# Virtual mass and drag in two-phase flow

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We study virtual mass and drag effects in a fluid suspension consisting of spherical particles immersed in an incompressible, nearly inviscid fluid. We derive average equations of motion for the fluid phase and the particle phase by the method of ensemble averaging. We show that the virtual mass and drag coefficients may be expressed exactly in terms of the dielectric constant of a corresponding dielectric suspension with the same distribution of particles. We make numerical predictions for the case of an equilibrium distribution of hard spheres.

# 1. Introduction

It is well known that a single body suspended in an incompressible, inviscid fluid experiences a reaction force when accelerated, due to the inertia of the surrounding fluid (Landau & Lifshitz 1961; Batchelor 1967; Milne-Thomson 1968; Lighthill 1986). For a spherical particle the corresponding virtual mass is just one half of the mass of the fluid displaced by the particle (Kelvin, see Lamb 1932, chap. 6). In the case of many particles the virtual mass is modified owing to hydrodynamic interactions. For a suspension of particles one may define a transport coefficient which plays the role of virtual mass in the average equation of motion. One wishes to find this coefficient as a function of the volume fraction occupied by particles. Similarly, for a slightly viscous fluid the drag coefficient is modified from its singleparticle value owing to hydrodynamic interactions. One wishes to find the drag coefficient appearing in the average equation of motion as a function of volume fraction.

The concentration dependence of the virtual mass has been studied by many authors (Zuber 1964; Buyevich 1971; van Wijngaarden 1976; Cook & Harlow 1984; Geurst 1985; Kok 1988; Biesheuvel & Spoelstra 1989). At low concentration one may consider the series expansion of the virtual mass in powers of the volume fraction. For the case of massless particles, or bubbles, van Wijngaarden (1976) has calculated the coefficient of the linear term in the expansion. For higher concentrations only approximate results have been obtained. In an early article Zuber (1964) derived an approximate expression for the virtual mass on the basis of a cell model. Van Wijngaarden's coefficient does not differ much from the value obtained by expansion of Zuber's expression in powers of the volume fraction.

In the following we show that the virtual mass and the drag coefficient may be expressed exactly in terms of the effective dielectric constant of a related dielectric suspension of spherical particles with the same geometric distribution. This mapping allows one to derive results for virtual mass and drag from results obtained for the

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dielectric problem. We find a different value for the coefficient calculated by van Wijngaarden. Zuber's expression for the virtual mass follows by approximating the dielectric constant of the related problem by its Clausius-Mossotti value. On the basis of the theory of dielectrics we propose an approximation scheme which allows accurate evaluation of the virtual mass and drag coefficient even at high volume fractions.

Our analysis is based on the linearized Navier-Stokes equations for an incompressible fluid. Thus we study only the limit of low Reynolds number. In the microscopic picture we consider a disordered configuration of identical spheres which are subject to small applied forces oscillating at a given frequency. As a consequence the spheres perform small oscillations about their rest positions. We study the limit of small viscosity, or equivalently the limit of high frequency. In this limit the fluid flow may be derived from a potential, except in a thin boundary layer surrounding each sphere. For the linear flow situation under consideration we obtain macroscopic equations of motion for the two-phase system by averaging over an ensemble of configurations. The virtual mass is found as an inertial coefficient in the linear macroscopic equations. The drag coefficient is found as a transport coefficient describing the friction between the two phases. Owing to hydrodynamic interactions both coefficients depend on the concentration of spheres.

In the following two sections we consider first the solution of the flow equations for a single sphere. Subsequently we study the hydrodynamic interactions between many spheres and perform the ensemble average. We show that the virtual mass and drag coefficient may be obtained from a related dielectric problem. The macroscopic equations may be written in a variety of different forms which lead to various possible definitions of the virtual mass. We indicate the relation between different definitions which have appeared in the literature.

# 2. Single-particle equation of motion

We consider a spherical particle of radius a and mass  $m_p$  immersed in an incompressible fluid of mass density  $\rho$ . We study first the case of an ideal fluid and neglect viscosity. We denote the local fluid flow velocity by v(r) and the fluid pressure by p(r). The instantaneous position of the centre of the sphere is denoted by R and its translational velocity by U. The sphere is assumed to be impermeable to the flow. Hence the velocity components normal to the surface must satisfy the kinematic boundary condition

$$v_{\mathbf{n}} = U_{\mathbf{n}} \quad \text{at} \quad |\mathbf{r} - \mathbf{R}| = a. \tag{2.1}$$

If the flow is irrotational the flow velocity can be derived from a scalar potential  $\phi$  according to

$$\boldsymbol{v} = -\boldsymbol{\nabla}\boldsymbol{\phi}.\tag{2.2}$$

Incompressibility of the fluid is expressed by  $\nabla \cdot \boldsymbol{v} = 0$ . As a consequence the potential  $\phi$  satisfies the Laplace equation  $\nabla^2 \phi = 0$ . If the flow at infinity is uniform the instantaneous flow pattern is

$$\boldsymbol{v}(\boldsymbol{r}) = \boldsymbol{v}_0 - \frac{1}{2} a^3 \frac{I - 3 \hat{\boldsymbol{r}}_0 \, \hat{\boldsymbol{r}}_0}{r_0^3} \cdot (\boldsymbol{U} - \boldsymbol{v}_0), \quad \boldsymbol{r}_0 > a, \qquad (2.3)$$

where  $r_0 = r - R$ . The force exerted on the sphere by the fluid is given by

$$\boldsymbol{K} = -\int p \boldsymbol{\dot{r}}_0 \,\mathrm{d}S,\tag{2.4}$$

where the integral is over a spherical surface just enclosing the particle. The force is found to be (Landau & Lifshitz 1961; Batchelor 1967)

$$\boldsymbol{K} = -\frac{1}{2}m_{\rm f}\frac{\mathrm{d}\boldsymbol{U}}{\mathrm{d}t} + \frac{3}{2}m_{\rm f}\frac{\mathrm{d}\boldsymbol{v}_0}{\mathrm{d}t},\tag{2.5}$$

where  $m_{\rm f} = (\frac{4}{3}\pi) \rho a^3$  is the mass of the displaced fluid. If an external force E is applied to the particle, then the acceleration of the particle is given by

$$(m_{\rm p} + \frac{1}{2}m_{\rm f})\frac{{\rm d}U}{{\rm d}t} = E + \frac{3}{2}m_{\rm f}\frac{{\rm d}v_0}{{\rm d}t}.$$
 (2.6)

This shows that the effective mass of the particle is the sum of its own mass  $m_p$  and the added mass  $\frac{1}{2}m_r$ . In the following we wish to investigate how the added mass is modified by hydrodynamic interactions in a system of many spheres. We wish to include the frictional force and consider a slightly viscous fluid described by the linearized equations of motion

$$\rho \frac{\partial \boldsymbol{v}}{\partial t} = \eta \nabla^2 \boldsymbol{v} - \nabla p, \quad \nabla \cdot \boldsymbol{v} = 0,$$
(2.7)

where  $\eta$  is the shear viscosity. The kinematic boundary condition (2.1) must then be supplemented with an equation relating the velocity components of fluid and particle tangential to the surface of the sphere. We use the mixed slip-no-slip boundary condition (Felderhof 1976)

$$\boldsymbol{v}_{t} - (\boldsymbol{U} + \boldsymbol{\Omega} \times \boldsymbol{r}_{0})_{t} = \frac{ca}{\eta} (\boldsymbol{\sigma} \cdot \boldsymbol{n})_{t} \text{ at } \boldsymbol{r}_{0} = a,$$
 (2.8)

where  $\Omega$  is the rotational velocity of the sphere. The proportionality constant c plays the role of slip parameter and  $(\boldsymbol{\sigma} \cdot \boldsymbol{n})_t$  is the normal-tangential component of the fluid stress tensor  $\boldsymbol{\sigma}$ , defined by

$$\sigma_{\alpha\beta} = \eta (\partial_{\alpha} v_{\beta} + \partial_{\beta} v_{\alpha}) - p \delta_{\alpha\beta}.$$
(2.9)

We consider small translational motions of the sphere about its rest position R due to an applied force E(t) and a uniform incident flow  $v_0(t)$ . Because of linearity it is convenient to do a Fourier decomposition in time and to consider harmonic time variation of the force  $E(t) = E_{\omega} \exp(-i\omega t)$  and incident flow  $v_0(t) = v_{0\omega} \exp(-i\omega t)$ . In the limit of small viscosity the solution of the flow equations (2.7) reads (Felderhof & Jones 1986)

$$\boldsymbol{v}_{\omega}(\boldsymbol{r}) = \boldsymbol{v}_{0\omega} - \frac{1}{2}a^{3} \frac{1 - 3\hat{\boldsymbol{r}}_{0} \hat{\boldsymbol{r}}_{0}}{r_{0}^{3}} \cdot (\boldsymbol{U}_{\omega} - \boldsymbol{v}_{0\omega}) + 3\left(1 + \frac{1}{2c}\right) \frac{\exp\left[-\alpha(r_{0} - \alpha)\right]}{\alpha r_{0}} (1 - \hat{\boldsymbol{r}}_{0} \hat{\boldsymbol{r}}_{0}) \cdot (\boldsymbol{U}_{\omega} - \boldsymbol{v}_{0\omega}), \quad r_{0} > a, \quad (2.10)$$

where  $\alpha = (-i\omega\rho/\eta)^{\frac{1}{2}}$ , Re ( $\alpha$ ) > 0. The slip parameter c takes the value c = 0 for noslip and the value  $c = \infty$  for perfect-slip boundary conditions. Clearly the first two terms on the right in (2.10) correspond to the solution (2.3). The last term describes a thin boundary layer near the surface. The force exerted by the fluid on the sphere now follows from

$$\boldsymbol{K}_{\omega} = \int \boldsymbol{\sigma}_{\omega} \cdot \boldsymbol{\hat{r}}_{0} \, \mathrm{d}S. \tag{2.11}$$

