

# Influence of Damping Constant on Models of Magnetic Hyperthermia

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In magnetic hyperthermia, the effectiveness for tumour cell destruction is measured by the specific loss power. Theoretically, within the medically accepted ranges for the amplitude and frequency of the applied magnetic field, if energy losses in nanofluids occur through magnetic relaxation processes, the specific loss power is calculated based on the linear response theory. In this theory, the specific loss power depends on the effective magnetic relaxation time of the colloidal nanoparticle system which involves either the Brownian relaxation time or the Néel relaxation time. All theoretical approaches to the Néel relaxation time show that it depends directly on the diffusional relaxation time and inversely on the smallest non-vanishing eigenvalue of the Fokker–Planck equation, where the damping constant is expressed one way or another by a value, generally unknown, which in most cases is approximated. This paper shows through a numerical experiment how the damping constant influences the specific loss power, referring to some benchmarks on how to choose the most accurate value of this constant in the case of magnetite nanoparticles, mostly used in magnetic hyperthermia applications. Following an uninspired choice of the damping constant value, the simulated or calculated data can deviate from the experimentally determined data, even if, in general, the model is correct and as close as possible to reality.

topics: magnetic hyperthermia, specific loss power, damping constant, numerical experiment

## 1. Introduction

The phenomena of magnetic relaxation in nanoparticle systems are intensively investigated today, especially for their biomedical implications, the most important of which being the cancer therapy by hyperthermia using magnetic nanofluids [1–3].

The effectiveness of a colloidal system of magnetic nanoparticles (MNPs), when converting the alternating magnetic field energy into heat able to rise the local temperature in the tumour tissue, is of major importance for the cancer therapy through magnetic hyperthermia. The system usually consists of MNPs dispersed in an aqueous medium, also known as a nanofluid. The heat transfer from the alternating magnetic field, whose parameters (amplitude, frequency) are ranging within medically accepted limits [4], to the tissue loaded with properly functionalized MNPs can be carried out by various mechanisms, depending on the type and size of the MNPs. In the case of nanoparticles with a magnetic oxide (usually ferrites) with an average size of less than 30 nm (currently considered to be the most suitable for such purposes), two heat transfer mechanisms must be considered [5]: magnetic hysteresis losses and magnetic relaxation processes. The effective mechanism depends on a relationship

between the magnetic relaxation time  $\tau$ , and the inverse of the alternating field frequency  $1/f$ , which defines the time window  $\tau_M$  of magnetic excitation ( $\tau_M$  can also be a measurement time window). In this regard, two regimes should be mentioned here [5]: a static regime  $\tau \gg \tau_M$ , wherein the main heat transfer is made by magnetic hysteresis losses and a dynamic regime  $\tau \ll \tau_M$ , wherein the main heat transfer is made by magnetic relaxation phenomena. The relaxation time depends on the volume of MNPs and, in the case of finite-size MNPs, on polydispersity. Both heat transfer mechanisms mentioned above can contribute: the first one for nanoparticles larger than a critical size and the second one for finer nanoparticles.

The effect of an external magnetic field on the longitudinal relaxation time of a nanoparticle system with uniaxial magnetic anisotropy can generally be studied by calculating the smallest non-vanishing eigenvalue (escape rate) using the Fokker–Planck equation, for the evolution in time of the magnetic moment density orientation [6]. The first study of this kind was carried out by Brown [7], whence the Brownian relaxation time model resulted, besides the simple Néel relaxation time model. A more general perturbation approach of the simple Néel expression is introduced by certain

models, such as the model of Aharoni and Eisenstein [8, 9], the model of Jones and Srivastava [10] and the model of Bessais et al. [11]. Later, Coffey studied the effect of an oblique magnetic field on the magnetic nanoparticle system [12] and, recently, the effect of a randomly oriented field has been studied [6]. Another theoretical approach to magnetic nanoparticle systems has been made by static and time-dependent micromagnetic simulations on various nanoparticle configurations [13].

The damping constant appears in all the expressions and models related to the Néel relaxation time. This paper aims through a numerical experiment to study how the value of this constant influences the specific loss power (SLP) as well as to constructively comment on how to choose this value, so as to be as close as possible to physical reality.

## 2. Influence of damping constant on SLP

A monodomain nanoparticle is in a uniform magnetisation state for any field applied. Each monodomain nanoparticle is characterised by a magnetic moment  $\mu_i$  which usually is given by [14]:

$$\boldsymbol{\mu}_i = M_s \nu_i \hat{e}_i, \quad (1)$$

where  $M_s$  is the spontaneous magnetisation,  $\nu_i$  is the particle volume and  $\hat{e}_i$  is the unit vector of the  $i$ -th magnetic moments.

