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Transverse shear-induced gradient diffusion in a dilute suspension of spheres

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We study the shear-induced gradient diffusion of particles in an inhomogeneous dilute suspension of neutrally buoyant spherical particles undergoing a simple shearing motion, with all inertia and Brownian motion effects assumed negligible. An expansion is derived for the flux of particles due to a concentration gradient along the directions perpendicular to the ambient flow. This expression involves the average velocity of the particles, which in turn is expressed as an integral over contributions from all possible configurations. The integral is divergent when expressed in terms of three-particle interactions and must be renormalized. For the monolayer case, such a renormalization is achieved by imposing the condition of zero total macroscopic flux in the transverse direction whereas, for the three-dimensional case, the additional constraint of zero total macroscopic pressure gradient is required. Following the scheme of Wang, Mauri & Acrivos (1996), the renormalized integral is evaluated numerically for the case of a monolayer of particles, giving for the gradient diffusion coefficient $0.077\gamma a^2\bar{c}^2$, where γ is the applied shear rate, a the radius of the spheres and $\bar{c}$ their areal fraction.

1. Introduction

The shear-induced diffusion of non-Brownian particles has recently been found to play an important role in many physical processes involving suspensions of particles dispersed in a viscous fluid (Acrivos 1995). In contrast to the Brownian diffusion of particles in a colloidal suspension, which is due to the thermal fluctuations of the interactions between the fluid and the particles, the shear-induced diffusion is due solely to the hydrodynamic interactions among the particles. Although, in principle, the process is deterministic, it can often be described as a diffusion process because of the random nature of the complicated hydrodynamic interactions. This shear-induced diffusion leads to a net migration of particles from regions of high concentration to regions of low and from regions of high shear rate to low. This has been shown to affect certain macroscopic properties of suspensions in a major way (Acrivos 1995).

In an earlier publication (Wang, Mauri & Acrivos 1996, hereafter referred to as I) we studied the shear-induced self-diffusion of both a liquid tracer and a tagged particle along the directions perpendicular to the ambient flow in a dilute suspension of neutrally buoyant smooth spheres of uniform concentration undergoing a simple shearing motion in the absence of inertia and Brownian motion effects and derived expressions for the corresponding tracer diffusivities to leading order in the particle concentration. Here, we shall examine the corresponding problems of gradient diffusion in a dilute suspension of smooth spheres and determine the corresponding gradient diffusion coefficients, which are key parameters in any constitutive equation that relates the particle flux to non-uniformities in the particle concentration profile. The analogous case involving suspensions of rough spheres was studied by da Cunha & Hinch (1996).

In the next section, the problem is formulated from both the Eulerian and the Lagrangian points of view. But, since integrals in the corresponding expressions turn out to be divergent, they are renormalized for a monolayer of spheres using the constraint of zero macroscopic flux as shown in § 3 and the renormalized integrals are then evaluated numerically based on the trajectory computation scheme described in I. In the final section, the divergent integral expressions for the case of a three-dimensional distribution of spheres are renormalized using the constraint of zero macroscopic pressure gradient together with the constraint of zero macroscopic volumetric flux.

2. Statement of the problem

Consider a dilute suspension of rigid smooth spheres undergoing a simple shear flow $\boldsymbol{U} = \gamma x_2 \boldsymbol{i}_1$ with negligible inertia and Brownian motion effects. Here, γ is the shear rate, x_2 is the component of the position vector along the direction of the gradient of the ambient flow, and $\boldsymbol{i}_1$ is the unit vector in the direction of the ambient flow. If the suspension is inhomogeneous, the average velocity of a test sphere relative to the bulk flow is, in general, not zero. To determine this velocity to leading order in the particle concentration, we only need to consider three-particle interactions. Recall that the interaction of two smooth spheres does not contribute to the drift velocity in the transverse directions owing to the symmetry of the geometry and the reversibility of the governing Stokes equations.

One way of defining the Eulerian average instantaneous velocity, $V^E_{p,j}$, of a test sphere A along the direction $\boldsymbol{i}_j$ ($\boldsymbol{i}_2$ and $\boldsymbol{i}_3$ are unit vectors in the direction of the gradient of the ambient flow and in the direction opposite to the vorticity of the ambient flow, respectively) is by taking the ensemble average of its velocity $V_j(\boldsymbol{0}|\boldsymbol{y},\boldsymbol{z})$ in the presence of two other spheres B and C, at a certain instant of time $t = t_i$,

$$V^E_{p,j} \equiv \int V_j(\boldsymbol{0}|\boldsymbol{y},\boldsymbol{z})P(\boldsymbol{y},\boldsymbol{z}|\boldsymbol{0})\mathrm{d}^3\boldsymbol{y}\mathrm{d}^3\boldsymbol{z}, \quad \boldsymbol{y} \equiv \boldsymbol{Y} - \boldsymbol{X}, \quad \boldsymbol{z} \equiv \boldsymbol{Z} - \boldsymbol{X}, \quad j = 2,3, \tag{1}$$

