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Screening in sedimenting suspensions

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Caflisch & Luke (1985) showed that, owing to the long range of the hydrodynamic interactions, the variance of the sedimentation velocity in a random suspension with uniform probability for the positions of the particles is divergent in the sense that it grows without bound as the macroscopic linear dimension of the settling vessel is increased. It is shown here, however, that a Debye-like screening of a particle's velocity disturbance, leading to a finite variance, will occur if the pair probability reflects a net deficit of one particle in the vicinity of each particle. The three-particle interactions, which determine the structure of a dilute, monodisperse suspension of spheres, lead to a deficit of neighbouring particles. The magnitude and range of this deficit are shown to be sufficient to lead to a Debye-like screening of the velocity disturbance at a radial distance of order $a\phi^{-1}$, where a is the particle radius and ϕ their volume fraction. A self-consistent approximation to the screened conditional average velocity field and pair distribution is presented. The screening leads to a variance of the particle velocity and a particle tracer diffusion coefficient that are finite and of order U^{s2} and $U^s a\phi^{-1}$, respectively, where U^s is the Stokes settling velocity of the particles in unbounded fluid.

1. Introduction

The sedimentation of a monodisperse, dilute suspension of non-Brownian particles in the absence of inertial effects is one of the basic, simple flows of suspensions. Neglecting all interparticle hydrodynamic interactions in the dilute limit would lead one to conclude that each particle's velocity $\boldsymbol{U}$ is equal to the Stokes settling velocity $\boldsymbol{U}^s = 2\Delta\rho a^2\boldsymbol{g}/9\mu$, where $\Delta\rho$ is the difference between the density of the particles and the fluid, a is the particle radius, μ is the fluid viscosity, and $\boldsymbol{g}$ is the acceleration due to gravity.

However, in attempting to calculate the first effects of particle interactions on the mean or variance of the sedimentation velocity in a random suspension by directly summing the influence of individual particles, one encounters divergent integrals. Batchelor (1972) introduced a renormalization that overcame the divergence problems inherent in determining the first correction to the mean sedimentation velocity in a dilute, random, monodisperse suspension. He found the average sedimentation velocity to be $\langle \boldsymbol{U}\rangle = \boldsymbol{U}^s(1-6.55\phi)$, where ϕ is the particle volume fraction and $\langle\;\rangle$ indicates an ensemble average over all possible particle configurations weighted by the probability of their occurrence.

Particles in a sedimenting suspension undergo a randomly fluctuating motion induced by the hydrodynamic disturbance caused by the surrounding particles. This randomly fluctuating motion is important because it is expected to give rise to a dispersion or mixing of chemical tracers and of the solid particles themselves. The variance $\langle U'^2 \rangle$ of the sedimentation velocity is the simplest measure of the particle velocity fluctuations. Here, $U' \equiv U - \langle U \rangle$ is the deviation of the particle's velocity from its average value. Caflisch & Luke (1985) showed that Batchelor's (1972) renormalization does not resolve the divergence difficulties associated with calculating the variance of the sedimentation velocity. Indeed, they showed that the variance, $\langle U'^2 \rangle = O(U^{s2} \phi L/a)$, grows in proportion to the characteristic linear dimension L of the settling vessel.

The divergence in the variance of the sedimentation velocity is particularly disturbing in view of the relationship of the solid-particle tracer diffusivity to the velocity variance. The diffusivity is given by a time integral of the particle's velocity correlation function, so it cannot be expected to be independent of L/a (and thus finite as $L/a \rightarrow \infty$) if the variance of the particle's settling velocity is not also finite as $L/a \rightarrow \infty$. A similar divergence arises if one attempts to calculate the effective diffusivity of a tracer in the suspending fluid of a random sedimenting suspension. Indeed Koch & Brady (1985) noted that any attempt to calculate the effective tracer diffusivity resulting from a random array of point forces of fixed magnitude leads to a divergent integral.

The variance of the fluid velocity and the effective tracer diffusivity are both well defined and independent of bed size in a random array of fixed particles (Koch & Brady 1985). The important difference between a random fixed array and a random sedimenting suspension is that in the fixed bed the particle velocities are maintained (at zero) and the forces acting on them are influenced by hydrodynamic interactions, while in a sedimenting suspension each particle's force is fixed (as the force of gravity acting on the particle's mass) and its velocity depends on hydrodynamic interactions. In a fixed bed the fluid velocity disturbance caused by a given particle will affect the external force required to keep the other particles fixed. This leads to a body force in the conditionally averaged momentum equation which is proportional to the velocity. Thus, the conditionally averaged velocity disturbance associated with one fixed particle satisfies Brinkman's equation and is screened at a distance $a\phi^{-\frac{1}{2}}$. As a result the conditionally averaged velocity disturbance decays like x^{-3} as the radial distance x goes to infinity, and the integrals required to determine the dispersion properties mentioned above are convergent. Direct hydrodynamic interactions, however, do not give rise to such screening of the particle velocity disturbance in a random sedimenting suspension.

The aforementioned results pertain to a random suspension of uniform probability. However, unless Brownian motion dominates over hydrodynamic interactions, there is no reason to expect that all accessible configurations of the suspension are equally probable. Thus, while Batchelor's (1972) result $U^s(1-6.55\phi)$ for the mean sedimentation velocity is applicable to Brownian suspensions, the average sedimentation velocity will generally depend on the suspension structure. A clear indication of this influence of structure is the fact that the first correction to the sedimentation velocity in a dilute periodic bed is $O(\phi^{\frac{1}{3}})$ (Hasimoto 1959) as opposed to the $O(\phi)$ correction in a random suspension with uniform probability.

It will be seen that a non-uniform suspension structure can have an even more profound effect on the variance of the velocity. In §2 we show that a suspension with a certain type of structure (one whose pair probability satisfies equation (2.14)) has a finite velocity variance, i.e. the variance is independent of L/a as $L/a \rightarrow \infty$. Physically, the criterion (2.14) corresponds to requiring a net average deficit of one particle in the vicinity of any given particle. If this criterion is satisfied, the ensemble-averaged velocity disturbance with one particle's position held fixed is screened in a manner that is analogous to the Debye screening of the electrical potential associated with a fixed ion in an ionic solution. It should be noted that the analogy between sedimenting suspensions and ionic solutions is limited to the effect of the pair probability on the variance of the velocity in the former case and the variance of the electrical potential in the latter case. The mechanisms controlling the pair probability in these two cases are, of course, different. In an ionic solution there is a clear mechanism – electrical attraction and repulsion – leading to a structure in which the electrical potential is screened. In a sedimenting suspension, it is not immediately obvious whether the hydrodynamic interactions that control the pair probability will cause a net increase or deficit in the number of particles near any given particle.

In §3 we address the problem of determining the pair probability in a monodisperse suspension of spheres. Even in the dilute limit the pair probability is controlled by three-particle interactions, because there is no relative motion between two isolated identical spheres. In this sense, sedimenting suspensions of monodisperse spherical particles are more difficult to treat theoretically than suspensions of non-spherical or polydisperse particles.

Batchelor & Wen (1982) determined the effect of two-particle interactions on the pair probability in a polydisperse suspension of non-Brownian spheres. In a polydisperse suspension two isolated particles do move relative to one another, so two-particle encounters occur in this case. The structure in the pair probability induced by these two-particle interactions is very short in range, decaying like a^6x^{-6} as $x \rightarrow \infty$. Batchelor & Wen (1982) related the pair probability to an integral along a particle trajectory of the divergence of the relative velocity. The non-random structure is short range, because the relative velocity is solenoidal except when the particles are close enough for multiple hydrodynamic reflections to be important.

The structure in the pair probability resulting from two-particle encounters in a polydisperse suspension decays too quickly with radial separation to affect the divergence problems discussed above. In order to obtain a deficit of one neighbouring particle in a dilute suspension, leading to Debye-like screening, the pair probability must decay at least as slowly as a^3x^{-3} as $x/a \rightarrow \infty$, so that the integral in (2.14) does not converge to an $O(\phi)$ result within an $O(a)$ radial separation. In a dilute suspension, a non-uniform structure must extend to a radial separation large compared with the particle radius before it can constitute an $O(1)$ net average particle deficit. Thus, although two-particle encounters occur in polydisperse suspensions of spheres, any long-range structure that could affect the divergence of the variance and diffusivity can only result from encounters of three or more particles.

In §3 we study the effects of three-particle interactions on the pair probability in a monodisperse suspension of spheres. It will be seen that the pair probability consists of an $O(n)$ short-range contribution and an $O(n\phi)$ long-range contribution. These will be treated in §§3.2 and 3.1, respectively. In §3.1.1, we determine the pair probability at separations x, such that $a \ll x \ll a\phi^{-1}$. From this study it is seen that the long-range contribution represents a net deficit of neighbouring particles and has sufficient range and magnitude to lead to screening of the velocity disturbance at a radial distance of order $a\phi^{-1}$. In §3.1.2, we present a self-consistent theory for the screening of the velocity disturbance and the pair probability at an $O(a\phi^{-1})$ separation. In §4, we discuss the effects of the predicted suspension structure on the properties of the suspension.

2. The effects of structure on suspension properties

It has been recognized for some time that the direct evaluation of the properties of sedimenting suspension by summing the effects of single particles and small groups of particles in a random suspension leads to non-convergent integrals. These convergence difficulties arise from the slow decay of the velocity disturbance caused by a particle settling in a viscous fluid. Because a particle moving through a fluid under the action of a body force acts as a source of momentum, it causes a velocity disturbance which decays like x^{-1} as $x \to \infty$, where x is the radial distance from the particle.

Caflisch & Luke (1985) showed that the finite size of a particle did not affect the convergence problems encountered in calculating the variance of the sedimentation velocity. Thus, in our discussion we shall adopt a point-particle approximation, in which the fluid velocity $\boldsymbol{u}$ and dynamic pressure p^* at a point $\boldsymbol{x}$ in the suspension satisfy

$$-\mu\nabla^2\boldsymbol{u}+\nabla p^* = \sum_{i=1}^{N} \boldsymbol{f}\delta(\boldsymbol{x}-\boldsymbol{r}_i), \tag{2.1a}$$

$$\nabla\cdot\boldsymbol{u} = 0, \tag{2.1b}$$

where ∇ is the Nablé operator, μ is the fluid viscosity, $\boldsymbol{r}_i$ is the position of the ith particle, and $\boldsymbol{f}=\frac{4}{3}\pi a^3\Delta\rho\boldsymbol{g}$ is the force of gravity acting on a particle. Here, $\Delta\rho$ is the density difference between the particle and fluid, and $\boldsymbol{g}$ is the acceleration due to gravity. In the point-particle approximation the particle velocity $\boldsymbol{U}(\boldsymbol{r}_1)$ is given by the sum of the Stokes settling velocity $\boldsymbol{U}^s$ of the particle in an unbounded, quiescent fluid and the fluid velocity $\boldsymbol{u}(\boldsymbol{r}_1)$ induced by all of the other particles, i.e. excluding the direct influence of the particle located at $\boldsymbol{r}_1$,

$$\boldsymbol{U}(\boldsymbol{r}_1) = \boldsymbol{U}^s + \left[\langle\boldsymbol{u}\rangle_1(\boldsymbol{x}\,|\,\boldsymbol{r}_1) - \frac{1}{8\pi\mu r'}\boldsymbol{f}\cdot\left(\boldsymbol{I}-\frac{\boldsymbol{r}'\boldsymbol{r}'}{(r')^2}\right)\right]_{\boldsymbol{x}=\boldsymbol{r}_1} \tag{2.2}$$

where $\boldsymbol{r}' \equiv \boldsymbol{x}-\boldsymbol{r}_1$. The term in the square brackets in (2.2) is the fluid velocity at $\boldsymbol{x}$ excluding the direct influence of the particle located at $\boldsymbol{r}_1$ (Saffman 1973). We shall use a number of types of configurational ensemble average in this analysis. $\langle M(\boldsymbol{x})\rangle$ indicates the unconditional ensemble average of a dependent variable M at a point $\boldsymbol{x}$ – the average over the ensemble of all possible configurations of the suspension with each configuration weighted by the probability that it would arise in a physical experiment. $\langle M\rangle_1(\boldsymbol{x}\,|\,\boldsymbol{r}_1)$ denotes the conditional average, i.e. the average over the subensemble of configurations in which a particle is located at $\boldsymbol{r}_1$. $\langle M\rangle_2(\boldsymbol{x}\,|\,\boldsymbol{r}_1,\boldsymbol{r}_2)$ is the conditional average with two particle positions held fixed at $\boldsymbol{r}_1$ and $\boldsymbol{r}_2$.

If one attempts to calculate the influence of surrounding particles on the mean sedimentation velocity of a given particle by directly summing the effects of each particle separately, one encounters a volume integral of the fluid velocity disturbance caused by a particle; this integral diverges like r'^2 as $r' \to \infty$. Batchelor (1972) overcame this convergence difficulty and calculated the first effects of interparticle hydrodynamic interactions on the sedimentation velocity in a dilute, random, monodisperse suspension of spheres. In the point-particle approximation Batchelor's renormalization is equivalent to adjusting the reduced pressure $p \equiv p^*-nfz$ to reflect the increase in the average density of the suspension by an amount nf/g due to the presence of the particles (Saffman 1973). In the above discussion, z is the vertical coordinate and n is the average number density of particles. Substituting the adjusted reduced pressure into (2.1*a*), one obtains

$$-\mu\nabla^2\boldsymbol{u}+\nabla p=\boldsymbol{f}\left[\sum_{i=1}^{N}\delta(\boldsymbol{x}-\boldsymbol{r}_i)-n\right]. \tag{2.3}$$

Performing the conditional ensemble average of the equations of motion, (2.3) and (2.1*b*), one obtains

$$-\mu\nabla^2\langle\boldsymbol{u}\rangle_1(\boldsymbol{x}\,|\,\boldsymbol{r}_1)+\nabla\langle p\rangle_1=\boldsymbol{f}[g(\boldsymbol{x}\,|\,\boldsymbol{r}_1)-n+\delta(\boldsymbol{x}-\boldsymbol{r}_1)], \tag{2.4a}$$

$$\nabla\cdot\langle\boldsymbol{u}\rangle_1(\boldsymbol{x}\,|\,\boldsymbol{r}_1)=0, \tag{2.4b}$$

where $g(\boldsymbol{x}\,|\,\boldsymbol{r}_1)$ is the number density of particles at $\boldsymbol{x}$ given a particle located at $\boldsymbol{r}_1$. Substituting the solution of (2.4) obtained using the fundamental solution of the Stokes equations into (2.2), we find that the average particle velocity is

$$\langle \boldsymbol{U}\rangle_1(\boldsymbol{r}_1\,|\,\boldsymbol{r}_1)=\boldsymbol{U}^s+\int \mathrm{d}\boldsymbol{x}\,[g(\boldsymbol{x}\,|\,\boldsymbol{r}_1)-n]\frac{1}{8\pi\mu r'}\boldsymbol{f}\cdot\left\{\boldsymbol{I}-\frac{\boldsymbol{r}'\boldsymbol{r}'}{r'^2}\right\}. \tag{2.5}$$

Far from the particle at $\boldsymbol{r}_1$ the pair probability $g(\boldsymbol{x}\,|\,\boldsymbol{r}_1)$ approaches the average number density n. If $g(\boldsymbol{x}\,|\,\boldsymbol{r}_1)-n$ decays faster than r'^{-2} as $r'\rightarrow\infty$, then the integral in the expression (2.5) for the average sedimentation velocity converges.