As a result one finds, for small viscosity,

$$\boldsymbol{K}_{\omega} = \left(\frac{3}{2}\,\mathrm{i}\omega \boldsymbol{m}_{\mathrm{f}} - \zeta\right)\left(\boldsymbol{U}_{\omega} - \boldsymbol{v}_{0\omega}\right) - \mathrm{i}\omega \boldsymbol{m}_{\mathrm{f}}\,\boldsymbol{U}_{\omega},\tag{2.12}$$

with the friction coefficient (Felderhof 1976)

$$\zeta = 6\pi\eta a \left(2 + \frac{1}{c}\right). \tag{2.13}$$

The linearized equation of motion for the particle may be written

$$\left[-\mathrm{i}\omega(m_{\mathrm{p}}+\frac{1}{2}m_{\mathrm{f}})+\zeta\right]\boldsymbol{U}_{\omega}=\boldsymbol{E}_{\omega}-\left(\tfrac{3}{2}\mathrm{i}\omega m_{\mathrm{f}}-\zeta\right)\boldsymbol{v}_{0\omega}. \tag{2.14}$$

This corresponds precisely to (2.6) with the additional effect of friction included.

We note that for perfect slip  $(c = \infty)$  the friction coefficient takes the value  $\zeta = 12\pi\eta a$ . This is precisely the value calculated by Levich (1962) from the energy dissipation in the irrotational flow pattern (2.3). The expression for the energy dissipation is quadratic in the flow velocity, but may be derived from the linear equation (2.7), so that our calculation is consistent with that given by Levich. As shown by Levich, in the case of irrotational flow the energy dissipation may be expressed in terms of a surface integral. The additional friction for finite values of the slip parameter is due to additional dissipation in the boundary layer described by the last term in (2.10), where the flow has non-zero vorticity.

The dimensionless parameter characterizing the importance of nonlinearity is given by the Reynolds number  $\rho a U/\eta$ . The dimensionless parameter characterizing the importance of the viscous stress for the rate of change of momentum is given by  $(|\alpha|a)^{-2} = \eta/\omega\rho a^2$ . The above equations are valid in the limit where both parameters are small. In the following we shall consider small motions of a many-sphere system under the same conditions. We shall derive a generalization of the equation of motion (2.14) with an added-mass term and a generalized friction coefficient dependent on the local density of spheres.

## 3. Potential flow about a sphere

For the study of hydrodynamic interactions it turns out to be convenient to reformulate the boundary-value problem. In this section we consider flow about a single sphere on the basis of the linear equations (2.7). The thin-boundary-layer effect is not treated explicitly, except in the calculation of the force exerted by the fluid on the sphere, where we shall use a slight generalization of (2.12). In this manner friction is included in the calculation.

For simplicity of notation we shall often omit the subscript  $\omega$ , it being understood that we are dealing with a single Fourier component at frequency  $\omega$ . We consider the potential flow problem for a single sphere centred at the origin. It follows from (2.3) that the potential due to its velocity U is given by

$$\phi_U(\mathbf{r}) = \frac{1}{2}a^3 \frac{\dot{\mathbf{r}}}{r^2} \cdot \mathbf{U}, \quad r > a.$$
 (3.1)

In the statistical averaging procedure to be applied in the many-sphere problem it is convenient to deal with equations valid everywhere in space. We specify the potential inside the sphere as

$$\phi(\mathbf{r}) = \phi_U(\mathbf{r}) = -\mathbf{r} \cdot \mathbf{U}, \quad \mathbf{r} < a. \tag{3.2}$$

Hence Laplace's equation is satisfied everywhere, except on the surface of the sphere. It is easily seen that the normal derivative of  $\phi$  is continuous across the surface, but that  $\phi$  itself is discontinuous. Hence we can write

$$\nabla^2 \phi = 4\pi\sigma \delta'(r-a), \tag{3.3}$$

where  $\sigma$  must be chosen such that

$$\phi|_{a+} - \phi|_{a-} = 4\pi\sigma. \tag{3.4}$$

In electrical language  $\sigma$  may be identified with the radial component of a surface polarization  $P^{s}$  (Jackson 1962). Explicitly

$$\boldsymbol{P}_t^{\mathrm{S}} = 0, \quad \boldsymbol{P}_r^{\mathrm{S}} = \boldsymbol{\sigma}. \tag{3.5}$$

For the potential  $\phi_U(\mathbf{r})$  given by (3.1) and (3.2) we find

$$\boldsymbol{P}_{\boldsymbol{u}}^{\mathrm{s}} = \frac{3a}{8\pi} \boldsymbol{\hat{r}} \boldsymbol{\hat{r}} \cdot \boldsymbol{U}. \tag{3.6}$$

The dipole moment of the sphere is given by

$$\boldsymbol{p}_U = \frac{1}{2} a^3 \boldsymbol{U} \tag{3.7}$$

in agreement with (3.1).

In the presence of an incident flow  $v_0 = -\nabla \phi_0$  we obtain the total potential

$$\phi(\mathbf{r}) = \phi_U(\mathbf{r}) + \phi_0(\mathbf{r}) + \phi_{\text{ind}}(\mathbf{r}). \tag{3.8}$$

The potential  $\phi_0(\mathbf{r})$  satisfies Laplace's equation and may be expanded in spherical harmonics,

$$\phi_0(\mathbf{r}) = \sum_{lm} A_{lm} r^l Y_{lm}(\theta, \varphi).$$
(3.9)

The induced potential  $\phi_{ind}(r)$  precisely cancels  $\phi_0$  for r < a, and for r > a has the harmonic expansion

$$\phi_{\rm ind}(\mathbf{r}) = \sum_{lm} B_{lm} \, r^{-l-1} Y_{lm}(\theta, \varphi), \quad r > a. \tag{3.10}$$

By use of the kinematic boundary condition (2.1) we find

Hence the induced surface polarization is given by

$$P_{\rm ind}^{\rm s} = \frac{1}{4\pi} \sum_{lm} \frac{2l+1}{l+1} A_{lm} a^l Y_{lm}(\theta, \varphi) \,\hat{\mathbf{r}}. \tag{3.12}$$

In particular, if the sphere is placed at rest in a uniform flow  $v_0$ , then a surface polarization

$$\boldsymbol{P}_{\text{ind}}^{\text{s}} = -\frac{3a}{8\pi} \hat{\boldsymbol{r}} \hat{\boldsymbol{r}} \cdot \boldsymbol{v}_{0}$$
(3.13)

is induced. The corresponding dipole moment is

$$\boldsymbol{p}_{\rm ind} = -\frac{1}{2}a^3 \boldsymbol{v}_0. \tag{3.14}$$

The total dipole moment for a moving sphere in an incident flow  $v_0(r)$  is given by

$$\boldsymbol{p} = \frac{1}{2}a^3(\boldsymbol{U} - \boldsymbol{v}_0(0)). \tag{3.15}$$

The higher-order multipole moments may be expressed in terms of the derivatives of the incident flow velocity  $v_0(r)$  at the origin.

