Energy Dissipation of Interacting Rigid Dipoles Driven by the RF-field in a Viscous Fluid

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***Abstract*—We have considered the response of an ensemble of uniaxial ferromagnetic nanoparticles placed into a fluid to an alternating field in the presence of thermal bath. The attention was paid to the absorbtion of the field energy as a result of nanoparticles rotation in a viscous fluid. The influence of the inter-particle interaction effects was studied numerically based on the effective Langevin equation. Using the Barnes-Hut algorithm and CUDA technology, the set of numerical results, which describes the frequency dependence of the power loss, has obtained for various system parameters.**

Keywords—ferrofluid; rigid dipole; effective Langevin equation; MD-simulation; CUDA

I. Introduction

Ferrofluids are media composed of magnetic nanoparticles of diameters in the range of 10-50 nm which are dispersed in a viscous fluid (for example, water or ethylene glycol) [1], [2]. These media combine the properties of both ferromagnetic solids and liquids. These unique characteristics make ferrofluids an attractive candidate for performing different tasks ranging from the delivery of rocket fuel into a spacecraft thrust chambers under zero-gravity conditions to high-precision drug delivery and magnetic fluid hyperthermia cancer therapy [3], [4]. Namely the last application motivated us to study the problem of energy dissipation of a ferromagnetic nanoparticle in a periodic magnetic field.

A viscous induced rotation of nanoparticles is the dominant mechanism of energy absorption and further ferrofluid heating for a highly anisotropic particle of radius more than 20nm and not very large frequencies ($10^3 - 10^6$MHz) [5]. To study this kind of motion, the rigid dipole model, where the magnetic moment is supposed to be locked into the crystal lattice [2], is widely used. Within this framework, a number of results for the dynamical and stochastic approximations were obtained. Here and after we are focused on the last case. Since the Langevin and Fokker-Plank formalism for such systems was developed [2], [7], it was applied successfully on repeated occasions. Thus, the power loss was found for a circularly-polarised [6], [8] and a linearly-polarised [9] fields. The convenient form of the Langevin equation for analytical and numerical treatment was established in [10]. Based on the numerical simulation, all these results were confirmed and extended in [8]. The work [9] deserves a special attention because of the attempt of account of the dipole interaction in the mean field approximation. However, the problem above can not be solved analytically and the numerical approach is demanded.

To these purposes, the molecular dynamics simulation method is often utilized. Based on it, a number of the magnetic properties of ferrofluids was investigated. Thus, in the works [11], [12] the ferrofluid structure and initial susceptibility were studied, in [12] the simulation was used for the size distribution determination, and the dynamical properties of ferrofluids were studied in [13]. It is rather unexpected, but to study the power loss, the model based on the Landau-Lifshitz equation, where only damping precession of the magnetic moment is taken into account [14], [15], is also used. Despite this approach is valid under some circumstances, it is used primarily because of the simpler equations of motion.

An excellent possibility of high-performance calculations on common PC, which is based on the use of graphics processing units for general-purpose computing (the so-called CUDA technology), was unveiled by Nvidia company [16]. And now a lot of scientific problems can be solved in inexpensive way and without special facilities like clusters or supercomputers. The collective dynamics of nanoparticles ensembles with the long-range dipole interaction is a suitable problem to test CUDA. Another decision to improve simulation performance is the approximate calculations of dipole fields, which mainly takes into account the nearest neighbours influence. The Barnes-Hut algorithm is the most well known in this regard; it was successfully implemented to ferrofluids simulation in [17]. Therefore, the main aim of the present study is to reveal the role of the dipole interaction in a ferrofluid heated by an alternating field using the MD simulation based on the effective Langevin equation [10] and technique presented in [17].

