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HYDRODYNAMIC SELF-DISPERSION IN A DILUTE SEDIMENTING SUSPENSION

Otávio Luís da Silva Rosa
Francisco Ricardo Cunha
University of Brasília, Department of Mechanical Engineering-FT
Campus Universitário, 70910-900 - Brasília, DF, Brasil

Abstract. *Microstructural changes, that is an important feature for the understanding of velocity fluctuations in sedimentation is investigated with numerical simulations. The simulations are used to describe hydrodynamic dispersion in a suspension of interacting particles sedimenting in a rectangular box with periodic sides and impenetrable bottom and top. It is observed how the positions of the particles evolve in a finite container. The suspension that was initially random in the gravity direction only, tends to be fully randomized as a result of the relative arrangements of the particles and the hydrodynamic interactions between them. The computer simulations suggest diffusivities dependent on the size of the simulated system but with anisotropy nearly independent of this.*

Keywords: *Velocity fluctuations, Sedimentation, Microstructure, Dispersion*

1. INTRODUCTION

Suspended particles subject to sedimentation do not generally move relative to the fluid with a constant velocity, but instead experience diffusion-like fluctuations in velocity due to interactions with neighboring and the resulting variations in the microstructure or configuration of the suspended particles. These velocity fluctuations are observed in non-Brownian suspension flows with very small particle Reynolds numbers. Such fluctuations have a long-time behavior characteristic of diffusion processes and their effect is now called hydrodynamic self-dispersion (Cunha 1995). This dispersion phenomenon is important for understanding of mixing process which inhibit separation (Davis 1996). The related phenomenon of shear-induced hydrodynamic diffusion in sheared monodisperse suspensions of spheres have been investigated experimentally (e.g. Leighton and Acrivos 1987) and theoretically (e.g. Cunha & Hinch 1996a). As a suspension contains many particles dispersed in a fluid, the motion of one particle creates velocity and dynamic pressure field that exert forces on the neighboring particles and affect their motion. At low Reynolds numbers such a disturbance flow is propagating via fluid by the mechanism of vorticity diffusion, and because of this we say that the particles interact hydrodynamically. Long-range multibody hydrodynamic interactions then play a key role in the motion of an individual sphere settling in the midst of a suspension of like non-Brownian spheres. After Batchelor (1972) the average velocity in sedimentation can be successfully predicted theoretically. In the present time, however, there is a discrepancy between experiments (Nicolai & Guazzelli 1995) and numerical simulations (Ladd 1993, Koch 1994 and Cunha 1995 and Ladd 1997) regarding the dependence of the sedimentation velocity variance on the container size. Experiments performed by Nicolai & Guazzelli (1995) using *well-stirred* suspensions showed that the values of the velocity fluctuations did not vary significantly when the vessel width was increased by a factor of four; in contrast, Cunha (1995) reported that computer simulations of velocity variances in sedimentation using random and independently initial distributions of particles in all directions of the space yield results which did increase with increasing the container size, following a parameter $O(U_s^2\phi\ell/a)$ according to the theory (Caflish & Luke 1985) and scaling argument proposed (Hinch 1988), where $U_s = 2\Delta\rho a^2 g/9\mu$ is the fall speed of an isolated sphere of radius a, ϕ is the volume fraction, and ℓ is the size of the container, μ is the fluid viscosity, $\Delta\rho$ denotes the difference between the density of the solid particles and fluid, μ is the fluid viscosity, $\mathbf{g}$ is the acceleration due to gravity. A well-stirred experimental suspension may not be a suspension where the particles are randomly positioned as assumed by those numerical simulations, though.

The screening mechanism of Koch and Shaqfeh (1991) is the only presently available theory that could lead to velocity fluctuations independent of the vessel size. However, the experiments (Nicolai et. al. 1995) and the lattice-Boltzmann numerical simulations of large-scale systems by Ladd (1997) were unable to verify the hypothesis that there is a deficit of one particle surrounding any given particle of the suspension assumed by this screening theory. At very low concentrations in a thin box, Segre, Herbolzheimer and Chaikin (1997) found a $\phi^{1/3}$ dependence, and an independence of the wider of the horizontal dimensions if it exceeded a certain correlation length $\ell_c = 10a\phi^{-1/3}$. Curiously this observed correlation length in the velocity fluctuations is somewhat greater than the narrower of the horizontal dimensions.