In a random suspension of point particles with uniform probability in a volume V, $g(\boldsymbol{x}\,|\,\boldsymbol{r}_1)\equiv(N-1)/V\approx n$ in the limit $N\rightarrow\infty$ with $n\equiv N/V$ held fixed. Thus,

$$\boldsymbol{U}(\boldsymbol{r}_1)=\boldsymbol{U}^s, \tag{2.6}$$

i.e. the average velocity is the same as the Stokes settling velocity of an isolated particle. Note that (2.6) has errors of order ϕ resulting from the neglect of the finite size of the particles. Batchelor (1972) accounted for the effects of finite particle size and found $\boldsymbol{U}=\boldsymbol{U}^s(1-6.55\phi)$ in a random, dilute monodisperse array of spheres. It should be noted that the correction -6.55ϕ applies only to a random array of particles with uniform probability. The structures in the non-Brownian suspensions studied here are determined by the hydrodynamic interactions between the particles and there is no reason to expect such suspensions to be random with uniform probability. Batchelor's (1972) method can be applied to other non-uniform structures, but the resulting correction will depend on the structure.

We shall now examine the problem of determining the variance of the fluid velocity in the suspension. Since the particle velocities are predominately determined by long-range interactions for which the point-particle approximation is appropriate, the variances of the fluid and particle velocities are equal at leading order. The variance of the fluid velocity is, by definition,

$$\langle u^2\rangle=\int \mathrm{d}\boldsymbol{C}_N\,P_N(\boldsymbol{C}_N)\,\boldsymbol{u}(\boldsymbol{x}\,|\,\boldsymbol{C}_N)\cdot\boldsymbol{u}(\boldsymbol{x}\,|\,\boldsymbol{C}_N), \tag{2.7}$$

where P_N is the probability density for finding the N particles in the configuration $\boldsymbol{C}_N$, and $\boldsymbol{C}_N$ denotes the positions of the N particles, i.e. $\boldsymbol{r}_1$, $\boldsymbol{r}_2$, etc. Approximating the fluid velocity induced by all N particles as the sum of the conditionally averaged velocities with one particle fixed, i.e.

$$\boldsymbol{u}(\boldsymbol{x}\,|\,\boldsymbol{C}_N)=\sum_{i=1}^{N}\langle\boldsymbol{u}\rangle_1(\boldsymbol{x}\,|\,\boldsymbol{r}_i)+\sum_{i=1}^{N}\sum_{j>i}\langle\boldsymbol{u}'\rangle_2(\boldsymbol{x}\,|\,\boldsymbol{r}_i,\boldsymbol{r}_j)+\ldots, \tag{2.8}$$

the velocity variance (2.7) becomes

$$\langle u^2\rangle \approx \int \mathrm{d}\boldsymbol{r}_1\, n\langle \boldsymbol{u}\rangle_1(\boldsymbol{x}\,|\,\boldsymbol{r}_1)\cdot\langle \boldsymbol{u}\rangle_1(\boldsymbol{x}\,|\,\boldsymbol{r}_1) - \int \mathrm{d}\boldsymbol{r}_1\, d\boldsymbol{r}_2\, n[g(\boldsymbol{r}_1\,|\,\boldsymbol{r}_2)-n]\langle \boldsymbol{u}\rangle_1(\boldsymbol{x}\,|\,\boldsymbol{r}_1)\cdot\langle \boldsymbol{u}\rangle_1(\boldsymbol{x}\,|\,\boldsymbol{r}_2). \quad (2.9)$$

Equation (2.9) is the first approximations to the variance in a dilute suspension. The next approximation in this expansion involves the conditionally averaged velocity with two-particle positions held fixed. In the point-particle approximation two-particle velocity correlations arise only from three-particle position correlations, i.e. from $g_3(\boldsymbol{x}\,|\,\boldsymbol{r}_2,\boldsymbol{r}_1)-g(\boldsymbol{x}\,|\,\boldsymbol{r}_1)-g(\boldsymbol{x}\,|\,\boldsymbol{r}_2)+n$, where $g_3(\boldsymbol{x}\,|\,\boldsymbol{r}_2,\boldsymbol{r}_1)$ is the particle density at $\boldsymbol{x}$ when two particle positions are held fixed at $\boldsymbol{r}_1$ and $\boldsymbol{r}_2$. In the Appendix, it is shown that the errors introduced by the approximation in (2.9) are small in the limit $\phi\rightarrow 0$. Note that the leading term (2.9) in the asymptotic expansion for the variance *is* affected by the pair probability, despite the fact that it involves only the conditionally averaged velocity with one particle fixed, cf. (2.4).

In a dilute random array with uniform probability, there are no three-particle position correlations so the equality in (2.9) is exact. Furthermore, the pair probability is equal to the number density, $g(\boldsymbol{r}_2\,|\,\boldsymbol{r}_1)=n$, in such an array, so the second integral in (2.9) is zero and the conditionally averaged velocity (2.4) is simply a Stokeslet which decays like x^{-1} with distance x away from the fixed particle.† Thus, the determination of the variance of the fluid velocity (2.9) in a random array requires the evaluation of a volume integral of the square of velocity due to a point force in pure fluid. This integral diverges like x as $x\rightarrow\infty$ indicating that the variance grows in proportion to the linear dimension of the suspension. Considerations of the finite size of the particles do not affect the convergence of (2.9). In fact, the largest effect associated with the finite particle size (which comes from the product of the Stokeslet with the potential dipole) makes an $O(x^{-4})$ contribution to the integrand in (2.7).

However, it will now be shown that a suspension with a certain type of structure has a finite velocity variance. First note that (2.4) may be solved upon Fourier transforming to obtain

$$\langle \hat{\boldsymbol{u}}\rangle_1(\boldsymbol{k}) = \frac{\boldsymbol{f}\cdot(\boldsymbol{I}-\boldsymbol{k}\boldsymbol{k}/k^2)}{(2\pi k)^2\mu}[1+\hat{\rho}(\boldsymbol{k})], \quad (2.10)$$

where ˆ indicates the transform, $\boldsymbol{k}$ is the transform variable corresponding to $\boldsymbol{x}-\boldsymbol{r}_1$, and $\rho\equiv g-n$ is the difference between the pair probability and the number density. The Dirac delta function can be used to write the right-hand side of (2.9) in terms of a single integral, i.e.

$$\langle u^2\rangle \approx \int \mathrm{d}\boldsymbol{r}_1\, \mathrm{d}\boldsymbol{r}_2\, n[\delta(\boldsymbol{r}_2-\boldsymbol{r}_1)-\rho(\boldsymbol{r}_2-\boldsymbol{r}_1)]\,\langle \boldsymbol{u}\rangle_1(\boldsymbol{x}\,|\,\boldsymbol{r}_1)\cdot\langle \boldsymbol{u}\rangle_1(\boldsymbol{x}\,|\,\boldsymbol{r}_2). \quad (2.11)$$

Using the convolution theorem twice, (2.11) may be written in terms of a single integral in Fourier space, i.e.

$$\langle u^2\rangle = n\int \mathrm{d}\boldsymbol{k}\; [1-\hat{\rho}(\boldsymbol{k})]\,\langle \hat{\boldsymbol{u}}\rangle_1(\boldsymbol{k})\cdot\langle \hat{\boldsymbol{u}}\rangle_1(-\boldsymbol{k}). \quad (2.12)$$

† Strictly speaking the pair probability in a suspension with uniform probability is $g(\boldsymbol{r}_2\,|\,\boldsymbol{r}_1) = n-1/V$. However, if we take the limit $V\rightarrow\infty$ first (before the limit $x\rightarrow\infty$), then $g\rightarrow n$. The contribution $-1/V$ is only important for separations x comparable with the linear dimension of the entire system. With a pair probability $g=n-1/V$, one still obtains an $O(U^{s2}L\phi/a)$ variance, although with a different coefficient than that calculated by Caflisch & Luke (1985) who omitted the $-1/V$ term.

Finally, substituting (2.10) into (2.12) one finds that the variance is

$$\langle u^2 \rangle = n \int \mathrm{d}\boldsymbol{k} \frac{[\boldsymbol{f}\cdot(\boldsymbol{I}-\boldsymbol{k}\boldsymbol{k}/k^2)]^2}{(2\pi k)^4 \mu^2}[1-\hat{\rho}(\boldsymbol{k})][1+\hat{\rho}(\boldsymbol{k})][1+\hat{\rho}(-\boldsymbol{k})]. \tag{2.13}$$

If the suspension is random with uniform probability, $\rho = 0$, and the integral in (2.13) diverges like k^{-1} as $k \rightarrow 0$, corresponding to a divergence of the real-space integral in (2.7) like x as $x \rightarrow \infty$. However, if $\hat{\rho} \sim -1+o(k)$ as $k \rightarrow 0$, then the integral in (2.13) converges and the variance of the fluid velocity is finite. The real-space equivalent of this criterion is that

$$\int_{x<R_i} \mathrm{d}\boldsymbol{x}[g(\boldsymbol{x}\,|\,\boldsymbol{0})-n] = -1+o(R_i^{-1}), \quad R_s \ll R_i \ll L. \tag{2.14}$$

Thus, the variance of the velocity has a finite value if and only if there is a net deficit of one particle in the suspension surrounding any given particle. We have denoted by R_s the radius of the region surrounding a given particle in which this net deficit exists, i.e. the radial distance at which (2.14) converges to -1. The transform space integral in (2.13) converges at a wavenumber $k \sim O(R_s^{-1})$ and so the variance is

$$\langle u^2 \rangle = O\left(\frac{\phi f^2 R_s}{\mu^2 a^3}\right) = O\left((U^s)^2 \phi \frac{R_s}{a}\right). \tag{2.15}$$

The relationship between the variance of the velocity and the structure of the suspension may be explained physically in the following manner. The force of gravity acting on a particle acts as a source of downward momentum, so that the velocity disturbance due to a particle sedimenting in pure fluid decays like x^{-1} as $x \rightarrow \infty$. However, when a particle settles in a suspension in which (2.14) is satisfied, the particle is surrounded by a region whose density is smaller than the average density of the suspension. This buoyant region acts as a source of upward momentum, which balances the downward momentum source in the particle so there is no net momentum source. Thus, outside the radial distance R_s over which this deficit occurs, the conditionally averaged velocity disturbance decays faster than x^{-1} and the integral in (2.13) converges. We shall refer to this phenomenon as screening of the velocity disturbance induced by a particle.

In order to obtain screening, it is essential that the net particle deficit surrounding each particle occur within a *finite* radial distance R_s. This point may be explained with reference to figure 1. Consider a suspension of N particles contained in a vessel of volume V. The integral in (2.14) would be identically equal to -1, if the integral were carried out over the entire volume V of the vessel. This is true because by fixing the position of one of the N particles, only $N-1$ particles are left to occupy the remaining volume. One can visualize this particle deficit as a 'ghost' particle. In a suspension of uniform probability the ghost particle is equally likely to be found at any point in the volume V, cf. figure 1(*a*). However, when (2.14) is satisfied, the ghost particle is within a radial distance R_s of the fixed particle, cf. figure 1(*b*). The screening of the conditionally averaged velocity in figure 1(*b*) occurs when the radial distance from the fixed particle is large compared to the radial distance R_s over which the particle deficit occurs. At such a large radial distance the fixed particle and the ghost particle appear to occupy a compact region of space which contains no net source of momentum and so the velocity disturbance decays more rapidly than x^{-1}. In the suspension with uniform probability in figure 1(*a*), the ghost particle is found with equal probability throughout the volume V. In such a suspension, it is not

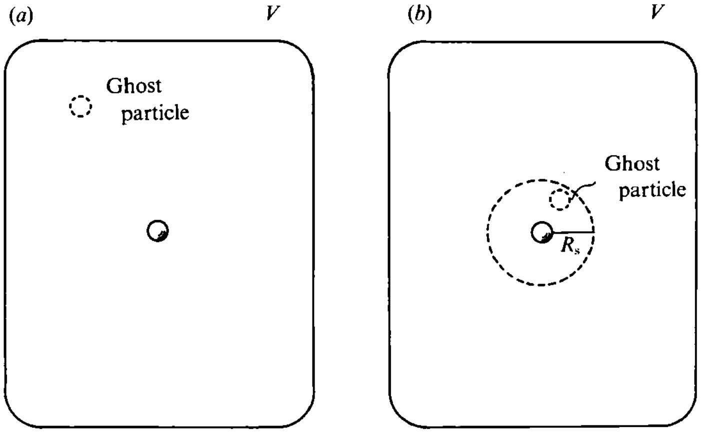

FIGURE 1. The structure of a suspension of N particles contained in a volume V is illustrated. The unbroken circle denotes one particle whose position is held fixed in the conditional ensemble average. Because one particle's position has been fixed, only N-1 particles inhabit the remaining volume. The one 'missing' or 'ghost' particle is represented by a circle with dotted lines. The structure with and without screening of the particles' velocity disturbances are respectively illustrated by (b) and (a).

possible to find a point within the suspension at which the particle and the ghost appear to occupy a compact region of space and the velocity disturbance is not screened. If we substitute in (2.15) the linear dimension L of the settling vessel for the lengthscale R_s over which the particle deficit occurs in a suspension with uniform probability, we recover Caflisch & Luke's (1985) result for the variance of the velocity in such a suspension.