II. Model and Methods

We consider the ensemble of equal spherical uniform ferromagnetic uniaxial nanoparticles of radius R, magnetization $\mathbf{M}$ ($\mathbf{M} = M = const$), and density D. Following [10], the rotational motion is described by the effective stochastic equations

$$\frac{d\theta_k}{d\tilde{t}} = (h_{kx}\cos\varphi_k + h_{ky}\sin\varphi_k)\cos\theta_k - h_{kz}\sin\theta_k + \frac{1}{\kappa}\cot\theta_k + \sqrt{\frac{2}{\kappa}}\,\mu_{k1}, \quad (1)$$

$$\frac{d\varphi_k}{d\tilde{t}} = \frac{1}{\tau_1}(h_{ky}\cos\varphi_k - h_{kx}\sin\varphi_k)\frac{1}{\sin\theta_k} + \sqrt{\frac{2}{\kappa}}\frac{1}{\sin\theta_k}\,\mu_{k2}, \quad (2)$$

$$\frac{d\boldsymbol{\rho}_k}{d\tilde{t}} = \frac{16\pi}{9}(\mathbf{f}_k^{dip} + \mathbf{f}_k^{sr}) + \sqrt{\frac{8}{3\kappa}}\mu_{k3}, \quad (3)$$

where θ, φ are the spherical coordinates of the nanoparticle magnetic moment, $\boldsymbol{\rho}$ is the vector defining the reduced coordinates of the nanoparticle, k is the index number of the nanopaticle in the ensemble, $\tilde{t} = t/\tau_1$ is the dimensionless time, $\tau_1 = 6\eta/M^2$ is the characteristic relaxation time, $\kappa = M^2V/k_{\mathrm{B}}T$ is the ratio of the magnetic and thermal energies, η is the fluid viscosity, V is the nanoparticle volume, k_{B} is the Boltzmann constant, T is the thermodynamical temperature, x, y, z denote the Cartesian components, $\mu_i = \mu_i(\tilde{t})$ $(i = 1, 2, 3)$ are the independent Gaussian white noises with zero means, $\langle\mu_i(\tilde{t})\rangle = 0$ and delta correlation functions, $\langle\mu_i(\tilde{t})\mu_i(\tilde{t}')\rangle = \delta(\tilde{t} - \tilde{t}')$.

The resulting reduced field ($\mathbf{h} = \mathbf{H}/M$) acting on the nanoparticle consists of the dipole field and the external one

$$\mathbf{h}_k = \sum_{j=1,j\neq k}^{N} \mathbf{h}_{kj}^{dip} + h[\cos(\widetilde{\omega}\tilde{t})\mathbf{e}_x + \sin(\widetilde{\omega}\tilde{t})\mathbf{e}_y], \quad (4)$$

$$\mathbf{h}_{kj}^{dip} = \frac{4\pi}{3}\frac{3\boldsymbol{\rho}_{kj}(\mathbf{u}_j\boldsymbol{\rho}_{kj}) - \mathbf{u}_j\rho_{kj}^2}{\rho_{kj}^5}, \quad (5)$$

where $\widetilde{\omega} = \omega\tau_1$ and h are the dimensionless frequency and amplitude of the external circularly polarized field, respectively, $\boldsymbol{\rho}_{kj}$ is the vector joining two nanoparticles, $\mathbf{u}_j = \mathbf{M}_j/M$ is the reduced magnetic moment of the j-th particle. Finally, the forces acting on the nanoparticle can be written as

$$\mathbf{f}_k^{dip} = \sum_{j=1,j\neq k}^{N}\left[3\frac{\boldsymbol{\rho}_{kj}(\mathbf{u}_j\mathbf{u}_k) + \mathbf{u}_k(\mathbf{u}_j\boldsymbol{\rho}_{kj}) + \mathbf{u}_j(\mathbf{u}_k\boldsymbol{\rho}_{kj})}{\rho_{kj}^5} - 15\frac{\boldsymbol{\rho}_{kj}(\mathbf{u}_k\boldsymbol{\rho}_{kj})(\mathbf{u}_j\boldsymbol{\rho}_{kj})}{\rho_{kj}^7}\right], \quad (6)$$

$$\mathbf{f}_k^{sr} = 24\varepsilon\sum_{j=1,j\neq k}^{N}\frac{\boldsymbol{\rho}_{kj}}{\rho_{kj}^2}\left[\left(\frac{\sigma}{\rho_{kj}}\right)^{12} - \left(\frac{\sigma}{\rho_{kj}}\right)^{6}\right], \quad (7)$$

where σ, ε are the parameters of the Lennard-Jones potential, which models a steric repulsion. The power loss is calculated in common way (see Eq.9 in Ref. [8]) with averaging over the whole ensemble. The dipole field calculation is performed as described in [17]. The system of equations 3 was solved by the fourth-order Runge-Kutta method with the time quantification step of 0.005τ in the range of 1000τ (τ is the field period). The video-card Nvidia GeForce 450 GTS was used for our simulations.