where $\boldsymbol{X}$, $\boldsymbol{Y}$ and $\boldsymbol{Z}$ refer to the absolute positions of A, B and C, respectively. Thus, $\boldsymbol{y}$ and $\boldsymbol{z}$ are the positions of sphere B and C relative to A, while $P(\boldsymbol{y},\boldsymbol{z}|\boldsymbol{0})$ denotes the probability density of finding a second sphere B at $\boldsymbol{y}$ and a third sphere C at $\boldsymbol{z}$ under the condition that there exists a test sphere at the origin. Since, from the solution of the creeping flow equations, $V_j(\boldsymbol{0}|\boldsymbol{y},\boldsymbol{z})$, the velocity of A is completely determined given $\boldsymbol{y}$ and $\boldsymbol{z}$, it is clear that the evaluation of $V^E_{p,j}$ requires knowledge of the probability density function $P(\boldsymbol{y},\boldsymbol{z}|\boldsymbol{0})$.

Now, it is obvious from the conservation of probability that

$$P(\boldsymbol{y},\boldsymbol{z}|\boldsymbol{X})\mathrm{d}^3\boldsymbol{y}\mathrm{d}^3\boldsymbol{z}|_{t_1} = P(\boldsymbol{y},\boldsymbol{z}|\boldsymbol{X})\mathrm{d}^3\boldsymbol{y}\mathrm{d}^3\boldsymbol{z}|_{t_2} \tag{2}$$

along any given three-particle trajectory for any two instants of time t_1 and t_2. The probability $P(\boldsymbol{y},\boldsymbol{z}|\boldsymbol{X})$ can then be determined by tracing the volume element $\mathrm{d}^3\boldsymbol{y}\mathrm{d}^3\boldsymbol{z}$ along a trajectory back to $t = -\infty$, where the probability density function for finding any one of the spheres is set equal to $n(\boldsymbol{x})$, the local average density of the particles. We further take n to be a linear function of position in the same direction $\boldsymbol{i}_j$ as that of the average velocity under study, hence

$$P_j^{-\infty}(\boldsymbol{x}) = n_0 + \frac{\mathrm{d}n}{\mathrm{d}x_j}x_j + \ldots, \quad j = 2, 3, \tag{3}$$

where n_0 and $\mathrm{d}n/\mathrm{d}x_j$ are treated as given constant parameters and the repeated index does *not* indicate summation. Thus, the probability density of the configuration B at $\boldsymbol{Y}^{-\infty}$ and C at $\boldsymbol{Z}^{-\infty}$ is given by the product of the probability density of finding each particle, or

$$P_j^{-\infty}(\boldsymbol{y}^{-\infty}, \boldsymbol{z}^{-\infty}|\boldsymbol{X}^{-\infty}) = n_0^2\left[1 + \frac{\mathrm{d}n}{n_0\mathrm{d}x_j}(y_j^{-\infty} + z_j^{-\infty} + 2X_j^{-\infty}) + \ldots\right], \tag{4}$$

with

$$X_j^{-\infty} = \int_{t_i}^{-\infty} V_j(\boldsymbol{X}|\boldsymbol{y}, \boldsymbol{z})\mathrm{d}t \tag{5}$$

being the displacement of the test sphere when the element moves along a trajectory from $t = t_i$ to its position at $t = -\infty$.

The analysis outlined above presupposes that, within the suspension, we can distinguish three widely separated length scales, specifically: (i) the micro-length scale equal to the particle radius a; (ii) the macro-length scale L, which refers to the linear dimensions of a macroscopic region, containing a large number of randomly distributed particles, wherein n and therefore the particle concentration c, vary according to (3) with $\mathrm{d}n/\mathrm{d}x_j$ being $O(n_0/L)$; and (iii) the linear dimension l, such that $a \ll l \ll L$, of an intermediate region surrounding the test particle but embedded in the macro-domain within which B and C must lie if they are to interact with A. We further suppose that B and C, which are otherwise indistinguishable from the remaining particles, enter this interaction region at random locations on its boundary on the upstream side of A, and that the probability of finding B and/or C at a given point is also given by (3). Furthermore, this implies that, at the end of their interaction with A, particles B and C re-enter the macroscopic region downstream of A where their positions again become indeterminate on account of their interaction with a large number of particles that are randomly distributed.