Fluctuations are often sensitive to subtle changes in the underlying suspension microstructure, which are difficult to observe experimentally. There should be some suspension microstructure that leads velocity fluctuations depending on the particle volume concentration only. Searching for other microstructures than the ones already simulated by Cunha (1995) and Cunha & Hinch (1996b), a new kind of initial distribution of particles is proposed and tested in this work. Numerical simulation is then used to examine the microstructural changes directly, as well to predict their effect on particle hydrodynamic dispersion.

2. DESCRIPTION AND FORMULATION OF THE PROBLEM

2.1. Scaling Argument

Consider a random monodisperse dilute suspension. Assume that $m = \Delta\rho a^3$ is the net particle mass, and a typical statistical fluctuation in density ρ'_ℓ, of a region of size

$\ell \gg a$ of such suspension scales as (Hinch 1988)

$$\rho'_\ell \sim m\sqrt{N}/\ell^3, \tag{1}$$

where $\sqrt{N}$ is a typical fluctuation in the number of the particles. Hence when a box of volume ℓ^3 containing N particles is divided into two equal parts by a vertical plane, one half of the box will contain $(\frac{N}{2} - \sqrt{N})$ particles, whereas the other half will contain $(\frac{N}{2} + \sqrt{N})$. This unbalance drives convection currents during the sedimentation process.

Now, the density number fluctuation leads to a fluctuation in the weight of $mg\sqrt{N}$. At the time scale it takes vorticity to diffuse over the length ℓ, $t \sim \rho\ell^2/\mu$, this buoyancy force is balanced by the viscous drag associated with the driven flow ${U'}_\ell$. Hence

$$mg\sqrt{N} \sim \mu\ell {U'}_\ell. \tag{2}$$

Then, by substituting $g \sim a^{-2}\mu U_s/\Delta\rho$ and $N \sim \phi(\ell/a)^3$ into the above equation, one obtains

$$U'^2_\ell \sim U_s^2\phi\frac{\ell}{a}. \tag{3}$$

If the particles velocity remain correlated by a time $O(\ell/{U'}_\ell)$, the hydrodynamic self-diffusivity scales as

$$D_\ell \sim aU_s\phi^{1/2}\left(\frac{\ell}{a}\right)^{3/2}. \tag{4}$$

The above scaling argument helps to explain how velocity fluctuations and hydrodynamic dispersion in a random dilute monodisperse suspension may be dependent on the system size (for more details see Cunha 1997), and why in this article we are proposing an initial non-homogeneous distribution of particles that is random in the gravity direction only. The main purpose of this rather unnatural kind of distribution is to try to eliminate the presence of convection-driven secondary flows in the horizontal directions, what in turns would influence the velocity fluctuations of the particles. For this end, the initial configurations used in all simulations now are generated placing the particles "regularly" in the horizontal directions, and random and independently in the vertical (parallel to the gravity) direction inside a rectangular box of dimensions $\ell \times \ell \times \ell$. The distribution is made so that each particle center is separated from the other ones by a distance greater than the dimensionless particle diameter in the same configuration, avoiding overlaps. A typical initial configuration can be seen in Fig. 1 of our previous article (Cunha & Silva Rosa 1998).

2.2. Governing Equations

Consider non-Brownian rigid spherical particles sedimenting in a incompressible Newtonian fluid of dynamic shear viscosity μ and density ρ, since the inertial effects of the fluid are negligible and the time scale are large compared to the viscous relaxation time (a^2/ν), the appropriate equations of the fluid motion (u, p) in the usual Eulerian description of an inertial frame are the pseudo-steady Stokes equations. Owing to the long range of the velocity disturbance in a dilute sedimenting suspension, the point-force approximation may be used for describing the hydrodynamic with fluid velocity governed by (Saffman 1973)