III. Results and Discussion

Since in the real applications the ensembles, not single nanoparticles, are used, the inter-particle interaction can essentially impact the response to the external field. And even if the volume fraction is small enough (for example, 1 %), due to the long-range dipole interaction and interaction caused by the surfactant covering of each nanoparticle, the results can be different compared with the single particle approximation. Firstly, the dipole interaction intends to join particles into dense clusters. Such formations are extremely undesirable on the reasons of further metabolism and excretion. Secondly, to prevent this clustering, the nanoparticles are coated with a surfactant providing repulsion. Competition of the above mentioned interactions can modify the specific power loss of each nanoparticle in a wide range, that is in focus of our investigation.

In a wide sense, the inter-particle interaction increases the magnetic energy and there are two consequences of this. From the one hand, the regular component of motion becomes strong due to the interaction, and the stochastic component, in contrast, is suppressed. And at a glance, such suppression can results in the increase in the power loss. From the other hand, the interaction fixes the nanoparticles magnetic moments that complicates the response to the external field. And this trend leads to the decrease in the power loss. Due to the long-range character of the dipole interaction, it is too hard to estimate its role in the energy absorption of the external periodic field by a ferrofluid. And to give a quantitative assessment, we performed a set of numerical simulations.

As it was mentioned above, the interaction is followed by the convergence of nanoparticles and cluster formation, which obstruct further translational and rotational motion of particles. At the same time, the clusters are an origin of a few phenomena affecting the power loss. Thus, each nanoparticle tries to reduce its energy and gets the equilibrium or quasi-equilibrium state. And due to the thermal fluctuations, the nanoparticles can switch between different states. These switchings break the alignment in the clusters and frustrate the nanoparticles magnetic moments. Under some circumstances this can leads to the increase in the power loss. Finally, large fluctuations can destroy the clusters completely and the nanoparticles response to the external field becomes better. As a result, the power loss increases that can be interpreted as the constructive role of thermal noise. We have studied in-depth all these phenomena including the influence of the system parameters on their conditions of occurrence. In this regard, the volume fraction, noise intensity, and surfactant characterizations are the most interesting.

Let us consider the process of cluster formation in detail. To minimize the magnetostatic energy, the nanoparticles should be closer to each other. Then the magnetic moment of each particle should be oriented along the resulting dipole field generated by other particles. Since the magnetic lines of force are closed curves, there are two trends. Firstly, the nanoparticles magnetic moments try to be aligned along one direction and this leads to the chain-like cluster formation. Secondly, the chain fragments tend to be arranged in the antiparallel way and attract each other forming the antiferromagnetic structure. The surfactant tries to prevent such agglomeration due to steric repulsion. The competition between these two types of interaction can lead to quite different results. We need to underline that since the magnetization is important for the performance of the hyperthermia method, it is reasonable to synthesize nanoparticles with magnetisation as large as it is possible. As a consequence, the intensity of the dipole interaction will increase and the clusters will become denser. Therefore, the actuality of the considered aspects will grow in time.

Different system parameters affect on the cluster formation process differently, and this influence is not always unambiguous. As a rule, increase in the volume fraction promotes the nanoparticles agglomeration that results in decay of the power

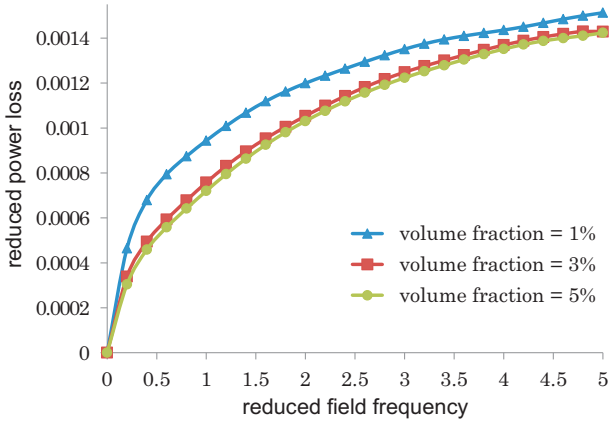

Fig. 1. Simulation results: the volume fraction influence without clusters. The system parameters: number of nanoparticles 4096, $M = 310$ G, $R = 10$ nm, $\eta = 0.006$ P, $h = 0.05$, $\kappa = 10$.

