# An averaged-equation approach to particle interactions in a fluid suspension

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(Received 18 December 1974 and in revised form 11 May 1977)

Earlier ideas are combined to produce a systematic approach both to forming the bulk equations of motion of a dilute suspension and to calculating the overall hydrodynamic interactions between the suspended particles. Equations governing averaged field quantities are derived by taking ensemble averages of the conservation laws and constitutive relations. The bulk equations thus produced contain a term in which the averaging is performed holding one particle fixed. If now the same prescription is applied to fields averaged with one particle fixed, equations are produced containing a term averaged with two particles fixed, and so on up an infinite hierarchy. The hierarchy can be truncated in an asymptotic analysis for small particle concentrations.

This approach to the mechanics of suspensions is illustrated by applying it to three problems which have already been well studied by different methods. The problems concern the first effects of hydrodynamic interactions on the bulk stress and sedimentation velocity of a free suspension, and on the permeability of a fixed bed. Earlier results are recovered in a new light. Multiparticle effects, which before have occurred as divergent sums, are seen to arise because the suspension described by the averaged equations assumes a viscosity and density different from the solvent, or in the case of the fixed bed because the suspension starts behaving as a porous medium instead of as a Newtonian solvent. A close connexion is thus revealed between the averaged-equation description of the interactions and a self-consistentfield model.

#### 1. Introduction

In this paper I wish to bring together several ideas and techniques which others have introduced to overcome particular problems, and to set them out in a systematic approach to analysing the mechanics of suspensions. The approach will be illustrated by calculating the bulk stress of a fluid suspension of rigid spheres to  $O(c^2)$ , where cis the volume fraction of particles. Additional sections will deal more briefly with the sedimentation rate of heavy spheres to O(c) and the permeability of a bed of fixed spheres to O(c). While I have already found the general approach useful in some new suspension problems, I think it is most easily explained in the familiar context of the three problems selected. Earlier results obtained using different methods by Batchelor (1972) for the sedimentation rate, Batchelor & Green (1972a) for the bulk stress and Childress (1972) for the permeability will be confirmed in §§7, 5 and 8 respectively.

The basic idea behind the approach is that the conservation laws and constitutive relations which govern one realization of the suspension should be averaged over the ensemble of realizations. This procedure produces equations governing averaged field quantities. Thus in §2 we shall see that the average velocity is governed by a bulk momentum equation (2.3) and bulk stress relations (2.5) with (2.7). The idea of seeking such a pair of bulk equations governing the average velocity emerges in Landau & Lifschitz (1959, p. 76). Earlier studies of suspensions had considered only the rate of energy dissipation and characterized it by an effective viscosity. Landau and Lifschitz, however, calculated a volume average of the full stress tensor, implicitly for use in the bulk momentum equation. The first explicit mention of a bulk momentum equation for a suspension appears to be a footnote in Batchelor (1970). In the same paper Batchelor also agreed with Hashin (1964) that an ensemble average would be logically preferable to the volume average used by Landau & Lifschitz and several authors since them, although for his immediate purposes there was no difference between the two types of average so Batchelor gave the majority of his discussion in terms of the volume average, which is easier to visualize. When we come to calculate average hydrodynamic interactions we shall need to use the more powerful ensemble average in order to form averages in situations which lack spatial stationarity.

The bulk constitutive relation, formed by ensemble averaging the different constitutive relations in the particles and fluid, is found to include a term in which the averaging is performed holding one particle fixed, see (2.5) and (2.7). Thus the bulk equations are not closed: they contain not only bulk quantities but also conditionally averaged quantities. This fact was first noticed by Tam (1969) when he attempted to take an ensemble average of a Green's function formulation of the third problem mentioned above, the 'fixed-bed' problem. Note, however, that Tam's Green's function formulation is divergent in an infinite suspension and also his assertion is erroneous that the conditionally and unconditionally averaged velocities are asymptotically equal as the number of particles becomes large.

To find the conditionally averaged field quantities I suggest that the basic idea of averaging the conservation laws and constitutive relations should simply be repeated, but now averaging over the sub-ensemble which has the one particle fixed. We shall see in §3 that this procedure produces a momentum equation (3.1) and constitutive relations (3.3) with (3.4) which govern the conditionally averaged velocity field. Earlier workers have obtained equations for the conditionally averaged velocity, but by methods other than just repeating the process used for the bulk equations. Employing some manipulations of ensemble averages developed by Saffman (1971) in another problem, Lundgren (1972) conditionally averaged the Stokes equations governing the flow of the fluid within the suspension. Childress (1972) in his second method and Saffman (1973) adopted the same technique in the simpler point-particle limit in which each particle can exert only a concentrated force on the fluid. Refinements of Lundgren's formulation have been made by Buyevich & Markov (1973) and Howells (1974). In an entirely different approach to the calculation of the effect of hydrodynamic interactions, Batchelor (1972) and Batchelor & Green (1972a) used no equation governing conditionally averaged fields. Thus, in the past, forming the bulk equations for the suspension and calculating the effect of hydrodynamic interactions have been treated separately with different

techniques. The systematic approach adopted in this paper, however, brings together the two problems in a natural way, and to some extent avoids introducing special additional techniques to tackle the hydrodynamic interactions.

If we follow the suggestion of the last paragraph and conditionally average the constitutive relations, a term requiring an average with two particles held fixed is produced, see (3.3) and (3.4). Equations governing this new average velocity with the two fixed particles can be derived by a further application of an ensemble average, now over the sub-ensemble with the two particles fixed, to the conservation laws and constitutive relations. As will now be evident, an infinite hierarchy of linked equations is thus generated by repeatedly averaging with one more fixed particle at each level. Essentially the same hierarchy was found by Lundgren (1972) and Childress (1972).

Different methods have been employed to truncate the hierarchy. Lundgren (1972) and Buyevich & Markov (1973) both made an *ad hoc* closure assumption that the linking term took a particular functional form with the same coefficients at the bulk level as at the first conditional level, these coefficients being determined by a self-consistency condition. This closure assumption can be justified only in the dilute-suspension limit which ignores hydrodynamic interactions.

In this paper (§§ 3 and 4) we truncate the hierarchy by noting that the linking terms become small as  $c \rightarrow 0$ . Thus at the required order of approximation in an asymptotic analysis for  $c \rightarrow 0$  we can neglect the term which would otherwise couple our truncated hierarchy to the higher orders we wish to omit. The idea for this closure scheme can be found in Childress (1972) in his second method and in Saffman (1973). Effectively they both ignored the coupling to the averaged field with three particles fixed, so that the two fixed particles appear to be surrounded by pure solvent. This latter problem, however, they solved only at the point-particle approximation, which was adequate for their leading-order terms but which is not adequate in this paper. (Saffman moreover worked with Fourier transforms which are not appropriate beyond the point-particle approximation.) Howells (1974) was the first to use the closure scheme beyond the point-particle approximation although he organized the solution of the truncated hierarchy in a slightly different way to that adopted here.

The truncated hierarchy of equations is solved as a system of coupled equations. One starts with the last member of the hierarchy, the one which has the most particles held fixed, because this equation does not depend on the others. The field so obtained gives the forcing term in the next equation up the hierarchy. One continues step by step up the hierarchy until the unconditionally averaged field is finally obtained.

In the problems for the sedimentation rate and bulk stress of a free suspension this programme for solving the truncated hierarchy proceeds with none of the divergence difficulties which Batchelor (1972) and Batchelor & Green (1972a) overcame by a special method for the calculation of interactions. The verification of their results should dispel any doubts which may exist concerning the rigour and uniqueness of their method of circumventing divergent quantities.

It is convenient and instructive not to follow precisely the programme advocated above for solving the truncated hierarchy. Some modification of the programme is moreover essential in the fixed-bed problem. The modification proves convenient because it enables the final result, e.g. the sedimentation rate, to be found without first obtaining the full details of the entire average velocity field outside a fixed particle. The modification is also instructive because it provides a link with the alternative theory of interactions used by Batchelor (1972) and Batchelor & Green (1972a) and with the self-consistent field theories.

The modification to the programme for solving the truncated hierarchy involves a rearrangement of each equation before it is solved. The most successful rearrangements are those which make the left-hand side of the equation reflect the basic physics governing the motion of the bulk suspension as distinct from that of the pure solvent, leaving the right-hand side of the equation to reflect the detailed corrections to that basic physics. Thus in the bulk stress and sedimentation problems the left-hand side of the governing equation is essentially that for Stokes flow with values of the density and viscosity modified from the pure solvent values to those corresponding to the suspension, see (6.2) and (7.4). As pointed out by Brinkman (1947), when we come to the fixed-bed problem this modification of the basic physics is crucial because the suspension acts not like a viscous fluid but instead like a porous medium, and hence the left-hand side of the governing equation is essentially that for Darcy flow through a porous medium, see (8.2).

#### 2. Bulk equations of motion

In this section we proceed from the continuum equations governing one realization of the suspension to the governing equations for the bulk medium. At the microstructural level it is assumed that the solvent is Newtonian and that the suspended particles may be described by continuum mechanics. In the usual notation let the density be  $\rho$ , the Eulerian velocity **u**, the Cauchy stress  $\sigma$  and the force density **f**. These will be defined everywhere whether in the fluid or in the particles. At the interface between the two phases the velocity should be continuous and the surface stress  $\sigma \cdot \mathbf{n}$  discontinuous only to accommodate any surface tension.

The governing equations for the full suspension fall into two classes, conservation laws and constitutive relations. For the purposes of the general exposition, the main pair associated with the rheology of the suspension will be treated, namely the momentum conservation law and the stress constitutive relation. Any other relevant law or relation can be tackled in the same manner. The Cauchy momentum equation is

$$(\partial/\partial t + \mathbf{u} \cdot \nabla) \rho \mathbf{u} = \nabla \cdot \boldsymbol{\sigma} + \mathbf{f}.$$
(2.1)

The constitutive relations depend on the particular suspension. Only incompressible phases will be considered here, so everywhere the velocity satisfies the solenoidal constraint

$$\nabla \cdot \mathbf{u} = \mathbf{0},\tag{2.2}$$

which has associated with it a locally indeterminate pressure field p (defined everywhere as minus one third the trace of the stress tensor). The solvent is always Newtonian so that, in the fluid,

$$\boldsymbol{\sigma} = -p\mathbf{I} + 2\mu\mathbf{e},$$

where the Eulerian strain rate is  $\mathbf{e} = \frac{1}{2}(\nabla \mathbf{u} + \nabla \mathbf{u}^{T})$ . The stress relation within the particles is left open. For rigid particles this relation is degenerate; the strain rate

vanishes and the stress becomes locally indeterminate. With this important case a possible one to be considered, it is desirable to have alternative bulk statements which avoid the details of the stress law within the particles.

The basic equations will now be averaged using the statistical ensemble average. As a temporary notation for this section, angled brackets are used to denote the average and a prime to denote the fluctuation of a quantity about its mean value. In subsequent sections the field quantities only occur when averaged and so the unnecessary averaging signs will be dropped then. The averaged momentum equation is

$$(\partial/\partial t + \langle \mathbf{u} \rangle \cdot \nabla) \langle \rho \mathbf{u} \rangle = \nabla \cdot (\langle \boldsymbol{\sigma} \rangle - \langle (\rho \mathbf{u})' \mathbf{u}' \rangle) + \langle \mathbf{f} \rangle.$$
(2.3)

Batchelor (1970) has suggested that the mean Reynolds stress might be best combined with the mean stress tensor to form a single bulk stress tensor, as grouped together above. Analytic progress can only be made easily, however, when the Reynolds number based on the flow around a suspended particle is small, in which case the viscous stress dominates the Reynolds stress. The restriction which is now made to low particle Reynolds numbers (applicable to small suspended particles) does not necessarily imply that the bulk flow cannot have a large Reynolds number: it can be large so long as the bulk-flow length scale greatly exceeds the particle length scale.

The bulk equations of motion would form a complete set if, by averaging, sufficient local relations could be obtained between the bulk velocity  $\langle \mathbf{u} \rangle$ , the bulk strain rate  $\langle \mathbf{e} \rangle$ , the bulk mass flux  $\langle \rho \mathbf{u} \rangle$ , the bulk density  $\langle \rho \rangle$ , the bulk stress  $\langle \boldsymbol{\sigma} \rangle$ , the bulk force  $\langle \mathbf{f} \rangle$ , and perhaps a few bulk variables characterizing the microstate along with their bulk conservation equations. The central rheological problem of the relation between the bulk stress and the bulk strain rate for force-free particles will be the main subject of the paper. The same procedure is applicable to the remaining relations as will be shown for the sedimentation velocity of heavy particles and the permeability of a fixed bed.

