Theoretical studies to elucidate the influence of magnetic dipolar interactions occurring in the magnetic nanoparticle systems, for biomedical applications

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Abstract. In recent years, the study of magnetic nanoparticles has been intensively developed not only for their fundamental theoretical interest, but also for their many technological applications, especially biomedical applications, ranging from contrast agents for magnetic resonance imaging to the deterioration of cancer cells via hyperthermia treatment. The theoretical and experimental research has shown until now that the magnetic dipolar interactions between nanoparticles can have a significant influence on the magnetic behaviour of the system. But, this influence is not well understood. It is clear that the magnetic dipolar interaction intensity is correlated with the nanoparticle concentration, volume fraction and magnetic moment orientations. In this paper, we try to understand the influence of magnetic dipolar interactions on the behaviour of magnetic nanoparticle systems, for biomedical applications. For the model, we considered spherical nanoparticles with uniaxial anisotropy and lognormal distribution of the sizes. The model involves a simulation stage of the spatial distribution and orientation of the nanoparticles and their easy axes of magnetic anisotropy, and an evaluation stage of the Néel relaxation time. To assess the Néel relaxation time, we are going to discretise and adapt, to the local magnetic field, the Coffey analytical solution for the equation Fokker-Planck describing the dynamics of magnetic moments of nanoparticles in oblique external magnetic field. There are three fundamental aspects of interest in our studies on the magnetic nanoparticles: their spatial & orientational distributions, concentrations and sizes.

1. Introduction

The physics of magnetic nanoparticles systems is of special scientific interest, and has attracted much attention in recent years due to the potential for various applications, from technology to medicine. In the literature, there are many theoretical and experimental studies, often contradictory, about the influence of magnetic dipolar interactions on the magnetic behaviour of the nanoparticle systems, of which we are going to point out some of them to have an image on the matter.

Mössbauer spectroscopy measurements [1], [2], [3] on weakly interacting $\gamma$-Fe$_2$O$_3$ nanoparticles showed a decrease of the relaxation time with increasing particle interaction. Other publication [4] reported on the specific absorption rate (SAR) of a maghemite-based ferrofluid, measured at 315 K, 3 kA/m, and 109 kHz. The magnetic characterization of three different concentrations of this ferrofluid
revealed the decrease of initial susceptibility values (liquid ferrofluid) and Néel relaxation times, $\tau_N$, with increasing concentration.

In the other hand, the AC susceptibility measurements at low frequency for dynamic properties of a system constituted by small iron grains dispersed in an amorphous alumina matrix [5] show that the relaxation time increases with increasing particle concentration. The same experimental results were found in case of $\gamma$-Fe$_2$O$_3$ nanoparticles dispersed in a polymer [6] investigated by means of Mössbauer spectroscopy measurements and AC susceptibility measurements in a large frequency range (10<$\nu$<104 Hz). An increase of the effective energy barrier with increasing interaction strength has been unambiguously seen in many other cases.

The previous theoretical attempts to consider the magnetic dipolar interactions were based on the Néel-Brown model [7]. The first attempt to introduce interactions in the Néel–Brown model was made by Shtrikmann and Wohlfarth who, by using a mean field approximation, predicted a Vogel–Fulcher law for the relaxation time in the weak interaction limit [8].

A more general approach was developed by Dormann et al. [5]. Taking into account the dipole-dipole interaction, the model predicted two magnetic regimes, according to the interaction strength. Firstly, for negligible or very weak interactions, the predictions matched the Néel-Brown model of superparamagnetism. Secondly, for appreciable dipolar interactions, the individual energy barriers were modified by their strength, the case being referred to as superparamagnetic regime modified by interactions. Here, an increase of Néel relaxation time with the strength of dipolar interactions (e.g., increasing particle concentration or decreasing particle distances) was predicted.

Note that the Shtrikman and Wohlfarth model [8] led to the same conclusions as the Dorman et. al. model [5]. In a third model, proposed by Mørup and Tronc [1] for the weak interaction limit, the opposite dependence of Néel relaxation time (decrease with increasing strength of the interactions) has been predicted. In order to untangle the apparent contradiction, Mørup [1] suggested that two magnetic regimes, governed by opposite dependencies of the Néel relaxation time, occur in the systems of interacting fine particles. At high temperatures and/or for weak interactions, the Néel relaxation time decreases with increasing interactions. In contrast, at high temperatures and/or for strong interactions, a transition occurs from the superparamagnetic state to a collective state, having most of the features of typical glassy behaviour. In this case, a freezing process occurs and the Néel relaxation time increases with increasing interactions. In the last few years, this collective state and its glassy properties are some of the most studied issues regarding the assemblies of strongly interacting nanoparticles [9], [10].

