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Nonlinear energy dissipation of magnetic nanoparticles in oscillating magnetic fields

D. Soto-Aquino [a], C. Rinaldi [b,*]

[a] ERC Incorporated, Air Force Research Laboratory, 10 E. Saturn Blvd., Edwards AFB, CA 93524, USA
[b] J. Crayton Pruitt Family Department of Biomedical Engineering and Department of Chemical Engineering, University of Florida, PO Box 116131, Gainesville, FL 32611-6131, USA

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ABSTRACT

The heating of magnetic nanoparticle suspensions subjected to alternating magnetic fields enables a variety of emerging applications such as magnetic fluid hyperthermia and triggered drug release. Rosensweig (2002) [25] obtained a model for the heat dissipation rate of a collection of non-interacting particles. However, the assumptions made in this analysis make it rigorously valid only in the limit of small applied magnetic field amplitude and frequency (i.e., values of the Langevin parameter that are much less than unity and frequencies below the inverse relaxation time). In this contribution we approach the problem from an alternative point of view by solving the phenomenological magnetization relaxation equation exactly for the case of arbitrary magnetic field amplitude and frequency and by solving a more accurate magnetization relaxation equation numerically. We also use rotational Brownian dynamics simulations of non-interacting magnetic nanoparticles subjected to an alternating magnetic field to estimate the rate of energy dissipation and compare the results of the phenomenological theories to the particle-scale simulations. The results are summarized in terms of a normalized energy dissipation rate and show that Rosensweig's expression provides an upper bound on the energy dissipation rate achieved at high field frequency and amplitude. Estimates of the predicted dependence of energy dissipation rate, quantified as specific absorption rate (SAR), on magnetic field amplitude and frequency, and particle core and hydrodynamic diameter, are also given.

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1. Introduction

Suspensions of magnetic nanoparticles, so-called magnetic fluids or ferrofluids, are attractive in a wide range of applications because of their ability to respond to a magnetic field. An important characteristic of these complex fluids is that in the presence of an alternating magnetic field (AMF) the suspension can generate heat [1–3]. This effect has led to development of promising applications such as cancer treatment by magnetic fluid hyperthermia [4–14], which takes advantage of the greater sensitivity of cancer tissue to heat. Another example is where magnetic nanoparticles are loaded into liposomes or coated with thermoresponsive polymers bearing a drug, which is released due to a change in the liposome or polymer triggered by energy dissipation due to the magnetic nanoparticle in an AMF [15–17]. In these biomedical applications, it is desirable to maximize the energy dissipation rate of the nanoparticles in order to minimize the amount of material that needs to be delivered to the intended tissue to achieve a desired effect. In contrast to this, there are other applications [18–22] where a temperature rise due to the action of an alternating field is counter-productive, as in the case of loudspeakers [23] where a layer of ferrofluid replaces the air gap between magnet and voice coil.

The heating power of magnetic nanoparticles is determined by several factors, including the magnetic material properties and the frequency and intensity of the alternating magnetic field. There are two relaxation mechanisms by which suspended magnetic nanoparticles respond to an alternating magnetic field and subsequently dissipate heat: physical rotation of the individual particles in the fluid (so-called Brownian relaxation) and the collective rotation of the atomic magnetic moments within each particle without the need for physical particle rotation (so-called Néel relaxation) [24]. For Brownian relaxation the rate of magnetization reorientation is determined by viscosity and particle size, and can be estimated from

$$\tau_B = \frac{3\eta V}{k_B T}, \tag{1}$$

where η is the fluid viscosity, V the volume of a single particle, k_B is

* Corresponding author.
E-mail address: carlos.rinaldi@bme.ufl.edu (C. Rinaldi).

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the Boltzmann constant, and T the absolute temperature. For Néel relaxation the characteristic time can be determined from

$$\tau_N = \tau_0 \exp\left(\frac{KV}{k_B T}\right), \tag{2}$$

where K denotes the magnetocrystalline anisotropy constant and τ_0 is a time constant. Both of these relaxation mechanisms are single particle effects and one typically assumes the shortest relaxation time dominates the process. Another potential mechanism for energy dissipation by magnetic nanoparticles in alternating magnetic fields is so-called hysteresis heating [2], which is expected for larger multi domain particles. Here we focus only on heating due to relaxation processes.

The principles underlying the relaxation heating of magnetic nanoparticles in a ferrofluid were described by Rosensweig [25]. However, as shown below, the assumptions of his analysis were strictly valid only in the limit of small applied magnetic field amplitude and frequency, due to the use of the phenomenological magnetization relaxation equation derived by Shliomis in 1972 [26], and because of the use of a linear magnetization assumption. As such, Rosensweig's model is often referred to as the linear response theory for energy dissipation by the magnetic nanoparticles. To amend this limitation, Rosensweig made an ad hoc substitution of the initial susceptibility for the so-called chord susceptibility in his final expressions for the volumetric energy dissipation rate, an often overlooked modification in subsequent papers that compare measurements to Rosensweig's theory. These limitations led Raikher and Stepanov [27] to investigate the absorption of AC field energy in suspensions of magnetic dipoles using a formulation based on solution of the Fokker–Planck equation, predicting that the expression due to Rosensweig ceases to be valid for large values of the applied field frequency. Beyond this, most subsequent analyses have been limited to applying the model of Rosensweig to predict the potential changes in temperature even under conditions where the assumptions used in deriving the model are no longer valid.

