The skin effect correction $-s$ is -19% according to our analytical estimates, which is close to the -23% determined in our numerical experiment. In conclusion, the value of the integral predicted using our model is less than 5% away from the experimental value, which comforts our approach.

Another convenient representation associated to the integral of the imaginary permeability is the “Efficient dynamic magnetization” $M_\mu(F)$ defined by Eq. (25). This quantity is represented on Fig. 4. Numerical results are obtained by numerical integration of the permeability calculated using Eq. (4). Analytical results are obtained from the saturation magnetization using Eq. (27) and the values of s , t and e computed from Eqs. (21)-(23). Analytical and numerical results are in excellent agreement, which validates our results. For the 0.1 μm thick layer, skin effect is negligible, and the graph illustrates that truncation effects decrease when F is increased. However, the adverse effect of summing to high frequencies is evidenced by the error bars that increase with frequency. On most permeability measurement systems for thin films, errors decrease when the thickness of the material increases, because there is more magnetic material in the cell. To account for that effect, the typical error $\Delta\mu$ has been decreased from 10 to 2 when the thickness increases from $2a=0.1 \mu\text{m}$ to $2a=1 \mu\text{m}$, and then further to $\Delta\mu=1$ when $2a=2 \mu\text{m}$. On the thicker films, it is remarkable to see that the integral provides the value of the saturation magnetization within 10% if $F>3.5 \text{ GHz}$ for the 1 μm thick film, and if $F>8 \text{ GHz}$ for the 2 μm thick film. Behind the profound changes on the magnetic losses due to skin effect that are evidenced on fig. 3, it appears that the integral quantity $\int \mu''(f).f.df$ is nearly an invariant.

V. EXPERIMENTAL VALIDATION

We have sputter-deposited amorphous CoZr thin films onto continuously transported 12 μm polyethylene terephthalate substrate. The base pressure inside the chamber before deposition was better than 1.10^{-6} mbar. During the process, the Ar pressure was fixed at 5.0^{-3} mbar. The residual magnetron field induces a uniaxial anisotropy parallel to the transportation direction. Four samples with various thicknesses, 0.3, 1.3, 1.7 and 2.1 μm , were fabricated. The layer thickness was tuned reducing the chilled roll speed and keeping the DC generator power constant. The saturation magnetization $4\pi M_s$ has been measured using a Vibrating Sample Magnetometer, and was found to be $11.3 \text{ kG} \pm 0.5 \text{ kG}$. The permeability has been measured using a thin film permeameter described elsewhere.³³ The typical error $\Delta\mu$ has been estimated around 20 for the thinnest film, and to 2 for the thicker ones.

The imaginary permeability measured on the 4 films is represented on Fig. 5. The thinnest film has a highly resonant permeability, and exhibits a secondary peak at higher frequency. This peak could be attributed to some inhomogeneity and the excitation of a higher frequency mode.²⁸ The thicker film exhibits a permeability with a very damped behaviour, and imaginary permeability levels down by a factor up to 4. The “Efficient dynamic

magnetization” $M_\mu(F) = \frac{1}{\pi} \sqrt{\frac{2}{\pi} \int_0^F \mu''(f).f.df}$ associated to these measurements is represented

on Fig. 6. As expected, this quantity is close to the saturation magnetization at high frequency. A better estimates of the saturation magnetization can be obtained using Eq. (27) with the corrections s and t computed from Eqs (21)-(22). These refined estimates are also shown on the graph, with their associated error bars. It can be seen that all the ranges of the estimates are comprised within the experimental error of the measured $4\pi M_s$. Larger measurement

uncertainty for the thin film permeability evidenced on Fig. 5 is responsible for a larger uncertainty on the integral quantity. It proves that the integral relation on thicker films may be very useful to obtain more precise experimental data. It is remarkable that despite the strong difference in the 4 spectra presented on Fig. 5, all the estimates derived using Eq. (27) coincides within the experimental errors.

VI. DISCUSSION

The fact that the integral depends on much fewer magnetic parameters than μ'' is appealing for the study of unsaturated materials. In particular, it allows the retrieval of information in composites and multilayer materials.

In the case of multilayers, Eq. (28) shows that the integral provides indication on the distribution of magnetization within the sample thickness. The microwave field is indeed a probe of the magnetization normal to it, and the integral $\int_0^F \mu''(f).f.df$ is an appropriate measure to quantify it. The corrections associated to integration up to finite frequencies are easily determined.