Other physical systems in which particles give rise to long-range fields similar to the long-range velocity fields in a sedimenting suspension are an ionic solution and an array of bodies with gravitational interactions. The charge of a single ion gives rise to an electrostatic potential which decays like x^{-1} as $x \rightarrow \infty$, and the gravitational potential due to a massive object also decays like x^{-1}. As a result the variance of the potential in a uniform distribution of either ions or massive objects is not finite. However, if the distribution of ions is such that there is an excess of oppositely charged ions and/or a deficit of like ions leading to a net counterbalancing opposite charge in the vicinity of any given ion, then the electrical potential will be screened. In an ionic solution, electrical repulsion and attraction lead to the net excess of oppositely charged ions and deficit of like ions required to yield Debye screening of the electrical potential (Lifschitz & Pitaevskii 1981).

It is not immediately obvious whether hydrodynamic interactions between uncharged, sedimenting particles will result in a net deficit or increase in particle density near a given particle. This is the primary issue to be addressed in §3 of this paper for interactions between spherical particles. It will be seen that interactions in suspensions of spherical particles do lead to a deficit of neighbouring particles. A self-consistent approximation of the process by which the coupling between momentum and particle conservation leads to a Debye-like screening is given in §3.1.2. However, in a companion paper (Koch & Shaqfeh 1989), it is shown that sedimenting spheroids tend to clump together and that a suspension of such particles is unstable to particle number density fluctuations. It appears that the question of whether a suspension is stable and has screening or is unstable depends on the details of the particle interactions.

3. Pair probability

In this section, we shall study the effects of hydrodynamic interactions on the pair probability in a dilute, monodisperse, homogeneous suspension. One would normally expect that the pair probability in a dilute suspension would be determined by two-particle interactions, encounters between three or more particles being much more rare. However, the relative position of two identical sedimenting spheres only changes under the influence of a third particle, so one must consider three-particle interactions even in the dilute limit.

In §2 we saw that a non-uniform long-range structure of the pair probability can have a large effect on such properties of a sedimenting suspension as the variance of sedimentation velocity and the self-diffusivity of the particles. It was shown that, if a long-range structure corresponding to a net deficit of one particle in the vicinity of each particle exists, namely (2.14), then the velocity disturbance caused by a particle is screened and the variance of the settling velocity and the self-diffusivity are finite. Equation (2.14) can only be satisfied if the pair probability is smaller than the particle number density and decays at least as slowly as a^3x^{-3} as $xa^{-1} \to \infty$.

Because we are primarily interested in the long-range structure of the pair probability it might seem reasonable to adopt a point-particle approximation for three-particle encounters. In this approximation each particle's velocity is taken to be the sum of its Stokes settling velocity and the fluid velocity disturbance induced at its position by the other particles treated as point forces (Saffman 1973). One would, therefore, neglect the effect of a particle's finite size on the fluid velocity disturbance it induces, and the effects of hydrodynamic reflections between particle which are only important when the particles' separation is $O(a)$.

In the absence of Brownian motion, the probability density $P_N(\boldsymbol{C}_N)$ for the positions of a group of N particles satisfies a conservation equation of the form

$$\frac{\partial P_N}{\partial t} + \sum_{i=1}^{N} \boldsymbol{\nabla}_i \cdot \boldsymbol{U}_i P_N = 0, \tag{3.1}$$

where $\boldsymbol{U}_i$ is the velocity of the ith particle, $\boldsymbol{\nabla}_i$ is the Nablé operator with the derivatives taken with respect to the position $\boldsymbol{r}_i$ of the ith particle. We shall adopt the normalization condition of Batchelor (1972), i.e.

$$\int \mathrm{d}\boldsymbol{x}_1 \ldots \mathrm{d}\boldsymbol{x}_k P_k(\boldsymbol{x}_1, \ldots, \boldsymbol{x}_k) = \frac{N!}{(N-k)!},$$

for all $k \leqslant N$. Because the fluid velocity is solenoidal and the Stokes settling velocity is a constant, we have $\boldsymbol{\nabla}_i \cdot \boldsymbol{U}_i = 0$ for all i in the point-particle approximation. As a result, a probability density, $P_N = N!/V^N$, that is constant independent of $\boldsymbol{r}_1, \ldots, \boldsymbol{r}_N$ or, in other words, random with uniform probability, is a solution of the particle conservation equation (3.1).† One particular set of initial and boundary conditions that would lead to such a constant solution is an initial condition that P_N is a constant in a unit cell with periodicity boundary conditions. For example, this indicates that if one performs an ensemble of numerical experiments in which N point particles are placed randomly with uniform probability in a periodic cell, the

† This solution is neutrally stable in the following sense. If we add a small disturbance g to the probability density P_N at $t = 0$, such that the maximum absolute value of g is less than ϵ, the maximum absolute value of the disturbance g will remain smaller than ϵ for all subsequent times. This simply results from the facts that the equation for P_N is a linear first order equation and the velocity is incompressible. The problem of determining P_N for point particles is equivalent to the advection of a passive, non-diffusive tracer by an incompressible flow in a $3N$-dimensional space.

probability density will remain uniform at all subsequent times. Thus, at least in this case, one must consider interactions in which at least two of the particles are within an $O(a)$ distance of one another, so that the point-particle approximation does not apply, in order to obtain a non-uniform structure.

Caflisch & Luke (1986) numerically simulated encounters between three identical spherical sedimenting particles, using the point-particle approximation. They then attempted to derive some information concerning the pair probability from these encounters, by assuming that the pair probability could be written in the form $g(\boldsymbol{x}\,|\,\boldsymbol{0}) = x^{\alpha}h(\cos\Theta)$, where α is an undetermined constant and Θ is the angle between $\boldsymbol{x}$ and the direction of gravity. Caflisch & Luke (1986) claimed to be able to determine that $h(\cos\Theta)$ reflected an excess of vertically oriented pairs without any knowledge of α. Although we have no reason to doubt the validity of Caflisch & Luke's simulations, we question their conclusions concerning the pair probability. Indeed, based on the preceding discussion, we believe they should not have obtained any non-uniform structure at all from their study.

We have seen that (at least in a random, homogeneous suspension with periodic boundary conditions in which the N-particle probability is initially uniform) a non-uniform probability density can only arise from those three-particle interactions in which at least two of the particles are separated by an $O(a)$ distance. On the other hand, encounters in which all three particles are within an $O(a)$ distance are much less frequent than those in which an isolated particle interacts with a close pair. Thus, we anticipate that the latter type of interaction will yield the largest contribution to the pair probability.

We consider, then, the joint probability $n^3m(\boldsymbol{x},\boldsymbol{r})$ for finding a close pair of particles, whose relative position $\boldsymbol{r}$ has an $O(a)$ magnitude, at a position $\boldsymbol{x}$ relative to a third, distant particle, i.e. $x \gg a$. The probability density for finding three particles none of which are close to one another is n^3. As a result, we have $\lim_{r\to\infty} m(\boldsymbol{x},\boldsymbol{r}) = 1$. In addition, the probability density of finding a close pair at a distance x from the third particle that is too great for the third particle to have affected the motion of the pair is $n^3\Omega(\boldsymbol{r})$, so that $\lim_{x\to\infty} m(\boldsymbol{x},\boldsymbol{r}) = \Omega(\boldsymbol{r})$. Here, $n^2\Omega(\boldsymbol{r})$ is the probability density of close pairs.

Neglecting the effects of interactions between four or more particles, the probability density $m(\boldsymbol{x},\boldsymbol{r})$ satisfies a conservation equation of the form

$$\frac{\partial m}{\partial t}+\boldsymbol{\nabla}\cdot(\boldsymbol{U}^{\mathrm{r}}\,m)+\boldsymbol{\nabla}_r\cdot(\mathring{\boldsymbol{r}}m) = 0, \tag{3.2}$$

where $\mathring{\boldsymbol{r}}$ is the relative velocity between the close pairs of particles and $\boldsymbol{U}^{\mathrm{r}}$ is the velocity of the pair relative to the distant, third particle, and $\boldsymbol{\nabla}$ and $\boldsymbol{\nabla}_r$ are the Nablé operators with the derivatives taken with respect to $\boldsymbol{x}$ and $\boldsymbol{r}$, respectively. It will be seen that the neglect of interactions between four or more particles is only justified for $x \ll a\phi^{-1}$. For larger separations $x = O(a\phi^{-1})$, the surrounding particles in the suspension will lead to a significant screening of the velocity disturbance caused by the distant particle, and will also lead to a significant relative motion of the close pair. Thus, at present, we shall restrict our attention to predicting the probability density for separations $a \ll x \ll a\phi^{-1}$.

When $x \gg a$, the relative velocity $\boldsymbol{U}^{\mathrm{r}}$ between the close pair and the third particle is approximately equal to the difference in their settling velocities in a quiescent, unbounded fluid, i.e.

$$\boldsymbol{U}^{\mathrm{r}} = (F-1)\,\boldsymbol{U}^{\mathrm{s}}+G\boldsymbol{U}^{\mathrm{s}}\cdot\frac{\boldsymbol{rr}}{r^2}, \tag{3.3}$$

where $F+G$ and F are inverse resistance coefficients for the motion of two identical particles parallel and perpendicular to their line of centres, respectively (Batchelor 1972; Stimson & Jeffery 1926; Goldman, Cox & Brenner 1966). F and G are functions of r and are always positive.

The relative velocity $\mathring{\boldsymbol{r}}$ between the two particles comprising the close pair is caused by the fluid velocity disturbance induced by the third particle. When the third particle is at a distance $a\phi^{-1} \gg x \gg a$, its fluid velocity disturbance may be approximated as that due to a Stokeslet, and, furthermore, it is approximately a linear shear flow on the scale of the close pair separation r. The relative velocity is then (Batchelor & Green 1972):

$$\mathring{r}_i = \frac{\partial u_i^s}{\partial x_j} r_j - \left[A \frac{r_i r_j}{r^2} + B\left(\delta_{ij} - \frac{r_i r_j}{r^2}\right)\right] E_{jk} r_k, \tag{3.4a}$$

where
$$\boldsymbol{E} = \tfrac{1}{2}[\nabla \boldsymbol{u}^s + \nabla \boldsymbol{u}^{s\dagger}], \tag{3.4b}$$

A and B are positive functions of r tabulated by Batchelor & Green (1972), and $A > B$ for all r. Another useful relation is

$$\nabla_r \cdot \mathring{\boldsymbol{r}} = W \frac{\boldsymbol{r}\cdot\boldsymbol{E}\cdot\boldsymbol{r}}{r^2} = W \frac{\boldsymbol{r}\cdot\nabla\boldsymbol{u}^s\cdot\boldsymbol{r}}{r^2}, \tag{3.5}$$

where $W = r(\mathrm{d}A/\mathrm{d}r) + 2\ (A-B)$ is tabulated by Batchelor (1977) and is always positive. The velocity $\boldsymbol{u}^s$ due to a Stokeslet satisfies the equations (Saffman 1973)

$$-\mu\nabla^2\boldsymbol{u}^s + \nabla p = \boldsymbol{f}\delta(\boldsymbol{x}), \tag{3.6a}$$

$$\nabla\cdot\boldsymbol{u}^s = 0. \tag{3.6b}$$

The solution of (3.6) in Fourier space is

$$\hat{\boldsymbol{u}}^s(\boldsymbol{k}) = \frac{(\boldsymbol{I} - \boldsymbol{k}\boldsymbol{k}/k^2)\cdot\boldsymbol{f}}{\mu(2\pi k)^2}. \tag{3.7}$$

Three-particle interactions affect the pair probability in two ways. First, a close pair of particles only undergoes relative motion under the influence of the shearing motion induced by a third particle. Thus, the probability that two particles are at a specified relative position of $O(a)$ magnitude is determined by the previous three-particle encounters experienced by the pair. This effect, treated in §3.2, leads to a short-range structure of the pair probability $g(\boldsymbol{r}_2|\boldsymbol{r}_1)$ that decays like na^6x^{-6} as $a/x \to \infty$. Here, $x \equiv |\boldsymbol{r}_2 - \boldsymbol{r}_1|$. Because this structure decays more rapidly than a^3x^{-3} as $x/a \to \infty$, it cannot lead to a Debye-like screening of the velocity disturbance, cf. (2.14). The second way in which three-particle interactions affect the pair probability is through the influence of a third particle that is currently within an $O(a)$ distance of either the one at $\boldsymbol{r}_2$ or at $\boldsymbol{r}_1$. Although the latter effect makes a small $O(n\phi)$ change in the pair probability, it will be seen in §3.1 that this change decays very slowly (like a/x) with radial separation, and can thus lead to Debye-like screening.

It is necessary to relate the pair probability g to the probability density m for groups of three particles. If we had an exact expression for the three-particle probability $P_3(\boldsymbol{r}_1, \boldsymbol{r}_2, \boldsymbol{r}_3)$, then the relation

$$ng(\boldsymbol{r}_2|\boldsymbol{r}_1) = \frac{1}{(N-2)}\int \mathrm{d}\boldsymbol{r}_3 P_3(\boldsymbol{r}_2 - \boldsymbol{r}_1, \boldsymbol{r}_3 - \boldsymbol{r}_2) \tag{3.8}$$

would yield an exact result for the pair probability. However, as noted by Rallison & Hinch (1986), an $O(1)$ estimate for P_3 in (3.8), for example $P_3 \approx n^3 m$, does not yield an $O(1)$ estimate for g. The difficulty is that using (3.8) with the estimate of the three-particle probability prescribed above only takes account of the effect of the *third* particle on the pair probability. The pair probability is actually affected by the presence of all the other N-2 particles in the suspension.