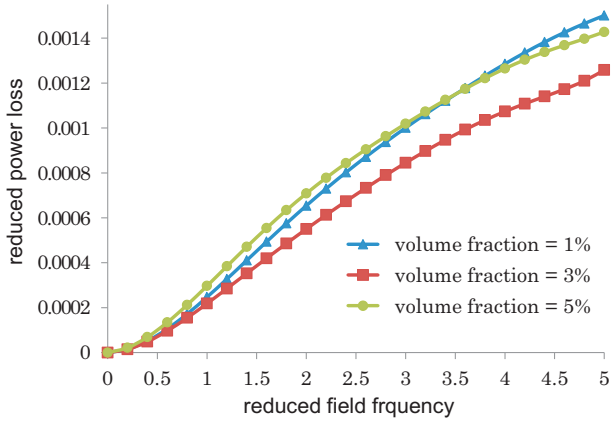

Fig. 2. Simulaion results: the volume fraction influence with clusters formation. The system parameters: number of nanoparticles 4096, $M = 310$ G, $R = 10$ nm, $\eta = 0.006$ P, $h = 0.05$, $\kappa = 25$.

loss (see Fig. 1). We explain this by various cluster types. When the volume fraction is small, the chain-like structures are formed. They are characterized by weaker interaction and more sensitive to the external field. For larger volume fraction, the short chain fragments join each other forming denser structures with the stronger interaction. And these aggregated structures have a weak response to the external field. But there are some exceptions from this trend. Firstly, when the noise intensity is small enough, the role of the nanoparticles concentration can be negligible. This happens because when the noise is small, the formed clusters for different volume fractions have the similar structure and remain stable. Secondly, the inverse relationship of the power loss on the volume fraction can occur that is shown in Fig. 2. When the inter-particle interaction in the clusters is strong, remagnetization of the whole cluster requires a stronger field, and, as a consequence, hysteresis loop widens. This is the origin of the increase in the power loss on the volume fraction for small frequencies (the curve for 5% exceeds the curve for 3% in Fig. 2). This effect disappears for larger frequencies, when the clusters do not reverse completely during the field period, because strong interaction suppresses the dynamics of each particle. Therefore, in contrary, the power loss becomes smaller in comparison with the case of a lower volume fraction.

When the temperature increases, the thermal fluctuations break the order, and this affects on the ferrofluid response differently. While the magnetic energy per one nanoparticle is larger than the thermal one, but not so large to exclude the essential fluctuations during the field period, a very interesting effect can occur. Each nanoparticle in cluster is in the equilibrium or quasi-equilibrium state, which is caused by the resulting dipole field. The nanoparticles magnetic moments fluctuate around these states predominantly. And when rare, but significant fluctuations occur, the nanoparticles perform transitions between the equilibrium states. Such phenomenon is similar to the relaxation of the magnetic moment in the fixed uniaxial nanoparticle, which was described in [19], or to the field-induced switching in the same particle, which was considered in [18]. The transition process proceeds fast enough, but during it each nanoparticle is in a frustrated state and characterized by a high energy in the external field. Therefore, the power loss increases, especially for high frequencies,

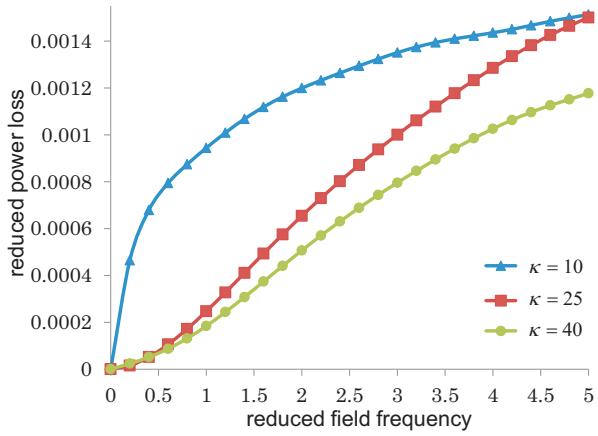

Fig. 3. Simulaion results: the influence of nanoparticles switching in clusters. The system parameters: number of nanoparticles 4096, $M = 310$ G, $R = 10$ nm, $\eta = 0.006$ P, $h = 0.05$, volume fraction 1%.

when the time of one transition becomes comparable with the field period. The phenomenon described requires a number of conditions, since a lot of factors influences the ratio between the thermal and deterministic energies, i.e. noise intensity, surfactant parameters and cluster types. Thus, in Fig. 3 this phenomenon is reflected in the behaviour of the curve for $\kappa = 25$. As seen, for small frequencies, the curves for $\kappa = 25$ and $\kappa = 40$ coincide. But while the frequency increases, the curve for $\kappa = 25$ tends to the curve for $\kappa = 40$ and intersects it.

