Microwave Absorption by a Rigid Dipole in a Viscous Fluid

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***Abstract*— We present a comprehensive theoretical and numerical study of the stochastic rotational motion of a high anisotropic ferromagnetic nanoparticle in a viscous carrier driven by a time-periodic field. The effects of energy absorption of circularly and linearly polarized fields are analyzed. The analytical expressions and numerical data for the probability density and the power loss are compared for different values of the system parameters. Their coincidence and divergence are discussed.**

Keywords*— *ferrofluid; rigid dipole; Fokker Planck equation; time-periodic field; power loss

I. Introduction

Ferrofluids are the complex media, which consist of ferromagnetic fine particles dispersed in a viscous liquid [1]. Such physical systems combine the basic properties of ferromagnetic solids, i.e. high permeability to magnetic fields, internal magnetization, and those of liquids, i.e. presence of surface tension, viscosity. Remarkable properties mentioned above make ferrofluids extremely attractive for such different fields as applied physics, microscale technologies, and medicine.

The most prominent applications are closely bound with the interaction with RF-field and absorption of its energy. One can underline a targeted hyperthermia in cancer therapy [2], when a great therapeutic effect is achieved by concentration of injected ferrofluid locally in injured tissue and further heating by a time-periodic magnetic field. Control of the heating rate is a critical issue here, and it is performed by the control of both fine particle parameters and field amplitude and frequency.

There are three ways for energy dissipation: mechanical rotation of a nanoparticle in a viscous carrier, damping precession of the nanoparticle magnetic moment within crystal lattice, and Eddy currents [3]. For a highly anisotropic particle of the radius of ~ 20 nm and not very large frequencies (~ 10^3-10^6 MHz) the first mechanism is a dominant and the other ones can be neglected.

The mechanical rotation is commonly described using a rigid dipole model [4], when the magnetic moment is supposed to be locked in the crystal lattice. The influence of the thermal bath essentially complicates the study of the phenomena of the RF-field energy absorption. Commonly, this problem is treated within the concept of complex magnetic susceptibility [3]. But when the magnetic energy becomes comparable with the thermal one, the relevant Langevin based approach along with the Fokker-Planck (FP) formalism [4] should be applied for a correct description.

Exact expressions for the probability density function (solution of the appropriate FP equation) and for the power loss of the rigid dipole in the linearly polarized field were reported in [5]. At the same time, in the case of the circularly polarized field action, the power loss was found in quasi-static approximation [6], while the probability density function was obtained in linear (with respect to the field frequency and amplitude) approximation [7]. Thus, to date, the role of the field parameters, liquid parameters, and temperature in the microwave absorption by a ferrofluid has not been studied fully.

II. Model Description

Following [7] we consider the mechanical rotation of a uniform spherical single-domain ferromagnetic nanoparticle of radius R, magnetization $\mathbf{M}$ ($|\mathbf{M}| = M = \text{const}$) and density ρ. In the deterministic case, this motion is governed by a pair of coupled equations

$$\dot{\mathbf{M}} = \boldsymbol{\omega} \times \mathbf{M}, \tag{1a}$$
$$J\dot{\boldsymbol{\omega}} = V\mathbf{M} \times \mathbf{H} - 6\eta V \boldsymbol{\omega}, \tag{1b}$$

where $\boldsymbol{\omega}$ is the angular velocity of the particle, J (= $8\pi\rho R^5/15$) is the moment of inertia of the particle, $\mathbf{H}$ is the external field, V is the nanoparticle volume, η is the viscosity of the fluid.

When the particle size is sufficiently small, the inertial term ($J\boldsymbol{\omega}$) can be neglected entirely for all practically interesting frequencies. Then, taking into account the random torque $\boldsymbol{\xi} = \boldsymbol{\xi}(t)$, which represents the thermal bath action, and Eq. (1a), one can obtain from Eq. (1b)

$$\dot{\mathbf{M}} = -\frac{1}{6\eta}\mathbf{M} \times (\mathbf{M} \times \mathbf{H}) - \frac{1}{6\eta V}\mathbf{M} \times \boldsymbol{\xi}, \tag{2}$$

which is commonly used to describe the stochastic rotation of ferromagnetic nanoparticles in a viscous fluid. In the spherical coordinates θ and φ (see Fig. 1), the Langevin equation (2) takes the following form:

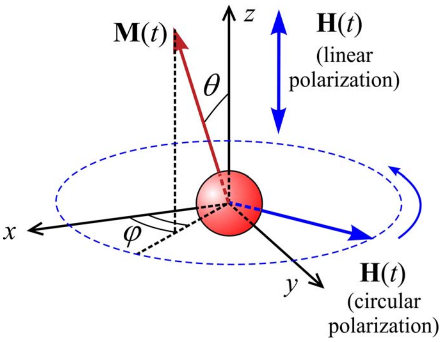

Fig. 1. (Color online) Sketch of the model. The nanoparticle with the frozen magnetization **M** is rotated about the origin of the Cartesian coordinate system in a viscous fluid under the action of the circularly or the linearly polarized external magnetic field **H**.

$$\dot{\theta}=\frac{1}{\tau_1}\left(h_x\sin\theta\cos\varphi+h_y\sin\theta\sin\varphi+h_z\cos\theta\right)\cot\theta-$$
$$-\frac{1}{\tau_1}h_z\frac{1}{\sin\theta}+\sqrt{\frac{2}{\tau_2}}\left(\zeta_y\cos\varphi-\zeta_x\sin\varphi\right), \quad (3a)$$
$$\dot{\varphi}=\frac{1}{\tau_1}\left(h_y\cos\varphi-h_x\sin\varphi\right)\frac{1}{\sin\theta}-$$
$$-\sqrt{\frac{2}{\tau_2}}\left(\zeta_x\cos\varphi+\zeta_y\sin\varphi\right)\cot\theta+\sqrt{\frac{2}{\tau_2}}\zeta_z, \quad (3b)$$

where $\boldsymbol{\zeta}=\boldsymbol{\xi}(12\eta V k_B T)^{0.5}$ is the rescaled random torque, k_B is the Boltzmann constant, T is the absolute temperature, $\tau_1=6\eta/M^2$, $\tau_2=6\eta V/k_B T$ are the characteristic times of the nanoparticle rotation induced by the external magnetic field and thermal torque, respectively ($\tau_2/2$ is the Brownian relaxation time), x, y, z are the Cartesian coordinates, $\mathbf{h}=\mathbf{H}/M$ is the reduced external field. The Cartesian components ζ_ν ($\nu=x, y, z$) are considered as the independent Gaussian white noises with zero means and correlation function $\langle\zeta_\nu(t)\zeta_\nu(t')\rangle=\Delta\delta(t-t')$, where angular brackets denote averaging over all realizations of the Wiener processes producing noises ζ_ν, Δ is the dimensionless noise intensity, and $\delta(t)$ is the Dirac δ-function.

Another way of description of the nanoparticle stochastic rotation is a statistical one and based on the FP equation. The FP equation, which corresponds to Eqs. (3), can be written in the form

$$\frac{\partial}{\partial t}P+\frac{1}{\tau_1}\frac{\partial}{\partial\theta}\left[\left(h_x\cos\varphi-h_y\sin\varphi\right)\cos\theta-h_z\sin\theta+\frac{1}{\kappa}\cot\theta\right]P+$$
$$+\frac{1}{\tau_1}\frac{1}{\sin^2\theta}\frac{\partial}{\partial\varphi}\left(h_y\cos\varphi-h_x\sin\varphi\right)P-\frac{1}{\tau_2}\frac{\partial^2 P}{\partial\theta^2}-$$
$$-\frac{1}{\tau_2}\frac{1}{\sin^2\theta}\frac{\partial^2 P}{\partial\varphi^2}=0, \quad (4)$$

where $P=P(\theta, \varphi, t)$ is the time-dependent probability density function for the rotational states of the nanoparticle (θ, φ are the polar and azimuthal angles, respectively),

$$\kappa=\tau_2/\tau_1=4\pi R^3M^2/3k_BT \quad (5)$$

is the relationship between the magnetic and thermal energies that shows the relative contribution of thermal fluctuations. The latter parameter is the most useful for the analysis of the role of temperature in the nanoparticle behavior including microwave absorption.