One may wonder whether it would not be easier first to determine the intrinsic permeability from measured permeabilities with skin effect, and after to determine the integral of the imaginary intrinsic permeability. Though this procedure is possible, it should be underlined that in the case the magnetic particles are made of different layers and/or domains with different permeabilities, this procedure is no longer valid. In contrast, the estimate of s for the skin effect correction on the integral is independent on the detailed magnetic parameters. It depends only of the $a^2\sigma$ product for the particle, and as a consequence it is a much more robust parameter. In the case of composites made of ferromagnetic powders, there is often a significant distribution in size of the particles. As a consequence the intrinsic permeability determined by the inversion of Eq. (1) taking an averaged value of a may have a limited validity, while the integral quantities can be exploited. In this case the averaged value of the corrective term s is directly related to $\langle a^2 \rangle$, as expressed by Eq. (18).

The relation $\sqrt{\frac{2}{\pi} \int_0^F \mu''(f).f.df} \approx \gamma 4\pi M_s$ may be useful to assess or to demonstrate the

measurement precision of thin film permeameters. In most cases, the uncertainty on the right member is essentially due to the gyromagnetic ratio γ , which is generally considered to be comprised between 2.8 and 3 GHz/kOe, and to some extent to the measurement precision of the saturation magnetization.

In some applications, $f.\mu''(f)$ is a quantity of direct interest. This is the case when magnetic losses are wanted for microwave attenuation, either for microwave filtering, electromagnetic compatibility or for Radar Absorbing Materials. The first order approximation of the reflection or transmission losses are in $f.\mu''(f)$. It has been shown that in the thin absorber limit, the performance of magnetic absorber is bounded by the integral.¹¹ As a consequence, it is an important result that moderate skin effect keeps the integral losses unaffected, though their frequency distribution is much affected. For a somewhat larger skin effect, the correction factor $-s$ may become significant. This leads to a decrease of the integrated losses.

VII. CONCLUSION

The integral $\int_0^F \mu''(f) \cdot f \cdot df$ is closely related to the saturation magnetization and the distribution of magnetization in soft magnetic materials and composites. While some properties of this quantity integrated up to infinite frequencies had already been outlined, this work provides practical results to deal with summation performed up to finite frequencies. It also extends previous results to the case where skin effect significantly alters the microwave response frequency. It may be viewed as a generalization of Snoek's law, since it expresses the tradeoffs between high permeability levels and high operation frequency.

The relations proposed in this work are useful guidelines for the conception of microwave materials. The integral quantity can be easily determined experimentally on many materials. Simple figure of merits deduced from this integral may be used to compare microwave materials.

It has been shown that this integral could also provide very straightforward information concerning the orientation and magnitude of the magnetization in thin films and multilayers. The microwave field acts as a "see through" probe of the magnetization transverse to the excitation. The integral on the imaginary permeability turn it into quantitative information.

VIII. APPENDIX

The dependence of the fields with time is assumed to be $\exp(+j\omega t)$, which is consistent with permeabilities that take the form $\mu' - j\mu''$, $\mu' > 0$. According to Eq. (9), a central issue is to estimate the quantity

$$\frac{1}{2} \int_0^\pi \mu(F e^{j\theta}) \cdot (F e^{j\theta})^2 \cdot d\theta$$

for a frequency F much larger than the resonance frequency F_0 . It is convenient to introduce the reduced frequency

$$\nu = \frac{F e^{j\theta}}{F_0} \quad (\text{A1})$$

Then

$$\frac{1}{2} \int_0^\pi \mu(F e^{j\theta}) \cdot (F e^{j\theta})^2 \cdot d\theta = \frac{1}{2} 4\pi\chi_0 F_0^2 \int_0^\pi \frac{\mu(\nu)}{4\pi\chi_0} \cdot \nu^2 \cdot d\theta \quad (\text{A2})$$

It is convenient to express the permeability given by Eq. (4) as a function of the reduced frequency ν :

$$\mu_G = 1 + \frac{4\pi\chi_0}{1 - \nu^2 + 2j\beta\nu} (1 + j\alpha'\nu), \quad (\text{A3})$$

where $\nu = f/F_0$ is a normalised frequency, $4\pi\chi_0$ is the initial susceptibility,

$$4\pi\chi_0 = \frac{F_M}{F_x} = \frac{F_M \cdot F_y}{F_0^2}; \quad (\text{A4a})$$

$$2\beta = \alpha \frac{F_x + F_y}{F_0}; \quad \alpha' = \alpha \frac{F_0}{F_y}. \quad (\text{A4b})$$

This holds provided the internal fields are small

$$H_k, H_{\text{int}} \ll 4\pi M_s \quad (\text{A4c})$$

It should be noted that when the Bloch-Blombergen damping term is used instead of the Gilbert damping term, the expression of the permeability has the same form as Eq. (A3), with $\alpha'=0$ and $\beta=1/T$, where T is the characteristic damping time. The development in $1/\nu$ of the susceptibility writes :

$$\frac{4\pi\chi_G(\nu)}{4\pi\chi_0} = \frac{-1}{\nu^2} \left[1 - \frac{2j\beta}{\nu} - \frac{1}{\nu^2} \right]^{-1} (1 + j\alpha'\nu)$$

$$\approx \frac{-1}{\nu^2} \left(1 + j\alpha'\nu + \frac{2j\beta'}{\nu} \right) \quad (\text{A5})$$

with

$$2\beta' = \alpha \frac{2F_x + F_y}{F_0}. \quad (\text{A6})$$

The terms in $\alpha'.\beta$ have been neglected in the above expression because the damping parameter is small and terms in α^2 are negligible.

