J. Fluid Mech. (1995), *vol.* 294, *pp.* 1–16

'Particle stress' in disperse two-phase potential flow

By H. F. BULTHUIS[1]†, A. PROSPERETTI[1] AND A. S. SANGANI[2]

[1] Department of Mechanical Engineering, The Johns Hopkins University, Baltimore, MD 21218, USA

[2] Department of Chemical Engineering and Materials Science, Syracuse University, Syracuse, NY 13244, USA

(Received 16 November 1993 and in revised form 9 December 1994)

The problem of determining the particle-phase stress in potential flow has been examined recently using two different procedures by Sangani & Didwania (1993*a*) and by Bulthuis (Appendix C of Zhang & Prosperetti 1994). The present study corrects errors in the expression given by Sangani & Didwania, recasts the expression given by Bulthuis in a form suitable for computation, and shows the equivalence of the results obtained by the two methods.

1. Introduction

Biesheuvel & Gorissen (1990) and Sangani & Didwania (1993*a*) (referred to as SD herein) derived average equations for a suspension of rigid massless spherical particles ('bubbles') in an incompressible potential flow. Their result for the disperse-phase average momentum equation may be written as

$$n\left(\frac{\partial \bar{I}}{\partial t}+\bar{w}\cdot\nabla\bar{I}\right) = -\nabla\cdot(n\boldsymbol{\tau})+n(f_v+f_g). \tag{1}$$

Here n is the particle number density, $\bar{I}$ and $\bar{w}$ their average impulse and velocity, f_v and f_g the average viscous and body forces to which they are subject, and $\boldsymbol{\tau}$ is the 'particle stress' consisting of a kinetic contribution $\boldsymbol{\tau}^k$ and a potential contribution $\boldsymbol{\tau}^p$,

$$\boldsymbol{\tau} = \boldsymbol{\tau}^k+\boldsymbol{\tau}^p. \tag{2}$$

The potential-interaction force on a representative bubble was decomposed in SD as a sum of pairwise interaction forces and its expression was derived using a procedure comonly used in statistical mechanics for determining the pressure or, equivalently, the equation of state of liquids (e.g. Irving & Kirkwood 1950; Rice & Gray 1965). In that context, one deals with a set of interacting particles (or molecules) in vacuum. For massless particles dispersed in a fluid, however, all the momentum is with the continuous phase and the analogy with molecules in vacuum is not obvious. For this reason, many investigators have followed a different approach in the study of two-

† Present address: J. M. Burgers Centre for Fluid Mechanics, Department of Applied Physics, University of Twente, 7500 AE Enschede, The Netherlands.

phase flows by taking systematic volume or ensemble averages of the two phases separately. This approach is commonly referred to as the two-fluid model, and has been described most recently by Zhang & Prosperetti (1994). In Appendix C of that study, Bulthuis also gave an expression for the particle stress. Both the approach and the resulting expressions for the particle stress appear quite different from those of SD, and it is the purpose of the present study to reconcile them.

Since we do not have anything to add to the expressions for f_v and f_g given in SD, and since their analysis is independent of the potential flow interaction that is our focus here, we shall ignore viscosity and body forces at the outset. Also, as in SD, our goal will be to derive the average equations correct to first order in the spatial derivatives of the average fields.

In §2, we present an approximate derivation of the particle stress using a method similar to that in SD. The potential interaction force between two particles separated by a distance r behaves as r^{-4} and, thus, the pair contribution to the stress behaves as r^{-3}. Since the volume integral of this quantity over all pairs in an infinite suspension is not absolutely convergent, one would need to devise a suitable renormalization scheme. SD formulated the problem in terms of N interacting particles in a periodic cell and expressed the velocity potential in terms of the periodic Green's function for the Laplace equation to avoid the renormalization of the average stress. This resulted in a derivation that appears somewhat complicated and too specialized to periodic suspensions. The method presented in §2 provides a much simpler alternative and is also useful in identifying the source of an error in the derivation presented in SD which we discuss in §4. The method is quite general and can be applied in principle to other suspension problems with long-range interactions.

In §3 we give an alternative derivation for the particle stress. This derivation is similar to that presented by Bulthuis in Appendix C of Zhang & Prosperetti (1994). According to this expression the stress is closely related to the fluid momentum flux $M_{ij} = \rho u_i u_j$, ρ being the density and u_i the velocity of the fluid. Evaluation of the stress starting from this expression, however, is not a trivial matter since it requires a detailed knowledge of the velocity field around many particles and a volume integration of the momentum flux. In fact, since the velocity disturbance caused by a particle at a distance r from its centre only decays as r^{-3}, one must once again devise a suitable renormalization procedure for evaluating the particle stress starting from this expression – an issue that was not addressed in Zhang & Prosperetti (1994). Thus, further modifications are necessary before their result can be compared with the expression derived by the previous method and in SD. We carry out detailed calculations for a special case of periodic suspensions considered in SD to show that the results obtained by the two methods are equivalent.

In §5 we summarize average equations for two-phase flows in which the potential interactions are of primary importance. Most studies on the subject suggest solving two sets of momentum equations, e.g. one for the disperse phase and the other for the mixture. An important point of this section is that in potential flow applications it is possible to replace the momentum equation for the mixture with a simpler kinematic description.

2. Derivation of the particle stress by the first method

Consider a suspension of N identical spherical particles in an inviscid incompressible fluid. Let $\mathscr{C}^N \equiv (\boldsymbol{x}^1, \boldsymbol{x}^2, \ldots, \boldsymbol{x}^N, \boldsymbol{w}^1, \boldsymbol{w}^2, \ldots, \boldsymbol{w}^N)$ denote the positions and velocities of the particles. Further, let $g^\alpha(\mathscr{C}^N, t)$ be any dynamic variable (e.g. the centre-of-mass velocity or the impulse) associated with a generic particle α. Then we define the ensemble average of g over all the particles by

$$n(\boldsymbol{x},t)\,\bar{g}(\boldsymbol{x},t)=\frac{1}{N!}\int \mathrm{d}\mathscr{C}^N P(\mathscr{C}^N,t)\sum_{\alpha=1}^{N}\delta(\boldsymbol{x}-\boldsymbol{x}^\alpha)\,g^\alpha(\mathscr{C}^N,t), \tag{3}$$

where $P(\mathscr{C}^N,t)$ is the probability density distribution for finding the configuration of particles in a neighbourhood of $\mathscr{C}^N$ at time t, and $n(\boldsymbol{x},t)$ the ensemble-averaged number density of particles, obtained by taking $g^\alpha=1$ in the above expression. Here, the normalization condition for $P(\mathscr{C}^N)$ is taken to be $\int P(\mathscr{C}^N)\,\mathrm{d}\mathscr{C}^N=N!$

With suitable initial conditions, dynamical equations for the particles and the continuous phase, and conditions 'at infinity' for the latter, it is then possible in principle to uniquely calculate the evolution of the system in time. Now it can be shown that $\bar{g}$ satisfies (Zhang & Prosperetti 1994)

$$\frac{\partial}{\partial t}(n\bar{g})+\boldsymbol{\nabla}\cdot(n\overline{\boldsymbol{w}g})=n\bar{\dot{g}}, \tag{4}$$

where $\dot{g}$ is the time derivative of g following the evolution of the entire system. In particular, for $g=1$, we find the conservation equation for the particle number density n:

$$\frac{\partial n}{\partial t}+\boldsymbol{\nabla}\cdot(n\bar{\boldsymbol{w}})=0, \tag{5}$$

using which (4) may be rewritten as

$$n\left(\frac{\partial\bar{g}}{\partial t}+\bar{\boldsymbol{w}}\cdot\boldsymbol{\nabla}\bar{g}\right)=-\boldsymbol{\nabla}\cdot[n(\overline{\boldsymbol{w}g}-\bar{\boldsymbol{w}}\bar{g})]+n\bar{\dot{g}}. \tag{6}$$