To take account of the effects of all N-2 other particles, it is necessary to relate the pair probability to the N-particle probability, i.e.

$$ng(\boldsymbol{r}_2 \mid \boldsymbol{r}_1) = \frac{1}{(N-2)!}\int \mathrm{d}\boldsymbol{r}_3 \dots \mathrm{d}\boldsymbol{r}_N\, P_N(\boldsymbol{C}_N). \tag{3.9}$$

An estimate for the N-particle probability density, that takes account of the variation of the probability density from its random value $N!/V^N$ when any of one of the N-2 other spheres are near either the one at $\boldsymbol{r}_2$ or the one at $\boldsymbol{r}_1$, is

$$\begin{aligned} P_N(\boldsymbol{C}_N) = {} & n^2\Omega(\boldsymbol{r}_2-\boldsymbol{r}_1)P_N(\boldsymbol{C}_N \mid \boldsymbol{r}_1,\boldsymbol{r}_2) \\ & + \sum_{i=3}^{N} n^3[m'(\boldsymbol{r}_1-\boldsymbol{r}_2,\boldsymbol{r}_1-\boldsymbol{r}_i)+m'(\boldsymbol{r}_i-\boldsymbol{r}_1,\boldsymbol{r}_2-\boldsymbol{r}_i)+m'(\boldsymbol{r}_1-\boldsymbol{r}_i,\boldsymbol{r}_2-\boldsymbol{r}_1)] \\ & \times P_N(\boldsymbol{C}_N \mid \boldsymbol{r}_1,\boldsymbol{r}_2,\boldsymbol{r}_i)+\dots, \end{aligned} \tag{3.10}$$

where $m'(\boldsymbol{r}_j-\boldsymbol{r}_k,\boldsymbol{r}_i-\boldsymbol{r}_k) \equiv m-\Omega(\boldsymbol{r}_i-\boldsymbol{r}_k)$. The terms omitted from (3.10) involve correlations between the positions of four or more particles. The order of the arguments of m' is important. The second argument of m' represents the relative position of the two particles constituting the close pair, while the first represents the separation of the close pair from the distant third particle. Thus, the three terms in the square brackets in (3.10) take account of the situations in which particle 2, particle i, and particle 1 are the 'third' distant particle, respectively. Substituting (3.10) into (3.9) and taking the limit $N \rightarrow \infty$ with $N/V \equiv n$ fixed, the pair probability is given by

$$g(\boldsymbol{r}_2 \mid \boldsymbol{r}_1) = n\Omega(\boldsymbol{r}_2-\boldsymbol{r}_1)+2n^2\int \mathrm{d}\boldsymbol{r}_3\, m'(\boldsymbol{r}_2-\boldsymbol{r}_1,\boldsymbol{r}_3-\boldsymbol{r}_2)+n^2\int \mathrm{d}\boldsymbol{r}_3\, m'(\boldsymbol{r}_1-\boldsymbol{r}_3,\boldsymbol{r}_2-\boldsymbol{r}_1). \tag{3.11}$$

The first term on the right-hand side of (3.11) is the $O(n)$ contribution to the pair probability resulting from configurations in which no third particle is close to either of the particles in the pair. It will be seen in §3.2 that this contribution asymptotes to a constant $\Omega \rightarrow 1$ at separations large compared to a. The second term represents the influence on the pair probability of a third particle that is currently within an $O(a)$ distance of one of the members of a pair. This term is only $O(n\phi)$ but decays slowly with radial separation x, giving the long-range structure of the pair probability investigated in §3.1. The third term representing the effect of a third particle on a close pair, for which $x = O(a)$, is both small, $O(n\phi)$, in magnitude and short range, and thus is of little interest.

3.1. *Long-range structure*

In this subsection, we examine the long-range structure of the pair probability that results from the net change in the number density of pairs of particles near any given particle. When a pair of particles falls into the region surrounding an isolated particle, the pair's relative position is influenced by the fluid velocity disturbance caused by the isolated particle. This velocity disturbance, which satisfies (3.6), decays like x^{-1}, with the separation x of the pair from the isolated particle. The relative velocity of the pair decays like x^{-2}, being proportional to the gradient of the fluid velocity disturbance, cf. (3.4). The change in the pair density is related to an integral along the pair's trajectory in $\boldsymbol{x}$ of the divergence with respect to $\boldsymbol{r}$ of the relative velocity, cf. (3.2), and, thus, this change in pair density decays like x^{-1}. Because the number density of particles that are within an $O(a)$ distance of another particle is small, $O(n\phi)$, the change in the number density of pairs of particles near the isolated particle yields an $O(n\phi a x^{-1})$ change in the pair density $g(\boldsymbol{x}\,|\,\boldsymbol{0})$. Although this contribution to the pair probability is small everywhere in a dilute suspension, it will represent an $O(1)$ net change in the number of neighbouring particles, thus affecting the conditionally averaged fluid velocity disturbance caused by a particle, at a radial distance $x = O(a\phi^{-1})$, cf. (2.14).

In §3.1.1, we shall study the structure of the pair probability at radial separations $a \ll x \ll a\phi^{-1}$. For separations in this range, the pair probability is determined by three-particle interactions. It will be seen that there is a net deficit of neighbouring particles at distances $a \ll x \ll a\phi^{-1}$. This suggests that the velocity disturbance will be screened at a radial separation $x = O(a\phi^{-1})$ at which many-particle interactions become important. In §3.1.2, we show how the coupling between the pair probability and the velocity disturbance can lead to screening at $x = O(a\phi^{-1})$.

3.1.1. *The pair probability at separations $a \ll x \ll a\phi^{-1}$*

In order to test for the possibility that the structure of the pair probability represents a long-range deficit of neighbouring particles sufficient to give a Debye-like screening of the velocity disturbance, we shall calculate the change in the number of particles within a sphere of radius R of a given particle. This net change, which will be denoted by H, is given by

$$H \equiv \int_{x \leqslant R} \mathrm{d}\boldsymbol{x}\, \rho(\boldsymbol{x}\,|\,\boldsymbol{0}), \tag{3.12}$$

where $\rho(\boldsymbol{x}\,|\,\boldsymbol{0}) \equiv g(\boldsymbol{x}\,|\,\boldsymbol{0}) - n$. We shall determine H for spheres of radii R, where $a \ll R \ll a\phi^{-1}$. On this lengthscale the effect of the particle density change on the velocity disturbance may be neglected and the preceding development of three-particle interactions is valid. If H is negative on such a lengthscale, screening may be anticipated at a larger $O(a\phi^{-1})$ radial distance.

Equation (3.12) may be written in terms of a volume integral over all space using the 'ball' function $\Pi(x/2R)$ of Bracewell (1978) to give

$$H = \int \mathrm{d}\boldsymbol{x}\, \Pi\!\left(\frac{x}{2R}\right) \rho(\boldsymbol{x}\,|\,\boldsymbol{0}), \tag{3.13}$$

where $\Pi(x/2R)$ is 1 for $x < R$ and 0 otherwise. Using the convolution theorem, (3.13) may be written as

$$H = \int \mathrm{d}\boldsymbol{k}\, \hat{\Pi}(-\boldsymbol{k})\, \hat{\rho}(\boldsymbol{k}), \tag{3.14}$$

where the Fourier transform of the ball function is (Bracewell 1978):

$$\hat{\Pi}(\boldsymbol{k}) = \frac{\sin(2\pi kR) - 2\pi kr \cos(2\pi kR)}{2\pi^2 k^3 R^3}. \tag{3.15}$$

Because we are interested in the long-range structure, and the change $m' \equiv m - \Omega(\boldsymbol{r})$ in the pair density decays like a/x, we may approximate (3.3) at steady state, neglecting terms smaller than a^2x^{-2}, as

$$\boldsymbol{U}^{\mathrm{r}} \cdot \nabla m' = -\nabla_r \cdot [\dot{\boldsymbol{r}}\Omega(\boldsymbol{r})]. \tag{3.16}$$

Equation (3.16) may be solved upon Fourier transforming in $\boldsymbol{x}$ to give

$$\hat{m}'(\boldsymbol{k},\boldsymbol{r}) = -\frac{1}{2\pi \mathrm{i}\boldsymbol{k}\cdot\boldsymbol{U}^{\mathrm{r}}}\nabla_r \cdot [\dot{\boldsymbol{r}}\Omega(\boldsymbol{r})]. \tag{3.17}$$

For simplicity we shall consider the case in which the close pair probability is random with uniform probability, i.e. $\Omega(\boldsymbol{r}) = 1$. The resulting calculation will show that there is a long-range deficit of particles under this condition. Using (3.11), (3.15), and (3.17), the expression (3.14) for the net change H in the number particles within a spherical volume of radius R near a given particle is given by

$$H = -\frac{2n^2 f}{\mu}\int \mathrm{d}\boldsymbol{r}\int \mathrm{d}\boldsymbol{k}\left[\frac{\sin(2\pi kR) - 2\pi kR\cos(2\pi kR)}{8\pi^4 k^5 R^3}\right]\left[\frac{W}{k_l U_l^{\mathrm{r}}}\right]\left[\frac{r_i r_j}{r^2}\right]k_i\left[\delta_{jk} - \frac{k_j k_k}{k^2}\right]\delta_{k3}, \tag{3.18}$$

where δ_{k3} is the unit vector in the direction of the gravitational acceleration. The integral with respect to $\boldsymbol{k}$ in (3.18) may be evaluated, using the relations

$$\int \sin\Theta\,\mathrm{d}\Theta\,\mathrm{d}\Phi\frac{k_j}{k_l U_l^{\mathrm{r}}} = \frac{4\pi U_j^{\mathrm{r}}}{U^{\mathrm{r}2}}, \tag{3.19a}$$

$$\int \sin\Theta\,\mathrm{d}\Theta\,\mathrm{d}\Phi\frac{k_i k_j k_k}{U_l^{\mathrm{r}} k_l k^2} = \frac{4\pi}{3U^{\mathrm{r}2}}(U_i^{\mathrm{r}}\delta_{jk} + U_j^{\mathrm{r}}\delta_{ik} + U_k^{\mathrm{r}}\delta_{ij}) - \frac{8\pi}{3U^{\mathrm{r}4}}U_i^{\mathrm{r}}U_j^{\mathrm{r}}U_k^{\mathrm{r}}, \tag{3.19b}$$

to give

$$H = -\frac{n^2 f R^2}{3\mu}\int \mathrm{d}\boldsymbol{r}\, W\left[2\frac{r_i r_j}{r^2}\frac{U_i^{\mathrm{r}}U_j^{\mathrm{r}}U_3^{\mathrm{r}}}{U^{\mathrm{r}4}} + \frac{r_i r_3}{r^2}\frac{U_i^{\mathrm{r}}}{U^{\mathrm{r}2}} - \frac{U_3^{\mathrm{r}}}{U^{\mathrm{r}2}}\right]. \tag{3.20}$$

In (3.19), Θ and Φ are the angular coordinates in a spherical coordinate system in Fourier space. The integrals in (3.19) contain a factor $(U_l^{\mathrm{r}}k_l)^{-1}$, and thus appear superficially to be conditionally convergent. However, the Fourier transform $(U_l^{\mathrm{r}}k_l)^{-1}$ is a generalized function and its integral is defined to be the Cauchy principle value (Lighthill 1980). The use of generalized functions may be avoided by solving (3.16) in real space using Lagrangian coordinates.

Substituting (3.3) for the relative velocity and integrating over the angular coordinates in $\boldsymbol{r}$, (3.20) becomes

$$H = -\frac{4\pi n^2 f R^2}{3\mu U^{\mathrm{s}}}\int_{2a}^{\infty} r^2\,\mathrm{d}r\frac{W}{F'}\Bigg[b^2 - (b^3 + b)\tan^{-1}(b^{-1}) + \frac{b^4(F'+G)^2}{F'^3}\left(3G + \left(\frac{F'}{b} - 3Gb\right)\tan^{-1}(b^{-1}) - \frac{F'+G}{b^2+1}\right)\Bigg], \tag{3.21}$$

where $b^2 \equiv F'^2/(G^2 + 2F'G)$, $F' \equiv F - 1$, and $F + G$ and F are inverse resistance coefficients, cf. (3.3). The integrand in (3.21) is positive over the entire range $\frac{1}{3} \leqslant (G/F') \leqslant 1$ of variation of the ratio G/F' with changing r. Thus, H is negative, indicating that there is a net deficit of particles near any given particle.

Figure 2 illustrates the physical mechanism leading to the computed deficit of

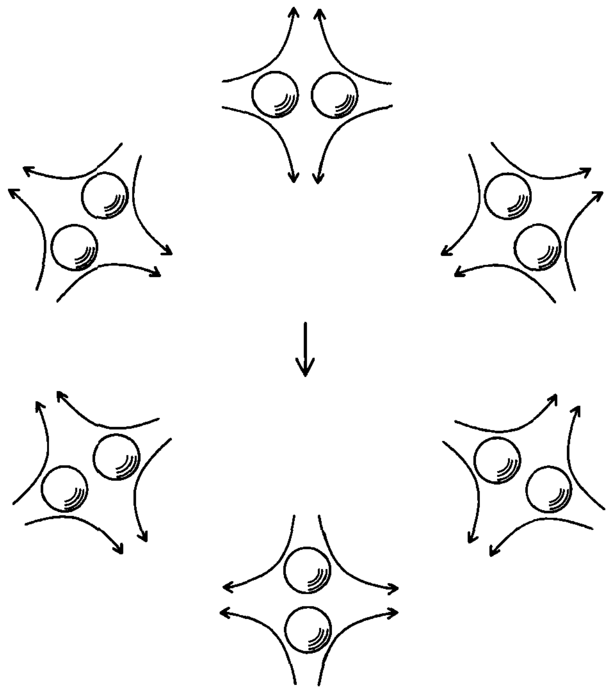

FIGURE 2. The physical mechanism leading to a deficit of close pairs near any given third particle is illustrated. At each position of the close pair relative to the third particle, the extensional component of the velocity disturbance caused by the third particle is illustrated. The pair probability is largest in each case along the compressional axis. The excess of relatively slowly sedimenting horizontal pairs above and rapidly sedimenting vertical pairs below the third particle leads to a net deficit of particles in the neighbourhood of the third particle.

neighbouring particles. The velocity disturbance caused by the third particle is approximately a Stokeslet in the vicinity of the close pair. We are interested in the first effects of a weak velocity disturbance on a pair passing a large distance from the third particle. It is the extensional component of the Stokeslet that determines this small change in the pair probability. The principle axis of extension is always parallel the radial separation vector $\boldsymbol{x}$, but the flow is converging above the Stokeslet and diverging below. Pairs of particles in an extensional flow are drawn together along the compressional axis and pulled apart along the extensional axis. However, as Batchelor (1977) noted in his study of the extensional viscosity of suspensions, the divergence of the relative velocity is negative in the compressional quadrant and positive in the extensional quadrant, corresponding to an apparent source of particle density in the compressional quadrant and a sink in the extensional quadrant. This leads to an increased pair density in the compressional quadrant in both the steady-state shearing of a Brownian suspension studied by Batchelor (1977) and the *transient* shearing of non-Brownian particles treated above. (The *steady* shearing of non-Brownian particles leads to an isotropic pair probability with an excess of close pairs.) The Stokeslet velocity disturbance leads, therefore, to an excess of horizontal pairs above and an excess of vertical particles below the third particle. The horizontal pairs above sediment less rapidly than the vertical pairs below, leading to a net deficit of pairs of particles near any given particle.