It is then necessary to obtain an appropriate development of the factor $A(ka)$ that accounts for the skin effect. The wavevector inside the ferromagnetic inclusion writes :

$$k = \sqrt{\varepsilon.\mu\omega/c}, \quad (\text{A7})$$

where $\omega=2\pi f$ is the pulsation corresponding to the frequency f , c the celerity of light, and ε the permittivity of the inclusion

$$\varepsilon = \frac{-j\sigma}{\omega.\varepsilon_0}, \quad (\text{A8})$$

where ε_0 is the dielectric constant of void and σ the conductivity. When skin effect is present but not overwhelming, it is possible to use the low order development of $A(ka)$ according to Eq. (7). In the case the upper integration frequency F is significantly larger than the gyromagnetic resonance frequency but not too large so that

$$F_0 \ll F \ll \frac{1}{2\pi\mu_0 a^2 \sigma}, \quad (\text{A9})$$

then

$$A(\nu) \approx 1 - jb.\nu.(1 + 4\pi\chi_B(\nu)), \quad (\text{A10})$$

with

$$b = \frac{2\pi\mu_0 F_0 a^2 \sigma}{p}. \quad (\text{A11})$$

The set of assumption on $|\nu| = F/F_0$ writes

$$1 \ll |\nu| \ll 1/(b.p), \quad (\text{A12})$$

The permeability in the presence of skin effect can be written

$$\mu(\nu) \approx 1 + 4\pi\chi(\nu).[1 - 2jb.\nu - jb.\nu.4\pi\chi_B(\nu)] - jb.\nu. \quad (\text{A13})$$

Taking into account that the damping parameter b is much less than unity, and keeping only the most significant terms leads to

$$\mu(\nu) \approx 1 - \frac{4\pi\chi_0}{\nu^2} \left[1 + 4b\beta' + jb \left(\frac{4\pi\chi_0}{\nu} - 2\nu \right) + j2 \frac{\beta'}{\nu} + j\alpha'\nu \right] - jb.\nu. \quad (\text{A14})$$

The integration of $\mu(\nu)$. ν on the semi-circle C can be expressed as the linear combination of integrals of ν^n with different powers n . These integrals are easily calculated using:

$$\frac{1}{2} \int_C \nu^n .d\theta = \frac{1}{2} |\nu|^n \int_0^\pi e^{jn\theta} .d\theta. \quad (\text{A15})$$

One finds

$$\int_0^F \mu''(f).f.df \approx \frac{\pi}{2} (F_M.F_y) \left[1 - \frac{2}{\pi} b \left(4\pi\chi_0 \frac{F_0}{F} + 2 \frac{F}{F_0} \right) - \frac{4\beta'}{\pi} \frac{F_0}{F} + \frac{2}{\pi} \alpha' \frac{F}{F_0} \right] + \frac{3}{2} \left(\frac{bF}{F_0} \right) .F^2. \quad (\text{A16})$$

Eq. (12) can be written under the following form

$$\int_0^F \mu''(f).f.df \approx \frac{\pi}{2} N_y (\gamma 4\pi M_s)^2 [1 - s - t + g] + S', \quad (\text{A17})$$

where s , S' , t and g are small corrective terms, with positive sign. s and S' are related to skin effect; t corresponds to the finite truncation; g corresponds to a small contribution that occurs in the case the Gilbert damping term is considered, but that is zero in the case the Bloch-Bloembergen damping model is considered. Each of these terms will be discussed in the following sections.

A. Gilbert damping correction

Previous work¹¹ had been conducted assuming a Bloch-Bloembergen damping. In the case the damping is described according to the Landau-Gilbert model, the integral up to infinite frequency diverges. The corrective terms in Eq. (13) writes:

$$g = \frac{2}{\pi} \alpha' \frac{F}{F_0} = \frac{2}{\pi} \alpha \frac{F}{F_y} = \frac{2}{\pi} \alpha \frac{F}{N_y F_M}. \quad (\text{A18})$$

The case where N_y is null or small is of no interest, since the dominant factor in the expression of the integral is proportional to N_y . As the upper integration bound F is expected to be lower than F_M , and because α is small (a few percents down to a fraction of percent are generally observed values), $g \ll 1$. This establishes that in practical case, the theoretical results obtained on $\int_0^F \mu''(f).f.df$ are independent on the damping term under consideration.