$\nabla \cdot \mathbf{u} = 0$ for the mass balance and

$$-\mu \nabla^2 \mathbf{u} + \nabla p = \sum_{\alpha} \mathbf{f}^{\boldsymbol{\alpha}} \delta(\mathbf{x} - \mathbf{x}_{\boldsymbol{\alpha}}) - \langle \mathbf{f} \rangle n, \tag{5}$$

corresponding to the momentum equation, where n is the number of particle, N, per unit of volume, $\delta(\mathbf{x} - \mathbf{x}_{\boldsymbol{\alpha}})$ denotes Dirac's delta distribution, and $\mathbf{f}^{\boldsymbol{\alpha}}$ is the hydrodynamic force exerted on the fluid by the particle α. In the absence of particle inertia, $\mathbf{f}^{\alpha} = \frac{4}{3}\pi a_{\alpha}^3(\rho_\alpha - \rho)\mathbf{g}$, which is just the net force of gravity with the buoyancy removed. $\langle \mathbf{f} \rangle = (1/N)\sum_\alpha \mathbf{f}^\alpha$ is the average force the particle exerted on the fluid. In particular, we are interested in the solution in which all components of the velocity field (u, v, w) are periodic in x and y with period ℓ, the horizontal components u, v periodic in z with period h, but the vertical component w satisfying an impenetrable bottom and top condition of vanishing vertical velocity.

2.3. Mobility Problem

The problem of N spherical particles free of inertia settling within an impenetrable container with periodic sides of dimensions $\ell \times \ell \times h$ has been formulated by Cunha (1995). Let $\mathbf{x}_\ell$ denotes the position of the particle m. Suppose an external force $\mathbf{f}^\ell$ is exerted on particle m and let $\mathbf{U}^\ell$ be its translation velocity. Then the appropriate formulation of hydrodynamic interaction which relates the velocities $\mathbf{U}^\ell$ and the forces $\mathbf{f}^\ell$ is given, in dimensionless terms, by

$$\mathbf{U}^\ell = \sum_{m=1}^{N} \left[\frac{3}{4} \sum_{\alpha} \mathbf{G}^{ps}(\mathbf{r}_\alpha, \mathbf{r}_{\alpha,s}, \mathbf{r}_{\alpha,i}) \cdot \mathbf{f}^m + \frac{3}{2\vartheta} \sum_{\beta}{}' \mathbf{J}^{rs}(\mathbf{k})\Theta \cdot \mathbf{f}^m \right], \tag{6}$$

with particle trajectories being obtained by integration of the kinematics equation

$$\frac{d\mathbf{x}^\ell}{dt} = \mathbf{U}^\ell, \qquad \mathbf{x}(0) = \mathbf{x}_o. \tag{7}$$

Here $\mathbf{G}^{ps}$ and $\mathbf{J}^{rs}$ are respectively the periodic stokeslet (Green Functions) in the physical and reciprocal spaces (Cunha 1995). These are obtained by Ewald summation using accurate computationally-eficient tabulation of incomplete gamma functions (Silva Rosa, 1998). $\mathbf{f}^m = -\frac{2}{9\mu} a^2 \frac{\Delta\rho}{U_s} \mathbf{g} = (0, 0, 1)$, ϑ is the volume of the periodic cell, and the relative position vectors of the singularities are defined as being $\mathbf{r}_{\alpha,s}$ and $\mathbf{r}_{\alpha,i}$. The Eqs. (6) and (7) will be applied to examine the dynamics of N point-particles sedimenting and interacting hydrodynamically within an impenetrable box. This type of equation represents a mobility problem with hydrodynamic interactions $O(N^2)$, calculated by using pairwise additivity (i.e. superposition of velocity) in the mobility matrix.

3. RESULTS AND DISCUSSIONS

In this section, it is presented the most relevant results of this work.

3.1. Statistical Analysis of the Suspension

The evolution Eqs. (6) and (7) are the heart of the dynamic simulation here. Given an initial configuration, the Eq. (7) is integrated in time to follow the evolution of the suspension microstructure. In order to use the simulations to determine macroscopic properties of the sedimenting suspension we derive below corresponding average expressions.