If an external force E is applied to the particle, then in the absence of an incident flow it acquires a velocity  $U_E$ , which may be calculated from (2.14). The corresponding dipole moment is

$$\boldsymbol{p}_E = \boldsymbol{\alpha}_E \boldsymbol{E} \tag{3.16}$$

with polarizability

$$\alpha_E = \frac{-1}{i\omega(m_f + 2m_p) - 2\zeta} a^3.$$
(3.17)

In an incident flow  $v_0 = -\nabla \phi_0$  the linearized equation of motion for the particle becomes (Felderhof 1976)

$$\left[-\mathrm{i}\omega(m_{\mathrm{p}} + \frac{1}{2}m_{\mathrm{f}}) + \zeta\right] U = E - \left(\frac{3}{2}\mathrm{i}\omega m_{\mathrm{f}} - \zeta\right) v_{0}(0), \tag{3.18}$$

a slight generalization of (2.14). Hence the total dipole moment may be written as

$$\boldsymbol{p} = \boldsymbol{p}_E + \hat{\boldsymbol{p}},\tag{3.19}$$

with **\hat{p}** given by

$$\hat{\boldsymbol{p}} = \hat{\alpha} \boldsymbol{v}_0(0), \qquad (3.20)$$

with polarizability

$$\hat{\alpha} = \frac{\mathrm{i}\omega(m_{\mathrm{f}} - m_{\mathrm{p}})}{\mathrm{i}\omega(m_{\mathrm{f}} + 2m_{\mathrm{p}}) - 2\zeta} a^{3}, \qquad (3.21)$$

as follows from (3.15) and (3.18). The surface polarization may be decomposed in similar manner,

$$\boldsymbol{P}^{\mathrm{S}} = \boldsymbol{P}^{\mathrm{S}}_{E} + \boldsymbol{\hat{P}}^{\mathrm{S}}, \tag{3.22}$$

where  $P_E^S$  is given by

$$\boldsymbol{P}_{E}^{s} = \frac{3\alpha_{E}}{4\pi a^{2}} \boldsymbol{\hat{r}} \boldsymbol{\hat{r}} \cdot \boldsymbol{E}$$
(3.23)

and  $\hat{P}^{s} = P_{U}^{s} + P_{ind}^{s} - P_{E}^{s}$  is linear in  $v_{0}(r)$ . In our theory the effect of friction appears only in the polarizabilities  $\alpha_{E}$  and  $\hat{\alpha}$ .

Finally we note that with the polarization

$$\boldsymbol{P}(\boldsymbol{r}) = \boldsymbol{P}^{\mathrm{S}}(\hat{\boldsymbol{r}})\,\delta(\boldsymbol{r}-\boldsymbol{a}) \tag{3.24}$$

and the velocity field  $\boldsymbol{v} = -\nabla \phi$  we may define the vector field

$$\boldsymbol{u} = \boldsymbol{v} + 4\pi \boldsymbol{P}. \tag{3.25}$$

In the electrical analogy u is the dielectric displacement. Hence the fields u and v satisfy the equations

$$\nabla \cdot \boldsymbol{u} = 0, \quad \nabla \times \boldsymbol{v} = 0. \tag{3.26}$$

These equations remain valid in the case of many spheres. The above concepts are useful in the study of hydrodynamic interactions between spheres.

### 4. Hydrodynamic interactions

We consider N identical spheres immersed in an incompressible fluid of infinite extent. The spheres make small movements about positions  $\mathbf{R}_1, \ldots, \mathbf{R}_N$  caused by applied forces  $E_1(t), \ldots, E_N(t)$  and by an irrotational flow  $\mathbf{v}_0(\mathbf{r}, t)$  incident from infinity. The fluid velocity  $\mathbf{v}(\mathbf{r}, t)$  and the pressure  $p(\mathbf{r}, t)$  are assumed to satisfy the

linear equations (2.7). We do a Fourier analysis in time and consider the limit of high frequency at fixed viscosity so that  $\eta/\omega\rho a^2$  is a small parameter. We neglect the thin boundary layer surrounding each sphere, except in its effect on the force exerted on the sphere by the fluid. Thus the whole flow is irrotational and we may put

$$\boldsymbol{v}_{\boldsymbol{0}\boldsymbol{\omega}} = -\boldsymbol{\nabla}\boldsymbol{\phi}_{\boldsymbol{0}\boldsymbol{\omega}}, \quad \boldsymbol{v}_{\boldsymbol{\omega}} = -\boldsymbol{\nabla}\boldsymbol{\phi}_{\boldsymbol{\omega}}. \tag{4.1}$$

This reduces the problem of hydrodynamic interactions to a problem in potential theory.

We employ the device introduced in the preceding section and describe the effect of each sphere on the flow in terms of a surface polarization. The potential  $\phi$  and the flow velocity v inside each sphere follow from its translational velocity. The total polarization is given by

$$\boldsymbol{P}(\boldsymbol{r}) = \sum_{j=1}^{N} \boldsymbol{P}_{j}(\boldsymbol{r}) = \sum_{j=1}^{N} \boldsymbol{P}_{j}^{\mathrm{S}}(\boldsymbol{r}) \,\delta(|\boldsymbol{r}-\boldsymbol{R}_{j}|-a). \tag{4.2}$$

The corresponding potential is

$$\phi(\mathbf{r}) = \phi_0(\mathbf{r}) + \int \frac{\mathbf{r} - \mathbf{r}'}{|\mathbf{r} - \mathbf{r}'|^3} \cdot \mathbf{P}(\mathbf{r}') \,\mathrm{d}\mathbf{r}'. \tag{4.3}$$

Each surface polarization  $P_i^s$  is decomposed as in (3.22) with  $P_{iE}^s$  given by

$$(3\alpha_E/4\pi a^2)\,\hat{r}\hat{r}\cdot E_j,$$

and  $\hat{\boldsymbol{P}}_{j}^{s}$  linear in the flow pattern incident on sphere j.

The flow velocity may be expressed as

$$\boldsymbol{v}(\boldsymbol{r}) = \boldsymbol{v}_0(\boldsymbol{r}) + \int \boldsymbol{G}_0(\boldsymbol{r} - \boldsymbol{r}') \cdot \boldsymbol{P}(\boldsymbol{r}') \, \mathrm{d}\boldsymbol{r}', \qquad (4.4)$$

with a Green's function which follows by differentiation of (4.3). The integral in (4.4) must be interpreted as

$$\int \boldsymbol{G}_{0}(\boldsymbol{r}-\boldsymbol{r}') \cdot \boldsymbol{P}(\boldsymbol{r}') \, \mathrm{d}\boldsymbol{r}' = -\frac{4\pi}{3} \boldsymbol{P}(\boldsymbol{r}) + \int_{\delta} \boldsymbol{G}_{0}(\boldsymbol{r}-\boldsymbol{r}') \cdot \boldsymbol{P}(\boldsymbol{r}') \, \mathrm{d}\boldsymbol{r}', \qquad (4.5)$$

where the symbol  $\delta$  on the second integral indicates that an infinitesimal sphere of radius  $\delta$  centred at r must be excluded from the integration. The flow pattern acting on sphere j may be expressed as

$$\boldsymbol{v}_{j}^{a}(\boldsymbol{r}) = \boldsymbol{v}_{0}(\boldsymbol{r}) + \sum_{k+j} \int \boldsymbol{G}_{0}(\boldsymbol{r} - \boldsymbol{r}') \cdot \boldsymbol{P}_{k}(\boldsymbol{r}') \, \mathrm{d}\boldsymbol{r}'.$$
(4.6)

The polarization on sphere j is given by

$$P_{j}(\mathbf{r}) = P_{jE}(\mathbf{r}) + \int \mathbf{M}(j; \mathbf{r}, \mathbf{r}') \cdot \mathbf{v}_{j}^{\mathrm{a}}(\mathbf{r}') \,\mathrm{d}\mathbf{r}'$$
(4.7)

with a linear operator  $\mathbf{M}(j)$  which may be found from (3.22). Substituting (4.6) we find that the polarization  $P(\mathbf{r})$  may be expressed in terms of a multiple scattering expansion with propagation between successive particles described by the Green's function  $\mathbf{G}_{0}$ .

For mathematical convenience we assume that the applied forces  $\{E_j\}$  may be derived from a vector field E(r), independent of the configuration  $R_1, \ldots, R_N$ , according to the rule

$$E_j = E(R_j) = \int E(r) \,\delta(r - R_j) \,\mathrm{d}r. \tag{4.8}$$

It is clear that the total polarization P(r) is linear in the field E(r) as well as in the incident flow  $v_0(r)$ , and may therefore be written

$$\boldsymbol{P}(\boldsymbol{r}) = \int \boldsymbol{\mathsf{T}}_{ME}(\boldsymbol{r}, \boldsymbol{r}'; \boldsymbol{R}_1, \dots, \boldsymbol{R}_N) \cdot \boldsymbol{E}(\boldsymbol{r}') \, \mathrm{d}\boldsymbol{r}' + \int \boldsymbol{\mathsf{T}}_{MM}(\boldsymbol{r}, \boldsymbol{r}'; \boldsymbol{R}_1, \dots, \boldsymbol{R}_N) \cdot \boldsymbol{v}_0(\boldsymbol{r}') \, \mathrm{d}\boldsymbol{r}' \quad (4.9)$$

with linear operators  $\mathbf{T}_{ME}$  and  $\mathbf{T}_{MM}$  dependent on the configuration. In shorthand notation

$$\boldsymbol{P} = \boldsymbol{\mathsf{T}}_{\boldsymbol{M}\boldsymbol{E}} \cdot \boldsymbol{E} + \boldsymbol{\mathsf{T}}_{\boldsymbol{M}\boldsymbol{M}} \cdot \boldsymbol{v}_{\mathbf{0}}. \tag{4.10}$$

The operators  $\mathbf{T}_{ME}$  and  $\mathbf{T}_{MM}$  may each be expressed in terms of a multiple scattering expansion.

