# Hydrodynamic Screening in Sedimenting Suspensions of non-Brownian Spheres

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It has been suggested [D. L. Koch and E. S. G. Shaqfeh, J. Fluid Mech. **224**, 275 (1991)] that the long-range hydrodynamic interactions in sedimenting suspensions of non-Brownian spheres are screened by changes in the pair correlation function. However, a large-scale numerical simulation, using more than 32 000 spheres, with full many-body hydrodynamic interactions, has found no evidence of the predicted changes in suspension microstructure. The absence of hydrodynamic screening in the simulations is shown to lead to divergent velocity fluctuations, in disagreement with recent experimental results [H. Nicolai and E. Guazzelli, Phys. Fluids **7**, 3 (1995)].

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In a sedimenting suspension, the root-mean-square fluctuations in particle velocity are of the same magnitude as the mean sedimentation velocity; for particles more than about 10 $\mu$m in diameter this hydrodynamic diffusion completely dominates the thermal Brownian motion. An apparently straightforward calculation has shown that for a suspension of randomly distributed particles the long-range hydrodynamic interactions cause the fluctuations in particle velocity to diverge linearly with the width of the container [1]. Although this result may seem surprising, there are experimental observations of just such a container-size dependence in very dilute sedimenting suspensions [2], where the solid volume fraction $\phi$ is less than 1%. On the other hand, more recent experiments, at a somewhat larger solid volume fraction ($\phi = 0.05$), found no systematic variation in the particle velocity fluctuations with container size [3]; the measured velocity fluctuations were also consistent with experiments using fluidized beds [4]. These theoretical and experimental findings raise the question of whether velocity fluctuations in sedimenting suspensions are controlled by the container size, or by the establishment of a nonrandom microstructure [5].

Koch and Shaqfeh have suggested that changes in pair correlations, induced by three-body hydrodynamic interactions, might lead to screening [5]. If long-range hydrodynamic interactions are screened, then the velocity fluctuations should be finite and independent of container size for sufficiently large vessels. However, the critical test of the theory is its prediction of a mass deficit, in other words, that the neighborhood around a test sphere contains precisely 1 particle less than if the same volume were filled uniformly with particles at the bulk suspension concentration. This prediction has not been tested experimentally because the particles used in the laboratory are too large to allow for a direct measurement of the structure factor by light scattering. Thus numerical simulations are the only technique available at present to study the microstructure (pair correlations) in sedimenting suspensions and to test for a mass deficit. The motivation for this work was to discover if hydrodynamic screening occurs in uniform sedimenting suspensions, in the absence of inhomogeneities introduced by cell boundaries. Thus large-scale numerical simulations, with more than 32 000 particles, have been carried out, using periodic boundary conditions to ensure that the suspension is uniform. The full many-body hydrodynamic interactions are accounted for, using the fluid equations for low Reynolds number (Stokes) flow. The numerical methods used in this work have allowed simulations of more than 100 times as many particles as in previous work [6]; for the first time we are able to simulate systems of macroscopic size and address fundamental questions about the effects of flow on the long-range microstructure of particle suspensions.

*Numerical method.*—The numerical method is based on a lattice-Boltzmann model of the fluid phase and a molecular dynamics simulation of the particle motion. The accuracy of the method has been established by extensive comparisons with theory, simulation, and experiment [7]. Moreover, for internal consistency we have run most of the calculations with two different mesh sizes, corresponding to an effective hydrodynamic radius ($a$) of the spheres of 1.39 and 2.443 lattice spacings (see Ref. [7] for a discussion of how the accuracy of the simulations depends on the effective hydrodynamic radius). Lastly, we have made several quantitative comparisons with earlier simulations of sedimentation [6], to verify that the new method does indeed reproduce known results for small systems ($N = 32$). In this work more than 32 000 spheres have been simulated; the cell width is about $70a$, comparable to laboratory experiments, and larger than the predicted hydrodynamic screening length in the Koch-Shaqfeh theory which suggests $\lambda \approx a/\phi$, or $\lambda \approx 10a$ in this case.