The determination of the structure factor of the suspension is the simpler way of obtaining the density number fluctuations. It is defined as being

$$F(\mathbf{k},t) = \frac{1}{N}\langle \hat{n}(\mathbf{k},t)\hat{n}^*(\mathbf{k},t)\rangle = \frac{1}{N}\left\langle \sum_{j,k}^{N} e^{2\pi i\mathbf{k}\cdot[\mathbf{x}_j(t)-\mathbf{x}_k(t)]}\right\rangle. \tag{8}$$

We were principally interested in looking at Fourier component corresponding to the box wave number $\mathbf{k} = (1/\ell, 1/\ell, 1/h)$ rather than particle-particle separation. The sedimentation velocity is given by the average velocity of the particles in the suspension defined as follows

$$\langle U_j\rangle = \left\langle \frac{1}{N}\sum_{i=1}^{N} U_j^i(t)\right\rangle \qquad j = 1,2,3. \tag{9}$$

The fluctuations in velocity of an individual particle in the suspension $U'^i_j(t) = U^i_j(t) - \bar{U}_j(t)$, is a measure of the deviation of the particle velocity from the mean velocity $\bar{U}_j$ calculated over all particles in the configuration $1/N\sum_{i=1}^{N} U_j^i(t)$ or at each time step in dynamical simulation. Hence, we define the variances of the sedimentation velocity as

$$\langle U_j^2(t)\rangle = \left\langle \frac{1}{N-1}\sum_{i=1}^{N}\left[U_j^i(t) - \bar{U}_j(t)\right]^2\right\rangle \qquad j = 1,2,3. \tag{10}$$

The normalized velocity fluctuations auto-correlation functions C_j were calculated in our simulations through the following expression

$$C_j = \frac{\langle U'_t(0)U'_t(t)\rangle}{\sqrt{\langle U'^2_j(0)\rangle\langle U'^2_j(t)\rangle}}, \tag{11}$$

and self-dispersion coefficients were calculated by integrating the velocity auto-correlation functions

$$D_j = \int_0^\infty \langle U'_j(0)U'_j(t)\rangle\, dt \qquad j = 1,2,3, \tag{12}$$

with the corresponding correlation τ^c times being estimated by the relation $\tau^c_j = \frac{D_j}{\langle U_j^2\rangle}$. Here the angle brackets denote a sum over all particles, and an average over all configurations or realizations (i.e. an average over time in dynamic simulation). A typical picture of the sedimentation process obtained with the dynamical simulation is displayed in Fig. 1. The picture was collected at different times throughout one arbitrary realization between those ones simulated with $\phi = 3\%$, $a/\ell = 0.05$ and 176 particles. Before the sedimentation began, the suspension microstructure was formed by 16 vertical parallel lines of 11 particles each, with stratification within the lines. When the sedimentation started, a mixing began to happen destroying the initial regular distribution in the horizontal directions. This mixing is due to the particle velocity fluctuations, that are caused here by the hydrodynamic interactions only. If the volume concentration was $\phi = 0\%$, there would not be any hydrodynamic fluctuation – what does not happen to thermal fluctuations (Brownian motion), which would exist even in zero particle concentration. The times presented in Figs 1 (a,b) are made non-dimensional by the reference scale a/U_s.

3.2. Time Developing of the Suspension

Microstructural changes, that is the variations in the relative arrangements of the particles, are among the most important and interesting features of the sedimentation process. The distribution of neighboring particles around a reference particle can be characterized by the structure factor of the suspension. Fig. 2 shows results of the statistics calculations of $F_{\perp}(\mathbf{k}, t)$ for $\phi = 3\%$ and $a/\ell = 0.05$. It is seen that the initial horizontal density fluctuations go way in time, indicating that the suspension tends to be randomized by the effects of the hydrodynamic interactions between the particles. The errobars increasing in the plot also indicates this randomization of the suspension.

Figura 1: Time development of the suspension showing the microstructure changes in the initial distribution of particles. Side view.

