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Alexis Chevalier, Jean-Luc Mattei, Marcel Le Floc'H. Ferromagnetic resonance of isotropic heterogeneous magnetic materials: theory and experiments. Journal of Magnetism and Magnetic Materials, 2000, Vol. 215, pp.66. hal-00491965

### HAL Id: hal-00491965 https://hal.univ-brest.fr/hal-00491965

Submitted on 19 Mar 2013

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Published in Journal of Magnetism and Magnetic Materials 215-216 (2000) 66-68

## Ferromagnetic resonance of isotropic heterogeneous magnetic materials: theory and experiments

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#### **Abstract**

Experimental variations of the ferromagnetic resonance (FMR) recorded on soft composite bodies are presented and their interpretation is undertaken. A successful application is performed for the Kittel expression of the FMR, initially written for an ellipsoid placed in vacuum, to a magnetic inclusion of the composite. This model includes the demagnetizing effects due to the magnetic inclusions. © 2000 Elsevier Science B.V. All rights reserved.

Keywords: Ferromagnetic resonance frequency; Demagnetizing effects; Composite materials

We have shown recently that ferrimagnetic composite bodies show a very different microwave behavior than one of the bulk materials [1,2]. The aim of this paper is to give an interpretation for the dependence of the ferromagnetic resonance (FMR) on the magnetic load of the composite.

The present study deals with composite bodies constituted of volume fraction C of ferrimagnetic soft particles mixed with a non-magnetic resin. The magnetic particles are  $Ni_{0.7}Zn_{0.3}Fe_2O_4$  and YIG grains, with diameters sized at 0.1 and  $10\,\mu\text{m}$ , respectively. It has been shown that the magnetization mechanisms in the obtained grains are controlled by spin rotations only [2]. (The powder synthesis is described in Ref. [2], and the preparation of the samples, together with the measurement technique of the dynamic susceptibility is detailed in Refs. [1,2].)

The magnetic grains are randomly dispersed in a non-magnetic matrix. They constitute clusters, the size of which is finite if the volume fraction C is below the percolation threshold  $C_p$ . Above  $C_p$  the percolating clus-

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ter is of infinite size [3]. Figs. 1 and 2 supply the experimental data of the ferromagnetic resonance frequency measured on the  $\mathrm{Ni}_{0.7}\mathrm{Zn}_{0.3}\mathrm{Fe}_2\mathrm{O}_4$  and YIG composites. It is known that demagnetizing fields play an important part in the values taken by the FMR in soft ferrites [4]. The samples we study are ring-shaped composites, allowing avoidance of macroscopic demagnetizing fields. However, we think that the *C*-dependent inner demagnetizing fields, i.e. concerning the clusters of aggregated magnetic particles, may act significantly on the FMR values.

The equation of motion of the magnetization vector M in the Landau-Lifshitz form is

$$\frac{\mathrm{d}\boldsymbol{M}}{\mathrm{d}t} = -\gamma(\boldsymbol{M} \times \boldsymbol{H}_0) - \alpha \frac{\gamma}{|\boldsymbol{M}|} [\boldsymbol{M} \times (\boldsymbol{M} \times \boldsymbol{H}_0)], \tag{1}$$

where  $H_0$  is the effective field responsible for the magnetic moments alignment,  $\gamma$  is the gyromagnetic ratio and  $\alpha$  is the damping parameter.

The precession of M around the internal field  $H_0$ , called ferromagnetic resonance, appears under the action of a weak alternating field h of frequency  $\omega$ . Solving Eq. (1) leads to the expression of the susceptibility tensor. The FMR arises at the frequency  $\omega_r$  for which the imaginary part of the diagonal terms is

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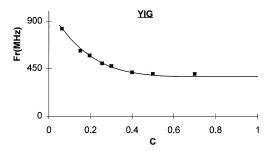


Fig. 1. Experimental (symbols) and theoretical (Eq. (6), full line) variations of the ferromagnetic resonance for YIG composites. ( $M_{\rm S}=140\,{\rm kA/m},\,\gamma H_0=400\,{\rm MHz}$ ).

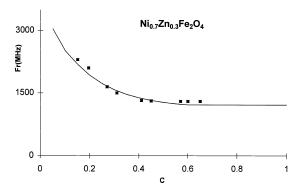


Fig. 2. Experimental (symbols) and theoretical (Eq. (6), full line) variations of the ferromagnetic resonance for  $Ni_{0.7}Zn_{0.3}Fe_2O_4$  composites. ( $M_S=410\,kA/m,\,\gamma H_0=750\,MHz$ ).

maximum. One finds

$$\omega_{\rm r} = \frac{\omega_0}{\sqrt{1 + \alpha^2}} \tag{2}$$

with  $\omega_0 = \gamma H_0$ . It must be noted that obtaining Eq. (2) does not require any assumption on the angle between  $H_0$  and h.

