THE PUBLISHING HOUSE
OF THE ROMANIAN ACADEMY

PROCEEDINGS OF THE ROMANIAN ACADEMY, Series A,
Volume 11, Number 2/2010, pp. 137–141

DYNAMIC MODELING OF FERRITE NANOPARTICLE SYSTEMS IN RADIOFREQUENCY MAGNETIC FIELDS

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Today the most studied magnetic materials are the magnetic materials based on the nanoparticles in solid or liquid matrix. The knowledge of the ferrite nanoparticles in high frequency magnetic fields insures the possibility to know the behavior of the magnetic materials based on nanoparticles. The nanocomposites based on ferrite nanoparticles and polymer dielectric matrix can replace successfully the usual high frequency ferrites. This work proposes a study by simulation of the ferrite nanoparticles behavior in high frequency magnetic fields.

Key words: Simulation; Relaxation time; Relaxation process; Fines particles; Dipolar interaction.

1. INTRODUCTION

The scientific and technological importance of nanometric particles magnetic disperse systems is justified by the possibility to achieve some advanced magnetic materials, from natural nano-structures, to artificial nanostructures: ferrofluids, high-density magnetic registration environments, magnetic sensors, hard & soft magnetic nano-composites used as electro-technical materials at usual frequencies and high frequency… The magnetic relaxation processes in nanoparticle systems have a decisive role in the new magnetic materials behavior based on nanoparticles in dinamic magnetic fields, aiming to use them in radio-frequency and microwaves instead of usual materials.

Due to the complexity of the problem, treating irregular arranging of particles in systems with competing dipolar magnetic interactions in thermal equilibrium and nonequilibrium, we are forced to use stochastic simulation method of magnetic relaxation time, since present analytical methods can not cope equivalently with such systems. The stochastic simulation method is widely applied in statistical physics, since it is a powerful tool for the (in principle) exact numerical calculation of thermodynamic properties of interacting many-particle systems.

We propose a simulation model of the magnetic relaxation process for ferrite nanoparticles systems which lead to a medium relaxation time determination, then, using the Debye's theory [1] can be obtained the frequency dependence of the components' complex magnetic susceptibilities, according to relations:

$$\chi' = \chi_\infty + \frac{\chi(0) - \chi_\infty}{1 + \omega^2 \tau^2}, \tag{1}$$

$$\chi'' = [\chi(0) - \chi_\infty] \cdot \frac{\omega\tau}{1 + \omega^2 \tau^2}, \tag{2}$$

where $\chi(0)$ represents the measured value of magnetic susceptibility at low frequency or in static regime, χ_∞ is the measured magnetic susceptibility at high frequency, ω is the angular frequency and τ is the magnetic relaxation time.

Depending on the magnetic material type for which the nanoparticles are the base constituent: ferrofluid in which the nanoparticles are clustered in the particle chain, layer or nanocomposite with solid dielectric matrix, than the simulation model of the magnetic relaxation process in ferrite nanoparticles system can used one-dimensional, bi-dimensional and three dimensional simulation model.

In the case of an ensemble of many-ultrafine particles, when analyzing the magnetization and demagnetization, it is very important to consider the dipolar magnetic interaction between particles. When this interaction is included, the problem becomes very complex and, until now, there is not exactly known an analytic solution [2]. For the medium relaxation time determination we use the three dimensional simulation stochastic model [3]. In this model we consider a distribution of the nanoparticles dimensions. We take into account of the dipolar magnetic interaction between nanoparticles [5, 6].

