Spin resonance in soft magnetic composite materials : a surprising effect of the magnetic load.

Alexis Chevalier, F. Le Floc'H, Jean-Luc Mattei, Philippe Talbot, Marcel Le Floc'H

Spin resonance in soft magnetic composite materials: a surprising effect of the magnetic load

A. Chevalier, F. Le Floc'h*, J.L. Mattei, Ph. Talbot and M. Le Floc'h

LEST, UMR 6616 du CNRS, 6 avenue Le Gorgeu, BP. 809, 29285 Brest cedex, France
** UMR 652, Chimie-Électrochimie Moléculaire-Chimie Analytique, 6 avenue Le Gorgeu, BP. 809, 29285 Brest cedex, France*

Abstract. The paper shows a successful extension to the high frequencies, namely in the spin resonance regions, of an analytical law firstly proposed to interpret the magnetic properties of soft magnetic composite materials in low fields and in the quasi-static domain. During the first investigations where the frequency did not yet taken into account, it was shown that the effective susceptibility of powder materials was only dependent on three parameters : The magnetic load, the chemical nature of the magnetic compound (through the intrinsic susceptibility) and the internal demagnetizing fields (through the effective shape factor). Dynamic effects were then examined by introducing in the intrinsic susceptibility parameter the dynamic model of Landau and Lifshitz. Experiments carried on soft ferrimagnetic powders (YIG and Ni-Zn spinels) between 10kHz and 20GHz, have led to a total agreement with the analytical law mentioned above.

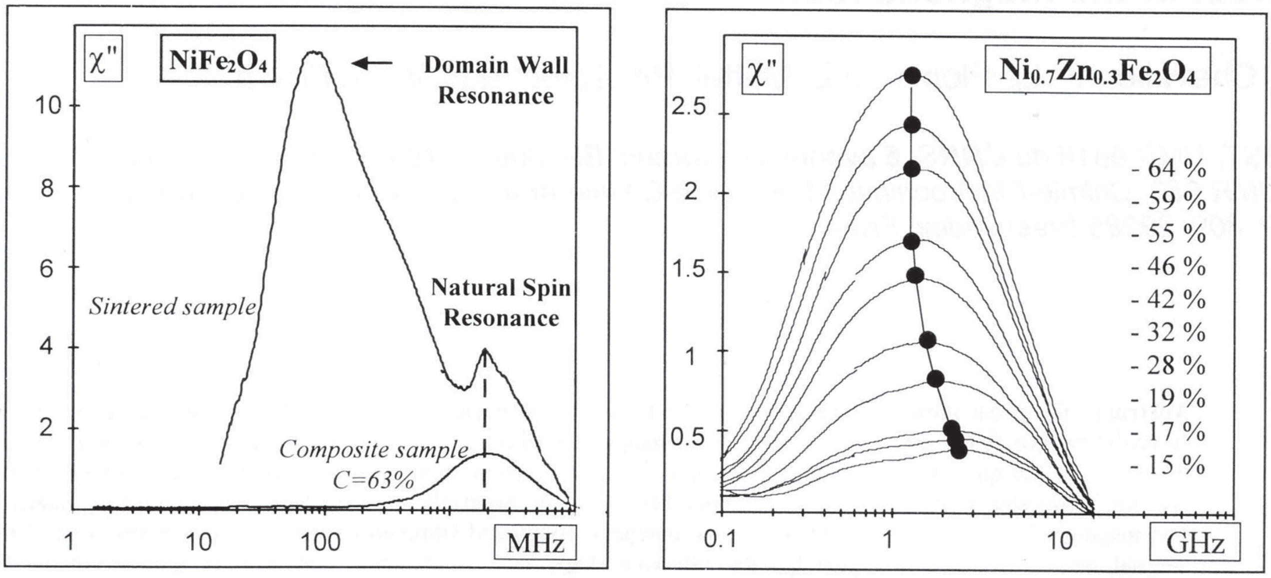
In order to describe the magnetic properties in soft magnetic composite materials, an analytical expression based on an average field theory has early been proposed and successfully supported by a number of experiments achieved in the quasi-static domain [1,2]. Although particle sizes were large enough to be divided in magnetic domains, it has been experimentally found that magnetization could satisfactorily be interpreted by the only rotational process. Actually this view is not really new since Rado, Wrigth and Emerson in 1950 [3], then Brown and Gravel in 1955 [4], had already observed that the domain wall contribution disappeared when bulk ferrites were transformed in powders then reshaped with the aid of a binder. The purpose of this work is now to extend our investigations concerning the static domain to the high frequencies and prove that the analytical expression mentioned above is able to interpret the quite general correlation we have experimentally found between the spin resonance (SR) frequencies in soft magnetic powders and their compacting ratio.