The models found in the literature, which take into consideration the magnetic dipolar interactions, do not provide a rigorous theoretical treatment of the subject and are based on a series of approximations, some of them being mathematically unjustified [1]. Moreover, the problem of their contradictions [1], [5], [11], [12] is much discussed, but without a conclusion that clearly leans toward a model or another one.

Regarding the action of the external magnetic field, the Néel-Brown model (basis of the models mentioned above) considers that the nanoparticle anisotropy axes are aligned with the external magnetic field. This condition is valid only in the intense magnetic fields. In the general case, at not very intense magnetic fields, and when the nanoparticles are dispersed in solid matrix, the anisotropy axes of nanoparticles are randomly distributed and the use of Néel-Brown model is no longer justified, although it is the starting point of introducing the magnetic dipolar interactions, because the local magnetic dipolar field has usually an oblique orientation to the anisotropy axis of the nanoparticle. Coffey et al. studied the external oblique magnetic field influence on the Néel relaxation time [13], succeeding to obtain a solution to the Fokker-Planck equation for nanoparticle magnetic moments dynamics, based on the Kramer’s theory [13]. This solution includes the solution of Néel-Brown model as particular case.

With regard to the high degree of complexity of the problem, it seems useful to consider the “numerical experiments”. For a system consisting of spherical magnetic nanoparticles with uniaxial magnetic anisotropy, we have to study the Coffey’s solution, which we discretize to introduce
the nanoparticle magnetic dipolar interactions. The 3D random distribution of the nanoparticle positions and magnetic anisotropy easy axes will be numerically simulated. The model allows obtaining the Néel relaxation time for each nanoparticle. The Néel relaxation time for the system is found through mediation. Our model is particularly suitable for predicting and understanding the Néel relaxation time trends in an interacting magnetic nanoparticle system, with a diameter of up to a few tens of nm.

2. About the magnetic dipolar interaction
A monodomain nanoparticle is in a state of uniform magnetization for any field applied. Each monodomain nanoparticle is characterized by a magnetic moment, which usually is given by [14]:

$$\mu_i = M_i v_i \hat{\mu}_i$$  \hspace{1cm} (1)

where $M_i$ is the spontaneous magnetization, $v_i$ is the particle volume and $\hat{\mu}_i$ is the unit vector of the magnetic moments.

Let $\hat{\mu}_i$ be the magnetic moment of the nanoparticle $i$, and $\hat{\mu}_j$ the magnetic moment of the nanoparticle $j$ with uniaxial anisotropy. The energy of the magnetic dipolar interaction between two nanoparticles ($i$ and $j$) is given by the relation [14]:

$$E_{ij} = \frac{\mu_0}{4\pi} \frac{\mu_i \mu_j}{r_{ij}^3} \left[ \hat{\mu}_i \cdot \hat{\mu}_j - 3 (\hat{\mu}_i \cdot \hat{r}_{ij}) (\hat{\mu}_j \cdot \hat{r}_{ij}) \right]$$ \hspace{1cm} (2)

where $r_{ij}$ is the distance between the centres of those two nanoparticles, $\hat{r}_{ij}$ is the versor of the direction connecting the nanoparticles $i$ and $j$, $\hat{\mu}_i$ and $\hat{\mu}_j$ are the unit vectors of the magnetic moments of the nanoparticles $i$ & $j$, and $\mu_0$ is the vacuum magnetic permeability.

Due to the long range character of the magnetic dipolar interaction, the interaction effect can be significant. If on the nanoparticle system is also acting an external magnetic field, then a local magnetic field is acting on each nanoparticle, which is the vectorial sum between the external magnetic field applied ($\vec{H}_{\text{ext}}$) and the internal magnetic dipolar field ($\vec{H}_{\text{id}}$) determined by the magnetic dipolar interactions among the nanoparticles.

$$\vec{H}_i = \vec{H}_{\text{ext}} + \vec{H}_{\text{id}}$$ \hspace{1cm} (5)

It will be of great help to develop a model of the interactions between uncoated and coated nanoparticles.

