# Microstructural transition in an ordered set of magnetic spheres immersed in a carrier liquid 

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#### Abstract

This work explores the physics of an ordered set of interacting spheres immersed in a carrier liquid. We present numerical simulations that compute the translational and rotational motion of $N$ interacting spheres based on classical principles of Stokesian dynamics. The spheres are assumed to be made of a magnetizable material, subjected to magnetic and hydrodynamic long range interactions. We explore structure transition using a Lagragian approach of a continuum volume of fluid containing micrometric magnetic particles. We present local maps of particle volume fraction within the calculation Lattice. In this condition, considering the presence and absence of an applied magnetic field, instantaneous snapshots of the local microstructure are taken. Thus, different possibilities of long range interactions are considered. We also complement these results with meaningful statistics of time series obtained through our simulations, such as the correlation time of velocity fluctuations and their self-correlation functions. The data analyzed in the present work sustain the fact that initially ordered neutrally buoyant suspensions have an anisotropic memory-like behavior in the direction of an applied field. It is also observed that particles tend to form small isotropic clusters in the absence of an external field. However, hydrodynamic interactions tend to disperse the particulate phase, avoiding the formation of clusters. This finding suggests that hydrodynamic interactions may play a relevant role on the magnetization dynamics of ferrofluids.


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## 1. Introduction

Geometrically ordered structures appear in several fields of human knowledge. Crystals, foams, and solid molecular arrangements are some examples of ordered structures in nature. In fluid mechanics an ordered set of spheres can be used as a simplified model of a porous media [1,2]. Several scientists have been interested in exploring the physics of ordered structures in fluid mechanics. A pioneer work of [3] explored the lubrication film between interacting spheres and discussed the physics of Boerlage four-ball testing instrument for lubricants and additives. This instrument is an example of practical usage of ordered sets of spheres in fluid mechanics. Another example found in the scientific literature regarding the study of ordered suspensions of spheres is the work of [4], in which he explored rheological properties of ordered suspensions of polystyrene latex spheres suspended in

[^0]water. More specifically, [4] used spectrophotometric and conductance stopped-flow techniques to measure the internal relaxation time of the suspension structure when subjected to a step strain flow. In the 1990s the rheology and formation of ordered suspensions and colloidal structures [5,6] was studied.

More recently, the work of Ten Cate and Sundaresan [7] explored the unsteady forces in ordered arrays of monodisperse spheres. The authors used Lattice-Boltzmann numerical simulations to understand the forces generated by fluid-structure interaction. Another recent work [8] provided the theoretical background to compute the flow of a single sphere moving in creeping flow between parallel walls. This is important due to the long range nature of hydrodynamic interactions to which solid bodies moving in lowReynolds number flow are subjected. The same laws and theoretical background must be used to develop numerical simulations of an ordered set of spheres in creeping flow (one of the goals of this work). In other recent works Yeo et al. [9,10] performed numerical simulations of highly concentrated non-Brownian suspensions of spheres in confined shear flow and observed ordering transition. The list of problems in fluid mechanics involving ordered structures passes through foams [11], granular flows [12], ordered arrays of
spheres in heat and mass transfer applications [13,14], crystals [15] and creeping flow in organized structures of rotating spheres.

In the present work, we intend to deal with a special type of ordered suspension. We consider a set of non-Brownian, neutrally buoyant magnetic spheres suspended in a viscous fluid. Our goal is to explore structure transition due to different physical mechanisms which include: dipolar interaction, an external applied field and long range hydrodynamic interactions, besides a combination of all the three mentioned mechanisms. For this purpose we use a research code developed by the author [17]. The code uses a robust Ewald summation technique [19] to compute long range interactions. The periodic summation technique is applied over the Rotne-Prager mobility tensor [20] as first introduced by the pioneer work of Beenakker [21]. The goal of this work is to explore structure transition in non-Brownian liquid-solid magnetic suspensions and to understand the underlying physical mechanisms behind the formation of particle clusters with specific geometries. We also intend to explore the long time statistical behavior of particle velocity fluctuations induced by long range interactions among the spheres.