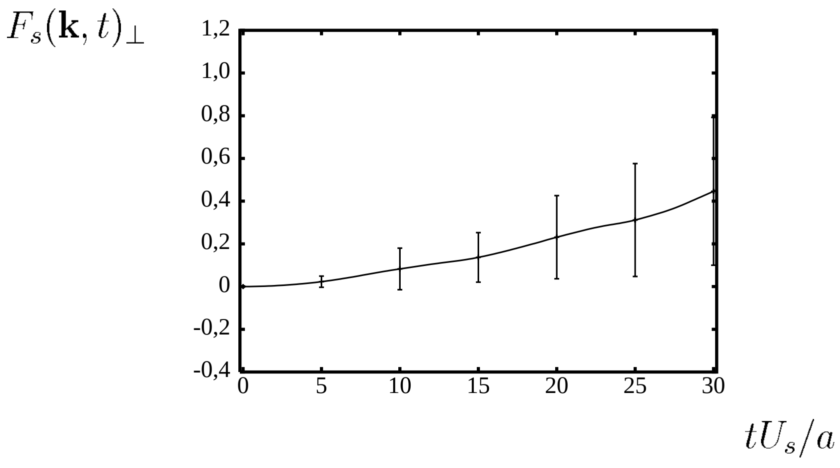

Figura 2: Time development of the dimensionless horizontal fluctuations in the density number of the particles as a function of dimensionless time.

The long-time behavior of the fluctuations are described by the velocity fluctuation autocorrelation functions and the hydrodynamic self-diffusivities both parallel and perpendicular to the gravity direction. The Figs. 3 and 4 shows the time development of the normalized velocity fluctuation auto-correlation functions for $\phi = 3\%$ and $a/\ell = 0.05$. The simulations reveal that these functions decay in time as a single exponential toward

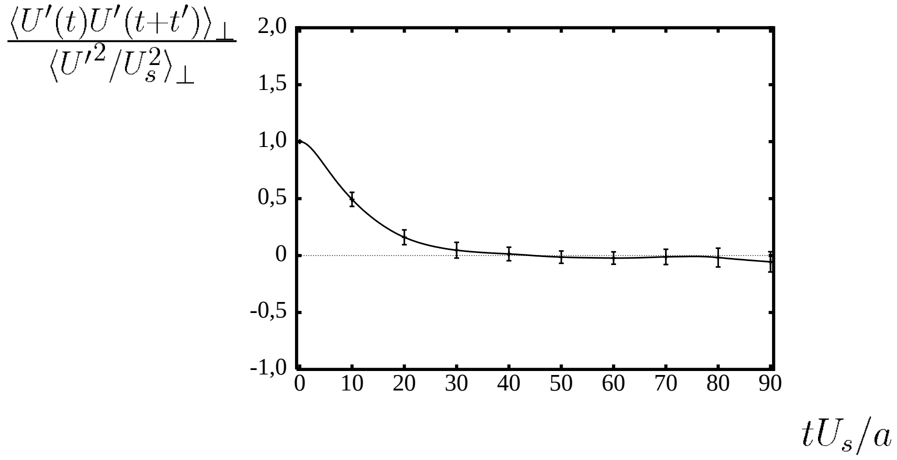

Figura 3: Normalized velocity fluctuation auto-correlation function perpendicular to the direction of gravity as a function of the dimensionless time. $\phi = 3\%$ and $a/\ell = 0.050$.

zero, indicating that particle velocity becomes totally uncorrelated after long time. The time to fall through $\ell \approx 60\ a$. The figures show also the error bars of these results. The results show that the aspect ratio $h/\ell = 3$ is sufficient to reach asymptotic and provide adequate data to determine the hydrodynamic self- diffusivities. This indicates that the particles has enough time to sample the horizontal cross section significantly before settling out so that the diffusivities are long time behavior ones. Note that this occurs in a time scale smaller than the one that many particles have already reach on the impenetrable bottom. The results also suggested a correlation time of the vertical fluctuations as being approximately the time to fall through 20 particles radius which is approximately twice bigger than the correlation of the horizontal fluctuations.

