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# Symmetry breaking of particle trajectories due to magnetic interactions in a dilute suspension

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#### ABSTRACT

This work presents a numerical study of the relative trajectories of two magnetic particles interacting in a dilute suspension. The suspension is composed of magnetic spherical particles of different radius and density immersed in a Newtonian fluid. The particles settle relative to one another under the action of gravity and, when in close proximity, exert on each other magnetic force and torque due to their permanent magnetization. The equations of motion for both translation and rotation of the particles are solved and particle inertia is included in the calculation. The numerical simulations are based on the direct computations of the hydrodynamic and of the magnetic interactions between the rigid particles in the regime of non-zero Stokes number. A detailed study of the relative trajectories of two magnetic particles in a dilute suspension allows us to explore irreversible interactions that lead to particle aggregation and particle migration induced by the breaking of the time reversibility of the creeping flow due to magnetic effects. The calculation shows that the rotation of the particles produced by magnetic interactions change significantly the dynamics of collisions of magnetic particle.

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#### 1. Introduction

In several chemical or industrial processes, it is important to separate solid material from a fluid phase. One possibility to accelerate the settling rate of particles in a suspension is to promote aggregation of the solid phase [11,24]. For isolated particles, the settling velocity is given by Stokes's law (e.g. [2]), and is proportional to the square of the particle radius. For submicron particles, Brownian forces exerted by the fluid molecules prevent differential sedimentation caused by gravity. When aggregates are formed, the tendency is to promote a faster separation process by the action of gravity. The mechanics of aggregation is still a subject of vast attention in the specialized literature (e.g. [1,12]).

The effect of pair interactions of two non-magnetic spheres for zero inertia was first determined by Batchelor and Green [3]. In fact, two-body interactions in a suspension that obey time-reversal symmetry cannot be the cause of a mechanism for mixing or diffusion. In a dilute suspension, the spheres will return to their initial streamlines at the end of all two-particle

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encounters, owing to the linearity of the flow when only viscous forces are present. The far-field magnetic interactions, which decay like  $1/r^4$  for force and  $1/r^3$  for torque, must yield permanent displacements that lead to fluctuations in the particle motion and, therefore, to irreversible trajectories.

Because zero Reynolds number flows are symmetrical under time reversal, one is forced to look elsewhere to understand non-equilibrium mixing enhancement. In some suspensions, there is an interparticle force capable to promote aggregation such as the case of magnetic suspensions. In these suspensions, collisions may be induced by differential sedimentation or Brownian motion and aggregates can be formed by magnetic interaction between the dipole moments of the particles. Recent examples of studies of magnetic aggregation are Mehta et al. [19], who have developed a method of detection and estimation of aggregates with the same size in otherwise stable magnetic fluids, and Shimada et al. [25], who reported successful extraction of magnetic clusters from a suspension, and the creation of a system that allows self-assembly.

One important physical property to study on the dynamics of aggregation in suspensions is the rate of doublet formation. One of the first works to approach the subject was developed by Smoluchowski [26], in which the particles were submitted only to a sticking force. Zeichner and Schowalter [29] included the effects of hydrodynamic interaction and interparticle force, computing

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the rate of aggregation of a dispersion of equal particles subject to simple shear and to uniaxial extensional flow. Curtis and Hocking [8] considered shear flows and reported experimental results showing the role of van der Waals forces in particle aggregation. In studies considering aggregation induced by gravity, Davis [9] investigated the influence of van der Waals and Maxwell slip in the collision efficiency.

Related problems of particle trajectory analysis considering spherical particles in simple shear to investigate the rheology of dilute suspension of rough particles have also been considered (e.g. [28]). The rheology will depend on the model chosen to describe the surface roughness of the particles and the contact between them (e.g. [5,10]). Cunha and Hinch [5] showed that even very small asperities on the particle surface (less than  $10^{-4}$  of a particle radius) might play a significant role during the encounters between a pair of particles. Other sources of symmetry breaking of particle trajectories were also studied in the literature, such as electrostatic pair interactions between two colloids [4], particle inertia [27] and deformation of particles, for instance in drops [18]. More recently, Ingber and Zinchenko [14] have studied, in dilute suspension of spheres, hydrodynamic interactions in Couette and Poiseuille flow at low Reynolds number.

Despite the progress in many aspects of this subject, there is still much to be understood regarding magnetic effects on particle encounters. Lacis and Gosko [17] have developed an algorithm to simulate the behavior of a polarized suspension under a magnetic field in the presence of hydrodynamic interactions. Cunha and Couto [7] have recently examined a magnetic suspension in the presence of hydrodynamic and magnetic interactions. They made calculations of the aggregate rate in a dilute polidisperse suspension composed of non-Brownian magnetic spherical particles. However, the simplification of an inertialess and torque-free particle (i.e. particles with no rotation) was an important restriction to explore the effects of magnetic interactions in full detail.

The present analysis consists in observing the behavior of the relative trajectories of the magnetic particles in order to study the intensity of the breaking of the reversibility of the trajectories associated with magnetic interactions, when the particles are not torque free, but feel the presence of either another magnetic particle, or an external magnetic field, through both forces and torques. In this work, we shall, therefore, present a formulation to study the dynamics of a very dilute magnetic suspension. The main differences of the present study and previous calculations (e.g. [6,7]) are: firstly, we consider particle rotation due to dipoledipole interactions. This adds a new equation for the orientation of the particles, for which the torque balance on the particles has to be known. Secondly, we account for the inertia of the particles and propose a resistance formulation of the problem, which implies in solving two second order equations for translation and rotation of the particles. We show typical plots of relative trajectories obtained from this model and use them to characterize the source for the diffusive behavior in a dilute suspension of magnetic spherical particles and its relevance to the problem of aggregation in magnetic suspensions.