3. Néel relaxation time of the nanoparticles interacting in local oblique magnetic field
The relaxation of magnetic nanoparticle systems is important for their applications. The Néel relaxation process is the solid state mechanism that occurs inside the nanoparticle, corresponding to the magnetic moment rotation between two equilibrium positions. The nanoparticle is fixed in space.

For the relaxation time assessment, we adapt an analytical Coffey solution for external magnetic fields [15], [13] - mean field method (eq. 5 and 4), in order to take into account the magnetic dipolar field on a nanoparticle. Below, we will use the notations:
\[ h_i = \frac{\mu_0 M_i H_i}{2K_{\text{eff}}} \] and \[ \sigma_i = \frac{K_{\text{eff}} V_i}{k_B T} \] (6)

where \( K_{\text{eff}} \) is the effective magnetic anisotropy constant of the \( i \)th nanoparticle, and \( H_i \) is assessed using the equations (4) and (5). The free diffusion time of magnetisation \( \tau'_{0N} \) for low damping \([16]\) is:

\[ \tau'_{0N} = \frac{\nu_i M_s}{2\gamma \alpha k_B T} \] (7)

where \( \alpha \) is the damping constant, and \( \gamma \) is the gyromagnetic ratio.

For most systems of ferro- and ferrimagnetic nanoparticles, the magnetic damping constant \( \alpha \) ranges around the low limit (\( \alpha \ll 1 \)) [17].

The Coffey analytical calculation is based on Kramer’s theory \([13]\) and in agreement with the numerical calculation and experimental results \([18]\). This calculation shows the relaxation time dependence on the magnetic damping constant \( \alpha \).

In these conditions, the relaxation time relation for an oblique magnetic field is \([15]\):

\[ \tau'_{\text{N(oblique field)}} = \frac{4\pi \tau'_{0N}(S_{11}^i + S_{12}^i)}{\sqrt{c_{i1}^{(1)}c_{i2}^{(1)} \cdot e^{-\Delta V_{i12}} + \sqrt{c_{i1}^{(2)}c_{i2}^{(2)} \cdot e^{-\Delta V_{i21}}}}} \] (8)

where \( \Delta V_{i12} \) and \( \Delta V_{i21} \) are the normalized energy barriers for magnetic moment re-orientations. In eq. (8):

\[ c_{i1}^{(p)} = 2\sigma_i (\cos 2\theta_{ip} + h_i \cos(\theta_{ip} - \Psi_i) \), c_{i2}^{(p)} = 2\sigma_i (\cos^2 \theta_{ip} + h_i \cos(\theta_{ip} - \Psi_i) \), with p=1,2

where \( \theta_{ip} \) are the solutions of the transcendental equation \( \sin 2\theta = 2h_i \sin(\Psi_i - \theta_i) \).

If \( h_i < h_{ci}(\Psi_i) < 1 \) (\( \Psi_i \) being the angle between \( \vec{H}_i \) and the easy anisotropy axis of the \( i \)th nanoparticle).

\[ \cos \theta_{i1,2} = \pm 1 \pm \frac{h_i^2}{2} \sin^2 \Psi_i + h_i^2 \sin^2 \Psi_i \cos \Psi_i \pm \frac{h_i^4}{16} \left( 1 + 3 \cos 2\Psi_i \right) \sin^2 \Psi_i + \frac{h_i^6}{32} \left( 19 + 156 \cos 2\Psi_i - 19 \cos 4\Psi_i \right) \sin^2 \Psi_i + \ldots \] (10)

\[ \Delta V_{i12} = \sigma_i \left[ 1 - 2h_i \left( \sin \Psi_i - \cos \Psi_i \right) + h_i^2 \sin^2 \Psi_i \left( \cos \Psi_i - \sin \Psi_i \right) + h_i^3 \sin^2 2\Psi_i + \frac{h_i^5}{32} \sin 2\Psi_i \left( 7 \cos 3\Psi_i - 3 \cos 3\Psi_i + 7 \sin 3\Psi_i - 3 \sin 3\Psi_i \right) + \frac{h_i^6}{2} \sin^2 2\Psi_i + \ldots \right] (11) \]

\[ \Delta V_{i21} = \sigma_i \left[ 1 - 2h_i \left( \sin \Psi_i + \cos \Psi_i \right) + h_i^2 \sin^2 \Psi_i \left( \cos \Psi_i + \sin \Psi_i \right) + h_i^3 \sin^2 2\Psi_i + \frac{h_i^5}{32} \sin 2\Psi_i \left( 7 \cos 3\Psi_i - 3 \cos 3\Psi_i + 7 \sin 3\Psi_i + 3 \sin 3\Psi_i \right) + \frac{h_i^6}{2} \sin^2 2\Psi_i + \ldots \right] (12) \]

\[ S_{i1,2} = \sigma_i \sqrt{h_i} \sin \Psi_i \left[ \frac{16 - 104}{3} h_i \sin \Psi_i + h_i^2 \left( 1 - 2 \cos 2\Psi_i \right) + \frac{h_i^3}{2} \sin \Psi_i \left( 45 + 51 \cos 2\Psi_i \right) + \ldots \right] \pm \ldots \] (13)