We have arrived at a description which is formally analogous to that encountered in creeping-flow hydrodynamics (Felderhof 1988). We proceed in the same way and arrive at a macroscopic description by averaging the equations over a probability distribution  $P(\mathbf{R}_1, \ldots, \mathbf{R}_N)$  which is assumed known. We assume that the spheres are distributed approximately uniformly in a volume  $\Omega$ . Thus we find, by averaging (4.10)

$$\langle \boldsymbol{P} \rangle = \langle \mathbf{T}_{ME} \rangle \cdot \boldsymbol{E} + \langle \mathbf{T}_{MM} \rangle \cdot \boldsymbol{v}_0.$$
 (4.11)

From (4.4) we find for the average flow velocity

$$\langle \boldsymbol{v} \rangle = \boldsymbol{v}_0 + \boldsymbol{G}_0 \cdot \langle \boldsymbol{P} \rangle. \tag{4.12}$$

Hence it follows that the average flow velocity satisfies

$$\nabla \cdot \langle \boldsymbol{v} \rangle = -4\pi \nabla \cdot \langle \boldsymbol{P} \rangle, \quad \nabla \times \langle \boldsymbol{v} \rangle = 0.$$
(4.13)

By use of the definition  $\boldsymbol{u} = \boldsymbol{v} + 4\pi \boldsymbol{P}$ , as in (3.25), we may rewrite these equations as

$$\nabla \cdot \langle \boldsymbol{u} \rangle = 0, \quad \nabla \times \langle \boldsymbol{v} \rangle = 0. \tag{4.14}$$

Substituting (4.12) in (4.11) and eliminating  $v_0$  we find

$$\langle P \rangle = \mathbf{X}_{ME} \cdot E + \mathbf{X}_{MM} \cdot \langle v \rangle, \qquad (4.15)$$

with the linear operators

$$\mathbf{X}_{ME} = (\mathbf{I} + \langle \mathbf{T}_{MM} \rangle \, \mathbf{G}_0)^{-1} \langle \mathbf{T}_{ME} \rangle, \quad \mathbf{X}_{MM} = \langle \mathbf{T}_{MM} \rangle \, (\mathbf{I} + \mathbf{G}_0 \langle \mathbf{T}_{MM} \rangle)^{-1}.$$
(4.16)

The corresponding kernels are short range in the difference  $\mathbf{r} - \mathbf{r}'$ , in contrast to the kernels  $\mathbf{T}_{ME}$  and  $\mathbf{T}_{MM}$ . Thus the relation (4.15) is local in nature, in contrast to (4.11).

We are also interested in the motion of the particles and therefore consider the particle current density, defined by

$$\boldsymbol{J}(\boldsymbol{r};\boldsymbol{R}_1,\ldots,\boldsymbol{R}_N) = \sum_{j=1}^N \boldsymbol{U}_j \,\delta(\boldsymbol{r}-\boldsymbol{R}_j). \tag{4.17}$$

The current density is also linear in E(r) and  $v_0(r)$ , and therefore we have, in analogy to (4.10),

$$\boldsymbol{J} = \boldsymbol{\mathsf{T}}_{JE} \cdot \boldsymbol{E} + \boldsymbol{\mathsf{T}}_{JM} \cdot \boldsymbol{v}_0. \tag{4.18}$$

Averaging this equation we obtain

$$\langle J \rangle = \langle \mathsf{T}_{JE} \rangle \cdot E + \langle \mathsf{T}_{JM} \rangle \cdot v_0. \tag{4.19}$$

Substituting (4.12) and eliminating  $v_0$  we find

$$\langle J \rangle = \mathbf{X}_{JE} \cdot E + \mathbf{X}_{JM} \cdot \langle v \rangle \tag{4.20}$$

with the linear operators

$$\mathbf{X}_{JE} = \langle \mathbf{T}_{JE} \rangle - \mathbf{X}_{JM} \, \mathbf{G}_0 \langle \mathbf{T}_{ME} \rangle, \quad \mathbf{X}_{JM} = \langle \mathbf{T}_{JM} \rangle (\mathbf{I} + \mathbf{G}_0 \langle \mathbf{T}_{MM} \rangle)^{-1}.$$
(4.21)

Again the corresponding kernels are short range in the difference r-r'. We have shown in earlier work (Cichocki & Felderhof 1988*a*) that the kernels may be evaluated from a renormalized cluster expansion.

## 5. Transport coefficients

In this section we consider a situation where the macroscopic fields show a slow spatial variation. Correspondingly the short-range transport kernels  $X_{ME}, \ldots, X_{JM}$ given by (4.16) and (4.21) may be replaced by delta-functions with prefactors which may be identified as transport coefficients. One may derive exact expressions for the transport coefficients by taking the thermodynamic limit  $N \to \infty$ ,  $\Omega \to \infty$  at constant density  $n = N/\Omega$ . In this limit the transport kernels become translationally invariant. We shall not carry out the procedure in full detail, because the present problem can be related to a corresponding dielectric problem. The theory of the dielectric constant of a polarizable suspension within the framework of the renormalized cluster expansion has been worked out in detail in Cichocki & Felderhof (1988b).

The relation to the dielectric problem becomes evident if we note that all intermediate scattering processes involve the single-body polarization operator  $\mathbf{M}$ , defined in (4.7). In electrical language this operator describes how a sphere is polarized by an incident electric field, which may be identified with the acting field  $v^{a}(\mathbf{r})$ . The response is characterized by a set of multipole polarizabilities  $\{\alpha_{i}\}$  for  $l = 1, 2, \ldots$  From (3.11) and (3.20) we find that these polarizabilities are given by

$$\alpha_1 = \hat{\alpha}, \quad \alpha_l = \frac{-l}{l+1} a^{2l+1} \quad \text{for} \quad l = 2, 3, \dots$$
 (5.1)

The multipole polarizabilities  $\alpha_l$  for  $l \ge 2$  are identical with those for a sphere of vanishing dielectric constant in vacuum. The dipole polarizability differs because it contains a contribution due to the velocity of the freely moving sphere caused by the incident flow. A collection of spheres with the given probability distribution and with given single particle polarizabilities has a perfectly well-defined dielectric constant  $\epsilon$ . In our case the polarizabilities are specified by (5.1). The transport coefficients of interest in the present problem may be expressed in terms of the dielectric constant. These relations are exact and therefore a calculation of the transport coefficients is reduced to a calculation of the dielectric constant of the corresponding dielectric system.

It is well known that the dielectric constant of an isotropic system of polarizable spheres is well approximated by the Clausius–Mossotti (CM) formula

$$\frac{\epsilon_{\rm CM} - 1}{\epsilon_{\rm CM} + 2} = \frac{4}{3}\pi n\hat{\alpha}.$$
(5.2)

It therefore makes sense to use this as a first approximation and to express the results in terms of the deviation from the Clausius-Mossotti value. The CM-formula (5.2) is based on Lorentz' approximation to the average local field acting on a selected particle. According to Lorentz this field is approximated by

$$\boldsymbol{v}_{\rm L} = \langle \boldsymbol{v} \rangle + \frac{4}{3} \pi \langle \boldsymbol{P} \rangle, \tag{5.3}$$

where  $\langle v \rangle$  is the average Maxwell field in electrical language. The exact expression for the average local field may be written

$$\boldsymbol{F} = \langle \boldsymbol{v} \rangle + \frac{4}{3} \pi \gamma \langle \boldsymbol{P} \rangle, \qquad (5.4)$$

where the coefficient  $\gamma$  expresses the correction to the Lorentz value. The corresponding exact expression for the dielectric constant is

$$\epsilon = 1 + \frac{4\pi n\hat{\alpha}}{1 - (\frac{4}{3}\pi) n\hat{\alpha}\gamma},\tag{5.5}$$

or equivalently

$$\frac{\epsilon - 1}{\epsilon + 2} = \frac{\left(\frac{4}{3}\pi\right)n\hat{\alpha}}{1 - \left(\frac{4}{3}\pi\right)n\hat{\alpha}(\gamma - 1)}.$$
(5.6)

In earlier work on the dielectric constant (Cichocki & Felderhof 1988b) we have expressed  $\gamma$  as

$$\gamma = 1 + \lambda + \mu, \tag{5.7}$$

and have derived exact statistical mechanical expressions for the coefficients  $\lambda$  and  $\mu$ .

We now express the transport coefficients of the hydrodynamic problem in terms of the coefficient  $\gamma$ . We begin by noting that in the long-wavelength limit the average polarization may be identified with the average dipole density,

$$\langle \mathbf{P}(\mathbf{r}) \rangle \approx \langle \sum_{j} \mathbf{p}_{j} \, \delta(\mathbf{r} - \mathbf{R}_{j}) \rangle.$$
 (5.8)

In a generalization of (3.15) the dipole moment of sphere j is given by

$$\boldsymbol{p}_{j} = \frac{1}{2} a^{3} [\boldsymbol{U}_{j} - \boldsymbol{v}_{j}^{a}(\boldsymbol{R}_{j})].$$
 (5.9)

Hence we obtain in the long-wavelength limit

$$\langle \boldsymbol{P} \rangle = \frac{1}{2} a^3 [\langle \boldsymbol{J} \rangle - n \boldsymbol{F}].$$
 (5.10)

Substituting the average local field from (5.4) we therefore find

$$\langle \boldsymbol{P} \rangle = \frac{a^3}{2 + \gamma \varphi} [\langle \boldsymbol{J} \rangle - n \langle \boldsymbol{v} \rangle], \qquad (5.11)$$

where  $\varphi = (\frac{4}{3}\pi) na^3$  is the volume fraction occupied by spheres. The relation (5.11) is exact in the long-wavelength limit.