Composites materials have been made by mixing non-magnetic (resin) with soft magnetic (YIG, Nickel and Nickel-Zinc ferrites) powders, then by pressing the mixtures in ring-shaped matrixes at room temperature. The (volume) magnetic fraction ranged from a few percents to about sixty percents. The particle sizes being ranged from 0.1 to 150 μm, particles have polydomain structures with isotropic properties well described by a scalar susceptibility. The complex susceptibility ($\chi' - j\chi''$) has been investigated at room temperature in the 10 kHz-20 GHz frequency band using a coaxial-line technique and vector network analysers.

It is well known that magnetization in soft magnetic bulk materials originates from both spin rotations and domain wall motions and that the latter strongly depends on the microstructure. Different works [5-10] have clearly shown the influence of the grain size on the susceptibility in polycrystalline soft ferrites. In particular, Globus the first [5], then Van der Zaag recently on Mn-Zn ferrites [10], have proved that the reduction of the grain size led to single-domain structures where magnetization was governed by the only rotational process.

In connection, and concerning now powder materials, it was experimentally proved that the two peaks of dynamic losses observable in bulk ferrites (domain wall resonance and rotational resonance), changed in only one peak (that of the spin rotation) when bulk materials were powdered [3].

Figure 1 gives a supplementary argument in favour of magnetization mechanisms only controlled by spin rotations. Indeed, the peak of losses present at 80 MHz in the sintered ferrite, no longer exists in the powdered sample. The high frequency peak is unchanged at 1.8 GHz which is a well known value of the Natural Spin Resonance (NSR) frequency of the nickel ferrite ($NiFe_2O_4$).



a) b)

Figure 1 - a : This result shows that the simple transformation of the bulk ferrite in a composite structure (particles of the same magnetic substance embedded in a binder with a volume magnetic fraction of 0.63) involves the disappearance of the domain wall contribution without any changes of the NSR frequency (1.8 GHz) - **b** : Effect of the magnetic fraction on the Spin Resonance (SR) frequency shown in the case of Ni-Zn ferrite powders ($Ni_{0.7}Zn_{0.3}Fe_2O_4$). The frequency shift observed beyond a certain threshold of concentration (C_p) has in fact a quite general character observable in any ferrimagnetic (insulating) matter.

Figure 1- b shows that the decreasing of the magnetic load also decreases the magnitude of losses with a shift of the SR frequency below a certain threshold of concentration. Let us remark that this threshold has already been introduced in investigations concerning the static domain [2]. Moreover, the literature gives a number of recent works on metallic powders in which this frequency shift can be pointed out [11].

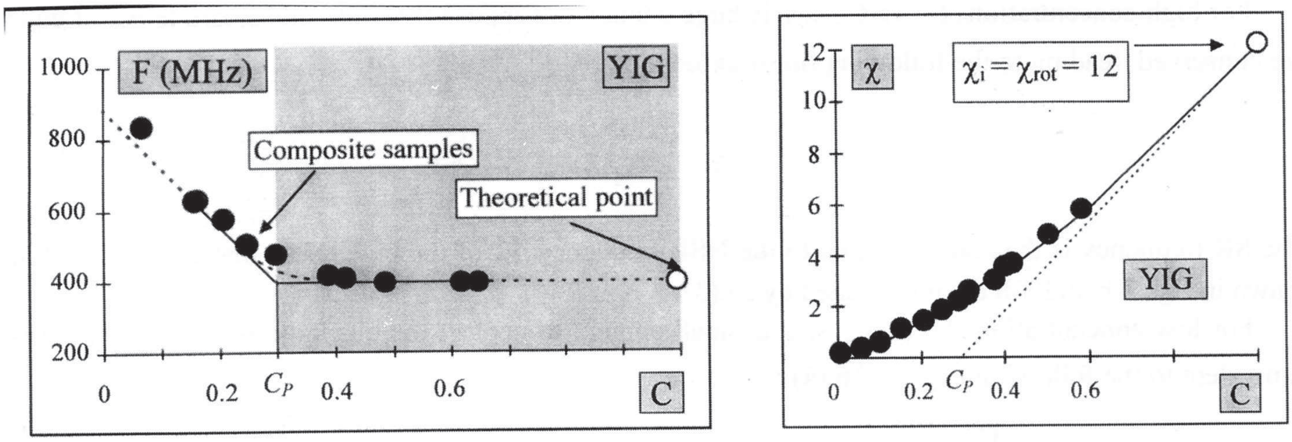
Figures 2-a and 3-a supply a more precise view of the relationship linking the SR frequency of composites to their magnetic loads (effect already visible in fig.1-b). Indeed, in all the materials studied, the increasing of the SR frequency with the decreasing of the magnetic fraction has systematically been observed, but surprisingly only below the threshold we have called C_p.