Since these simulations solve time-dependent fluid equations, the results depend on the mean particle velocity. The simulations can be characterized by a Reynolds number $R_W = UW/\nu$, based on the width of the cell $W$ and the mean flow velocity $U$ ($\nu$ is the kinematic viscosity of the fluid). $R_W$ is a measure of the range of distance scales over which the hydrodynamic interactions spread

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before a solid particle sediments a significant fraction of that distance. It is based on the cell width to allow the range of the hydrodynamic interactions to grow with the dimensions of the cell. The leading order correction to Stokes flow around an isolated sphere is of order $R_W/4$ at the boundary of the periodic unit cell. It was determined empirically that the results for small systems ($N = 128$) were independent of particle velocity for $R_W < 1$. In the simulations reported here $R_W$ was kept essentially constant at $R_W \approx 0.4$; as the systems got larger the effective gravitational force was reduced in proportion. By comparison, in the laboratory experiments [3] $R_W \approx 0.03$, roughly a factor of 10 less.

To reduce the expected screening length, while still keeping the suspension reasonably dilute, the simulations were run at a volume fraction $\phi = 0.1$. Since our earlier studies of sedimentation, it has been found that the velocity fluctuations and hydrodynamic diffusion coefficients depend on box *shape* as well as box size [8]. Diffusion in a cubic box is considerably more anisotropic than if the box is elongated; we have used a 4:1 height to width ratio throughout, which is very close to the asymptotic limit of a very elongated box [8]. A wide range of system sizes was used, from $N = 128$ to $N = 32\,768$, to compare the scaling of the velocity fluctuations with the predictions for a random long-range microstructure and with the predictions for screened hydrodynamic interactions. Each simulation was run for about 500 Stokes times, where a Stokes time $(a/U_0)$ is the time it takes an isolated sphere (velocity $U_0$) to fall one particle radius. Data from the first 150–200 Stokes times, during which time the suspension microstructure adjusts to the steady state, were discarded; results were obtained by averaging over the remaining 300–350 Stokes times. For the smaller systems we ensemble averaged over a number of different initial conditions as well. The largest calculations involved about 4.5 million fluid nodes for nearly 1 million time steps. Both the memory requirements ($\approx 10^9$ bytes) and the computation time ($\approx 10^{15}$ floating point operations) necessitated parallel computing. A problem of this size takes about 1000 h on 32 nodes of the Meiko CS2, or about 200 h on 32 nodes of the IBM SP2.

*Hydrodynamic screening.*—In slow sedimentation both particle inertia and fluid inertia can be ignored; in a typical experiment the particle Reynolds number is of the order of $10^{-4}$ [3,4]. Thus the velocity of a particle, at any instant, is completely determined by the configuration of its neighbors. In addition to the direct hydrodynamic interactions between the spheres, there is a pressure gradient generated by the base of the container, which balances exactly the total gravitational force on the particles; i.e.,

$$\int_V \nabla p(\mathbf{r})d\mathbf{r} = N\mathbf{F}, \tag{1}$$

where $\mathbf{F}$ is the gravitational force on each of the (identical) spheres and $N$ is the number of spheres in the sample volume $V$. In the homogeneous suspensions modeled by the simulations, the pressure gradient is replaced by an equivalent uniform force density, $\mathbf{f} = N\mathbf{F}/V$, applied throughout the sample. On *average*, the backflow of fluid induced by the pressure gradient (or force density) cancels the contributions of the hydrodynamic interactions at large distances, so that the mean sedimentation velocity is finite and independent of container size [9] and shape [10]. However, contributions of distant particles to the velocity *fluctuations* of a test sphere fall off as $R^{-2}$, and, when summed over all particles in the system (apart from the test sphere), give a contribution to the variance that is proportional to the linear dimensions of the container [1]. For a sample with periodic boundary conditions, the *diverging* contribution to the velocity fluctuations can be written as a sum over wave vectors $\mathbf{k}$ that are commensurate with the periodic box [6]

$$\langle \mathbf{UU} \rangle = \frac{NS(k \to 0)}{V^2} \sum_{\mathbf{k} \neq 0} \mathbf{F} \cdot \mathbf{T}(\mathbf{k})\mathbf{T}(\mathbf{k}) \cdot \mathbf{F}\,. \tag{2}$$