3. The monolayer case

First, we consider the monolayer case where the motion of all the particles is confined to a plane perpendicular to $\boldsymbol{i}_3$, and compute the average velocity of A in the direction $\boldsymbol{i}_2$. Therefore, similar to the three-dimensional case, the Eulerian average velocity of a test particle can be written as

$$\bar{V}_p^E = \iint V_2(\boldsymbol{0}|\boldsymbol{y}, \boldsymbol{z})P(\boldsymbol{y}, \boldsymbol{z}|\boldsymbol{0})\mathrm{d}y_1\mathrm{d}y_2\mathrm{d}z_1\mathrm{d}z_2, \tag{6}$$

where the overbar denotes that the quantity corresponds to the monolayer case.

For computational convenience, the above four-fold integral is converted to a three-fold integral by integrating along the trajectories of the spheres. It is clear from (2) that, although the element $\mathrm{d}y_1\mathrm{d}y_2\mathrm{d}z_1\mathrm{d}z_2$ changes along a trajectory, the quantity $P(\boldsymbol{y}, \boldsymbol{z}|\boldsymbol{X})\mathrm{d}y_1\mathrm{d}y_2\mathrm{d}z_1\mathrm{d}z_2$ or, because $\mathrm{d}y_1 = v_1^B\mathrm{d}t_i$, where v_1^B is the component of the velocity of particle B in the direction of the ambient flow, the rate $P(\boldsymbol{y}, \boldsymbol{z}|\boldsymbol{X})v_1^B\mathrm{d}y_2\mathrm{d}z_1\mathrm{d}z_2$ remains the same and changes only from one trajectory to another. Therefore, since

$P(\boldsymbol{y},\boldsymbol{z}|\boldsymbol{X})$ is presumed known at $t=-\infty$, we can convert (6) into

$$\bar{V}_p^E = \int v_1^B(-\infty)\mathrm{d}y_2^{-\infty}\mathrm{d}z_1^{-\infty}\mathrm{d}z_2^{-\infty} \int V_2 P^{-\infty}(\boldsymbol{y}^{-\infty},\boldsymbol{z}^{-\infty}|\boldsymbol{X}^{-\infty})\mathrm{d}t_i,$$

where the superscript $-\infty$ denotes the value of the corresponding quantity at $t=-\infty$. To avoid counting two identical configurations twice, we denote as B the sphere which first arrives at a reference plane x_1^r =constant, and the other as C. After substituting (4) into the above expression and noting that the term containing only the uniform distribution drops out because it does not lead to any net particle migration, (6) can be written as

$$\bar{V}_p^E = \bar{n}_0\frac{\mathrm{d}\bar{n}}{\mathrm{d}x_2}\int v_1^B(-\infty)\mathrm{d}y_2^{-\infty}\mathrm{d}z_1^{-\infty}\mathrm{d}z_2^{-\infty}\int_{-\infty}^{+\infty}[(y_2^{-\infty}+z_2^{-\infty})V_2+2X_2^{-\infty}V_2]\mathrm{d}t_i.$$

Substituting $V_2=-\mathrm{d}X_2^{-\infty}/\mathrm{d}t_i$ from (5), the second term in the square bracket becomes, on integration with respect to $\mathrm{d}t_i$,

$$\int_{-\infty}^{+\infty}2X_2^{-\infty}V_2\mathrm{d}t_i = -\int_{-\infty}^{+\infty}2X_2^{-\infty}\frac{\mathrm{d}X_2^{-\infty}}{\mathrm{d}t_i}\mathrm{d}t_i = -(\Delta X_2)^2,$$

where ΔX_2, the total displacement of the test sphere due to an encounter with spheres B and C, is given by

$$\Delta X_2 = X_2^{-\infty}(t_i\to\infty) = \int_{-\infty}^{+\infty}V_2(\boldsymbol{X}|\boldsymbol{y},\boldsymbol{z})\mathrm{d}t. \tag{7}$$

Thus, the contribution of the second term is related to the self-diffusion coefficient $\bar{D}_p^S$ and hence the average velocity can be expressed as

$$\bar{V}_p^E = -\frac{2}{\bar{n}_0}\bar{D}_p^S\frac{\mathrm{d}\bar{n}}{\mathrm{d}x_2}+\bar{n}_0\frac{\mathrm{d}\bar{n}}{\mathrm{d}x_2}\int\Delta X_2(y_2^{-\infty}+z_2^{-\infty})v_1^B(-\infty)\mathrm{d}y_2^{-\infty}\mathrm{d}z_1^{-\infty}\mathrm{d}z_2^{-\infty}, \tag{8}$$

where the particle self-diffusivity $\bar{D}_p^S$ is given by (cf. I),

$$\bar{D}_p^S = \tfrac{1}{2}\bar{n}_0^2\int(\Delta X_2)^2 v_1^B(-\infty)\mathrm{d}y_2^{-\infty}\mathrm{d}z_1^{-\infty}\mathrm{d}z_2^{-\infty}, \tag{9}$$

which is of order $\bar{c}^2$, with $\bar{c}$ being the areal fraction occupied by the particle phase. It is easy to show that the above is consistent with the expression given by da Cunha & Hinch (1996) for the flux of particles in a slightly non-uniform dilute suspension of rough spheres, which they obtained by considering only two-particle interactions.