The equation of motion for a generic particle positioned at $\boldsymbol{x}^\alpha$ in the absence of viscous and gravitational forces is

$$m\dot{\boldsymbol{w}}^\alpha=-\int_{|\boldsymbol{x}^\alpha-\boldsymbol{z}|=a}\mathrm{d}S_z\,\hat{\boldsymbol{n}}p(\boldsymbol{z},t;N), \tag{7}$$

with m the mass of the particle. After expressing the continuous-phase pressure p from the Bernoulli integral, we have

$$m\dot{\boldsymbol{w}}^\alpha=\rho\int_{|\boldsymbol{x}^\alpha-\boldsymbol{z}|=a}\mathrm{d}S_z\,\hat{\boldsymbol{n}}\left(\frac{\partial\phi_C}{\partial t}+\tfrac{1}{2}\boldsymbol{u}_C\cdot\boldsymbol{u}_C\right), \tag{8}$$

where ρ is the density of the continuous phase, ϕ_C is the continuous phase flow potential, $\hat{\boldsymbol{n}}$ the unit outward normal on the surface of the particle, and $\boldsymbol{u}_C=\boldsymbol{\nabla}\phi_C$ the velocity of the fluid. Upon introducing the impulse $\boldsymbol{I}$ according to the usual definition

$$\boldsymbol{I}^\alpha=-\rho\int_{|\boldsymbol{x}^\alpha-\boldsymbol{z}|=a}\mathrm{d}S_z\,\hat{\boldsymbol{n}}\phi_C, \tag{9}$$

and applying a standard transport theorem, we find

$$m\dot{\boldsymbol{w}}^\alpha+\dot{\boldsymbol{I}}^\alpha=\rho\int_{|\boldsymbol{x}^\alpha-\boldsymbol{z}|=a}\mathrm{d}S_z\,\hat{\boldsymbol{n}}(\tfrac{1}{2}\boldsymbol{u}_C\cdot\boldsymbol{u}_C-\boldsymbol{w}^\alpha\cdot\boldsymbol{u}_C). \tag{10}$$

For any closed surface the integral $\int \mathrm{d}S_z(\partial_i\phi_C)\,\hat{n}_j$ is symmetric in the indices i and j so that, in view of the kinematic boundary condition $\boldsymbol{u}_C\cdot\hat{\boldsymbol{n}} = \boldsymbol{w}^\alpha\cdot\hat{\boldsymbol{n}}$ on the particle surface, the preceding expression may equivalently be written as

$$m\dot{\boldsymbol{w}}^\alpha + \dot{\boldsymbol{I}}^\alpha = \boldsymbol{F}^\alpha, \tag{11}$$

where

$$\boldsymbol{F}^\alpha = \rho\int_{|\boldsymbol{x}^\alpha-\boldsymbol{z}|=a} \mathrm{d}S_z\,\hat{\boldsymbol{n}}\cdot(\tfrac{1}{2}u_C^2\,\mathbf{1} - \boldsymbol{u}_C\,\boldsymbol{u}_C), \tag{12}$$

and $u_C^2 = \boldsymbol{u}_C\cdot\boldsymbol{u}_C$, with $\mathbf{1}$ the identity tensor. The left-hand side of (11) expresses the change of the apparent momentum $\boldsymbol{p}$ that can be attributed to the particle,

$$\boldsymbol{p}^\alpha = m\boldsymbol{w}^\alpha + \boldsymbol{I}^\alpha. \tag{13}$$

Upon averaging the equation of motion (11) according to (3) and using (6) to express $\bar{\boldsymbol{p}}$, we find

$$n\left[\frac{\partial\bar{\boldsymbol{p}}}{\partial t} + \bar{\boldsymbol{w}}\cdot\boldsymbol{\nabla}\bar{\boldsymbol{p}}\right] = -\boldsymbol{\nabla}\cdot[n(\overline{\boldsymbol{wp}} - \bar{\boldsymbol{w}}\bar{\boldsymbol{p}})] + n\bar{\boldsymbol{F}}, \tag{14}$$

where, according to (3), we have

$$n(\boldsymbol{x},t)\,\bar{\boldsymbol{F}}(\boldsymbol{x},t) = \frac{1}{N!}\int \mathrm{d}\mathscr{C}^N P(\mathscr{C}^N,t)\sum_\alpha^N \boldsymbol{F}^\alpha\delta(\boldsymbol{x}-\boldsymbol{x}^\alpha). \tag{15}$$

Equation (14), with minor modifactions accounting for the finite mass of the particles, is in agreement with the corresponding momentum equation derived by Biesheuvel & Gorissen (1990) and SD (cf. (1) with $\boldsymbol{\tau}^k \equiv \overline{\boldsymbol{wp}} - \bar{\boldsymbol{w}}\bar{\boldsymbol{p}}$ and $n\bar{\boldsymbol{F}} = -\boldsymbol{\nabla}\cdot\boldsymbol{\sigma}, \boldsymbol{\sigma} = n\boldsymbol{\tau}^p$). The latter investigators determined $\boldsymbol{\sigma}$ for a set of N interacting particles placed inside the unit cell of a periodic array to avoid the difficulty associated with the long-range nature of the interaction force among particles. As mentioned in the Introduction, this makes the resulting expression for the stress rather too specialized. In what follows, we shall present a simpler and more general method for arriving at the expression for the particle stress. In this part of the present study, we shall adopt a simple dipole approximation for the potential flow around N incompressible particles and write

$$\phi_C(\boldsymbol{x}) = \phi_\infty(\boldsymbol{x}) - \sum_{\alpha=1}^N D_m^\alpha\,\partial_m|\boldsymbol{x}-\boldsymbol{x}^\alpha|^{-1}, \tag{16}$$

where D_m^α is the induced dipole due to the presence of particle α, $\partial_m = \partial/\partial x_m$ is an abbreviated notation for partial differentiation, and ϕ_∞ is the potential due to sources other than the dipoles of the particles. A more complete expression for the velocity potential can be written by including higher-order multipoles if desired. However, as shown in Sangani & Didwania (1993*b*), the dipole approximation is adequate in many potential flow applications, and furthermore the difficulties associated with the long-range nature of the interactions are important only for the dipole interactions.

For the purpose of relating the induced dipoles to the velocity of the particles, we expand ϕ_C near the centre of a representative particle α in vector harmonics:

$$\phi_C = \boldsymbol{s}\cdot[\boldsymbol{\nabla}\phi^r(\boldsymbol{x}^\alpha) + \boldsymbol{D}^\alpha s^{-3}] + \ldots, \tag{17}$$

where $\boldsymbol{s} = \boldsymbol{x} - \boldsymbol{x}^\alpha$ and ϕ^r is the regular part of ϕ at $\boldsymbol{x}^\alpha$, i.e.

$$\phi^r(\boldsymbol{x}) = \phi_\infty(\boldsymbol{x}) - \sum_{\beta\neq\alpha} D_m^\beta\,\partial_m|\boldsymbol{x}-\boldsymbol{x}^\beta|^{-1}. \tag{18}$$

Application of the boundary condition $\hat{\boldsymbol{n}}\cdot\boldsymbol{\nabla}\phi_C = \hat{\boldsymbol{n}}\cdot\boldsymbol{w}^\alpha$ at the surface of the particle gives

$$\boldsymbol{D}^\alpha = \tfrac{1}{2}a^3[\boldsymbol{u}^r(\boldsymbol{x}^\alpha) - \boldsymbol{w}^\alpha], \tag{19}$$

where a is the radius of the particle and $\boldsymbol{u}^r = \nabla\phi^r$.

Now the force on the particle is approximately given by (see e.g. Milne Thomson 1968)

$$F_i^\alpha = 4\pi\rho D_m \partial_m u_i^r(\boldsymbol{x}^\alpha). \tag{20}$$

A more precise expression for the force on a particle in the midst of many others may be obtained by including higher-order multipoles induced by the presence of the particle (see e.g. (78)–(79) in SD where $C_{n+1,m}$ is related to the derivatives of the regular part of the velocity at $\boldsymbol{x}$).

Upon combining (20) with (15) we obtain

$$n\bar{F}_i(\boldsymbol{x}) = \frac{4\pi\rho}{N!}\int \mathrm{d}\mathscr{C}^N P(\mathscr{C}^N, t) \sum_{\alpha=1}^{N} \delta(\boldsymbol{x}-\boldsymbol{x}^\alpha) D_m^\alpha \partial_m u_i^r(\boldsymbol{x}^\alpha, t; \mathscr{C}^N). \tag{21}$$

Substituting for $\boldsymbol{u}^r = \nabla\phi^r$ from (18) into (21), and noting that ϕ_∞ is independent of $\mathscr{C}^N$, we find

$$n\bar{F}_i(\boldsymbol{x}) = -\frac{4\pi\rho}{N!}\int \mathrm{d}\mathscr{C}^N P(\mathscr{C}^N, t) \sum_{\alpha=1}^{N} \sum_{\beta\neq\alpha}^{N} \delta(\boldsymbol{x}^\alpha-\boldsymbol{x}) D_m^\alpha D_n^\beta \partial_{imn}\frac{1}{r^{\alpha\beta}} + 4\pi\rho n\bar{D}_m(\boldsymbol{x})\partial_{im}\phi_\infty, \tag{22}$$

where $r^{\alpha\beta} = |\boldsymbol{x}^\alpha - \boldsymbol{x}^\beta|$ and the partial derivatives are taken with respect to $r^{\alpha\beta}$. Now, given ϕ_∞ and the position and velocity of all the particles, the dipole moment of each particle is in principle determined. Therefore, specifying $\boldsymbol{w}^\alpha$ is equivalent to specifying $\boldsymbol{D}^\alpha$, and vice versa. Since the velocities of the particles do not explicitly appear in the above calculation, it is convenient to think of $\mathscr{C}^N$ as being equivalently specified in terms of $\boldsymbol{x}^\alpha$ and $\boldsymbol{D}^\alpha$. After averaging over $N-2$ particles, the above expression yields