The process by which all nanoparticles return to thermodynamic equilibrium is called the magnetic relaxation. In a system with ultrafine magnetic nanoparticles, the alignment of the magnetic moments in the direction of the external field can occur in two distinct ways [15]. In the first case, the magnetic moments of the monodomain nanoparticles remain fixed and under the action of the field, each nanoparticle rotates so that the orientation of the magnetic moment approaches the direction of the field. In the second case, the magnetic moment of the nanoparticle rotates while the nanoparticle remains fixed. Due to these two possibilities, two relaxation processes occur: the Brown relaxation, correlated with the nanoparticle rotation, and the Néel relaxation, correlated with the magnetic moment rotation inside the nanoparticle. The effective relaxation time for the  $i$ -th nanoparticle can be described as follows [16]:

$$\frac{1}{\tau_{\text{eff}}^i} = \frac{1}{\tau_N^i} + \frac{1}{\tau_B^i}, \quad (2)$$

where  $\tau_N^i$  is the Néel relaxation time and  $\tau_B^i$  is the Brownian relaxation time.

For spherical particles, the Brownian relaxation time is usually described by [17]:

$$\tau_B^i = \frac{3\nu_H^i \eta}{k_B T}, \quad (3)$$

where  $k_B$  is the Boltzmann constant,  $T$  is the temperature,  $\nu_H^i$  is the hydrodynamic volume of the  $i$ -th nanoparticle and  $\eta$  is the dynamic viscosity coefficient.

The most important feature of the magnetic dipolar interaction is its long range character. In the presence of an external magnetic field, each nanoparticle is affected by a local magnetic field. This field is the vectorial sum of the external magnetic field applied  $\mathbf{H}_{\text{ext}}$  and the internal magnetic dipolar field  $\mathbf{H}_{\text{id}}$  determined by the magnetic dipolar interactions among the nanoparticles [14], i.e.,

$$\mathbf{H}_i = \mathbf{H}_{\text{ext}} + \mathbf{H}_{\text{id}}, \quad (4)$$

where

$$\mathbf{H}_{\text{id}} = \frac{1}{4\pi} \sum_{j,j \neq i} \frac{\mu_j}{r_{ij}^3} [3\hat{r}_{ij}(\hat{e}_i \cdot \hat{r}_{ij}) - \hat{e}_j], \quad (5)$$

where  $r_{ij}$  is the distance between the centres of two nanoparticles,  $\hat{r}_{ij}$  is the versor of the direction connecting the  $i$ -th and  $j$ -th nanoparticle, and  $\hat{e}_i$  is the unit vector of the  $i$ -th magnetic moments.

An analytical Coffey solution for external magnetic fields [12] can be adapted as the Néel relaxation time in order to take into account the magnetic dipolar field acting on a nanoparticle. Further, it can be discretized [14], working at the nanoparticle level. We use the following notations:

$$h_i = \frac{\mu_0}{2} \frac{M_s H_i}{K_i^{\text{eff}}} \quad (6)$$

and

$$\sigma_i = \frac{K_i^{\text{eff}} \nu_i}{k_B T}, \quad (7)$$

where  $\mu_0$  is the vacuum magnetic permeability,  $K_i^{\text{eff}}$  is the effective magnetic anisotropy constant of the  $i$ -th nanoparticle and  $H_i$  is assessed using (5) and (6). Thus, the free diffusion time of magnetisation  $\tau_{0N}^i$  for low damping [18] is

$$\tau_{0N}^i = \frac{\nu_i M_s}{2\gamma \alpha k_B T}, \quad (8)$$

where  $\alpha$  is the damping constant and  $\gamma$  is the gyro-magnetic ratio.

In (8), every value for MNP is known, except for  $\alpha$ . Our calculation shows the dependence of the relaxation time on the magnetic damping constant  $\alpha$ . With these conditions, the relaxation time relation for an oblique magnetic field is [18]:

$$\tau_N^i(\text{oblique field}) = \frac{4\pi\tau_{0N}^i (S_{i1}^{-1} + S_{i2}^{-1})}{\sqrt{c_{i1}^{(1)} c_{i2}^{(1)} e^{-\Delta V_{i12}} + \sqrt{c_{i1}^{(2)} c_{i2}^{(2)} e^{-\Delta V_{i21}}}}, \quad (9)$$

where  $\Delta V_{i12}$  and  $\Delta V_{i21}$  are the normalized energy barriers for magnetic moment re-orientations. One also has

$$c_{i1}^{(p)} = 2\sigma_i \cos(2\theta_{ip}) + h_i \cos(\theta_{ip} - \psi_i),$$

$$c_{i2}^{(p)} = 2\sigma_i \cos^2(\theta_{ip}) + h_i \cos(\theta_{ip} - \Psi_i) \quad (10)$$

where  $p = 1, 2$  while  $\theta_{ip}$  is the solution of the transcendental equation  $\sin(2\theta_i) = 2h_i \sin(\Psi_i - \theta_i)$ .