The integral in (8) is not absolutely convergent, as can be seen by noting that $\Delta X_2 \sim O(1/\bar{r}^5)$ for large $|\boldsymbol{y}|$ and $|\boldsymbol{z}|$ with $\bar{r}$ being a typical distance between any two of the spheres which participate in the encounter (cf. § 5 of I). As shown by Batchelor (1972) for the analogous situation of a sedimenting suspension, such a divergence arises from the fact that the integral (8) for $\bar{V}_2^E$ is expressed in terms of the positions of only two particles rather than, as should be the case for any mean quantity, over the configuration space of the whole suspension. Fortunately, again following Batchelor (1972), we can renormalize such a divergent integral by applying a global constraint inherent in the formulation of the problem, in this case the constraint of zero total areal flux in the $\boldsymbol{i}_2$ direction for the suspension as a whole, i.e.

$$\bar{J}_p+\bar{J}_f=0, \tag{10}$$

where $\bar{J}_p$ and $\bar{J}_f$ are the areal fluxes for the particle phase and the fluid phase, respectively.

To obtain an expression for $\bar{J}_p$ we recall that the particle flux consists of two parts: a convective contribution $\bar{c}\bar{V}_p^E$ where $\bar{V}_p^E$ is the particle drift velocity encountered earlier, and a purely diffusive flux equal to $-\bar{D}_p^S \mathrm{d}\bar{c}/\mathrm{d}x_2$ where $\bar{D}_p^S$ is the particle tracer diffusivity in a homogeneous suspension. To see why this diffusive term must appear in the expression for $\bar{J}_p$, consider the case of a suspension of uniform concentration in which, however, some of the particles are colored. Under these conditions, the drift velocity is, of course, zero. But, if the concentration c of these coloured particles is not uniform, their flux does not vanish and is given by the diffusive expression $-\bar{D}_p^S \mathrm{d}\bar{c}/\mathrm{d}x_2$ noted above. Consequently,

$$\bar{J}_p = \bar{c}\bar{V}_p^E - \bar{D}_p^S \frac{\mathrm{d}\bar{c}}{\mathrm{d}x_2}$$

and, similarly,

$$\bar{J}_f = (1-\bar{c})\bar{V}_f^E - \bar{D}_f^S \frac{\mathrm{d}(1-\bar{c})}{\mathrm{d}x_2},$$

where $\bar{V}_f^E$ and $\bar{D}_f^S$ are, respectively, the average Eulerian liquid velocity and liquid self-diffusivity with $\bar{D}_f^S$ given by

$$\bar{D}_f^S = \tfrac{1}{2}\bar{n}_0^2 \int (\Delta X_2^*)^2 v_1^B(-\infty)\mathrm{d}y_2^{-\infty}\mathrm{d}z_1^{-\infty}\mathrm{d}z_2^{-\infty}, \tag{11}$$

where ΔX_2^* denotes the displacement of a fluid point due to its encounter with two spheres B and C.

From this constraint, we find that $\bar{V}_f^E \sim O(\bar{c}^2 \mathrm{d}\bar{c}/\mathrm{d}x_2)$, which is of smaller order than the velocity $\bar{V}_p^E$ given by (8). On the other hand, by means of a procedure similar to that used in arriving at (8) for $\bar{V}_p^E$, we can obtain that

$$\bar{V}_f^E = -\frac{2}{\bar{n}_0}\bar{D}_f^S \frac{\mathrm{d}\bar{n}}{\mathrm{d}x_2} + \bar{n}_0 \frac{\mathrm{d}\bar{n}}{\mathrm{d}x_2}\int \Delta X_2^*(y_2^{-\infty}+z_2^{-\infty})v_1^B(-\infty)\mathrm{d}y_2^{-\infty}\mathrm{d}z_1^{-\infty}\mathrm{d}z_2^{-\infty}, \tag{12}$$

which is of order $\bar{c}\mathrm{d}\bar{c}/\mathrm{d}x_2$. But, since $\bar{V}_f^E$ must vanish to $O(\bar{c}\mathrm{d}\bar{c}/\mathrm{d}x_2)$, we have, by subtracting the above from the right-hand side of (8), that

$$\begin{aligned}\bar{V}_p^E = &-\frac{2}{\bar{n}_0}(\bar{D}_p^S - \bar{D}_f^S)\frac{\mathrm{d}\bar{n}}{\mathrm{d}x_2} \\ &+ \bar{n}_0 \frac{\mathrm{d}\bar{n}}{\mathrm{d}x_2}\int (\Delta X_2 - \Delta X_2^*)(y_2^{-\infty}+z_2^{-\infty})v_1^B(-\infty)\mathrm{d}y_2^{-\infty}\mathrm{d}z_1^{-\infty}\mathrm{d}z_2^{-\infty} + O\left(\bar{c}^2\frac{\mathrm{d}\bar{c}}{\mathrm{d}x_2}\right).\end{aligned} \tag{13}$$

