Journal of Magnetism and Magnetic Materials 426 (2017) 206–210

Contents lists available at ScienceDirect

Journal of Magnetism and Magnetic Materials

journal homepage: www.elsevier.com/locate/jmmm

The effect of the magnetic nanoparticle's size dependence of the relaxation time constant on the specific loss power of magnetic nanoparticle hyperthermia

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ARTICLE INFO

Keywords:
Landau-Lifshitz-Gilbert Equation
Linear response theory
Magnetic hyperthermia
Magnetic nanoparticles
Specific loss power

ABSTRACT

Magnetic nanoparticle hyperthermia is a cancer treatment in which magnetic nanoparticles (MNPs) are subjected to an alternating magnetic field to induce heat in the tumor. The generated heat of MNPs is characterized by the specific loss power (SLP) due to relaxation phenomena of the MNP. Up to now, several models have been proposed to predict the SLP, one of which is the Linear Response Theory. One parameter in this model is the relaxation time constant. In this contribution, we employ a macrospin model based on the Landau-Lifshitz-Gilbert equation to investigate the relation between the Gilbert damping parameter and the relaxation time constant. This relaxation time has a pre-factor τ_0 which is often taken as a fixed value ranging between 10^{-8} and 10^{-12} s. However, in reality it has small size dependence. Here, the influence of this size dependence on the calculation of the SLP is demonstrated, consequently improving the accuracy of this estimate.

1. Introduction

In oncology, hyperthermia refers to the heating of organs or tissues to temperatures ranging from 42 °C to 46 °C where it causes the death of cancer cells [1]. One possible way to locally apply heat to cancerous regions is by means of magnetic nanoparticle hyperthermia [2]. There, magnetic nanoparticles (MNPs) are injected in the body, and subsequently guided towards the cancer cells. This can be achieved by various mechanisms. They can for instance be directed by external fields [3], or, alternatively the particles can be coated with a biological marker which binds them to cancer cells [4]. Once the particles are at the desired location, they are subjected to an alternating magnetic field to induce a temperature increase in the particles and the tumor tissue. The heat generated by MNPs is provided by the dissipated power of the MNPs as they run through a hysteresis loop. This is quantified by the specific loss power (SLP), also known as the specific absorption rate (SAR) which is a measure of power dissipated per unit mass of the magnetic material. The heating properties of the MNPs depend on three major mechanisms. The first is called Néel relaxation [5], where the magnetization within the MNPs is excited by thermal fluctuations and irreversibly jumps over energy barriers due to the anisotropy of the material. Next, in Brownian relaxation the MNPs as a whole rotate due to their Brownian motion in their suspension. Finally, when the externally applied fields are sufficiently large to suppress the energy barriers between different anisotropy directions, a third, temperature independent, mechanism exists [6]. Next to the temperature, the size, the magnetic anisotropy and the saturation magnetization of the MNPs also the amplitude and the frequency of the magnetic field determine the relative strength of each loss mechanism. To gain further insight in the different processes and their combined effect on the SLP, a significant number of measurements have been carried out recently using various experimental setups [3,4,7]. Furthermore, also a number of increasingly complex analytical or numerical descriptions of the physics behind these processes are available [5,8]. Specifically, the theory behind the Néel relaxation mechanism is well established but unfortunately the resulting equations can only be solved analytically in specific limits, e.g. for very large energy barriers, and otherwise one has to rely on numerical calculations. Recently, a macrospin model based on the Landau-Lifshitz-Gilbert (LLG) equation has been employed to investigate the dynamics of the MNPs when subjected to an externally

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http://dx.doi.org/10.1016/j.jmmm.2016.11.079
Received 26 May 2016; Received in revised form 9 November 2016; Accepted 17 November 2016
Available online 21 November 2016
0304-8853/

applied magnetic field [9]. The LLG equation contains a phenomenological damping term, whose size is determined by the Gilbert damping constant [10], α, which accommodates for all loss mechanisms. The damping constant α is related to τ_0, which provides a typical relaxation time used in the description of the Néel relaxation process, and is the inverse of an attempt frequency [11]. Usually, τ_0 is described as constant taking values between 10^{-8} and 10^{-12} s [7]. However, this large range is inconvenient to accurately determine the losses related to the hysteresis loop of MNPs and furthermore, τ_0 has a size dependence which is often neglected. In this contribution, the relation between τ_0 and α is investigated. Based on a macrospin model [12], an empirical relation is determined and subsequently used in the SLP calculations of MNP samples with a lognormal size distribution [13].

2. Methods

2.1. Macrospin model

To investigate the magnetic dynamics of the MNPs, a model based on the LLG equation is used [12]

$$\frac{d\boldsymbol{m}}{dt} = -\frac{\mu_0\gamma}{1+\alpha^2}(\boldsymbol{m}\times \mathbf{H}_{\mathbf{eff}} + \alpha\boldsymbol{m} \times (\boldsymbol{m} \times \mathbf{H}_{\mathbf{eff}})) \tag{1}$$

where $\boldsymbol{m}$ denotes the magnetization vector of the considered MNP, normalized to the saturation magnetization $\boldsymbol{m}=\frac{M}{M_s}$, $\mathbf{H}_{\mathbf{eff}}$ is the effective field acting on each MNP and takes into account the demagnetizing, thermal, external and anisotropy field contributions [9], μ_0 is the vacuum permeability and $\gamma = 1.7595\ 10^{11}\ \frac{\text{rad}}{\text{Ts}}$ denotes the gyromagnetic ratio.