It is defined the normalized effective energy barriers for nanoparticle magnetic moment re-orientations

\[ \Delta V_{\text{eff}} = \frac{\Delta V_{i12} + \Delta V_{i21}}{2} \] (14)

The relation (8) can be written:
\begin{align*}
\frac{1}{\tau_{\text{N(oblige, field)}}} &= \frac{S_{11} \cdot S_{12} \sqrt{c_{11}} \sqrt{c_{12}}}{4\pi \varepsilon_0 N (S_{11} + S_{12})} e^{-\Delta V_{112}} + \frac{S_{11} \cdot S_{12} \sqrt{c_{11}} \sqrt{c_{12}}}{4\pi \varepsilon_0 N (S_{11} + S_{12})} e^{-\Delta V_{211}} = f_{12} e^{-\Delta V_{112}} + f_{21} e^{-\Delta V_{211}} 
\end{align*}

(15)

where \( f_{12} \) and \( f_{21} \) are the attempt frequency factors.

The equations (8)-(13) indicate that the dipolar interactions have a complex effect, which leads to the misalignment of the moments with respect to the local easy axis.

4. Results and discussions

We simulate the random 3D arrangement of the nanoparticles in a face-centred cubic lattice. Among all the possible positions, the particles have been located in randomly selected suitable locations, according to the ratio between the volumes of the considered nanoparticles and the whole volume of the simulation box. The periodic boundary conditions have been applied on all the sides of the simulation box. A random 3D distribution of the anisotropy easy axes is also used. We are going to study a system of 1000 uncoated magnetite nanoparticles, with uniaxial magnetic anisotropy, effective magnetic anisotropy constant of \( 2.5 \times 10^4 \) J/m\(^3\) and easy axes of anisotropy, with random distribution. We consider spontaneous magnetization of \( 4.46 \times 10^5 \) A/m, simulation temperature \( T = 298.15 \) and external magnetic field intensity of 15 kA/m, typical value for magnetic hyperthermia applications.

As can be seen from the equation (4), the magnetic dipolar interactions can have an influence determined by their nanoparticle concentrations and sizes.

4.1 The long range and anisotropic character of magnetic dipolar interaction

The long range and anisotropic character of the magnetic dipolar interaction is reflected in the fact that the magnetic dipolar field, respectively the magnetic dipolar energy of a nanoparticle, depends on its neighbours’ contributions. To highlight this character, we will study how the strength of the average magnetic dipolar field depends on the number of nanoparticles found in the system, at the constant parameters of the simulation. The local magnetic dipolar field on the nanoparticle (4) will be simulated either using the Ewald method [19] or by direct summation. We are going to consider nanoparticles of 12 nm with the volume fraction \( f = 0.24 \), in the absence and in the presence of external magnetic field. The results are shown in the Figures 1 and 2.

![Figure 1](image1.png)

**Figure 1.** Average local magnetic dipolar field intensity versus number of nanoparticles

a) by direct summation, b) by applying the Ewald summation method
In Figure 1, we can see that the average local magnetic dipolar field intensity varies with the number of nanoparticles in the system. If over the magnetic dipolar field is superimposed an external magnetic field (whose magnitude, in this case, is 15 kA/m, typical for magnetic hyperthermia), it can be seen that the magnetic dipolar field influences are still visible – Figure 2.

4.2 The influence of magnetic dipolar interactions on Néel relaxation time

We take into account three types of nanoparticles, i.e. small-sized (6 nm), medium-sized (12 nm) and large-sized (18 nm). In the first instance, we consider that the sizes have no dispersion. We will assess the Néel relaxation time on each nanoparticle, using the relations (8)-(13), at different values of the nanoparticle volume fraction, ranging from 0.02 to 0.3. We obtain an average value of the Néel relaxation time. The results are shown in Figure 3. It is noted that, for small and large sizes (Figures 3a and 3c), the average Néel relaxation time increases with increasing concentration, whereas, for medium-sized nanoparticles, the average Néel relaxation time decreases with increasing concentration. At small sizes of the nanoparticles, this increase is very sensitive, so that the Néel relaxation time can be considered to be approximately constant. At large sizes, the increase should be taken into account.