A second expression for the average polarization may be derived from the relation

$$\boldsymbol{p}_j = \boldsymbol{\alpha}_E \boldsymbol{E}_j + \hat{\boldsymbol{\alpha}} \boldsymbol{v}_j^{\mathrm{a}}(\boldsymbol{R}_j). \tag{5.12}$$

Hence we obtain in the long-wavelength limit

$$\langle \boldsymbol{P} \rangle = n \alpha_E \boldsymbol{E} + n \hat{\boldsymbol{\alpha}} \boldsymbol{F}. \tag{5.13}$$

Substituting from (5.4) we find

$$\langle \boldsymbol{P} \rangle = \chi \boldsymbol{E} + \kappa \langle \boldsymbol{v} \rangle, \tag{5.14}$$

with coefficients

$$\chi = n\alpha_E [1 - \frac{4}{3}\pi n\hat{\alpha}\gamma]^{-1}, \quad \kappa = n\hat{\alpha} [1 - \frac{4}{3}\pi n\hat{\alpha}\gamma]^{-1}.$$
(5.15)

Again the relation (5.14) is exact in the long-wavelength limit.

Equating (5.11) and (5.14) and making use of (3.17) and (3.21) we find the relation

$$\left[-\mathrm{i}\omega(m_{\mathrm{p}}+m_{\mathrm{a}})+\zeta_{a}\right]\langle J_{\omega}\rangle = nE_{\omega}+\left[-\mathrm{i}\omega(m_{\mathrm{f}}+m_{\mathrm{a}})+\zeta_{\mathrm{a}}\right]n\langle v_{\omega}\rangle, \tag{5.16}$$

where the added mass  $m_a$  is given by

$$m_{\rm a} = \frac{1 - \gamma \varphi}{2 + \gamma \varphi} m_{\rm f}, \tag{5.17}$$

and the effective friction coefficient  $\zeta_a$  by

$$\zeta_{\rm a} = \frac{2}{2 + \gamma \varphi} \zeta. \tag{5.18}$$

The relation (5.16) may be regarded as a generalization of the single-particle equation of motion (2.14) to a macroscopic equation of motion for the many-particle system.

To conclude this section we note that substitution of (5.14) into (4.14) yields

$$\boldsymbol{\nabla} \cdot [\boldsymbol{\epsilon} \langle \boldsymbol{v}_{\omega} \rangle] = -4\pi \boldsymbol{\nabla} \cdot (\boldsymbol{\chi} \boldsymbol{E}_{\omega}), \qquad (5.19)$$

where we have used the relation

$$\epsilon = 1 + 4\pi\kappa, \tag{5.20}$$

which follows from (5.5) and (5.15). It is evident from (5.19) that in general the relation between  $\langle v(\mathbf{r}) \rangle$  and  $E(\mathbf{r})$  is highly non-local. The determination of  $\langle v \rangle$  for given E in finite geometry is equivalent to the solution of a problem in electrostatics.

## 6. Average equations of motion

In this section we study the average equation of motion (5.16) in more detail. The equation is exact in the long-wavelength limit, with transport coefficients  $m_{\rm a}$  and  $\zeta_{\rm a}$  given by (5.17) and (5.18), where the coefficient  $\gamma$  must be found from the equivalent dielectric constant by use of (5.5).

We define the average particle velocity  $U_{\omega}$  and the mass-averaged fluid velocity  $V_{\omega}$  with the equations

$$\langle J_{\omega} \rangle = n U_{\omega}, \quad \langle u_{\omega} \rangle = \varphi U_{\omega} + (1 - \varphi) V_{\omega}.$$
 (6.1)

Both  $U_{\omega}(\mathbf{r})$  and  $V_{\omega}(\mathbf{r})$  are velocity fields with slow spatial variation. The field  $\langle u_{\omega} \rangle$  may be identified with the average volume velocity field of the mixture.

By use of (3.25) and (5.11) we find

$$4\pi \langle \boldsymbol{P}_{\omega} \rangle = \frac{3\varphi(1-\varphi)}{2-3\varphi+\gamma\varphi} (\boldsymbol{U}_{\omega}-\boldsymbol{V}_{\omega}).$$
(6.2)

Using also (6.1) we may cast (5.16) in the form

$$-\mathrm{i}\omega m_{\mathrm{p}} U_{\omega} + [-\mathrm{i}\omega m_{\mathrm{v}} + \zeta_{\mathrm{v}}] (U_{\omega} - V_{\omega}) = E_{\omega} - \mathrm{i}\omega m_{\mathrm{f}} V_{\omega}, \qquad (6.3)$$

with the virtual mass

$$m_{\rm V} = \frac{1 - \gamma \varphi}{2 - 3\varphi + \gamma \varphi} m_{\rm f} \tag{6.4}$$

and the corresponding friction coefficient

$$\zeta_{\rm V} = \frac{2(1-\varphi)}{2-3\varphi+\gamma\varphi}\zeta.$$
(6.5)

The equation of motion (6.3) may be cast in several different forms. First we note that it follows from (3.25), (6.1) and (6.2) that

$$m_{\rm f}(V_{\omega} - \langle v_{\omega} \rangle) = \varphi m_{\rm V}(U_{\omega} - V_{\omega}). \tag{6.6}$$

FLM 225

We recall that we have continued the field  $v_{\omega}(r)$  inside the particles where it is identical with the translational velocity. Correspondingly we may also continue the pressure inside the particles and define it from the equation

$$\mathbf{i}\omega\rho\boldsymbol{v}_{\omega} = \boldsymbol{\nabla}\boldsymbol{p}_{\omega}.\tag{6.7}$$

This equation now holds everywhere in space. Averaging (6.7) and substituting in (6.6) we obtain

$$-\mathrm{i}\omega\rho\,\boldsymbol{V}_{\omega} = -\,\boldsymbol{\nabla}\langle\boldsymbol{p}_{\omega}\rangle - \mathrm{i}\omega\boldsymbol{n}\boldsymbol{m}_{\mathrm{V}}(\boldsymbol{U}_{\omega} - \boldsymbol{V}_{\omega}). \tag{6.8}$$

Using this on the right-hand side of (6.3) we may rewrite that equation as

$$-\mathrm{i}\omega m_{\mathrm{p}} U_{\omega} + \left[-\mathrm{i}\omega(1-\varphi) m_{\mathrm{V}} + \zeta_{\mathrm{V}}\right] (U_{\omega} - V_{\omega}) = E_{\omega} - \frac{4}{3}\pi a^{3} \nabla \langle p_{\omega} \rangle.$$
(6.9)

This equation is similar to equations of motion derived by Drew (1983) and by Geurst (1985, 1986). We note that (6.8) may be regarded as the average equation of motion for the fluid. By linear combination of (6.8) and (6.9) we find

$$-\mathrm{i}\omega[(1+\varphi C_{\mathrm{V}})\,m_{\mathrm{p}}+(1-\varphi)\,m_{\mathrm{f}}]\,(\boldsymbol{U}_{\omega}-\boldsymbol{V}_{\omega})+\zeta_{\mathrm{V}}(\boldsymbol{U}_{\omega}-\boldsymbol{V}_{\omega})=\boldsymbol{E}_{\omega}-\frac{4}{3}\pi a^{3}\frac{m_{\mathrm{f}}-m_{\mathrm{p}}}{m_{\mathrm{f}}}\boldsymbol{\nabla}\langle\boldsymbol{p}_{\omega}\rangle,$$
(6.10)

where the virtual-mass coefficient  $C_{\rm v}$  is defined as  $C_{\rm v} = m_{\rm v}/m_{\rm f}$ . The last term on the right may be regarded as a buoyancy force. We emphasize that the above equations must be used in combination with (4.14) and (5.19).

A slightly different form of (6.3) may be derived in terms of the velocity difference

$$\boldsymbol{U} - \langle \boldsymbol{u} \rangle = (1 - \varphi) (\boldsymbol{U} - \boldsymbol{V}). \tag{6.11}$$

This leads to

$$-\mathrm{i}\omega m_{\mathrm{p}} U_{\omega} + [-\mathrm{i}\omega m_{\mathrm{v}}' + \zeta_{\mathrm{v}}'] (U_{\omega} - \langle u_{\omega} \rangle) = E_{\omega} - \mathrm{i}\omega m_{\mathrm{f}} V_{\omega}, \qquad (6.12)$$

with transport coefficients

$$m'_{\rm V} = \frac{m_{\rm V}}{1-\varphi}, \quad \zeta'_{\rm V} = \frac{\zeta_{\rm V}}{1-\varphi}.$$
 (6.13)

The form (6.12) may be compared with equation (7.4) of Biesheuvel & van Wijngaarden (1984). We recall that the Levich value  $12\pi\eta a$  for the friction coefficient  $\zeta$  follows by use of the perfect-slip parameter  $c = \infty$  in (2.13).