$$n\bar{F}_i(\boldsymbol{x}) = -4\pi\rho\int \mathrm{d}\mathscr{C}^2 P(2,t)\,\delta(\boldsymbol{x}-\boldsymbol{x}^1) D_m^1 D_n^2 \partial_{imn}\frac{1}{r} + 4\pi\rho n\bar{D}_m \partial_{jm}\phi_\infty, \tag{23}$$

with $P(2,t)$ the probability of finding particles at positions $\boldsymbol{x}^1$ and $\boldsymbol{x}^2$ with dipole strengths $\boldsymbol{D}^1$ and $\boldsymbol{D}^2$. In the above equation, $\boldsymbol{r} = \boldsymbol{x}^1 - \boldsymbol{x}^2$. Since the particles 1 and 2 are distinct from each other, $P(2;t) = 0$ for $|\boldsymbol{x}^1-\boldsymbol{x}^2| \leqslant 2a$. Outside this sphere of radius $2a$, we write

$$P(2,t) = P(\boldsymbol{x}^1, \boldsymbol{D}^1, t)\,P(\boldsymbol{x}^2, \boldsymbol{D}^2, t) + P(\boldsymbol{x}^1, \boldsymbol{D}^1, t)\,\Delta P(2,t), \tag{24}$$

where $\Delta P(2,t)$ is the difference between the conditional and unconditional probability density functions,

$$\Delta P(2,t) = P(\boldsymbol{x}^2, \boldsymbol{D}^2 \,|\, \boldsymbol{x}^1, \boldsymbol{D}^1, t) - P(\boldsymbol{x}^2, \boldsymbol{D}^2, t). \tag{25}$$

Upon splitting $\bar{\boldsymbol{F}}$ into two parts according to this decomposition of the pair probability distribution we write

$$\bar{\boldsymbol{F}} = \bar{\boldsymbol{F}}^{mf} + \bar{\boldsymbol{F}}^{sr}. \tag{26}$$

The first term, that may be referred to as the mean-field contribution to the force, is given by

$$n\bar{F}_i^{mf}(\boldsymbol{x}) = 4\pi\rho n\bar{D}_m(\boldsymbol{x})\left[\partial_{im}\phi_\infty - \int_{r>2a} \mathrm{d}^3 r\, n\,\bar{D}_n(\boldsymbol{x}-\boldsymbol{r})\,\partial_{imn}\frac{1}{r}\right]. \tag{27}$$

The second term in (26) is

$$n\bar{F}_i^{sr}(\boldsymbol{x}) = -4\pi\rho\int_{r>2a} \mathrm{d}\mathscr{C}^2 P(\boldsymbol{x}^1, \boldsymbol{D}^1, t)\,\Delta P(2,t)\,\delta(\boldsymbol{x}-\boldsymbol{x}^1) D_m^1 D_n^2 \partial_{imn}\frac{1}{r}. \tag{28}$$

The mean field force given by (27) depends on ϕ_∞. We must now eliminate ϕ_∞ and obtain a result in terms of the averaged quantities at $\boldsymbol{x}$. To do this we define the potential ϕ_D inside the particles to be a regular solution of Laplace's equation satisfying

$$\phi_D^\alpha = \phi_C \quad \text{on} \quad |\boldsymbol{x}-\boldsymbol{x}^\alpha| = a. \tag{29}$$

Thus, for example, ϕ_D inside particle α is given by (cf. (17))

$$\phi_D^\alpha(\boldsymbol{x}) = \boldsymbol{s}\cdot[\boldsymbol{u}^r(\boldsymbol{x}^\alpha)+\boldsymbol{D}^\alpha a^{-3}]+\ldots \qquad (\boldsymbol{s} = \boldsymbol{x}-\boldsymbol{x}^\alpha). \tag{30}$$

Now we define the ensemble-average potential $\langle\phi\rangle$ by

$$\langle\phi\rangle(\boldsymbol{x}) = \langle\chi\phi_C+(1-\chi)\,\phi_D\rangle, \tag{31}$$

and the ensemble-averaged velocity of the mixture $\boldsymbol{U}$ by

$$\boldsymbol{U}(\boldsymbol{x}) = \langle\chi\nabla\phi_C+(1-\chi)\,\boldsymbol{w}\rangle, \tag{32}$$

where $\chi(\boldsymbol{x})$ is an indicator function for the continuous phase such that χ equals unity when $\boldsymbol{x}$ lies in the continuous phase and zero otherwise and $\boldsymbol{w}$ is the velocity of the particle when $\boldsymbol{x}$ is inside a particle. It may be noted that, inside particle α, $\boldsymbol{w}^\alpha \neq \nabla\phi_D^\alpha$ in general. Note also that the above definition of the average potential is the same as the macroscopic potential defined by Wallis (1991).

By using the fact that $\phi_C = \phi_D$ at the surface of the particles and the definition (19) of the induced dipole due to the presence of a particle in the vicinity of $\boldsymbol{x}$, it is now easy to show that

$$\boldsymbol{U} = \boldsymbol{G}-4\pi n\bar{\boldsymbol{D}}, \tag{33}$$

where $\boldsymbol{G} = \nabla\langle\phi\rangle$. To obtain this result we need to evaluate the ensemble average $\langle(1-\chi)(\boldsymbol{w}-\nabla\phi_D)\rangle$. This we do by a volume integral over the test particle centred at $\boldsymbol{x}$. This relation is therefore incorrect for inhomogeneous mixtures but, as explained in SD, it is sufficient to evaluate (33) for homogeneous mixtures to determine the averaged equations correct to first order in the spatial derivatives of the fields.

Since both the fluid and the particle phases are incompressible, $\boldsymbol{U}$ is solenoidal, and taking the divergence of (33) therefore yields

$$\nabla\cdot\boldsymbol{G} = \nabla^2\langle\phi\rangle = 4\pi\nabla\cdot(n\bar{\boldsymbol{D}}). \tag{34}$$

It may be noted that, while ϕ_C and ϕ_∞ satisfy the Laplace equation, the average potential $\langle\phi\rangle$ satisfies a Poisson equation.

To obtain a relation between $\langle\phi\rangle$ and ϕ_∞ we now imagine the system of N particles to be bounded by a surface $\partial\Omega$ and the flow to be induced by specifying the normal component of $\boldsymbol{U} = \nabla\langle\phi\rangle-4\pi n\bar{\boldsymbol{D}}$ over this surface. Then, using Green's identity, we obtain

$$\langle\phi\rangle(\boldsymbol{x}) = \phi_\infty(\boldsymbol{x})+\int_\Omega \mathrm{d}^3x'\,n\bar{\boldsymbol{D}}(\boldsymbol{x}')\cdot\nabla'\frac{1}{|\boldsymbol{x}-\boldsymbol{x}'|}, \tag{35}$$

with ϕ_∞, the potential due to singularities at 'infinity', given by

$$\phi_\infty(\boldsymbol{x}) = \frac{1}{4\pi}\int_{\partial\Omega}\mathrm{d}S\hat{\boldsymbol{n}}\cdot\left\{\frac{\boldsymbol{U}(\boldsymbol{x}')}{|\boldsymbol{x}-\boldsymbol{x}'|}-\langle\phi\rangle(\boldsymbol{x}')\,\nabla'\frac{1}{|\boldsymbol{x}-\boldsymbol{x}'|}\right\}. \tag{36}$$

The second term in (35) may be referred to as the reactive potential. Note that both terms in the right-hand side of (35) become large as the volume occupied by Ω, or, equivalently, N, becomes large, while $\langle\phi\rangle$ remains finite. This circumstance is reminiscent of O'Brien's (1978) macroscopic boundary integral approach to renormalization but the present method is more general as the boundary conditions on $\langle\phi\rangle$ are unrestricted. Now from (35) the second partials of ϕ_∞ are expressed in terms of $\langle\phi\rangle$ as

$$\partial_{im}\phi_\infty(\boldsymbol{x}) = \partial_{im}\langle\phi\rangle(\boldsymbol{x}) + \int \mathrm{d}^3r\, n\bar{D}_n(\boldsymbol{x}-\boldsymbol{r})\,\partial_{imn}\frac{1}{r}. \tag{37}$$

Since the integral here is convergent at infinity, we have removed the reference to Ω. Substituting for ϕ_∞ into (27) we have

$$n\bar{F}_i^{mf} = 4\pi\rho n\bar{D}_m\left[\partial_{im}\langle\phi\rangle + \int_{r\leqslant 2a} \mathrm{d}^3r\, n\bar{D}_n(\boldsymbol{x}-\boldsymbol{r})\,\partial_{imn}\frac{1}{r}\right]. \tag{38}$$

