Microstructure in a settling suspension of hard spheres

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We report direct observations of the structure factor in a settling suspension, using numerical simulations based on a lattice-Boltzmann model of the fluid. We find that the horizontal density fluctuations in bounded suspensions are strongly suppressed by the settling process, vanishing as k^2 at long wavelengths. Our measurements of the structure factor confirm the qualitative predictions of one of several competing theories, although this theory does not yet explain why container walls are so important. Our results contradict the idea that a settling suspension is inevitably stratified by hydrodynamic dispersion at the suspension-supernatent interface.

The dynamics of suspensions at low Reynolds numbers are controlled by the distribution of particle positions, which is sufficient to determine the particle velocities at any instant of time. In a settling suspension, subtle shifts in the pair correlation function can have a dramatic effect on the macroscopic behavior. It is well known that the velocity fluctuations in a settling suspension are dependent on container size if the particles are randomly distributed [1]. However, it has been suggested [2] that there are rearrangements of the suspension microstructure during settling which have the effect of suppressing the long-wavelength density fluctuations. The idea of some kind of microstructural rearrangement is now widely accepted, since there is no other viable explanation for experimental results showing that velocity fluctuations are independent of system size [3,4]. However, the mechanism for the supposed microstructural rearrangements is highly controversial. Theoretical explanations include three-body hydrodynamic interactions [2], convection of density fluctuations [5–7], and the effects of vertical walls [8]. Recent numerical simulations have shown that none of these ideas are completely correct, and that the container must be bounded at the top and bottom in order for the divergence of the velocity fluctuations to be suppressed [9,10]. However, even here there is disagreement; Ladd [9] has suggested that the density fluctuations drain to the interfaces, along the lines originally proposed by Hinch [5] and examined in more detail by Levine *et al.* [7], while Mucha *et al.* [10,11] have proposed that there is a weak vertical variation in the particle concentration (stratification) caused by hydrodynamic dispersion at the suspension-supernatent interface. Luke has shown that such a concentration gradient can prevent the divergence of the velocity fluctuations, even when the pair distribution is random [12].

In this paper we report direct observations of the structure factor in a settling suspension, using numerical simulations based on a lattice-Boltzmann fluid [13,14]. It has not been possible to measure the structure factor of a settling suspension of non-Brownian particles experimentally, since the particles are too large for light scattering measurements. Fluc-

tuations in particle concentration have been measured within a cylindrical or rectangular window [15], but this only gives an angle average of the pair distribution. In numerical simulations it is possible to calculate $S(\mathbf{k})$ as a function of both wavelength and direction. This is important since some theories predict that the structure factor becomes highly anisotropic [7], with the horizontal fluctuations vanishing at long wavelengths, while the vertical fluctuations remain finite. Our simulations show that horizontal density fluctuations are indeed strongly suppressed when there are container walls at the top and bottom, but not when the suspension is periodic in the vertical direction. This is consistent with our earlier observation [9] that the velocity fluctuations are size independent in bounded containers, but size dependent in periodic systems. Our measurements of the structure factor agree qualitatively with the predictions of Levine *et al.* [7], although their theory does not explain why the container walls are so important. Our results contradict the idea that a settling suspension is inevitably stratified by hydrodynamic dispersion at the suspension-supernatent interface; we find that the particle concentration is uniform in the bulk region to within statistical errors, and any possible gradient is too small to have a noticeable effect on the velocity fluctuations. This does not discount the possibility that stratification occurs at very low particle concentrations or in polydisperse suspensions.

