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This article reports on an isotropic model of the magnetic susceptibility based on an average field theory and proposes to predict the dynamic behaviors of powder magnetic materials. It was essentially built around a so-called effective demagnetizing factor capable of taking the particle shapes into account. So, for a population of randomly distributed particles of anisotropic shapes like, for instance, needles or flakes, we show that the effective demagnetizing factor of this population of particles can be significantly lowered with regard to the well known value of 1/3 classically used to represent the isotropy state. This phenomenon is interpreted as the natural tendency of particles to form clusters to which a moving demagnetizing factor must be assigned. Taking then the aggregation process of particles into account, the ability of the model to predict the dynamic properties of many composite magnetic materials is successfully demonstrated. Our development is illustrated by experimental results concerning a nickel–zinc ferrimagnetic ($Ni_{0.7}Zn_{0.3}Fe_2O_4$) powder. © *2001 American Institute of Physics.* [DOI: 10.1063/1.1389520]

I. INTRODUCTION

Over the last years the study of composite materials aroused growing interest. We report here on microwave applications of composite materials consisting of soft magnetic particles embedded in insulating binders.

Several theories, reviewed by Greffe and Grosse,[1] have been proposed to describe the dielectric properties of composite materials. One of the most known among them, the effective medium theory (EMT), has been developed by Bruggeman[2] and predicts the average susceptibility of inhomogeneous media. In a previous article[3] we showed that, in the simple case of a two-phase mixture where only one of them was magnetic, the EMT leads to a simple and useful scalar expression as follows:

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

The magnetic susceptibility χ of the mixture depends on the intrinsic susceptibility χ_i of the magnetic substance as well as on the magnetic volume fraction C, and then on the so-called effective shape factor N for the global representation of the microscopic demagnetizing effects. The next part of our study details how the factor N can be connected to the shapes of particles and to their distributions in space.

II. THEORETICAL DEVELOPMENT

In order to simply calculate the effective factor N, we assume that the particles of concern can be replaced by ellipsoids whose the associated triplet of demagnetizing constants (N_x, N_y, N_z) is fixed by the ratios of the respective lengths of their semi-axes. One can, then, calculate the diagonal tensor[4] of the internal field, H_i, for such ellipsoid particles of permeability, μ_i, embedded in a homogeneous medium of permeability, μ (Fig. 1).

The isotropic character of the mixture is, next, introduced through the assumption of the random distribution of particles in space. The mean internal field $\langle H_i \rangle$ is derived from the average of the three earlier components

$$\langle H_i \rangle = \tfrac{1}{3}(H_{i/x} + H_{i/y} + H_{i/z}). \qquad (2)$$

Since the demagnetizing factors satisfy the conditions: $N_x + N_y + N_z = 1$ and $N_x = N_y$ then, effective data on particle shapes are reduced to the only parameter N_z. The internal field $\langle H_i \rangle$, then, becomes

$$\langle H_i \rangle = \left[\frac{2}{\mu + (\mu_i - \mu)(1-N_z)/2} + \frac{1}{\mu + (\mu_i - \mu)N_z} \right] \frac{\mu H}{3}. \qquad (3)$$

In the meaning of the effective medium theory applied to the isotropic case, we choose to define[5,6] the effective shape factor N by taking for $\langle H_i \rangle$ the same formal expression as that of the H_i components

$$\langle H_i \rangle = \frac{H}{1 + (\mu_i - \mu/\mu)N}. \qquad (4)$$

The equality between relations (3) and (4), then, allows us to connect N, first to the real parameter of shape N_z of the magnetic particles, and second to the volume fraction C. The effective shape factor N is then given by

$$N = \frac{2 + 3\delta N_z(1-N_z)}{6 + \delta + 3\delta N_z}, \qquad (5)$$

where $\delta = \chi_i/(1+\chi)$ is a so-called factor of contrast, which itself depends on the volume fraction C.