The integral term can be evaluated, taking into account the generalized nature of the derivatives of $1/r$:

$$\partial_{imn}\frac{1}{r} \rightarrow -\frac{4\pi}{5}(\delta_{im}\partial_n + \delta_{in}\partial_m + \delta_{mn}\partial_i)\,\delta(\boldsymbol{r}) \quad \text{for} \quad r \rightarrow 0, \tag{39}$$

to obtain

$$n\bar{F}_i^{mf} = 4\pi\rho n\bar{D}_j\partial_j G_i - \frac{(4\pi)^2}{10}\rho(\delta_{ij}\delta_{mn} + \delta_{im}\delta_{jn} + \delta_{in}\delta_{jm})\,\partial_j(n^2\bar{D}_m\bar{D}_n). \tag{40}$$

The first term is the force on a dipole with strength $\bar{\boldsymbol{D}}$ in a flow field with rate of strain $\nabla\boldsymbol{G}$ and corresponds to the ponderomotive force introduced by Kelvin for the analogous electrostatic problem (Mazur & de Groot 1956; de Groot & Mazur 1962). This 'ponderomotive' force can be written as the divergence of a stress, equivalent to the macroscopic Maxwell stress σ_{ij}^M in electrostatics,

$$4\pi\rho n\bar{D}_j\partial_j G_i = -\rho\,\partial_j(G_iG_j - \tfrac{1}{2}\delta_{ij}G_kG_k - 4\pi nG_i\bar{D}_j) \equiv -\rho\,\partial_j\sigma_{ij}^M. \tag{41}$$

Here use has been made of the relation (33) and of the facts that $\boldsymbol{U}$ is solenoidal and $\boldsymbol{G}$ is irrotational.

The force due to the short-range interactions (cf. (28)) can also be written in divergence form using the procedure introduced by Irving & Kirkwood (1950) and also described in Biesheuvel & Gorissen (1990). Since the force between two particles, being proportional to $\partial_{imn}r^{-1}$, is an odd function of $\boldsymbol{r}$, one can replace $\delta(\boldsymbol{x}-\boldsymbol{x}^1)$ by $\frac{1}{2}[\delta(\boldsymbol{x}-\boldsymbol{x}^1)-\delta(\boldsymbol{x}-\boldsymbol{x}^2)]$. On using a Taylor series expansion of this delta function difference:

$$\delta(\boldsymbol{x}-\boldsymbol{x}^1) - \delta(\boldsymbol{x}-\boldsymbol{x}^2) = -r_j\partial_j\delta(\boldsymbol{x}-\boldsymbol{x}^1) + \ldots \quad (\boldsymbol{r} = \boldsymbol{x}^1 - \boldsymbol{x}^2), \tag{42}$$

we obtain

$$n\bar{F}_i^{sr} = -\partial_j\sigma_{ij}^{sr}, \tag{43}$$

with

$$\sigma_{ij}^{sr} = -2\pi\rho\int_{r>2a}\mathrm{d}\mathscr{C}^2 P(\boldsymbol{x}^1, \boldsymbol{D}^1, t)\,\Delta P(2,t)\,\delta(\boldsymbol{x}-\boldsymbol{x}^1)\,r_j\,\partial_{inm}\frac{1}{r}D_m^1 D_n^2. \tag{44}$$

As long as $\Delta P \rightarrow 0$ as $|\boldsymbol{x}^1 - \boldsymbol{x}^2| \rightarrow \infty$, the integral converges. Combining equations (26), (40), and (43) we obtain

$$n\bar{\boldsymbol{F}} = 4\pi\rho n\bar{\boldsymbol{D}}\cdot\nabla\boldsymbol{G} - \nabla\cdot\boldsymbol{\sigma}^*, \tag{45}$$

with

$$\sigma_{ij}^* = \frac{(4\pi)^2}{10}\rho n^2\bar{D}_m\bar{D}_n(\delta_{ij}\delta_{mn} + \delta_{im}\delta_{jn} + \delta_{in}\delta_{jm}) + \sigma_{ij}^{sr}. \tag{46}$$

The first term in $\boldsymbol{\sigma}^*$ originates from the exclusion of an average dipole distribution within a sphere of radius $2a$. Both $\boldsymbol{\sigma}^*$ and $\boldsymbol{\sigma}^{sr}$ can be referred to as short-range contributions to the stress. In the case of a uniform spatial distribution of particles, for which $\Delta P(2,t) = 0$ for $r > 2a$, the tensor due to short-range interactions σ_{ij}^{sr} is identically zero. In other words, the mixture acts just like a 'dielectric' continuum outside a sphere of radius $2a$ and producing a reactive field at its centre resulting in

$$\sigma_{ij}^{*} = \frac{(4\pi)^2}{10}\rho n^2 \bar{D}_m \bar{D}_n(\delta_{ij}\delta_{mn} + \delta_{im}\delta_{jn} + \delta_{in}\delta_{jm}). \tag{47}$$

In general, however, the stress $\boldsymbol{\sigma}^{sr}$ is non-zero, and one must use appropriate means to determine $P(2,t)$ as a part of the problem for given macroscopic flow conditions. For dilute suspensions one may use, for example, kinetic theory (e.g. van Wijngaarden & Kapteyn 1990; van Wijngaarden 1993; Kumaran & Koch 1993; Sangani *et al.* 1994) to determine these quantities but, for dense suspensions, one must typically use dynamic simulations. For this reason, it is of interest to determine the particle stress for a given configuration of N particles in a periodic box, a typical situation in simulations (e.g. Sangani & Didwania 1993*b*). It may be noted that this is an exceptional case where ΔP does not become vanishingly small at infinity, and thus the term 'short range' contribution is somewhat misleading here. In this section we will only treat the case $N = 1$ but the extension to arbitrary N is straightforward.

Substituting in (44) the expression for ΔP appropriate for the present periodic spatial distribution we obtain

$$\sigma_{ij}^{sr} = -\frac{2\pi\rho}{\mathscr{V}} D_n D_m \int_{r>2a} \mathrm{d}^3 r\, r_j \partial_{inm} \frac{1}{r}\left(\sum_L \delta(\boldsymbol{r}-\boldsymbol{r}_L) - \frac{1}{\mathscr{V}}\right), \tag{48}$$

where $\mathscr{V}$ is the volume of the unit cell. Upon making use of the definition of the regular part of the periodic Green's functions S_1 and S_2 (Hasimoto 1959; Sangani & Acrivos 1983), we have

$$S_1^r(0) = \lim_{\epsilon\to 0}\int_{r>\epsilon} \mathrm{d}^3 r \frac{1}{r}\left(\sum_L \delta(\boldsymbol{r}-\boldsymbol{r}_L) - \frac{1}{\mathscr{V}}\right), \tag{49}$$

$$S_2^r(0) = \lim_{\epsilon\to 0}\int_{r>\epsilon} \mathrm{d}^3 r \frac{r}{2}\left(\sum_L \delta(\boldsymbol{r}-\boldsymbol{r}_L) - \frac{1}{\mathscr{V}}\right). \tag{50}$$

Following a procedure similar to that described in SD, noting that the integral between ϵ and $2a$ does not contribute, and using the identity

$$r_j \partial_{imn}\frac{1}{r} = \partial_{ijmn} r - (\delta_{ij}\partial_{mn} + \delta_{jm}\partial_{in} + \delta_{jn}\partial_{im})\frac{1}{r}, \tag{51}$$

we obtain

$$\sigma_{ij}^{*} = -\frac{2\pi\rho}{\mathscr{V}} D_n D_m [2\,\partial_{ijmn} S_2^r(0) - (\delta_{ij}\partial_{mn} + \delta_{jm}\partial_{in} + \delta_{jn}\partial_{im}) S_1^r(0)]. \tag{52}$$

This result will be compared in §4 with that given by SD.