The detailed calculation of the relation between the bulk stress and bulk strain rate is left to the four following sections. The remainder of this section addresses the problem of averaging the constitutive law, which is differently defined in the two phases. The laws in the two phases may be conveniently combined into one as

$$\boldsymbol{\sigma}(\mathbf{x}) = -p(\mathbf{x})\mathbf{I} + 2\mu \mathbf{e}(\mathbf{x}) + \mathbf{s}(\mathbf{x}, \mathscr{C}) \quad \text{for all} \quad \mathbf{x}, \tag{2.4}$$

where **s** is a generalized function which is zero if **x** is a point in the fluid while if **x** is in a particle **s** is the extra stress above the value given by the fluid law applied at **x**. While the particle extra stress **s** will be specified by a constitutive relation for the particles, its value will depend ultimately on the full configuration of the particles  $\mathscr{C}$  through the complete solution at the microscale of the boundary-value problem. The constitutive relations once combined into the single equation (2.4) are easily averaged:

$$\langle \boldsymbol{\sigma} \rangle (\mathbf{x}) = -\langle p \rangle (\mathbf{x}) \mathbf{I} + 2\mu \langle \mathbf{e} \rangle (\mathbf{x}) + \langle \mathbf{s} \rangle (\mathbf{x}).$$
 (2.5)

The bulk stress exceeds the pure solvent law by the particle contribution  $\langle \mathbf{s} \rangle$ . The bulk pressure field  $\langle p \rangle$  is locally indeterminate and associated with the bulk solenoidal constraint

$$\nabla \cdot \langle \mathbf{u} \rangle = 0. \tag{2.6}$$

The preceding manipulations have correctly exposed the need in the subsequent calculations to average quantities only within the particles, because s vanishes in the fluid. To express further this dependence on the particle interiors, consider s to be non-zero at x because it lies in a particle centred at  $x_1$ . Then

$$\langle \mathbf{s} \rangle \left( \mathbf{x} \right) = \int_{|\mathbf{x}_1 - \mathbf{x}| \leqslant a} \langle \mathbf{s} \rangle \left( \mathbf{x} | \mathbf{x}_1 \right) P(\mathbf{x}_1) \, dV_1, \tag{2.7}$$

where the integral is performed over the finite volume such that  $\mathbf{x}$  can lie in a particle centred at  $\mathbf{x}_1$ ,  $P(\mathbf{x}_1)$  is the probability density function for a single particle being centred at  $\mathbf{x}_1$ , and  $\langle \mathbf{s} \rangle \langle \mathbf{x} | \mathbf{x}_1 \rangle$  is the particle extra stress at  $\mathbf{x}$  averaged over the subclass of realizations which have a particle centred at  $\mathbf{x}_1$ . If there are several species of particles, e.g. of different size, shape, orientation or constitution, then the single vector  $\mathbf{x}_1$  for the particle centre should be extended to label the species, as well as the position. Such a generalization will be suppressed.

At this stage it is appropriate to restrict the suspension to be locally homogeneous, by which is meant that all the probability density functions vary under translations (but not necessarily under configurational alterations) only on a bulk length scale which greatly exceeds the microstructural length scale. The microstructural length scale is the one on which a relevant quantity like  $\langle \mathbf{u} \rangle \langle \mathbf{x} | \mathbf{x}_1 \rangle$  varies with  $\mathbf{x} - \mathbf{x}_1$ . In many cases the microstructural length scale is found a posteriori to be simply the particle size. In the problem of the permeability of the fixed bed, however, it is found to be a larger interaction length. Present experience indicates rather surprisingly that in dilute suspensions the particle separation is not a relevant microstructural length scale unless there is a dependence on it in the probability density functions. Thus the particle separation need not be smaller than the bulk length scale when using ensemble averaging, whereas it must in the volume-averaging approach. Of course in a practical application where one would like the theoretical ensemble average to approximate to a single experimental measurement which smoothed the data over a stationary dimension (volume, surface, length or time), then the bulk scale should exceed the sample size and this in turn should exceed the particle separation.

With the assumption of local homogeneity, the probability function  $P(\mathbf{x}_1)$  in (2.7) may be regarded as a constant, equal to the value at  $\mathbf{x}$  about which  $\mathbf{x}_1$  ranges by the particle size, and may thus be taken outside the integral. Corrections for some slight inhomogeneity could be made by replacing the constant  $P(\mathbf{x})$  with a few further terms of the Taylor series for  $P(\mathbf{x}_1)$  taken about  $\mathbf{x}_1 = \mathbf{x}$ . As this leads to the scantily studied area of non-local rheology, such corrections will always be neglected. The local homogeneity also allows a small translation of  $\mathbf{x} - \mathbf{x}_1$  in the two arguments of the conditionally averaged particle extra stress  $\langle \mathbf{s} \rangle (\mathbf{x} + (\mathbf{x} - \mathbf{x}_1) | \mathbf{x})$ . Thus the particle contribution to the bulk stress becomes

$$\langle \mathbf{s} \rangle \left( \mathbf{x} \right) = P(\mathbf{x}) \int_{|\mathbf{x}' - \mathbf{x}| \leq a} \langle \mathbf{s} \rangle \left( \mathbf{x}' | \mathbf{x} \right) dV', \qquad (2.8)$$

where the integral extends over all points  $\mathbf{x}'$  within a particle centred at  $\mathbf{x}$ . This expression shows that the ensemble average reduces to a volume average when there is local homogeneity. Batchelor (1970) showed that the last volume integral could be replaced by a surface integral, which is more useful for rigid particles. Using the

low-Reynolds-number condition and the divergence theorem, he obtained for force free particles ( $\mathbf{f} = 0$  inside the particles)

$$\int_{|\mathbf{x}'-\mathbf{x}| \leq a} \langle \mathbf{s} \rangle (\mathbf{x}'|\mathbf{x}) dV' = \text{an isotropic term of no interest} + \oint_{|\mathbf{x}'-\mathbf{x}|=a} \{ \langle \mathbf{\sigma} \rangle (\mathbf{x}'|\mathbf{x}) \cdot \mathbf{n}'\mathbf{x}' - \mu[\langle \mathbf{u} \rangle (\mathbf{x}'|\mathbf{x}) \mathbf{n}' + \mathbf{n}' \langle \mathbf{u} \rangle (\mathbf{x}'|\mathbf{x})] \} dS', \quad (2.9)$$

the square-bracketed term making no contribution for rigid particles. Hereafter all field quantities  $\mathbf{u}, \mathbf{e}, \mathbf{\sigma}, p$  and  $\mathbf{s}$  occur as ensemble averaged quantities, perhaps with some particles remaining fixed. The unnecessary averaging signs will therefore be dropped.

### 3. Bulk stress in the dilute limit

The expression for the bulk stress derived in the preceding section, (2.5) with (2.7), does not represent a closed description of the bulk suspension because the extra field  $\mathbf{s}(\mathbf{x}|\mathbf{x}_1)$  was introduced. In this section a closed description will be derived which is asymptotically correct when the suspension is dilute. The description will be illustrated with a calculation of the bulk stress for small, rigid, spherical particles free of external forces and couples. In more complicated suspensions, the bulk stress can depend not only on the bulk strain rate but also on a microstructural variable which is governed by further equations for the microstructural dynamics. Such complications have been avoided to expose more clearly the general approach.

So far two restrictions have been placed on the suspension, restrictions of inertialess particle dynamics and local homogeneity. No correction terms for these effects will be considered. While at this stage the problem is linear and statistically stationary, it is still intractable. The random geometry of the particle positions is too difficult to analyse. A further approximation of low particle concentration is now made. The particles are required to be widely spaced compared with their overall length. For similar sized particles not of extreme shape, this amounts to keeping the particle volume fraction c small. For particles of extreme shape the volume fraction must be made much smaller, so that the volume fraction of the smallest spherical envelopes around each particle is small. As well as being dilute the suspension is additionally required to be reasonably random; in particular, the probability of n different particles being in a specific configuration should be  $O(c^n)$  and should vary between different configurations only with the length scale of the particle size and nothing larger, for example the particle separation.

The particle contribution to the bulk stress  $\mathbf{s}(\mathbf{x})$  requires, according to (2.7), the derivation of the average particle stress at  $\mathbf{x}$  conditioned to one particle being centred at  $\mathbf{x}_1$ ,  $\mathbf{s}(\mathbf{x}|\mathbf{x}_1)$ . There are two alternative routes to this conditionally averaged quantity. The approach followed by Batchelor is to find the quantity for each configuration  $\mathbf{s}(\mathbf{x}|\mathscr{C})$  by some plausible arguments about negligible effects in the dilute limit, and then to make the conditional average using if necessary a renormalization of any convergence difficulties. The other approach, suggested by Brinkman and followed here, is to derive equations governing the mean fields like  $\mathbf{s}(\mathbf{x}|\mathbf{x}_1)$ , and then to solve these new equations neglecting terms which become small in the dilute limit.

The equations governing the mean field at  $\mathbf{x}$  conditioned to a particle being centred at  $\mathbf{x}_1$  are derived following the same process used in the preceding section for the unconditioned mean fields: the conservation law and the constitutive relation are simply conditionally averaged. Averaging the momentum conservation law (2.1) for inertialess and force-free particle dynamics gives

$$\nabla \cdot \boldsymbol{\sigma}(\mathbf{X}|\mathbf{X}_1) = 0. \tag{3.1}$$

The incompressibility constraint (2.2) becomes

$$\nabla \cdot \mathbf{u}(\mathbf{x}|\mathbf{x}_1) = 0. \tag{3.2}$$

The derivation of these last two equations has exploited the power of the ensemble average over other types of average. Only the ensemble average may be freely commuted with the space and time differential operators. Conditionally averaging the constitutive equations in the combined form (2.4) gives

$$\boldsymbol{\sigma}(\mathbf{x}|\mathbf{x}_1) = -p(\mathbf{x}|\mathbf{x}_1) \,\mathbf{I} + 2\mu \mathbf{e}(\mathbf{x}|\mathbf{x}_1) + \mathbf{s}(\mathbf{x}|\mathbf{x}_1), \tag{3.3}$$

where, using an equation similar to (2.7), the particle extra stress may be expressed as

$$\mathbf{s}(\mathbf{x}|\mathbf{x}_1) = \int_{|\mathbf{x}_2 - \mathbf{x}| \leq a} \mathbf{s}(\mathbf{x}|\mathbf{x}_1, \mathbf{x}_2) P(\mathbf{x}_2|\mathbf{x}_1) dV_2.$$
(3.4)

In this last expression the integral is performed over the finite volume such that  $\mathbf{x}$  lies in a particle centred at  $\mathbf{x}_2$ ,  $P(\mathbf{x}_2|\mathbf{x}_1)$  is the probability density function that there is a particle centred at  $\mathbf{x}_2$  given that there is one centred at  $\mathbf{x}_1$ , and  $\mathbf{s}(\mathbf{x}|\mathbf{x}_1,\mathbf{x}_2)$  is the particle extra stress at  $\mathbf{x}$  averaged over that subclass of the full ensemble which has particles centred at  $\mathbf{x}_1$  and at  $\mathbf{x}_2$ . When  $\mathbf{x}_1 = \mathbf{x}_2$ , there is certainly a particle centred at  $\mathbf{x}_2$  given that there is one centred at  $\mathbf{x}_1$ . (The definition of  $P(\mathbf{x}_2|\mathbf{x}_1)$  used above does not require the particles at  $\mathbf{x}_1$  and  $\mathbf{x}_2$  to be different.) The effect of this sole delta function in  $P(\mathbf{x}_2|\mathbf{x}_1)$  is to make sure that  $\boldsymbol{\sigma}(\mathbf{x}|\mathbf{x}_1)$  is given by the particle constitutive relation when  $\mathbf{x}$  lies within the particle centred at  $\mathbf{x}_1$ . If an alternative definition of  $P(\mathbf{x}_2|\mathbf{x}_1)$  had been used with a requirement that the particles at  $\mathbf{x}_1$  and  $\mathbf{x}_2$  were different, then the right-hand side of (3.4) would have to be replaced by two expressions, one valid when  $\mathbf{x}$  lies inside the particle at  $\mathbf{x}_1$  and one valid when it is outside.

In the attempt to close the bulk equations (2.3) and (2.5)-(2.7) by deriving the equations (3.1)-(3.4) which govern the conditionally averaged fields with one particle fixed, a field  $\mathbf{s}(\mathbf{x}|\mathbf{x}_1,\mathbf{x}_2)$  averaged with two particles fixed has been introduced. Repeating the process for this two-particles-fixed field introduces a three-particles-fixed field; continuing, the complete infinite hierarchy can be produced. A truncation is needed and can be achieved rationally using the diluteness. From the 'reasonable-randomness' assumption, a typical conditional probability density such as  $P(\mathbf{x}_n|\mathbf{x}_1,...,\mathbf{x}_{n-1})$  multiplied by the volume of a particle is of the order of the small volume fraction c so long as all the n particles are different. A series of levels of approximation is produced by neglecting the small O(c) particle extra stress terms at different stages in the infinite hierarchy. At the very lowest level, the  $\mathbf{s}(\mathbf{x})$  term in (2.5) is neglected because it is O(c) smaller than the other terms in (2.5) through the  $P(\mathbf{x}_1)$  factor in (2.7). This level of approximation says that the bulk suspension

ultimately behaves like the pure solvent as  $c \to 0$ . At the next level of approximation, the  $\mathbf{s}(\mathbf{x}|\mathbf{x}_1)$  term in (3.3) is neglected when  $\mathbf{x}$  does not lie in the particle at  $\mathbf{x}_1$ , because it is O(c) smaller than the other terms in (3.3) through the  $P(\mathbf{x}_2|\mathbf{x}_1)$  factor in (3.4). This level of approximation, which brings in the first particle contribution to the bulk stress, will be solved in the remainder of this section. The following section proceeds to one further level in which the first particle interactions enter. There is no guarantee that the small neglected terms in the equations will have a small effect. Such a hope would be supported (but not proven) by continuing to the higher approximations. In fact, in the permeability problem, small O(c) terms do produce a larger but still small,  $O(c^{\frac{1}{2}})$  effect.