As shown in [8], the following Langevin equations

$$\dot{\theta}=\frac{1}{\tau_1}\left(h_x\cos\varphi+h_y\sin\varphi\right)\cos\theta-\frac{1}{\tau_1}h_z\sin\theta+$$
$$+\frac{1}{\tau_2}\cot\theta+\sqrt{\frac{2}{\tau_2}}\mu_1, \quad (6a)$$
$$\dot{\varphi}=\frac{1}{\tau_1}\left(h_y\cos\varphi-h_x\sin\varphi\right)\frac{1}{\sin\theta}+\sqrt{\frac{2}{\tau_2}}\frac{1}{\sin\theta}\mu_2 \quad (6b)$$

also correspond to the Eq. (4). Here $\mu_i=\mu_i(t)$ ($i=1, 2$) are the independent Gaussian white noises of the unit intensity with zero means, and δ correlation functions. Therefore, Eq. (3) and Eq. (6) are equivalent in the statistical sense, so, we can use more suitable for the numerical simulation Eq. (6) instead of Eq. (3).

We consider two types of the external time-periodic field:

$$\mathbf{h}=\mathbf{e}_x h\cos\Omega t+\mathbf{e}_y h\sin\Omega t, \quad (7a)$$
$$\mathbf{h}=\mathbf{e}_z h\cos\Omega t, \quad (7b)$$

are the circularly and linearly polarized, respectively. Here $\mathbf{e}_x$, $\mathbf{e}_y$, $\mathbf{e}_z$ are the unit vectors of the Cartesian coordinates, h is the field amplitude, t is the time, and Ω is the field frequency. The nanoparticle driven by the field (7a) or (7b) performs spherical motion or rotation with immobilized center of mass. Interaction with a viscous media leads to dissipation of the mechanical energy of the particle and further heating of the surrounding environment. These losses are compensated by the absorption of energy of the external field.

The power loss per period, characterizing the absorption process during the forced nanoparticle rotation, in accordance with the definition can be written as

$$Q=\frac{V\Omega}{2\pi}\cdot\int_0^{2\pi/\Omega}dt\mathbf{H}\frac{\partial\mathbf{M}}{\partial t}. \quad (8)$$

Trajectories of the nanoparticle are random due to the interaction with the thermal bath. Therefore, the dimensionless averaged over trajectories power loss q is of interest from the point of view of description of the heating and microwave absorption

$$q=\frac{\tau_1}{M^2V}\int_0^{\pi}d\theta\int_0^{2\pi}d\varphi QP=\frac{\Omega\tau_1}{2\pi M^2}\int_0^{\pi}d\theta\int_0^{2\pi}d\varphi P\int_0^{2\pi/\Omega}dt\mathbf{H}\frac{\partial\mathbf{M}}{\partial t}. \quad (9)$$

III. Results and Discussion

A. Circularly Polarized Field

For the external field of the type (7a), the FP equation (4) in the stationary case ($\partial P/\partial t = 0$) has the approximate solution for $\kappa h \ll 1$ and $\kappa\Omega \ll 1$ [7]. The probability density function here is given by the expression

$$P_0 = \frac{\sin\theta}{4\pi}\left[1 + \kappa h \sin\theta\cos\psi - \frac{\kappa^2 h^2}{6}\left(1 - 3\sin^2\theta\cos^2\psi\right)\right] + \\ + \frac{1}{8\pi}\kappa^2 h\Omega\sin^2\theta\sin\psi\,, \qquad (10)$$

where $\psi = \varphi - \Omega t$ is the lag angle between vectors **H** and **M**.

The average value of **MH** should remain constant for the specified system parameters. Even though the quasi-periodic mode of motion [9] is generated in the noise-free approximation, thermal fluctuations should suppress it and the condition $\langle \mathbf{MH} \rangle = \text{const}$ holds. Therefore, the following Soto-Aquino [10] equation (9) can be rewritten as

$$q = \frac{\Omega\tau_1}{2\pi M^2}\int_0^{\pi} d\theta \int_0^{2\pi} d\varphi P \int_0^{2\pi/\Omega} dt \mathbf{M}\frac{\partial \mathbf{H}}{\partial t} \qquad (11)$$

that is more suitable for numerical calculation using effective Langevin equation (6). Then, substituting Eq. (10) into Eq. (11) and performing the necessary integration procedures, we obtain the average power loss q in the form

$$q = \kappa^2 h^2 \Omega^2 \tau_1^2 / 6\,. \qquad (12)$$

To confirm these findings, we carried out the numerical calculation, where Eq. (6) was solved using the fourth-order Runge-Kutta method and power loss was calculated via the sum of increments $\Delta t \mathbf{M} d\mathbf{H}/dt$ for each iteration with the time step Δt. For averaging over the trajectories we performed 262 144 runs with different initial conditions and different noise vectors for random torque calculation. Each run lasted 20 field periods. The video-card Nvidia GeForce 450 GTS and CUDA technology were used for high-performance numerical simulation.

