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A numerical perspective on the relation between particle rotational inertia and the equilibrium magnetization of a ferrofluid

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ABSTRACT

This work explores, from a numerical perspective, the role of particle rotational inertia on the magnetization dynamics of ferrofluids. A robust numerical method is used for this purpose. The numerical research code is based on the use of a convergent long range dipolar interactions technique. These interactions are computed through a sophisticated Ewald summation procedure. The balance of linear and angular momentum is solved for $\mathcal{N}$ ensembles containing N particles each. Long range dipolar magnetic torques are solved in a periodic system of Lattices, spread in physical and reciprocal spaces to assure the convergence on the calculation of the suspension transport properties. A small effect of particle rotational inertia is considered. The system of equations of N particles distributed randomly in space is solved simultaneously for different parallel realizations in order to achieve a meaningful statistics of our many-body system. The results are focused on the behavior of the suspension magnetization for different particle concentrations and intensities of rotational inertia. The physical parameter used to express this effect is the particle rotational Stokes number. The simulations indicate that, from a numerical perspective, rotational inertia may induce a relevant, but often neglected, effect on the magnetization equilibrium of a ferrofluid. This finding is relevant for the community of numericists interested in using Langevin Dynamics applied to dipolar suspensions. We propose an expression with a correction on the effect of the particle rotational inertia to compute the magnetization of a magnetic fluid. The results obtained in this work are compatible with some insights previously pointed out in former scientifical works.

1. Introduction

A magnetic fluid is a colloidal, stable suspension of nanometric magnetic particles immersed in a viscous carrier liquid [1,2]. The suspension stability is achieved due to two mechanisms. The first important stabilizing mechanism is associated with the size of the particles. In the nanoscale, the particles are subjected to translational and rotational Brownian forces and torques [3,4]. These random fluctuations end up randomizing the suspension microstructure, avoiding differential sedimentation and particle aggregation due to van der Waals or dipolar interparticle forces [5,6]. The second stabilizing mechanism is related to the presence of surfactants used to induce steric repulsion and avoid particle aggregation.

Since the discovery of ferrofluids in the 60s, scientists and engineerings have been using them in many relevant applications. These applications include: drug targeting [7], stabilization of fluidized beds [8,9], magnetic pumping [10], cappilary ascension and fluid displacement in porous media [11], optimization of heat transfer processes [12], tumor treatment through magnetic hiperthermia [13] and the list goes on.

In order to understand the microstructural behavior of a magnetic fluid, we often reccur to the use of numerical simulations. These simulations are usually performed under the framework of Langevin Dynamics. This technique consists in solving stochastical differential equations for the translational and rotational velocities of each particle on the simulation domain [6]. The use of this technique allows us to explore not only the behavior of transport properties of the suspension, but also to study structure transition [14] without the costs and complications of building expensive experimental setups. The idea behind the formulation of the Langevin stochastic differential equation is quite simply. The greater challenge is to properly formulate the expressions for long range interacting forces and torques.

For interacting particulate systems, it is well known that depending on the transport property of interest, numerical problems regarding convergence may appear for slow-decay

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interactions [15]. This problem is more severe for hydrodynamic interactions, which decay with $1/r$, begin r the distance between the center of the particles. When dealing with magnetic suspensions, the slow decay of long range dipolar torques – $1/r^3$ – also requires a special treatment to assure the correct computation of the suspension magnetization [6]. This special treatment consists in writting the expressions for magnetic torques in a periodic space using the Ewald summation technique [16]. This procedure emulates an infinite system (in the thermodynamic limit) with a finite number of interacting particles and is described in a very clear manner in a series of works of Wang et al. [17–19]. Recently, the amount of numerical works focused on exploring the microstructural dynamic behavior of magnetic fluids has increased substantially [20–23]. The approach of Langevin Dynamics applied to the study of magnetization dynamics of ferrofluids seems to be a quite effective technique. Recent works based on this method [6,18] were able to recover with a good agreement some theoretical predictions on the equilibrium magnetization of concentrated ferrofluids [24–26]. It is important to highlight that these numerical works have not explored splicitly the role of particle rotational inertia. Moreover, it is common sense that the approximation of vanishing mass seems to have no impact whatsoever on the equilibrium magnetization of a ferrofluid. Consider a suspension of Fe_2O_3 particles, with an average diameter of $d = 10$ nm and density $\rho_p = 5.240$ kg/m^3, and the mass of a single particle will be approximately $m \approx 2 \times 10^{-21}$ kg. This quick calculation gives us an idea of the smallness of the mass of a single magnetic particle in a ferrofluid. The vanishing mass limit implies in a vanishing inertia limit. However, it is known that the consideration of a small, but non-zero, artificial effect of particle inertia is convenient for numerical purposes when simulating Brownian systems [27]. The influence of this small effect of rotational inertia applied to Brownian dipolar suspensions has not been explored so far. Indeed, on the scientific literature there are only few works that studied how particle rotational inertia affects the imaginary suceptibility of ferrofluids under alternating magnetic fields [29,30]. But the numerical behavior of the steady state equilibrium magnetization under a static field has not been explored as a function of the intensity of particle rotational inertia.

In order to fulfill this gap, this work explores the role of particle rotational inertia on the magnetization dynamics of ferrofluids under the presence of long range dipolar interactions. The idea that rotational inertia may display an important role on the behavior of magnetic suspensions, was previously mentioned in a recent work of the author [31] in another context. In the mentioned paper, the authors found out that aggregation rates in noncolloidal dilute magnetic suspensions are severe influenced by particle rotational inertia. This work inteds to explore the influence of this mechanism on the magnetization dynamics of colloidal magnetic suspensions for non-diluted magnetic fluids from a numerical perspective.