Evaluating the integral in (3.21) by numerical quadrature, using cubic spline interpolations of the values tabulated by Batchelor (1977) for W and by Goldman *et al.* (1966) for the inverse resistance coefficients $F+G$ and F, we find the net deficit of particles within a radius R to be

$$H = -11.1\,\phi^2 R^2/a^2. \tag{3.22}$$

The description of particle interactions used in this subsection and, thus, (3.22) are valid for radial distances $a \ll R \ll a\phi^{-1}$.

In this subsection, we have shown that there is a long-range particle deficit, when the close particle pair probability is uniform, $\Omega = 1$. This calculation suggests that such a deficit will exist in the actual suspension. However, it does not preclude the possibility that a structure in the close pair probability could develop for which there is an excess rather than a deficit of neighbouring particles. If such an excess did develop, it would be expected to lead to an instability of the type described in Koch & Shaqfeh (1989) for sedimenting spheroids. In the following analysis, we shall develop a self-consistent theory for the screening of the particle velocity disturbance due to a deficit of neighbouring particles.

3.1.2. *Screening of the velocity disturbance at $O(a\phi^{-1})$ radial separations*

The particle deficit within a sphere of radius R becomes $O(1)$, when $R = O(a\phi^{-1})$. At this radial separation one can no longer treat three interacting particles as if they were surrounded by a quiescent, pure fluid. One change in the preceding analysis that needs to be made to treat three-particle interactions when $x = O(a\phi^{-1})$ is to replace the velocity disturbance of an isolated Stokeslet (3.6) with the averaged velocity $\langle \boldsymbol{u}\rangle_1(\boldsymbol{x}\,|\,\boldsymbol{0})$ conditioned to a particle fixed at the origin. This conditionally averaged velocity satisfies (2.4). The conditionally averaged velocity is affected not only by the downward gravitational force acting on the particle at the origin but also by the upward buoyancy force caused by the particle deficit in the surrounding suspension. Thus, at $x = O(a\phi^{-1})$, the conditionally averaged velocity disturbance seen by a pair of particles decreases and so the deficit in the pair probability begins to decrease. It will be shown that this coupling between the conditionally averaged velocity and the pair probability will lead to a net deficit of one particle and a screening of the velocity disturbance.

In addition, the presence of many intervening particles changes the nature of the relative trajectories of the three particles. At separations $x = O(a\phi^{-1})$, the relative velocity between the close pair and the third particle are changed by an $O(U^s)$ amount due to interactions with the other particles in the suspensions. Since the velocity in the suspension remains correlated over an $O(a\phi^{-1})$ distance, the motion of particles on lengthscales larger than $a\phi^{-1}$ may be described by an effective diffusivity tensor. An exact description of the relative motion on lengthscales of $O(a\phi^{-1})$ would require a non-local diffusivity tensor (cf. Koch & Brady 1987) that is a function of relative position and the direction of gravity. In this section, we shall adopt a simpler model, in which the relative motion of the close pair and the third particle induced by the other particles in the suspension is described by a constant isotropic diffusivity whose value will be calculated in a self-consistent manner. Even for this relatively simple model the analysis of the flow is quite involved.

Thus, we want to solve (2.10) and (3.11) for the conditional-average velocity disturbance and the pair probability self-consistently. At large separations, $x \gg a$, ρ in (3.11) is well approximated by the most slowly decaying contribution, i.e.

$$\rho = 2n^2 \int \mathrm{d}\boldsymbol{r}\, m'(\boldsymbol{x},\boldsymbol{r}). \tag{3.23}$$

The three-particle probability is given by (3.16) with an additional term describing the relative diffusion between the close pair and third particle induced by the other particles, i.e.

$$\boldsymbol{U}^{\mathrm{r}} \cdot \nabla m' - 2D\nabla^2 M' = -\nabla_r \cdot [\mathring{\boldsymbol{r}} \Omega(\boldsymbol{r})], \tag{3.24}$$

where D is the effective diffusivity of an isolated particle. In writing (3.24), we have taken the diffusivity of the close pair to be the same as that of an isolated particle and we have neglected any correlation between the diffusive motions of the close pair and the third particle. The solution of (3.24) upon Fourier transforming in $\boldsymbol{x}$ is

$$\hat{m}'(\boldsymbol{k}, \boldsymbol{r}) = -\frac{1}{2\pi \mathrm{i}\boldsymbol{k} \cdot \boldsymbol{U}^{\mathrm{r}} + 8\pi^2 Dk^2} \nabla_r \cdot [\mathring{\boldsymbol{r}} \Omega(\boldsymbol{r})]. \tag{3.25}$$

In (3.24) and (3.25), the relative velocity of the close pair $\mathring{\boldsymbol{r}}$ is that caused by a partially screened Stokeslet rather than the 'unscreened' Stokeslet used in §3.1.1. In other words, $\boldsymbol{u}^{\mathrm{s}}$ in (3.4) and (3.5) is replaced by $\langle \boldsymbol{u} \rangle_1$. Substituting (2.10) and (3.5) into (3.25) and restricting our attention at present to the case in which there is no short-range structure, i.e. $\Omega = 1$, we obtain

$$\hat{m}' = -\frac{\boldsymbol{k}(\boldsymbol{f} \cdot (\boldsymbol{I} - \boldsymbol{kk}/k^2)) : (\boldsymbol{rr}/r^2)\, W(1+\hat{\rho})}{(\boldsymbol{k} \cdot \boldsymbol{U}^{\mathrm{r}} - 4\pi \mathrm{i} Dk^2)(2\pi k)^2 \mu}. \tag{3.26}$$

Substituting (3.26) into (3.23) and solving for $\hat{\rho}$, one obtains

$$\hat{\rho} = \frac{-\beta a^{-2} \phi^2}{(2\pi k)^2 + \beta a^{-2} \phi^2}, \tag{3.27}$$

where

$$\beta = \frac{27}{4\pi} \int \mathrm{d}\boldsymbol{r} \frac{k_i \left(\delta_{j3} - \dfrac{k_3 k_j}{k^2}\right) \dfrac{r_i r_j}{r^2} W}{k_k U_k^{\mathrm{r}} - 4\pi \mathrm{i} k^2 (D/U^{\mathrm{s}})}, \tag{3.28}$$

and we have non-dimensionalized r with a and U^{r} with U^{s}. An expression for the conditional-average velocity disturbance can be obtained by substituting (3.27) into (2.10) to give

$$\langle \hat{\boldsymbol{u}} \rangle_1 = \frac{\boldsymbol{f} \cdot (\boldsymbol{I} - \boldsymbol{kk}/k^2)}{\mu[(2\pi k)^2 + \beta a^{-2} \phi^2]}. \tag{3.29}$$

The variance of the velocity in the suspension is given by (2.13), which upon substitution from (3.27), becomes

$$\langle u^2 \rangle = n \int \mathrm{d}\boldsymbol{k} \frac{(\boldsymbol{f} \cdot (\boldsymbol{I} - \boldsymbol{kk}/k^2))^2 \, (2\beta(\boldsymbol{k})\, a^{-2} \phi^2 + (2\pi k)^2)}{\mu^2 [(2\pi k)^2 + \beta(\boldsymbol{k})\, a^{-2} \phi^2]^2 \, [(2\pi k)^2 + \beta(-\boldsymbol{k})\, a^{-2} \phi^2]}. \tag{3.30}$$

The screening of the velocity field in (3.29) and the pair probability disturbance in (3.27) occur because of the terms $\beta a^{-2} \phi^2$ in the denominators of these expressions. For wavenumbers $k \gg a^{-1}\phi$, corresponding to radial separations $x \ll a\phi^{-1}$, the viscous terms $(2\pi k)^2$ in the denominators of (3.27) and (3.29) dominate. Thus, at distances shorter than the screening length $a\phi^{-1}$, the velocity and pair probability disturbances decay like $1/r$ (or $1/k^2$ in Fourier space). At larger separations $x \gg a\phi^{-1}$, corresponding to smaller wavenumbers, the terms $\beta a^{-2} \phi^2$, arising from the particle deficit dominate in the denominators of (3.27) and (3.29). As a result the velocity and pair probability disturbances are screened at separations larger than the screening length.

Upon substituting for the relative velocity from (3.3) and using the definitions $k_3/k = \cos\Theta$, $r_i k_i/rk \equiv \cos\Theta'$, and $r_3/r \equiv \cos\Theta\cos\Theta' + \sin\theta\ \sin\Theta'\ \cos\phi$, (3.28) becomes

$$\beta = \frac{27}{4\pi}\int_2^\infty r^2\,\mathrm{d}r\frac{W}{G}\int_0^\pi \sin\Theta'\,\mathrm{d}\Theta'\int_0^{2\pi}\mathrm{d}\phi\frac{\sigma\cos\phi}{1+\sigma\cos\phi}, \tag{3.31a}$$

where

$$\sigma \equiv \frac{G\sin\Theta\cos\Theta'\sin\Theta'}{F'\cos\Theta + G\cos\Theta\cos^2\Theta' - 2\mathrm{i}KD/U^{\mathrm{s}}}, \tag{3.31b}$$

and $K \equiv 2\pi k$. The integral over the angle ϕ in (3.31) can be evaluated by contour integration in the complex plane. This is accomplished by introducing a change of variables $z = \mathrm{e}^{\mathrm{i}\phi}$ and integrating over the unit circle in the complex plane for z. The result of this integration is

$$\beta = \frac{27}{4\pi}\int_2^\infty r^2\,\mathrm{d}r\frac{W}{G}\int_0^\pi \sin\Theta'\,\mathrm{d}\Theta'\,2\pi\left(1-\frac{H(1-|z_+|)}{(1-\sigma^2)^{\frac{1}{2}}}+\frac{H(1-|z_-|)}{(1-\sigma^2)^{\frac{1}{2}}}\right), \tag{3.32}$$

where $H(x)$ is the Heaviside step function and $z_\pm = -1/\sigma \pm (1-\sigma^2)^{\frac{1}{2}}/\sigma$. The function β can be evaluated for any particular value of the Fourier space coordinates, Θ and K, and the diffusivity D by two simultaneous numerical quadratures.

To complete the theoretical description, we must adopt a self-consistent approximation for the effective diffusivity D. The diffusivity tensor is given by a time integral of the correlation function for the particle's velocity $\boldsymbol{U}$, i.e.

$$\boldsymbol{D} = \int_0^\infty \mathrm{d}\tau\,\langle \boldsymbol{U}'(t+\tau)\,\boldsymbol{U}'(t)\rangle, \tag{3.33}$$

where $\boldsymbol{U}' \equiv \boldsymbol{U} - \boldsymbol{U}^{\mathrm{s}}$. The particle velocity fluctuation $\boldsymbol{U}'$ results from the velocity disturbances caused by the surrounding particles, i.e. $\langle \boldsymbol{u}\rangle_1\,(\boldsymbol{x}\,|\,\boldsymbol{r}_1)$. The test particle at $\boldsymbol{x}$ samples variations in the velocity disturbance caused by a second particle at $\boldsymbol{r}_1$, due to the relative motion induced by the surrounding particles. In keeping with our simplified description of this process we shall treat this sampling as if the two particles diffuse independently with a local isotropic diffusivity. An approximation to the diffusivity that neglects non-local and anisotropic effects as well as the correlations between particle positions and velocities is

$$3D = n\int \mathrm{d}\boldsymbol{r}_1\int \mathrm{d}\boldsymbol{x}'\int_0^\infty d\tau\,P(\boldsymbol{x},t+\tau\,|\,\boldsymbol{x}',t)\,\langle \boldsymbol{u}\rangle_1(\boldsymbol{x}\,|\,\boldsymbol{r}_1)\cdot\langle \boldsymbol{u}\rangle_1(\boldsymbol{x}'\,|\,\boldsymbol{r}_1), \tag{3.34}$$

where the transition probability $P(\boldsymbol{x},t+\tau\,|\,\boldsymbol{x}',t)$ for the relative position of the two particles satisfies the equation

$$\frac{\partial P}{\partial \tau} - 2D\nabla^2 P = \delta(\boldsymbol{x}-\boldsymbol{x}')\,\delta(\tau). \tag{3.35}$$

Solving (3.35) after integrating with respect to τ and Fourier transforming with respect to $\boldsymbol{x}-\boldsymbol{x}'$, substituting the result into (3.34), and using the product and convolution theorems gives

$$D = \frac{n}{3}\int \mathrm{d}\boldsymbol{k}\,\frac{\langle \boldsymbol{u}\rangle_1(\boldsymbol{k})N\langle \boldsymbol{u}\rangle_1(-\boldsymbol{k})}{2D(2\pi k)^2}. \tag{3.36}$$