The problem for the bulk stress correct to O(c) has been reduced to the following. The values of  $\mathbf{s}(\mathbf{x}|\mathbf{x}_1)$  when  $\mathbf{x}$  lies in the particle at  $\mathbf{x}_1$  must be found to O(1) so that the integral (2.7) can be evaluated. The averaged fields  $\boldsymbol{\sigma}(\mathbf{x}|\mathbf{x}_1)$  and  $\mathbf{e}(\mathbf{x}|\mathbf{x}_1)$  are found by solving (3.1) and (3.2) together with the approximation

$$\sigma(\mathbf{x}|\mathbf{x}_1) = \begin{pmatrix} \text{particle} \\ \text{fluid} \end{pmatrix} \text{law when } \mathbf{x} \text{ is } \begin{pmatrix} \text{inside} \\ \text{outside} \end{pmatrix} \text{ particle at } \mathbf{x}_1, \quad (3.5)$$

which includes an O(c) negligible error outside the particle. There are the usual boundary conditions on the surface of the particle at  $\mathbf{x}_1$ . Finally a connexion is made between the conditionally averaged fields and the bulk state through the application of the boundary condition on  $\mathbf{u}(\mathbf{x}|\mathbf{x}_1)$  when  $\mathbf{x}$  is far from the particle at  $\mathbf{x}_1$ . In order that the conditionally averaged state can sum to the bulk state, it is necessary to impose

$$\mathbf{u}(\mathbf{x}|\mathbf{x}_1) \sim \mathbf{u}(\mathbf{x}) \quad \text{as} \quad |\mathbf{x} - \mathbf{x}_1| \to \infty.$$
 (3.6)

This condition assumes that the equations governing  $\mathbf{u}(\mathbf{x}|\mathbf{x}_1)$  allow it to settle down to the asymptotic state of  $\mathbf{u}(\mathbf{x})$ , the suspension is reasonably random and is of infinite extent compared with the finite volume of the particle. For the bulk-stress problem, the appropriate form of the infinity condition (3.6) is found using the local homogeneity as

when

$$\begin{aligned} \mathbf{u}(\mathbf{x}|\mathbf{x}_1) &\sim \mathbf{u}(\mathbf{x}_1) + (\mathbf{x} - \mathbf{x}_1) \cdot [\boldsymbol{\omega}(\mathbf{x}_1) + \mathbf{e}(\mathbf{x}_1)], \\ &|\mathbf{x} - \mathbf{x}_1| \geqslant \text{particle size}, \end{aligned}$$
(3.7)

where  $\boldsymbol{\omega}$  and  $\boldsymbol{e}$  are the bulk vorticity and strain rate. This completes the specification of the problem for the bulk stress to O(c).

The details of the calculation for the bulk stress are well known and are only presented briefly for rigid spheres of radius a which are not pre-stressed. The solution for the velocity field  $\mathbf{u}(\mathbf{x}|\mathbf{x}_1)$  from (3.1), (3.2), (3.5) and (3.7) is, with  $\mathbf{r} = \mathbf{x} - \mathbf{x}_1$ ,

$$\mathbf{u}(\mathbf{x}|\mathbf{x}_1) = \mathbf{u}(\mathbf{x}_1) + \mathbf{r} \cdot \boldsymbol{\omega}(\mathbf{x}_1) \text{ for } r \leq a,$$

and  $\mathbf{u}(\mathbf{x}|\mathbf{x}_1) = \mathbf{u}(\mathbf{x}_1) + \mathbf{r} \cdot \mathbf{\omega}(\mathbf{x}_1)$  $+ \mathbf{e}(\mathbf{x}_1) : \{\mathbf{r}\mathbf{l}(1 - (a/r)^5) + (5/2a^2) \mathbf{rrr}((a/r)^5 - (a/r)^7)\}$  for  $r \ge a$ .

The stress in the rigid sphere is found to be

$$\boldsymbol{\sigma}(\mathbf{X}|\mathbf{X}_1) = -p(\mathbf{X}_1)\,\mathbf{I} + 5\mu\mathbf{e}(\mathbf{X}_1) + O(c),$$

so that the particle extra stress is simply

$$\mathbf{s}(\mathbf{x}|\mathbf{x}_1) = 5\mu\mathbf{e}(\mathbf{x}_1) + O(c), \quad r \leq a.$$

This is now substituted into (2.7), where by local homogeneity  $\mathbf{e}$  and  $P(\mathbf{x}_1)$  evaluated at  $\mathbf{x}_1$  can be approximated by their values at  $\mathbf{x}$ . Thus Einstein's (1906) result

$$\boldsymbol{\sigma}(\mathbf{x}) = -p(\mathbf{x})\,\mathbf{I} + 2\mu(1 + \frac{5}{2}c)\,\mathbf{e}(\mathbf{x})$$

correct to O(c) is recovered.

#### 4. Towards the bulk stress to $O(c^2)$

The preceding two sections have been addressed to the first fundamental problem in the study of suspensions, which is the description of the bulk suspension. Very familiar material has been restated with the simple change to an ensemble average. The advantage of the ensemble-average approach will now become apparent as attention is transferred to the second fundamental problem for suspensions of analysing the first effects of hydrodynamic interactions. No longer is the second problem tackled by a completely different approach to the first, but the same approximation process used for the first problem is continued just one further stage for the second.

To find the bulk stress  $\sigma$  in (2.5) correct to  $O(c^2)$  it is necessary to evaluate the integral in (2.7) with  $\mathbf{s}(\mathbf{x}|\mathbf{x}_1)$  known correctly to O(c) when  $\mathbf{x}$  lies in the particle centred at  $\mathbf{x}_1$ . The one-particle-fixed fields are found by solving the boundary-value problem (3.1)-(3.3) and (3.7), for which it is necessary to evaluate the integral in (3.4) with  $\mathbf{s}(\mathbf{x}|\mathbf{x}_1,\mathbf{x}_2)$  known correctly to O(1) when  $\mathbf{x}$  lies in the particle centred at  $\mathbf{x}_2$  given there is a different particle centred at  $\mathbf{x}_1$ . The two-particles-fixed fields are found by solving a similar boundary-value problem:

$$\nabla \cdot \boldsymbol{\sigma}(\mathbf{x}|\mathbf{x}_1,\mathbf{x}_2) = 0, \quad \nabla \cdot \mathbf{u}(\mathbf{x}|\mathbf{x}_1,\mathbf{x}_2) = 0,$$

with the usual boundary conditions on the surfaces of the two particles, a boundary condition far from the two particles given by

 $\mathbf{u}(\mathbf{x}|\mathbf{x}_1,\mathbf{x}_2) \sim \mathbf{u}(\mathbf{x}_1) + (\mathbf{x} - \mathbf{x}_1) \cdot (\mathbf{\omega}(\mathbf{x}_1) + \mathbf{e}(\mathbf{x}_1)), \text{ when } |\mathbf{x} - \mathbf{x}_1|, |\mathbf{x} - \mathbf{x}_2| \ge a,$ and with an averaged constitutive equation given with adequate accuracy by

$$\boldsymbol{\sigma}(\mathbf{x}|\mathbf{x}_1,\mathbf{x}_2) = \begin{pmatrix} \text{particle} \\ \text{fluid} \end{pmatrix} \text{law when } \mathbf{x} \text{ is } \begin{pmatrix} \text{inside either} \\ \text{outside both} \end{pmatrix} \text{ of the particles at } \mathbf{x}_1 \text{ and } \mathbf{x}_2.$$

The error in the constitutive equation occurs outside both the particles and is O(c) small. At the present level of approximation the mean fields with two particles fixed are those for two particles surrounded by pure solvent. The required solution of the Stokes flow past two spheres is well known and has been reviewed by Batchelor & Green (1972b).

With the two-particles-fixed fields known at the accuracy required, the integral in (3.4) can be evaluated for  $\mathbf{s}(\mathbf{x}|\mathbf{x}_1)$ . Rearranging (3.1) and (3.3) then gives a momentum equation at the level of one particle held fixed: for  $\mathbf{x}$  inside the particle at  $\mathbf{x}_1$ , i.e.  $|\mathbf{x} - \mathbf{x}_1| \leq a$ ,

 $\nabla \cdot \boldsymbol{\sigma}(\mathbf{x}|\mathbf{x}_1) = 0,$ 

and, for **x** outside the particle at  $\mathbf{x}_1$ ,  $|\mathbf{x} - \mathbf{x}_1| \ge a$ ,

$$-\nabla p(\mathbf{x}|\mathbf{x}_1) + \mu \nabla^2 \mathbf{u}(\mathbf{x}|\mathbf{x}_1) = -\nabla \cdot \int_{|\mathbf{x}_2 - \mathbf{x}| \leq a} \mathbf{s}(\mathbf{x}|\mathbf{x}_1, \mathbf{x}_2) P(\mathbf{x}_2|\mathbf{x}_1) dV_2.$$
(4.1)

The known right-hand side of (4.1) produces the first corrections to the bulk stress due to hydrodynamic interactions between the particles in the suspension.

There is an alternative form for (4.1) which is more convenient for rigid particles because it uses data from the two-particle problem evaluated only on the surface of the particles. To derive the alternative form, the function  $\mathbf{s}(\mathbf{x}|\mathbf{x}_1,\mathbf{x}_2)$  inside the particle at  $\mathbf{x}_2$  is first replaced by an integral of a generalized function

$$\mathbf{s}(\mathbf{x}|\mathbf{x}_1,\mathbf{x}_2) = \int_{|\mathbf{x}'-\mathbf{x}_2| \leq a} dV' \mathbf{s}(\mathbf{x}'|\mathbf{x}_1,\mathbf{x}_2) \,\delta(\mathbf{x}'-\mathbf{x}), \quad |\mathbf{x}-\mathbf{x}_2| \leq a.$$

Thus the right-hand side of (4.1) may be written in  $|\mathbf{x} - \mathbf{x}_1| \ge a$  as

$$-\int_{|\mathbf{x}_2-\mathbf{x}_1|\geq 2a} dV_2 P(\mathbf{x}_2|\mathbf{x}_1) \int_{|\mathbf{x}'-\mathbf{x}_2|\leqslant a} dV' \mathbf{s}(\mathbf{x}'|\mathbf{x}_1,\mathbf{x}_2) \cdot \nabla \delta(\mathbf{x}'-\mathbf{x}).$$

Using the definition of the particle extra stress as the stress above the fluid law, the zero divergence of  $\sigma(\mathbf{x}|\mathbf{x}_1,\mathbf{x}_2)$  and the divergence theorem, we obtain

$$\begin{split} &\int_{|\mathbf{x}_{2}-\mathbf{x}_{1}|\geq 2a} dV_{2} P(\mathbf{x}_{2}|\mathbf{x}_{1}) \left\{ \oint_{|\mathbf{x}'-\mathbf{x}_{2}|=a} dS' \boldsymbol{\sigma}(\mathbf{x}'|\mathbf{x}_{1},\mathbf{x}_{2}) \cdot \mathbf{n}' \boldsymbol{\delta}(\mathbf{x}'-\mathbf{x}) \right. \\ &\left. + \int_{|\mathbf{x}'-\mathbf{x}_{2}|\leq a} dV' \left[ -p(\mathbf{x}'|\mathbf{x}_{1},\mathbf{x}_{2}) \nabla \boldsymbol{\delta}(\mathbf{x}'-\mathbf{x}) + 2\mu \mathbf{e}(\mathbf{x}'|\mathbf{x}_{1},\mathbf{x}_{2}) \cdot \nabla \boldsymbol{\delta}(\mathbf{x}'-\mathbf{x}) \right] \right\}. \end{split}$$

For rigid particles the last term in the volume integral contributes nothing to the value of that integral. Finally removing the generalized functions produces in  $|\mathbf{x} - \mathbf{x}_1| \ge a$ 

$$-\nabla \int_{|\mathbf{x}_{1}-\mathbf{x}_{1}|\geq 2a} p(\mathbf{x}|\mathbf{x}_{1},\mathbf{x}_{2}) P(\mathbf{x}_{2}|\mathbf{x}_{1}) dV_{2} + \oint_{|\mathbf{x}_{2}-\mathbf{x}|=a} \sigma(\mathbf{x}|\mathbf{x}_{1},\mathbf{x}_{2}) \cdot \mathbf{n}_{2} P(\mathbf{x}_{2}|\mathbf{x}_{1}) dS_{2}.$$
 (4.2)

The first term can be combined with the pressure term on the left-hand side of (4.1) and is of no consequence because it vanishes on the surface of the particle at  $\mathbf{x}_1$ . This alternative form for use with rigid particles is equivalent to that used by Howells (1974) (his (2.1) and (2.2)), presented in that paper with no formal derivation.

An immediately appealing method for finding the O(c) correction driven by the right-hand side of (4.1) is to solve separately for each fixed second particle at  $\mathbf{x}_2$  and then to sum over all the  $\mathbf{x}_2$  with a probability weighting. The alternative form (4.2) shows more clearly the linearity in  $P(\mathbf{x}_2|\mathbf{x}_1)$  of the right-hand side, a linearity which must therefore extend to the solution of the boundary-value problem. The suggested method for finding the required  $\mathbf{s}(\mathbf{x}|\mathbf{x}_1)$  amounts to nothing more than commuting the inversion operator with the probability sum. Each fixed particle at  $\mathbf{x}_2$  forces in (4.1) the two-particle solution which is already known. Thus the method yields

$$\mathbf{s}(\mathbf{x}|\mathbf{x}_1) = \int_{|\mathbf{x}_2 - \mathbf{x}_1| \ge 2a} \mathbf{s}(\mathbf{x}|\mathbf{x}_1, \mathbf{x}_2) P(\mathbf{x}_2|\mathbf{x}_1) dV_2.$$
(4.3)

The advantage of the method is that it bypasses the full solution of the mean fields with one particle fixed, e.g. including detailed knowledge of  $\mathbf{u}(\mathbf{x}|\mathbf{x}_1)$  to O(c). Unfortunately this method does not work: it is not possible to commute the inversion operator with the probability sum. The trouble is that the infinite integral (4.3) does not converge. The origin of the non-convergence is in the treatment of the forcing in

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(4.1) when  $\mathbf{x}$  lies far from the particle at  $\mathbf{x}_1$ . Far from a particle at  $\mathbf{x}_1$ , a particle at  $\mathbf{x}_2$  acts as a constant force-dipole as it resists deforming with the suspension. The single force-dipole induces a stresslet velocity field. The suggested method finds the effects of the constant force-dipole density by summing the induced stresslet velocity fields. Owing to the long-range nature of the stresslets, this Green's function technique would only work if the strength of the force-dipoles decayed rapidly away from the particle at  $\mathbf{x}_1$ . The failure of the Green's function approach does not mean, however, that no solution exists to the averaged field equation (4.1), but merely than an alternative technique is required for solving the boundary-value problem.

Although the appealing method described above failed, the programme as originally outlined would have been successful. The trouble arose in an attempt to bypass the full  $\mathbf{u}(\mathbf{x}|\mathbf{x}_1)$  problem by combining the known  $\mathbf{s}(\mathbf{x}|\mathbf{x}_1,\mathbf{x}_2)$  with the probability weight  $P(\mathbf{x}_2|\mathbf{x}_1)$ . A constant distribution of force-dipoles has zero spatial gradient, and in particular zero divergence. Thus if the right-hand side of (4.1) had been evaluated in strict accordance with the approach, the troublesome terms would have not contributed to the forcing in (4.1). It should be noted that the unforced solutions to (4.1) are controlled by the boundary condition at infinity. Now the constant forcedipole density represents a stress distribution, so that it has no effect with an infinity condition on velocity, whereas there would be an effect with an infinity condition on stress.