The values of N are calculated by solving Eqs. (1) and (5) together. Figure 2 shows that the effective shape factor N depends both on the magnetic load C and on the real shape factor N_z of particles. For separated spherical particles N is equal to 1/3 over the whole volume fraction range. On the other hand, in all other cases, i.e., needle- or flake-type par-
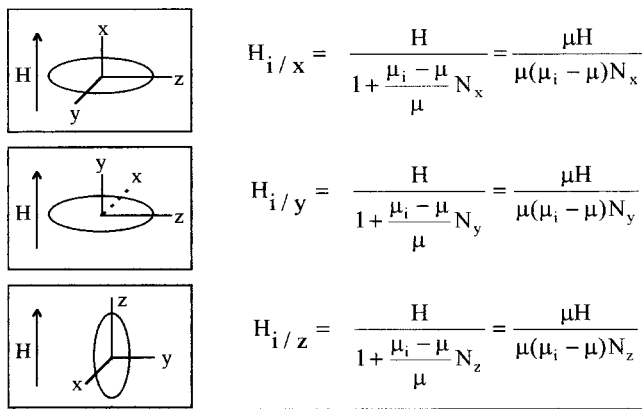

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FIG. 1. Components of the internal field H_i.

FIG. 3. Experimental variations of the real part χ' of susceptibility of $Ni_{0.7}Zn_{0.3}Fe_2O_4$ magnetic composites.

ticles, N is smaller than $1/3$ so that the shape anisotropy of particles, even for an isotropic mixture, always has the direct consequence to systematically lower the effective shape factor of the mixture.

We will now show the ability of the above analytical model [Eqs. (1) and (5)] to predict the dynamic properties of soft magnetic powders. In the example chosen, the model is applied to the extraction, from experimental data, of the intrinsic susceptibility χ_i of the magnetic matter.

III. EXPERIMENTAL RESULTS

The fine magnetic powders used in our study were synthesized in our laboratory by a wet method[7] in order to be sure of the very high chemical purity of the resulting fine powders. Observations by electron microscopy showed that most of the particles were regular and spherical with diameters close to 0.1 μm. However, in agreement with the earlier theory, one should keep in mind that clusters can strongly deviate the magnetic properties of the macroscopic body.

Composites materials were made by mixing nonmagnetic (epoxy resin) with soft magnetic ($Ni_{0.7}Zn_{0.3}Fe_2O_4$ ferrite) powders. Then, they were shaped and pressed at room temperature in ring-shaped matrices to avoid the sample de-

magnetizing fields. The magnetic volume fraction ranged within a few percent to about 60%. Thermal densifications were not used in this study.

The initial dynamic susceptibility of the composite material $[\chi(\omega)=\chi'(\omega)-j\chi''(\omega)]$ was investigated at room temperature in the frequency range 50 MHz–20 GHz by using a Hewlett-Packard APC7 coaxial-line technique and network analyzers. The results are shown in Figs. 3 (real parts) and 4 (imaginary parts) for different magnetic fractions in the earlier mentioned range. The reduction of the magnetic load lowers the magnitude of the susceptibility. This behavior is general and usually expected.

However, as the magnetic resonance frequency (MRF) is spotted by the maximum of the imaginary part of susceptibility, its behavior is clearly different: first, it remains unchanged, then increases continuously below a certain threshold C_p close to 30%.

We consider this resonance frequency shift as being a direct consequence of the increase of the microscopic demagnetizing fields in the mixture in connection with the modifications of dipolar-type interactions on the one hand, and of the particle arrangements on the other hand. This is quite consistent with other studies which have also clearly highlighted the basic role of the demagnetizing effects[8] on the dynamic properties of the magnetic susceptibility in composite materials. As the volume fraction decreases, the local demagnetizing fields add to the total anisotropy field and consequently increase the resonance frequency. In the next
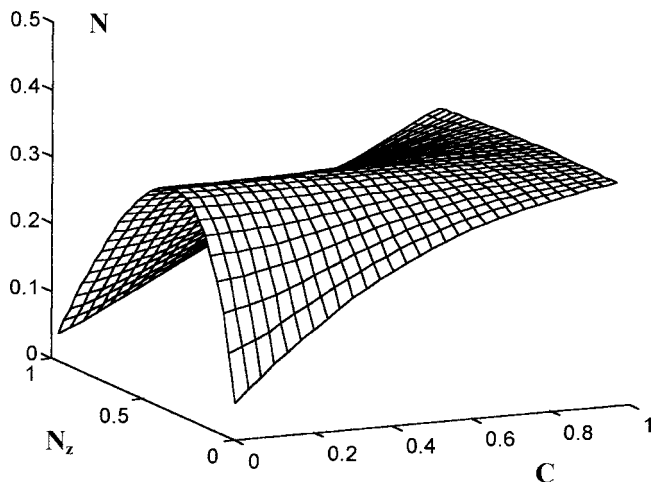