An examination of (3.28) indicates that the real part of β (denoted β_{R}) is an even function of $\boldsymbol{k}$, while the imaginary part of β (denoted β_I) is odd. As a result, the integrand in (3.36) is real. Integrating (3.36) over the meriodonal angle in the spherical coordinate system in Fourier space gives

$$D^2 = \frac{9(U^s a\phi^{-1})^2}{4\pi}\int_0^\infty \mathrm{d}K \int_0^1 \mathrm{d}x \frac{(1-x^2)}{(K^2+\beta_R)^2+\beta_I^2}, \tag{3.37}$$

where $K \equiv 2\pi k a\phi^{-1}$ and $x \equiv \cos\Theta$. The integrand in (3.37) has an integrable singularity at $K = 0$ and $x = 1$. It was possible to integrate the singular part of the integral in (3.37) with respect to K analytically by contour integration in the complex plane. This integration yielded an integral in x that was also singular. Integrating the singular part of the latter integral analytically, we obtained

$$D^2 = (U^s a\phi^{-1})^2(d_2+d_{11}+d_{12}), \tag{3.38a}$$

where

$$d_2 \equiv \frac{9}{4\pi}\int_0^\infty \mathrm{d}K \int_0^1 \mathrm{d}x\,(1-x^2)\frac{\beta_{I0}^2-\beta_I^2+\beta_{R0}^2-\beta_R^2+2K^2(\beta_{R0}-\beta_R)}{((K^2+\beta_R)^2+\beta_I^2)((K^2+\beta_{R0})^2+\beta_{I0}^2)} \tag{3.38b}$$

$$d_{11} \equiv 9\pi/(32\alpha_1^{\frac{3}{2}}), \tag{3.38c}$$

$$d_1 \equiv \frac{9}{8\alpha_1}\int_0^1 \mathrm{d}x \frac{2\beta_{R0}^{\frac{1}{2}}-(\gamma^2+4\beta_{R0})^{\frac{1}{2}}}{2\beta_{R0}^{\frac{1}{2}}(\gamma^2+4\beta_{R0}^{\frac{1}{2}})}, \tag{3.38d}$$

and where β_{R0} and β_{I0}, the asymptotic behaviour of the real and imaginary parts of β respectively in the dual limit $K \to 0$ and $x \to 1$, are given by

$$\beta_{R0} = \alpha_1(1-x^2), \quad \beta_{I0} = \gamma K = K\alpha_2 D(1-x^2). \tag{3.38e,f}$$

The constants, $\alpha_1 = 11.3$ and $\alpha_2 = 159$, were determined by an analytic integration with respect to Θ' and a numerical integration with respect to r of the asymptotic behaviour of β obtained from (3.32). Note that (3.38) is an implicit equation for D, since β depends on D. Thus, to determine D, we performed a numerical quadrature of (3.38*b*), which in turn required a numerical quadrature of (3.32) to determine β at each step. We also performed the numerical integration of (3.38*d*). We then solved (3.38) iteratively repeating the numerical integrations at each step. This gave the result

$$D = 0.52\, U^s a\phi^{-1}. \tag{3.39}$$

Thereafter, we completed the numerical integration of (3.30) with (3.32) and found the variance of the velocity to be

$$\langle u^2\rangle = 4.7\, U^{s2}. \tag{3.40}$$

Note that the variance is $O(U^{s2})$ even though the volume fraction of particles is very small. This results from the long range of the velocity disturbance caused by each particle. The velocity at a given point in space is affected by the $O(\phi^{-2})$ particles within a screening length of that point – each particle making an $O(U^{s2}a^2/x^2) = O(U^{s2}\phi^2)$ contribution to the variance.

3.2. *Short-range structure*

In addition to the long-range structure investigated in the preceding section, three-particle interactions also cause a short-range structure to the pair probability on an $O(a)$ lengthscale. Although the long-range structure has a profound influence on the suspension properties, it would be difficult to observe directly because it represents only an $O(n\phi)$ change in the pair probability. The short-range structure of the pair probability represents an $O(n)$ change from its random value, so it may be more easily observed.

As a close pair of particles sediments, it encounters many isolated particles whose velocity disturbances lead to a relative motion of the pair. It will be assumed subject to *a posteriori* justification that the largest effect on the close pair probability comes from third particles that are a distance large compared to a from the pair.

In order to derive an equation for the evolution of the probability density $n^2\Omega(\boldsymbol{r}_1-\boldsymbol{r}_2)$ for finding two particles at positions $\boldsymbol{r}_1$ and $\boldsymbol{r}_2$ separated by a distance $|\boldsymbol{r}_1-\boldsymbol{r}_2| = O(a)$, we start from the conservation equation (3.1) for the N-particle probability density. An estimate for the N-particle probability density for cases where $|\boldsymbol{r}_1-\boldsymbol{r}_2| = O(a)$, which takes account of the effects of interactions of the close pair with distant third particles is, cf. (3.10),

$$P_N = n^2\Omega(\boldsymbol{r}_1-\boldsymbol{r}_2)P_N(\boldsymbol{C}_N\,|\,\boldsymbol{r}_1,\boldsymbol{r}_2)+\sum_{i=3}^{N} n^3m'(\boldsymbol{r}_i-\boldsymbol{r}_1,\boldsymbol{r}_1-\boldsymbol{r}_2)P_N(\boldsymbol{C}_N\,|\,\boldsymbol{r}_1,\boldsymbol{r}_2,\boldsymbol{r}_i). \quad (3.41)$$

Substituting (3.41) into (3.1), integrating over the coordinates $\boldsymbol{x}_3$ to $\boldsymbol{x}_N$, and applying the divergence theorem and the condition of no particle flux through the boundaries of the volume V gives

$$\frac{\partial\Omega(\boldsymbol{r})}{\partial t}+\boldsymbol{\nabla}_r\cdot\int \mathrm{d}\boldsymbol{x}\,\mathring{\boldsymbol{r}}(\boldsymbol{x},\boldsymbol{r})(\Omega(\boldsymbol{r})+nm'(\boldsymbol{x},\boldsymbol{r})) = 0. \quad (3.42)$$

The analysis in this subsection of the factors controlling the relative position of a close pair in a sedimenting suspensions bears significant similarities to the analysis in Shaqfeh & Koch (1988) of the orientation of non-spherical particles flowing through a fixed bed. Here the relative position $\boldsymbol{r}$ plays a role analogous to the particle orientation in the aforementioned analysis. It was shown in Shaqfeh & Koch (1988) that the effects of the velocity fluctuations in a fixed bed on the particle orientation could be expressed in terms of an effective rotary diffusivity and a rotational drift velocity. Similarly, it will now be seen that the effect of the velocity disturbance caused by distant particles in a sedimenting suspension is to give rise to an effective relative diffusion tensor and a relative drift velocity of a close pair of sedimenting particles.

The first term in the integrand in (3.42) integrates to zero because the relative velocity of the close pair is an odd function of the separation from the third particle. In other words, the mean relative velocity of the pair due to all the other particles in the suspension is zero. This is a consequence of the absence of an average shear flow in the suspension.

It will be seen that the largest contribution to the flux in (3.42) arises from distant third particles, i.e. those for which $x \gg a$. Equation (3.16) may be solved to give an expression for m' valid when $x \gg a$,

$$m'(\boldsymbol{x},\boldsymbol{r}) = \frac{1}{U^r}\int_{-\infty}^{\eta} \mathrm{d}\eta'\,\boldsymbol{\nabla}_r\cdot[\mathring{\boldsymbol{r}}(\boldsymbol{x}',\boldsymbol{r})\,\Omega(\boldsymbol{r})], \quad (3.43)$$

where η is a coordinate measured along the direction parallel to the velocity $\boldsymbol{U}^r$ of the pair relative to the third particle. Equation (3.43) is simply the inverse Fourier transform of (3.17).

Substituting (3.43) into (3.42), the equation for the pair probability is

$$\frac{\partial \Omega}{\partial t}+\nabla_r\cdot[\mathring{r}^{\mathrm{h}}\Omega-\boldsymbol{d}^{\mathrm{h}}\cdot\nabla_r\Omega]=0, \tag{3.44a}$$

where the effective relative diffusivity $\boldsymbol{d}^{\mathrm{h}}$ and the relative drift velocity $\mathring{\boldsymbol{r}}^{\mathrm{h}}$ for the pair are given by

$$\boldsymbol{d}^{\mathrm{h}}\equiv\frac{n}{U^{\mathrm{r}}}\int \mathrm{d}\boldsymbol{x}\,\mathring{\boldsymbol{r}}(\boldsymbol{x},\boldsymbol{r})\int_{-\infty}^{\eta}\mathrm{d}\eta'\,\mathring{\boldsymbol{r}}(\boldsymbol{x}',\boldsymbol{r}), \tag{3.44b}$$

and

$$\mathring{\boldsymbol{r}}^{\mathrm{h}}\equiv\frac{n}{U^{\mathrm{r}}}\int \mathrm{d}\boldsymbol{x}\,\mathring{\boldsymbol{r}}(\boldsymbol{x},\boldsymbol{r})\int_{-\infty}^{\eta}\mathrm{d}\eta'\,\nabla_r\cdot[\mathring{\boldsymbol{r}}(\boldsymbol{x}',\boldsymbol{r})]. \tag{3.44c}$$

Equation (3.44*b*) indicates that the effective relative diffusion tensor for the pair is given by the integral of the relative velocity correlation function along the pair's trajectory. The drift velocity in (3.44*c*) is related to the correlation of the pair's relative velocity with the dilatation of the relative velocity experienced along the trajectory.

Substituting (3.4) and (3.5) for the relative velocity and its divergence, (3.44*b, c*) may be written as

$$\mathring{r}_i^{\mathrm{h}}=A_{jkmn}\,W\frac{r_m r_n}{r^2}\left[(\tfrac{1}{2}B-1)\,\delta_{ij}r_k+\tfrac{1}{2}B\delta_{ik}\,r_j+(A-B)\frac{r_i r_j r_k}{r^2}\right], \tag{3.45a}$$

and

$$d_{il}^{h}=A_{jkmn}\left[(1-\tfrac{1}{2}B)\,\delta_{ij}r_k+(B-A)\frac{r_i r_j r_k}{r^2}-\tfrac{1}{2}B\delta_{ik}\,r_j\right]$$
$$\times\left[(1-\tfrac{1}{2}B)\,\delta_{lm}r_n+(B-A)\frac{r_l r_m r_n}{r^2}-\tfrac{1}{2}B\delta_{ln}\,r_m\right], \tag{3.45b}$$

where

$$A_{jkmn}\equiv\frac{n}{U^{\mathrm{r}}}\int \mathrm{d}\boldsymbol{x}\,\frac{\partial u_j}{\partial x_k}(\boldsymbol{x})\int_{-\infty}^{\eta}\mathrm{d}\eta'\,\frac{\partial u_m}{\partial x_n}(\boldsymbol{x}'). \tag{3.45c}$$

The fourth-order tensor A_{jkmn} is the integral over the pair's trajectory of the self-correlation of the gradient of the velocity disturbance caused by the third particle. A similar velocity gradient correlation function was obtained by Shaqfeh & Koch (1988) in a study of particle orientation in flows through fixed beds. The A_{jkmn} obtained here differs slightly from that of Shaqfeh & Koch (1988), because here the integral is carried out along a trajectory which depends on the pair's relative position.

The velocity gradient correlation has the symmetry $A_{jkmn}=A_{mnjk}$. This symmetry may be noted by using the convolution theorem to rewrite (3.45*c*) in terms of an integral in Fourier space, cf. equation (39) of Shaqfeh & Koch (1988). Using the symmetry $A_{jkmn}=A_{mnjk}$, (3.45*c*) may be integrated by parts to give

$$A_{jkmn}=\frac{n}{2U^{\mathrm{r}}}\int \mathrm{d}\boldsymbol{\rho}\left[\int_{-\infty}^{\infty}\mathrm{d}\eta\,\frac{\partial u_j}{\partial x_k}\right]\left[\int_{-\infty}^{\infty}\mathrm{d}\eta\,\frac{\partial u_m}{\partial x_n}\right], \tag{3.46}$$

where $\boldsymbol{\rho}$ is the two-dimensional position vector in the plane perpendicular to the relative velocity $\boldsymbol{U}^{\mathrm{r}}$. Applying the convolution theorem, (3.46) becomes

$$A_{jkmn}=\frac{n}{2U^{\mathrm{r}}}\int \mathrm{d}\xi\,\widehat{\frac{\partial u_j}{\partial x_k}}(\xi)\,\widehat{\frac{\partial u_m}{\partial x_n}}(-\xi), \tag{3.47}$$

where ξ is the transform variable in the plane perpendicular to U^r, i.e. $\xi_i \equiv (\delta_{ij} - U_i^r U_j^r / U^{r2}) k_j$. Substituting (3.7) for the velocity disturbance in (3.47) one obtains, after some rearrangement,

$$A_{jkmn} = \frac{27\phi U^{s2}}{4\pi a U^r} \int_{\phi a^{-1}}^{a^{-1}} \frac{d\xi}{\xi} \int_0^{2\pi} d\Phi \, \bar{\xi}_k (\delta_{jp} - \bar{\xi}_j \bar{\xi}_p) \delta_{p3} \bar{\xi}_n (\delta_{mq} - \bar{\xi}_m \bar{\xi}_q) \delta_{q3}, \qquad (3.48)$$

where $\bar{\xi}_k \equiv \xi_k / \xi$, Φ is the angle between ξ_k and an arbitrary vector in the plane perpendicular to U^r. The integral with respect to ξ in (3.48) should range over the entire plane, i.e. $0 \leqslant \xi < \infty$. However, this integral is conditionally convergent in the limits $\xi \to 0$ and $\xi \to \infty$. It should be recalled that the approximate equation (3.16) used to find the three-particle probability, and the approximate expression (3.7) for the relative velocity are only valid at radial separations $a \ll x \ll a\phi^{-1}$, corresponding to wavenumbers in the range $\phi a^{-1} \ll \xi \ll a^{-1}$. The conditional convergence in the limit $\xi \to \infty$ $(x \to 0)$ results from our approximation of the velocity disturbance caused by the third particle as a Stokeslet. The actual velocity gradients in (3.47) are well behaved at distances $x = O(a)$, and the contributions to A_{jkmn} from this region are $O(\phi U^s / a)$. The conditional convergence in the limit $\xi \to 0 (x \to \infty)$ arises because we have used the unscreened velocity disturbance (3.7) caused by the third particle. In §3.1 it was seen that the velocity disturbance caused by a particle is affected by the deficit of surrounding particles at a distance $x = O(a\phi^{-1})$. Provided that this deficit results in a smooth transition from the $O(U^s a x^{-1})$ decay of the velocity disturbance with radial distance when $x \ll a\phi^{-1}$ to a more rapid decay when $x \gg a\phi^{-1}$, the contributions to A_{jkmn} arising from distances $x \geqslant O(a\phi^{-1})$ will be $O(\phi U^s / a)$. The dominant $O[\phi U^s (\ln \phi^{-1}) / a]$ contribution to A_{jkmn} will then arise from intermediate radial distances. This contribution is given by (3.48).