Two successful methods for going directly from  $\mathbf{s}(\mathbf{x}|\mathbf{x}_1,\mathbf{x}_2)$  to the bulk stress to  $O(c^2)$  will be presented. The first method can also be used in the problem of the sedimentation of heavy particles, and generates Batchelor's (1972) convergent-integral expressions. The second method maintains to a further degree Brinkman's philosophy of averaged materials and is more powerful, being able to tackle additionally the problem of the permeability of a fixed bed. Both methods subtract the difficult state at infinity from the equation, i.e. roughly speaking the constant force-dipole density. The subtraction is chosen to be simple and to have an effect on the averaged field which can be easily calculated by an alternative technique to the inappropriate Green's function one. The difference between the subtracted state at infinity and the actual state at a point  $\mathbf{x}$  near the particle at  $\mathbf{x}_1$  is a remainder term which is designed to be convergently invertable by the Green's function technique. The two methods differ in their subtlety at detecting the state at infinity.

#### 5. The first renormalization<sup>†</sup>

The most convenient form of (4.1) for discussing the renormalization is, in  $|\mathbf{x} - \mathbf{x}_1| \ge a$ ,

$$-\nabla p_{f}(\mathbf{x}|\mathbf{x}_{1}) + \mu \nabla^{2} \mathbf{u}(\mathbf{x}|\mathbf{x}_{1}) = \int_{|\mathbf{x}_{2}-\mathbf{x}_{1}| \geq 2a} dV_{2} P(\mathbf{x}_{2}|\mathbf{x}_{1}) \\ \times \oint_{|\mathbf{x}'-\mathbf{x}_{3}|=a} dS' \boldsymbol{\sigma}(\mathbf{x}'|\mathbf{x}_{1},\mathbf{x}_{2}) \cdot \mathbf{n}' \delta(\mathbf{x}'-\mathbf{x}), \quad (5.1)$$

where  $p_f$  is the average pressure in the fluid alone, counting zero pressure when the realization has x inside a particle. The new form is suited to considering the individual contributions from each  $x_2$ -particle.

<sup>†</sup> The word 'renormalization' is used in analogy with its use in quantum field theory.

When the  $\mathbf{x}_2$ -particle is far from the  $\mathbf{x}_1$ -particle, it is effectively an isolated particle in an unbounded solvent. The fluid velocity surrounding the  $\mathbf{x}_2$ -sphere is then

$$\mathbf{v}(\mathbf{x}) = \mathbf{u}(\mathbf{x}_1) + (\mathbf{x} - \mathbf{x}_1) \cdot \nabla \mathbf{u}(\mathbf{x}_1) - \frac{5}{6}a^3(1 + \frac{1}{10}a^2\nabla_2^2) (\mathbf{e}(\mathbf{x}_1) \cdot \nabla_2) \cdot \mathbf{I}(\mathbf{x} - \mathbf{x}_2),$$

where  $\boldsymbol{\mathscr{I}}$  is the Stokeslet interaction tensor

$$\mathscr{I}(\mathbf{r}) = r^{-1}\mathbf{I} + r^{-3}\mathbf{r}\mathbf{r}.$$

The disturbance velocity field due to the  $\mathbf{x}_2$ -sphere is governed by Stokes' equation applied everywhere with a singularity in the forbidden region  $|\mathbf{x} - \mathbf{x}_2| < a$ :

$$-\nabla p + \mu \nabla^2 \mathbf{v} = 8\pi \mu_6^5 a^3 (1 + \frac{1}{10} a^2 \nabla_2^2) \,(\mathbf{e}(\mathbf{x}_1) \,.\, \nabla_2) \,\delta(\mathbf{x} - \mathbf{x}_2).$$

At the centre of the  $\mathbf{x}_2$ -sphere there is a force-dipole (or stresslet) and a degenerate octupole.

The subtraction from the integral on the right-hand side of (5.1) for the first renormalization is a uniform distribution of force dipoles and octupoles with values corresponding to an isolated particle in the unbounded solvent and with density  $P(\mathbf{x}_1)$  in the excluded shell  $|\mathbf{x} - \mathbf{x}_1| \ge 2a$ . Thus in  $|\mathbf{x} - \mathbf{x}_1| \ge a$ 

$$-\nabla p_{f}(\mathbf{x}|\mathbf{x}_{1}) + \mu \nabla^{2} \mathbf{u}(\mathbf{x}|\mathbf{x}_{1}) = 5c\mu \int_{|\mathbf{x}_{2}-\mathbf{x}_{1}| \ge 2a} dV_{2} \left(1 + \frac{1}{10}a^{2}\nabla_{2}^{2}\right) \mathbf{e}(\mathbf{x}_{1}) \cdot \nabla_{2} \delta(\mathbf{x}-\mathbf{x}_{2}) + \int_{|\mathbf{x}_{2}-\mathbf{x}_{1}| \ge 2a} dV_{2} \left\{P(\mathbf{x}_{2}|\mathbf{x}_{1}) \oint_{|\mathbf{x}'-\mathbf{x}_{2}|=a} dS' \boldsymbol{\sigma}(\mathbf{x}'|\mathbf{x}_{1},\mathbf{x}_{2}) \cdot \mathbf{n}' \delta(\mathbf{x}'-\mathbf{x}) - P(\mathbf{x}_{1}) \frac{20\pi\mu a^{3}}{3} \left(1 + \frac{1}{10}a^{2}\nabla_{2}^{2}\right) \left(\mathbf{e}(\mathbf{x}_{1}) \cdot \nabla_{2}\right) \delta(\mathbf{x}-\mathbf{x}_{2})\right\}.$$
(5.2)

The two contributions to the  $O(c^2)$  bulk stress correction, one from the first integral in (5.2) of constant multipoles and the other from the remainder second integral, will now be calculated separately.

The volume distribution of constant dipoles and octupoles can be replaced by a surface distribution of monopoles and quadrupoles

$$-5c\mu \oint_{|\mathbf{x}_2-\mathbf{x}_1|=2a} dS_2 \mathbf{e}(\mathbf{x}_1) \cdot \mathbf{n}_2 (1+\frac{1}{10}a^2\nabla_2^2) \,\delta(\mathbf{x}-\mathbf{x}_2).$$

The surface integral at infinity is rejected using the boundary conditions at infinity on the velocity. This dubious move is the weak point of the first method. It is, however, correct and will be explained and justified by the second renormalization. The velocity field induced by the surface distribution of multipoles is

$$\mathbf{v} = \frac{5}{6}a^{3}P(\mathbf{x}_{1}) \oint_{|\mathbf{x}_{2}-\mathbf{x}_{1}|=2a} dS_{2} \mathbf{e}(\mathbf{x}_{1}) \cdot \mathbf{n}_{2}(1 + \frac{1}{10}a^{2}\nabla_{2}^{2}) \cdot \mathscr{I}(\mathbf{x}-\mathbf{x}_{2}).$$
(5.3)

This velocity is singular at  $|\mathbf{x} - \mathbf{x}_1| = 2a$ . All the singularity is, however, cancelled by an identical singularity in the remainder term, and moreover only the solution near  $|\mathbf{x} - \mathbf{x}_1| = a$  will be needed in calculating the bulk stress contribution by (2.9). The velocity field (5.3) results from the multipoles acting in the infinite solvent alone. In order to maintain the rigid boundary conditions on the particle centred at  $\mathbf{x}_1$ , an extra image term is also required. The image term in the  $\mathbf{u}(\mathbf{x}|\mathbf{x}_1)$  field can fortunately be bypassed by applying an appropriate Faxén law.

The Faxén law, as derived by Batchelor & Green (1972a), states that the contribution to the bulk stress integral

$$\oint_{|\mathbf{x}-\mathbf{x}_1|=a} dS \boldsymbol{\sigma} \cdot \mathbf{n} \mathbf{x}$$

for a free rigid sphere placed in a pre-existing unbounded flow  $\mathbf{v}(\mathbf{x})$  is

$$\frac{10}{3}\pi a^3\mu(1+\frac{1}{10}a^2\nabla^2)\left(\nabla\mathbf{v}+\nabla\mathbf{v}^{\mathrm{T}}\right)$$

evaluated at the sphere centre  $\mathbf{x} = \mathbf{x}_1$ . Note the similarity between Faxén's law and the velocity field outside an isolated particle given at the beginning of this section. The similarity is rooted in the reciprocal theorem for Stokes flow.

Applying the Faxén law to the velocity field (5.3) yields a bulk stress contribution

$$\begin{split} P(\mathbf{x}_1) \frac{10}{3} \pi a^3 \mu (1 + \frac{1}{10} a^2 \nabla^2) \frac{5}{6} a^3 P(\mathbf{x}_1) \oint_{|\mathbf{x}_2 - \mathbf{x}_1| = 2a} dS_2 \mathbf{e} \cdot \mathbf{n}_2 \\ & \times (1 + \frac{1}{10} a^2 \nabla_2^2) \cdot (\nabla \mathscr{I} + \nabla \mathscr{I}^T) \quad \text{at} \quad \mathbf{x} = \mathbf{x}_1. \end{split}$$

This can be evaluated by straightforward manipulation and it is  $5\mu c^2 \mathbf{e}$ . The  $\nabla^2$  term does not in fact contribute to the integral.

The correction velocity field forced by the remainder integral in (5.2) is, in

$$\begin{aligned} |\mathbf{x} - \mathbf{x}_1| &\ge a, \\ \int_{|\mathbf{x}_1 - \mathbf{x}_1| &\ge 2a} dV_2 \{ P(\mathbf{x}_2 | \mathbf{x}_1) \mathbf{U}_2(\mathbf{x}; \mathbf{x}_1, \mathbf{x}_2) + P(\mathbf{x}_1) \frac{5}{6} a^3 (1 + \frac{1}{10} a^2 \nabla_2^2) \left( \mathbf{e}(\mathbf{x}_1) \cdot \nabla_2 \right) \cdot \mathscr{I}(\mathbf{x} - \mathbf{x}_2) \} \end{aligned}$$

plus an extra image term associated with the second part which maintains the rigid boundary conditions on the  $\mathbf{x}_1$ -particle. The  $\mathbf{U}_2$  velocity field is the additional velocity in the solution of the pure solvent flow around the two particles at  $\mathbf{x}_1$  and  $\mathbf{x}_2$  in excess of the pure solvent flow around the single particle at  $\mathbf{x}_1$ . This velocity field will contribute to the bulk stress integral (2.9) at each fixed  $\mathbf{x}_2$  a term  $\mathbf{S}_2(\mathbf{x}_1, \mathbf{x}_2)$ , where again  $\mathbf{S}_2$  is the excess of the quantity for two particles above the value with just one isolated particle. At each fixed  $\mathbf{x}_2$  the second part of the above velocity integrand contributes to the bulk stress integral (2.9) according to the Faxén law

$$\begin{split} \frac{10}{3}\pi a^3 \mu (1 + \frac{1}{10}a^2\nabla^2) \frac{5}{6}a^3 P(\mathbf{x}_1) \left(1 + \frac{1}{10}a^2\nabla_2^2\right) (\mathbf{e}(\mathbf{x}_1) \cdot \nabla_2) \cdot (\nabla \mathscr{I} + \nabla \mathscr{I}^T), \\ & \text{evaluated at} \quad \mathbf{x} = \mathbf{x}_1, \\ & = -\frac{20}{3}\pi a^3 \mu P(\mathbf{x}_1) \left(1 + \frac{1}{10}a^2\nabla^2\right) \mathbf{E}_1(\mathbf{x};\mathbf{x}_2) \quad \text{evaluated at} \quad \mathbf{x} = \mathbf{x}_1. \end{split}$$

 $\mathbf{E}_1(\mathbf{x}; \mathbf{x}_2)$  is the strain rate at  $\mathbf{x}$  in excess of the bulk strain rate due to a single sphere placed at  $\mathbf{x}_2$  in unbounded pure solvent. Gathering together the two parts of the remainder, their contribution to the bulk stress integral is

$$\int_{|\mathbf{x}_1-\mathbf{x}_2| \ge 2a} dV_2 \{ P(\mathbf{x}_2 | \mathbf{x}_1) \, \mathbf{S}_2(\mathbf{x}_1, \mathbf{x}_2) - 5c\mu[(1 + \frac{1}{10}a^2\nabla^2) \, \mathbf{E}_1(\mathbf{x} ; \mathbf{x}_2)]_{\mathbf{x}=\mathbf{x}_1} \}.$$

The Laplacian term can be subtracted leaving an absolutely convergent integral because  $\nabla^2 \mathbf{E}_1 = O(|\mathbf{x} - \mathbf{x}_2|^{-5})$ . The integral contribution of the Laplacian can then be seen to vanish, by first performing the integral over a sphere  $|\mathbf{x}_2 - \mathbf{x}_1| = \text{constant}$ .

The final form for the bulk stress to  $O(c^2)$  as derived by the first renormalization is

$$\sigma(\mathbf{x}) = -p(\mathbf{x}) \mathbf{I} + 2\mu \mathbf{e}(\mathbf{x}) + 5\mu c \mathbf{e}(\mathbf{x}) + 5\mu c^2 \mathbf{e}(\mathbf{x}) + P(\mathbf{x}) \int_{|\mathbf{x}_1 - \mathbf{x}| \ge 2a} dV_2 \{ P(\mathbf{x}_2 | \mathbf{x}) \mathbf{S}_2(\mathbf{x}, \mathbf{x}_2) - 5\mu c \mathbf{E}_1(\mathbf{x}; \mathbf{x}_2) \}.$$
 (5.4)

This result can be immediately identified with Batchelor & Green's (1972*a*) result, in their (4.2). In their (4.1), they find the  $5\mu c^2 \mathbf{e}(\mathbf{x})$  term comes from an integral in  $|\mathbf{x}_2 - \mathbf{x}_1| < 2a$  of an extended definition of the second term in the integrand of the above (5.4).