**Figure 3.** Average Néel relaxation time versus volume fraction of nanoparticles without radii dispersion

a) small-sized; b) medium-sized; c) high-sized nanoparticles
To understand this dependence, we will simulate the local magnetic field on the nanoparticle and its average throughout the system, as well as the average normalized energy barriers and the effective average normalized energy barriers of nanoparticle reorientation. In Figure 4, we can see that the average values of the local magnetic dipolar field a) and the local magnetic dipolar field b) increase with increasing concentration of nanoparticles for all three categories of nanoparticle sizes.

**Figure 4.** Average local magnetic dipolar intensity versus the volume fraction of nanoparticles a) small-sized; b) medium-sized; c) high-sized nanoparticles

Regarding the dependence of the average normalized energy barriers and the effective average normalized energy barriers on the volume fraction, Figure 5, Figure 6, we can see their decrease with increasing concentration for all the size categories of nanoparticles.

**Figure 5.** Average normalized energy barriers versus volume fraction of nanoparticles

Analysing the Figures 3-5, we can conclude that, in case of nanoparticles without size dispersions or with small dispersions, for medium sizes, the decrease of energy barriers with increasing concentration influences significantly the decrease of relaxation time with increasing concentration. In this case, the dependence of the attempt frequency factors on the local magnetic field, in the relation (15), has no major influence. In case of very small and quite large nanoparticles, the values of these frequency factors change the shape of the relaxation time dependence on the nanoparticle concentration. So, their approximation through constant values, as appearing in most models in the literature, is not very appropriate.
Moreover, we keep the nanoparticle volume fraction at a constant value and assess the Néel relaxation time according to the standard deviation parameter of nanoparticle diameters – Figure 7.

We consider lognormal distribution of the diameters, with the standard deviation $\sigma_d$, where $\sigma$ is the standard deviation parameter. If the nanoparticle sizes are dispersed, the Néel relaxation time increases sharply with increasing standard deviation parameter of the nanoparticle diameters - Figure 7. We keep the standard deviation for size distribution at a constant value, for example $0.1\,d_m$, and assess the dependence of the Néel relaxation time on the nanoparticle volume fraction. It is noted that the size dispersion does not affect the type of Néel relaxation time dependence on the nanoparticle concentration, but only the values – Figure 8.
5. Conclusions

In this paper, we tried to clarify the effect of magnetic dipolar interactions on the Néel relaxation time for nanoparticle systems with medical applications, i.e. not very concentrated and not subjected to very intense external magnetic fields. The Néel relaxation time has been assessed by using the Coffey solution in oblique magnetic field, discretized and adapted to local magnetic field on the nanoparticle. The 3D random distribution of the nanoparticle positions and the random distribution of the easy axes anisotropy orientations were obtained by numerical simulation.

Considering the nanoparticle sizes without dispersion or with small dispersion, we found the fact that, in case of small-sized (≤ 6 nm) and large-sized (≥ 18 nm) nanoparticles, the average Néel relaxation time increases with increasing concentration, whereas, in case of medium nanoparticle sizes, the average Néel relaxation time decreases with increasing concentration. The increase of Néel relaxation time with increasing nanoparticle concentration is sensitive at small sizes, so it can be considered approximately constant, but this increase should be taken into account at large sizes of the nanoparticles. In case of medium sizes, the influence of the energy barrier variation with the concentration prevails, so the attempt frequency factors have no major influence. In case of small and large sizes, the attempt frequency factors dependence on concentration (through the local magnetic field on particle) and on nanoparticle sizes, can overturn the concentration dependence of the Néel relaxation time, being significant at large nanoparticle sizes.

If the nanoparticle sizes show dispersion, the Néel relaxation time increases strongly with increasing standard deviation parameter of the nanoparticle diameters, the more strongly the larger are the nanoparticle sizes. Also, we found that the size distribution does not affect the type of Néel relaxation time dependence on the nanoparticle concentration, but only the values.

The study presents a theoretical tool to control the parameters influencing the dynamics of magnetic moments of the nanoparticle systems, with biomedical applications, and not only.

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