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To cite this version:

HAL Id: hal-00656158
http://hal.univ-brest.fr/hal-00656158
Submitted on 19 Mar 2013

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Pressure Dependence of the Frequency Permeability Spectra of Soft Ferrite Composite Materials: A Method of Measuring the Natural Ferromagnetic Resonance Frequency

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Experimental studies on the complex susceptibility of soft magnetic composite materials (magnetostrictive Ni-Zn and Ni-Zn-Co spinels) in their region of ferromagnetic resonance (FMR) show that the FMR is an increasing function of the applied pressure. It is shown that powders could advantageously replace their sintered materials counterpart to measure without any ambiguity the natural ferrimagnetic resonance (NFMR) frequencies of bulk materials. It is also shown that such soft ferrite composites can be realized in order to shift FMR to higher values, and to obtain low magnetic losses up to 900 MHz. High frequencies applications can be envisaged.

Index Terms—Ferrimagnetic materials, ferrites, microwave materials, natural ferrimagnetic resonance measurements.

I. INTRODUCTION

This study deals with investigations on the ferromagnetic resonance (FMR) of various types of spinel ferrites powders. In particular, it is shown how powders might advantageously replace sintered materials to measure their natural ferrimagnetic resonance (NFMR) frequency. The studied materials are different in their chemical nature (Ni-Zn and Ni-Zn-Co-In spinels) as well as in their preparation mode (powders obtained either by grinding sintered ferrites or by co-precipitation method). Regarding ground powders, it is well known that residual stresses are able to partially screen the intrinsic magnetic properties by introducing additional anisotropy terms. This is true in particular for stress sensitive materials for which the FMR frequency deviates far from NFMR value. The experimental value of the effective anisotropy field \( H_k \) is then hardly accessible. Therefore, through a study of the stamping effects observed on the high frequency susceptibility in the FMR region, this work shows how powders provide an efficient way to overcome the problem of the uncontrolled internal stresses. This study also shows that the shifting to higher frequencies of the dispersive region (and of the associated magnetic losses) allow to considerate stressed ferrites as potential candidates for frequencies applications up to 900 MHz.

II. EXPERIMENTS

Nano-sized particles (diameter = 30 nm) of spinel ferrite \( \text{Ni}_{0.7}\text{Zn}_{0.3}\text{Fe}_2\text{O}_4 \), \( \text{Ni}_{0.4}\text{Zn}_{0.6}\text{Fe}_2\text{O}_4 \), and \( \text{Ni}_{0.2}\text{Zn}_{0.8}\text{Co}_{0.2} \text{Fe}_{1.98}\text{O}_4 \) were prepared by co-precipitation method. More details on synthesis process and on structural characterization can be found in [1]. Soft magnetic composite materials were prepared by mixing epoxy resin with ferrite particles (with a magnetic load C at about 0.5). This value must be carefully chosen. Actually, it is an experimental fact that inner demagnetizing effects turn to outer effects when the fraction in magnetic matter goes through a particular value [2], [3]. For heterogeneous and randomly structured materials, cooperative phenomena in the composite arise at a percolation threshold \( C_p \) that lies between filler fractions from 20 to 40-vol.% [2], [4]. Therefore, in order to avoid such inner demagnetizing effects on the FMR value, C value was chosen above the percolation threshold. It has been demonstrated that, above \( C_p \) the resonance frequency no longer depends on the magnetic load [5], [13]. The mixture was moulded under uniaxial stress, with various applied pressure values. It must be obvious that for further experiments, the sample thickness will be reduced by the applied pressure; consequently the magnetic load will increase. However this will not affect the measured resonance frequency, precisely because the magnetic load remains well above the percolation threshold. Because C is well above the percolation threshold, the magnetic particles are expected to form a percolating cluster. Therefore a pressure applied on the sample would lead the magnetic particles to be strongly fitted into each other. The anisotropy induced by the pressure is therefore not expected to relax, and this is will be well confirmed by the experiments.

A Hewlett Packard HP 8753ES network analyzer and APC7 coaxial-line setup was used for the measurements of the complex permeability (\( \mu = \mu' - \mu'' \)) in the frequency range [10 MHz–6 GHz]. The measured FMR frequencies \( f_R \) are obtained from permeability spectra measurements (maximum of the imaginary part), for various applied pressures. The accuracy on the measured values of \( f_R \) is always better than 50 MHz. The reproducibility of the presented experiments have been checked, on these spinels ferrites, as well as on other soft ferrites (not shown in this paper).
energy writes: \( E = (K + 3/2\lambda_\gamma \sigma_1) \sin^2 \varphi \). \( K \) is the magnetocrystalline anisotropy constant, \( \lambda_\gamma \) is the magnetostriction constant, and \( \sigma_1 \) the local stress on the grains (correlated to the moulding pressure \( P \)). Assuming that the angle \( \varphi \) between the magnetization and the easy axis is low, an effective anisotropy field can be defined by [10]: \( H_{\text{eff}} = -\mu_0 M_S H_{\text{eff}} \cos \varphi \). Straightforward calculations on [111] axe (the studied materials present four easy axes) leads to

\[
H_{\text{eff}} = \frac{4}{3} \left( K + 3/2\lambda_\gamma \sigma_1 \right) M_S .
\]  