In this equation $S(k \to 0)$ is the long-wavelength limit of the structure factor, and $\mathbf{T}(\mathbf{k}) = (\mathbf{1} - \mathbf{kk}/k^2)/\eta k^2$ is the periodic Green's function for hydrodynamic interactions in a fluid of viscosity $\eta$. The sum in Eq. (2) is proportional to $V^{4/3}$; thus any divergence in the velocity variance is directly connected to the behavior of long-wavelength density fluctuations, described by $S(k \to 0)$. If the particle positions are uncorrelated at separations beyond a few particle radii, then $S(k \to 0)$ is finite and the velocity fluctuations diverge. If, on the other hand, long-wavelength density fluctuations are for some reason suppressed, and $S(k \to 0) = 0$, then the velocity fluctuations will be finite [5], even for very large samples. An analogous situation occurs in electrolyte solutions where long-wavelength fluctuations in charge density are suppressed by Debye-Hückel screening. The charges rearrange so that an ion and its environment are overall electrically neutral on length scales greater than the Debye-Hückel screening length. Thus in charged systems there are long-range pair correlations (on the order of the screening length) which are such that the charge-charge structure factor vanishes in the long-wavelength limit. Koch and Shaqfeh [5] have proposed that a similar effect could suppress the velocity fluctuations in sedimenting suspensions. Here the pressure gradient in the background fluid plays the same role as the counterions in an electrolyte solution. The equivalent Koch-Shaqfeh screening of the hydrodynamic interactions requires that a sedimenting sphere and its neighbors are neutrally buoyant with respect to the bulk suspension. In terms of the pair correlation function $g(\mathbf{r})$, hydrodynamic screening requires that the mass deficit,

$$n(\lambda) = \frac{N}{V} \int_{V_\lambda} [g(\mathbf{r}) - 1]d\mathbf{r}\,, \tag{3}$$

is precisely equal to $-1$. The integral in Eq. (3) is over a spherical volume of radius $\lambda$, where $\lambda$ is much greater than the particle radius $a$, but much less than the size of the container. Thus for hydrodynamic screening to occur the number of neighbors in a *submacroscopic* volume $V_\lambda$ around a test sphere must be exactly 1 particle less than if that same volume were filled uniformly with particles [5]. Using the relation between the structure factor and the pair correlation function, $S(\mathbf{k}) = 1 + (N/V) \int \cos(\mathbf{k} \cdot \mathbf{r})[g(\mathbf{r}) - 1]d\mathbf{r}$, it is clear that Eq. (3) corresponds to the condition $S(k \to 0) = 0$.

In order to test the validity of the Koch-Shaqfeh theory, we have determined the value of the mass deficit $n(r)$ at distance scales larger than the expected screening length, $\lambda \approx 10a$, at the simulated volume fraction ($\phi = 0.1$); the results are shown in Fig. 1. There is some mass deficit, even for the most-probable (or equilibrium) distribution of nonoverlapping spheres, because of excluded volume effects. However, the large-$r$ asymptotic value of the deficit for the most-probable distribution is always greater than $-1$; at $\phi = 0.1$, $n(r \to \infty) = -0.54$. By contrast, if there is hydrodynamic screening, then the Koch-Shaqfeh theory predicts that $n(r \to \infty)$ is exactly $-1$. If the deficit is less than 1 particle or, in other words, if $n(r \to \infty) > -1$, there will not be a complete screening of the hydrodynamic interactions, and the velocity fluctuations will diverge. Results for $n(r)$ from a numerical simulation of 32 768 sedimenting spheres are shown in Fig. 1. The pair correlation function is qualitatively different from what would be expected if there were hydrodynamic screening (shown by the dashed curve). It can be seen that $n(r)$ is essentially constant in the range $15a < r < 35a$; its large-$r$ asymptotic value, $n(r \to \infty) = -0.6$, is close to that for the most-probable distribution. Our calculations would detect the onset of screening, if it existed, for screening lengths up to about $50a$, which is an order of magnitude larger than the theoretical prediction. More accurate simulations, using a smaller number ($N = 7424$) of larger spheres ($a = 2.443$ lattice spacings), have also been carried out; in this case we can calculate pair correlations out to about $r = 20a$ (see Fig. 1). These results also show no sign of hydrodynamic screening; the mass deficit is the same as for the smaller spheres (within statistical errors).