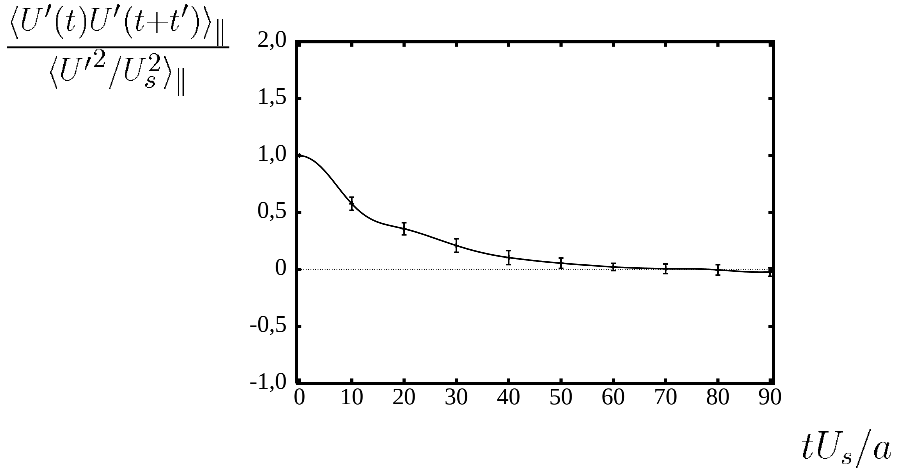

Figura 4: Normalized velocity fluctuation auto-correlation function parallel to the direction of gravity as a function of the dimensionless time. $\phi = 3\%$ and $a/\ell = 0.050$.

The result corresponding to the hydrodynamic diffusivities both parallel and perpendicular to the gravity direction as function of time is displayed in Fig. 5. The numerical simulation here determined values of $D_{\|} \approx 3aU_s$ which are slightly smaller than those reported by experiments and closely to the numerical result, $D_{\|} \approx 2aU_s$, of our previous simulations with fully random suspension considering the same parameter: $h/\ell = 3$, $a/\ell = 0.05$ and $\phi = 3\%$ (Cunha & Hinch 1996b). The experiments by Ham &

Homsy (1988) found this coefficient increasing from about $2aU_s$ at $\phi = 2.5\%$ to $6aU_s$ at 5%, and the more recent experiments by Nicolai et al. (1995) reported such self-diffusivity as being approximately $5aU_s$ at 5%. One possible explanation for this difference is that the comparison with experiments should be made with the understanding that the values of our diffusivities depend on system size (see Fig. 6). The laboratory experiments are also at dilute limit ($\phi = 5\%$), but they have a box about 100 times the particle diameter, which is obviously impossible to copy here due to the limited size of our numerical system for larger number of particles. It should be mentioned that the vertical diffusivity obtained in the present simulation is much smaller than that one predicted by hydrodynamic screening theory of Koch and Shaqfeh (1991) who found $D_{\parallel} = 0.52\phi^{-1}$ (≈ 17 at $\phi = 3\%$).

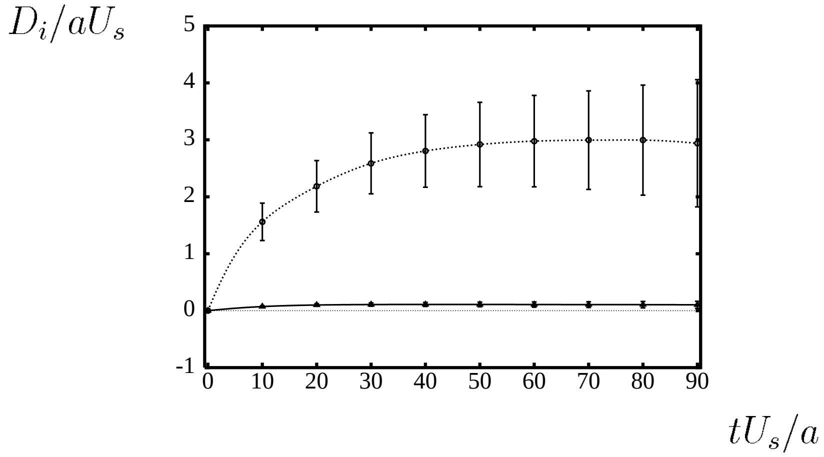

Figura 5: Dimensionless hydrodynamic self-diffusivities as a function of time. $\phi = 3\%$ and $a/\ell = 0.050$. $\triangle$: horizontal direction; $\circ$: vertical direction.