It means that materials which would be magnetically loaded just above this value (about 0.3), should have the same SR frequency as the bulk substance itself. Indeed, the values of the NSR frequencies deduced from this remarkable property (400 MHz for YIG and 1850 MHz for Ni-ferrite) agree well with those usually found in the literature [12]. Similar behaviours have also been observed in nickel-zinc ferrite composites with nevertheless an obvious contribution of the technological stresses [13].

We have established [1] an analytical relation linking the effective susceptibility χ of the mixture to both the chemical nature of the magnetic matter (through its intrinsic susceptibility χ_i) and the heterogeneous aspect of the medium (through an effective shape factor N) :

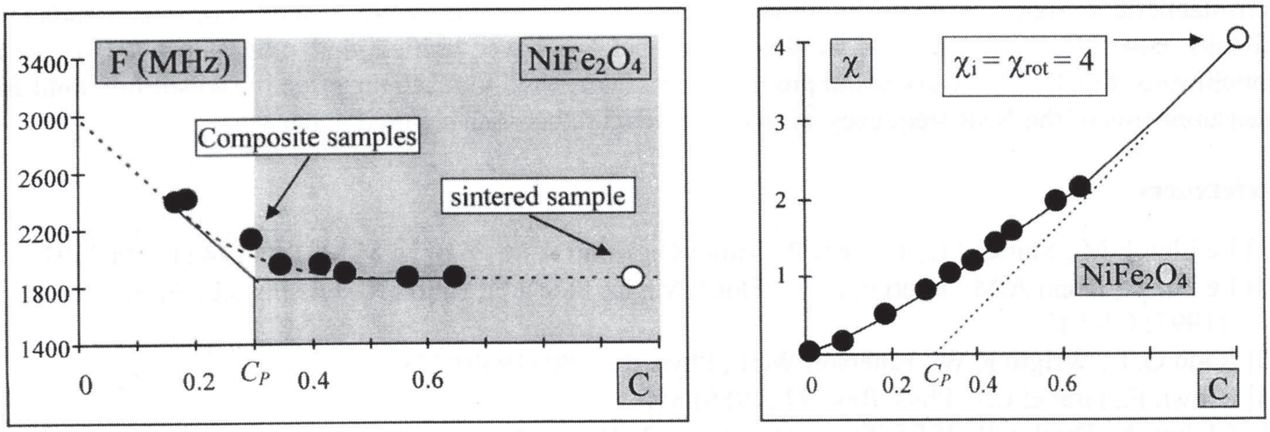
$$(1-N)\chi^2+[1+(N-C)\chi_i]\chi-C\chi_i=0 \quad (1)$$

C is the magnetic fraction, and N is connected to the shapes and distributions of particles. χ_i characterizes the rotational susceptibility. Indeed, the values of χ_i determined by comparing eq.(1) and experimental results in figs. 2-b and 3-b, lead to those usually assigned to the rotational susceptibilities (χ_{rot}) in the compounds investigated (YIG, $NiFe_2O_4$) [14-16].



a) b)

Figure 2 - a : Magnetic load dependence of the SR frequency in YIG composites. Better than in figure 1-b, the existence of the threshold C_p is clearly proved. Consequently, it may be said that magnetic powders with a compacting ratio just higher than 0.3 have the same SR frequency as the bulk itself (see the theoretical point) - **b :** Effect of the magnetic load on the static susceptibility in YIG composites [2]. Let notice that experimental values (solid circles) agree remarkably with the predictions (dashed curve) of our model (eq.1 [1]). Let notice also that the value 12 (open circle) deduced from eq.1 is close to that usually attributed to the rotational susceptibility of YIG [16].



a) b)

Figure 3 - a : Magnetic load dependence of the SR frequency in Ni ferrite composites. The remarks are the same as in fig.2-a - **b : :** Effect of the magnetic load on the static susceptibility. Let notice that the value 4 (open circle) deduced from eq.(1) is consistent with the experiment (full circles), and close to that usually attributed to the rotational susceptibility of $NiFe_2O_4$ [14,15].