Since it can easily be shown that the difference $\Delta X_2 - \Delta X_2^*$ is of order smaller than $1/\bar{r}^5$, with $\bar{r}$ denoting the typical distance between any two spheres (cf. § 5 of I), the integral in the above expression is convergent and can be evaluated numerically.

The expression for $\bar{V}_p^E$ given by (13) can also be obtained more directly starting from the particle flux expression as given by the Fokker–Planck equation

$$\bar{J}_p = \bar{c}\bar{V}_p^L - \frac{\mathrm{d}}{\mathrm{d}x_2}\bar{D}_p^S \bar{c} = \bar{c}\bar{V}_p^L - \bar{c}\frac{\mathrm{d}\bar{D}_p^S}{\mathrm{d}\bar{c}}\frac{\mathrm{d}\bar{c}}{\mathrm{d}x_2} - \bar{D}_p^S\frac{\mathrm{d}\bar{c}}{\mathrm{d}x_2},$$

with all terms of order $\bar{c}^2\mathrm{d}\bar{c}/\mathrm{d}x_2$, where $\bar{V}_p^L$ is the Lagrangian particle velocity given by

$$\bar{V}_p^L = \frac{\bar{c}}{\pi^2}\frac{\mathrm{d}\bar{c}}{\mathrm{d}x_2}\int \Delta X_2(y_2^{-\infty}+z_2^{-\infty})v_1^B \mathrm{d}y_2^{-\infty}\mathrm{d}z_1^{-\infty}\mathrm{d}z_2^{-\infty}.$$

On the other hand, the Lagrangian expression for the fluid flux is

$$\bar{J}_f = (1-\bar{c})\bar{V}_f^L - (1-\bar{c})\frac{\mathrm{d}\bar{D}_f^S}{\mathrm{d}\bar{c}}\frac{\mathrm{d}\bar{c}}{\mathrm{d}x_2} - \bar{D}_f^S\frac{\mathrm{d}(1-\bar{c})}{\mathrm{d}x_2}.$$

But since the second term is of $O(\bar{c}\mathrm{d}\bar{c}/\mathrm{d}x_2)$, the requirement that $\bar{J}_p + \bar{J}_f = 0$ gives that, to order $\bar{c}\mathrm{d}\bar{c}/\mathrm{d}x_2$,

$$\bar{V}_f^L = \frac{\bar{c}}{\pi^2}\frac{\mathrm{d}\bar{c}}{\mathrm{d}x_2}\int \Delta X_2^*(y_2^{-\infty}+z_2^{-\infty})v_1^B \mathrm{d}y_2^{-\infty}\mathrm{d}z_1^{-\infty}\mathrm{d}z_2^{-\infty} = \frac{\mathrm{d}\bar{D}_f^S}{\mathrm{d}\bar{c}}\frac{\mathrm{d}\bar{c}}{\mathrm{d}x_2}.$$

Therefore, by subtracting the above expression for $\bar{V}_f^L$ from that for $\bar{V}_p^L$, we have that

$$\bar{V}_p^E = \bar{V}_p^L - \frac{\mathrm{d}\bar{D}_p^S}{\mathrm{d}\bar{c}}\frac{\mathrm{d}\bar{c}}{\mathrm{d}x_2} = -\left[\frac{\mathrm{d}}{\mathrm{d}\bar{c}}(\bar{D}_p^S - \bar{D}_f^S)\right]\frac{\mathrm{d}\bar{c}}{\mathrm{d}x_2} + \frac{\bar{c}}{\pi^2}\frac{\mathrm{d}\bar{c}}{\mathrm{d}x_2}\int(\Delta X_2 - \Delta X_2^*)v_1^B(y_2^{-\infty}+z_2^{-\infty})\mathrm{d}y_2^{-\infty}\mathrm{d}z_1^{-\infty}\mathrm{d}z_2^{-\infty},$$

which is the same as (13) obtained via the Eulerian description.