If  $h_i < h_{ic}(\Psi_i) < 1$  (where  $\Psi_i$  is the angle between  $\mathbf{H}_i$  and the easy anisotropy axis of the  $i$ -th nanoparticle), then [18]:

$$\begin{aligned} \cos(\theta_{i1,2}) = \pm 1 \mp \frac{1}{2} h_i^2 \sin^2(\psi_i) + h_i^3 \sin^2(\psi_i) \cos(\psi_i) \mp \frac{13 + 11 \cos(2\psi_i)}{16} h_i^4 \sin^2(\psi_i) \\ + \frac{3 + \cos(2\psi_i)}{2} h_i^5 \sin^2(\psi_i) \cos(\psi_i) \mp \frac{183 + 156 \cos(2\psi_i) - 19 \cos(4\psi_i)}{64} h_i^6 \sin^2(\psi_i) + \dots \end{aligned} \quad (11)$$

$$\begin{aligned} \Delta V_{i12} = \sigma_i \left[ 1 - 2h_i (\sin(\Psi_i) - \cos(\Psi_i)) + h_i^2 + \frac{h_i^3}{2} \sin(2\Psi_i) (\cos(\Psi_i) - \sin(\Psi_i)) + \frac{h_i^4}{2} \sin^2 2\Psi_i \right. \\ \left. + \frac{h_i^5}{32} \sin(2\Psi_i) (7 \cos(\Psi_i) - 3 \cos(3\Psi_i) - 7 \sin(\Psi_i) - 3 \sin(3\Psi_i)) + \frac{h_i^6}{2} \sin^2(2\Psi_i) + \dots \right] \end{aligned} \quad (12)$$

$$\begin{aligned} \Delta V_{i21} = \sigma_i \left[ 1 - 2h_i (\sin(\Psi_i) + \cos(\Psi_i)) + h_i^2 + \frac{h_i^3}{2} \sin(2\Psi_i) (\cos \Psi_i + \sin(\Psi_i)) + \frac{h_i^4}{2} \sin^2(2\Psi_i) \right. \\ \left. + \frac{h_i^5}{32} \sin(2\Psi_i) (7 \cos(\Psi_i) - 3 \cos(3\Psi_i) + 7 \sin(\Psi_i) + 3 \sin(3\Psi_i)) + \frac{h_i^6}{2} \sin^2(2\Psi_i) + \dots \right] \end{aligned} \quad (13)$$

$$\begin{aligned} S_{i1,2} = \sigma_i \sqrt{h_i \sin(\Psi_i)} \left[ 16 - \frac{104h_i}{3} \sin(\Psi_i) + h^2 (1 - 21 \cos(2\Psi_i)) + \frac{h_i^3}{2} \sin(\Psi_i) (45 + 51 \cos(2\Psi_i)) + \dots \right] \\ \pm 2\pi\sigma_i h_i^2 \sin(2\Psi_i) \left[ 4 - 3h_i \sin(\Psi_i) - 2h_i^2 \sin^2(\Psi_i) + \dots \right]. \end{aligned} \quad (14)$$

The normalized effective energy barriers for the nanoparticle magnetic moment re-orientations are defined as

$$\Delta V_i^{\text{eff}} = \frac{\Delta V_{i12} + \Delta V_{i21}}{2}. \quad (15)$$

The relation (9) can be now expressed as

$$\begin{aligned} \frac{1}{\tau_{0N}^i(\text{oblique field})} = \frac{S_{i1} S_{i2}}{4\pi\tau_{0N}^i(S_{i1} + S_{i2})} \\ \times \left[ \sqrt{c_{i1}^{(1)} c_{i2}^{(1)}} e^{-\Delta V_{i12}} + \sqrt{c_{i1}^{(2)} c_{i2}^{(2)}} e^{-\Delta V_{i21}} \right] = \\ f_{12} e^{-\Delta V_{i12}} + f_{21} e^{-\Delta V_{i21}}, \end{aligned} \quad (16)$$

where  $f_{12}$  and  $f_{21}$  are the attempt frequency factors.