B. Skin effect correction

Let us examine in more detail the corrective terms associated to skin effect in Eq. 13. The term S' is independent of the magnetization of the sample.

$$S' = \frac{3}{2} \left(\frac{bF}{F_0} \right) .F^2. \quad (\text{A19})$$

It corresponds to the well-known fact that conductive particles may exhibit non unit permeability because of eddy currents. Though the integral of the imaginary permeability times frequency diverges at infinity, it should be noted that if the integral is performed only up to a frequency F that is not too large, according to Eq. (A9), then $S' \ll F^2$. In the case of ferromagnetic materials, it is convenient to express S' as a perturbation of the main term in Eq. (A17):

$$S' = \frac{\pi}{2} N_y F_M^2 .s'. \quad (\text{A20})$$

The expression for s' is:

$$s' = \frac{3}{\pi N_y} \left(\frac{bF}{F_0} \right) \cdot \left(\frac{F}{F_M} \right)^2 = \frac{6\mu_0 a^2 \sigma}{p} \cdot \frac{F^3}{F_M^2}. \quad (\text{A21})$$

s' is negligible provided (A19) is met and $F \ll F_M$. Let us now examine the s corrective term in 13,

$$s = \frac{2}{\pi} b \left(4\pi\chi_0 \frac{F_0}{F} + 2 \frac{F}{F_0} \right) \approx \frac{4\mu_0 a^2 \sigma}{p} N_y \frac{F_M^2}{F}. \quad (\text{A22})$$

It is straightforward that s is positive. It means that skin effect tends to decrease the value of the integral.

C. Finite frequency summation correction

Stopping the integration to some finite upper frequency affects the value of the integral, even if no skin effect is present. The t term in Eq. (A17) accounts for this truncation effect. It writes:

$$t = \frac{4\beta' F_0}{\pi F} \approx \frac{2}{\pi} \alpha (2N_x + N_y) \frac{F_M}{F}. \quad (\text{A23})$$

This correction factor is much less than unity provided

$$F \gg \alpha F_M = \alpha (\gamma 4\pi M_s). \quad (\text{A24})$$

Even for a very large saturation magnetization material such as CoFe with $4\pi M_s = 24\text{k Oe}$, for a typical value of $\alpha = 2\%$, Eq. (A24) requires that the upper integration frequency is such that $F \gg 1.4\text{ GHz}$, which is an easily met condition.

D. Effect of experimental measurements errors

When using the integral is computed from experimental permeability data, the error on the sum increases when the upper integration bound is extended. An error term has to be added to the right member of Eq. (A17). In order to be able to compare directly the error $\pm e$ to the other terms $-s$ and $-t$, it is convenient to write this additional term as

$$\frac{\pi}{2} N_y F_M^2 \cdot e.$$

It is straightforward to show that

$$|e| \leq \frac{\Delta\mu}{\pi N_y} \left(\frac{F}{F_M} \right)^2, \quad (\text{A25})$$

where $\Delta\mu$ is the maximum error on the measured permeability. The relative error is small provided

$$F \ll (\gamma 4\pi M_s) / \sqrt{\Delta\mu}. \quad (\text{A26})$$

Figure Caption

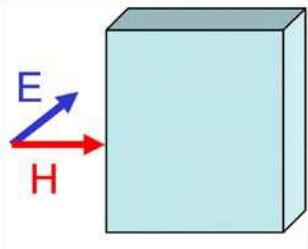
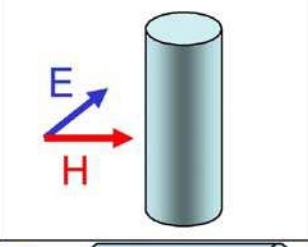
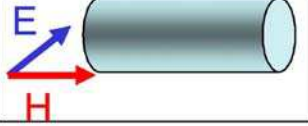

inclusion type	sketch	$A(ka)$	2 nd order approx.
plate		$\frac{\tan(ka)}{ka}$	$1 + \frac{(ka)^2}{3}$
cylinder ⊥ H		$\frac{J_1(ka)}{ka.J_0(ka) - J_1(ka)}$	$1 + \frac{(ka)^2}{4}$
cylinder // H		$\frac{2J_1(ka)}{ka.J_0(ka)}$	$1 + \frac{(ka)^2}{8}$
sphere		$\frac{2(\tan(ka) - ka)}{ka + ((ka)^2 - 1)\tan(ka)}$	$1 + \frac{(ka)^2}{10}$

FIG. 1. Various inclusions shapes, with the associated functions $A(ka)$ used for the renormalization of the permeability in the presence of skin effect; k is the wavevector inside of the inclusion, and a is its radius (or half thickness in the case of a plate). The expression of the 2nd order approximation of $A(ka)$ is also given.