4. Numerical results for a monolayer

We used the same numerical scheme as in I to compute the average velocity of a sphere in a monolayer by evaluating the integral in (13). However, in view of the fact that the difference $\Delta X_2 - \Delta X_2^*$ in the integrand is numerically much smaller than either of these two terms, special care had to be taken in evaluating the integral because even a small residue error in either ΔX_2 or ΔX_2^* will lead to a significant numerical uncertainty. After evaluating the integral and using the expressions given by (9) and (11), for $\bar{D}_p^S$ and $\bar{D}_f^S$, respectively, we find

$$\bar{V}_p^E = -0.045\gamma a^2\bar{c}\frac{\mathrm{d}\bar{c}}{\mathrm{d}y},$$

$$\bar{V}_f^L = \frac{\mathrm{d}\bar{D}_f}{\mathrm{d}\bar{c}}\frac{\mathrm{d}\bar{c}}{\mathrm{d}y} = 0.134\gamma a^2\bar{c}\frac{\mathrm{d}\bar{c}}{\mathrm{d}y},$$

while

$$\bar{V}_p^L = \bar{V}_p^E + \frac{\mathrm{d}\bar{D}_p}{\mathrm{d}\bar{c}}\frac{\mathrm{d}\bar{c}}{\mathrm{d}y} = +0.019\gamma a^2\bar{c}\frac{\mathrm{d}\bar{c}}{\mathrm{d}y}.$$

The reason why $\bar{V}_p^L$, the Lagrangian average particle velocity, is positive, which appears counter intuitive, is still not clear.

Finally, we obtain the particle gradient diffusion coefficient for a monolayer of particles

$$\bar{D}_p^G = 0.077a^2\gamma\bar{c}^2,$$

which is defined by

$$\bar{J}_p = \bar{c}\bar{V}_p^E - \bar{D}_p^S\frac{\mathrm{d}\bar{c}}{\mathrm{d}x_2} \equiv -\bar{D}_p^G\frac{\mathrm{d}\bar{c}}{\mathrm{d}x_2}. \qquad (14)$$

5. Renormalization for the three-dimensional distribution of particles

In § 3, we were able to renormalize the expression for the average velocity of a test sphere in a monolayer of spheres using the constraint of zero bulk flux in the transverse direction. For the case of a three-dimensional distribution of spheres, however, this constraint is not sufficient to yield a convergent expression for the average velocity. Specifically, after applying this constraint to the three-dimensional case, the Eulerian average velocity becomes

$$V^E_{p,j} = -\frac{2}{n_0}(D^S_{p,j} - D^S_{f,j})\frac{\mathrm{d}n}{\mathrm{d}x_2} + n_0\frac{\mathrm{d}n}{\mathrm{d}x_j}\int(\Delta X_j - \Delta X^*_j)(y_j^{-\infty} + z_j^{-\infty})v_1^B(-\infty)\mathrm{d}y_2^{-\infty}\mathrm{d}y_3^{-\infty}\mathrm{d}z_1^{-\infty}\mathrm{d}z_2^{-\infty}\mathrm{d}z_3^{-\infty} + O\left(c^2\frac{\mathrm{d}c}{\mathrm{d}x_2}\right). \tag{15}$$

But, since the difference $\Delta X_2 - \Delta X_2^*$ is of $O(1/\bar{r}^7)$ for large $\bar{r}$ the above expression is still not convergent and an additional renormalization is required. To obtain this additional constraint, we first note that the $O(1/\bar{r}^7)$ term in $\Delta X_2 - \Delta X_2^*$ for large $\bar{r}$ arises from the difference of the velocity of a test sphere, $V_2(\boldsymbol{X}|\boldsymbol{y},\boldsymbol{z})$, from that of a fluid tracer, $V_2^*(\boldsymbol{X}|\boldsymbol{y},\boldsymbol{z})$, due to their interactions with two other spheres B and C. Specifically, following the reflection method, the velocity disturbance due to an isolated sphere, say B, will be reflected by another sphere C, producing a velocity disturbance $\boldsymbol{u}'$, which is of $O(1/\bar{r}^5)$. This disturbance influences the velocity of the test particle, which can be either a test sphere or a fluid tracer. However, according to Faxén's law, the response velocity of a test sphere differs from that of a fluid tracer by an amount equal to $\frac{1}{6}\nabla^2\boldsymbol{u}'$. It is the cumulative effect of this response velocity difference on the reflected disturbance arising from all the sphere pairs in the suspension that renders the integral in (15) divergent. But, this sum is related to the average pressure gradient in the suspension in view of the fact that the difference $\frac{1}{6}\nabla^2\boldsymbol{u}'$ in the response velocity is related to the corresponding pressure gradient disturbance by the usual Stokes equation $\mu\nabla^2\boldsymbol{u}' = \nabla p'$. On the other hand, since, from a macroscopic point of view, the bulk pressure gradient in the transverse directions must be zero even in the presence of a concentration gradient in this direction, it is clear that this constraint of zero bulk pressure gradient in the transverse direction can be used to remove the non-convergence in the average velocity expression by subtracting the corresponding divergent parts and evaluating the remaining convergent integral (Batchelor 1972).