The structure factors were calculated directly from particle positions obtained in a simulation of 72 000 monodisperse non-Brownian spheres, settling in a tall thin container of dimensions $50a \times 50a \times 1000a$, where a is the particle radius. The container was bounded at the top and bottom by rigid no-slip walls, while periodic conditions were used at the sides. It has been previously shown that the boundaries at the top and bottom of the cell have a profound effect on the velocity fluctuations in a settling suspension [9]; for comparison purposes we have also calculated the structure factor in a settling suspension with periodic boundary conditions on all faces. In both cases, the particle volume fraction was set at $\phi = 13\%$, so that the mean interparticle spacing $a\phi^{-1/3}$ is almost exactly $2a$. The height of the bounded cell allowed for lengthy simulations, in excess of 1500 Stokes times, where the Stokes time $t_s = a/U_0$ and U_0 is the settling veloc-
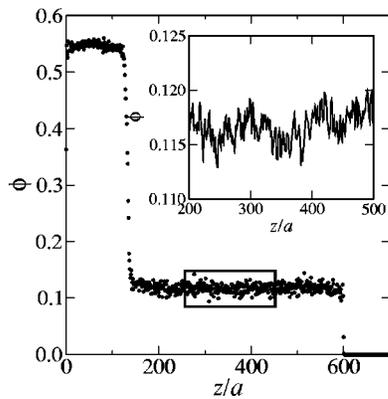
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FIG. 1. The particle volume fraction at steady state as a function of height. The dots indicate the volume fraction at a single instant of time, $t=1200t_s$, averaged across the horizontal plane and over a height of a. The data shows that the suspension-supernatent interface is sharp and the density is uniform over the region $200-500a$. The expanded view of this region shows the density profile averaged over the steady-state time window $1000<t/t_s<1400$. The density in the viewing window ($250-450a$) is uniform to within statistical uncertainties; any residual density gradient is negligible, with a characteristic length in excess of 10^4a, or more than ten times the height of the container.

ity of an isolated particle. This is sufficient to establish a steady state over a fairly substantial viewing window, at least $200a$ high, located about one-third of the way up the column (see Fig. 1). We have checked that the measured structure factor is insensitive to the exact location and size of the viewing window. The steady state structure factor was determined over a time interval $1000<t/t_s<1400$ from the onset of the settling process.

The numerical simulations were based on a lattice-Boltzmann model of the underlying fluid dynamics [13,14], and include recent improvements to the calculation of the lubrication forces and to the overall stability of the algorithm [16]. The simulations were carried out at a Reynolds number Re=0.06, based on the mean settling velocity. Results from simulations at a lower Reynolds number, Re=0.03, were indistinguishable from those at Re=0.06, which is consistent with previous observations [9] that inertial effects are negligible in this system when Re<0.1. The other key factor controlling the accuracy of the numerical simulation is the number of grid points occupied by each solid particle, which is a measure of the resolution of the particle surface on the fluid grid. In these simulations the particle radius was set to two grid spacings, since test calculations have shown that the mean settling velocity is then insensitive to further increases in particle size.

Figure 1 shows that the density profile is uniform in the bulk, with a sharp interface between the suspension and supernatant fluid. A sharp interface has also been observed experimentally (see Ref. [17] for example), but recently it has been suggested that hydrodynamic dispersion at the suspension-supernatent interface could lead to a weak stratification of the particles and a nonuniform density in the bulk [10,11]. Stratification has been suggested as a mechanism for suppressing density fluctuations [12], but the density profile
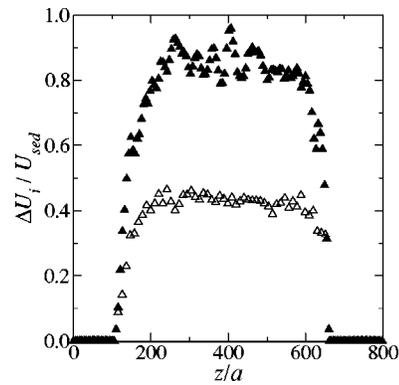

FIG. 2. Particle velocity fluctuations as a function of height. The vertical (solid symbols) and horizontal (open symbols) velocity fluctuations have been averaged over the horizontal plane and over a time window of $1000<t/t_s<1400$.

in the inset to Fig. 1 contradicts the recent suggestion [11] that stratification can be a general explanation for the saturation of velocity fluctuations. As can be seen, any mean variation in density is well below the statistical noise, even after averaging over several hundred Stokes times. Our data suggest that any residual density gradient must have a characteristic length of at least 10^4a, or ten times the container height. Moreover, the stratification theory predicts that the velocity fluctuations vary with height [11], since the density gradient is largest near the suspension-supernatent interface. Again this is contradicted by our measured velocity fluctuations, which are constant throughout the viewing window, as shown in Fig. 2. Hydrodynamic dispersion does cause a spreading of the interface, but this is compensated by hindered settling, which convects the less dense regions at a higher velocity than the high density regions and thereby sets up a convective flux in opposition to the diffusive flux. Balancing the convective and diffusive concentration fluxes with respect to a frame moving with the mean settling velocity, we estimate an interface thickness, $D_\parallel/U' \approx 3a$, at this volume fraction. Here the vertical dispersion coefficient, $D_\parallel=4.1U_{sed}a$, was taken from our simulations, and $U'=-\phi dU/d\phi \approx 5\phi U_0$ is the velocity of a density perturbation with respect to the mean settling speed. Our simulations do not rule out the possible significance of stratification in very dilute suspensions ($\phi<1\%$), such as are commonly used in particle-image-velocimetry measurements [18,10], but they do exclude it as a general mechanism for hydrodynamic screening.