FIG. 2. Effective shape factor N as a function of the magnetic load C and of the particle shape factor (N_z). Theoretical values are obtained by solving the couple of Eqs. (1) and (5) for $\chi_i = 20$.
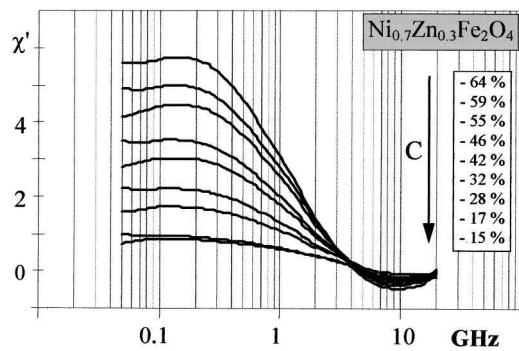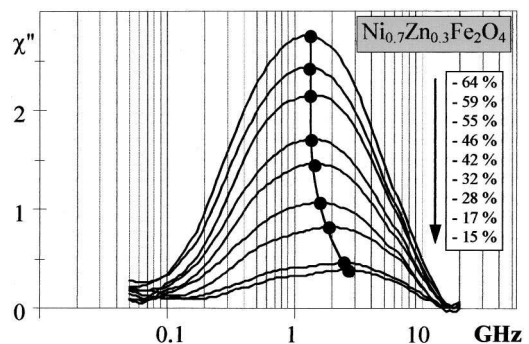

FIG. 4. Experimental variations of the imaginary part χ'' of susceptibility of $Ni_{0.7}Zn_{0.3}Fe_2O_4$ magnetic composites. Full circles (●) spot the resonance frequency shift.
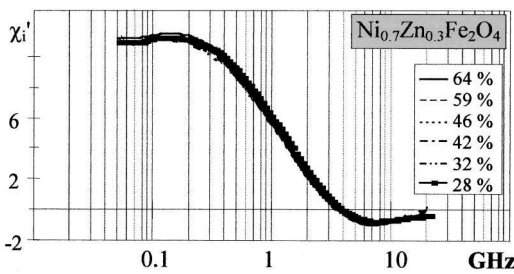
FIG. 5. Real part of the dynamic intrinsic susceptibility χ_i' obtained with the inversion of experimental data of Fig. 3.

FIG. 7. Experimental and theoretical values of the effective shape factor N obtained to superimpose dynamic spectra of Figs. 5 and 6.

section we will demonstrate that the model proposed in this study takes properly this internal effect into account.

IV. COMPARISONS WITH THE MODEL

To process experimental data in order to extract the intrinsic susceptibility of the magnetic matter, we use Eq. (1) in which χ_i is taken as the unknown variable, whereas C and χ come from measurements. N is then adjusted under the obvious condition that it must remain less than or equal to 1/3. This leads to the noteworthy transformation of Figs. 3 and 4 into the reduced curves of Figs. 5 and 6. One should, indeed, note that, over the whole frequency range and for the different magnetic fractions, there is a unique solution for N; in consequence, the model we use [see Eq. (1)] can describe both the decreasing magnitude of the susceptibility and the shift in the MRF observable in composites magnetic materials.

Figure 7 shows the experimental values of the earlier solution for N. They correspond to the expected intrinsic susceptibility and are absolutely consistent with the theoretical curves predicted in Fig. 2. They are not only less than 1/3, but they also depend on the volume fraction.