So long as the two-particle probability density asymptotes to the one-particle probability according to  $P(\mathbf{x}_2|\mathbf{x}_1| = P(\mathbf{x}_1) + O(|\mathbf{x}_2 - \mathbf{x}_1|^{-2})$  for large separations of the two particles, then the integrand in (5.4) is  $O(|\mathbf{x}_2 - \mathbf{x}_1|^{-5})$  and so absolutely convergent. A full discussion of the binary probability problem was given by Batchelor & Green (1972*a*). They found for free spheres the steady distribution of  $P(\mathbf{x}_2|\mathbf{x}_1)$  asymptotes to  $P(\mathbf{x}_1)$  with an error  $O(|\mathbf{x}_2 - \mathbf{x}_1)^{-6})$ , and so it does not affect the integrand until  $O(|\mathbf{x} - \mathbf{x}_1|^{-9})$ . The leading term in the integrand is  $O(|\mathbf{x}_2 - \mathbf{x}_1|^{-5})$ . By first performing the surface integral on  $|\mathbf{x}_2 - \mathbf{x}| = \text{constant}$ , the leading term is found to contribute nothing to the integral, and is in fact identical to the earlier discarded  $\nabla^2 \mathbf{E}_1$  term. The leading term to contribute to the integral is  $O(|\mathbf{x}_2 - \mathbf{x}_1|^{-6})$ . From the method of reflexions for calculating the far-field interactions, the term corresponds to the change in value of the  $\mathbf{x}_2$ -particle's dipole strength due to the straining field it experiences from the  $\mathbf{x}_1$ -particle acting as isolated in a pure solvent. This observation is exploited in the second renormalization.

#### 6. The second renormalization

The first renormalization has shown that the averaged-equation approach can be used to calculate the bulk stress to  $O(c^2)$ , producing the same result as Batchelor & Green (1972*a*). The first renormalization, however, would not succeed in the permeability problem. A second, more powerful, renormalization of the bulk stress problem is now presented which carries further the averaged-equation concept.

The second renormalization recognizes that the right-hand side of (4.1) or (5.1) represents the difference between the bulk material and the pure solvent. It is the bulk material that characterizes the behaviour of the conditionally averaged fields far from the fixed particles. At the O(c) level required here, the bulk material seen by  $\mathbf{u}(\mathbf{x}|\mathbf{x}_1)$  is a Newtonian fluid with Einstein's effective viscosity

$$\boldsymbol{\sigma}(\mathbf{x}|\mathbf{x}_1) \to -p(\mathbf{x}|\mathbf{x}_1) | + 2\mu(1 + \frac{5}{2}c) \, \mathbf{e}(\mathbf{x}|\mathbf{x}_1) + O(c^2), \quad \text{as} \quad |\mathbf{x}_2 - \mathbf{x}_1| \to \infty.$$
(6.1)

This O(c) increase in the effective viscosity is taken from the right-hand side of (5.1) in the excluded shell  $|\mathbf{x} - \mathbf{x}_1| \ge 2a$  and is combined with the left-hand side. The second renormalization is thus in  $|\mathbf{x} - \mathbf{x}_1| \ge a$ 

$$-\nabla p_{f}(\mathbf{x}|\mathbf{x}_{1}) + \mu \nabla^{2} \mathbf{u}(\mathbf{x}|\mathbf{x}_{1}) + \int_{|\mathbf{x}_{2}-\mathbf{x}_{1}| \geq 2a} dV_{2} \frac{5}{2} \mu c \nabla_{2}^{2} \mathbf{u}(\mathbf{x}_{2}|\mathbf{x}_{1}) \,\delta(\mathbf{x}-\mathbf{x}_{2}) \\ + \left( \oint_{|\mathbf{x}_{2}-\mathbf{x}_{1}| \geq 2a} - \oint_{\infty} \right) dS_{2} \,5\mu c \mathbf{e}(\mathbf{x}_{2}|\mathbf{x}_{1}) \cdot \mathbf{n}_{2} \,\delta(\mathbf{x}-\mathbf{x}_{2}) \\ = \int_{|\mathbf{x}_{2}-\mathbf{x}_{1}| \geq 2a} dV_{2} \left\{ P(\mathbf{x}_{2}|\mathbf{x}_{1}) \oint_{|\mathbf{x}'-\mathbf{x}_{2}| = a} dS' \,\mathbf{\sigma}(\mathbf{x}'|\mathbf{x}_{1},\mathbf{x}_{2}) \cdot \mathbf{n}' \delta(\mathbf{x}-\mathbf{x}') \\ - 5\mu c \mathbf{e}(\mathbf{x}_{2}|\mathbf{x}_{1}) \cdot \nabla_{2} \,\delta(\mathbf{x}-\mathbf{x}_{2}) \right\}.$$
(6.2)

The surface distribution on the left-hand side at  $|\mathbf{x}_2 - \mathbf{x}_1| = 2a$  has the effect of ensuring that the surface stress  $\sigma(\mathbf{x}|\mathbf{x}_1) \cdot \mathbf{n}$  is continuous across the surface when using the

Einstein viscosity outside and the pure solvent viscosity inside. The surface integral at infinity signifies a similar increase in the bulk stress due to the enhanced viscosity at O(c). This latter term corresponds to the dubiously dismissed integral in the first renormalization. As the boundary conditions at infinity are applied to the strain rate  $\mathbf{e}(\mathbf{x}|\mathbf{x}_1)$  and not to the stress  $\boldsymbol{\sigma}(\mathbf{x}|\mathbf{x}_1)$  this term has no effect.

Comparing the second renormalization (6.2) with the first (5.2) it is seen that the degenerate octupole has been dropped and, more significantly, the strength of the dipoles in the subtraction has become dependent upon the unknown field  $\mathbf{e}(\mathbf{x}|\mathbf{x}_1)$ . The problem posed by (6.2) is thus an integro-differential equation. When solving the equation to O(c), it is possible to consider separately the contributions due to the O(c) terms on the two sides of the equations.

The left-hand side of (6.2) describes the Stokes flow of a liquid with a viscosity  $\mu$  inside  $a < |\mathbf{x}_2 - \mathbf{x}_1| < 2a$  and a viscosity  $\mu^* = \mu(1 + \frac{5}{2}c)$  in  $2a < |\mathbf{x}_2 - \mathbf{x}_1|$ . When the strain rate at infinity is given, the jump in the viscosity alters the stress within the  $\mathbf{x}_1$ -particle. With some straightforward but tiresome algebra the bulk stress integral (2.9) can be evaluated for the two viscosity fluids as

$$\frac{20\pi a^{3}\mu}{3}\mathbf{e}(\mathbf{x}_{1})\frac{40\frac{\mu^{*}}{\mu}\left(153+127\frac{\mu^{*}}{\mu}\right)}{3663+6004\frac{\mu^{*}}{\mu}+1533\left(\frac{\mu^{*}}{\mu}\right)^{2}}$$

Substituting the Einstein viscosity for  $\mu^*$  reduces this to

$$\frac{20\pi a^{3}\mu}{3}\mathbf{e}(\mathbf{x})\left\{1+\frac{103}{64}\,c+O(c^{2})\right\}.$$

To calculate the O(c) correction driven by the right-hand side of (6.2), the Green's function for the pure solvent may be used to invert the left-hand side, and the dilute approximation  $\mathbf{e}(\mathbf{x}_1) + \mathbf{E}_{\mathbf{I}}(\mathbf{x};\mathbf{x}_1)$  may be used to approximate  $\mathbf{e}(\mathbf{x}|\mathbf{x}_1)$  in the integrand. Thus the O(c) correction velocity field is

$$\int_{|\mathbf{x}_{2}-\mathbf{x}_{1}| \ge 2a} dV_{2} \{ P(\mathbf{x}_{2}|\mathbf{x}_{1}) \mathbf{U}_{2}(\mathbf{x};\mathbf{x}_{1},\mathbf{x}_{2}) - P(\mathbf{x}_{1}) \frac{5}{6}a^{3}([\mathbf{e}(\mathbf{x}_{1}) + \mathbf{E}_{1}(\mathbf{x}_{2};\mathbf{x}_{1})] \cdot \nabla_{2}) \cdot \mathscr{I}(\mathbf{x}-\mathbf{x}_{2}) \}$$

with an extra image field for the second term associated with maintaining the rigid boundary condition on the  $\mathbf{x}_1$ -particle. As in the first renormalization the  $\mathbf{U}_2$  term, which is the additional velocity due to the presence of the second  $\mathbf{x}_2$ -particle, contributes to the bulk stress integral by  $\mathbf{S}_2(\mathbf{x}_1, \mathbf{x}_2)$ . Using the appropriate Faxén law the second term contributes

The third argument in  $\mathbf{E}_1$  denotes that a strain rate of  $\mathbf{e}(\mathbf{x}_1) + \mathbf{E}_1(\mathbf{x}_2; \mathbf{x}_1)$  instead of  $\mathbf{e}(\mathbf{x}_1)$  has been used for the boundary condition at infinity in the isolated particle problem which defines  $\mathbf{E}_1$ . Thus this second term from the right-hand side represents the bulk stress from an isolated particle experiencing the additional strain rate at  $\mathbf{x}_1$  due to an isolated particle at  $\mathbf{x}_2$  experiencing the strain rate  $\mathbf{e}(\mathbf{x}_1) + \mathbf{E}_1(\mathbf{x}_2; \mathbf{x}_1)$ .

Gathering together the contributions from the two sides of (6.2), the second renormalization has derived an expression for the bulk stress to  $O(c^2)$ 

$$\begin{aligned} \mathbf{\sigma}(\mathbf{x}) &= -p(\mathbf{x}) \,\mathbf{I} + 2\mu (1 + \frac{5}{2}c) \,\mathbf{e}(\mathbf{x}) + \frac{515}{64} \,\mu c^2 \,\mathbf{e}(\mathbf{x}) \\ &+ P(\mathbf{x}) \int_{|\mathbf{x}_1 - \mathbf{x}_1| \ge 2a} dV_2 \left\{ P(\mathbf{x}_2 | \mathbf{x}) \,\mathbf{S}_2(\mathbf{x}, \mathbf{x}_2) - 5\mu c \,\mathbf{E}_1(\mathbf{x}; \mathbf{x}_2; \mathbf{e}(\mathbf{x}) + \mathbf{E}(\mathbf{x}_2; \mathbf{x})) \right\}. \end{aligned}$$
(6.3)

The difference between this result and the result (5.4) from the first renormalization is

$$5\mu c \left\{ \frac{39}{64} c\mathbf{e}(\mathbf{x}) - P(\mathbf{x}_1) \int_{|\mathbf{x}_1 - \mathbf{x}_1| \ge 2a} dV_2 \mathbf{E}_1(\mathbf{x}; \mathbf{x}_2; \mathbf{E}_1(\mathbf{x}_2; \mathbf{x})) \right\}.$$

Further tedious algebra confirms that this difference vanishes identically, and so provides a check on the expressions given in the two sections.

The integral in (6.3) converges absolutely so long as  $P(\mathbf{x}_2|\mathbf{x}_1)$  asymptotes to  $P(\mathbf{x}_1)$  like any non-positive power of large  $|\mathbf{x}_2 - \mathbf{x}_1|$ . Using the steady  $P(\mathbf{x}_2|\mathbf{x}_1)$  distribution for free spheres, there are no probability effects until  $O(|\mathbf{x}_2 - \mathbf{x}_1|^{-9})$ . After integrating around on surfaces of constant  $|\mathbf{x}_2 - \mathbf{x}_1|$  to remove an  $O(|\mathbf{x}_2 - \mathbf{x}_1|^{-5})$  term, the leading contributor in the integrand is  $O(|\mathbf{x}_2 - \mathbf{x}_1|^{-8})$ . This represents two effects found by the method of reflexions approach to far-field interactions. The dipole strength of the sphere at  $\mathbf{x}_2$  has a component  $\frac{2}{3}\pi a^5 \mu \nabla_2^2 \mathbf{e}(\mathbf{x}_2|\mathbf{x}_1)$  in addition to that proportional to  $\mathbf{e}(\mathbf{x}_2|\mathbf{x}_1)$  which gave the Einstein viscosity. There is also a quadrupole with a strength proportional to  $\nabla_2 \nabla_2 \mathbf{u}(\mathbf{x}_2|\mathbf{x}_1)$ . These two effects are not included in the characterization of the bulk material, because on the bulk length scale they produce small effects of the order of the particle length divided by bulk length. They would, moreover, complicate the statement of the problem by raising the order of the differential equations.

The second renormalization is more powerful than the first, the leading contributing term in (5.4) being  $O(|\mathbf{x}_2 - \mathbf{x}_1|^{-6})$  compared with  $O(|\mathbf{x}_2 - \mathbf{x}_1|^{-8})$  in (6.3). The term which has been removed represents the change in the dipole strength of the  $\mathbf{x}_2$ -particle due to the dipole field of the particle at  $\mathbf{x}_1$ . The improved decay is of no immediate use here or in the sedimentation problem, but is the crucial difference between the success and failure of the two normalizations in the permeability problem.

The first renormalization subtracts the leading-order terms of the first reflexion, while the second renormalization removes the leading-order terms of the second reflexion (compare (5.4) and (6.3)). The higher-order terms of these reflexions can be removed using higher-order multipoles. The third- and higher-order reflexions cannot, however, be represented by subtractions in a differential equation for the averaged field and must therefore remain on the right-hand side. The solution of the unaveraged boundary-value problem is nonlinear in terms of the position of the boundaries. Averaging on the other hand is a linear operation. In the differential equation for the averaged field, the effects of the particles are linearly superposed by the averaging. This linear superposition is applicable up to the second reflexion, but cannot represent the essentially nonlinear third reflexion. In two associated transport properties of two-phase materials, the electrical or thermal conductivity and the elastic moduli, successive reflexions can be made orders of magnitude smaller if the particles and surrounding medium have nearly equal properties. Thus to a certain order in the near equality of their properties, but at arbitrary volume concentration, the interactions between the particles can be represented by a differential equation for the averaged field.