The constraint of zero bulk pressure gradient in the transverse directions can be expressed as an ensemble average

$$\langle\nabla p\rangle_j = \boldsymbol{i}_j\cdot\int\nabla p(\boldsymbol{Y},\boldsymbol{Z})P(\boldsymbol{Y},\boldsymbol{Y})\mathrm{d}^3\boldsymbol{Y}\mathrm{d}^3\boldsymbol{Z} = 0, \tag{16}$$

where ∇p is the pressure gradient at a test point $\boldsymbol{X}^t$ located at the origin for the configuration of two spheres B and C at $\boldsymbol{Y}$ and $\boldsymbol{Z}$ respectively, while $P(\boldsymbol{Y},\boldsymbol{Z})$ denotes the probability density of this configuration. Note that the above integral is taken over all possible configurations as long as the spheres B and C do not overlap, including the configuration where the sample point lies within one of the spheres. Since the midpoint of the centreline of B and C does not move in the $\boldsymbol{i}_j$ direction, the probability density function $P(\boldsymbol{Y},\boldsymbol{Z})$ is given by

$$P(\boldsymbol{Y},\boldsymbol{Z}) = P_j^{-\infty}(\boldsymbol{Y}^{-\infty},\boldsymbol{Z}^{-\infty})q(|\boldsymbol{Y}-\boldsymbol{Z}|), \tag{17}$$

with $q(|\boldsymbol{Y}-\boldsymbol{Z}|)$ being the probability factor given by Batchelor & Green (1972).

Now, the domain of integration in (16) can be divided into two parts depending on whether the sample point lies outside or inside the spheres, i.e.

$$\langle \nabla p \rangle_j = \boldsymbol{i}_j \cdot \left[\int_{outside} +2 \int_{inside\ one\ of\ the\ spheres} \right]. \tag{18}$$

The first integral

$$I_1 = \boldsymbol{i}_j \cdot \int_{outside} \nabla p(\boldsymbol{Y}, \boldsymbol{Z}) P(\boldsymbol{Y}, \boldsymbol{Z}) \mathrm{d}^3 \boldsymbol{Y} \mathrm{d}^3 \boldsymbol{Z}, \tag{19}$$

is related to the divergent part in the expression (15) as mentioned above. For the second integral, it is convenient to use $\boldsymbol{r} \equiv \boldsymbol{X}^t - \boldsymbol{Y}$ and $\boldsymbol{z} \equiv \boldsymbol{Z} - \boldsymbol{Y}$ to yield

$$I_2 = 2\boldsymbol{i}_j \cdot \int_{1<r} \nabla p(\boldsymbol{r}, \boldsymbol{z}) P(\boldsymbol{r}, \boldsymbol{z}) \mathrm{d}^3 \boldsymbol{r} \mathrm{d}^3 \boldsymbol{z} \tag{20}$$

with

$$P(\boldsymbol{r}, \boldsymbol{z}) = n_0^2 \left[1 + \frac{1}{2n_0} \frac{\mathrm{d}n}{\mathrm{d}x_j} (z_j - 2r_j) + \ldots \right] q(z). \tag{21}$$

The integral I_2, which is over the domain inside one of the spheres, say B, can be determined by converting the volume integral into a surface integral by Gauss's theorem, as done for similar problems (Batchelor & Green 1972).

Substituting (21) into the expression for I_2 yields

$$I_2 = n_0 \frac{\mathrm{d}n}{\mathrm{d}x_j} \boldsymbol{i}_j \cdot \int_{z>2} q(z) \mathrm{d}^3 \boldsymbol{z} \int_{r<1} (z_j - 2r_j) \nabla p \mathrm{d}^3 \boldsymbol{r}. \tag{22}$$

Applying Gauss's theorem to the integral with respect to $\mathrm{d}^3\boldsymbol{r}$ gives

$$I_2 = n_0 \frac{\mathrm{d}n}{\mathrm{d}x_j} \int_{z>2} q(z) \mathrm{d}^3 \boldsymbol{z} \left[\boldsymbol{i}_j \cdot \int_{r=1} (z_j - 2r_j) p \boldsymbol{n} \mathrm{d}S + 2 \int_{r<1} p \mathrm{d}^3 \boldsymbol{r} \right]. \tag{23}$$