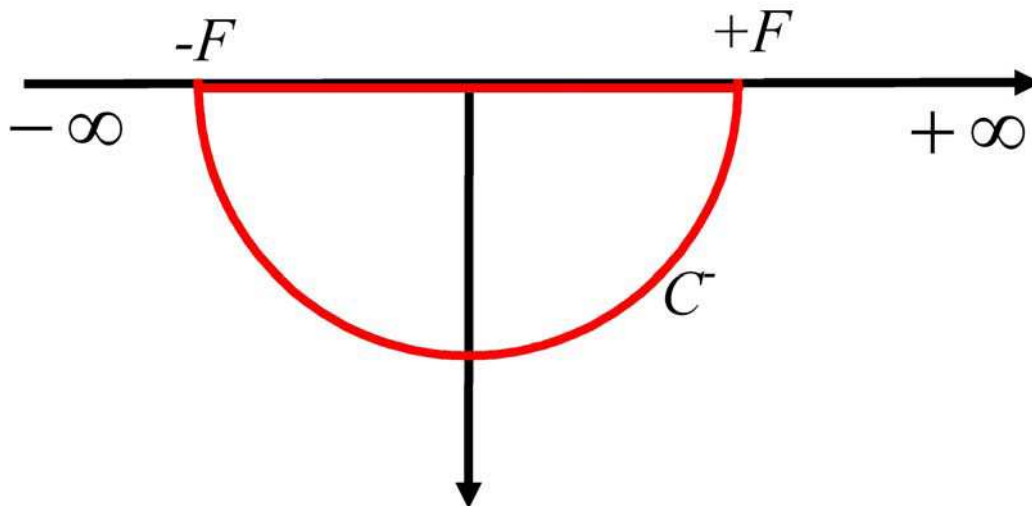


FIG. 2. Sketch of a closed integration contour in the complex frequency plane.