Returning to the second renormalization (6.1), a connexion can be observed between the rigorous asymptotic theory of hydrodynamic interactions and a certain selfconsistent field model. As far as the left-hand side of (6.2) is concerned, the conditionally averaged fields are governed by the equations of a pure solvent in

$$a \leq |\mathbf{x} - \mathbf{x}_1| \leq 2a$$

and the bulk material in  $2a \leq |\mathbf{x} - \mathbf{x}_1|$ . The bulk material (6.1) is identified selfconsistently in (2.7) by an integral over the same conditionally averaged fields. In the calculations of this section this process appears in a rather degenerate form because of the diluteness approximation, although it becomes more apparent at the next,  $O(c^3)$  level. The left-hand side of (6.2) thus represents the self-consistent field model in the excluded shell version. The correct rigorous theory differs from the self-consistent model through a non-zero right-hand side in (6.2). There must be a difference because the self-consistent model uses no information about either the probability of particle configurations or the flow past nearby particles. At the  $O(c^2)$  level these two effects can be seen in action in the right-hand side of (6.2). The binary distribution  $P(\mathbf{x}_2|\mathbf{x}_1)$ multiplying the first term in the integrand need not be, and in the steady state for free particle is not, equal to the uniform distribution  $P(\mathbf{x}_1)$  used in the second, subtraction term. The exact two-particle solution  $U_2$  (at  $O(c^2)$ ) differs from that represented by variable strength dipoles, although in the method of reflexions there is agreement up to the fourth term. Thus the excluded shell version of the self-consistent model takes the natural, and perhaps best, assumptions in the lack of such vital information.

## 7. The sedimentation velocity to O(c)

The ensemble-average approach to suspensions advocated in the first sections of the paper can also be applied to calculating the sedimentation velocity of heavy particles relative to the average velocity of the suspension. Particle sedimentation enters the equations of motion for the bulk suspension when the conservation of mass is considered. The bulk averaged conservation of mass law gives the rate of change of the bulk density as the divergence in the bulk mass flux. A constitutive equation relating the bulk mass flux to the bulk density and bulk velocity also involves the density difference between the two phases, the particle concentration and the sedimentation velocity. The sedimentation velocity further obviously enters the bulk equations of motion in the conservation of the particle concentration c, which should be viewed as a bulk variable specifying the microstructural state.

The ensemble-average approach to the mass and particle conservation problems throws up the natural definition of the sedimentation velocity for rigid particles

$$\mathbf{V} = \mathbf{u}(\mathbf{x}|\mathbf{x}) - \mathbf{u}(\mathbf{x}).$$

Note how the bulk problem brings in the one-particle-fixed average fields, which in turn bring in the two-particles-fixed fields and so on through the infinite hierarchy. A closure of the problem is made by the diluteness approximation. At the very lowest level of approximation the suspension is a pure solvent with no particles and so there is no sedimentation problem. At the next level of approximation, corresponding to § 3 in the bulk-stress problem, the one-particle-fixed average field  $\mathbf{u}(\mathbf{x}|\mathbf{x}_1)$  is effectively that of an isolated particle at  $\mathbf{x}_1$  surrounded by a pure solvent. This standard problem gives a sedimentation velocity

$$\mathbf{V} \sim \mathbf{V}_1 = \frac{2}{9}a^2\mu^{-1}(\rho_p - \rho_f)\mathbf{g},$$

where  $\rho_p$  and  $\rho_f$  are the particle and fluid densities, and **g** the gravitational acceleration.

At one further level of approximation, the first hydrodynamic interactions between the particles enter and this is the principal concern of the section. As discussed at this same level for the bulk-stress problem in §4, the equations governing the two-particlesfixed field  $\mathbf{u}(\mathbf{x}|\mathbf{x}_1, \mathbf{x}_2)$  are those of an effectively pure solvent surrounding two particles at  $\mathbf{x}_1$  and  $\mathbf{x}_2$ , which is a classical Stokes flow problem (e.g. see Goldman, Cox & Brenner 1966). With the two-particles-fixed field considered known, the problem for the one-particle-fixed field accurate to O(c) is: inside the particle,  $|\mathbf{x} - \mathbf{x}_1| \leq a$ ,

$$abla \cdot \mathbf{\sigma}(\mathbf{x}|\mathbf{x}_1) + 
ho_p \mathbf{g} = 0,$$

and outside the particle,  $|\mathbf{x} - \mathbf{x}_1| \ge a$ ,

$$\nabla \cdot \mathbf{u}(\mathbf{x}|\mathbf{x}_1) = 0,$$

$$-\nabla p_{f}(\mathbf{x}|\mathbf{x}_{1}) + \mu \nabla^{2} \mathbf{u}(\mathbf{x}|\mathbf{x}_{1}) + \rho_{f} \mathbf{g}$$

$$= -\int_{|\mathbf{x}_{2}-\mathbf{x}| \geq a} dV_{2} P(\mathbf{x}_{2}|\mathbf{x}_{1}) \left(\rho_{p} - \rho_{f}\right) \mathbf{g} - \nabla \cdot \int_{|\mathbf{x}_{2}-\mathbf{x}| \leq a} dV_{2} P(\mathbf{x}_{2}|\mathbf{x}_{1}) \,\boldsymbol{\sigma}(\mathbf{x}|\mathbf{x}_{1},\mathbf{x}_{2})$$

$$= \int_{|\mathbf{x}_{2}-\mathbf{x}_{1}| \geq 2a} dV_{2} P(\mathbf{x}_{2}|\mathbf{x}_{1}) \oint_{|\mathbf{x}'-\mathbf{x}_{1}| = a} dS' \boldsymbol{\sigma}(\mathbf{x}'|\mathbf{x}_{1},\mathbf{x}_{2}) \cdot \mathbf{n}' \delta(\mathbf{x}'-\mathbf{x}),$$

$$(7.1)$$

subject to the usual boundary conditions of continuity of averaged velocity and surface stress on the  $x_1$ -particle and the boundary condition far from the  $x_1$ -particle

$$\mathbf{u}(\mathbf{x}|\mathbf{x}_1) \sim \mathbf{u}(\mathbf{x}), |\mathbf{x} - \mathbf{x}_1| \ge a.$$

If either of the alternative right-hand sides of (7.1) were evaluated as functions of position, then the left-hand side could be inverted directly. Difficulties arise in finding  $\mathbf{u}(\mathbf{x}|\mathbf{x}_1)$  when (7.1) is inverted separately for each  $\mathbf{x}_2$ -particle and an attempt is made to sum over the  $\mathbf{x}_2$ -particles with a  $P(\mathbf{x}_2|\mathbf{x}_1)$  probability weighting. The difficulties of convergence can be overcome using the two renormalization procedures presented earlier.

For the first renormalization the flow outside an isolated falling  $\mathbf{x}_2$ -sphere is studied :

$$\mathbf{U}_{1}(\mathbf{x}|\mathbf{x}_{2}) = \frac{1}{6}a^{3}\mu^{-1}(\rho_{p}-\rho_{f})\,\mathbf{g}(1+\frac{1}{6}a^{2}\nabla_{2}^{2})\,.\,\mathscr{I}(\mathbf{x}-\mathbf{x}_{2}).$$

This shows that the flow is generated by a force monopole and a degenerate quadrupole acting from the centre of the  $\mathbf{x}_2$ -particle. The first renormalization consists of sub-tracting a uniform distribution with density  $P(\mathbf{x}_1)$  in the excluded volume  $|\mathbf{x} - \mathbf{x}_2| \ge 2a$  of force poles and quadrupoles with values corresponding to an isolated  $\mathbf{x}_2$ -particle in an unbounded solvent. Thus in  $|\mathbf{x} - \mathbf{x}_1| \ge a$ 

$$-\nabla p_{f}(\mathbf{x}|\mathbf{x}_{1}) + \rho \nabla^{2} \mathbf{u}(\mathbf{x}|\mathbf{x}_{1}) + \rho_{f} \mathbf{g} = -\int_{|\mathbf{x}_{2}-\mathbf{x}_{1}| \geq 2a} dV_{2} c(\rho_{p}-\rho_{f}) \mathbf{g}(1+\frac{1}{6}a^{2}\nabla_{2}^{2}) \delta(\mathbf{x}-\mathbf{x}_{2}) + \int_{|\mathbf{x}_{2}-\mathbf{x}_{1}| \geq 2a} dV_{2} \left\{ P(\mathbf{x}_{2}|\mathbf{x}_{1}) \oint_{|\mathbf{x}'-\mathbf{x}_{2}|=a} dS' \boldsymbol{\sigma}(\mathbf{x}'|\mathbf{x}_{1},\mathbf{x}_{2}) \cdot \mathbf{n}' \delta(\mathbf{x}'-\mathbf{x}) + c(\rho_{p}-\rho_{f}) \mathbf{g}(1+\frac{1}{6}a^{2}\nabla_{2}^{2}) \delta(\mathbf{x}-\mathbf{x}_{2}) \right\}.$$
(7.2)

The right-hand-side contributions to the sedimentation are considered in turn from the force poles in the first integral, the force quadrupoles in the first integral, and finally the second integral.

The force monopole distribution is a uniform change in density by  $\Delta \rho = c(\rho_p - \rho_f)$ in  $|\mathbf{x} - \mathbf{x}_1| \ge 2a$ , which is most conveniently re-organized into the superposition of  $\Delta \rho$  everywhere,  $0 < |\mathbf{x} - \mathbf{x}_1|$ ,  $-\Delta \rho$  inside the particle,  $0 < |\mathbf{x} - \mathbf{x}_1| < a$ , and  $-\Delta \rho$  in the excluded region  $a < |\mathbf{x} - \mathbf{x}_1| < 2a$ . The uniform change everywhere produces an O(c) change in the pressure gradient but no motion (because velocity and not stress boundary conditions must be imposed far from the particle). The change of the particle density reduces its buoyancy by (1-c) and therefore also the sedimentation velocity by  $-c\mathbf{V}_1$ . The density change in the excluded region induces a velocity field

$$-\int_{0<|\mathbf{x}_1-\mathbf{x}_1|<2a} dV_2 P(\mathbf{x}_1) \frac{1}{6} a^3 \frac{\rho_p - \rho_f}{\mu} \mathbf{g} \cdot \mathbf{\mathscr{I}}(\mathbf{x}-\mathbf{x}_2)$$

plus an extra image term to maintain the rigid boundary condition on the  $x_1$ -particle. The contribution to the sedimentation velocity of the  $x_1$ -particle can be found without calculating the image terms using Faxén's first law, giving

$$-\left(1+\tfrac{1}{6}a^2\nabla^2\right)\int_{a<|\mathbf{x}_2-\mathbf{x}_1|\leqslant 2a}dV_2P(\mathbf{x}_1)\tfrac{1}{6}a^3\frac{\rho_p-\rho_f}{\mu}\mathbf{g}\,.\,\mathcal{I}(\mathbf{x}-\mathbf{x}_2),\quad\text{evaluated at}\quad\mathbf{x}=\mathbf{x}_1,$$

or  $-\frac{9}{2}cV_1$  by straightforward manipulation.

The volume distribution of quadrupoles in the first integral on the right-hand side of (7.2) can be replaced by a surface distribution of dipoles

$$\oint_{|\mathbf{x}_1-\mathbf{x}_1|=2a} dS_2 c_6^1 a^2 (\rho_p - \rho_f) \mathbf{g}(\mathbf{n}_2 \cdot \nabla_2) \, \delta(\mathbf{x} - \mathbf{x}_2).$$

There is also a similar surface distribution at infinity, which is ignored as justified only by the second renormalization. Applying Faxén's first law directly to the velocity induced by the dipole surface distribution gives a contribution to the sedimentation velocity

$$(1+\tfrac{1}{6}a^2\nabla^2)\oint_{|\mathbf{x}_2-\mathbf{x}_1|=2a}dS_2 c\,\frac{a^2}{48\pi}\frac{\rho_p-\rho_f}{\mu}\,\mathbf{g}(\mathbf{n}_2\cdot\nabla_2)\,.\,\mathcal{I}(\mathbf{x}-\mathbf{x}_2),\quad\text{evaluated at}\quad\mathbf{x}=\mathbf{x}_1,$$

or  $\frac{1}{2}cV_1$  by straightforward manipulation.

In the second integral on the right-hand side of (7.2) the  $\sigma(\mathbf{x}|\mathbf{x}_1, \mathbf{x}_2)$  surface distribution creates the isolated two-particle velocity field  $\mathbf{u}(\mathbf{x}|\mathbf{x}_1, \mathbf{x}_2)$  with a sedimentation velocity  $\mathbf{V}_1 + \mathbf{V}_2(\mathbf{x}_1, \mathbf{x}_2)$ , where  $\mathbf{V}_2$  is used for the incremental velocity due to the second particle. By design the multipoles create the isolated particle velocity field  $\mathbf{U}_1(\mathbf{x}; \mathbf{x}_2)$ , with an associated image term to maintain the rigid boundary conditions on the  $\mathbf{x}_1$ -particle. The image terms can be bypassed by applying Faxén's first law for the sedimentation velocity.

Combining all the contributions from the different parts of the right-hand side of (7.2), the sedimentation velocity correct to O(c) may be expressed as

$$\mathbf{V} \sim \mathbf{V}_{1} - 5c\mathbf{V}_{1} + \int_{|\mathbf{x}_{2} - \mathbf{x}_{1}| \ge 2a} dV_{2} \{ P(\mathbf{x}_{2} | \mathbf{x}_{1}) \, \mathbf{V}_{2}(\mathbf{x}_{2}, \mathbf{x}_{1}) - P(\mathbf{x}_{1}) \, [(1 + \frac{1}{6}a^{2}\nabla^{2}) \, \mathbf{U}_{1}(\mathbf{x}; \mathbf{x}_{1})_{\mathbf{x} = \mathbf{x}_{1}}] \}.$$
(7.3)

The remainder term is absolutely convergent if  $P(\mathbf{x}_2|\mathbf{x}_1)$  asymptotes to  $P(\mathbf{x}_1)$  quicker than  $O(|\mathbf{x}_2 - \mathbf{x}_1|^{-2})$ . The result (7.3) from the first renormalization agrees with Batchelor (1972), identifying his  $\mathbf{V}' + \mathbf{V}''$  and  $\mathbf{W}$  with the above  $-5c\mathbf{V}_1$  and remainder integral.