Yet another form of the equation of motion is

$$-\mathrm{i}\omega m_{\mathrm{p}} \boldsymbol{U}_{\omega} + [-\mathrm{i}\omega m_{\mathrm{L}} + \zeta_{\mathrm{L}}] (\boldsymbol{U}_{\omega} - \langle \boldsymbol{u}_{\omega} \rangle) = \boldsymbol{E}_{\omega} - \mathrm{i}\omega m_{\mathrm{f}} \langle \boldsymbol{u}_{\omega} \rangle, \qquad (6.14)$$

with the virtual mass

$$m_{\rm L} = \frac{1 + 3\varphi - \gamma\varphi}{2 - 3\varphi + \gamma\varphi} m_{\rm f}, \tag{6.15}$$

and the corresponding friction coefficient

$$\zeta_{\rm L} = \zeta_{\rm V}' = \frac{2}{2 - 3\varphi + \gamma \varphi} \zeta. \tag{6.16}$$

One may derive (6.14) from (6.12) by use of (6.11). We use the subscript L, because the same transport coefficients appear in the longitudinal equation of motion to be derived shortly. The equation of motion (6.14) is closely related to equation (4.12) of van Wijngaarden (1976), who considered in particular  $m_p = 0$ . The relations (6.13) and (6.15) imply that one must be careful in comparing values for the virtual mass appearing in the literature. From (6.4) we find that the virtualmass coefficient  $C_{\rm v} = m_{\rm v}/m_{\rm f}$  has the exact expression

$$C_{\rm v} = \frac{1 - \gamma \varphi}{2 - 3\varphi + \gamma \varphi}.\tag{6.17}$$

We note that if  $\gamma$  is approximated by the Clausius-Mossotti value  $\gamma = 1$ , then the virtual-mass coefficient  $C_{\rm V}$  takes the value  $\frac{1}{2}$ , independent of the volume fraction. Similarly we define the virtual-mass coefficient  $C_{\rm L}$  by means of the relation  $m_{\rm L} = C_{\rm L} m_{\rm f}$ . From (6.15) and (6.17) we find the relation

$$C_{\rm v} = (1 - \varphi) C_{\rm L} - \varphi.$$
 (6.18)

The friction coefficient  $\zeta_{\rm L}$  may be written

$$\zeta_{\rm L} = \frac{2}{3} (1 + C_{\rm L}) \, \zeta = \frac{2}{3} \frac{1 + C_{\rm V}}{1 - \varphi} \, \zeta. \tag{6.19}$$

We compare with an expression for the virtual mass derived by Zuber (1964) from a cell model. It is instructive to repeat his calculation within our present framework and to include friction. Consider a spherical particle at the centre of a sphere of radius b and under the influence of a small force E. The resulting flow has the potential

$$\phi(\mathbf{r}) = \frac{\mathbf{p} \cdot \hat{\mathbf{r}}}{r^2} - \mathbf{A} \cdot \mathbf{r}. \tag{6.20}$$

From the boundary condition  $v_r = 0$  at r = b it follows that  $A = -2p/b^3$ . We substitute this in the two equations

$$\boldsymbol{p} = \alpha_E \boldsymbol{E} + \hat{\alpha} \boldsymbol{A} = \frac{1}{2} a^3 (\boldsymbol{U} - \boldsymbol{A}). \tag{6.21}$$

Eliminating p we obtain

$$\left[-\mathrm{i}\omega m_{\mathrm{p}} - \mathrm{i}\omega \frac{b^{3} + 2a^{3}}{b^{3} - a^{3}} + \zeta \frac{b^{3}}{b^{3} - a^{3}}\right] U = E.$$
(6.22)

Choosing the radius b such that  $a^3/b^3 = \varphi$  we find Zuber's induced mass

$$m_{\rm Z} = \frac{1}{2} m_{\rm f} \frac{1+2\varphi}{1-\varphi} \tag{6.23}$$

and the corresponding friction coefficient

$$\zeta_{\rm Z} = \frac{\zeta}{1 - \varphi}.\tag{6.24}$$

It is clear from the above derivation that Zuber's induced mass cannot be identified with the virtual mass  $m_{\rm V}$ , since in the defining equation (6.22) the fluid velocity does not occur.

We can define an induced mass without the limitations of the cell model by use of (5.19). We consider a suspension of infinite extent which on average is uniform and impose a plane-wave force field

$$\boldsymbol{E}(\boldsymbol{r},t) = \boldsymbol{E}_{\boldsymbol{a}\omega} \exp\left(\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{r} - \mathrm{i}\omega t\right). \tag{6.25}$$

7-2

It follows from (5.19) that in the long-wavelength limit the corresponding velocity field has amplitude

$$\langle \boldsymbol{v}_{\boldsymbol{q}\boldsymbol{\omega}} \rangle = -\frac{4\pi\chi}{\epsilon} \hat{\boldsymbol{q}} \hat{\boldsymbol{q}} \cdot \boldsymbol{E}_{\boldsymbol{q}\boldsymbol{\omega}}. \tag{6.26}$$

More explicitly we find from (3.21) and (5.5) that

$$\left[-\mathrm{i}\omega m_{\mathrm{f}}(1+3\varphi-\gamma\varphi)-\mathrm{i}\omega m_{\mathrm{p}}(2-3\varphi+\gamma\varphi)+2\zeta\right]\langle\boldsymbol{v}_{q\omega}\rangle = -3\varphi\hat{\boldsymbol{q}}\hat{\boldsymbol{q}}\cdot\boldsymbol{E}_{q\omega}.$$
 (6.27)

Substituting this in (5.16) we obtain for the longitudinal component of the particle current density

$$[-\mathrm{i}\omega(m_{\mathrm{p}}+m_{\mathrm{L}})+\zeta_{\mathrm{L}}]\hat{q}\hat{q}\cdot\langle J_{q\omega}\rangle = n\hat{q}\hat{q}\cdot E_{q\omega}, \qquad (6.28)$$

with  $m_{\rm L}$  given by (6.15) and  $\zeta_{\rm L}$  by (6.16). The corresponding transverse relation reads

$$[-\mathrm{i}\omega(m_{\mathrm{p}}+m_{\mathrm{a}})+\zeta_{\mathrm{a}}](\boldsymbol{l}-\hat{\boldsymbol{q}}\hat{\boldsymbol{q}})\cdot\langle\boldsymbol{J}_{\mathrm{q}\omega}\rangle = n(\boldsymbol{l}-\boldsymbol{q}\boldsymbol{q})\cdot\boldsymbol{E}_{\boldsymbol{q}\omega}.$$
(6.29)

If  $\gamma$  is approximated by the Clausius-Mossotti value  $\gamma = 1$ , then  $m_{\rm L}$  becomes identical with Zuber's expression (6.23) and  $\zeta_{\rm L}$  becomes identical with (6.24). As one would expect the cell model yields a mean field result.

We note that (6.27) may be written alternatively as

$$[-\mathrm{i}\omega(m_{\mathrm{p}}+m_{\mathrm{L}})+\zeta_{\mathrm{L}}]\hat{q}\hat{q}\cdot\langle v_{q\omega}\rangle = \frac{-3\varphi}{2-3\varphi+\gamma\varphi}\hat{q}\hat{q}\cdot E_{q\omega}.$$
(6.30)

By means of (3.25), (6.1), (6.2) and (6.28) this may be seen to be consistent with (4.14). The latter equation implies that the longitudinal component of the mean volume flux  $q \cdot \langle u_{q\omega} \rangle$  vanishes.

One-dimensional motions in the direction of the wavevector are purely longitudinal. For such motions we may cast (6.28) in the alternative form

$$\left[-\mathrm{i}\omega(m_{\mathrm{p}}+m_{\mathrm{L}})+\zeta_{\mathrm{L}}\right]U_{q\omega}^{\mathrm{L}}=E_{q\omega}^{\mathrm{L}},\tag{6.31}$$

where we have used the notation  $U_{q\omega}^{\rm L} = \hat{\boldsymbol{q}} \cdot \boldsymbol{U}_{q\omega}$ . Comparing this with Batchelor's equation of motion (2.15) for a fluidized bed (Batchelor 1988) we see that his virtual-mass coefficient  $C(\varphi)$  must be identified with  $C_{\rm L} = m_{\rm L}/m_{\rm f}$ .

Finally we note that for longitudinal motions we may use the equation  $\langle u^L \rangle = 0$  to cast the particle equation of motion (6.31) in the form

$$-\mathrm{i}\omega m_{\mathrm{p}} U_{q\omega}^{\mathrm{L}} + [-\mathrm{i}\omega m_{\mathrm{L}} + \zeta_{\mathrm{L}}] (1-\varphi) (U_{q\omega}^{\mathrm{L}} - V_{q\omega}^{\mathrm{L}}) = E_{q\omega}^{\mathrm{L}}.$$
(6.32)

The above analysis shows that the equations may be written in a variety of different forms. The restriction to one-dimensional flow introduces transport coefficients  $m_{\rm L}$  and  $\zeta_{\rm L}$ , which differ from the coefficients  $m_{\rm V}$  and  $\zeta_{\rm V}$  appearing in the three-dimensional equation of motion.

