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► To cite this version:

Jean-Luc Mattei, Philippe Laurent, A. Chevalier, M. Le Floc'H. Application of an Effective Medium Theory to Composite Materials with Randomly Dispersed Particles of Specific Shapes. Journal de Physique IV Proceedings, 1997, 07 (C1), pp.C1-547-C1-548. 10.1051/jp4:19971225 . jpa-00254898

HAL Id: jpa-00254898

<https://hal.science/jpa-00254898>

Submitted on 1 Jan 1997

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Application of an Effective Medium Theory to Composite Materials with Randomly Dispersed Particles of Specific Shapes

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Abstract. The Effective Medium Theory used to describe the magnetic properties of randomized heterogeneous materials includes as parameters the intrinsic permeabilities μ_i of the magnetic particles and an effective shape factor $\bar{N}$. This paper compares the experimental variations of $\bar{N}$ to the theoretical variations predicted by an EMT. It is shown that the *effective shape factor* $\bar{N}$ cannot be seen as the *mean shape factor* of the composite components. The values of $\bar{N}$ belong to the range $]0, 1/3[$, and are strongly correlated to the permeabilities μ_i . In the low concentration range, random distributions of needle-like and flake-like particles both lead to low values for $\bar{N}$. The result $\bar{N}=1/3$ is obtained in the whole concentration range only in the case of randomly dispersed spheres. The values of the μ_i might be kept constant up to a volumic fraction c in magnetic matter equal to 50%, according to previous results we got. The experimental results presented here are well described by this theory up to $c=50\%$.

1. INTRODUCTION: THE EFFECTIVE SHAPE FACTOR

This work deals with the static magnetic permeability of randomly inhomogeneous materials, constituted of identically shaped soft particles. Our aim is to reach a better understanding of the parameters acting in the Effective Medium Theory (EMT), which is the most widely encountered among the Bruggeman-type laws used to describe the behaviour of such materials within the long-wavelength limit.

It is well known that solving Laplace's equation, in the case of an ellipsoidal particle (permeability μ_i) embedded in an homogeneous medium (permeability $\bar{\mu}$) when a uniform field H is applied, leads to the following expression of the internal field H_i :

$$H_i = \frac{\bar{\mu}}{\bar{\mu} + \bar{N}(\mu_i - \bar{\mu})} H_0 \quad (1)$$

Here, $\bar{N}$ is a diagonal tensor, named shape factor of the particle, the components of which along the three principal axes are N_α ($\alpha=x,y,z$). This dipolar approximation is commonly used to express that the composite medium is viewed as effectively homogeneous. In the case of a random dispersal of the ellipsoids axes along the three directions x, y, z , the EMT leads to the two following relations:

$$\frac{1}{3} \sum_i c_i \sum_{\alpha=x,y,z} \frac{\bar{\mu}}{\bar{\mu} + N_\alpha(\mu_i - \bar{\mu})} H_0 = H_0 \quad (2)$$

$$\sum_i c_i \frac{\bar{\mu}}{\bar{\mu} + \bar{N}(\mu_i - \bar{\mu})} H_0 = H_0 \quad (3)$$

The relation (2) means that the i^{th} class of particles (volumic fraction c_i , permeability μ_i) constituting the actual material acts in a same manner along each directions. The relation (3) (a Bruggeman-type law) shows that a single scalar shape factor $\bar{N}$ is associated with all the classes of particles; c_i and μ_i keep the same meaning as in relation (2). In the simplest case of a single class (permeability μ_1) of ellipsoids of revolution ($N_x + N_y + N_z = 1$, and $N_x = N_y$) dispersed in a non magnetic host medium ($\mu_2 = 1$), the equality between the two previous relations leads to the general result:

$$\bar{N} = \frac{2\delta + 3N_z(1 - N_z)}{6\delta + (3N_z + 1)} \quad \text{with} \quad \delta = \frac{\bar{\mu}}{\mu_1 - \mu} \quad (4)$$