3. Derivation of the particle stress by the second method

We now present an alternative derivation of the particle stress according to the method of Bulthuis in Appendix C of Zhang & Prosperetti (1994). This might be particularly welcomed by the readers who feel uncomfortable with the use of Taylor series expansion of the delta function (cf. (42)) used in the first derivation. The starting point of this derivation is (14). Upon substituting for $\boldsymbol{F}$ from (12), and after integration over the position of $N-1$ particles and the velocities of N particles we obtain

$$n(\boldsymbol{x},t)\,\bar{\boldsymbol{F}}(\boldsymbol{x},t)=\rho\int \mathrm{d}^3w P(\boldsymbol{x},\boldsymbol{w},t)\int_{|\boldsymbol{x}-\boldsymbol{z}|=a}\mathrm{d}S_z\,\hat{\boldsymbol{n}}\cdot\langle\tfrac{1}{2}u_C^2\,\mathbf{1}-\boldsymbol{u}_C\boldsymbol{u}_C\rangle_1(\boldsymbol{z},t\,|\,\boldsymbol{x},\boldsymbol{w}), \quad (53)$$

where $P(\boldsymbol{x},\boldsymbol{w},t)$ is the reduced one-point probability density function and the bracket $\langle\ldots\rangle_1(\boldsymbol{z},t\,|\,\boldsymbol{x},\boldsymbol{w})$ denotes the continuous-phase average conditional on the presence of a particle with position $\boldsymbol{x}$ and velocity $\boldsymbol{w}$. For potential flow, $\nabla\cdot(\frac{1}{2}u_C^2\,\mathbf{1}-\boldsymbol{u}_C\boldsymbol{u}_C)$ vanishes so that we have the identity

$$\nabla\cdot\langle\chi(\tfrac{1}{2}u_C^2\,\mathbf{1}-\boldsymbol{u}_C\boldsymbol{u}_C)\rangle-\int\mathrm{d}^3w\int_{|\boldsymbol{x}-\boldsymbol{y}|=a}\mathrm{d}S_y\,P(\boldsymbol{y},\boldsymbol{w},t)\,\hat{\boldsymbol{n}}\cdot\langle\tfrac{1}{2}u_C^2\,\mathbf{1}-\boldsymbol{u}_C\boldsymbol{u}_C\rangle_1(\boldsymbol{x},t\,|\,\boldsymbol{y},\boldsymbol{w})=0. \quad (54)$$

This identity is a consequence of the fact that the gradient of the indicator function is a delta distribution with a pole at the surface of the particles multiplied by the unit outward normal vector $\hat{\boldsymbol{n}}$ at the surface (see e.g. Zhang & Prosperetti 1994). Adding (54) to the right-hand side of (53), we find

$$\begin{aligned} n\bar{\boldsymbol{F}} &= \nabla\cdot\rho\langle\chi(\tfrac{1}{2}u_C^2\,\mathbf{1}-\boldsymbol{u}_C\,\boldsymbol{u}_C)\rangle \\ &\quad+\rho\int\mathrm{d}^3wP(\boldsymbol{x},\boldsymbol{w},t)\int_{|\boldsymbol{x}-\boldsymbol{z}|=a}\mathrm{d}S_z\,\hat{\boldsymbol{n}}\cdot\langle\tfrac{1}{2}u_C^2\,\mathbf{1}-\boldsymbol{u}_C\boldsymbol{u}_C\rangle_1(\boldsymbol{z},t\,|\,\boldsymbol{x},\boldsymbol{w}) \\ &\quad-\rho\int\mathrm{d}^3w\int_{|\boldsymbol{x}-\boldsymbol{y}|=a}\mathrm{d}S_y\,P(\boldsymbol{y},\boldsymbol{w},t)\,\hat{\boldsymbol{n}}\cdot\langle\tfrac{1}{2}u_C^2\,\mathbf{1}-\boldsymbol{u}_C\boldsymbol{u}_C\rangle_1(\boldsymbol{x},t\,|\,\boldsymbol{y},\boldsymbol{w}). \end{aligned} \quad (55)$$

The two integrals in this equation do not exactly cancel because, in the first one, the integration is carried out over the surface of a particle centred at $\boldsymbol{x}$, while in the second one it is over all the particles that touch the point $\boldsymbol{x}$. However, while the second integrand depends strongly on the distance from the particle centre, it depends only weakly on the position $\boldsymbol{y}$ of the centre itself and therefore a Taylor series expansion can be carried out in this variable. For brevity, denote the conditionally averaged quantity in the second integrand by $F_1(\boldsymbol{x},t\,|\,\boldsymbol{y},\boldsymbol{w})$ and let $\boldsymbol{x}=\boldsymbol{y}+\boldsymbol{s}$. Then we have (Hinch 1977)

$$\begin{aligned} P(\boldsymbol{y},\boldsymbol{w},t)\,F_1(\boldsymbol{y}+\boldsymbol{s},t\,|\,\boldsymbol{y},\boldsymbol{w}) &= P(\boldsymbol{x},\boldsymbol{w},t)\,F_1(\boldsymbol{x}+\boldsymbol{s},t\,|\,\boldsymbol{x},\boldsymbol{w}) \\ &\quad-\boldsymbol{s}\cdot\nabla_x[P(\boldsymbol{x},\boldsymbol{w},t)\,F_1(\boldsymbol{x}+\boldsymbol{s},t\,|\,\boldsymbol{x},\boldsymbol{w})]+\ldots. \end{aligned} \quad (56)$$

Since the integration variable $\boldsymbol{z}$ equals $\boldsymbol{x}+\boldsymbol{s}$, the first term cancels the first integral in (55) and the final result may be written as

$$n\bar{\boldsymbol{F}}=-\nabla\cdot\boldsymbol{\sigma}, \quad (57)$$

with the particle stress given by

$$\boldsymbol{\sigma}=\boldsymbol{M}=\tfrac{1}{2}\mathrm{Tr}\,[\boldsymbol{M}]\,\mathbf{1}+\rho\int\mathrm{d}^3wP(\boldsymbol{x},\boldsymbol{w},t)\int_{|\boldsymbol{x}-\boldsymbol{z}|=a}\mathrm{d}S_z\,\boldsymbol{s}\,\hat{\boldsymbol{n}}\cdot\langle\boldsymbol{u}_C\boldsymbol{u}_C-\tfrac{1}{2}u_C^2\,\mathbf{1}\rangle_1(\boldsymbol{z},t\,|\,\boldsymbol{t},\boldsymbol{w}). \quad (58)$$

Here

$$\boldsymbol{M}=\rho\langle\chi\boldsymbol{u}_C\,\boldsymbol{u}_C\rangle \quad (59)$$

is the average momentum flux of the continuous phase which is seen to play an important role in $\boldsymbol{\sigma}$. This result for the potential part of the particle stress has been derived with the only assumptions of the flow being incompressible and irrotational.

According to (58) the particle stress is not necessarily symmetric. This circumstance should cause no concern as the usual arguments on the symmetry of the stress tensor (see e.g. Batchelor 1967) are irrelevant here since $\overline{I\times w}\neq 0$ in general. As a matter of fact, it can be shown that the antisymmetric part of the stress tensor $\boldsymbol{\sigma}$ equals the antisymmetric part of $\overline{nwI}$. Also, in the limit of zero particle density, we obtain

$$\sigma = \rho(\boldsymbol{u}_C \boldsymbol{u}_C - \tfrac{1}{2}u_C^2\, \mathbf{1}), \tag{60}$$

which is analogous to the electrostatic Maxwell stress (cf. (41) with $\boldsymbol{u}_C = \boldsymbol{G}$ and $n = 0$). The divergence of this stress is zero, indicating that the medium is force-free in the absence of particles. The presence of the particles modifies the (macroscopic) stress, depending on the microstructure, the number density, and the properties of the particles.

The explicit evaluation of this general expression for the potential stress $\boldsymbol{\sigma}$ requires the momentum flux in the continuous phase. A naive way to evaluate this correct to $O(n)$ would be to write $\boldsymbol{u}_C = \boldsymbol{G}+\boldsymbol{u}^*$ outside a test particle with the disturbance velocity $\boldsymbol{u}^*$ induced by the presence of the particle decaying as $1/r^3$ and then evaluate $\langle \chi \boldsymbol{u}_C \boldsymbol{u}_C\rangle$ by carrying out the integration over the fluid space outside the test particle. This, however, leads to a non-absolutely convergent integral. Thus proper renormalization would be needed in evaluating this even to $O(n)$. In fact, this has already been done by van Wijngaarden (1976) who used Batchelor's renormalization technique to obtain the momentum flux correct to $O(n)$. His approach, however, cannot be easily generalized to higher order in the particle number density, and we will follow a different approach here in which the ensemble average over the continuous phase is first related to an average over the particle phase.