The specific loss power (SLP) is the electromagnetic power lost per nanofluid mass unit [19], expressed in watt per kilogram. In biomedical applications, where the energy losses occur through magnetic relaxation processes, SLP can be calculated in the framework of the linear response theory [20].

Thus, for the nanofluid [21] one applies:

$$\begin{aligned} \text{SLP} = \frac{p}{f\rho} = 3\pi\mu_0\nu H_0^{\text{ext}} \frac{M_s}{\rho N} \\ \times \sum_{i=1}^N \left( \coth(\xi_i) - \frac{1}{\xi_i} \right) \frac{2\pi\nu\tau_{\text{eff}}^i \varepsilon_i}{1 + (2\pi\nu\tau_{\text{eff}}^i)^2}, \end{aligned} \quad (17)$$

where the magnetic volume fraction of the nanoparticles reads as

$$f_m = \frac{f}{1 + \frac{2\delta}{d_m - 2\delta}}. \quad (18)$$

Here,  $\rho$  is the density of the nanoparticle material,  $f$  is the volume fraction of nanoparticles,  $\delta$  is the coating thickness,  $\nu$  is the frequency,  $H_0^{\text{ext}}$  is the magnitude of the applied field,  $N$  is the number of nanoparticles and  $\tau_{\text{eff}}^i$  is the effective magnetic relaxation time for the  $i$ -th nanoparticle. Therefore, it follows that

$$\xi_i = \frac{\mu_0\nu_0 M_s H_0^{\text{ext}}}{k_B T} \quad (19)$$

and  $\varepsilon_i$  is given by [22–25]:

$$\varepsilon_i = \frac{\left(\frac{\sigma_i}{3.4}\right)^{1.47} \cos^2(\psi_i) + \frac{1}{3}}{\left(\frac{\sigma_i}{3.4}\right)^{1.47} + 1}. \quad (20)$$

As can be deduced from (6)–(8), the damping constant influences the Néel relaxation time through the free diffusion time of magnetisation  $\tau_{0N}^i$ . In many specialized works the dependence of  $\tau_{0N}^i$  on the damping constant  $\alpha$  (e.g., such as (7)) is provided in the range  $10^{-8} \div 10^{-12}$  s [26, 27]. The width of this range appears an inconvenience in the case of the numerical experiments which are working with analytical expressions of the Néel relaxation time. Therefore, choosing a value for the damping constant  $\alpha$  can greatly influence the results, as will be shown below.

### 3. Results and discussion

The influence of the magnetic nanoparticle system properties on the Néel relaxation time was studied experimentally by magnetorelaxometry (MRX) measurements [28]. Thus, it was shown that there is a clear trend of  $\alpha$  decrease with decreasing size for thin films of iron oxide [29]. This effect is also seen in other materials [30, 31] and in [29] an upper limit for  $\alpha$  in iron oxide thin films of 0.0365 was found. In the case of MRX measurements made on iron oxide particles, there is estimated an interval for the damping constant ranging from 0.0005 to 0.002 [14], i.e., significantly lower values compared to the value of 0.07 for this bulk material.

For our numerical study, we consider the case of a colloidal system consisting of 500 spherical

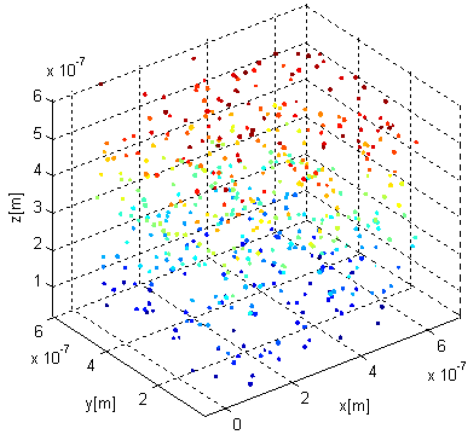


Fig. 1. Positions of nanoparticles inside the test cube.