V. DISCUSSION

We showed in the previous section that the effective shape factor N depends on both the real particle shape (N_z) and the magnetic fraction C [Eq. (5)]. Thus, theoretically in the case of spherical particles, the effective shape factor N must be equal to 1/3 and not depend on the magnetic fraction (see Fig. 2). However, experimental values are systematically below 1/3 and magnetic fraction-dependent even in the case of spherical magnetic particles (Fig. 7). This discrepancy, in fact, results from the unavoidable aggregations of
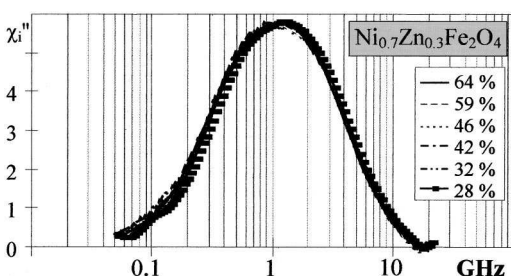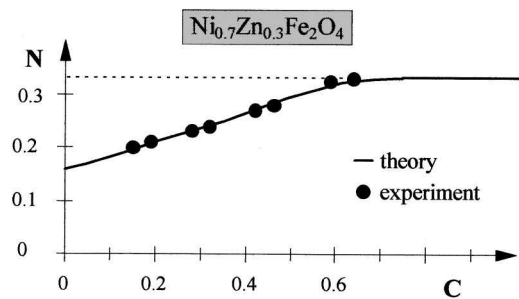
particles during the powder mixing processes, particularly when they are of very small sizes. By way of consequence the clusters are perceived as new particles distorted and bigger than the others. It is then obvious that the shape distribution of such aggregates cannot be evaluated by direct (microscopy) observations, especially because they are sensitive to the changes in the magnetic load. This is, in fact, quite a general problem which largely goes beyond the limits of this study.

It is, however, quite possible from simple and practical assumptions to build an intuitive function of distribution of the cluster shapes. First, any aggregation of individual ellipsoidal particles necessarily results in clusters of ellipsoidal shapes with a demagnetizing factor N_z (see Fig. 8). Second, the probability $P(N_z)$ to find clusters with a given shape depends on the magnetic fraction as follows: at low concentrations, the particles tend to remain linked in small chain-like aggregates so as they are equivalent to prolate ellipsoids,[9] and then the composite becomes equivalent to a mixture of needle-type particles for which N_z is by definition less than 1/3. On the contrary, at high concentrations, the clusters grow in any direction and progressively tend to become spherical: by definition, N_z is here equal to 1/3. In our study we considered that the probability to develop flake-like aggregates for which N_z is by definition higher than 1/3 was unrealistic.

FIG. 6. Imaginary part of the dynamic intrinsic susceptibility χ_i'', obtained with the inversion of experimental data of Fig. 4.

cluster	associated ellipsoid		c/a	N_z
◯			1	1/3
◯◯			2	0.173
◯◯◯			3	0.108
◯◯◯◯◯			2.5	0.089
◯◯◯◯◯◯			0.3	0.661

FIG. 8. Identification of aggregates with ellipsoid.

FIG. 9. Statistical distribution $P(N_z)$ of aggregates shape (N_z) with the volume fraction C.

When the particle concentration in a given mixture is increasing, aggregates are rapidly formed; their shapes are, then, continuously changing from chains to spheres up to the concentration unity where the medium becomes isotropic. A further work would be in deriving a mathematical function of the distribution $P(N_z)$ to have finally a completely predicting model. This is an interesting challenge because it is known that the dynamic properties of magnetic powder materials are largely influenced by such a term.

The earlier assumptions are sufficient to design a convincing profile of the distribution law $P(N_z)$. Indeed, as the magnetic concentration increases, the function $P(N_z)$ must have a maximum whose position is shifted towards the high values of N_z up to 1/3 (strictly speaking) when the magnetic fraction reaches unity. Simultaneously, the profile of $P(N_z)$ must become narrower and narrower. Figure 9, finally, shows the histogram of $P(N_z)$ which was used to realize the very satisfactory fit of Fig. 7.

For an exact computation of the effective shape factor N, Eq. (3) can be now easily expressed as a continuous sum acting on N_z as follows:

$$\langle H_i \rangle = \int_0^1 \left[\frac{2}{\mu + (\mu_i - \mu)(1 - N_z)/2} \right.$$
$$\left. + \frac{1}{\mu + (\mu_i - \mu)N_z} \right] \frac{\mu H}{3} P(N_z) dN_z. \tag{6}$$

VI. CONCLUSION

An extension of the effective medium theory was proposed to predict the dynamic behaviors of isotropic composite materials. This model is capable of describing experimental results, namely the variation of magnitude of permeability, together with the shift of the resonance frequency. While taking into account the formation of particle aggregates, we derived the intrinsic permeability of magnetic matter independently of the magnetic fraction of the mixture. An immediate and interesting extension of this work would be the determination of an analytical function of the magnetic fraction of the mixture to take the grain structure contribution into account.

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