Let $\boldsymbol{u} = \nabla\phi$ be the gradient of the microscopic potential introduced after (29). Note that with this definition $\boldsymbol{u} = \nabla\phi_D^\alpha$ inside the particle α, and thus $\boldsymbol{u} \neq \boldsymbol{w}^\alpha$, where $\boldsymbol{w}^\alpha$ is the velocity of the particle. Upon setting $\boldsymbol{u} = \boldsymbol{G}+\boldsymbol{u}^*$, with $\boldsymbol{G} \equiv \nabla\langle\phi\rangle$ and $\langle \boldsymbol{u}^*\rangle = \boldsymbol{0}$, we find

$$\sigma_{ij} = \sigma_{ij}^\dagger + \sigma_{ij}^*, \tag{61}$$

where

$$\rho^{-1}\sigma_{ij}^\dagger = G_i G_j - \tfrac{1}{2}\delta_{ij} G_k G_k + \langle \chi(G_i u_j^* + G_j u_i^* - \delta_{ij} G_k u_k^*)\rangle$$
$$+\int \mathrm{d}^3 w P(\boldsymbol{x},\boldsymbol{w};t)\int_{|\boldsymbol{x}-\boldsymbol{z}|=a} \mathrm{d}S_z\, \hat{n}_k s_j \langle G_i u_k^* + G_k u_i^* - \delta_{ik} G_m u_m^*\rangle_1 (\boldsymbol{z},t\,|\,\boldsymbol{x},\boldsymbol{w}), \tag{62}$$

and
$$\rho^{-1}\sigma_{ij}^* = \langle \chi T_{ij}\rangle + \int \mathrm{d}^3 w P(\boldsymbol{x},\boldsymbol{w};t)\int_{|\boldsymbol{x}-\boldsymbol{z}|=a} \mathrm{d}S_z\, \hat{n}_k s_j \langle T_{ik}\rangle_1 (\boldsymbol{z},t\,|\,\boldsymbol{x},\boldsymbol{w}), \tag{63}$$

with
$$T_{ij} = u_i^* u_j^* - \tfrac{1}{2}\delta_{ij} u_k^* u_k^*. \tag{64}$$

Now, since $\langle \chi \boldsymbol{u}^*\rangle = -\langle (1-\chi)\boldsymbol{u}^*\rangle = -\langle (1-\chi)(\nabla\phi_D - \boldsymbol{G})\rangle$, we can convert the average over the continuous phase in (62) to a volume integral over a test particle. Combining this with the last term in (62) and using (30) and (19) to evaluate the resulting integral we obtain

$$\rho^{-1}\sigma_{ij}^\dagger = G_i G_j - \tfrac{1}{2}\delta_{ij} G_k G_k - 4\pi n G_i \bar{D}_j = \rho^{-1}\sigma_{ij}^M. \tag{65}$$

The divergence of the 'Maxwell stress' $\sigma_{ij}^\dagger$ corresponds to the 'ponderomotive' force, as can be seen from equation (41). In dilute suspensions, where the interaction among particles can be neglected to leading approximation, the stress $\boldsymbol{\sigma}^*$ can be evaluated by directly integrating T_{ij} over the fluid volume outside the test particle as this quantity decays as $1/r^6$, r being the distance from the centre of the particle. Alternatively, we may use the identity

$$T_{ij} = \partial_k(x_j T_{ik}) - x_j \partial_k T_{ik}, \tag{66}$$

together with $\partial_k T_{ik} = 0$ in the fluid, to convert the volume integral of T_{ij} to an integral over the surface of the test particle. This integral can then be seen to cancel exactly the surface integral in (63), showing thereby that $\boldsymbol{\sigma}^*$ is smaller than $O(n)$. In other words, to $O(n)$, we have

$$\boldsymbol{\sigma} = \boldsymbol{\sigma}^M + O(n^2). \tag{67}$$

This result agrees with that derived by the first method. Now instead of proceeding with the $O(n^2)$ calculation for a specified spatial and velocity distribution of the particles as in the previous section, we shall consider the problem of determining $\boldsymbol{\sigma}^*$ for the case of N particles in a periodic box. In the framework of the dipole approximation the periodic flow disturbance induced by the particles can be written (cf. SD)

$$\boldsymbol{u}^*(\boldsymbol{x}) = -\sum_{\alpha=1}^{N} \boldsymbol{D}^\alpha \cdot \nabla\nabla S_1(\boldsymbol{r}^\alpha), \tag{68}$$

where $\boldsymbol{r}^\alpha = \boldsymbol{x} - \boldsymbol{x}^\alpha$. Upon substituting (68) in the definition (63) of $\boldsymbol{\sigma}^*$ we may write

$$\boldsymbol{\sigma}^* = \sum_{\alpha=1}^{N} \sum_{\gamma=1}^{N} \boldsymbol{\sigma}^{\alpha\gamma}, \tag{69}$$

with

$$\rho^{-1}\mathscr{V}\sigma^{\alpha\gamma} = \int_{V_f} \boldsymbol{T}^{\alpha\gamma}\,\mathrm{d}V + \sum_{\nu=1}^{N} \int_{S^\nu} \boldsymbol{T}^{\alpha\gamma}\cdot\hat{\boldsymbol{n}}\boldsymbol{r}^\nu\,\mathrm{d}S. \tag{70}$$

Here V_f denotes the volume occupied by the fluid, $\mathscr{V}$ the volume of the unit cell, and

$$T_{ij}^{\alpha\gamma} = \tfrac{1}{2}D_m^\alpha D_n^\gamma[\partial_{im} S_1(\boldsymbol{r}^\alpha)\,\partial_{jn} S_1(\boldsymbol{r}^\gamma) + \partial_{jm} S_1(\boldsymbol{r}^\alpha)\,\partial_{in} S_1(\boldsymbol{r}^\gamma) - \delta_{ij}\,\partial_{km} S_1(\boldsymbol{r}^\alpha)\,\partial_{kn} S_1(\boldsymbol{r}^\gamma)]. \tag{71}$$

Consider first the pair $\alpha \neq \gamma$. For such a pair we first convert the integral over the surface of particle ν to a volume integral inside the particle using the identity (66) and the divergence theorem. Thus we obtain

$$\rho^{-1}\mathscr{V}\sigma_{ij}^{\alpha\gamma} = \int_{\mathscr{V}} T_{ij}^{\alpha\gamma}\,\mathrm{d}V + \sum_{\nu=1}^{N} \int_{V^\nu} r_j^\nu\,\partial_k T_{ik}^{\alpha\gamma}\,\mathrm{d}V. \tag{72}$$

In calculating T_{ij} inside the particles, consistently with the divergence theorem, we must use the analytic extension of T_{ij} evaluated from outside the particle, i.e. we must continue to use the expression (68) for $\boldsymbol{u}^*$. Using

$$\nabla^2 S_1(\boldsymbol{x}) = 4\pi[\mathscr{V}^{-1} - \delta(\boldsymbol{x})], \tag{73}$$

it is readily seen that

$$\partial_k T_{ik}^{\alpha\gamma} = -2\pi D_m^\alpha D_n^\gamma[\partial_{im} S_1(\boldsymbol{r}^\alpha)\,\partial_n \delta(\boldsymbol{r}^\gamma) + \partial_{in} S_1(\boldsymbol{r}^\gamma)\,\partial_m \delta(\boldsymbol{r}^\alpha)]. \tag{74}$$

Therefore, the last term in (72) is integrated readily to obtain

$$2\pi D_m^\alpha D_n^\gamma[\delta_{jn}\,\partial_{im} S_1(\boldsymbol{x}^\gamma - \boldsymbol{x}^\alpha) + \delta_{jm}\,\partial_{in} S_1(\boldsymbol{x}^\alpha - \boldsymbol{x}^\gamma)]. \tag{75}$$

Now, to evaluate the first term on the right-hand side of (72), we substitute $\nabla^2 S_2(\boldsymbol{r}^\alpha)$ for $S_1(\boldsymbol{r}^\alpha)$ in (71), where S_2 is a spatially periodic function satisfying $\nabla^2 S_2 = S_1$ (Hasimoto 1959). Integrating by parts four times and discarding at every step the integrals of periodic functions times $\hat{\boldsymbol{n}}$ on the surface of the unit cell, we obtain

$$\int_{\mathscr{V}} T_{ij}^{\alpha\gamma}\,\mathrm{d}V = \tfrac{1}{2}D_m^\alpha D_n^\gamma \int_{\mathscr{V}} \mathrm{d}V[2\,\partial_{ijmn} S_2(\boldsymbol{r}^\alpha) - \delta_{ij}\,\partial_{mn} S_1(\boldsymbol{r}^\alpha)]\,\nabla^2 S_1(\boldsymbol{r}^\gamma). \tag{76}$$

This can be further simplified by using (73) to find

$$\int_{\mathscr{V}} T_{ij}^{\alpha\gamma}\,\mathrm{d}V = -2\pi D_m^{\alpha} D_n^{\gamma}[2\,\partial_{ijmn} S_2(\boldsymbol{x}^{\alpha}-\boldsymbol{x}^{\gamma}) - \delta_{ij}\,\partial_{mn} S_1(\boldsymbol{x}^{\alpha}-\boldsymbol{x}^{\gamma})], \tag{77}$$

where we have made use of the fact that the volume integrals of S_1 and S_2 or any of their derivatives over the unit cell vanish.