## 2. Formulation

We consider a set of $N$ magnetic spheres immersed in a viscous Newtonian liquid with zero initial velocity. The spheres are neutrally buoyant and are initially distributed in an ordered manner. They are subjected to magnetic and hydrodynamic long range interactions. We assume that the mass of each sphere is small, thus they have negligible inertia and can be treated through a mobility problem. The dimensionless equation that computes the velocity of each sphere is given by
$\boldsymbol{u}_{i}=\boldsymbol{M}_{i, i}^{s} \cdot \boldsymbol{f}_{i}+\sum_{j \neq i, j=1}^{N} \boldsymbol{M}_{i, j}^{p} \cdot \boldsymbol{f}_{j}$,
with
$\boldsymbol{f}_{i}=\boldsymbol{f}_{i}^{r}+\boldsymbol{f}_{i}^{c}+\boldsymbol{f}_{i}^{m}$.
Here $\boldsymbol{u}_{i}$ is the velocity of an arbitrary particle $i, \boldsymbol{M}_{i, i}^{S}$ is the self-mobility matrix, $\boldsymbol{M}_{i, j}^{p}$ represents the pair-mobility matrix, expressed as two sums in the physical $\mathcal{L}$ and reciprocal $\hat{\mathcal{L}}$ spaces, $\boldsymbol{f}_{i}$ and $\boldsymbol{f}_{j}$ denote the forces acting on particles $i$ and $j$ respectively, $\boldsymbol{f}_{i}^{r}, \boldsymbol{f}_{i}^{c}$ and $\boldsymbol{f}_{i}^{m}$ are the repulsive, contact and magnetic forces acting on an arbitrary particle $i$. We do not account for gravitational forces, since we consider a neutrally buoyant suspension. The matrices $\boldsymbol{M}_{i, i}^{S}$ and $\boldsymbol{M}_{i, j}^{p}$ are given by
$\boldsymbol{M}_{i, i}^{s}=\left(1-6 \xi \pi^{-1 / 2}+\frac{40}{3} \xi^{3} \pi^{-1 / 2}\right) \boldsymbol{I}$,
and
$\boldsymbol{M}^{i j}\left(\boldsymbol{r}^{\prime}\right)=\sum_{\boldsymbol{x} \in \mathcal{L}} \boldsymbol{M}^{1}\left(\boldsymbol{r}^{\prime}+\boldsymbol{x}\right)+\frac{1}{V} \sum_{\boldsymbol{k} \in \hat{\mathcal{L}}, \boldsymbol{k} \neq 0} \boldsymbol{M}^{2}(\boldsymbol{k}) \cos \left(\boldsymbol{k} \cdot \boldsymbol{r}^{\prime}\right)$.
where $\xi=\pi^{2} / V^{3}$ is a parameter set to accelerate the convergence of the sum, $V$ is the volume of a single Lattice, $I$ is the identity tensor, $\boldsymbol{r}^{\prime}$ is the vector connecting the edges of the central Lattice with the edge of the surrounding Lattices, $\boldsymbol{x}$ is the position of an arbitrary particle with respect to the origin of the system and $\hat{\boldsymbol{k}}=\boldsymbol{k} / k$ is the dimensionless wavenumber, related to the spectrum of particle positions by the relation $\boldsymbol{k}=2 \pi /\left(\boldsymbol{x}+\boldsymbol{r}^{\prime}\right)$. The mobility matrices $\boldsymbol{M}^{1}$ and $\boldsymbol{M}^{2}$ are given by the classic work of Beenakker [21].

The forces $\boldsymbol{f}_{i}^{r}, \boldsymbol{f}_{i}^{c}$ necessary to calculate each particle velocity as expressed in (1) are given in their dimensionless versions as
$\boldsymbol{f}_{i}^{r}=\Lambda\left|\boldsymbol{u}_{i}\right| e^{\left(-\epsilon_{i j} / \mathcal{Y}\right)} \hat{\boldsymbol{e}}_{r}, \quad \boldsymbol{f}_{i}^{c}=P_{c} \epsilon_{i j}^{3 / 2} \hat{\boldsymbol{e}}_{r}$,
where $\Lambda, \mathcal{Y}$ and $P_{c}$ are calibration constants of our model [17]. The variable $\epsilon_{i j}$ represents the dimensionless distance between the surfaces of particles $i$ and $j$. The expression used to compute magnetic forces due to long range dipolar interactions is given in details by Gontijo et al. [17,18].

For the angular motion, neglecting rotational inertia we have
$\boldsymbol{\omega}_{i}=\boldsymbol{T}_{m}^{i}$,
where $\omega_{i}$ denotes the angular velocity of an arbitrary particle $i$ and $\boldsymbol{T}_{m}^{i}$ represents magnetic torques due to Longe range dipolar interactions between the particles and to field-particle interaction. The complete expression for $\boldsymbol{T}_{m}^{i}$ is provided in the works of Gontijo et al. [17,18].

The physical parameters that appear in the dimensionless system of governing equations are denoted by $\varphi_{m}$ and $\psi_{m}$. These parameters are defined respectively as follows:
$\varphi_{m}=\frac{\mu_{0} m_{d}^{2}}{8 \pi^{2} \eta a^{5} U_{0}} \quad$ and $\quad \psi_{m}=\frac{\mu_{0} m_{d} H_{0}}{6 \pi \eta a^{2} U_{0}}$,
where $U_{0}$ is a typical velocity of the problem (i.e. in a magnetic suspension $U_{0}$ could be the average velocity of approximation between two magnetic particles, during the formation of a dimmer), $\mu_{0}$ is the magnetic permeability of the free space, $m_{d}$ is the magnitude of the magnetic dipole moment of a single particle, $H_{0}$ is a typical value of the imposed external magnetic field, $a$ is the radius of the particle and $\eta$ is the viscosity of the surrounding fluid.

## 3. Results and discussions

Fig. 1a shows the initial local volume fraction distribution within the calculation domain (primary Lattice in the physical space) for


Fig. 1. (a) Side view in the plane $z y$ of the local concentration field for $t=0$. (b) Initial distribution of particles.

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