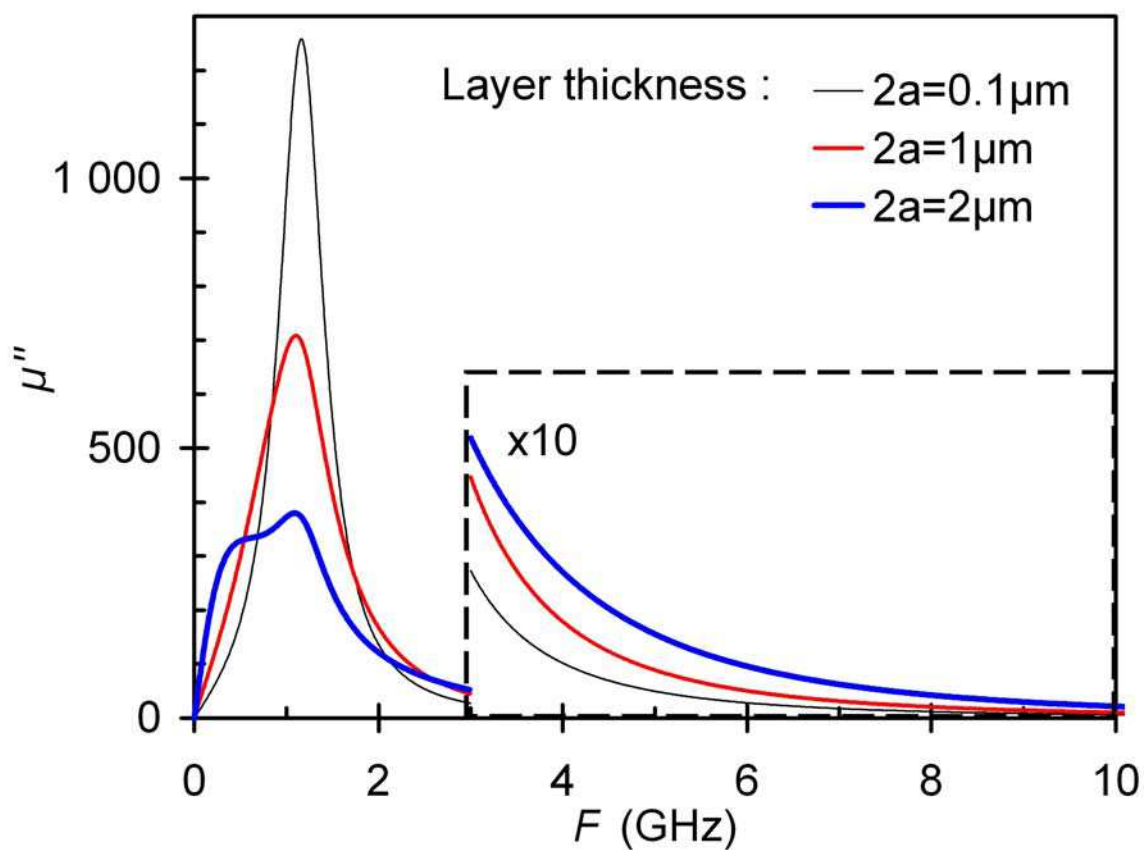


FIG. 3 Imaginary permeability μ'' computed for films with thickness $2a$ ranging from $0.1 \mu\text{m}$ to $2 \mu\text{m}$, using Landau-Gilbert model and taking into account skin effect.

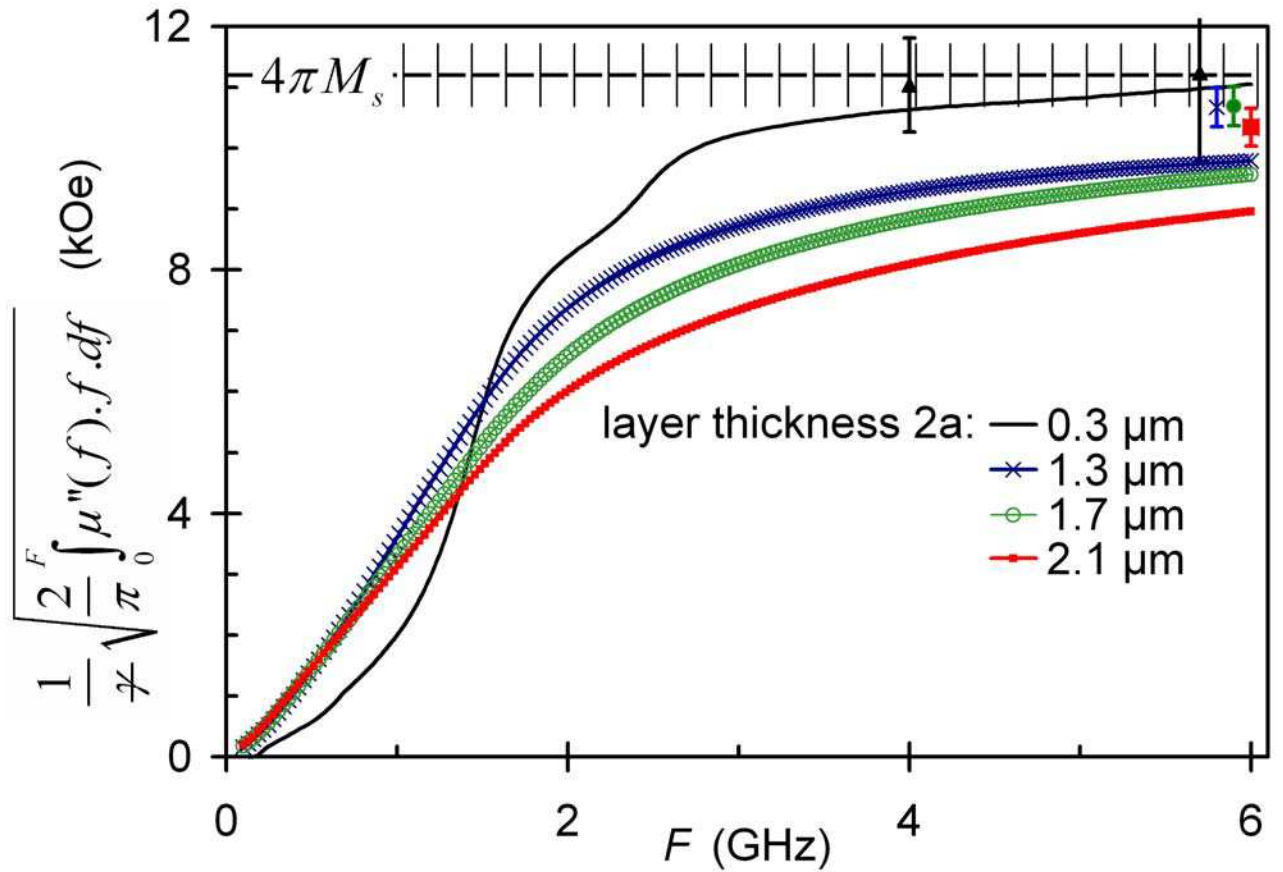


FIG 4. Efficient dynamic magnetization $M_\mu(F) = \frac{1}{\alpha} \sqrt{\frac{2}{\pi} \int_0^F \mu''(f) \cdot f \cdot df}$ as a function of the upper integration frequency for the calculated permeabilities represented on Fig. 3, obtained either by numerical integration (symbols), or from the analytical estimates (lines). The error bars that would arise from typical measurements errors on the permeability are also represented. The dashed line is the saturation magnetization.