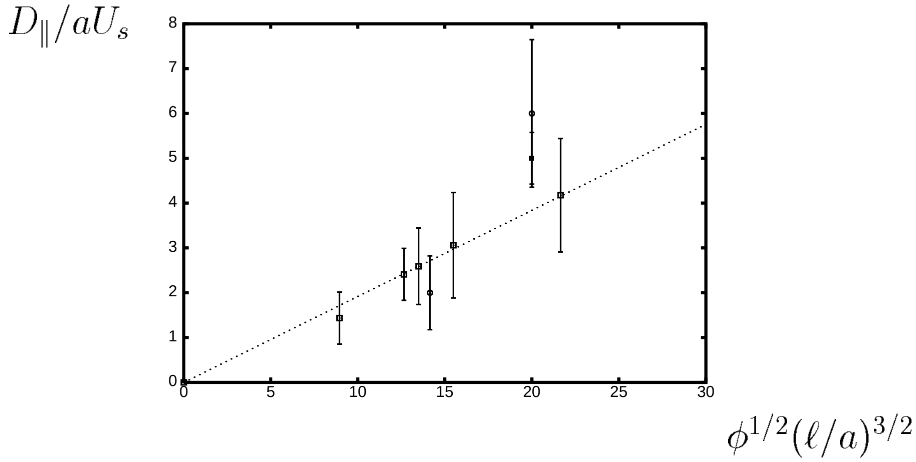

Figura 6: Vertical dimensionless hydrodynamic self-diffusivity as a function of the parameter $\phi^{1/2}(\ell/a)^{3/2}$. The dashed line is the linear fit $D_{\parallel} = 0.19\ aU_s\phi^{1/2}(\ell/a)^{3/2}$.

Figure 6 shows the plot of the self-hydrodynamic diffusivity parallel to the direction of the gravity as a function of the scaling $\phi^{1/2}(\ell/a)^{3/2}$ predicted in §§2.1. Accordingly the numerical results for the diffusivity increases linearly with this parameter as a consequence of the suspension randomization that occurs after times order of the correlation time. The observed values of the numerical diffusivities agree with experimental measurement of Ham & Homsy (1988) and Nicolai et al. (1994), but differently the experiments have diffusivities independent of the system size.

The dispersion process is observed strongly anisotropic. The major feature to note is the constant anisotropy in self diffusivities, about 25, that appear to be independent of the system size. This value of diffusivity anisotropy is about is about twice larger than the result, $D_{\|}/D_{\perp} \approx 10$, found by previous simulations with suspension on the same conditions but fully random (Cunha & Hinch 1996b). This difference should be attributed to the lower velocity fluctuations in the horizontal direction that occured in the present simulations as a consequence of the more regular initial arrangements of the particle. Moreover, it is important to notice that our results still reproduce more realistic amount of anisotropy than simulations results of fully periodic cubic cells by Ladd (1993) and Koch (1994). They predicted ratio of diffusivities $O(100)$, differently the experimental observations with anisotropy $\approx 5aU_s$ (Nicolai et al. 1995). This suggests that an impenetrable box do play an important role in simulating a sedimentation process. Physically, large fluctuations in the vertical velocity that increases with the size of the box are due to large horizontal density fluctuations, just $\sqrt{N}$ statistical fluctuations. When a periodic bottom is considered the heavy part of the suspension falls indefinitely preserving at each instant the density excess. On the other hand if it is imposed an impenetrable boundary, then there must be a convection current down on the heavy side, along the bottom, up the light side and cross the top.

4. CONCLUDING REMARKS

The numerical results reveal that after times closely to velocity fluctuation correlation time the more regular initial distribution considered now was strongly randomized as result of the hydrodynamic interactions between the particles. The magnitude of the diffusivity parallel to gravity agreed well with those predicted experimentally for dilute suspension, but it depends on the size of the numerical system, increasing like $O(\phi^{1/2}(\ell/a)^{3/2})$. The simulations showed degree of anisotropy in hydrodynamic self-diffusivity independent of the system size.

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