The above calculations need some modifications for the pair $\alpha = \gamma$ since the divergence of $T_{ik}^{\alpha\alpha}$ will now involve the product of generalized functions (i.e. a delta function and its derivatives) with singular functions which cannot be integrated. Note that in this case $T_{ij}^{\alpha\alpha} \sim r^{-6}$ as $r = |\boldsymbol{x}-\boldsymbol{x}^{\alpha}| \rightarrow 0$. Of course, the overall result for the contribution from $T_{ij}^{\alpha\alpha}$ is finite and hence this strong singularity should not contribute to the final result. Indeed, it can be shown that the contribution from this pair is the same as for the other pairs except that S_1 and S_2 must now be replaced by their regular parts. Thus, the final result for σ_{ij}^{*} is

$$\sigma_{ij}^{*} = -\frac{2\pi\rho}{\mathscr{V}} \sum_{\alpha=1}^{N} \sum_{\gamma=1}^{N} D_m^{\alpha} D_n^{\gamma}[2\,\partial_{ijmn} S_2(\boldsymbol{x}^{\alpha}-\boldsymbol{x}^{\gamma}) - (\delta_{ij}\,\partial_{mn} + \delta_{jn}\,\partial_{im} + \delta_{jm}\,\partial_{in})\, S_1(\boldsymbol{x}^{\alpha}-\boldsymbol{x}^{\gamma})], \tag{78}$$

with the convention that, for $\alpha = \gamma$, the derivatives of S_1 and S_2 must be evaluated after removing the singularities $1/r$ and $r/2$ from these functions. It is easily seen that the above expression reduces to the result derived in the previous section by taking $N = 1$. The above procedure can now be extended in a straightforward manner to include the higher-order multipoles in the many-particle interaction problem. However, since this was already done correctly in SD, we shall not pursue it here.

It is interesting to note that the exact pair decomposition for the multiparticle stress tensor derived in SD appears in a very natural manner starting with the expression for σ_{ij} derived here. More specifically, the pair decomposition is a simple consequence of the stress tensor being bilinear in $\boldsymbol{u}^{*}$, with $\boldsymbol{u}^{*}$ written as a sum of disturbances induced by each particle in the suspension.

4. Comparison with SD

A comparison of the particle stress derived here with that in SD shows that that expression is incorrect. The ponderomotive force or, equivalently, the stress tensor $\sigma_{ij}^{\dagger}$, which depends explicitly on $\boldsymbol{G}$, is missing in that study. In addition, the short-range tensor σ_{ij}^{*} is also not exactly the same. In SD, this stress has an additional contribution which the authors referred to as τ_{ij}^{ren}.

The source of the first error can be easily spotted by comparing the derivation presented in SD with the derivation presented in §2. In SD, a very specific form of ϕ_{∞} was chosen, namely $\phi_{\infty} = \boldsymbol{G}\cdot\boldsymbol{x}$, with $\boldsymbol{G}$ treated as a constant. Since the second derivative of this ϕ_{∞} is identically zero, the mean-field part of the stress is missing. The authors realized that their calculation did not account for the force on particles in straining flow and added a note in proof in which they mentioned that an additional force due to straining motion must be added. For this they took $\phi_{\infty} = G_j x_j + a_{jk} x_j x_k$ and by an incorrect volume averaging procedure equated a_{jk} to the mean strain $E_{jk} = \frac{1}{2}(\partial_k U_j + \partial_j U_k)$ instead of $\partial_k G_j$ as found here. Thus the term $-\frac{4}{3}\pi a^3 \rho (1+\frac{1}{2}C_E) E_{ij} V_j$, with $\boldsymbol{V} = \bar{\boldsymbol{w}} - \boldsymbol{U}$, that they suggested should be added to the disperse-phase momentum equation, should have $\partial_i G_j$ instead of E_{ij}. Based on the equality of the coefficient C_E for the straining motion force and the added mass coefficient C_a in the case of periodic arrays, those authors further conjectured that $C_a = C_E$ for random arrays as well. We see that this conjecture was correct; in fact, from (45) and (82) below, it appears rather naturally from the derivations presented here.

The second error arose in equating ϕ_∞ to $\langle\phi\rangle$. As we have seen here the difference between these two quantities corresponds to the reactive potential. It can be shown that the additional contribution from this neglected potential for cubic unit cells exactly cancels τ_{ij}^{ren}.

Thus, in summary, had SD chosen $\phi_\infty(\boldsymbol{x})$ instead of $\boldsymbol{G}\cdot\boldsymbol{x}$ in their (47) and related the contribution from ϕ_∞ to $\langle\phi\rangle$ and the reactive potential as is done here, they would have obtained the correct expression for the particle stress.

5. Conclusions and summary of average equations

We have obtained the potential contribution to the so-called particle, or disperse-phase, stress tensor $\boldsymbol{\sigma}$ in two ways. In the approach of §2, the interaction force between particles was decomposed into the sum of pairwise interactions. By approximately representing each particle with a dipole, we were able to write the particle stress in terms of the pair-probability function for the particle positions and dipole moments. In the approach of §3, the continuous phase was viewed as the carrier of a stress $\rho(\boldsymbol{u}_C\,\boldsymbol{u}_C - \frac{1}{2}u_C^2\,\mathbf{1})$. By a direct averaging of this quantity the explicit relation (58) between the particle stress and the momentum flux $\boldsymbol{M}$ was established. These results correct the expression for $\boldsymbol{\sigma}$ given in SD by adding a contribution of first order in the particle number density that was missed in that study.

Within the dipole approximation both of our approaches have been shown to give identical results. Beyond the dipole approximation, the expressions become very complicated and we refer the reader to SD which correctly includes these higher-order contributions.

With our expression for $\boldsymbol{\sigma} = \boldsymbol{\sigma}^\dagger + \boldsymbol{\sigma}^*$ we may write the disperse-phase average momentum equation as

$$n\left(\frac{\partial\bar{\boldsymbol{p}}}{\partial t} + \bar{\boldsymbol{w}}\cdot\nabla\bar{\boldsymbol{p}}\right) = 4\pi\rho n\bar{\boldsymbol{D}}\cdot\nabla\boldsymbol{G} - \nabla\cdot[n\boldsymbol{\tau}^k + \boldsymbol{\sigma}^*], \tag{79}$$

where $\boldsymbol{\tau}^k = \overline{\boldsymbol{w}\boldsymbol{p}} - \bar{\boldsymbol{w}}\bar{\boldsymbol{p}}$ and $\boldsymbol{p}$ is the apparent momentum of a particle which, in the case of a massless bubble, reduces to its impulse. In order to illustrate the dependence of $\boldsymbol{\sigma}^*$ upon the particle probability distribution function and to verify that it indeed gives an $O(n^2)$ contribution, we have carried out approximate calculations for periodic and random arrays. We did not consider the separate probiem of determining the kinetic part $n\boldsymbol{\tau}^k$ of the stress, and we omitted forces due to viscosity, gravitation, and collisions. Given suitable models for these effects (see Sangani & Didwania 1993*b*; Sangani, Zhang & Prosperetti 1994) one would be able to calculate the evolution of the system.

The present results have been derived in terms of auxiliary variables such as $\boldsymbol{p}$, $\boldsymbol{I}$, $\boldsymbol{D}$, $\boldsymbol{G}$, etc. For applications, these auxiliary quantities must be related to measurable fields such as the average particle velocity $\bar{\boldsymbol{w}}$ and the mean flow rate $\boldsymbol{U}$. For example, the average impulse can be related to $\boldsymbol{U}$ and $\bar{\boldsymbol{w}}$ by introducing the added mass coefficient C_a according to the definition of SD, namely

$$\bar{\boldsymbol{I}} = \rho\frac{4\pi a^3}{3}[\tfrac{1}{2}C_a(\bar{\boldsymbol{w}} - \boldsymbol{U}) - \boldsymbol{U}]. \tag{80}$$

The average dipole moment can be expressed in terms of the average impulse by writing ϕ^{α}_{D} instead of ϕ_C in the integral (9), applying the divergence theorem, and averaging, to find

$$\bar{I} = -\rho\frac{4\pi a^2}{3}\left(\bar{w}+\frac{3}{a^3}\bar{D}\right), \tag{81}$$

or, combining the two foregoing relations,

$$\bar{D} = \tfrac{1}{3}a^3(1+\tfrac{1}{2}C_a)(U-\bar{w}). \tag{82}$$

Furthermore, from (33),

$$G = U+\frac{4\pi a^3 n}{3}(1+\tfrac{1}{2}C_a)(U-\bar{w}). \tag{83}$$

It is perhaps unnecessary to point out that C_a, like $\boldsymbol{\sigma}^*$, depends on a particle probability distribution function that must be determined independently from dynamical simulations or kinetic theory arguments.

A number of analytical results are available for the added mass coefficient in dilute as well as dense suspensions with specified spatial and velocity distribution of particles (e.g. van Wijngaarden 1976; Biesheuvel & Spoelstra 1989; Sangani *et al.* 1991; Zhang & Prosperetti 1994). Also, the added mass coefficient is directly related to the polarizability, 'inverse porosity' and exertia as defined by Wallis (1991). For example, Wallis's coefficient δ for the dipole density is defined by

$$4\pi n\bar{D} = \delta(U-\bar{w}) \tag{84}$$

(the extra factor -4π with respect to Wallis's definition arises due to a difference in the definition of the dipole strength). Comparison with (82) shows that δ is related to the added mass coefficient C_a according to $\delta = \frac{4}{3}\pi a^3 n(1+\frac{1}{2}C_a)$. The above expressions should, of course, be modified when the suspension microstructure is anisotropic because, in that case, the added mass coefficient and consequently all the related coefficients are tensors of rank two.