We consider that the nanoparticle system is situated in an external magnetic field. The study of the behavior of the nanoparticle system starts when the action of the external magnetic field ends ($t = 0$) and when the magnetic moments are parallel with their anisotropy axis and with the direction of the magnetic field. For every nanoparticle of the system of which position is generated. It is given $\vec{r}_{ij}$ the positional vectors that unite the centre of the nanoparticle i with the centers of the nanoparticles neighbours j. The r_{ij} size of these vectors which represent the distances between nanoparticle i and neighbor nanoparticle j, for a random display of nanoparticles in the 3D volume, are generated by the transforming Box Mueller [7]:

$$r_{ijk} = r_{ijkmed} + \sigma_{ijk}\sqrt{-\ln(r\text{and}1)}\cdot\left[\cos(2\pi r\text{and}2) + \sin(2\pi r\text{and}2)\right]. \tag{3}$$

In eq. (3) σ_{ijk} is the distribution variance of distances between nanoparticles and k being the order of the nearest neighbors for a given particle i. For simplicity, we express the distribution variance of distances as:

$$\sigma_{ijk}=r_{ijk\text{med}}\cdot v_r, \tag{4}$$

where v_r is variance parameter and $r_{ijk\text{med}}$ is the r_{ijk} mean value.

Then it can be simulated the energy barriers, the probability of the nanoparticle to pass from a stabile state to another stabile state, then their relaxation time.

2. STUDY OF THE FERRITE NANOPARTICLES' BEHAVIOR IN HIGH FREQUENCY MAGNETIC FIELDS

The simulations were performed on a system with spherical fine particles of magnetite with uniaxial anisotropy, anisotropy constant K = 19,000 J/m^3, spontaneous magnetization $M_s = 4.7\cdot10^5$ A/m, of which most probable value of the particle diameter $d_m = 9.848$ nm. The particle's first order neighbors for i-th particle, for very high density systems, are the neighbors that are placed at one diameter distance from the central point of the i-th particle (Fig. 1). Using the volume fraction definition, if we note with:

$$f = \frac{V_{powder}}{V_{test}} \tag{5}$$

the volume fraction of particles, for a high-density (tangent particles one with another) system we can write [8] by Garcia relation:

$$f = 0.32\cdot f_0, \tag{6}$$

$$f_0 = \frac{8K\pi}{\mu_0 M_s^2}. \tag{7}$$

Using (5), (6) and (7) we obtain the number of k order neighbors by using the following relation:

$$n_k = 0.32(2k+1)^3 f_0. \tag{8}$$

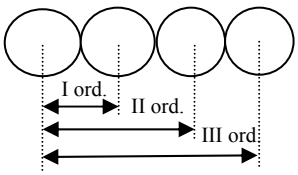


Fig. 1 – Orders of the nearest neighbors for very high-density systems.

For the diluted systems, we keep the number of neighbors of I, II and III order and we use a parameter D – distance between two particles one next to another (not between their central points), which is bigger as the system is more diluted (Fig. 2).

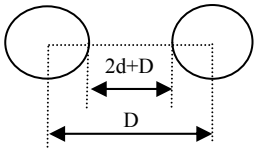
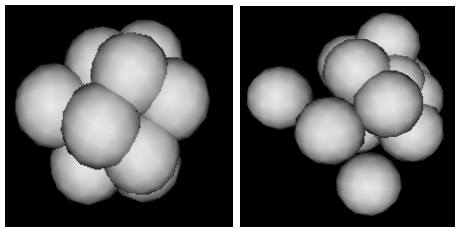


Fig. 2 – The first order of the nearest neighbors for a diluted system.

In the first approximation we considered that the particles are of the same diameter and the temperature of system is 300 K.

We simulated the nano-particle arrangement from the first two neighbourhood ranks, for different values of the nano-particle concentration and for different values of the distribution variance of distances between nano-particles σ_{ijk}. In Fig.3 are simulated the disposals of the 14 nano-particles of rank I around a given nano-particle, for different values of the distance variance parameter v_r.



v_r=0.2 v_r=0.4

Fig. 3 – Disposal of rank I nano-particles into a sample with concentration of $9.9 \cdot 10^{23}$ part/m^3, for different values of the distance variance parameter vr.