Fig. 1 corroborates the validity of our analytical estimation because of good agreement with the numerical results. The divergence increases while the field frequency grows that also corresponds to the main condition for obtaining Eq. (10). The simulation was performed for the following values of the system parameters: $M = 3.38 \cdot 10^5$ A/m (γ-Fe_2O_3, maghemite), $\eta = 5.0 \cdot 10^{-3}$ Pa/s (human blood), $R = 10$ nm, $T = 314$ K.

The role of thermal fluctuations in the microwave absorption is traced from Fig. 2. As it was expected, when the thermal energy grows relatively to the magnetic one, the efficiency of the nanoparticle heating decays. On the other hand, the difference between the power losses for various κ strongly depends on the field frequency. Such a difference is most pronounced for small frequencies, while with increasing frequency this difference

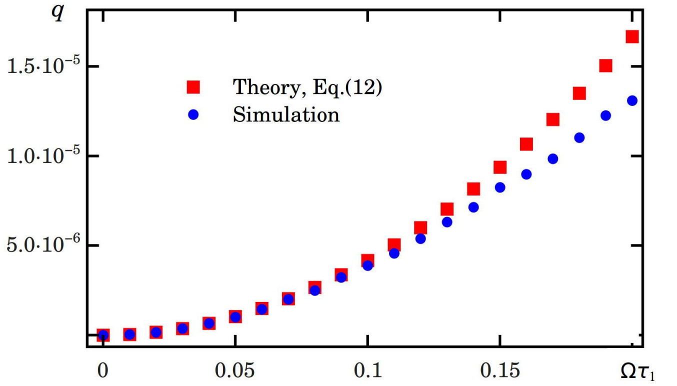

Fig. 2. (Color online) The comparison of the analytical and numerical results for the case of the circularly polarized field action.

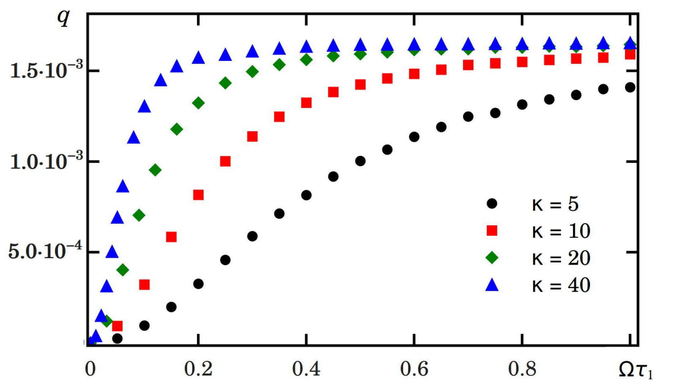

Fig. 3. (Color online) The numerical results for different values of κ when the circularly polarized field is applied.

decreases for different noise intensities, and the dynamic approximation becomes valid. Then, while frequency grows, the power loss tends to constant.

B. Linearly Polarized Field

When the external field of the type (6b) is applied, the FP equation takes the following form due to the symmetry of the problem:

$$\frac{df}{dt} = \frac{1}{\tau_2}\left[\frac{1}{\sin\theta}\frac{\partial}{\partial\theta}\left(\sin\theta\frac{\partial f}{\partial\theta} + f\cdot\kappa h\sin^2\theta\cos\Omega t\right)\right]. \qquad (13)$$

One needs to underline that the latter equation entirely coincides (up to the constants) with the corresponding formula in [5]. The solution of Eq. (13) is obtained by expansion in Legendre polynomials and harmonics [5]

$$f = 0.5 + \sum_{l=0}^{\infty}\left[\sum_{n=0}^{\infty} A_{ln}\cos n\Omega t + \sum_{n=0}^{\infty} B_{ln}\sin n\Omega t\right]\mathrm{P}_l(x)\,, \qquad (14)$$

where $x = \cos\theta$, $\mathrm{P}_l(x)$ is the Legendre polynomials, n, l are the whole numbers. Finally, the constants can be found from the algebraic sets of equations [5]