In addition to the above equations for the particle phase, we need an additional set of equations for U, the ensemble-averaged mixture velocity. SD and other previous investigators (e.g. Biesheuvel & van Wijngaarden 1984) propose the following mixture equations for the suspension of massless particles:

$$\nabla\cdot U = 0, \tag{85}$$

$$\rho\left[\frac{\partial}{\partial t}(1-\beta)\,U^L_i+\frac{\partial}{\partial x_j}[(1-\beta)\,U^L_i\,U^L_j]\right] = -\frac{\partial P}{\partial x_i}+\rho(1-\beta)\,g_i-\frac{\partial}{\partial x_j}\beta\Sigma_{ij}, \tag{86}$$

where β is the volume fraction of particles, $(1-\beta)\,U^L_i = U_i-\beta\bar{w}_i$, and Σ_{ij} is the stress tensor for the mixture. Here P is the mixture pressure which must be treated as an additional unknown, unlike $\boldsymbol{\sigma}$ and $\boldsymbol{\Sigma}$ which through suitable constitutive relations are to be taken as functions of U, $\bar{w}-U$, and their spatial derivatives. When the boundary conditions for the macroscopic flow are specified in terms of U, potential flow effects are of primary importance, and the determination of P is unimportant, one may use an alternative, simpler, kinematic description of the mixture:

$$\nabla\cdot U = 0, \quad \nabla\times[\,U+4\pi n\bar{D}] = 0 \tag{87}$$

obtained by taking a curl of (33). In this case the complete set of equations describing the two-phase flow consists of equations (5), (13), (79), (80), (82), (83) and (87).

It is useful to write (79) in terms of the volumetric flow rate U instead of G as

$$n\left(\frac{\partial \bar{\boldsymbol{p}}}{\partial t}+\bar{\boldsymbol{w}}\cdot\nabla\bar{\boldsymbol{p}}\right) = 4\pi\rho n\bar{\boldsymbol{D}}\cdot\nabla U+4\pi\rho n\bar{\boldsymbol{D}}\times\nabla\times U-\nabla\cdot[n\boldsymbol{\tau}^k+n\boldsymbol{\tau}^p], \tag{88}$$

where

$$n\tau^p_{ij} = \sigma^*_{ij}-8\pi^2\rho(n\bar{D})^2\delta_{ij}. \tag{89}$$

For one-dimensional flow U is independent of x and the first two terms in the right-hand side of (88) vanish. In this case the particle momentum is therefore only dependent on $\boldsymbol{\tau}^k$ and $\boldsymbol{\tau}^p$. For random arrays our calculations with the point-dipole approximation give

$$n\tau^p_{ij} = (4\pi n)^2\rho[-\tfrac{2}{5}\bar{D}^2\delta_{ij}+\tfrac{1}{5}\bar{D}_i\bar{D}_j]. \tag{90}$$

Note that, for dipole moments aligned in the x-direction so that $\bar{D}_i = \bar{D}\delta_{i1}$, this yields

$$n\tau^p_{11} = -\tfrac{1}{5}\rho(4\pi n\bar{D})^2, \quad n\,\mathrm{Tr}\,(\boldsymbol{\tau}^p) = -\rho(4\pi n\bar{D})^2. \tag{91}$$

In the case of a simple cubic array of particles, our equations (52) and (89) together with the fact that $\partial_{ij}S^r_1(0) = (4/3\mathscr{V})\pi\delta_{ij}$, yield instead

$$n\tau^p_{11} = \lambda\rho(4\pi n\bar{D})^2, \quad n\,\mathrm{Tr}\,(\boldsymbol{\tau}^p) = -\rho(4\pi n\bar{D})^2, \tag{92}$$

with $\lambda = -(\mathscr{V}/4\pi)\,\partial_{1111}S^r_2(0) \approx 0.1716$. This result for τ^p_{11} is interestingly in agreement with that given in SD because the two errors mentioned in §4 exactly cancel each other. On the other hand, the trace of $\boldsymbol{\tau}^p$ as given in SD is in error.

It may be noted that while we have discussed specific results for the two very special cases of periodic suspension and random suspension with uniform dipole distribution, the general expressions derived here have wider applicability and can be used to determine stress in a more general flow by combining it with either a suitable kinetic theory or numerical simulations.

The authors express their gratitude to Dr D. Z. Zhang for his participation in the early stages of this work. H.B. is also grateful to Professor L. van Wijngaarden and Dr A. Biesheuvel for many discussions and continuous encouragement. The work of the first two authors has been supported by DOE and NSF under grants DE-FG02-89ER14043 and CTS-8918144 respectively. A.S.S. has been supported by NSF under grants CTS-9118675 and CTS-9307723.

REFERENCES

Batchelor, G. K. 1967 *An Introduction to Fluid Dynamics*. Cambridge University Press.

Biesheuvel, A. & Gorissen, W. C. M. 1990 Void fraction disturbances in a uniform bubbly fluid. *Intl J. Multiphase Flow* **16**, 211–231.

Biesheuvel, A. & Spoelstra, S. 1989 The added mass coefficient of a dispersion of spherical gas bubbles in liquid. *Intl J. Multiphase Flow* **15**, 911–924.

Biesheuvel, A. & Wijngaarden, L. van 1984 Two-phase flow equations for a dilute dispersion of gas bubbles in liquid. *J. Fluid Mech.* **148**, 301–318.

Groot, S. R. de & Mazur, P. 1962 *Non-equilibrium Thermodynamics*. North-Holland.

Hasimoto, H. 1959 On the periodic fundamental solutions of the Stokes equations and their applications to viscous flow past a cubic array of spheres. *J. Fluid Mech.* **5**, 317–329.

Hinch, E. J. 1977 An averaged-equation approach to particle interactions in a fluid suspension. *J. Fluid Mech.* **83**, 695–720.

Irving, J. H. & Kirkwood, J. G. 1950 The statistical mechanical theory of transport processes. IV. The equations of hydrodynamics. *J. Chem. Phys.* **18**, 817–829.

KUMARAN, V. & KOCH, D. L. 1993 The rate of coalescence in a suspension of high Reynolds number, low Weber number bubbles. *Phys. Fluids* A **5**, 1135–1140.

MAZUR, P. & GROOT, S. R. DE 1956 On the pressure and ponderomotive force in a dielectric. *Physica* **22**, 657–669.

MILNE-THOMSON, L. M. 1968 *Theoretical Hydrodynamics*, 5th edn. Macmillan.

O'BRIEN, R. W. 1978 A method for the calculation of the effective transport properties of suspensions of interacting particles. *J. Fluid Mech.* **91**, 17–39.

RICE, S. A. & GRAY, P. 1965 *The Statistical Mechanics of Simple Liquids*. John Wiley & Sons.

SANGANI, A. S. & ACRIVOS, A. 1983 The effective conductivity of a periodic array of spheres. *Proc. R. Soc. Lond.* A **386**, 263–275.

SANGANI, A. S. & DIDWANIA, A. K. 1993*a* Dispersed-phase stress tensor in flows of bubbly liquid at large Reynolds numbers. *J. Fluid Mech.* **248**, 27–54 (referred to herein as SD).

SANGANI, A. S. & DIDWANIA, A. K. 1993*b* Dynamic simulations of flows of bubbly liquid at large Reynolds number. *J. Fluid Mech.* **250**, 307–337.

SANGANI, A. S., KANG, S.-Y., KOCH, D. L. & TSAO, H.-K. 1994 Numerical simulations and kinetic theory for simple shear motion of bubbly suspensions. In *Proc. IUTAM Symp. on Waves in Liquid/Gas and Liquid/Vapor Two-Phase System, Kyoto* (ed. L. van Wijngaarden & S. Morioka). To be published by Kluwer.

SANGANI, A. S., ZHANG, D. Z. & PROSPERETTI, A. 1991 The added mass, Basset, and viscous drag coefficients in nondilute bubbly liquids undergoing small amplitude oscillatory motion. *Phys. Fluids* A **3**, 2955–2970.

WALLIS, G. B. 1991 The averaged Bernoulli equation and macroscopic equations of motion for the potential flow of a two-phase dispersion. *Intl J. Multiphase Flow* **17**, 683–695.

WIJNGAARDEN, L. VAN 1976 Hydrodynamic interaction between gas bubbles in liquid. *J. Fluid Mech.* **77**, 27–44.

WIJNGAARDEN, L. VAN 1993 The mean rise velocity of pairwise-interacting bubbles in liquid. *J. Fluid Mech.* **251**, 55–78.

WIJNGAARDEN, L. VAN & KAPTEYN, C. 1990 Concentration waves in dilute bubble/liquid mixtures. *J. Fluid Mech.* **212**, 111–137.

ZHANG, D. Z. & PROSPERETTI, A. 1994 Averaged equations for inviscid disperse two-phase flow. *J. Fluid Mech.* **267**, 185–219.