Obviously, I_2 is independent of the nature of the material inside the sphere B as long as its surface is rigid. So we can use any constitutive relation to determine the volume integral inside B. One convenient choice is to assume that the sphere is made of an infinitely viscous fluid with no surface tension on the interface. For this choice, the pressure field p is a harmonic function without any singularity inside the sphere. Therefore, its average over the volume inside the sphere equals the average on the surface of the sphere

$$\frac{1}{\frac{4}{3}\pi} \int_{r \leqslant 1} p \mathrm{d}^3 \boldsymbol{r} = \frac{1}{4\pi} \int_{r=1} p \mathrm{d}S \tag{24}$$

and thus

$$I_2 = n_0 \frac{\mathrm{d}n}{\mathrm{d}x_j} \boldsymbol{i}_j \cdot \int_{z>2} q(z) \mathrm{d}^3 \boldsymbol{z} \int_{r=1} (\tfrac{2}{3} + z_j r_j - 2r_j^2) p \mathrm{d}S. \tag{25}$$

Since, for large z, the leading term for p is due to the reflection by C of the disturbance induced by the presence of B, which is of order $1/z^6$, the above integral is absolutely convergent and can be evaluated numerically.

Using the same procedure, as in § 3 we can convert (19) into

$$I_1 = n_0 \frac{\mathrm{d}n}{\mathrm{d}x_j} I_{10} + n_0 \frac{\mathrm{d}n}{\mathrm{d}x_j} \int \Delta X_j'(y_j^{-\infty} + z_j^{-\infty}) v_1^B(-\infty) \mathrm{d}y_2^{-\infty} \mathrm{d}y_3^{-\infty} \mathrm{d}z_1^{-\infty} \mathrm{d}z_2^{-\infty} \mathrm{d}z_3^{-\infty}, \tag{26}$$

where

$$\Delta X_j' = \int_{-\infty}^{+\infty} \tfrac{1}{6} \nabla^2 V_j'(\boldsymbol{X} | \boldsymbol{y}, \boldsymbol{z}) \mathrm{d}t, \tag{27}$$

and

$$I_{10} = \int \alpha(y_j^{-\infty} + z_j^{-\infty}) v_1^B(-\infty) \mathrm{d}y_2^{-\infty} \mathrm{d}y_3^{-\infty} \mathrm{d}z_1^{-\infty} \mathrm{d}z_2^{-\infty} \mathrm{d}z_3^{-\infty}, \tag{28}$$

with

$$\alpha = \int_{-\infty}^{+\infty} \tfrac{1}{6} X_j^{-\infty} \nabla^2 V_j'(\boldsymbol{X}|\boldsymbol{y},\boldsymbol{z}) \mathrm{d}t. \tag{29}$$

Here, $V_j'(\boldsymbol{X}|\boldsymbol{y},\boldsymbol{z})$ is the velocity disturbance at a fluid point located at $\boldsymbol{X}$ with sphere B at $\boldsymbol{y}$ and C at $\boldsymbol{z}$. $\Delta X_j'$ and α can be computed just as ΔX_j by integrating along the trajectory for any given initial configurations. The integral in the expression for I_{10} is obviously convergent in view of the fact that $\alpha \sim O(1/\bar{r}^9)$, which can be seen by noting that $X_j^{-\infty}$ is of $O(1/\bar{r}^2)$ for large $\bar{r}$.

The origin of the non-convergence of the integral in (26) is the same as that for the integral in (15). Hence, we can remove the non-convergent part in (15) using the constraint (16) together with (25), (26) and (28) to yield

$$\begin{aligned} V_{p,j}^E = & -\frac{2}{n_0}(D_{p,j}^S - D_{f,j}^S)\frac{\mathrm{d}n}{\mathrm{d}x_j} - n_0\frac{\mathrm{d}n}{\mathrm{d}x_j} I_{10} - I_2/6 \\ & + n_0 \frac{\mathrm{d}n}{\mathrm{d}x_j} \int (\Delta X_j - \Delta X_j^* - \tfrac{1}{6}\Delta X_j')(y_j^{-\infty} + z_j^{-\infty}) v_1^B(-\infty) \mathrm{d}y_2^{-\infty} \mathrm{d}y_3^{-\infty} \mathrm{d}z_1^{-\infty} \mathrm{d}z_2^{-\infty} \mathrm{d}z_3^{-\infty}. \end{aligned} \tag{30}$$

The term $\Delta X_j - \Delta X_j^* - \frac{1}{6}\Delta X_j'$ in the integrand decays faster than $1/\bar{r}^8$, which can be seen by considering the hydrodynamic interactions among three spheres in a simple shear flow for large $\bar{r}$ in terms of the reflection method. Thus, the integral is now convergent.

In conclusion, we can renormalize the average velocity expression by using together the constraints of zero bulk flux and zero bulk pressure gradient and obtain convergent expressions for the Eulerian average particle velocity in the transverse directions which can be evaluated numerically. This in turn can be used to determine the gradient diffusivity through (14). This has not been attempted, however, because of the intense computational effort that is required.

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