Figure 3 shows the static structure factor, $S(\mathbf{k})$, in bounded and periodic suspensions steadily settling under gravity. A settling suspension with periodic boundaries in all three directions has a random microstructure at long wavelengths [$S(k\rightarrow 0)\neq 0$], with a structure factor that is finite at all wavelengths and more or less isotropic; in other words it is qualitatively similar to an equilibrium suspension. These observations are consistent with previous simulations [19,20], although in that work the pair distribution function was analyzed rather than the structure factor. On the other hand, if rigid boundaries are imposed at the top and bottom of the vessel, the structure factor shows that the long wave-
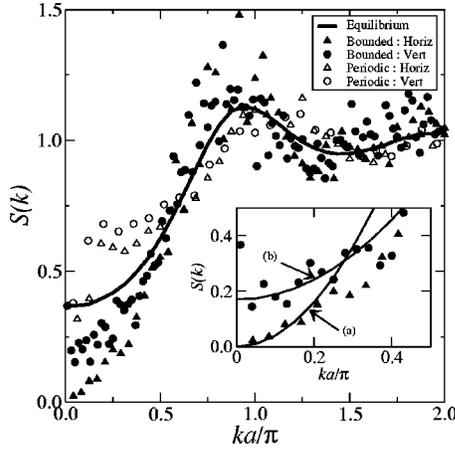
FIG. 3. The structure factor $S(\mathbf{k})$ for bounded (solid symbols) and periodic suspensions (open symbols) at steady state; i.e., averaged over the time window $1000 < t/t_s < 1400$. The solid line is the averaged equilibrium structure factor obtained by a Monte Carlo simulation with 10^6 moves per particle. The inset clarifies the long wavelength behavior of the bounded system; the solid lines indicate quadratic fits to the data: (a) horizontal component (triangles) $S(k_\perp) \approx 0.4(ka)^2$ and (b) vertical component (circles) $S(k_\parallel \to 0) \approx 0.17$.

FIG. 4. Time evolution of density fluctuations in the horizontal plane. The structure factor $S(k_\perp)$ is shown over different time intervals: (a) (circles) $0 < t/t_s < 200$, (b) (squares) $200 < t/t_s < 400$, (c) (triangles) $400 < t/t_s < 600$. The solid line indicates the equilibrium structure factor. The behavior at long wavelength is shown by the inset. The data is an ensemble average over four different starting conditions.

length pair correlations are strongly damped, especially in the horizontal direction where the density fluctuations appear to vanish at sufficiently long wavelengths.

Levine *et al.* [7] have proposed that there are two qualitatively distinct nonequilibrium phases for settling suspensions, an unscreened phase characterized by a random microstructure and a screened phase where the horizontal density fluctuations are damped out at long wavelengths. They derived an expression for the nonequilibrium structure-factor,

$$S(\mathbf{k}) = \frac{N_\perp k_\perp^2 + N_\parallel k_\parallel^2}{D_\perp k_\perp^2 + D_\parallel k_\parallel^2 + \gamma k_\perp^2/k^2}, \quad (1)$$

which is consistent in functional form with the structure factor obtained in our numerical simulations (Fig. 3). The renormalized parameters N_i and D_i together with the damping coefficient γ were calculated from coupled field equations describing the evolution of the particle concentration and fluid velocity. According to the theory, the phase boundary is determined by the anisotropy in the renormalized noise $N_\perp/N_\parallel$ and diffusivity $D_\perp/D_\parallel$.