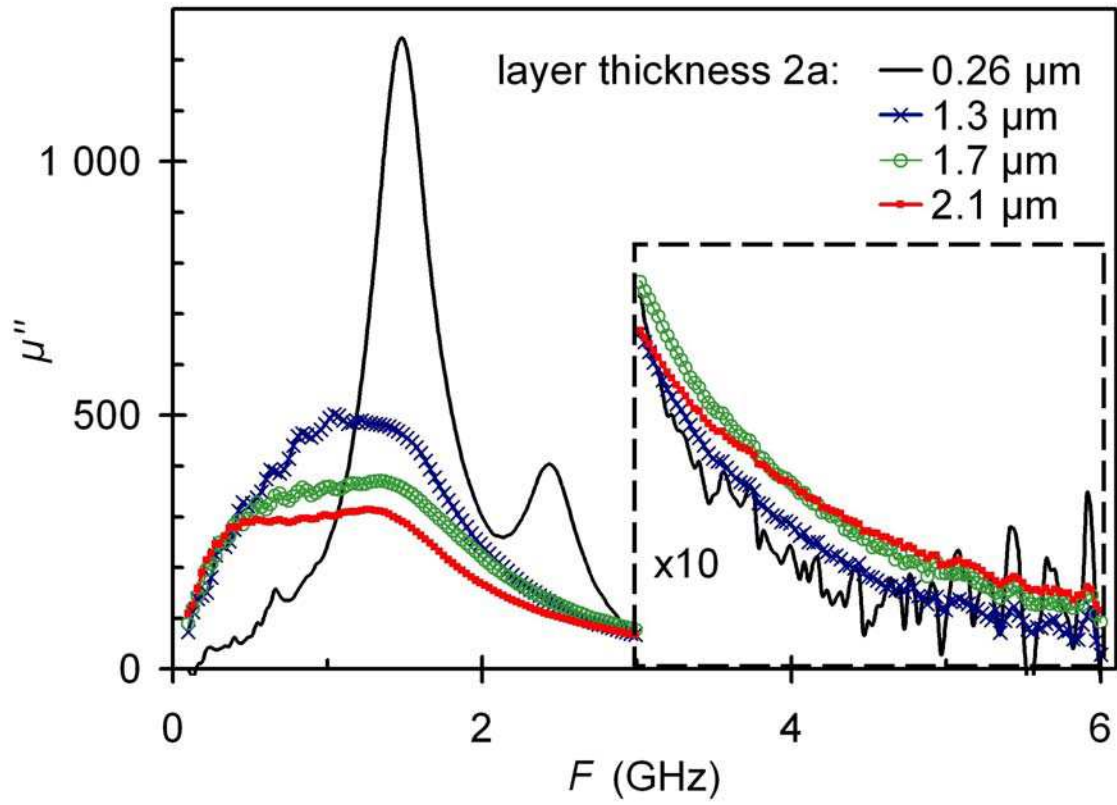


FIG. 5. Imaginary permeability μ'' measured on CoZr amorphous thin films with different thicknesses.