Expressions for the drift velocity and the effective diffusivity tensor may be obtained by integrating (3.48). However, the analysis is very complicated. Here, in order to obtain a qualitative picture of the effects of three-particle interactions on the pair probability we shall make the following simplification. We shall consider cases in which the velocity of the pair relative to the third particle is in the vertical direction. This situation arises in a suspension of sedimenting particles when the pair is oriented either parallel or perpendicular to gravity. The approximation that the relative velocity is in the vertical direction is expected to give the correct qualitative behaviour for a pair of sedimenting spheres, since their velocity relative to a third particle is never more than about 18° from the direction of the gravitational acceleration.

When the relative velocity is parallel to the gravitational acceleration the integral in (3.48) may be performed to give, cf. (45) of Shaqfeh & Koch (1988),

$$A_{jkmn} = \frac{27\phi U^{s2}}{4U^r} \ln \phi^{-1} (\delta_{kn} - \delta_{k3} \delta_{n3}) \delta_{j3} \delta_{m3}. \qquad (3.49)$$

Substituting (3.49) into the expression (3.45a) for the drift velocity and evaluating the r- and Θ-components, one obtains

$$\dot{r}_r^{\mathrm{h}} = -\frac{27\phi U^{s2}}{4U^r} \ln \phi^{-1} \, W(1-A) \, r \cos^2 \Theta \sin^2 \Theta, \qquad (3.50a)$$

$$\dot{r}_\Theta^{\mathrm{h}} = \frac{27\phi U^{s2}}{4U^r} \ln \phi^{-1} \, W r \cos \Theta \sin \Theta [1 - \tfrac{1}{2}B + (B-1) \cos^2 \Theta]. \qquad (3.50b)$$

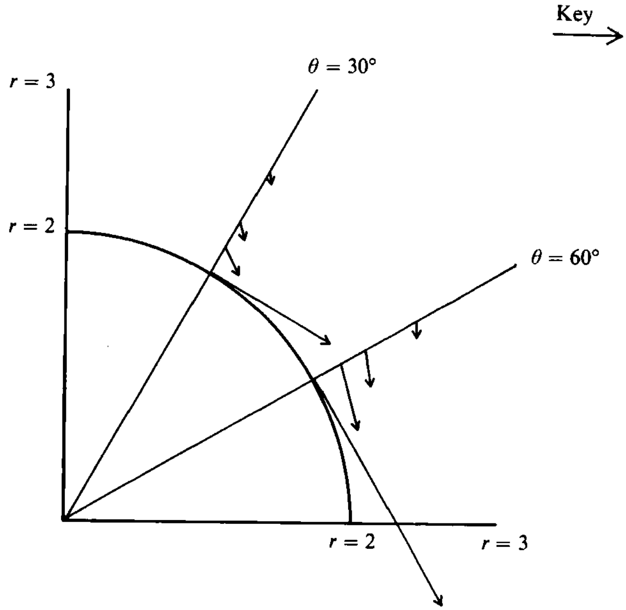

FIGURE 3. The drift velocity vector $\dot{\boldsymbol{r}}^{\mathrm{h}}$ is plotted for various relative positions $\boldsymbol{r}$. The scale is indicated by the arrow at the upper right of the figure which denotes a vector of magnitude $27\phi U^{s2}a \ln \phi^{-1}/4U^{r}$. Here, r is non-dimensionalized with a.

The drift velocity vector as a function of relative position is illustrated in figure 3. The drift velocity goes to zero at vertical and horizontal relative positions. Since A is less than 1 for all values of r, the radial component of the drift is always inward. B takes on values between 0 and 0.4060 (Batchelor & Green 1972), so the angular component of the drift is in the positive (negative) Θ-direction when the angle Θ between the separation vector and the gravitational acceleration is less (greater) than $\frac{1}{2}\pi$. Thus, the radial component of the drift velocity favours close pairs and the angular component tends to form horizontal pairs.

The relative diffusion tensor obtained by substituting (3.49) into (3.45*b*) is

$$\begin{aligned} d_{il}^{\mathrm{h}} = \frac{27\phi U^{s2}}{4U^{r}}(\ln \phi^{-1})\, W\{r^2 \delta_{i3}\delta_{l3}[(1-\tfrac{1}{2}B)^2(1-\alpha^2)-\tfrac{1}{4}B^2\alpha^2+B(1-\tfrac{1}{2}B)\alpha^2] \\ +\tfrac{1}{4}B^2\alpha^2 r^2 \delta_{il} + r_i r_l[(B-A)^2\alpha^2(1-\alpha^2)-B(B-A)\alpha^2] \\ +(rr_i\delta_{l3}+\delta_{i3}rr_l)[(1-\tfrac{1}{2}B)(B-A)(\alpha-\alpha^3)-\tfrac{1}{2}B(1-\tfrac{1}{2}B)\alpha+\tfrac{1}{2}B(B-A)\alpha^3]\}, \quad (3.51) \end{aligned}$$

where $\alpha \equiv \cos\Theta$. The relative diffusion tensor grows like r^3 as $r/a \rightarrow \infty$, because the relative velocity between the two particles constituting the pair grows like r and the velocity of the pair relative to the third particle decays like r^{-1}. It should be recalled, however, that the description adopted for the three-particle interactions here is only valid when the close pair are separated by an $O(a)$ distance. At larger separations, the relative velocity between the particles will be different from the values given by (3.3) and (3.4). For $r \gg R_s$, the pair diffusivity will asymptote to twice the single-particle tracer diffusivity, whose magnitude is estimated in (3.39).

Note that the drift velocity is non-zero only for small radial separations at which the relative velocity is not solenoidal. Because the drift velocity involves the correlation of the $O(r)$ relative velocity with the $O(r^{-6})$ divergence of the velocity weighted by the $O(r^{-1})$ inverse of the relative velocity, $(U^{r})^{-1}$, it decays like r^{-6} as

$r \to \infty$. The term involving the drift velocity in (3.44a) then acts like a compact source quadrupole of pairs, particle conservation precluding the presence of a source and the evenness of $\Omega(\boldsymbol{r})$ precluding the presence of a dipole of particles. This quadrupole leads to a flux $-\boldsymbol{d}^{\mathrm{h}} \cdot \nabla_r \Omega$ that decays like r^{-4}. Since the diffusivity grows like r^3, the pair probability must decay like r^{-6} as $r \to \infty$. This confirms our *a priori* assumption that the $O(n)$ 'short-range' structure of the pair probability decays more rapidly than r^{-3} and thus cannot give rise to screening of the velocity disturbance (cf. (2.14)).

The assumptions made concerning the pair probability by Caflisch & Luke (1986) are at variance with the preceding observation. They assumed that the pair probability had an $O(n)$ structure that extended to large separations $r \gg a$, and that this structure could be studied using the point-particle approximation. It was noted above, however, that a random suspension with no structure in the pair probability is a solution of the particle conservation equation (3.1) in the point-particle approximation. Note also that Caflisch & Luke (1986) suggested that there is an excess of vertical pairs, while our calculations indicate an excess of horizontal pairs.

4. Conclusions

In this paper we have proposed a mechanism by which the velocity disturbances caused by sedimenting particles in suspension may be screened. In §2 we considered in detail the calculation of the variance of the fluid velocity in a sedimenting suspension. In examining the convergence properties of the velocity variance, it was sufficient to use a point-particle approximation, because this approximation captured the long-range velocity disturbances. It was shown that the velocity variance is finite (i.e. independent of the size of the vessel) if the pair probability exhibits a net deficit of one particle in a compact region of space surrounding any given particle, i.e. if it satisfies (2.12). If the pair probability does not satisfy (2.12), then the variance of the velocity is divergent in the sense that it grows linearly with the linear size of the settling vessel. Thus, in a suspension of particles with no inertia the variance can only be independent of the size of the settling vessel if (2.12) is satisfied. The results in §2 for the effect of structure on the variance of the fluid velocity apply to sedimenting particles of any shape, because the leading contribution to the variance depends only on the magnitude of the force of gravity acting on the particle.

Because it was not immediately obvious whether a structure that satisfies (2.12) could develop in a sedimenting suspension, we studied the structure in a monodisperse sedimenting suspension of non-Brownian spheres in §3. It was determined that the pair probability in a sedimenting suspension of spherical particles consists of two contributions. (i) An $O(n)$ contribution, which goes to a constant within an $O(a)$ radial distance. In §3.2, it was shown that close pairs with their relative position perpendicular to gravity were favoured over close pairs whose relative position is parallel to gravity. (ii) In §3.1 three-particle interactions were shown to result in an $O(n\phi)$ contribution to the pair probability. Despite its small magnitude this contribution plays a crucial role in determining the macroscopic properties of the suspension. This contribution to the pair probability consists of a deficit of particles that decays very slowly (as ax^{-1}) with radial position. This long-range deficit of neighbouring particles leads to a screening of a particle's velocity disturbance in a sedimenting suspension in a manner analogous to the Debye screening of an ion's electrical potential in an ionic solution by the surrounding ion cloud. Because of its small magnitude the particle deficit can lead to Debye-like screening of the velocity disturbance only at a large $O(a\phi^{-1})$ radial distance.

The Debye-like screening of the velocity disturbance caused by a sedimenting particle has important effects on the properties of the suspension, many of which would depend on the size of the suspension in the absence of screening. The detailed velocity disturbance and pair probability at $O(a\phi^{-1})$ radial separations have only been calculated through a self-consistent approximation. Nonetheless it is possible to make predictions concerning the orders of magnitude of the suspension properties based on a knowledge of the screening length, $a\phi^{-1}$, alone. The simplest measure of the fluctuations in the velocities of the particles and fluid in a suspension are the variances of the sedimentation velocity and the fluid velocity, respectively. The calculation of these velocity variances involves summing the squares of the velocity disturbances of all the particles in a suspension. Since the velocity disturbance decays as $U^{s} a x^{-1}$, this leads to a volume integral of a quantity that decays like $\phi U^{s2} a^2 x^{-2}$, cf. (2.8). In general, the order of magnitude of the velocity variances will be $U^{s2}\phi R_s/a$, where R_s is the radial distance to which the velocity disturbance caused by a particle propagates. In a random suspension whose pair probability does not satisfy (2.12), the integral for the velocity variance only converges when one reaches the vessel walls, so R_s is the minimum dimension of the settling vessel L and the variances are $O(\phi U^{s2} L/a)$, as predicted for a suspension of uniform probability by Caflisch & Luke (1985). It has been seen that a suspension of spherical particles develops a structure that can lead to screening of the velocity disturbance at an $O(a\phi^{-1})$ radial distance, leading to $O(U^{s2})$ velocity variances.

Attempts to calculate the effective diffusivities of either the sedimenting particles or chemical tracers in the fluid phase also lead to divergent integrals. These diffusivities are given by time integrals of the particles' velocity correlation functions. The order of magnitude of the effective diffusivities may be estimated as the product of the variance of the velocity and the correlation time t_c, i.e. the time over which a solid or fluid particle's velocity correlation function decays. The fluid velocity will remain correlated for the $O(R_s/U^s)$ time that it takes a particle to fall through the interaction volume of radius R_s surrounding the fluid particle. Thus, the effective diffusivity for a chemical tracer in the fluid phase is $O(\langle u^2\rangle R_s/U^s)$ or $O(U^s a\phi^{-1})$.

To estimate the order of magnitude of the effective tracer diffusivity of a solid particle, D, we first note that the variance of the solid-particles' velocity (like the variance of the fluid velocity) is $O(U^{s2})$. However, the process by which a solid-particle's velocity becomes uncorrelated is different from the process by which a fluid-particle's velocity becomes uncorrelated. In the absence of hydrodynamic interactions all of the particles would settle at the same speed. Thus, it is necessary to take into account the effects of hydrodynamic interactions with third particles on the relative motion of two particles in order to see how they move away from and stop influencing one another. To estimate the order of magnitude of the correlation time, we treat the effect of these interactions with third particles as an effective diffusion and estimate the relative diffusion coefficient as being of the same order of magnitude as the solid-particle tracer diffusivity D. The correlation time is then $t_c = O(R_s^2/D)$. Solving for the correlation time self-consistently with $D = O(\langle U^2\rangle t_c)$, we obtain $D = O(\langle U^2\rangle^{\frac{1}{2}} R_s) = O(U^s a\phi^{-1})$. Thus, both the fluid-phase and solid-particle tracer diffusivities are predicted to grow like $U^s a\phi^{-1}$ as $\phi \rightarrow 0$.

This predicted scaling of the effective diffusivities only holds when the screening length $a\phi^{-1}$ is much smaller than the minimum linear dimension of the settling vessel L. If the settling vessel is tall and narrow, then a particle's velocity may become uncorrelated before it settles out, even when the sidewalls limit the range of the particle interactions. In this case, a diffusive behaviour may still be expected. When the vessel sidewalls are limiting, the effective diffusivity scales like $D = O(\langle U^2 \rangle t_c)$, where $\langle U^2 \rangle = O(U^{s2}\phi L/a)$ and the correlation time is $t_c = O(L/U^s)$ for the fluid-phase tracer and $t_c = O(L^2/D)$ for the solid particles. Thus, the effective diffusivity for a fluid-phase tracer is $O(U^s \phi L^2/a)$ and the solid-particle effective diffusivity is $O(U^s \phi^{\frac{1}{2}} L^{\frac{3}{2}}/a^{\frac{1}{2}})$. In both cases, the effective diffusivity goes to zero if one takes the limit $\phi \rightarrow 0$ with the vessel dimension L held fixed.