In the second renormalization the equations governing the one-particle-fixed averaged field  $\mathbf{u}(\mathbf{x}|\mathbf{x}_1)$  are made to reflect the properties of the bulk material far from the  $\mathbf{x}_1$ -particle. At O(c) the bulk material has a density  $\rho_f(1-c) + \rho_p c$ , and a viscosity  $\mu(1+\frac{5}{2}c)$ . To avoid creating a divergent integral in the eventual remainder integral, the weight of each  $\mathbf{x}_2$  particle is not located at its centre but distributed uniformly over the surface of the  $\mathbf{x}_2$ -particle. Taking the bulk material terms from the right-hand side of (7.1c) onto the left-hand side yields the following second renormalization in  $|\mathbf{x} - \mathbf{x}_1| \ge a$ :

$$-\nabla p_{f}(\mathbf{x}|\mathbf{x}_{1}) + \mu \nabla^{2} \mathbf{u}(\mathbf{x}|\mathbf{x}_{1}) + \rho_{f} \mathbf{g} + \int_{|\mathbf{x}_{1} - \mathbf{x}_{1}| \geq 3a} dV_{2} c(\rho_{p} - \rho_{f}) \mathbf{g} \delta(\mathbf{x} - \mathbf{x}_{2}) + \int_{a < |\mathbf{x}_{1} - \mathbf{x}_{1}| < 3a} dV_{2} c(\rho_{p} - \rho_{f}) \frac{|\mathbf{x}_{2} - \mathbf{x}_{1}|^{2} + 2a|\mathbf{x}_{2} - \mathbf{x}_{1}| - 3a^{2}}{4|\mathbf{x}_{2} - \mathbf{x}_{1}| a} \mathbf{g} \delta(\mathbf{x} - \mathbf{x}_{2}) + \int_{|\mathbf{x}_{1} - \mathbf{x}_{1}| \geq 2a} dV_{2} \frac{5}{2} \mu c \nabla^{2} \mathbf{u}(\mathbf{x}_{2}|\mathbf{x}_{1}) \delta(\mathbf{x} - \mathbf{x}_{2}) + \left( \oint_{|\mathbf{x}_{2} - \mathbf{x}_{1}| = 2a} - \oint_{\infty} \right) dS_{2} 5 \mu c \times \mathbf{e}(\mathbf{x}_{2}|\mathbf{x}_{1}) \cdot \mathbf{n}_{2} \delta(\mathbf{x} - \mathbf{x}_{2}) = \int_{|\mathbf{x}_{1} - \mathbf{x}_{1}| \geq 2a} dV_{2} \left\{ \oint_{|\mathbf{x}' - \mathbf{x}_{1}| = a} dS' [P(\mathbf{x}_{2}|\mathbf{x}_{1}) \mathbf{\sigma}(\mathbf{x}'|\mathbf{x}_{1} \cdot \mathbf{x}_{2}) \cdot \mathbf{n}_{2} + P(\mathbf{x}_{1}) \frac{1}{3} a(\rho_{p} - \rho_{f}) \mathbf{g} ] \delta(\mathbf{x}' - \mathbf{x}) - 5 \mu c \mathbf{e}(\mathbf{x}_{2}|\mathbf{x}_{1}) \cdot \nabla_{2} \delta(\mathbf{x} - \mathbf{x}_{2}).$$
(7.4)

The left-hand side of the renormalization represents the excluded-shell version of the self-consistent field model of the problem. The remainder integral includes local effects in the probability distribution,  $P(\mathbf{x}_2|\mathbf{x}_1)$ , and in the hydrodynamics  $\sigma(\mathbf{x}|\mathbf{x}_1, \mathbf{x}_2)$  which cannot be included in the self-consistent field formulations.

It is possible to solve (7.4) correct to O(c) by considering independently the effects arising from the change in the density, the change in the viscosity, and the remainder integral. The uniform density  $\rho_f(1-c) + \rho_p c$  in  $|\mathbf{x} - \mathbf{x}_1| > 3a$  leads, by now familiar methods, to a correction in the sedimentation velocity of  $-13c\mathbf{V}_1$ , while the continuously varying density in  $a < |\mathbf{x} - \mathbf{x}_1| < 3a$  gives a correction of  $8c\mathbf{V}_1$ . The jump to the Einstein viscosity in  $|\mathbf{x} - \mathbf{x}_1| > 2a$  with the continuity of surface stress at 2acauses a correction of  $-\frac{205}{128}c\mathbf{V}_1$ . From the remainder integral, the  $\sigma(\mathbf{x}|\mathbf{x}_1,\mathbf{x}_2)$  distribution generates the isolated two-particle velocity field  $\mathbf{U}_2(\mathbf{x};\mathbf{x}_1,\mathbf{x}_2)$  with the additional sedimentation rate  $\mathbf{V}_2(\mathbf{x}_1,\mathbf{x}_2)$ . The surface distribution of the weight of the  $\mathbf{x}_2$ -particle would create in an unbounded solvent the isolated particle flow  $\mathbf{U}_1(\mathbf{x};\mathbf{x}_2)$ , an image term being needed in the presence of the  $\mathbf{x}_1$ -particle. In the final remainder term an adequate approximation to the required  $\mathbf{e}(\mathbf{x}_2|\mathbf{x}_1)$  is the solution for a single particle in an unbounded solvent  $\mathbf{E}_1(\mathbf{x}_2;\mathbf{x}_1)$ .

Combining all the various contributions, the sedimentation velocity may be expressed correct to O(c) as

$$\mathbf{V} \sim \mathbf{V}_{1} - \frac{845}{128}c\mathbf{V}_{1} + \int_{|\mathbf{x}_{1} - \mathbf{x}_{1}| \ge 2a} dV_{2} \{ P(\mathbf{x}_{2}|\mathbf{x}_{1}) \, \mathbf{V}_{2}(\mathbf{x}_{1};\mathbf{x}_{2}) - P(\mathbf{x}_{1}) \, [(1 + \frac{1}{6}a^{2}\nabla^{2}) \, (\mathbf{U}_{1}(\mathbf{x};\mathbf{x}_{1}) + \frac{5}{6}a^{3}(\mathbf{E}_{1}(\mathbf{x};\mathbf{x}_{2}) \, \cdot \nabla_{2}) \, \cdot \, \boldsymbol{\mathscr{I}}(\mathbf{x} - \mathbf{x}_{2})]_{\mathbf{x} = \mathbf{x}_{1}} \}.$$
(7.5)

By straightforward manipulation the result of the second renormalization can be shown to be equal to the result of the first. The advantage of the remainder integral in (7.5) is that the leading term in the integrand is  $O(|\mathbf{x}_2 - \mathbf{x}_1|^{-6})$  compared with the weaker  $O(|\mathbf{x}_2 - \mathbf{x}_1|^{-4})$  in (7.3), assuming that the binary probability function asymptotes sufficiently fast. In the method of reflexions for far-field interactions, the new leading term comes from three effects: a correction to the dipole strength of the  $\mathbf{x}_2$ -particle proportional to  $\nabla_2^2 \mathbf{e}(\mathbf{x}_2|\mathbf{x}_1)$ , a quadrupole at  $\mathbf{x}_2$  proportional to  $\nabla_2 \nabla_2 \mathbf{u}(\mathbf{x}_2|\mathbf{x}_1)$ , and an octupole at  $\mathbf{x}_2$  proportional to  $\mathbf{e}(\mathbf{x}_2|\mathbf{x}_1)$ . These effects cannot easily be included in any improved renormalization.

#### 8. The permeability to O(c)

The averaged-equation formulation is finally applied to a model of a porous medium. A dilute suspension of rigid spheres is considered in which the particles are held fixed by suitable body forces acting in the particles. The problem is to calculate the permeability of the porous medium, i.e. the mean body force required to hold the particles fixed when the volume flux through the fixed bed is given. The dilute fixed bed is not a realistic model of a porous medium, but more a mathematical test for the theories of hydrodynamic interactions. The first renormalization procedure of (which recovered Batchelor's method) fails the test, and the second renormalization procedure of § 6 (which recognizes that the effective medium differs from the pure solvent) is needed.

For simplicity let the body forces  $f(x|\mathscr{C})$  be only those acting in the particles necessary to hold them fixed. Then the required drag per unit volume is f(x). By the linearity of the Stokes-flow problem

$$\mathbf{f}(\mathbf{x}) = -\mu \alpha^2 \mathbf{u}(\mathbf{x}),$$

where  $\alpha^{-1}$  is Darcy's pore size of the porous medium. The problem for the permeability becomes one of expressing  $\alpha^2$  in terms of a and c. Because **f** has been chosen to vanish outside the particles, it is possible to write

$$\mathbf{f}(\mathbf{x}) = \int_{|\mathbf{x}_1 - \mathbf{x}| \leq a} dV_1 P(\mathbf{x}_1) \mathbf{f}(\mathbf{x} | \mathbf{x}_1),$$

which by local homogeneity becomes

$$\mathbf{f}(\mathbf{x}) = P(\mathbf{x}_1) \, \mathbf{F},$$

where  $\mathbf{F}$  is the average total force acting on a particle centred at  $\mathbf{x}$ . In the dilute theory in which each particle is effectively surrounded by unbounded solvent,

and so 
$$\mathbf{F} \sim \mathbf{F}_1 = -6\pi\mu a \mathbf{u}(\mathbf{x}_1)$$
$$\alpha^2 \sim 6\pi a P(\mathbf{x}_1) = \frac{9}{2}ca^{-2}.$$

Corrections to this estimate from hydrodynamic interactions are now sought.

The equations of motion governing the averaged fields with one particle fixed at  $\mathbf{x}_1$  are: outside that particle,  $|\mathbf{x} - \mathbf{x}_1| \ge a$ ,

$$\nabla \cdot \mathbf{u}(\mathbf{x}|\mathbf{x}_{1}) = 0,$$
  
$$-\nabla p_{f}(\mathbf{x}|\mathbf{x}_{1}) + \mu \nabla^{2} \mathbf{u}(\mathbf{x}|\mathbf{x}_{1})$$
  
$$= \int_{|\mathbf{x}_{1}-\mathbf{x}_{1}| \geq 2a} dV_{2} P(\mathbf{x}_{2}|\mathbf{x}_{1}) \oint_{|\mathbf{x}'-\mathbf{x}_{2}|=a} dS' \boldsymbol{\sigma}(\mathbf{x}'|\mathbf{x}_{1},\mathbf{x}_{2}) \cdot \mathbf{n}' \delta(\mathbf{x}'-\mathbf{x}), \quad (8.1)$$

subject to boundary conditions

$$\begin{aligned} \mathbf{u}(\mathbf{x}|\mathbf{x}_1) &\sim \mathbf{u}(\mathbf{x}_1), \quad |\mathbf{x} - \mathbf{x}_1| \geqslant a, \\ \mathbf{u}(\mathbf{x}|\mathbf{x}_1) &= 0, \qquad |\mathbf{x} - \mathbf{x}_1| = a. \end{aligned}$$

If the right-hand side of (8.1) is evaluated at O(c) using the  $\sigma(\mathbf{x}|\mathbf{x}_1, \mathbf{x}_2)$  for two isolated spheres surrounded by pure solvent, it is found to be a constant plus a term like  $|\mathbf{x} - \mathbf{x}_1|^{-1}$  far from the  $\mathbf{x}_1$ -particle. The constant term can be accommodated by a change in the pressure gradient, but the decaying  $|\mathbf{x} - \mathbf{x}_1|^{-1}$  body force causes an infinite velocity field in a viscous fluid. The direct method of tackling a truncated hierarchy for a dilute suspension therefore breaks down with the fixed bed. The cause of the trouble is that the conditionally averaged fields far from the fixed particles should be governed by the equations of the effective material which is a porous medium and not a viscous fluid. The left-hand side of (8.1) can be made to reflect the effective porous medium by adding to both sides of the equations a velocity-dependent body force  $-\mu \alpha^2 \mathbf{u}(\mathbf{x}|\mathbf{x}_1)$ . With this modification the direct method (of evaluating the new right-hand side to O(c) and then inverting the new left-hand-side operator) successfully finds an approximation to  $\mathbf{u}(\mathbf{x}|\mathbf{x}_1)$ .

The direct method evaluates the complete velocity field  $\mathbf{u}(\mathbf{x}|\mathbf{x}_1)$ , and then finds the required force  $\mathbf{F}$  on the  $\mathbf{x}_1$ -particle. An expression for  $\mathbf{F}$  is now sought which bypasses the unnecessary details of the full solution for  $\mathbf{u}(\mathbf{x}|\mathbf{x}_1)$ . The probability sum over the two-particle contributions is divergent so a renormalization is needed. The second renormalization must be used because the first renormalization cannot reflect the change in physics from a viscous fluid to a porous medium. The main subtraction for the porous medium is a body force proportional to the local velocity,  $P(\mathbf{x}_1) 6\pi \rho a \mathbf{u}(\mathbf{x} | \mathbf{x}_1)$ . To avoid a minor convergence difficulty due to this drag being exerted on the surface of the particle rather than at its centre, a uniform distribution of quadrapoles at the particle centres with strengths proportional to  $\mathbf{u}(\mathbf{x}|\mathbf{x}_1)$  must be included. Adhering strictly to the second renormalization procedure, a dipole proportional to  $\nabla \mathbf{u}(\mathbf{x}|\mathbf{x}_1)$ should be included. This term reflects the change at O(c) of the bulk stress from the pure solvent value, the change being asymmetric because torques must be applied to stop the particles rotating. At large distances from the  $\mathbf{x}_1$ -particle the porousmedium character dominates the non-Newtonian stress, and so this dipole subtraction is not needed.