The ferrimagnetic resonance frequency \( F_R \) is given by \( F_R = \gamma H_{\text{eff}} \) where \( \gamma \) is the gyromagnetic ratio (\( \gamma = 35.185 \text{ MHz/kA} \cdot \text{m}^{-1} \)), that is

\[
F_R = F_0 + \frac{2\gamma}{\mu_0} \left( \frac{\lambda_\gamma}{M_S} \right) \sigma_1 .
\]  

Here \( F_0 \) is the NFM frequency of the bulk which is linked to the magnetocrystalline anisotropy field \( H_K \) through the gyromagnetic ratio: \( F_0 = \gamma H_K \).

Relation (2) applies for a material in which the particles would be submitted to internal stresses \( \sigma_1 \) all applied along the magnetization direction (say along one of the easy axes). Although it might be difficult to theoretically correlate the internal stress \( \sigma_1 \) to the moulding pressure \( P \), our experiments allow to think that, inside the domain of strength investigated, \( \sigma_1 \) is proportional to \( P \) (\( \sigma_1 \approx \alpha P \)) so that an empirical law fairly similar to (2) can be expressed as

\[
F_R = F_0 + \alpha \frac{2\gamma}{\mu_0} \left( \frac{\lambda_\gamma}{M_S} \right) P
\]  

where \( \alpha \) is an unknown factor to be determined experimentally. The physical meaning of the factor \( \alpha \) is sketched in Fig. 3: because of the existing voids in the ferrite compact, the local strengths are most probably higher than the applied strength itself. Its values might depend on mechanical properties of the particles. All our experiments on \( \text{Ni}_6\text{Zn}_{1-x}\text{Co}_x\text{Fe}_2\text{O}_4 \) ferrites (where \( x \) belongs to \([0.3, 0.7]\)) led to an experimental value \( \alpha \approx 2 \). Relation (3) establishes a linear variation of the FMR with the applied pressure \( P \) from which one would get the ratio \( \lambda_\gamma M_S \) and the NFM frequency \( F_0 \) of the bulk. The same behavior was observed for \( \text{Ni}_6\text{Zn}_{1-x}\text{Fe}_2\text{O}_4 \) composites (magnetic load \( C = 0.5 \)). The obtained data are summarized in Table I. The magnetostriiction constants were obtained after magnetization measurements by SQUID experiments. They are in agreement with published data [7]. The obtained values for \( H_{\text{eff}} \) are also consistent with published data on the magnetocrystalline anisotropy constant \( K \) [11].

The region below 50 MPa, where the FMR is constant, but higher than usual values for bulk materials, may correspond to permanent internal stresses that exist before moulding.

Another benefit of this study is that it may open to soft ferrites the field of microwave applications up to 1 GHz. Actually spinel ferrite materials usually exhibit high magnetic losses limiting their potential to be used as high frequency devices below 400 MHz.

Fig. 4 shows the effect of the moulding pressure on the permeability. Thanks to adequate manufacturing process and to the magnetostriective effect demonstrated on Figs. 1 and 2, soft ferrite composites can be realized in order to shift FMR to.
TABLE I

<table>
<thead>
<tr>
<th>Sample type</th>
<th>$M_s$ (kA/m)</th>
<th>$\lambda_s \times 10^6$</th>
<th>$f_R$ (GHz)</th>
<th>$H_K$ (kA/m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ni$<em>{0.7}$Zn$</em>{0.3}$Fe$_2$O$_4$</td>
<td>345</td>
<td>-6.5</td>
<td>0.27</td>
<td>7.5</td>
</tr>
<tr>
<td>Ni$<em>{0.7}$Zn$</em>{0.3}$Fe$_2$O$_4$</td>
<td>410</td>
<td>-17.5</td>
<td>0.75</td>
<td>21.3</td>
</tr>
<tr>
<td>Ni$<em>{0.3}$Zn$</em>{0.7}$Co$<em>{0.2}$In$</em>{0.02}$Fe$_{1.98}$O$_4$</td>
<td>420</td>
<td>-21</td>
<td>2.68</td>
<td>76.2</td>
</tr>
</tbody>
</table>

IV. CONCLUSION

Soft magnetic composite materials have FMR frequencies which tend towards the NFMR of bulks (sintered materials). Powders seem to be, thus, a very interesting aspect of matter to develop useful methods for investigating anisotropy fields by high frequency measurements. Concerning the difficult question of stresses induced by the moulding phase, it has been shown how a simple technique of extrapolation can lead to the expected NFMR frequency. Because of their low magnetic losses up to 900 MHz these composites can be envisaged as devices for high frequencies applications.

REFERENCES