Although our calculations indicate that there are no long-range pair correlations in homogeneous sedimenting suspensions, significant changes in the short-range microstructure are induced by the sedimentation process. A comparison of pair correlations for most-probable and steady-state sedimentation microstructures is also shown in Fig. 1. The enhancement in pair correlations near contact can be explained by consideration of three-body hydrodynamic interactions [5]. At somewhat larger separations, $5a < r < 15a$, the pair probability decreases more rapidly than for the most-probable distribution (see Fig. 1), perhaps by the mechanism suggested by Koch and Shaqfeh [5]. However, it seems that the correlation times for the three-body hydrodynamic interactions are not long enough to lead to sufficient deficit for hydrodynamic screening.

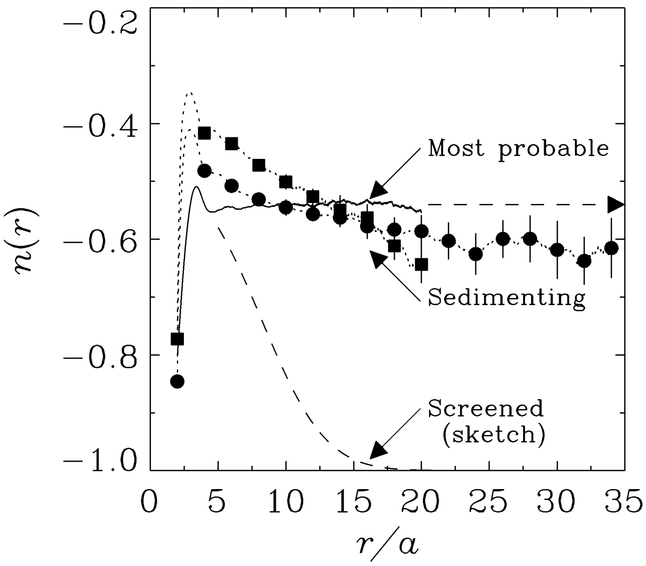


FIG. 1. Long-range pair correlations in a steadily sedimenting suspension at a volume fraction $\phi = 0.1$. The number of particles in a spherical volume surrounding a test sphere, minus the average number of particles in the same volume, $n(r)$, is plotted for a system with a periodic repeat width of $70a$ (circles). The 4:1 unit cell contained 32 768 spheres, with an effective hydrodynamic radius $a = 1.39$ lattice spacings. Data from a run of 7424 spheres, with an effective hydrodynamic radius $a = 2.443$ lattice spacings, are also shown (squares). The statistical errors for both sets of data are similar and are indicated by the vertical bars. The pair correlation function for the most-probable distribution of nonoverlapping spheres at $\phi = 0.1$ is also shown for comparison (solid line); its large-$r$ asymptotic value, $n(r \to \infty) = S(k \to 0) - 1 = -0.54$, can be determined from the Carnahan-Starling equation of state for hard spheres, and is shown by the horizontal arrow. A rough sketch of a screened pair correlation function, with a screening length of approximately $10a$, is shown by the dashed curve.

*Velocity fluctuations.*—If there is no screening of the hydrodynamic interactions, as the simulations strongly suggest, then the velocity fluctuations should diverge linearly with the width of the container, as described by Eq. (2). It can be seen in Fig. 2 that the simulated velocity fluctuations do diverge approximately linearly with the periodic repeat width $W$; the actual numerical values are in reasonable quantitative agreement with the theory for the most-probable distribution of spheres [Eq. (2)]. The relatively small differences between simulation and theory are most likely due to finite size effects.