The chosen renormalized version of (8.1) is thus in  $|\mathbf{x} - \mathbf{x}_1| \ge a$ 

$$-\nabla p_{f}(\mathbf{x}|\mathbf{x}_{1}) + \mu \nabla^{2} \mathbf{u}(\mathbf{x}|\mathbf{x}_{1}) - P(\mathbf{x}_{1}) 6\pi\mu a \mathbf{u}(\mathbf{x}|\mathbf{x}_{1}) - \int_{|\mathbf{x}_{2}-\mathbf{x}_{1}| \ge 2a} dV_{2} \frac{3}{4} \mu c \nabla^{2}_{2} \mathbf{u}(\mathbf{x}|\mathbf{x}_{1}) \delta(\mathbf{x}-\mathbf{x}_{2}) - \left( \oint_{|\mathbf{x}_{2}-\mathbf{x}_{1}| = 2a} - \oint_{\infty} \right) dS_{2} \frac{3}{2} \mu c \times \mathbf{e}(\mathbf{x}_{2}|\mathbf{x}_{1}) \cdot \mathbf{n}_{2} \delta(\mathbf{x}-\mathbf{x}_{2}) = -\int_{2a \ge |\mathbf{x}_{2}-\mathbf{x}_{1}| \ge a} dV_{2} P(\mathbf{x}_{1}) 6\pi\mu a \mathbf{u}(\mathbf{x}_{2}|\mathbf{x}_{1}) \delta(\mathbf{x}-\mathbf{x}_{2}) - \left( \oint_{|\mathbf{x}_{2}-\mathbf{x}_{1}| \ge 2a} - \oint_{\infty} \right) dS_{2} \frac{3}{4} \mu c [\mathbf{u}(\mathbf{x}_{2}|\mathbf{x}_{1}) \mathbf{n}_{2} \cdot \nabla_{2} \delta(\mathbf{x}-\mathbf{x}_{2}) + \mathbf{n}_{2} \cdot (\nabla_{2} \mathbf{u}(\mathbf{x}_{2}|\mathbf{x}_{1}))^{\mathrm{T}} \delta(\mathbf{x}-\mathbf{x}_{2})] + \int_{|\mathbf{x}_{2}-\mathbf{x}_{1}| \ge 2a} dV_{2} \left\{ P(\mathbf{x}_{2}|\mathbf{x}_{1}) \oint_{|\mathbf{x}'-\mathbf{x}_{3}| = a} dS' \mathbf{\sigma}(\mathbf{x}'|\mathbf{x}_{1},\mathbf{x}_{2}) \cdot \mathbf{n}' \delta(\mathbf{x}'-\mathbf{x}) - P(\mathbf{x}_{1}) 6\pi\mu a \mathbf{u}(\mathbf{x}_{2}|\mathbf{x}_{1}) (1 + \frac{1}{6}a^{2} \nabla_{2}^{2}) \delta(\mathbf{x}-\mathbf{x}_{2}) \right\}.$$
(8.2)

The contributions to the drag on the  $\mathbf{x}_1$ -particle from the different small terms will be considered separately, working accurate to O(c). With the right-hand side set to zero and the boundary condition at infinity satisfied, the porous-medium term on the left-hand side of (8.2) increases the drag to

$$\mathbf{F}_{1}[1+(3/\sqrt{2})c^{\frac{1}{2}}+\frac{3}{2}c],$$

while the other extra terms on the left-hand side, describing a jump in viscosity to  $\mu(1-\frac{3}{4}c)$  outside the excluded shell, would alone decrease the drag to  $\mathbf{F}_1(1-\frac{123}{256}c)$ . At O(c) the effects of the small porosity and the small jump in viscosity can be calculated separately and added together. Because the small O(c) porous-medium term does produce a larger,  $O(c^{\frac{1}{2}})$  effect, corrections to that term must be considered later.

The corrections from the two subtraction terms on the right-hand side of (8.2) can be calculated at the desired accuracy by using the pure solvent operator for the left-hand side of (8.2). The volume distribution of monopoles in the excluded shell yields a contribution  $\mathbf{F}_1 \frac{89}{64}c$ . The surface distributions at infinity represent the antisymmetric part of the bulk stress and the increased pressure gradient at infinity. As velocity and not force boundary conditions are imposed at infinity, the surface distributions at infinity should be dropped. The surface distributions at  $|\mathbf{x}_2 - \mathbf{x}_1| = 2a$ produce corrections  $-\mathbf{F}_1 \frac{171}{1024}c$  from the monopoles and  $+\mathbf{F}_1 \frac{125}{2048}c$  from the dipoles.

The remainder term in (8.2) is tackled at each fixed  $\mathbf{x}_2$  and then summed. If the solution for two isolated particles in unbounded pure solvent is used for  $\sigma(\mathbf{x}|\mathbf{x}_1, \mathbf{x}_2)$  and if this term is inverted using the pure solvent left-hand side, then the contribution to the drag is  $P(\mathbf{x}_2|\mathbf{x}_1) \mathbf{F}_2(\mathbf{x}_1; \mathbf{x}_2)$ , where  $\mathbf{F}_2$  is the extra drag on the  $\mathbf{x}_1$ -particle due to a particle being at  $\mathbf{x}_2$  in an otherwise pure solvent. The subtraction term yields a contribution

$$P(\mathbf{x}_1) 6\pi\mu a(1 + \frac{1}{6}a^2\nabla^2) \mathbf{U}_1(\mathbf{x};\mathbf{x}_2;\mathbf{u}(\mathbf{x}_1) + \mathbf{U}_1(\mathbf{x}_2;\mathbf{x}_1))$$

evaluated at  $\mathbf{x} = \mathbf{x}_1$ , where the last argument of  $\mathbf{U}_1$  indicates that the pure solvent disturbance flow outside the  $\mathbf{x}_2$ -particle is calculated with a flow at infinity of

$$\mathbf{u}(\mathbf{x}_1) + \mathbf{U}_1(\mathbf{x}_2; \mathbf{x}_1)$$

instead of the otherwise assumed  $\mathbf{u}(\mathbf{x})$ . The combination of the two remainder contributions is not summable. There is a divergent term in  $P(\mathbf{x}_2|\mathbf{x}_1) F(\mathbf{x}_1, \mathbf{x}_2)$  equal to

$$P(\mathbf{x}_1) \, 6\pi\mu a (\tfrac{3}{4}a)^3 \, \mathbf{u}(\mathbf{x}_1) \, . \, \boldsymbol{\mathscr{I}}(\mathbf{x}_1 - \mathbf{x}_2) \, . \, \boldsymbol{\mathscr{I}}(\mathbf{x}_2 - \mathbf{x}_1) \, . \, \boldsymbol{\mathscr{I}}(\mathbf{x}_1 - \mathbf{x}_2)$$

which comes from the third reflexion. The divergence could not have been avoided by subtracting the change in the bulk stress relation from the solvent to the effective medium.

The final divergence is resolved by realizing that the trouble occurs in the region  $|\mathbf{x}_2 - \mathbf{x}_1| \ge a$ . In this region gradients are becoming smaller so that the pure solvent Laplacian term on the left-hand side of (8.2) is becoming comparable with the porous medium term. Thus for long-range effects it is not legitimate to treat the left-hand side as the pure solvent operator. When the porous medium term is retained on the left-hand side of (8.2), the final Stokeslet interaction tensor in the triple product is turned into a shielded Stokeslet interaction tensor  $\mathscr{I}(\mathbf{r}, \alpha)$  given by Howells (1974) as

$$\mathscr{I}(\mathbf{r},\alpha) = \mathbf{I} \frac{2}{\alpha^2 r^3} [(1 + \alpha r + \alpha^2 r^2) e^{-\alpha r} - 1] + \mathbf{rr} \frac{2}{\alpha^2 r^5} [3 - (3 + 3\alpha r + \alpha^2 r^2) e^{-\alpha r}],$$

where at the required accuracy  $\alpha^2 a^2 = \frac{9}{2}c$ . Changing the third interaction tensor to a shielded type does make the third reflexion term convergent, with a contribution to the drag on the  $\mathbf{x}_1$ -particle equal to  $\mathbf{F}_1(\frac{135}{64}c\ln c + 7.266c)$ . The  $O(c\ln c)$  contribution from this modification of the third reflexion is correct but that part of the O(c) contribution which comes from the far field,  $|\mathbf{x}_2 - \mathbf{x}_1| \ge a$ , is incorrect. In the far field it is not possible to use the solution for two particles in a pure solvent. Taking into account the porous medium term on the left-hand side of the  $\mathbf{u}(\mathbf{x}|\mathbf{x}_1, \mathbf{x}_2)$  version of (8.2) turns the first two Stokeslet interaction tensors into the shielded type. With all three interaction tensors of the shielded type, the third reflexion contribution to the  $\mathbf{x}_1$ -particle drag is  $\mathbf{F}_1(\frac{185}{64}c\ln c + 11.987c)$ . The third reflexion is subtracted from the remainder integral and the above separately calculated contribution added.

The small porous-medium term in the  $\mathbf{u}(\mathbf{x}|\mathbf{x}_1, \mathbf{x}_2)$  problem has one further effect at O(c). In pure solvent the drag on the  $\mathbf{x}_2$ -particle becomes  $-6\pi\mu a \mathbf{u}(\mathbf{x}_2|\mathbf{x}_1)$  as the two particles become widely separated. The porous medium term increases this by a factor  $1 + (3/\sqrt{2})c^{\frac{1}{2}} + O(c \ln c)$ . Thus working correct to O(c) the porous medium subtraction term on both sides of (8.2) should be  $-P(\mathbf{x}_1) 6\pi\mu a(1 + (3/\sqrt{2})c^{\frac{1}{2}}) \mathbf{u}(\mathbf{x}|\mathbf{x}_1)$ . With this modified porous medium term, the first calculated drag correction (coming from just the porous medium term on the left-hand side of (8.2)) becomes

$$\mathbf{F}_{1}[1+(3/\sqrt{2})c^{\frac{1}{2}}+\frac{15}{4}c].$$

Combining the many contributions to the permeability the O(c) result is

$$\begin{split} \mathbf{f}(\mathbf{x}_{1}) &= -\,6\pi\mu a P(\mathbf{x}_{1}) \left[ \mathbf{u}(\mathbf{x}_{1}) \left( 1 + (3/\sqrt{2}) \, c^{\frac{1}{2}} + \frac{1\,3\,5}{6\,4} c \ln c + 16\cdot54\,1c \right) \right. \\ &+ \int_{|\mathbf{x}_{1} - \mathbf{x}_{1}| \,\geq\, 2a} dV_{2} \left\{ -\,P(\mathbf{x}_{2}|\mathbf{x}_{1}) \, \mathbf{F}(\mathbf{x}_{1};\mathbf{x}_{2})/6\pi\mu a + P(\mathbf{x}_{1}) \left[ (\frac{3}{4}a)^{3} \,\mathscr{I}^{3}(\mathbf{x}_{1} - \mathbf{x}_{2}) \right. \\ &- \left. (1 + \frac{1}{6}a^{2}\nabla^{2}) \, \mathbf{U}_{1}(\mathbf{x};\mathbf{x}_{2};\mathbf{u}(\mathbf{x}_{1}) + \mathbf{U}_{1}(\mathbf{x}_{2};\mathbf{x}_{1})) \right]_{\mathbf{x} = \mathbf{x}_{1}} \right\} \right]. \end{split}$$

The remainder integral converges in the region  $|\mathbf{x}_2 - \mathbf{x}_1| \ll \alpha^{-1}$  so long as the binary probability distribution asymptotes to the number density  $P(\mathbf{x}_1)$  faster than

$$O(|\mathbf{x}_2 - \mathbf{x}_1|^{-2}).$$

If the probability distribution does not affect the convergence, then the integral has a leading term  $O(|\mathbf{x}_2 - \mathbf{x}_1|^{-4})$  coming from the fourth reflexion and the bulk stress correction, which was not subtracted in the chosen renormalization.

The  $O(c^{\frac{1}{2}})$  term was first found by Brinkman (1947), who solved (8.2) with no righthand side and only the porous medium correction term on the left-hand side. Childress (1972) first derived the  $O(c \ln c)$  term which comes from the third reflexion. The value of the O(c) term does not agree with any derived earlier. Howells (1974) partially corrected Childress's value and effectively gives 16.457 instead of the above 16.541. It is not clear whether this small difference arises from the computation of the integrals or from a more fundamental error.

#### REFERENCES

BATCHELOR, G. K. 1970 J. Fluid Mech. 41, 545.
BATCHELOR, G. K. 1972 J. Fluid Mech. 52, 245.
BATCHELOR, G. K. & GREEN, J. T. 1972a J. Fluid Mech. 56, 401.
BATCHELOR, G. K. & GREEN, J. T. 1972b J. Fluid Mech. 56, 375.

BRINKMAN, H. C. 1947 Appl. Sci. Res. A1, 27.

BUYEVICH, YU. A. & MARKOV, V. G. 1973 Prikl. Mat. Mekh. 37, 1059 (translated in J. Appl. Math. Mech. 37, 1005).

CHILDRESS, S. 1972 J. Chem. Phys. 56, 2527.

EINSTEIN, A. 1906 Ann. Phys. 19, 289.

GOLDMAN, A. J., COX, R. G. & BRENNER, H. 1966 Chem. Engng Sci. 21, 1151.

HASHIN, Z. 1964 Appl. Mech. Rev. 17, 1.

LANDAU, L. D. & LIFSCHITZ, E. M. 1959 Fluid Mechanics. Pergamon.

Howells, I. D. 1974 J. Fluid Mech. 64, 449.

LUNDGREN, T. S. 1972 J. Fluid Mech. 51, 273.

SAFFMAN, P. G. 1971 Stud. Appl. Math. 50, 93.

SAFFMAN, P. G. 1973 Stud. Appl. Math. 52, 115.

TAM, C. K. W. 1969 J. Fluid Mech. 38, 537.