#### 2. Formulation of the problem

The flow examined here consists of pair of spherical particles of radius  $a_1$  and  $a_2$ , densities  $\rho_1$  and  $\rho_2$ , immersed in a viscous fluid of viscosity  $\eta$ . The particles have masses  $\mathcal{M}_1$  and  $\mathcal{M}_2$  and interact magnetically and hydrodynamically. They are free to rotate with angular velocities  $\omega_1$  and  $\omega_2$  and have individual magnetic dipole moments  $\mathbf{m}_1$  and  $\mathbf{m}_2$ . The particles are forced to encounter due to differential sedimentation in an environment

where the gravity acceleration is equal to  $\mathbf{g}$ , and also due to the presence of an external magnetic field  $\mathbf{H}_0$ .

The linear momentum equations that describe the motion of each particle are given by

$$\mathcal{M}_{1} \frac{d\boldsymbol{u}_{1}}{dt} = \frac{4}{3} \pi \Delta \rho_{1} \boldsymbol{g} a_{1}^{3} + \left[ 6\pi \eta a_{1} \left( \frac{6D_{t}}{\delta \tau} \right)^{1/2} \right] \boldsymbol{\xi} + \boldsymbol{f}_{h1} + \boldsymbol{f}_{m1}, \tag{1}$$

$$\mathcal{M}_2 \frac{d\mathbf{u}_2}{dt} = \frac{4}{3} \pi \Delta \rho_2 \mathbf{g} a_2^3 + \left[ 6\pi \eta a_2 \left( \frac{6D_t}{\delta \tau} \right)^{1/2} \right] \boldsymbol{\xi} + \boldsymbol{f}_{h2} + \boldsymbol{f}_{m2}, \tag{2}$$

where the sub indices 1 and 2 represent the properties of particles 1 and 2, respectively,  ${\bf u}$  denotes the particle velocity, t is time,  $\Delta \rho_1 = \rho_1 - \rho_f$  and  $\Delta \rho_2 = \rho_2 - \rho_f$  represent the density difference between the carrier fluid and each of the particles,  $D_t$  is the coefficient of Brownian diffusion, given by the classical fluctuation-dissipation theorem [24],  $\delta \tau$  is a typical time-step related to the Brownian motion,  $\xi$  represents a random unitary vector associated with Brownian forces,  ${\bf f}_h$  denotes the forces associated to hydrodynamic interactions on each particle and  ${\bf f}_m$  are the forces resulting from magnetic interactions.

The governing equations describing the rotational motion of the particles are expressed in terms of the torque balance equations for both particles 1 and 2, which gives

$$J_{1}\frac{d\omega_{1}}{dt} = -8\pi\eta a_{1}^{3}\omega_{1} + \left[8\pi\eta a_{1}^{3}\left(\frac{6D_{r}U_{s}}{a_{1}\delta\tau}\right)^{1/2}\right]\xi + \mathbf{T}_{m1},\tag{3}$$

$$J_{2}\frac{d\omega_{2}}{dt} = -8\pi\eta a_{2}^{3}\omega_{2} + \left[8\pi\eta a_{2}^{3} \left(\frac{6D_{r}U_{s}}{a_{2}\delta\tau}\right)^{1/2}\right]\xi + \mathbf{T}_{m2}.$$
 (4)

Here, J is the polar moment of inertia of the particles,  $D_r$  is the coefficient of rotational Brownian diffusion coefficient,  $T_m$  is the torque due to magnetic interaction of the particles and  $U_s = 2\Delta \rho_1 g a_1^2/9\eta$  is Stokes velocity of particle 1. Note that this formulation does not consider torques induced by hydrodynamic interactions. The calculation of the magnetic and hydrodynamic forces and torques are discussed in the following sections.

#### 2.1. Hydrodynamic interactions

The calculation of the hydrodynamic forces acting on the particles is based on a resistance formulation, as proposed by Jeffrey and Onishi [15]. Since Stokes equations are linear and quasi-steady, there is a linear relation between the velocity of the particles and the forces originated from the hydrodynamic interactions. The forces  $f_{h1}$  and  $f_{h2}$ , in Eqs. (1) and (2), are given by Jeffrey and Onishi [15]

$$\begin{bmatrix} \boldsymbol{f}_{h1} \\ \boldsymbol{f}_{h2} \end{bmatrix} = \eta \begin{bmatrix} \boldsymbol{A}_{11} & \boldsymbol{A}_{12} \\ \boldsymbol{A}_{21} & \boldsymbol{A}_{22} \end{bmatrix} \begin{bmatrix} \boldsymbol{u}_1 \\ \boldsymbol{u}_2 \end{bmatrix}, \tag{5}$$

where  $A_{11}$ ,  $A_{12}$ ,  $A_{21}$  and  $A_{22}$  are second rank tensors, given in index notation by

$$A_{ii}^{\alpha\beta} = X_{\alpha\beta}^{A} \hat{r}_{i} \hat{r}_{i} + Y_{\alpha\beta}^{A} (\delta_{ii} - \hat{r}_{i} \hat{r}_{i}). \tag{6}$$

Here, the indices  $\alpha$  and  $\beta$  denote the position that these tensors occupy in the resistance matrix, the indices i and j are the free indices,  $\hat{r}$  is the unit vector connecting the center of the particles and  $X_{\alpha\beta}^A$  and  $Y_{\alpha\beta}^A$  are called the resistance functions that depend on the dimensionless distance between the center of the particles s and on the density ratio of the particles  $\lambda$ ,

$$s = \frac{2r}{a_1 + a_2} \quad \text{and} \quad \lambda = \frac{\rho_2}{\rho_1}. \tag{7}$$

A detailed derivation of the resistance functions can be found in Jeffrey and Onishi [15].

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