Although the results for the largest system ($N = 32\,768$) are in quite good agreement with experiment [11], there are two significant qualitative differences between simulation and experiment. First, the vertical to horizontal anisotropy in the simulated velocity fluctuations is close to 10:1, which is characteristic of a random

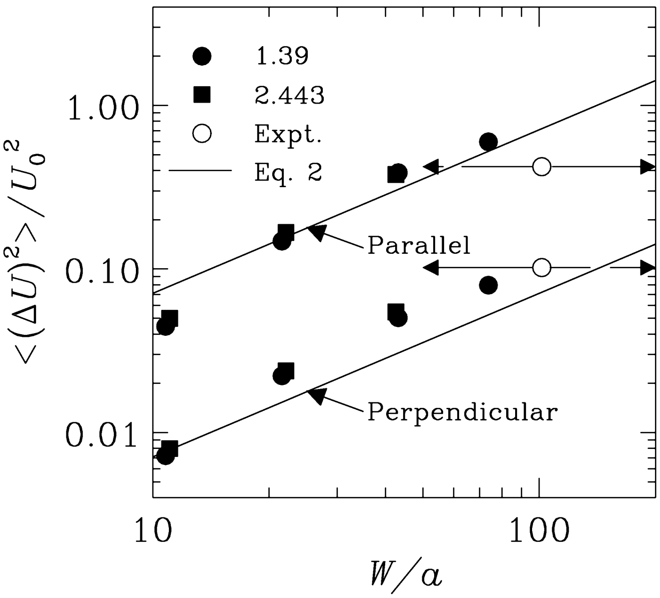


FIG. 2. Particle velocity fluctuations as a function of box width ($W$) in a steadily sedimenting suspension at a volume fraction $\phi = 0.1$. The data are normalized by the sedimentation velocity, $U_0$, of an isolated sphere under the same applied force. Simulation results for two different particle sizes are shown; the solid line is the theoretical scaling (linear in $W/a$) for the most-probable distribution of spheres [Eq. (2)]. The statistical errors in the simulation data are in the range 2%–4%, smaller than the plotting symbols in the figure. Experimental results at a volume fraction $\phi = 0.1$ (open symbols) are shown for comparison [11]. The experimental data are located along the $W$ axis by the smallest dimension of the experimental vessel. The range of container widths for which no systematic change in variance was observed experimentally (however, at $\phi = 0.05$) [3] is from $W/a = 50-200$, as indicated by the horizontal arrows.

microstructure at large particle separations. The simulated anisotropy is independent of system size (see Fig. 2), but is significantly larger than the experimental measurement of about a 4:1 anisotropy. The second and most important difference illustrated in Fig. 2 is that the velocity fluctuations for a homogeneous sedimenting suspension diverge over ranges of container size, where they are observed experimentally to be constant [3]. The data in Fig. 2 indicate that the velocity fluctuations measured in the simulations are independent of the particle radius (or mesh size) used in the lattice-Boltzmann simulations, and are thus not likely to be further modified by more accurate calculations.

The pair correlation function for a sedimenting suspension has been computed to distances well beyond the expected screening length, $\lambda \approx 10a$. We find no evidence of a mass deficit sufficient for hydrodynamic screening, nor of long-range pair correlations that might eventually lead to such a deficit. Moreover, the velocity fluctuations diverge with the width of the periodic unit cell, as would be expected for a random long-range microstructure. Our results suggest that other mechanisms must be uncovered to account for the experimental observations in Refs. [3] and [4]. The inhomogeneities introduced by the experimental apparatus may affect the velocity fluctuations in at least two different ways. First, long-range perturbations to the microstructure may be induced by the container walls, which may in turn lead to hydrodynamic screening; second, there may be large-scale fluctuations in volumetric flow, which, in the present calculations, have been suppressed by the periodic boundary conditions. New simulations, explicitly including container walls, are planned to address these questions.

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