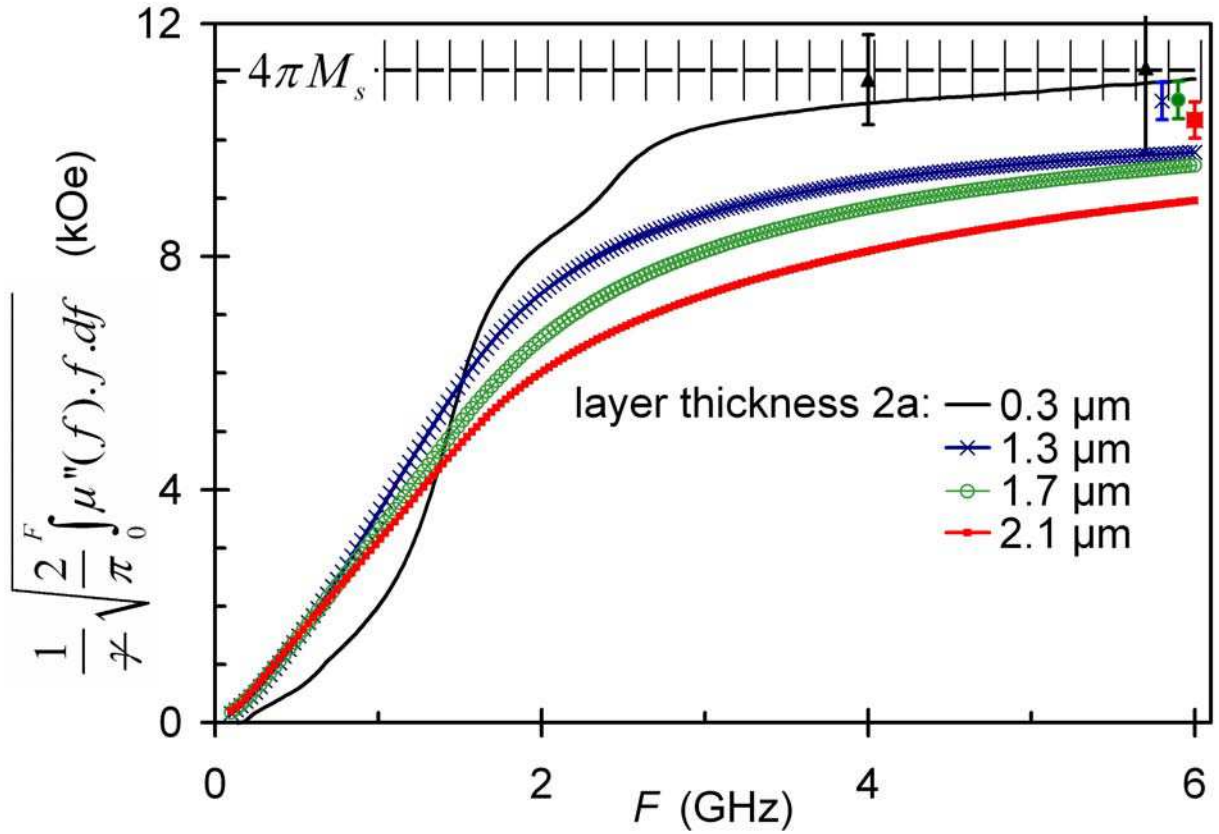


FIG. 6. Efficient dynamic magnetization $M_\mu(F) = \frac{1}{\varphi} \sqrt{\frac{2}{\pi_0} \int_0^F \mu''(f) \cdot f \cdot df}$ as a function of the upper integration frequency associated to the measured permeabilities represented on Fig. 5, and comparison with the value of the saturation magnetization (dashed line with error bars). Values of the saturation magnetization extrapolated from $M_\mu(F)$ using the analytical estimates are also represented, with appropriate error bars.

Table Caption

Lower bound	Upper bound	Eq.	Comment
$F_0 \ll F$	$F \ll (2\pi\mu_0 a^2 \sigma)^{-1}$	(A9)	Required for the validity of the whole approach
		(A4c)	$H_k, H_{\text{int}} \ll 4\pi M_s$ also required
	$F \ll F_M / \alpha$	(A18)	Effect of gilbert damping +g is small
	$F \ll F_M$	(A21)	Skin effect term s' is small
$4\mu_0 a^2 \sigma F_M^2 / p \ll F$		(A22)	Skin effect term -s is small
$\alpha F_M \ll F$		(A24)	Effect of truncation -t is small
	$F \ll F_M / \sqrt{\Delta\mu}$	(A25)	Impact $\pm e$ of experimental errors is small

Table I. Guidelines for choosing the upper integration frequency F on $\int_0^F \mu''(f).f.df$, as a function of the gyromagnetic resonance frequency F_0 , $F_M = \gamma 4\pi M_s$, and other parameters.

Lower bound	Upper bound	Parameter	Eq.	Analytical Value	Numerical value
$F_0 = 1.2$ GHz	$(2\pi\mu_0 a^2 \sigma)^{-1} = 165$ GHz	$\int_0^F \mu''(f).f.df$		$1.05 \cdot 10^3$ GHz ²	$0.99 \cdot 10^3$ GHz ²
	$F_M / \alpha = 1500$ GHz	+g	(A18)	0.3%	0.4%
	$F_M = 30$ GHz	+s'	(A21)	0.05%	$\ll 0.1\%$
$\frac{4\mu_0 a^2 \sigma F_M^2}{p} = 1.2$ GHz		-s	(22)	-19%	-23%
$\alpha F_M = 0.6$ GHz		-t	(21)	-6.4%	-6.5%
	$F_M / \sqrt{\Delta\mu} = 30$ GHz	$\pm e$	(23)	$\pm 1.3\%$	

Table II. Numerical estimation of the lower and upper frequency bounds for F associated to Table I in the case of a 2 μ m thick film; for $F=6$ GHz, values of the different corrective terms accounting for the relative difference between $\int_0^F \mu''(f).f.df$ and $\frac{\pi}{2}(\gamma 4\pi M_s)^2$ obtained both analytically and numerically.

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