Finally, when the thermal energy is comparable with the magnetic one, the thermal fluctuations completely prevent the cluster formation. Despite the noise suppresses the response of each nanoparticle to an external field, for the interacting ensemble the noise leads to quite different results. Since the nanoparticles in dense clusters are strongly bonded and weakly exposed to the external field, the thermal fluctuations increase the nanoparticle response, and, correspondingly, the energy absorbed from the external periodic field. We interpret this as the constructive role of noise. The results of the set of simulations confirming this phenomenon are depicted in Fig. 4. As seen, the power loss curves for a larger noise intensity are above than the curves for a smaller one. This

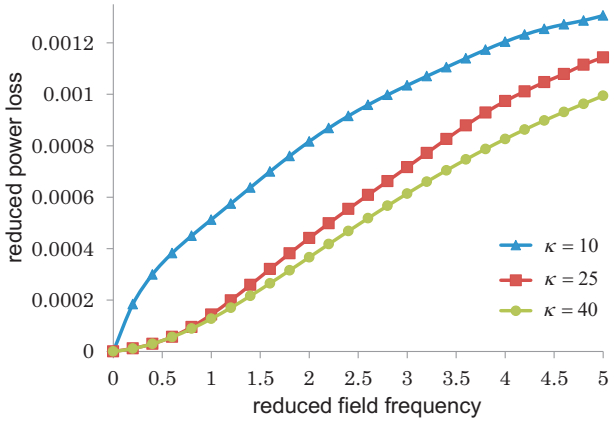

Fig. 4. Simulaion results: the influence of thermal fluctuations breaking the clusters. The system parameters: number of nanoparticles 4096, $M = 310$ G, $R = 10$ nm, $\eta = 0.006$ P, $h = 0.05$, volume fraction 3%.

is especially expressed for high frequencies, when the field-induced oscillations of the nanoparticles leading to destruction of the clusters. At the same time, at low frequencies the curve for $\kappa = 25$ almost coincides with the curve for $\kappa = 40$ because in both cases the similar clusters are formed, and they are not completely broken by the thermal noise.

IV. Conclusions

We have considered the response of an ensemble of uniaxial ferromagnetic nanoparticles placed into a fluid to an alternating field in the presence of thermal bath. The attention was paid to the absorbtion of the field energy as a result of the nanoparticles rotation in a viscous fluid. The influence of the inter-particle interaction effects was studied numerically based on the effective Langevin equation. Using the Barnes-Hut algorithm and CUDA technology, the set of numerical results, which describes the frequency dependence of the power loss, has been obtained for various system parameters.

Since the power loss of the ensemble is defined by the dynamics of each particle, the inter-particle dipole interaction has a critical impact to the ensemble susceptibility to an external periodic field. Due to the cluster formation, each nanoparticle is in the strong in the characteristics can cause quite different structures of the ensemble. Therefore, the power loss is sensitive to the system parameters. The interaction effects especially are actual for low frequencies, when clusters inverse the magnetization during the field period.

The thermal noise and the inter-particle interaction are the competing factors. The fluctuations can partially break the nanoparticles order in the clusters or destruct the clusters completely. In the first case, the effect of significant increase in the power loss for high frequencies is observed. We explain this by rare switchings of the nanoparticles in the clusters between the quazi-equilibrium states formed by the resulting dipole field. The switching process is characterised by a frustrated state, within which the energy consumption is high enough. The resulting power loss here can be almost equal to the value of the single particle limit.

When the noise intensity is large, the clusters are destroyed, but in the interacting ensemble this leads to the power loss increase as a consequence of the better response of the nanoparticles to an external field. And under certain other conditions, the larger noise intensity corresponds to the larger power loss values. Therefore, we talk about the constructive role of thermal noise.

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