Unfortunately, there are no experimental measurements of the variance of either the fluid or particle velocities in a sedimenting suspension. Ham & Homsy (1988) have obtained experimental values for the effective solid-particle tracer diffusivity in a nearly monodisperse sedimenting suspension of spherical particles. The measurement was taken by following the motion of a single marked particle in the suspension. The effective diffusivity was defined in terms of the time rate of change of the particle's mean-square displacement. A direct comparison of the present theory to these measurements is made uncertain by two factors: (i) the volume fraction range 0.025–0.1 in the experiments may not be sufficiently dilute to exhibit the asymptotic behaviour predicted here; and (ii) the 90% confidence limits on the experimentally measured values of the diffusivity indicated that the diffusivity was only known to within about a factor of two. The experimentally measured effective diffusivity increased with decreasing volume fraction as the volume fraction was varied from 0.1 to 0.08 and 0.05, in qualitative agreement with our prediction $D = O(U^s a \phi^{-1})$.

When the volume fraction was further decreased to 0.025, the experimental value for the effective diffusivity decreased. A possible explanation for this decrease is that the settling vessel was not sufficiently large to see the asymptotic value of the diffusivity at this lower volume fraction. The observed decrease in the effective diffusivity with a decrease in the volume fraction for 0.05 to 0.025 is qualitatively consistent with the prediction above for the behaviour of a suspension in which the vessel sidewalls limit the range of the particle interactions. The ratio of the column radius to the particle radius in the experiments was $L/a = 100$; this was not varied significantly in the experiments. The maximum in the experimentally observed diffusivity came at a volume fraction for which the length $a\phi^{-1}$ was smaller than, but of the same order of magnitude as, the vessel radius. Our scaling arguments are only sufficient to predict that the maximum in the diffusivity should occur when L/a is of the same order of magnitude as ϕ^{-1}. A further piece of evidence that would suggest that Ham & Homsy's lowest volume fraction results were influenced by the finite lateral dimensions of the vessel is the fact that the distance (denoted H_∞ by the authors) for the particle's velocity to lose correlation with its initial value was comparable with the vessel diameter.

It has been noted that Batchelor's (1972) calculation of the first correction $-6.55\phi U^s$ to the mean sedimentation velocity can be expected to apply to a suspension of Brownian particles that possess the uniform pair probability stipulated in his calculation but not to a suspension of non-Brownian particles that possess a non-uniform structure. The $O(n)$ short-range structure of the pair probability consisting of an excess of close pairs may be expected to lead to an $O(\phi U^s)$ increase of the average sedimentation velocity over that calculated by Batchelor (1972). The effect of the long-range structure on the average sedimentation velocity may be determined from (2.5). The long-range contribution to the difference between the pair probability and the number density, i.e. $g - n$, is $O(n\phi a r'^{-1})$ for $a \ll r' \ll a\phi^{-1}$ (cf. the discussion at the beginning of §3.1). For r' larger than $a\phi^{-1}$, $g - n$ decays more rapidly owing to the screening of velocity and density perturbations. Thus, the integral in (2.5) grows like $n\phi U^{\mathrm{s}}Ra$ with the radial extent R to which the integration is carried out until R exceeds $a\phi^{-1}$. The integral then converges to an $O(U^{\mathrm{s}}\phi)$ value. The long-range deficit, then, is expected to give rise to an $O(\phi U^{\mathrm{s}})$ decrease in the mean sedimentation velocity. Thus, we predict that the first correction to the average settling velocity caused by hydrodynamic interactions in a dilute suspension of non-Brownian, sedimenting spheres is $O(\phi U^{\mathrm{s}})$ as calculated by Batchelor but with a different numerical coefficient. It is interesting to note that, if Debye-like screening occurred at a smaller radial distance, for example at a distance $a\phi^{-\alpha}$ with $0 < \alpha < 1$, this first correction would be larger, $O(\phi^{\alpha}U^{\mathrm{s}})$.

An unfortunate consequence of the long-range nature of the interparticle interactions in a dilute sedimenting suspension is that it would be exceedingly difficult to accurately numerically simulate such a suspension. Since the interactions between particles separated by a distance up to the Debye-like screening length are important, one would require a number of particles that is large compared to ϕ^{-2} to perform an accurate numerical simulation of the dynamics of a three-dimensional suspension.

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Appendix: Justification for the neglect of higher-order velocity correlations in the expression (2.8) for the velocity variance

The expression (2.8) used to approximate the variance of the fluid velocity is the first term in an asymptotic expansion in the dilute limit for the velocity variance. This first term involves the effects of individual particles on the velocity variance, while the kth term involves effects of groups of k particles. To produce this expansion we can expand the detailed fluid velocity as a sum of the effects of groups of one, two, etc. particles, i.e.

$$\boldsymbol{u}(\boldsymbol{x}\,|\,C_N) = \sum_{i=1}^{N} \langle \boldsymbol{u}\rangle_1(\boldsymbol{x}\,|\,\boldsymbol{r}_i) + \sum_{i=1}^{N}\sum_{j>i} \langle \boldsymbol{u}'\rangle_2(\boldsymbol{x}\,|\,\boldsymbol{r}_i,\boldsymbol{r}_j) + \sum_{i=1}^{N}\sum_{j>i}\sum_{k>j} \langle \boldsymbol{u}''\rangle_3(\boldsymbol{x}\,|\,\boldsymbol{r}_i,\boldsymbol{r}_j,\boldsymbol{r}_k) + \ldots + \langle \boldsymbol{u}^{N-1}\rangle_N(\boldsymbol{x}\,|\,C_N). \quad \text{(A 1)}$$

Each term in (A 1) represents only the additional effects of a group of k particles that cannot be expressed in terms of $k-1$ particle correlations. Thus,

$$\langle \boldsymbol{u}'\rangle_2(\boldsymbol{x}\,|\,\boldsymbol{r}_i,\boldsymbol{r}_j) \equiv \langle \boldsymbol{u}\rangle_2(\boldsymbol{x}\,|\,\boldsymbol{r}_i,\boldsymbol{r}_j) - \langle \boldsymbol{u}\rangle_1(\boldsymbol{x}\,|\,\boldsymbol{r}_i) - \langle \boldsymbol{u}\rangle_1(\boldsymbol{x}\,|\,\boldsymbol{r}_j), \quad \text{(A 2a)}$$

$$\langle \boldsymbol{u}''\rangle_3(\boldsymbol{x}\,|\,\boldsymbol{r}_i,\boldsymbol{r}_j,\boldsymbol{r}_k) \equiv \langle \boldsymbol{u}\rangle_3(\boldsymbol{x}\,|\,\boldsymbol{r}_i,\boldsymbol{r}_j,\boldsymbol{r}_k) - \langle \boldsymbol{u}'\rangle_2(\boldsymbol{x}\,|\,\boldsymbol{r}_i,\boldsymbol{r}_j) - \langle \boldsymbol{u}'\rangle_2(\boldsymbol{x}\,|\,\boldsymbol{r}_i,\boldsymbol{r}_k) - \langle \boldsymbol{u}'\rangle_2(\boldsymbol{x}\,|\,\boldsymbol{r}_k,\boldsymbol{r}_j) - \langle \boldsymbol{u}\rangle_1(\boldsymbol{x}\,|\,\boldsymbol{r}_i) - \langle \boldsymbol{u}\rangle_1(\boldsymbol{x}\,|\,\boldsymbol{r}_j) - \langle \boldsymbol{u}\rangle_1(\boldsymbol{x}\,|\,\boldsymbol{r}_k), \quad \text{(A 2b)}$$

etc. Equation (2.8) is the result obtained for the variance if the expansion (A 1) is substituted for the first factor of the velocity $\boldsymbol{u}(\boldsymbol{x}\,|\,C_N)$ in (2.7) and only terms involving the conditional average with one particle fixed are retained. If we substitute (A 1) for the first factor of the velocity in (2.7) and truncate the expansion (A 2) at the second term, we obtain

$$\langle u^2\rangle \approx \int \mathrm{d}\boldsymbol{r}_1\, n\,\langle \boldsymbol{u}\rangle_1(\boldsymbol{x}\,|\,\boldsymbol{r}_1)\cdot\langle \boldsymbol{u}\rangle_1(\boldsymbol{x}\,|\,\boldsymbol{r}_1)$$

$$+\int \mathrm{d}\boldsymbol{r}_1\,\mathrm{d}\boldsymbol{r}_2\, ng(\boldsymbol{r}_1\,|\,\boldsymbol{r}_2)\,\langle \boldsymbol{u}\rangle_1(\boldsymbol{x}\,|\,\boldsymbol{r}_2)\cdot\langle \boldsymbol{u}'\rangle_2(\boldsymbol{x}\,|\,\boldsymbol{r}_1,\boldsymbol{r}_2)$$

$$+\int \mathrm{d}\boldsymbol{r}_1\,\mathrm{d}\boldsymbol{r}_2\,\tfrac{1}{2}ng(\boldsymbol{r}_1\,|\,\boldsymbol{r}_2)\,\langle \boldsymbol{u}'\rangle_2(\boldsymbol{x}\,|\,\boldsymbol{r}_1,\boldsymbol{r}_2)\cdot\langle \boldsymbol{u}'\rangle_2(\boldsymbol{x}\,|\,\boldsymbol{r}_1,\boldsymbol{r}_2). \qquad \text{(A 3)}$$

Integrating the second term over the coordinate $\boldsymbol{r}_1$ and using the definition of the conditional average, (A 3) becomes

$$\langle u^2\rangle \approx \int \mathrm{d}\boldsymbol{r}_1\, n\langle \boldsymbol{u}\rangle_1(\boldsymbol{x}\,|\,\boldsymbol{r}_1)\cdot\langle \boldsymbol{u}\rangle_1(\boldsymbol{x}\,|\,\boldsymbol{r}_1)$$

$$-\int \mathrm{d}\boldsymbol{r}_1\,\mathrm{d}\boldsymbol{r}_2[ng(\boldsymbol{r}_1\,|\,\boldsymbol{r}_2)-n^2]\,\langle \boldsymbol{u}\rangle_1(\boldsymbol{x}\,|\,\boldsymbol{r}_2)\cdot\langle \boldsymbol{u}\rangle_1(\boldsymbol{x}\,|\,\boldsymbol{r}_1)$$

$$+\int \mathrm{d}\boldsymbol{r}_1\,\mathrm{d}\boldsymbol{r}_2\,\tfrac{1}{2}ng(\boldsymbol{r}_1|\boldsymbol{r}_2)\,\langle \boldsymbol{u}'\rangle_2(\boldsymbol{x}\,|\,\boldsymbol{r}_1,\boldsymbol{r}_2)\cdot\langle \boldsymbol{u}'\rangle_2(\boldsymbol{x}\,|\,\boldsymbol{r}_1,\boldsymbol{r}_2). \qquad \text{(A 4)}$$

The first two terms term in (A 4) were used to obtain the approximate expression (2.13) for the variance in §2. It is our purpose here to show that the last term, which involves two-particle velocity correlations, is smaller than the $O(U^{s2})$ terms retained in the body of the paper.

To this end, we derive conservation equations of the two-particle velocity disturbance $\langle \boldsymbol{u}'\rangle_2(\boldsymbol{x}\,|\,\boldsymbol{r}_1,\boldsymbol{r}_2)$. Such equations may be obtained by taking the conditional average of (2.4) with two particle positions fixed and subtracting from it the conditional averages of (2.4) with one particle fixed at $\boldsymbol{r}_1$ and with one particle fixed at $\boldsymbol{r}_2$, to give

$$-\mu\nabla^2\langle \boldsymbol{u}'\rangle_2(\boldsymbol{x}\,|\,\boldsymbol{r}_1,\boldsymbol{r}_2)+\boldsymbol{\nabla}\langle p'\rangle_2 = \boldsymbol{f}[g_3(\boldsymbol{x}\,|\,\boldsymbol{r}_1,\boldsymbol{r}_2)-g(\boldsymbol{x}\,|\,\boldsymbol{r}_1)-g(\boldsymbol{x}\,|\,\boldsymbol{r}_2)+n], \qquad \text{(A 5a)}$$

$$\boldsymbol{\nabla}\cdot\langle \boldsymbol{u}'\rangle_2(\boldsymbol{x}\,|\,\boldsymbol{r}_1,\boldsymbol{r}_2) = 0. \qquad \text{(A 5b)}$$

Note that the two-particle velocity correlations in (A 5a) are induced only by three-particle position correlations. It was seen in §3 that the three-particle probability density differs from the sum of the two-particle probabilities only if at least two of the particles are within an $O(a)$ distance, cf. (3.17) and the discussion following (3.1). Thus, the right-hand side of (A 4a) and the two-particle velocity disturbance $\langle \boldsymbol{u}'\rangle_2(\boldsymbol{x}\,|\,\boldsymbol{r}_1,\boldsymbol{r}_2)$ are only non-zero when $\boldsymbol{r}_1$ and $\boldsymbol{r}_2$ are within an $O(a)$ distance of one another.

We are now ready to estimate the orders of magnitude of the errors, represented by the final term in (A 4), incurred by neglecting the two-particle velocity correlations. The first of the two spatial integrals in the final term to be performed, the one in $\boldsymbol{r}_1$, will converge within an $O(a)$ distance of the point $\boldsymbol{r}_2$, because beyond this distance the two-particle velocity disturbance decays to zero. One is then left with an integral over $\boldsymbol{r}_2$ of the $O(n^2)$ pair distribution function times the product of the $O(U^s\phi a/r_2)$ two-particle velocity disturbance and the $O(U^s a/r_2)$ one-particle velocity disturbance. The two-particle velocity disturbance is driven by the $O(\phi)$ source of momentum in (A 5a). The integral with respect to $\boldsymbol{r}_2$ is expected to converge at the $O(R_s)$ radial distance at which the velocity disturbances are screened. Its value is then $O(U^{s2}\phi^3 R_s/a)$, and is thus small compared with the $O(U^{s2}\phi R_s/a)$ term evaluated in §2. In making the point-particle approximations we have also neglected $O(U^{s2}\phi)$ contributions associated with the finite size of the particles. These finite-size effects are larger than any of the contributions arising from two-particle velocity correlations.

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