Moreover, as shown first by Kittel [5], the condition for resonance depends on the shape of the body. The Kittel equation, applicable to an ellipsoid in the saturated magnetic state, placed in the empty space, and with demagnetizing factors denoted by  $N_x^0, N_y^0$ , and  $N_z^0$ , is

$$\omega_{K} = \sqrt{(\omega_{0} + \omega_{m}(N_{y}^{0} - N_{z}^{0}))} \times \sqrt{(\omega_{0} + \omega_{m}(N_{x}^{0} - N_{z}^{0}))}.$$
(3)

In this relation,  $\omega_{\rm m}=\gamma M_{\rm S}$ , the z-axis is in the direction of the field  $H_0$  and the y- and x-axis are perpendicular to the z-axis. Therefore, by substituting  $\omega_0$  by  $\omega_{\rm K}$  in Eq. (2),

the FMR of an ellipsoidal body with damping is given by

$$\omega_{\rm r} = \frac{\omega_{\rm K}}{\sqrt{1 + \alpha^2}}.\tag{4}$$

Our purpose is to improve a possible application of this relation to isotropic composite materials. This extension requires a discussion about the following two assumptions, written in order to fit best the situation preluding to the setting of Eq. (3): (1) the composite material must be modeled by a single ellipsoidal particle imbeded in a homogeneous medium, the physical properties of which can be eventually derived by an effective medium theory (EMT); (2) the magnetic particles must behave as single-domain grains.

Considering the first assumption leads one to examine what are the demagnetizing factors  $N_{\alpha}$  for an ellipsoidal particle immersed in a homogeneous medium which is not the vacuum, but rather characterized by a permeability  $\mu_{\rm e} > 1$ . As demonstrated in Ref. [6], the demagnetizing factors associated to an ellipsoidal body (with a magnetic permeability  $\mu_{\rm i}$ ) immersed in vacuum ( $\mu_{\rm e} = 1$ ) are equal to the shape factors  $A_{\alpha}$ . Whereas, if the magnetic body is immersed in a magnetic medium, the demagnetizing factors will be  $N_{\alpha}$ . The factors  $A_{\alpha}$  and  $N_{\alpha}$  are linked by the following relation [6]:

$$N_{\alpha} = \frac{\mu_{\rm i} - \mu_{\rm e}}{\mu_{\rm e}(\mu_{\rm i} - 1)} A_{\alpha}.\tag{5}$$

We have shown that this expression is a fairly good description of a computed variation of  $N_{\alpha}$  [7]. Eq. (5) expresses that  $N_{\alpha}$ , and therefore the demagnetizing field, decreases when the magnetic properties of the surrounding medium tend towards the one of the magnetic ellipsoid.

As indicated above,  $\mu_e$  has been written by using a Bruggeman-type EMT [8]. The reason for this is that, as is well known, this EMT predicts the non-trivial phenomenon of percolation. Indeed, the percolation can intuitively be suspected to be of some importance in the demagnetizing effects appearing in the magnetic clusters of the composite. Moreover, the actual randomly dispersed particles system is then treated as uninteracting particles immersed in a uniform medium of permeability  $\mu_e$ . This feature fits well to the situation described by Eq. (3) of a single body immersed in a uniform medium. The second assumption is also acceptable in the presented cases, by considering the grain size for Ni<sub>0.7</sub>Zn<sub>0.3</sub>Fe<sub>2</sub>O<sub>4</sub>, but also the spectra of the dynamic permeability for the two presented materials. Indeed, these spectra do not reveal any Polder Smit effect which would indicate, through the appearance of a resonance band, the existence of interacting magnetic domains [1]. (Actually, a broadening of the resonance band, terminating at  $\omega_{\rm M} = \gamma M_{\rm S}$ , is a well-known experimental fact in the field of the microwave behavior of soft ferrites. This effect is explained by the precession motion of the magnetizations. It induces magnetic poles on each side of the domain walls of a multidomain particle. The linked demagnetizing effects enlarge the resonance band [4,9].) An extended form of the Kittel expression can be got by combining Eqs. (3)–(5). The proposed expression for the FMR of an isotropic composite material is the following:

$$\omega_{R} = \frac{1}{\sqrt{1 + \alpha^{2}}} \times \sqrt{\omega_{0} + \omega_{m} \frac{\mu_{i} - \mu_{e}}{\mu_{e}(\mu_{i} - 1)} (A_{y} - A_{z})} 
\times \sqrt{\omega_{0} + \omega_{m} \frac{\mu_{i} - \mu_{e}}{\mu_{e}(\mu_{i} - 1)} (A_{x} - A_{z})}.$$
(6)

This relation provides quite satisfactory fits of the experimental results (Figs. 1 and 2). The classical value of  $\alpha=0.3$  has been found to give the best results. The shape factors  $A_{\alpha}$  have been found by inverting the relation between  $\mu_{\rm e}$  and C derived from the EMT [1], and assuming the clusters to be of ellipsoidal shape  $(A_x=A_y,A_x+A_y+A_z=1)$ . As a conclusion, an analytical relation has been proposed which described the experimental variations of the FMR measured on composite materials, as a function of their volume fraction C in magnetic

matter. It is based on the description of the inner demagnetizing effects for a composite body of infinite size. The application of the found relation to the two presented experimental studies is promising. Further experiments will be carried out.

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