### 7. Numerical results

In the preceding sections we have derived exact expressions for the transport coefficients  $m_{\rm V}$ ,  $\zeta_{\rm V}$ ,  $m_{\rm L}$  and  $\zeta_{\rm L}$  in terms of the parameter  $\gamma$  defined in (5.4). In order to find numerical results for  $\gamma$  we must study the dielectric constant of a static system of spheres with polarizabilities given by (5.1). In this section we evaluate  $\gamma$  exactly to first order in the volume fraction. Also we propose a scheme which allows approximate calculation of  $\gamma$  even at high volume fractions.

First we consider the low-density value of the coefficient  $\gamma$ . We write

$$\gamma = 1 + \delta, \quad \delta = \delta_0 + O(\varphi).$$
 (7.1)

The low-density value  $\delta_0$  may be evaluated exactly from the solution of a pair problem. From equations (3.35), (5.15) and (5.16) of Felderhof, Ford & Cohen (1982) we find

$$\delta_{0} = \left(\frac{a^{3}}{\hat{\alpha}}\right)^{2} \int_{2a}^{\infty} \mathrm{d}R \, R^{2} g_{0}(R) \left[a_{10}(R) + 2a_{11}(R) - 3\left(\frac{\hat{\alpha}}{a^{3}}\right)\right],\tag{7.2}$$

where  $g_0(R)$  is the low-density value of the pair correlation function, and  $a_{10}(R)$  and  $a_{11}(R)$  are distance-dependent amplitudes which follow from the solution of the pair problem. We shall consider hard-sphere statistics, so that the pair correlation function is given by

$$g_0(R) = \theta(R - 2a). \tag{7.3}$$

According to equation (3.11) of Felderhof *et al.* (1982) the amplitudes  $a_{10}(R)$  and  $a_{11}(R)$  are found as solutions of the set of equations

$$\sum_{l'=1}^{\infty} M_{ll'}^m a_{l'm} = \delta_{l1}, \quad l = 1, 2, \dots, \quad m = 0, 1,$$
(7.4)

where

$$M_{ll'}^{m} = \frac{a^{2l+1}}{\alpha_l} \delta_{ll'} - (-1)^m \binom{l+l'}{l+m} \binom{a}{R}^{l+l'+1}.$$
(7.5)

In our hydrodynamic problem  $\alpha_1 = \hat{\alpha}$ , and the multipole polarizabilities  $\alpha_l$  for  $l \ge 2$  are given by (5.1). We follow the procedure of Felderhof *et al.* (1982) and write  $\delta_0$  as the sum

$$\delta_0 = \delta_{0D} + \delta_{0M}, \tag{7.6}$$

where  $\delta_{0D}$  is the value found in the dipole approximation, and  $\delta_{0M}$  represents the correction from the higher-order multipole moments. In the dipole approximation one finds

$$\delta_{0D} = \frac{2}{3} \log \frac{8a^3 + \hat{\alpha}}{8a^3 - 2\hat{\alpha}}.$$
(7.7)

The correction from higher-order multipoles is given by

$$\delta_{0M} = \left(\frac{a^3}{\dot{\alpha}}\right)^2 \int_{2a}^{\infty} \mathrm{d}R \, R^2 g_0(R) \left[b_{10}(R) + 2b_{11}(R)\right] \tag{7.8}$$

with the functions  $b_{1m}(R)$  defined by

$$b_{1m} = a_{1m} - (M_{11}^m)^{-1}. (7.9)$$

The functions may be evaluated from equations (5.9)-(5.11) of Felderhof *et al.* (1982). From those equations it follows that the functions  $a_{1m}$  may be expressed in terms of the solutions  $a'_{1m}$  of a related problem with modified dipole polarizability  $a'_1$ , but with the same multipole polarizabilities  $a_l$  for  $l \ge 2$ . Using the abbreviation  $M_m \equiv M_{11}^m$  we find

$$a_{1m} = a'_{1m} [1 + (M_m - M'_m) a'_{1m}]^{-1}.$$
(7.10)

In the present case it is of interest to choose  $\alpha'_1 = -\frac{1}{2}a^3$ . Then the related dielectric problem concerns a system of perfectly insulating spheres, i.e. spheres with vanishing dielectric constant embedded in vacuum. For that system the integral (7.8), with

Â	$\delta_{0\mathrm{D}}$	$\delta_{_{0M}}$	$\delta_0$	$k_{v}$	$k_{\rm L}$
-0.5	-0.122	-0.095	-0.216	0.324	3.324
-0.4	-0.098	-0.095	-0.193	0.289	3.289
-0.3	-0.074	-0.095	-0.169	0.253	3.253
-0.2	-0.049	-0.096	-0.145	0.218	3.218
-0.1	-0.025	-0.096	-0.121	0.182	3.182
0	0	-0.097	-0.097	0.145	3.145
0.2	0.051	-0.098	-0.048	0.072	3.072
0.4	0.103	-0.100	0.002	-0.004	2.996
0.6	0.157	-0.103	0.054	-0.081	2.919
0.8	0.212	-0.106	0.106	-0.159	2.841
1.0	0.270	-0.110	0.160	-0.241	2.759

TABLE 1. Table of coefficients  $\delta_{0D}$ ,  $\delta_{0M}$  and their sum  $\delta_0$  for various values of the mass parameter  $\hat{\beta} = (m_{\rm f} - m_{\rm p})/(m_{\rm f} + 2m_{\rm p})$ . We also list  $k_{\rm v} = -\frac{3}{2}\delta_0$  and  $k_{\rm L} = 3 - \frac{3}{2}\delta_0$ 

 $b_{1m}(R)$  replaced by  $b'_{1m}(R)$ , has already been evaluated. We denote the corresponding coefficient as  $\delta^{\infty}_{0M}$ . By comparison with (5.1) we see that this coefficient is identical with  $\delta_{0M}$  in the limit  $m_p/m_f = \infty$ .

In our numerical calculations of  $\delta_0$  we consider the limit of vanishing viscosity. In table 1 we list the values of  $\delta_{0D}$ ,  $\delta_{0M}$  and their sum  $\delta_0$  for a range of values of the parameter

$$\hat{\beta} = \left(\frac{\hat{\alpha}}{a^3}\right)_{\eta=0} = \frac{m_{\rm f} - m_{\rm p}}{m_{\rm f} + 2m_{\rm p}}.$$
(7.11)

We also list the value of the product  $k_{\rm V} = -\frac{3}{2}\delta_0$  which appears in the expansion of the virtual-mass coefficient  $C_{\rm V} = m_{\rm V}/m_{\rm f}$  in powers of the volume fraction

$$C_{\rm V} = \frac{1}{2} \left[ 1 - \frac{3}{2} \delta_0 \varphi + O(\varphi^2) \right], \tag{7.12}$$

as follows from (6.17). We note that the case of gas bubbles may be identified with the limit  $m_p = 0$ , i.e.  $\hat{\beta} = 1$ . Furthermore we list the value of the product  $k_{\rm L} = 3 - \frac{3}{2}\delta_0$  which appears in the expansion of the coefficient

$$C_{\rm L} = \frac{1}{2} \left[ 1 + (3 - \frac{3}{2}\delta_0)\varphi + O(\varphi^2) \right], \tag{7.13}$$

as follows from (6.18).

As shown below (6.16), van Wijngaarden's (1976) virtual-mass coefficient should be identical with  $C_{\rm L}$  for  $m_{\rm p} = 0$ . Thus his coefficient k should equal  $3 - \frac{3}{2}\delta_0^0$ , where the superscript indicates that  $m_{\rm p} = 0$ . A comparison with the corresponding entry in our table for  $k_{\rm L}$  yields  $k_{\rm L}^0 = 2.759$ , in good agreement with van Wijngaarden's value 2.78. Apparently van Wijngaarden uses the equivalent of (7.10) in his derivation. In later work (Biesheuvel & van Wijngaarden 1984) the value of the coefficient was corrected to 3.32, which seems to correspond to the value 3.324 for  $m_{\rm p} = \infty$  in our table.