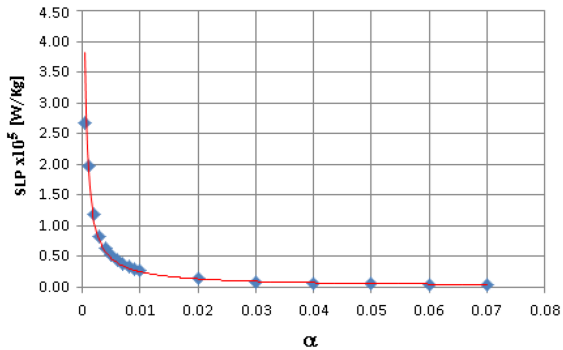


Fig. 2. SLP versus the damping constant  $\alpha$ .

magnetite nanoparticles with spontaneous magnetisation of  $4.46 \times 10^5$  A/m and uniaxial magnetic anisotropy of  $2.5 \times 10^4$  J/m<sup>3</sup>. The nanoparticles whose sizes have a lognormal distribution with the average diameter  $d_m = 10$  nm and standard deviation  $0.1d_m$  are dispersed in water whose dynamic viscosity is  $8.9 \times 10^{-4}$  Pa s. The temperature is 300 K and the coating thickness of the nanoparticles is 3 nm. We consider the volume fraction of the nanoparticles  $f = 0.05$ . The external magnetic field intensity is set to 15 kA/m along the  $z$  axis and the frequency to 300 kHz. These parameters fall within the allowed range for biomedical applications.

We consider a random placement of the nanoparticles in a face-centred cubic grid with random orientations of the anisotropy axes and nanoparticle magnetic moments (see Fig. 1).

As a numerical calculation procedure, we use the relations (3), (9) and (2) to calculate the Néel relaxation time, the Brownian relaxation time, and the effective magnetic relaxation time, respectively, and (16) to calculate SLP. The values of the damping constant  $\alpha$  vary between 0.0005 and the corresponding value for bulk material in the case of magnetite. Thus, we obtain the results presented in Fig. 2. It can be seen that the variation of the damping constant values quite strongly affects the SLP values

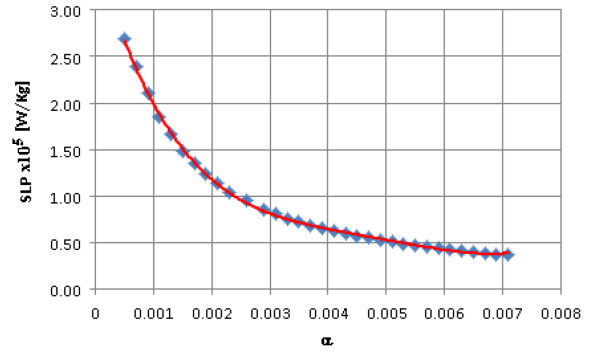


Fig. 3. SLP versus the damping constant  $\alpha$  over the range of variation suggested by the MRX experiments.

from about 268 458 W/kg to 3854 W/kg, requiring therefore paying an increased attention to the value used in the calculations related to the damping constant  $\alpha$ .

Figure 3 shows the variation of SLP versus the damping constant  $\alpha$  over a range of values closer to that suggested by the MRX experiments carried out on magnetite nanoparticle systems, 0.0005–0.002 [14].

It turned out that the values for SLP simulated with the model used in this paper for the range of damping constant values suggested by the MRX experiments carried out on magnetite nanoparticle systems, are in agreement with the values measured on such nanoparticle systems existing in the literature [32–35].

## 4. Conclusions

This paper provides a numerical experiment based on a simulation model of how the values of the damping constant  $\alpha$  influence the values of SLP in the case of systems of the nanoparticles suspended in a liquid matrix. The positions, orientations of the anisotropy axes and the orientations of the magnetic moments of the nanoparticles are considered to be random. The Néel relaxation time calculated using the Coffey model in the oblique magnetic field adapted to the local magnetic field on the nanoparticle and the Brownian relaxation time calculated using Brown's theory are entering into an effective magnetic relaxation time that influences SLP calculated based on the linear response theory (LRT).

Following the numerical simulations, we saw that SLP is strongly influenced by the value of the damping constant  $\alpha$  and, moreover, the SLP values simulated with the values of the damping constant  $\alpha$ , suggested by the MRX experiments on magnetite nanoparticle systems, are in agreement with the experimentally determined SLP values published in the literature. Choosing a value for the damping constant  $\alpha$  appropriate for the bulk material leads to results far from reality even if the numerical simulation models quite accurately describe the

behaviour of the real system. Thus, it is recommended to choose for the damping constant  $\alpha$  only values that match the values suggested by the MRX experiments carried out on nanoparticle systems.

The results obtained in this paper are useful for optimising the numerical modelling of the process of magnetic hyperthermia with nanoparticles, the process intensively studied by the scientific community because it is of real interest as a non-invasive method used in cancer therapy.

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