2. Formulation of the problem

We consider a suspension of N rigid, spherical, magnetic particles denoted with a lower index i, where $1 \leqslant i \leqslant N$. The particles have radii a and dipole moments with orientation $\hat{d}_i$ as illustrated in Fig. 1. In a context of Langevin dynamics, where we are interested in determining transport properties of magnetic suspensions, the physics is ruled by a competition of Brownian motion and dipolar interactions. Hence, the equations of motion for an arbitrary i particle are based on the linear momentum balance equation (translational motion) and the angular momentum balance equation (rotational motion) accounting for Brownian and magnetic forces and torques.

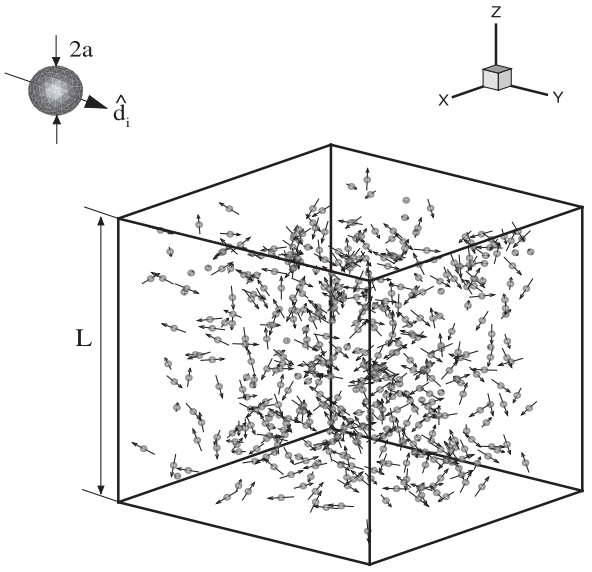

Fig. 1. Sketch of the problem.

For the translational motion of a non massive, neutrally buoyant particle ($m \ll 1$, where m is the mass of the particle and $\rho_s = \rho_f$, being ρ_s and ρ_f the densities of the particles and the fluid respectively), located in an arbitrary position $\boldsymbol{x^i}$ in space, subjected to a Stokes drag and magnetic and Brownian forces, we have

$$\boldsymbol{u^i} = \frac{1}{6\pi\mu a}(\boldsymbol{f_b^i} + \boldsymbol{f_m^i}), \frac{d\boldsymbol{x^i}}{dt} = \boldsymbol{u^i} \tag{1}$$

where μ is the fluid's viscosity, $\boldsymbol{u}^i$ is the velocity of the i^{th} particle, $\boldsymbol{f_b}$ and $\boldsymbol{f_m}$ represent the Brownian and magnetic forces respectively. The last term on the RHS of the balance of linear momentum (1) computes magnetic dipolar interactions between the particles. In our model we use a periodic system of Lattices to assure the computation of convergent magnetic interactions. The computation of magnetic dipolar interaction forces is made in a non-periodic fashion given its fast decay ($1/r^4$). The expressions for the magnetic forces due to dipolar interactions and for the Brownian forces are given [6] by

$$\boldsymbol{f_m} = \sum_{\substack{j=1 \\ i \neq j}}^{N} \frac{3\mu_0 m_i m_j}{4\pi r_{ij}^4} \left[(\boldsymbol{d_i} \cdot \boldsymbol{d_j})\hat{\boldsymbol{r}}_{\boldsymbol{ij}} + (\boldsymbol{d_i} \cdot \hat{\boldsymbol{r}}_{\boldsymbol{ij}})\boldsymbol{d_j} + (\boldsymbol{d_j} \cdot \hat{\boldsymbol{r}}_{\boldsymbol{ij}})\boldsymbol{d_i} - 5(\boldsymbol{d_i} \cdot \hat{\boldsymbol{r}}_{\boldsymbol{ij}})(\boldsymbol{d_j} \cdot \hat{\boldsymbol{r}}_{\boldsymbol{ij}})\hat{\boldsymbol{r}}_{\boldsymbol{ij}} \right],$$

$$\boldsymbol{f_b} = 6\pi\mu a \sqrt{\frac{6\mathcal{D}}{\delta\tau}}\boldsymbol{\xi}, \tag{2}$$

here μ_0 is the magnetic permeability of the free space ($\mu_0 = 4\pi 10^{-7} H.m^{-1}$), m_i and m_j are the intensity of the dipole moments of particles i and j, r_{ij} is the distance between the centers of particles i and j, $\boldsymbol{d_i}$ and $\boldsymbol{d_j}$ are the orientation of particles i and j dipole moments (unitary vectors), $\hat{\boldsymbol{r}}_{\boldsymbol{ij}} = (\boldsymbol{x_i} - \boldsymbol{x_j})/r_{ij}$, being $\boldsymbol{x_i}$ and $\boldsymbol{x_j}$ the positions of particles i and j in the free space, $\mathcal{D}$ is the Stokes–Einstein Brownian diffusion coefficient [3], $\delta\tau$ represents a Brownian time-step, which in the SIMS code is the simulation time-step and $\boldsymbol{\xi}$ is a random unitary vector with uniform distribution in the interval $[-1, 1]$. We will also assume in this work homogeinety in the particles material, so for any pair (i,i), we have $m_i = m_j = m$. The calculation of the particles position $\boldsymbol{x}$ is made by solving the kinematic equation