In the investigation of the frequency behaviour, constant χ_i has been supposed to obey the Landau and Lifshitz equation [17] :

$$\frac{d\vec{M}}{dt} = \gamma\left(\vec{M} \wedge \vec{H}\right) - \frac{\alpha\gamma}{M}\vec{M} \wedge \left(\vec{M} \wedge \vec{H}\right) \qquad (2)$$

Magnetization $\vec{M}$ precesses around the internal field $\vec{H}$ under the action of a weak alternating field. γ is the gyromagnetic ratio and α the damping constant.

The theoretical SR frequency curves [18] derived from the combination of eqs.(1) and (2) are plotted in figs.2-a and 3-a (dashed lines). It is clear that theory agrees well with the experiments. In particular, the increasing of the SR frequency with the decreasing of the magnetic load below the threshold C_p is also well described by the model :

For high concentrations ($C >> C_p$), χ is large enough to suppose that only the square terms of eq.(1) are conserved, leading to the following linear expression of χ :

$$\chi = \frac{C - N}{1 - N} \chi_i \quad (3)$$

The SR frequency is then simply equal to the NSR frequency F_0 of the bulk. Note that the straight lines drawn in figs.2-b and 3-b are represented by eq.(3).

For low concentrations ($C << C_p$), χ is small enough to neglect the quadratic terms. Eq.(1) is then equivalent to the following expression (4) :

$$\chi = \frac{C\chi_i}{1 + (N - C)\chi_i} \quad (4) \qquad \text{and} \qquad F = Fo\sqrt{1 + (N - C)\chi_{rot}} \quad (5)$$

Eq.(5) is the SR frequency derived from eq (4). Note the similarity with the classical formulation of Kittel [19]. Eq.(5) shows that the SR frequency F is maximum for $C = 0$, then decreases up to $C = N$ (numerically equal to about 0.33). Of course, we conclude that $C_p = N$.

The validity of the analytical model (eq.1) has been extended to the dynamic properties of soft ferrimagnetic powders with no additional terms to interpret the major behaviours. The Landau and Lifshitz equation has been used in this paper, but we have verified that other laws led to similar conclusions. Finally, this work could provide an useful method to reach the effective anisotropy field from measurements of the NSR frequency by using powders rather than sintered materials.

References

[1] Le Floc'h M., Mattei J.L., Laurent P., Minot O., Konn A.M., J. of M.M.M. **140-144** (1995) 2191.
[2] Le Fur P., Konn A.M., Laurent P., Le Floc'h M., Le Floc'h F., Binon P., J.de Phys.IV France **7** (1997) C1-545.
[3] Rado G.T., Wrigth R.W., Emerson W.H., Phys. Rev. **80** (1950) 273.
[4] Brown F., Gravel Ch., Phys. Rev. **97** (1955) 55.
[5] Globus A., Duplex P., IEEE Trans. On Magn., **2** (1966) 441-445.
[6] Perduijn D., Peloschek H.P.,Proc. Beit. Cearm. Soc., **10** (1968) 263-273.
[7] Roess E., Hanke I., Moser E., Z. Angew. Phys., **17** (1964) 59.
[8] Beer A., Schwarz J., IEEE Trans. On Magn., **2** (1966) 470.
[9] Johnson M.T., Visser E.G., IEEE Trans. On Magn., **26** (1990) 1987-1989.
[10] Van der Zaag P.J., Ruigrok J.J.M., Noordermeer A., Van Delden M.H.W., Por P.T., Rekveldt M. Th., Donnet D.M., Chapman J.N., J. Of Appl. Phys.,**74** (1993) 4085-4095.
[11] Berthault A., Rousselle D., G. Zerah, J. of M.M.M. **112** (1992) 477.
[12] Globus A., J. de Phys. France, **38** (1977) C1-1 C1-15.
[13] Chevalier A., Le Floc'h F., Konn-Martin A.M., Le Floc'h M., «Influence of the moulding pressure on the magnetic properties of soft ferrite composite materials», SMM13-Grenoble September 1997.
[14] Globus A., C.R. Acad. Sc., **257** (1963) 1752-1755.
[15] Globus A., Duplex P., Guyot M., IEEE Trans. On Magn., **MAG-7** (1971) 617-622.
[16] Globus A., Duplex P., C.R. Acad. Sc., **262** (1966) 1188-1191.
[17] Landau L., Liftshitz E., Phys. Z. Sowjetunion **8** (1935) 153.
[18] Laurent P., Viau G., Konn A.M., Gelin Ph., Le Floc'h M., J. of M.M.M. **160** (1996) 63-64.
[19] Kittel C., Yager W.A. and Merritt F.R., Physica, **15,** (1949) 256.