In this contribution we approach this problem from an alternative point of view by solving the phenomenological magnetization relaxation equation of Shliomis [26] exactly for the case of arbitrary magnetic field amplitude and frequency and by solving the magnetization relaxation equation of Martsenyuk, Raikher, and Shliomis (MRSh) [28] numerically. The latter has been found to accurately describe the magnetic field and shear rate dependence of the so-called magnetoviscosity of dilute ferrofluids [29–31]. Rotational Brownian dynamics simulations of non-interacting particles subjected to an oscillating magnetic field are compared with these solutions of the phenomenological models. We note at the outset that the MRSh equation and the Brownian dynamics simulations used below are strictly valid only for suspensions of nanoparticles that relax by the Brownian relaxation mechanism and for which there are no particle–particle interactions. While these limitations preclude direct comparison of our results to a variety of experimental measurements with particles that relax by the Néel relaxation mechanism or for which interactions are significant, or indeed dominant, the results are nevertheless valuable in providing insight into the effects of non-linear magnetization on the energy dissipation rate of ferrofluids in alternating magnetic fields. The results are summarized in terms of a non-dimensional energy dissipation rate, which is a function of the applied field amplitude, parameterized by the Langevin parameter. Finally, numerical estimates for the rate of energy dissipation, as specific absorption rate, are given as a function of particle size and magnetic field amplitude and frequency.

2. Energy dissipation by relaxation losses

In a suspension of magnetic nanoparticles the Brownian or Néel mechanisms both lead to an apparent superparamagnetic behavior. If the field, $\mathrm{H}=H_0\mathbf{i}_z$, is suppressed, the magnetization relaxes to a new equilibrium state. In the case of an external oscillating magnetic field, $\mathrm{H}=H_0\ \cos(\Omega t)\mathbf{i}_z$, the dipole moment of the particles follows the oscillations of the magnetic field with a phase-lag between the field and the particle, due to the finite magnetic relaxation time of the particles. During this process the magnetic field exerts work on the magnetic particles, which is ultimately converted to heat.

There are various formulations for the rate of magnetic work and the thermodynamics of magnetizable systems [32]. However, in all of these the magnetic work term is the product of magnetic field and magnetization. For a cycle of the magnetic field these formulations can be shown to be equivalent to the expression

$$W_m = \mu_0 \int_0^{2p} \mathbf{H}\cdot\frac{d\mathbf{M}}{dt}dt, \tag{3}$$

where $2p$ is the period of the cycle.

We consider the case where the internal energy is constant during application of the alternating magnetic field, such that the energy of the work done by the magnetic field is dissipated as heat, thus

$$Q = W_m = \mu_0 \int_0^{2p} \mathbf{H}\cdot\frac{d\mathbf{M}}{dt}dt. \tag{4}$$

Then, we may define the average rate of energy dissipation per cycle of period $2p$ as

$$\left\langle \dot{Q} \right\rangle = \frac{1}{2p}\mu_0 \int_0^{2p} \mathbf{H}\cdot\frac{d\mathbf{M}}{dt}dt. \tag{5}$$

We limit attention to a linearly polarized field, such that $\mathbf{H}$ and $\mathbf{M}$ are collinear to obtain

$$\left\langle \dot{Q} \right\rangle = \frac{1}{2p}\mu_0 \int_0^{2p} H\frac{dM}{dt}dt. \tag{6}$$

It will be convenient for later calculations to rewrite Eq. (6) in the equivalent form

$$\left\langle \dot{Q} \right\rangle = -\frac{1}{2p}\mu_0 \int_0^{2p} M\frac{dH}{dt}dt. \tag{7}$$

In many practical cases the alternating magnetic field is sinusoidal

$$H = H_0\ \cos(\Omega t), \tag{8}$$

with amplitude H_0 and radian frequency Ω. In that case (7) reduces to

$$\left\langle \dot{Q} \right\rangle = \frac{\mu_0 H_0 \Omega}{2p} \int_0^{2p} M\ \sin(\Omega t)dt, \tag{9}$$

with $p = \pi/\Omega$.

Next we consider the fact that the magnetization of a suspension of magnetic nanoparticles cannot exceed the saturation magnetization $M_s = \phi M_d$ and that it would seem appropriate to scale time with respect to the nanoparticle relaxation time τ, hence we introduce the dimensionless variables

$$\tilde{M} = M/M_s;\ \tilde{\Omega} = \Omega\tau;\ \tilde{t} = t/\tau, \tag{10}$$

and obtain

$$\left\langle \dot{Q} \right\rangle = \frac{\mu_0 H_0 \phi M_d\, \Omega}{2}\frac{1}{\tilde{p}} \int_0^{2\tilde{p}} \tilde{M}\ \sin(\tilde{\Omega}\tilde{t})d\tilde{t}, \tag{11}$$

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