These calculations, using the eq. (1) and (2) are performed under the hypothesis of weak or strong dipolar interaction. In the weak interaction case, which corresponds to a concentration of the particles of $2.24 \cdot 10^{23}$ part/m^3, the distance between the i-th particle and the nearest neighbors is $2d_m$, $4d_m$ to the next nearest neighbors, and $6d_m$ to the third order particles. For a strong interaction, which corresponds to a concentration of the particles of $1.16 \cdot 10^{24}$ part/m^3 the distance between the i-th particle and the nearest neighbors is d_m, $2d_m$, and $3d_m$ respectively. The frequency dependence of the complex magnetic susceptibility components is seeing in Figs. 4-5.

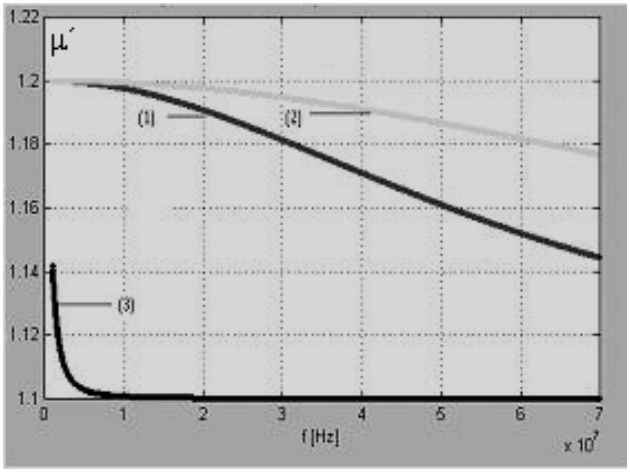


Fig. 4 – The frequency dependence of the real magnetic susceptibility at 293 K (spherical particles with identical volume): 1) – without interaction, 2) – with interaction, 3) – very strong interaction.

Now, we consider that particle dimensions have a lognormal distribution. For the generation of random values of particle diameters, we used the algorithm presented in [4, 6]. The method of generating a lognormal distribution is based on the relation:

$$d_i = \mathrm{e}^{\sigma_{\ln d} \cdot u_i + \ln d_0} \tag{9}$$

In eq. (9) u_i are the values of a normal distribution on (0,1), with the mean equal to ln d_0 and the variance equal $\sigma^2_{\ln d}$.

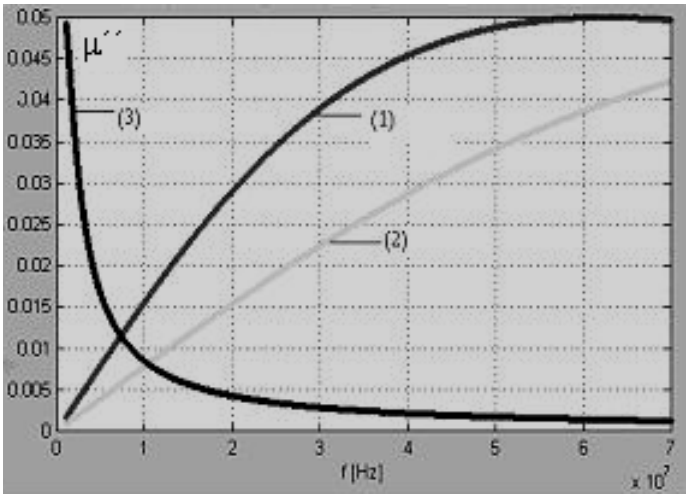


Fig. 5 – The frequency dependence of the imaginary magnetic susceptibility at 293 K (spherical particles with identical volume): 1) – without interaction, 2) – with interaction, 3) – very strong interaction.-

In eq. (9) we have considered that the mean of the distribution is $\ln d_0 = -18.4409$ and the variance is $\sigma^2_{\ln d} = 0.0025 \left(\sigma_{\ln d} = 0.05\right)$. The most probable value of the particle diameter $d_m = 9.848$ nm corresponds to a volume of $5 \cdot 10^{-25}$ m^3.

In the simulation algorithm, we first generate the random variables, which are used in the model, and then we computed the energy barriers of each particle and relatation time. Finally, from eqs. (1) and (2) the frequency dependence of the complex magnetic susceptibility components – see in Figs. 6 and 7.