In order to obtain an approximate expression for the coefficient  $\delta$ , applicable at high density, we appeal again to the theory of the dielectric constant. We first consider a system of spheres of dielectric constant  $\epsilon_2$  embedded in a medium of dielectric constant  $\epsilon_1$ . The geometrical distribution is assumed to be the same as before. The effective dielectric constant  $\epsilon^*$  of this system is given by an expression similar to (5.6):

$$\frac{\epsilon^* - \epsilon_1}{\epsilon^* + 2\epsilon_1} = \frac{\beta\varphi}{1 - \beta\varphi[\lambda(\beta) + \mu(\beta)]},\tag{7.14}$$

φ	0.1	0.2	0.3	0.4	0.5
ζ,	0.021	0.040	0.059	0.084	0.141
8,	0.0192	0.0276	0.0282	0.0235	0.0161

TABLE 2. Values of the three-point parameter  $\zeta_2$  and the Kirkwood-Yvon coefficient  $s_2$  for various values of the volume fraction  $\varphi$ 

with reduced polarizability  $\beta$  given by

$$\beta = \frac{\epsilon_2 - \epsilon_1}{\epsilon_2 + 2\epsilon_1},\tag{7.15}$$

and with coefficients  $\lambda(\beta)$  and  $\mu(\beta)$  appropriate to this particular system. To first order in  $\beta$  the sum  $\lambda(\beta) + \mu(\beta)$  is given by

$$\lambda^{(1)} + \mu^{(1)} = 9K\beta/\varphi^2, \tag{7.16}$$

where K is a correlation integral involving two- and three-particle correlation functions (Felderhof 1982). It is related to the three-point parameter  $\zeta_2$  introduced by Beran (1965) for general two-phase geometries by

$$K = \frac{2}{9}\varphi(1-\varphi)\,\zeta_2. \tag{7.17}$$

The correlation integral K depends only on the geometrical distribution of spheres and is independent of their dielectric properties. In table 2 we list the values of  $\zeta_2$  for various volume fractions, as calculated by Torquato & Lado (1986) for a system of identical spheres with hard-sphere statistics. According to Torquato (1985) substitution of the approximate value (7.16) in (7.14) provides a highly accurate expression for  $\epsilon^*$ .

The multipole polarizabilities (5.1) for  $l \ge 2$  in the hydrodynamic system are identical with those of the dielectric system with  $\epsilon_2 = 0$  and  $\epsilon_1 = 1$ , but the hydrodynamic system has dipole polarizability  $\hat{\alpha} = \hat{\beta}a^3$ , rather than  $\alpha'_1 = -\frac{1}{2}a^3$ . This implies that the dipole contribution to the correlation integral K is modified. From the structure of the correlation integral K it follows that a reasonable approximation to the coefficient  $\delta = \lambda + \mu$  in our hydrodynamic problem is given by

$$\delta \approx -\frac{9}{2}(K - K_{\rm D})/\varphi^2 + \delta_{\rm D}^{(1)}, \qquad (7.18)$$

where  $K_{\rm D}$  is the dipole approximation to K, and  $\delta_{\rm D}^{(1)}$  is the value of  $\delta$  calculated in dipole approximation to first order in  $\hat{\alpha}$ . These coefficients are given by

$$K_{\rm D} = \frac{1}{9} s_2 \varphi, \quad \delta_{\rm D}^{(1)} = s_2 \hat{\alpha} / \varphi a^3,$$
 (7.19)

where  $s_2$  is given by the so-called Kirkwood-Yvon integrals for a system of polarizable point dipoles. The coefficient  $s_2$  has been evaluated recently by computer simulation of a hard-sphere system at several volume fractions (Cichocki & Felderhof 1989). We list a number of values in table 2. Combining (7.17)-(7.19) we find the approximate value for  $\delta$ :

$$\delta_{\rm app} = -\frac{1-\varphi}{\varphi}\zeta_2 + \frac{s_2}{2\varphi} \bigg[ 1 + 2\frac{\dot{\alpha}}{a^3} \bigg]. \tag{7.20}$$

$\hat{\beta}^{\varphi}$	0+	0.1	0.2	0.3	0.4	0.5
-0.5	-0.211	-0.189	-0.160	-0.138	-0.126	-0.141
-0.4	-0.186	-0.170	-0.146	-0.128	-0.120	-0.138
-0.3	-0.161	-0.151	-0.132	-0.119	-0.114	-0.135
-0.2	-0.136	-0.131	-0.119	-0.109	-0.108	-0.131
-0.1	-0.111	-0.112	-0.105	-0.100	-0.103	-0.122
0	-0.086	-0.093	-0.091	-0.091	-0.097	-0.125
0.2	-0.036	-0.055	-0.063	-0.072	-0.085	-0.118
0.4	0.014	-0.016	-0.036	-0.053	-0.073	-0.112
0.6	0.064	0.022	-0.008	-0.034	-0.061	-0.106
0.8	0.114	0.061	0.019	-0.015	-0.050	-0.099
1.0	0.164	0.099	0.047	0.003	-0.038	-0.093

TABLE 3. Values of the approximate value  $\delta_{app}$  of the coefficient  $\delta$ , as given by (7.20), for various values of the mass parameter  $\hat{\beta}$  and the volume fraction  $\varphi$ 

In table 3 we list  $\delta_{app}$  for five volume fractions and eleven values of  $\hat{\alpha}/a^3$  in the limit of vanishing viscosity for a system with hard-sphere statistics. The low-density value of the above expression is (Felderhof 1982)

$$\delta_{\text{app},0} = -\frac{7}{24} + \frac{3}{16} \ln 3 + \frac{1}{4} \frac{\dot{a}}{a^3} \approx -0.08568 + \frac{1}{4} \frac{\dot{a}}{a^3}.$$
 (7.21)

We find good agreement with the values of  $\delta_0$  listed in table 1. This gives confidence that (7.20) also provides a good approximation to  $\delta = \gamma - 1$  at high volume fractions.

### 8. Discussion

We have shown above that the virtual mass and drag coefficient of a fluid suspension consisting of spherical particles immersed in a nearly inviscid fluid may be expressed exactly in terms of the effective dielectric constant of a related suspension with particular electrical properties and with the same geometrical distribution. We have derived average equations of motion for the fluid suspension by the method of ensemble averaging. In the averaging we have assumed an isotropic distribution.

The linearized equations of motion on the macroscopic scale are given by (4.14), (5.19), (6.1), (6.3), (6.6) and (6.8). The transport coefficients  $\epsilon$ ,  $\chi$ ,  $m_V$  and  $\zeta_V$  in these equations are local quantities. That is, they may be evaluated in terms of the local volume fraction and the particle distribution functions. Thus our equations may be extended without difficulty to spatially non-uniform suspensions.

The relation between the imposed force field  $E_{\omega}(\mathbf{r})$  and the resulting velocity fields is highly non-local. In order to find the velocity fields one first determines the average field  $\langle v_{\omega}(\mathbf{r}) \rangle$  from (4.14) and (5.19). Subsequently one finds the particle velocity field  $U_{\omega}(\mathbf{r})$  from (5.16) and (6.1), which may be combined to give

$$\left[-\mathrm{i}\omega(m_{\mathrm{p}}+m_{\mathrm{a}})+\zeta_{\mathrm{a}}\right]U_{\omega}=E_{\omega}+\left[-\mathrm{i}\omega(m_{\mathrm{f}}+m_{\mathrm{a}})+\zeta_{\mathrm{a}}\right]\langle v_{\omega}\rangle. \tag{8.1}$$

Finally one finds the fluid velocity field  $V_{\omega}(r)$  from (6.6), which may be rewritten as

$$V_{\omega} = \frac{1}{1 + \varphi C_{\mathrm{V}}} \langle \boldsymbol{v}_{\omega} \rangle + \frac{\varphi C_{\mathrm{V}}}{1 + \varphi C_{\mathrm{V}}} U_{\omega}. \tag{8.2}$$

The non-locality arises in the connection between  $\langle v_{\omega} \rangle$  and  $E_{\omega}$ . The remaining relations (8.1) and (8.2) are local. In non-uniform situations the relation between  $\langle v_{\omega} \rangle$  and  $E_{\omega}$  strongly depends on the geometry, just as in Maxwell theory.

In formulating the above relationships we have used local transport coefficients on the basis of a long-wavelength approximation. As we have shown in §4, more generally the theory leads to short-range transport kernels. In principle the corresponding wavevector-dependent transport coefficients may again be studied by use of the dielectric analogy.

It is evident from the theory developed above that a macroscopic description of the two-phase system involves transport coefficients which can be evaluated only on the basis of a microscopic picture. We have limited ourselves to a linear theory, so that we describe small deviations from rest. However, once the transport coefficients have been found, we may use them with confidence in nonlinear equations of motion. In this regard the situation is similar to the hydrodynamics of simple liquids (Hansen & McDonald 1986). The transport coefficients of viscosity and heat conductivity of a simple liquid may be evaluated on the molecular level from linear response theory. Subsequently these coefficients are used in the complete set of nonlinear hydrodynamic equations.

Our linear response approach leads to transport coefficients  $m_{\rm V}$  and  $\zeta_{\rm V}$  which are frequency-dependent. We have treated the group  $\eta/\omega\rho a^2$  as a small parameter and to be consistent we must finally expand in powers of this parameter. The first-order term in the expansion of the virtual mass may be regarded as a contribution to the effective friction coefficient.

Finally we emphasize that our description of hydrodynamic interactions in nearly inviscid fluids may also be used in the study of the dynamics of two or more particles. For any configuration  $\mathbf{R}_1, \ldots, \mathbf{R}_N$  of N particles our formalism allows the calculation of a configuration-dependent mass matrix and friction matrix. One finds these matrices by considering the high-frequency limit in the response to oscillatory applied forces for the given configuration. Once the matrices have been determined one can construct the full nonlinear equations of motion.

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