This expression shows that the effective parameter $\bar{N}$ cannot be seen as a mean demagnetizing factor (in this case, one would get $\bar{N}=1/3$). Its values belong to the range $]0, 1/3[$, depending both on μ_1 and on $\bar{\mu}$ (or on the volumic fraction c_1). For particles dispersed at random, the result $\bar{N}=1/3$ is reached in the whole concentration range only for spheres, and for needles and flakes at the concentration $c=1$. In the low concentration range, and for μ_1 large enough, δ will be small and $\bar{N}$ will be quite independent of μ_1 ; then the two extremal values of N_z , that is $N_z < 1$ (needles) and $N_z > 0$ (flakes), will both lead to $\bar{N} \rightarrow 0$. The value of μ_1 has been shown to be very constant for $c < 50\%$, and its physical origin associated to spin rotation [1].

2. EXPERIMENTAL RESULTS

Figure 1: Experimental and theoretical variations of the effective shape factor $\bar{N}$. In the low concentration range, $\bar{N}$ is low as well as for needles as for flakes. $\bar{N}=1/3$ is reached only in the case of randomly dispersed spheres. The results given by the EMT (eq. (4)) have been obtained for the following shape factors: $N_z=0.75$ (flake-like particles), $N_z=0.01$ (needle-like particles); for the spheres, two situations have been simulated (see section 3).

Three types of composites have been studied. Each of them is made of well defined shaped particles: either spherically shaped (sendust, $\mu_1=61$, $N_z=1/3$), or needle-like shaped (iron, $\mu_1=61$, $N_z \approx 0.007$), or flake-like shaped (sendust, $\mu_1=26$, $0.8 < N_z < 0.6$). The above particles shape factors have been determined through optical observations. The experimental values of the effective demagnetizing factor $\bar{N}$ are shown in fig.1. They have been obtained by measuring $\bar{\mu}$ (the measurement technique is described in ref.[2]), and then by using relation (3). Three areas have been distinguished on the concentration axis, in order to allow an easier reading of the results. However the values of c at which the boundary areas take place must be taken only as indications. In the low concentration range the interparticles interactions are usually viewed as neglectible. In the medium concentration range the particles clustering is more pronounced, the cluster sizes increase with the volumic fraction c in magnetic matter. The higher limit of this second region corresponds roughly to the maximum filling fraction ($c=50-60\%$) of a random packing of equally-sized hard particles [3]. The high concentration range (third area), which starts with this filling coefficient, cannot be reached unless by applying high pressures on the composite. Such a treatment, which bends out of shape the particles, will certainly lead to modify the physical origin of the magnetic permeability μ_1 . In this paper, we will focus the discussion on the low and the medium concentration ranges defined above.

3. DISCUSSION AND CONCLUSION

Figure 1 shows the general fairly good agreement obtained between the experimental and the theoretical results, for a volumic fraction in magnetic matter up to $c=50\%$. The experimental values obtained for $c < 1$ (ie: for $\mu \neq 1$) give $\bar{N} < 1/3$, as predicted by relation (4). At higher concentrations, the experimental variation concerning the two types of composites made of non spherical particles seem to be consistent with their expected theoretical asymptotic behaviours towards $\bar{N}=1/3$. The values given to the shape factors N_z in relation (4) to fit these experimental results are in good accordance with the dimensions of the actual particles (see caption of figure1 and section 2). For the type of composites made of spherical particles, two situations have been simulated. The first one (fig.1, greasy dashed line), which modelises non clustering particles, is obtained for $N_z=1/3$ in relation (4). It leads to the constant value $\bar{N}=1/3$ throughout the concentration range. However, a better reproduction of the experimental results is reached when the magnetic matter is subdivided into isolated spheres and chains of spheres (fig.1, fine dashed line). Some percents of this kind of clusters lower the theoretical value of $\bar{N}$ from $1/3$ to 0.3 .

Other experimental results, got on composites made of YIG (with a dispersion in the shape of the magnetic particles), confirm the general character of relation (4).

The simplest EMT is suitable to describe, in a large concentration range, the experimental results presented above. However, the effective shape factor $\bar{N}$ (a parameter of the EMT) might not be confused with the mean shape factor of the particles. In the concentration range under study, the spatial randomization of the particles is sufficient to explain on its own the experimental values of the effective shape factor.

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