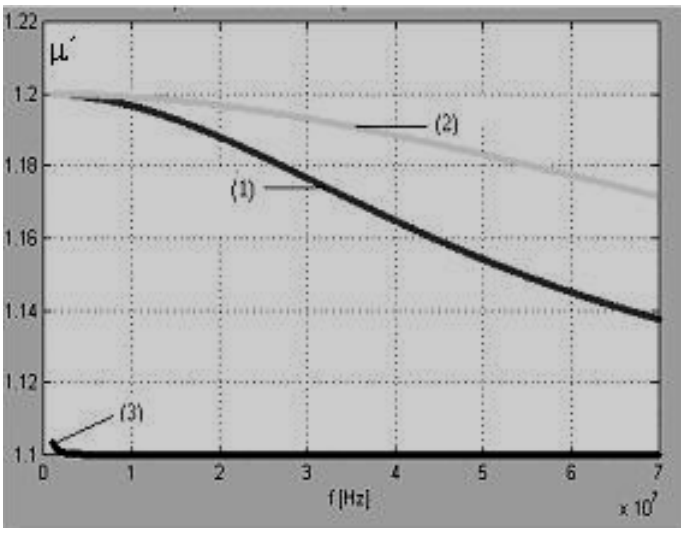


Fig. 6 – The frequency dependence of the real magnetic susceptibility at 293 K (spherical particles with lognormal volume distribution (with the variance 0.0025): 1) – without interaction, 2) – with interaction, 3) – very strong interaction.

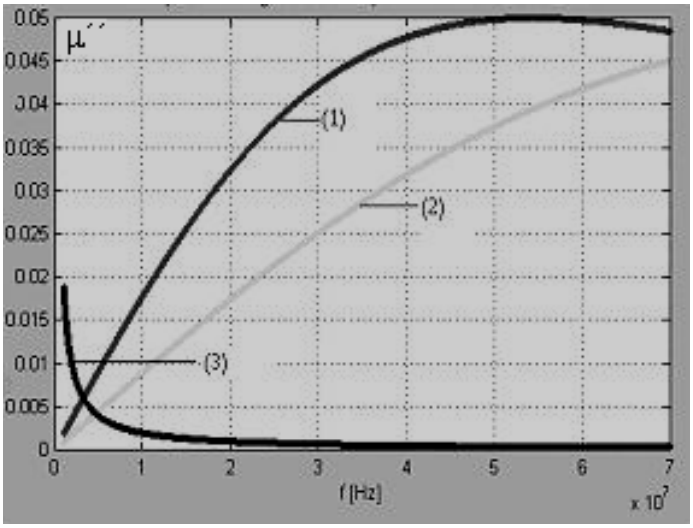


Fig. 7 – The frequency dependence of the imaginary magnetic susceptibility at 293 K (spherical particles with lognormal volume distribution (with the variance 0.0025): 1) – without interaction, 2) – with interaction, 3) – very strong interaction.

3. CONCLUSIONS

With the increasing interest in realising new magnetic materials, the magnetic behaviour of the nanoparticles disperse systems became an important problem either from the experimental point of view or from the theoretical one.

In this paper it has been carried out a simulation to determine the dependence of the complex magnetic susceptibilities components for a fine particles system on frequency based on the three-dimensional model for relaxation process [3], similar to real situation. In this model it has been considered that the particles are randomly arranged in a volume. Considering the dipolar magnetic interaction between particles it can be observed an influence for the dependence between complex magnetic susceptibilities components and the frequency.

For a weak interaction (concentration = $2.24 \cdot 10^{23}$ part/m^3) it results a removal of the maximum imaginary component of the complex susceptibility to a high frequency. For a very strong interaction, the maximum imaginary component of the complex susceptibility moves to a low frequency.

These conclusions are in accordance with the majority experimental data for nanoparticles systems [9, 10].

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Received November 13, 2009