The structure factor data can be used to extract ratios of the parameters that appear in Eq. (1); namely $N_\perp/\gamma = 0.4a^2$, and $N_\parallel/D_\parallel = 0.17$. We obtained $N_\perp/\gamma$ and $N_\parallel/D_\parallel$ from the low-k behavior of the horizontal and vertical density fluctuations, but since our data is rather noisy, it is impossible to extract a meaningful value of $D_\perp/\gamma$, which appears as a quartic correction to the asymptotic k^2 dependence of the structure factor. Nevertheless, for the sake of completeness we will use our best estimate, $D_\perp/\gamma = 0.5a^2$, to determine the ratio $N_\perp/N_\parallel \approx 0.7$. When combined with tracer-diffusion measurements of $D_\perp/D_\parallel = 0.14$, this suggests we are near the transition between screened and unscreened phases [7]. Unfortunately our data is not sufficiently precise to enable a
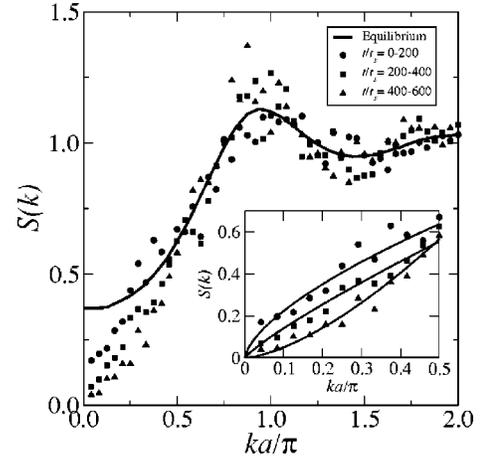
definitive conclusion to be drawn. Significantly larger systems sizes will be necessary for a quantitative comparison with the predictions of the theory, with greatly increased computational requirements.

Although the structure factors measured in the simulations are consistent with the predictions of Levine *et al.* [7], their theory postulates that all the important dynamics occurs in the bulk, independent of the macroscopic boundary conditions. Although this is a logical assumption, our numerical simulations show that it is incorrect. Simulations with periodic boundary conditions do not show the same damping of the horizontal density fluctuations, as would be expected if the model proposed in Ref. [7] were correct in all essentials. Instead, our simulations suggest that the container bottom and the suspension-supernatant interface act as sinks of fluctuation energy, as suggested earlier by Hinch [5]. Random density fluctuations convect to one of these two interfaces and are absorbed by the density gradient at the interface. The data shown in Fig. 4 support this conclusion, albeit not conclusively. Here we show the structure factor in the viewing window during its evolution from an equilibrium state to the steady state. Despite the limited time averaging (a total of $800t_s$ for each plot), the implication is that the long wavelength fluctuations decay fastest. If so, this is evidence for the immediate convection of large-scale density fluctuations [5], rather than the establishment of a density gradient by hydrodynamic diffusion [11]. We have previously proposed [9] that there is a transition between the mechanism proposed by Hinch [5], describing the decay of the fluctuation energy stored in the initial configuration, to the steady-state behavior described by Levine *et al.* [7]. However, it is not clear how to explain the absence of screening in periodic suspensions within the context of this theory. It would be interesting to study the convection-diffusion model proposed in Ref. [7] in the presence of container walls, although this would almost certainly require a numerical calculation.

It is illuminating to compare the convection-diffusion theory of Ref. [7] with point-particle simulations [10,11], with which it has much in common. The Oseen velocity field is divergence free, and therefore the steady-state solution of the Smoluchowski equation is always a random distribution [2]. In Refs. [10,11] the screening is generated by transient vertical variations in particle concentration, rather than by a steady-state change in pair correlations. The key difference is that the theory in Ref. [7] adds strongly anisotropic concentration fluctuations, which are an empirical representation of the supposed effects of the many-body hydrodynamic interactions. To obtain a hydrodynamically screened phase, the theory requires that small-scale concentration fluctuations are largest in the horizontal plane, $N_\perp^0 \gg N_\parallel^0$ [7]. By contrast, in a random suspension the density fluctuations are isotropic on all length scales. Our simulations show that there is a pronounced change in the anisotropy as the settling proceeds, so the character of the noise must change as the suspension evolves to steady state. It would be interesting to discover how this process is affected by container walls.

In this paper we have presented what is, to our knowledge, the first determination of the structure factor in a steadily settling suspension. Our results confirm certain predictions of Ref. [7], in terms of the behavior of both the horizontal and vertical density fluctuations. Nevertheless, the theory does not yet explain why the hydrodynamic interactions in spatially homogeneous (fully periodic) suspensions are not screened. The simulations show that hydrodynamic screening is possible within a homogenous "bulk" region of a bounded suspension. Thus they do not support the recent contention [11] that hydrodynamic dispersion leads to a macroscopic density gradient, and that this gradient is necessary for hydrodynamic screening. We suggest that it will be fruitful to explore the connection between the multibody hydrodynamic interactions in a settling suspension and anisotropic noise in a convection-diffusion model.

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