$$-n\Omega A_{ln} = \frac{1}{\tau_2}\left[-l(l+1)B_{ln} + \frac{\kappa h}{2}\left(\frac{l(l+1)}{2l-1}\left(B_{l-1,n-1}+B_{l-1,n+1}\right) - \frac{l(l+1)}{2l+3}\left(B_{l+1,n-1}+B_{l+1,n+1}\right)\right)\right], \quad (15a)$$

$$n\Omega B_{ln} = \frac{1}{\tau_2}\left[-l(l+1)A_{ln} + \frac{\kappa h}{2}\left(\frac{l(l+1)}{2l-1}\left((1+\delta_{n1})A_{l-1,n-1}+A_{l-1,n+1}\right) - \frac{l(l+1)}{2l+3}\left((1+\delta_{n1})A_{l+1,n-1}+A_{l+1,n+1}\right)\right)\right] \quad (15b)$$

for $l \geq 1, \; n \geq 1$,

$$A_{l0} = \frac{\kappa h}{2}\left[\frac{1}{2l-1}A_{l-1,1} - \frac{1}{2l+3}A_{l+1,1}\right] \text{ for } (l \geq 1). \quad (15c)$$

In addition, $A_{00} = 0.5$, $A_{0n} = 0$, $B_{l0} = 0$, $B_{0n} = 0$.

The power loss q in this case after substituting of Eq. (14) into Eq. (11), taking into account the relationships $H_x M_x = 0$, $H_y M_y = 0$, $M_z = M{\cdot}x$, and performing all integration procedures can be written in the simple form

$$q = h\Omega\tau_1 B_{11}/3 . \quad (16)$$

Here, the coefficient B_{11} should be found by the solution of Eqs. (15) numerically. Eq. (16) is confirmed by the numerical simulation based on Eq. (6) in the way described above. The simulation results are illustrated in Fig. 4 that completely coincides with the analytical ones in a whole range of the field frequencies. Also, as it was expected, the high frequency asymptotic values of the power loss q for the case of the linearly polarized field are two times smaller than for the circularly polarized one.

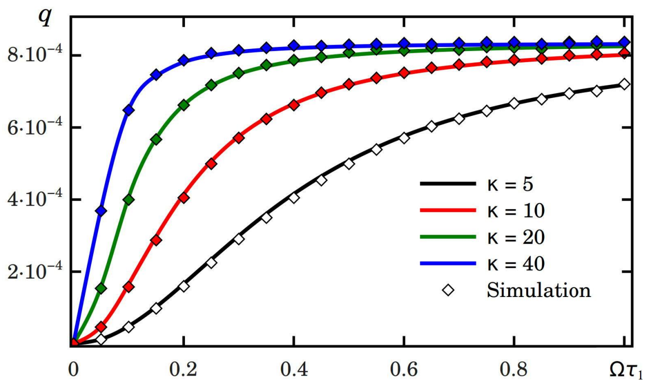

Fig. 4. (Color online) The numerical and analytical results for different values of κ when the linearly polarized field is applied.

IV. CONCLUSIONS

We present the comprehensive study of the interaction of the spherical uniform ferromagnetic nanoparticle of magnetization M placed into the fluid of viscosity η with the time-periodic external magnetic field of amplitude h and frequency Ω. To this end, the Langevin and Fokker-Planck equations are utilized together with the rigid dipole model. The advantage of our investigation consists in the consequent account of the thermal fluctuations using both the numerical and analytical approaches. In this way we confirm numerically the analytical results for the parameters where the derived expressions are valid and obtain the data in a whole range of the parameters.

We characterize the environment heating that is a result of the interaction of the external field with the nanoparticle by the dimensionless power loss q. The analytical expressions for q were obtained for the circularly and linearly polarized external fields. Their dependences on the system parameters such as Ω, h, type of polarization, η, and thermal noise intensity were examined in full both theoretically and numerically. In particular, it was established that the dependences $q(\Omega)$ exhibit saturated behavior and tend to constant with increasing frequency. The influence of the thermal fluctuations also decays with the frequency. Thus, the most efficient heating of the ferrofluid by the time-periodic magnetic field takes place when the condition $\Omega\tau_1 \sim 1$ holds, where $\tau_1 = 6\eta/M^2$ is the characteristic time of the nanoparticle rotation induced by the field.

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