The LLG equation is numerically integrated for an ensemble of nanoparticles with the simulation tool Vinamax [9]. In all simulations, single core magnetite MNPs are considered with magnetic material properties M_s=446 kA/m, K=25 kJ/m³, at a temperature of 300 K. The MNP concentrations are assumed to be sufficiently low to neglect the dipolar interactions between the MNPs [11] and that the MNPs are sufficiently small to consider them to be single domain particles [13]. In all simulations an ensemble of at least 10000 MNPs is studied with easy axis orientations being randomly distributed. Either the 5th order Dormand-Prince method or the 7th order Fehlberg method are used to integrate the LLG equation. Both solvers use fixed time steps, which is required for the implementation of the randomly fluctuating thermal field [9]. Finally, for each simulation we carefully checked that an appropriate time step was used.

In real MNP samples, the MNPs often have a lognormal size distribution [13], as dictated by their production process [16]. Such distributions are therefore considered here as well. Eq. (2) shows the probability density function of the MNP radius r

$$p(r) = \frac{1}{\sigma r\sqrt{2\pi}} e^{\left(-\frac{\ln^2(r/\mu)}{2\sigma^2}\right)} \tag{2}$$

which can be interpreted as the distribution whose logarithm is normally distributed with μ and σ being the mean radius and standard deviation, respectively.

2.2. Magnetorelaxometry

Magnetorelaxometry is a method to characterize MNPs by measuring the decaying net magnetic moment of the MNP sample after it has been magnetized in an external field [17]. The decaying magnetic signal is described by

$$M(t) = M_0 e^{-\frac{t}{\tau}} \tag{3}$$

where M_0 denotes the magnetization of the sample at t=0 and τ the effective relaxation time constant given by $\frac{1}{\tau}=\frac{1}{\tau_N}+\frac{1}{\tau_B}$.
τ_N and τ_B denote the Néel and Brownian relaxation time respectively.

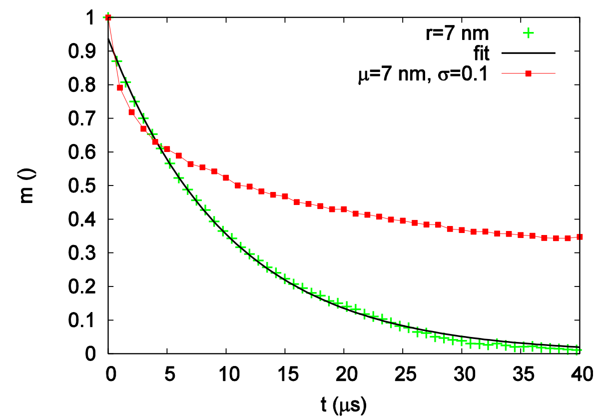

Fig. 1. MRX simulation of 10000 MNPs with α=0.01 with fixed radius of 7 nm (green symbols) or lognormal size distribution (red line). The black line shows the closest fit of Eq. (3) to the data to extract τ_N from the simulation. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

For immobilized MNPs only the Néel relaxation mechanism is relevant. Since the purpose is to attain the effect of the typical relaxation time τ_0 used in the description of the Néel relaxation process, the Brownian relaxation time will be no longer considered.

The Néel relaxation time

$$\tau_N = \tau_0 e^{\frac{KV}{k_B T}} \tag{4}$$

is mainly determined by the ratio between the energy barrier, given by the product of the anisotropy constant K and the volume $V=4\pi r^3/3$ of the MNPs, and the available thermal energy, which is the product of the Boltzmann constant k_B, and the temperature T.

As mentioned earlier, τ_0 is typically taken as a constant between 10^{-8} and 10^{-12} s. However, this quantity is size dependent as can be seen from Eq. (5), derived by Brown [15], in the high barrier limit $KV \gg k_B T$

$$\tau_0 = \frac{1+\alpha^2}{2\alpha\gamma}\sqrt{\frac{\pi k_B T M_s^2}{4K^3 V}} \tag{5}$$

With Vinamax, it is possible to simulate MRX experiments [11], and the result of one such simulation is shown in Fig. 1. The green symbols depict the relaxing signal of a sample consisting of MNPs with a fixed radius of 7 nm. The black curve shows a fit to this data with an equation of the form given by Eq. (3), allowing the extraction of τ_N from these simulations. Because also the energy barrier and temperature are known, τ_0 can thus be obtained with the help of Eq. (4).

When a sample with a lognormal size distribution is considered, the magnetic moment is no longer described by an exponentially decaying function but is given by a weighted sum of such functions [11]:

$$M(t) = \int_0^\infty M_0 e^{\frac{-t}{\tau_N(r)}} p(r)\, dr \tag{6}$$

A typical signal for μ=7 nm and σ=0.1 is depicted in Fig. 1 in red. It clearly exhibits a decaying magnetic moment that is no longer described by a simple decaying exponential. Each lognormal distribution gives rise to a characteristic shape, which allows to experimentally recovering the lognormal distribution of MNP samples from MRX data [18].

2.3. Linear response theory

Linear response theory (LRT) is a theoretical model which describes the dynamic response of an ensemble of MNPs to a time-varying external field [14]. When a time-varying magnetic field $H(t)=H_0\cos(\omega t)$ is applied with angular frequency ω and amplitude H_0 the magnetization response is given as follows: