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A method for the calculation of the effective transport properties of suspensions of interacting particles

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The early attempts at calculating effective transport properties of suspensions of interacting spherical particles resulted in non-absolutely convergent expressions. In this paper we provide a physical interpretation for these convergence difficulties and we present a new method of determining the effective transport properties which clarifies difficulties in existing methods.

This method, which is described for simplicity in the context of the thermal conduction problem, is based on an expression that gives the temperature gradient ∇T at a point $\mathbf{x}$ in the matrix in terms of integrals over the surrounding particles and an integral over a large surface Γ which encloses $\mathbf{x}$ and which we term the 'macroscopic boundary'. Without the integral over Γ, this expression for ∇T would be non-absolutely convergent, for the contribution to $\nabla T(\mathbf{x})$ from a distant particle is proportional to $1/r^3$, where r is the distance of the particle from $\mathbf{x}$. On comparing the expression for ∇T with the formula used by Rayleigh (1892) in his investigation of the effective conductivity of a cubic array of spheres, we find that Rayleigh's convergence difficulties arose simply from an incorrect assessment of the macroscopic boundary integral.

By combining the expression for $\nabla T(\mathbf{x})$ with a formula for the dipole strength of a sphere placed in an ambient temperature field, we obtain a convergent expression relating the dipole strength of a sphere to integrals over the surrounding particles. An expression for the effective conductivity of a random suspension of spheres correct to $O(\phi^2)$ is obtained simply by averaging this expression for the thermal dipole strength. By a similar procedure we obtain expressions for the effective viscosity and effective elastic moduli correct to $O(\phi^2)$. Most of these results have been obtained by earlier workers using a 'renormalization' procedure due to Batchelor; the method presented here has the advantage that the renormalization quantity arises naturally from the macroscopic boundary integral referred to earlier, so there is no uncertainty about its choice.

1. Introduction

A sample of a suspension or a composite material which contains a large number of particles embedded in a continuous matrix may in many cases be treated as a homogeneous material, and be assigned 'effective' properties which are related to, but which may be quite different from, the properties of the matrix or the particles. In this paper we present a new method of determining the effective transport properties of a suspension which will clarify the difficulties in existing methods described

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by Jeffrey (1977). The transport property of interest may be the conductivity (electrical or thermal), the static dielectric constant, the viscosity or the elastic moduli. This method will be described in the context of the thermal conduction problem, for the mathematical details are relatively simple in this case. In the final section we shall briefly describe the application of the method to the elasticity and viscosity problem.

In the work that follows we make use of the formalism which has recently been developed (see Batchelor 1974 for a review) for dealing with the problem of calculating effective transport properties. For statistically homogeneous materials, the effective transport properties can be defined in terms of volume-averaged quantities; in the case of thermal conduction the relevant quantities are the average temperature gradient $\langle \nabla T \rangle$ and flux density $\langle \mathbf{F} \rangle$, defined by

$$\langle \nabla T \rangle = \frac{1}{V}\int_V \nabla T \, dV \tag{1.1}$$

and

$$\langle \mathbf{F} \rangle = \frac{1}{V}\int_V \mathbf{F} \, dV, \tag{1.2}$$

where V is a volume large enough to contain many particles. Since the equations which govern the temperature distribution in the suspension are linear, it follows that $\langle \mathbf{F} \rangle$ and $\langle \nabla T \rangle$ are linearly related; that is

$$\langle \mathbf{F} \rangle = -\mathbf{k}^* . \langle \nabla T \rangle, \tag{1.3}$$

where $\mathbf{k}^*$ is termed the 'effective conductivity tensor'. Our aim is to derive an expression for $\mathbf{k}^*$.

The quantity $\mathbf{k}^*$ can be related to an average over the particles in V. To obtain this relation, we write the expression (1.2) for $\langle \mathbf{F} \rangle$ in the form

$$\langle \mathbf{F} \rangle = \frac{1}{V}\int_{V-\Sigma V_i} \mathbf{F} \, dV + \frac{1}{V}\sum_i \int_{V_i} \mathbf{F} \, dV, \tag{1.4}$$

where ΣV_i is the portion of V occupied by particles. In the matrix we have

$$\mathbf{F} = -k \nabla T,$$

where k is the matrix conductivity, and on substituting this relation in the integral over the matrix in (1.4) we get

$$\langle \mathbf{F} \rangle = -k \langle \nabla T \rangle + n \langle \mathbf{S} \rangle, \tag{1.5}$$

where n is the number density of particles. The quantity $\langle \mathbf{S} \rangle$ is the average of the dipole strength $\mathbf{S}$ of a particle in V, where

$$\mathbf{S} = (1-\alpha^{-1})\int_{V_p} \mathbf{F} \, dV = (1-\alpha^{-1})\int_{A_p} \mathbf{x}\mathbf{F} . \hat{\mathbf{n}} \, dA. \tag{1.6}$$

Here αk is the particle conductivity, V_p and A_p are the volume and the surface of the particle, and $\hat{\mathbf{n}}$ is the unit normal directed outwards from the particle surface. The quantity $\langle \mathbf{S} \rangle$ is linearly related to $\langle \nabla T \rangle$, thus the conductivity may be determined from the average dipole strength with the aid of the expressions (1.3) and (1.5) for $\langle \mathbf{F} \rangle$.

Analogous results to those described above may be obtained for the other transport properties (Batchelor 1974). This formalism has been used in a number of recent investigations of the effective transport properties of suspensions and composite materials (Batchelor 1970; Batchelor & Green 1972*b*; Jeffrey 1973; Chen & Acrivos 1978).

If the suspension is very dilute, the interactions between the particles may be neglected and the effective transport properties can be calculated from expressions for the dipole strength of a particle alone in an infinite matrix. At higher volume fractions the problem of determining the effective transport properties is more difficult, for the particle interactions must be taken into account.

2. The effect of particle interactions

2.1. *Previous theoretical investigations*

The earliest study of the effect of particle interaction on conductivity was carried out by Rayleigh (1892), who obtained an expression for the conductivity of spheres in a cubic array. This expression provides the first few terms in the expansion of the conductivity as a power series in a/d, where a is the sphere radius and d denotes the centre-to-centre distance between nearest neighbours in the array. In order to calculate the effect of surrounding particles on the dipole strength of a reference sphere, Rayleigh assumed that the ambient temperature gradient at the position of the centre of the reference sphere is equal to the average temperature gradient plus the sum of the fields produced by the surrounding spheres.† However, this sum is non-absolutely convergent; that is, the result depends on the order in which the contributions from the (infinite number of) surrounding spheres are summed. Rayleigh noted this, but nevertheless summed the contributions in a particular order, giving no real justification for doing so.

Since that time there have been a number of investigations into the effective conductivity and static dielectric constant of various regular arrays. Levine (1966) and Zuzovsky & Brenner (1977) avoided the convergence problem by formulating methods which rely on the fact that the temperature gradient is periodic, while other investigators adopted Rayleigh's method without any attempt to justify the procedure for evaluating the non-absolutely convergent sum (Meredith & Tobias 1960; Bertaux, Bienfait & Jolivet 1975). This flaw in Rayleigh's method was finally removed by McKenzie & McPhedran (1978), who introduced a modified version of the method which is free of convergence difficulties. In this paper we show why the convergence problem arose in the first place, and we present a simple divergence-free method of calculating the effective transport properties which applies *both* to regular arrays and to random arrays of spherical particles. For the case of a regular array, this method is shown to be equivalent to that used by McKenzie & McPhedran.

The problem of determining the effective transport properties of a random array of interacting spheres is more difficult than that for a regular array, and most of the theoretical investigations have been concerned with the problem of calculating the perturbation in the average dipole strength $\langle \mathbf{S} \rangle$ caused by particle interactions in dilute suspensions.

The probability that a particle in a dilute random suspension will have m neighbours within a distance of several radii is of order ϕ^m, where ϕ is the particle volume fraction.

† We are referring here to equation (62) of Rayleigh's paper; the validity of the above interpretation of that equation will become clearer in § 3, where the Rayleigh method is analysed in more detail.

If we assume that the interactions between particles fall off sufficiently rapidly with increasing separation, then the perturbation in $\langle \mathbf{S} \rangle$ is due mainly to pair interactions ($m = 1$) when $\phi \ll 1$. Provided that this assumption is valid, the average dipole strength may apparently be written as

$$\langle \mathbf{S} \rangle = \mathbf{S}^0 + \int \mathbf{S}'(\mathbf{r})\, p(\mathbf{r}|\mathbf{0})\, dV(\mathbf{r}), \tag{2.1}$$

where $p(\mathbf{r}|\mathbf{0})\,dV(\mathbf{r})$ is the probability that the centre of a particle lies within the volume dV surrounding the point $\mathbf{r}$, given that the centre of the reference sphere is at the origin. The term $\mathbf{S}^0$ denotes the dipole strength in the absence of particle interaction, and $\mathbf{S}'$ is the amount by which the dipole strength of the reference sphere is altered by the presence of another sphere at $\mathbf{r}$, neglecting all other particles. Unfortunately, the term $\mathbf{S}'(\mathbf{r})$ falls off as $1/|\mathbf{r}|^3$ as $|\mathbf{r}| \to \infty$, and the integral in (2.1), like the sum encountered by Rayleigh, is not absolutely convergent.

In order to obtain an expression for $\langle \mathbf{S} \rangle$ in terms of convergent integrals, Batchelor (see 1974 for review) devised a technique based on the observation that, associated with each of the transport problems, there is a quantity which had the same far-field dependence as $\mathbf{S}'$ and which has a known average. We shall call this quantity the 'renormalizing quantity'. By combining the general formula for $\langle \mathbf{S} \rangle$ with the expression for the average of the renormalizing quantity, it is possible to relate $\langle \mathbf{S} \rangle$ to an integral which *is* dominated by pair interactions for $\phi \ll 1$, and thus a formula for $\langle \mathbf{S} \rangle$ correct to $O(\phi)$ is obtained. This method, called here the 'renormalization technique', has been employed in the derivation of expressions for the average velocity of sedimentation of spheres to order ϕ (Batchelor 1972), the effective viscosity to order ϕ^2 of a suspension of rigid spheres in a Newtonian liquid (Batchelor & Green 1972*b*) and the effective conductivity of a random suspension of spheres to order ϕ^2 (Jeffrey 1973).

Although the renormalization technique has been very successful, some workers have found that the correct choice of a renormalizing quantity is not always straightforward, a matter which we discuss in § 6. In order to avoid the need for renormalizing, Willis & Acton (1976) devised an alternative method for obtaining a convergent expression for the average dipole strength of a random suspension of spheres, based on an integral-equation formulation for the elasticity problem. Unfortunately, this method is considerably more complicated than the renormalization procedure, and like that procedure it does not provide insight into the cause of the convergence difficulties referred to earlier. This latter point is important, for the non-absolutely convergent terms such as Rayleigh's sum yield finite answers (which depend on the method of evaluation) and there is a temptation to regard the convergence problem as a mere inconvenience.

2.2. *Outline of the present paper*

In this paper we present a method which resembles, but which is simpler than, Willis & Acton's, and which may be applied to any of the transport problems associated with suspensions of spherical particles in random or regular arrays. The method is straightforward, and it enables us to provide a physical interpretation for the convergence difficulties described in the previous section.

The procedure is based on a relation between the temperature gradient at a point $\mathbf{x}$ in a suspension and integrals over the surfaces of the surrounding particles, together with an integral over a surface Γ which encloses $\mathbf{x}$ and which has the following properties: (i) it is sufficiently large to contain many particles and (ii) at each point on Γ the local radii of curvature of the surface are much greater than the length scales associated with the fluctuations in the temperature field (typically of the order of the particle diameter). We call Γ a 'macroscopic boundary', since the length scales associated with the surface are much larger than those associated with the microstructure. For such a surface it is shown that the local temperature and flux density which appear in the integral over Γ may be replaced by the averaged quantities $\langle T\rangle$ and $\langle \mathbf{F}\rangle$.

On applying the divergence theorem to this integral, we obtain a term which may be regarded as the field due to a continuous distribution of dipoles over the volume enclosed by Γ.† The contribution to $\nabla T(\mathbf{x})$ from particles which lie in a distant volume is cancelled by the contribution from the continuous distribution of dipoles contained within that volume, hence the expression for $\nabla T(\mathbf{x})$ converges. Thus when the integral over the macroscopic boundary is taken into account, there are no convergence problems associated with the expression for $\nabla T(\mathbf{x})$. In § 4 we show that Rayleigh's convergence difficulties arose from an incorrect assessment of this macroscopic boundary term. By combining the expression for $\nabla T(\mathbf{x})$ with a formula for the dipole strength of a sphere placed in an ambient temperature field we obtain a convergent expression for the dipole strength of a reference sphere, which shows explicitly the effect of distant particles.

In § 5 we employ this result in a study of the problem of conduction through a random suspension of spheres. Jeffrey's (1973) formula for the conductivity is obtained simply by taking the average of the expression for the dipole strength of a spherical particle. With this method there is no need for renormalizing, for the renormalizing quantity arises naturally from the macroscopic boundary term referred to earlier.

In § 6 we show how this method may be applied to the elasticity and viscosity problems, and we obtain expressions for the effective elastic moduli and viscosity, correct to $O(\phi^2)$. Finally, we discuss Chen & Acrivos's (1978) calculation of the effective modulus of compression. In this case the renormalization procedure is complicated by the fact that there are three renormalizing quantities to choose from. No such complications arise if the method presented here is used, since the renormalization quantity is obtained directly from the macroscopic boundary term.

3. An expression for the thermal dipole strength of a sphere in a statistically homogeneous suspension

In this section we derive an important expression for the dipole strength of a spherical particle in terms of integrals over the surrounding particles, together with the dipole-field term referred to in the previous section. We begin by noting that, since the temperature field satisfies Laplace's equation, the temperature at a point $\mathbf{x}$ in the matrix of a suspension may be related to integrals over the surrounding particles with the aid of Green's Third Identity (Protter & Weinberger 1967, pp. 84), viz.

$$T(x) = \frac{1}{4\pi}\sum_i \oint_{A_i}\left\{\frac{\mathbf{F}'}{kr} + T'\nabla'\frac{1}{r}\right\}.\hat{\mathbf{n}}\,dA + \frac{1}{4\pi}\oint_{\Gamma}\left\{\frac{\mathbf{F}'}{kr} + T'\nabla'\frac{1}{r}\right\}.\hat{\mathbf{n}}\,dA, \tag{3.1}$$

† The resulting equation for $\nabla T(\mathbf{x})$ is analogous to Willis & Acton's expression (2.7) for the strain in an elastic matrix.

where $r = |\mathbf{x}-\mathbf{x}'|$, $\mathbf{F}' = \mathbf{F}(\mathbf{x}')$, $T' = T(\mathbf{x}')$ and $\nabla' = \partial/\partial\mathbf{x}'$. Here Γ denotes a closed surface enclosing $\mathbf{x}$, A_i denotes the surface of the ith particle contained in Γ, and $\hat{\mathbf{n}}$ is the unit normal directed into the matrix. If the surface Γ passes through the ith particle, then A_i denotes the closed surface of the part of the particle that lies within Γ.

With the aid of the divergence theorem we can convert the integrals over the surfaces of the particles to volume integrals:

$$\oint_{A_i} \left\{\frac{\mathbf{F}'}{kr} + T'\nabla' \frac{1}{r}\right\} . \hat{\mathbf{n}} dA = \int_{V_i} \frac{\boldsymbol{\tau}'}{k} . \nabla' \frac{1}{r} dV, \tag{3.2}$$

where V_i is the volume of the ith particle and

$$\boldsymbol{\tau}(\mathbf{x}) = \mathbf{F}(\mathbf{x}) + k\nabla T(\mathbf{x}) \tag{3.3}$$

may be called the 'extra flux density'. This quantity is related to the polarization stress in elasticity and the polarization vector in dielectric theory. From (1.6) we see that the dipole strength of a particle is

$$\mathbf{S}^i = \int_{V_i} \boldsymbol{\tau} dV,$$

and on comparing the expressions (1.5) and (3.3) for $\langle \mathbf{F} \rangle$ and $\boldsymbol{\tau}$ we find

$$\langle \boldsymbol{\tau} \rangle = n \langle \mathbf{S} \rangle. \tag{3.4}$$

From (3.3) it can be seen that the extra flux density $\boldsymbol{\tau}(\mathbf{x})$ is zero at points which lie in the matrix. Hence the sum of the volume integrals obtained by substituting (3.2) in the expression (3.1) for $T(x)$ may be written as a single integral over the entire volume V contained within Γ, and (3.1) becomes

$$T(x) = \frac{1}{4\pi} \int_V \frac{\boldsymbol{\tau}'}{k} . \nabla' \frac{1}{r} dV + \frac{1}{4\pi} \oint_\Gamma \left\{\frac{\mathbf{F}'}{kr} + T'\nabla' \frac{1}{r}\right\} . \hat{\mathbf{n}} dA. \tag{3.5}$$

Since we shall be concerned here with temperature gradients, we differentiate (3.5) to obtain an expression for the temperature gradient at a point in the matrix, viz.

$$\nabla T(\mathbf{x}) = -\frac{1}{4\pi k} \int_V \boldsymbol{\tau}' . \nabla'\nabla' \frac{1}{r} dV - \frac{1}{4\pi k} \oint_\Gamma \left\{\nabla' \frac{1}{r} \mathbf{F}' + kT'\nabla'\nabla' \frac{1}{r}\right\} . \hat{\mathbf{n}} dA, \tag{3.6}$$

where the minus signs on the right-hand side come from replacing ∇ by $-\nabla'$. A similar result which applies when the point $\mathbf{x}$ lies within a particle is used in § 5; in this case the integral over V must be modified since $\boldsymbol{\tau}$ is non-zero at $r = 0$.

Equation (3.6) holds for any closed surface Γ enclosing $\mathbf{x}$, and we now consider the form which this equation takes if Γ is a 'macroscopic boundary' (see § 2). If the distance from $\mathbf{x}$ to the nearest point on Γ is much greater than the distance between neighbouring particles, then the functions $\nabla' r^{-1}$ and $(\nabla'\nabla' r^{-1}) . \hat{\mathbf{n}}$ are approximately constant over a portion of Γ which cuts through many particles and so is large enough to determine an average. Thus the integral over Γ in (3.6) becomes

$$\frac{1}{4\pi k} \oint_\Gamma \left\{\nabla' \frac{1}{r} \langle \mathbf{F} \rangle + k\langle T' \rangle \nabla'\nabla' \frac{1}{r}\right\} . \hat{\mathbf{n}} dA.$$

It should be noted that the quantities $\langle \mathbf{F} \rangle$ and $\langle \nabla T \rangle$ are constant for a statistically homogeneous material, hence these terms could be taken outside the integral sign.

Substituting the above expression for the macroscopic boundary integral in (3.6), we find that the temperature gradient is given by

$$\nabla T(\mathbf{x}) = -\frac{1}{4\pi k}\int_V \boldsymbol{\tau}' \,.\, \nabla'\nabla' \frac{1}{r}\, dV - \frac{1}{4\pi k}\oint_\Gamma \left\{\nabla' \frac{1}{r}\langle \mathbf{F}\rangle + k\langle T'\rangle \nabla'\nabla' \frac{1}{r}\right\}.\hat{\mathbf{n}}dA. \quad (3.7)$$

Since the term $\nabla'\nabla' r^{-1}$ is of order r^{-3}, the integral over V in (3.7) is not absolutely convergent when taken by itself. However, the presence of the macroscopic boundary term causes the expression for $\nabla T(\mathbf{x})$ to converge, as we shall now show. We apply the divergence theorem to the surface integral in (3.7), giving

$$\oint_\Gamma \left\{\nabla' \frac{1}{r}\langle \mathbf{F}\rangle + k\langle T'\rangle \nabla'\nabla' \frac{1}{r}\right\}.\hat{\mathbf{n}}dA = -\oint_{V'} \{\langle \mathbf{F}\rangle + k\nabla\langle T'\rangle\}.\nabla'\nabla' \frac{1}{r}\, dV$$
$$-\oint_{\Gamma_s} \left\{\nabla' \frac{1}{r}\langle \mathbf{F}\rangle + k\langle T'\rangle \nabla'\nabla' \frac{1}{r}\right\}.\hat{\mathbf{n}}dA, \quad (3.8)$$

where Γ_s denotes the surface of a sphere which lies within Γ and is centred on $\mathbf{x}$, and V' denotes the volume which lies between the surfaces Γ and Γ_s. From the expression (1.5) for the bulk flux density, we see that the volume integral in (3.8) may be written as

$$-\int_{V'} n\langle \mathbf{S}\rangle \,.\, \nabla'\nabla' \frac{1}{r}\, dV. \quad (3.9)$$

This quantity, divided by $4\pi k$, may be regarded as the temperature gradient at $\mathbf{x}$ due to a uniform distribution of dipoles throughout V'.

On evaluating the integral over Γ_s in (3.8) and substituting the resulting expression for the macroscopic boundary integral in (3.7), we obtain

$$\nabla T(x) = \langle \nabla T\rangle - \frac{n}{3k}\langle \mathbf{S}\rangle - \frac{1}{4\pi k}\int_{V_s} \boldsymbol{\tau}' \,.\, \nabla'\nabla' \frac{1}{r}\, dV - \frac{1}{4\pi k}\int_{V'} \{\boldsymbol{\tau}' - n\langle \mathbf{S}\rangle\} \,.\, \nabla'\nabla' \frac{1}{r}\, dV, \quad (3.10)$$

where V_s ($= V - V'$) denotes the volume of the sphere with surface Γ_s.

To show that the integral over V' in (3.10) converges, we note that for large r the quantity $\nabla'\nabla' r^{-1}$ is approximately constant over regions which contain many particles, and hence to leading order we may replace $\boldsymbol{\tau}'$ in the integrand by its average value, $n\langle \mathbf{S}\rangle$ [see (3.4)], provided that r is sufficiently large. Thus as r increases, the integrand diminishes more rapidly than r^{-3}, and the integral converges.

Since $\boldsymbol{\tau} = 0$ in the matrix, we may neglect the integral over V_s in (3.10) if that volume lies entirely in the matrix. Thus in the limit as $V_s \to 0$, (3.10) assumes the more compact form

$$\nabla T(\mathbf{x}) = \langle \nabla T\rangle - \frac{n}{3k}\langle \mathbf{S}\rangle - \frac{1}{4\pi k}\int_{V'} \{\boldsymbol{\tau}' - n\langle \mathbf{S}\rangle\} \,.\, \nabla'\nabla' \frac{1}{r}\, dV, \quad (3.11)$$

which holds for points $\mathbf{x}$ which lie in the matrix.

An integral equation analogous to (3.11) was first obtained by Korringa (1973) for the elasticity problem, and this result was used by Willis & Acton (1976) for the calculation of effective elastic moduli. The derivation of (3.11) given here differs from that used by Korringa, in that we do not require the temperature to be given by $\langle \nabla T\rangle . \mathbf{x}$ on the boundary of the suspension; instead we have used the fact that the material and the applied field are statistically homogeneous.

In order to obtain the required expression for the dipole strength of sphere j in terms of integrals over the surrounding particles, we begin by writing (3.10) in the form

$$\nabla T(\mathbf{x}) = \nabla T^E(\mathbf{x}) - \frac{1}{4\pi k}\int_{V_j} \boldsymbol{\tau}' . \nabla'\nabla' \frac{1}{r}\, dV, \tag{3.12}$$

where

$$\nabla T^E(\mathbf{x}) = \langle \nabla T\rangle - \frac{n}{3k}\langle \mathbf{S}\rangle - \frac{1}{4\pi k}\sum_{\substack{i\\ i\neq j}} \int_{V_i} \boldsymbol{\tau}' . \nabla'\nabla' \frac{1}{r}\, dV + \frac{1}{4\pi k}\int_{V'} n\langle \mathbf{S}\rangle . \nabla'\nabla' \frac{1}{r}\, dV \tag{3.13}$$

and the sum extends over the spheres enclosed by the surface Γ, except sphere j. In deriving this expression we have replaced the integrals of $\boldsymbol{\tau}' . \nabla'\nabla' r^{-1}$ which appear in (3.10) by a sum of integrals over the particles using the fact that $\boldsymbol{\tau} = 0$ in the matrix.

The form of (3.12) is the same as that of the expression for the temperature-gradient field surrounding a single particle (sphere j) in an infinite matrix, $T^E(\mathbf{x})$ representing the temperature field in the absence of the particle. Now we may obtain the required expression for the dipole strength of sphere j from the formula for the dipole strength of a sphere placed in an ambient temperature field T^E, viz.

$$\mathbf{S} = -4\pi a^3 \beta k \nabla T^E(\mathbf{x}_0), \tag{3.14}$$

where $\beta = (\alpha-1)/(\alpha+2)$, $\mathbf{x}_0$ denotes the centre of the sphere and a is the sphere radius. The derivation of this formula is given in the appendix. In low-Reynolds number hydrodynamics, expressions like (3.14) which relate sphere properties to parameters of the ambient field are known as 'Faxén formulae'.

The required expression for the dipole strength is obtained by combining the result (3.14) with the expression (3.13) for ∇T^E, which gives

$$\mathbf{S}^j = \mathbf{S}^0 + \beta\phi\langle \mathbf{S}\rangle + \beta a^3 \sum_{\substack{i\\ i\neq j}} \int_{V_i} \boldsymbol{\tau}' . \nabla'\nabla' \frac{1}{r}\, dV - \beta a^3 \int_{V'} n\langle \mathbf{S}\rangle . \nabla'\nabla' \frac{1}{r}\, dV, \tag{3.15}$$

where

$$\mathbf{S}^0 = -4\pi a^3 \beta k \langle \nabla T\rangle \tag{3.16}$$

is the dipole strength of the reference sphere in the absence of particle interaction. The expression (3.15) is valid if the volume V' has a spherical inner surface, concentric with sphere j.

If the radius of this inner surface Γ_s is much larger than the average distance between neighbouring particles, the contribution to $\mathbf{S}^j$ from particles which lie within V' is approximately cancelled by the dipole-field term

$$\beta a^3 \int_{V'} n\langle \mathbf{S}\rangle . \nabla'\nabla' \frac{1}{r}\, dV,$$

hence (3.15) may be written in the form

$$\mathbf{S}^j = \mathbf{S}^0 + \beta\phi\langle \mathbf{S}\rangle + \beta a^3 \sum_{\substack{i\\ i\neq j}} \int_{V_i} \boldsymbol{\tau}' . \nabla'\nabla' \frac{1}{r}\, dV, \tag{3.17}$$

where the sum now only extends over the particles which lie within Γ_s. An expression analogous to (3.17) forms the starting point for the derivation of the Clausius–Mosotti formula for the dielectric constant of a dense material (Panofsky & Phillips 1962); in this case the expression is obtained by assuming that the suspension which lies outside Γ_s may be treated as a continuous dielectric, and the term $\beta\phi\langle \mathbf{S}\rangle$ arises from the polarization charges on the surface Γ_s.

4. Rayleigh's non-convergent sum

In this section we show why Rayleigh was able to obtain the correct expression for the conductivity of a cubic array of spheres by summing a non-absolutely convergent series in a particular order. To calculate the dipole strength of a sphere in a cubic array, Rayleigh assumed that the external field $\nabla T^E(\mathbf{x})$ is given by $\langle \nabla T\rangle$ plus the sum of the fields produced by the surrounding spheres. Rayleigh replaced the surrounding particles by a dipole and several higher-order poles, but for our purposes it is sufficient to consider the approximate form of Rayleigh's expression obtained by replacing the surrounding spheres by dipoles, viz.

$$\nabla T^E(\mathbf{x}) = \langle \nabla T\rangle - \frac{1}{4\pi k}\sum_{\substack{i\\ i\neq j}} \mathbf{S}\,.\,\nabla'\nabla'\,\frac{1}{r^i}, \tag{4.1}$$

where $\mathbf{S}$ denotes the (uniform) dipole strength of the spheres and r^i is the distance from $\mathbf{x}$ to the centre of sphere i. (In Rayleigh's notation, $|\nabla T_E(\mathbf{x})|$, $|\langle \nabla T\rangle|$ and $-|S|/4\pi k$ are denoted by A_1, H and B_1 respectively. The expression (4.1) is equivalent to the first of the equations (62) in Rayleigh's paper, with the higher-order multipole terms such as B_3 removed.) On combining (4.1) with the Faxén-type formula (3.14), we get

$$\mathbf{S} = \mathbf{S}^0 + \beta a^3 \mathbf{S}\,.\sum_{\substack{i\\ i\neq j}} \nabla'\nabla'\,\frac{1}{r^i}. \tag{4.2}$$

As mentioned earlier, the sum of $\nabla'\nabla'(r^i)^{-1}$ over the surrounding spheres is non-absolutely convergent, and unless the order of summation is specified the expression is meaningless. From (3.13) it can be seen that, in formulating the expression for ∇T^E, Rayleigh incorrectly assessed the contribution from the macroscopic boundary (referred to, rather obscurely, as 'the potential due to the sources at infinity other than the spheres', p. 489), and this is why he obtained a non-convergent sum.

The correct expression for $\mathbf{S}$ is obtained from (3.15), viz.

$$\mathbf{S} = \mathbf{S}^0 + \beta\phi\mathbf{S} + \beta a^3 \mathbf{S}\,.\left\{\sum_{\substack{i\\ i\neq j}} \nabla'\nabla'\,\frac{1}{r^i} - n\int_{V'} \nabla'\nabla'\,\frac{1}{r}\,dV\right\}, \tag{4.3}$$

where we have replaced the surrounding particles by dipoles. This expression converges as the volume V' becomes infinite, thanks to the dipole-field term

$$n\int_{V'} \nabla'\nabla'\,\frac{1}{r}\,dV.$$

On comparing (4.3) with Rayleigh's expression (4.2), we see that both this dipole-field term and the term $\beta\phi\mathbf{S}$ are absent.

In order to evaluate the expression (4.2) Rayleigh summed the terms in the following way: he first calculated the sum over the spheres contained in an infinitely long cylinder of square cross-section. The axis of the cylinder was chosen to coincide with one of the axes of the lattice and $\langle\nabla T\rangle$ was taken to be parallel to this axis. By letting the cross-section of the cylinder become infinite, Rayleigh obtained a value for the sum. With the aid of (4.3) we can now see why this particular order of summation leads to the correct result. We let the volume V' in (4.3) denote the volume of the cylinder described above. On applying the divergence theorem to the dipole-field term in (4.3), we get

$$\mathbf{S}\,.\int_{V'}\nabla'\nabla'\frac{1}{r}\,dV = -|\mathbf{S}|\int_{\Gamma}\frac{\partial}{\partial x}\left(\frac{1}{r}\right)\hat{\mathbf{n}}\,dA - \mathbf{S}\,.\int_{\Gamma_s}\nabla'\left(\frac{1}{r}\right)\hat{\mathbf{n}}\,dA, \qquad (4.4)$$

where x denotes the component of $\mathbf{r}$ in the direction of $\mathbf{S}$, Γ denotes the surface of the cylinder, and as usual Γ_s is a sphere centred on the point $r = 0$. For a cubic lattice $\mathbf{S}$ is parallel to $\langle\nabla T\rangle$, and since $\langle\nabla T\rangle$ is parallel to the cylinder axis it can be seen from symmetry considerations that the integral over Γ in (4.4) is zero. Evaluating the integral over Γ_s in (4.4), we get

$$\mathbf{S}\,.\int_{V'}\nabla'\nabla'\frac{1}{r}\,dV = \frac{4\pi}{3}\,\mathbf{S},$$

and on substituting this result in (4.3), we obtain Rayleigh's expression (4.2) for the dipole strength. Thus by summing the terms in a special way, Rayleigh was able to obtain the correct value for the dipole strength from an improper expression.

For an array in which the lattice vectors are not mutually orthogonal, Rayleigh's procedure runs into difficulties, for $\langle\mathbf{S}\rangle$ and $\langle\nabla T\rangle$ are not in general parallel for this type of material, and it is not easy to see how the sum in (4.2) should be evaluated in this case. No such difficulties are encountered in the application of (4.3) presented here, since the expression is absolutely convergent. Higher-order terms in the expression for $\mathbf{k}^*$ for this type of material may be obtained by replacing the spheres surrounding the reference sphere by dipoles and a number of higher-order poles, and coupling equation (3.15) for $\mathbf{S}^j$ with Rayleigh's expressions (62) for the higher-order multipole strengths of the reference sphere; the latter expressions are correct, for the contribution from distant spheres falls off faster than $1/r^3$, and there is no macroscopic boundary term.

This method is similar to that adopted by McKenzie & McPhedran (1978) in their study of the electrical conductivity of a simple cubic array of spheres. These authors modified Rayleigh's expression (4.1) by replacing the average field $\langle\nabla T\rangle$ by a term $\mathbf{E}_0$, referred to as 'the external electric field'. By using the argument which was employed on the derivation of the Clausius–Mossoti formula (described at the end of § 3) they show that

$$\mathbf{E}_0 = \langle\nabla T\rangle + \mathbf{E}_p,$$

where

$$\mathbf{E}_p = \frac{1}{4\pi k}\int_{\Gamma}\nabla'\frac{1}{r}\langle\mathbf{S}\rangle\,.\,\hat{\mathbf{n}}\,dA.$$

These results correspond to equations (24) and (19) respectively of McKenzie & McPhedran's paper. On applying the divergence theorem to the above integral and substituting the resulting expression for $\mathbf{E}_0$ in place of $\langle\nabla T\rangle$ in (4.1), we obtain the correct expression for $\nabla T^E(\mathbf{x})$. The method presented here is similar to McKenzie & McPhedran's method in that both make use of an expression for $\nabla T^E(\mathbf{x})$ of the form

(3.13). However the latter method is limited to conductivity-type problems for regular arrays of spheres.

Although our method may be used for the determination of other transport properties of regular arrays (O'Brien 1977), we shall not present the details here; instead we turn to the more important problem of determining the effective transport properties of a random suspension of spheres.

5. Conduction through a random array of spheres

In this section we describe a procedure for obtaining the effective conductivity of a random suspension of spheres without the need for renormalizing. Our aim is to obtain an expression for the effective conductivity tensor $\mathbf{k}^*$, correct to $O(\phi^2)$, where as usual ϕ denotes the particle volume fraction. From the expression (1·5) for the average flux density, it can be seen that to obtain the required formula for $\mathbf{k}^*$ we need an expression for the average dipole strength $\langle \mathbf{S}\rangle$ correct to $O(\phi)$.

We begin by writing equation (3.15) for the dipole strength of the reference sphere in the form

$$\mathbf{S}^j = \mathbf{S}^0 + \beta\phi\langle \mathbf{S}\rangle + 4\pi\beta a^3 k \sum_{\substack{i \\ i\neq j}} \nabla\theta(\mathbf{x}_i) - \beta a^3 \int_{V'} n\langle \mathbf{S}\rangle \,.\, \nabla'\nabla' \frac{1}{r}\, dV, \qquad (5.1)$$

where

$$\nabla\theta(\mathbf{x}_i) = \frac{1}{4\pi k}\int_{V_i} \boldsymbol{\tau}' \,.\, \nabla'\nabla' \frac{1}{r}\, dV \qquad (5.2)$$

may be regarded as the contribution to the external field at the centre of the reference sphere (sphere j) from sphere i, and $\mathbf{x}_i$ denotes the position of the centre of sphere i.

For convenience, we shall take our origin at the centre of the reference sphere. On taking the ensemble average of (5.1), we get

$$\langle \mathbf{S}\rangle = \mathbf{S}^0 + \beta\phi\langle \mathbf{S}\rangle + \beta a^3 \int_{r=2a}^{\infty} \left\{4\pi k\langle\nabla\theta(\mathbf{r}|\mathbf{0})\rangle p(\mathbf{r}|\mathbf{0}) - n\langle \mathbf{S}\rangle \,.\, \nabla\nabla \frac{1}{r}\right\} dV, \qquad (5.3)$$

where $\langle\nabla\theta(\mathbf{r}|0)\rangle$ is the average value of $\nabla\theta(\mathbf{r})$, averaged over all configurations for which there is a sphere at $\mathbf{r}$ and another at the origin. In deriving this result we have taken the inner surface of V' in (5.1) to coincide with the surface of the reference sphere $r = 2a$.

If there is no long-range order in the suspension, we have

$$p(\mathbf{r}|\mathbf{0}) \sim n \quad \text{as} \quad r \to \infty,$$

and since

$$4\pi k\langle\nabla\theta(\mathbf{r}|\mathbf{0})\rangle \sim \langle \mathbf{S}\rangle \,.\, \nabla\nabla r^{-1} \quad \text{as} \quad r\to\infty,$$

the integral in (5.3) converges.

We can write (5.3) in a more convenient form with the aid of the relation

$$\langle \mathbf{S}(\mathbf{0}|\mathbf{r})\rangle = \mathbf{S}^0 + 4\pi k\beta a^3\langle\nabla\theta(\mathbf{r}|\mathbf{0})\rangle + \Big[\beta\phi\langle \mathbf{S}\rangle - \beta a^3 \int_{V(\mathbf{r})} n\langle \mathbf{S}\rangle \,.\, \nabla\nabla\frac{1}{r}\, dV$$
$$+ \beta a^3 \int_{V'-V(\mathbf{r})} \left\{4\pi k\langle\nabla\theta(\mathbf{r}'|\mathbf{0},\mathbf{r})\rangle p(\mathbf{r}'|\mathbf{0},\mathbf{r}) - n\langle \mathbf{S}\rangle \,.\, \nabla\nabla\frac{1}{r}\right\} dV\Big], \qquad (5.4)$$

obtained by averaging (5.1) over all configurations for which there is a sphere at $\mathbf{0}$ and one at $\mathbf{r}$. Here $V(\mathbf{r})$ denotes the volume of a spherical particle, centred on $\mathbf{r}$, the term $\langle \mathbf{S}(\mathbf{0}|\mathbf{r})\rangle$ denotes the average dipole strength of a sphere centred on the origin, given that there is a sphere at $\mathbf{r}$, and similarly $\langle \nabla\theta(\mathbf{r}'|\mathbf{0},\mathbf{r})\rangle$ is the average value of $\nabla\theta(\mathbf{r}')$ averaged over all configurations for which there are spheres at $\mathbf{0}$, $\mathbf{r}$ and $\mathbf{r}'$. $p(\mathbf{r}'|\mathbf{0},\mathbf{r})$ is the probability that a sphere lies in a unit volume about $\mathbf{r}'$, given that there are spheres at $\mathbf{0}$ and $\mathbf{r}$. Since the quantities n and $p(\mathbf{r}'|\mathbf{0},\mathbf{r})$ are $O(\phi)$, we expect that the square-bracketed terms in (5.4) will also be $O(\phi)$, hence we may write (5.4) in the form

$$4\pi k\beta a^3\langle \nabla\theta(\mathbf{r}|\mathbf{0})\rangle = \langle \mathbf{S}(\mathbf{0}|\mathbf{r})\rangle - \mathbf{S}^0 + O(\phi).$$

On substituting this expression for $\langle \nabla\theta(\mathbf{r}|\mathbf{0})\rangle$ in (5.3), we obtain

$$\langle \mathbf{S}\rangle = \mathbf{S}^0 + \beta\phi\langle \mathbf{S}\rangle + \int_{r=2a}^{\infty}\left[\{\langle \mathbf{S}(\mathbf{0}|\mathbf{r})\rangle - \mathbf{S}^0\}p(\mathbf{r}|\mathbf{0}) - \beta a^3 n\langle \mathbf{S}\rangle . \nabla\nabla\frac{1}{r}\right]dV + O(\phi^2). \tag{5.5}$$

To evaluate the integral in (5.5) we require an expression for $\langle \mathbf{S}(\mathbf{0}|\mathbf{r})\rangle$. If we neglect terms $O(\phi)$, then $\langle \mathbf{S}(\mathbf{0}|\mathbf{r})\rangle$ is equal to the dipole strength $\mathbf{S}(\mathbf{0}|\mathbf{r})$ of one of a pair of spheres with separation vector $\mathbf{r}$ alone in an infinite matrix with the far-field boundary condition $\nabla T(\mathbf{x}) \to \langle \nabla T\rangle$ at points far from the particles. To show this, we take the average of the expression (3.11) for $\nabla T(\mathbf{x})$ over the configurations for which there is one sphere at the origin and one sphere at $\mathbf{r}$; this gives

$$\langle \nabla T(\mathbf{x}|\mathbf{0},\mathbf{r})\rangle = \langle \nabla T\rangle - \sum_{i=1}^{2}\frac{1}{4\pi k}\int_{V^i}\langle \boldsymbol{\tau}(\mathbf{x}'|\mathbf{0},\mathbf{r})\rangle . \nabla'\nabla'\frac{1}{r}\,dV + O(\phi), \tag{5.6}$$

where the sum extends over the two spheres at $\mathbf{0}$ and $\mathbf{r}$ respectively. For a pair of spheres alone in an infinite matrix, $\nabla T(\mathbf{x})$ is given by

$$\nabla T(\mathbf{x}) = \langle \nabla T\rangle - \sum_{i=1}^{2}\frac{1}{4\pi k}\int_{V^i}\boldsymbol{\tau}(\mathbf{x}') . \nabla'\nabla'\frac{1}{r}\,dV.$$

This expression has the same form as (5.6) with the $O(\phi)$ term removed, and it is not difficult to show from (5.6) and the analogous expressions for $\nabla T(\mathbf{x})$ in particles 1 and 2 that the averaged field $\langle \nabla T(\mathbf{x}|\mathbf{0},\mathbf{r})\rangle$ satisfies the same differential equation and boundary conditions, to $O(\phi)$, as the temperature gradient field for the two-sphere problem; hence

$$\langle \mathbf{S}(\mathbf{0}|\mathbf{r})\rangle = \mathbf{S}(\mathbf{0}|\mathbf{r}) + O(\phi).$$

On substituting this result in (5.5) we get

$$\langle \mathbf{S}\rangle = \mathbf{S}^0 + \beta\phi\mathbf{S}^0 + \int_{r=2a}^{\infty}\left[\{\mathbf{S}(\mathbf{0}|\mathbf{r}) - \mathbf{S}^0\}p(\mathbf{r}|\mathbf{0}) - \beta a^3 n\mathbf{S}^0 . \nabla\nabla\frac{1}{r}\,dV\right] + O(\phi^2). \tag{5.7}$$

With the aid of the expressions (1.5) and (3.16) for $\langle \mathbf{F}\rangle$ and $\mathbf{S}^0$, we find from (5.7) that the bulk flux density is given by

$$\langle F\rangle = -k\langle \nabla T\rangle - 3\phi k\beta\langle \nabla T\rangle(1+\phi\beta) + n\int_{r=2a}^{\infty}\left[\{\mathbf{S}(\mathbf{0}|\mathbf{r}) - \mathbf{S}^0\}p(\mathbf{r}|\mathbf{0}) - \beta n a^3\mathbf{S}^0 . \nabla\nabla\frac{1}{r}\right]dV. \tag{5.8}$$

We can write this expression in the same form as Jeffrey's (1973) result (equation (3.13) in that paper), by noting that the temperature gradient at a point $\mathbf{r}$ caused by a single sphere at $\mathbf{0}$ in an infinite matrix is given by

$$\nabla T(\mathbf{r}|\mathbf{0}) = \langle \nabla T\rangle + \frac{\mathbf{S}^0}{4\pi k} . \nabla\nabla\frac{1}{r}.$$

Combining this result with (5.8), we get

$$\langle \mathbf{F} \rangle = -k\langle \nabla T\rangle - 3\phi k\beta \langle \nabla T\rangle (1+\phi\beta) \\ + n\int_{r=2a}^{\infty} [\{\mathbf{S}(\mathbf{0}|\mathbf{r}) - \mathbf{S}^0\} p(\mathbf{r}|\mathbf{0}) - 4\pi k n a^3 \beta \{\nabla T(\mathbf{r}|\mathbf{0}) - \langle \nabla T\rangle\}]\, dV,$$

which is the same as Jeffrey's expression for $\langle \mathbf{F} \rangle$.

Jeffrey obtained this result using the renormalization method described in § 1. To apply that technique it is necessary to obtain a renormalizing quantity. The method presented here has the advantage that this renormalizing quantity arises naturally from the integral over the macroscopic boundary in the expression (3.7) for $\nabla T(\mathbf{x})$, and it is now clear that the convergence difficulties encountered in the past simply do not arise when the macroscopic boundary term is included.

In their investigation into the effective elastic moduli of a random suspension of spheres, Willis & Acton (1976) make use of an integral equation which is analogous to the expression (3.11) for $\nabla T(\mathbf{x})$. With the aid of this expression, the authors obtain an integral equation for the polarization stress within a particle (equation (2.8) of that paper) of which the counterpart in the thermal problem would be†

$$\boldsymbol{\tau}(\mathbf{x}) = -3\beta \left[k\langle \nabla T\rangle - \frac{n}{3}\langle \mathbf{S}\rangle - \frac{1}{4\pi}\sum_i \int_{V_i} \boldsymbol{\tau} . \nabla'\nabla' \frac{1}{r}\, dV \right. \\ \left. + \frac{1}{4\pi}\int_{V'} n\langle \mathbf{S}\rangle . \nabla'\nabla' \frac{1}{r}\, dV \right], \quad (5.9)$$

where as usual $\beta = (\alpha-1)/(\alpha+2)$, and the sum extends over the spheres contained in V'.

On the assumption that only pair interactions need be considered in order to calculate $\langle \mathbf{S}\rangle$ to $O(\phi)$, Willis & Acton replace this expression for the polarization stress by

$$\boldsymbol{\tau}(\mathbf{x}) = -3\beta \left[k\langle \nabla T\rangle - \frac{n}{3}\langle \mathbf{S}\rangle - \frac{1}{4\pi}\sum_{i=1}^{2} \int_{V_i} \boldsymbol{\tau}' . \nabla'\nabla' \frac{1}{r}\, dV \right. \\ \left. + \frac{1}{4\pi}\int_{V'} n\langle \mathbf{S}\rangle . \nabla'\nabla' \frac{1}{r}\, dV \right], \quad (5.10)$$

where the sum here extends over only two of the spheres in V', one of which encloses $\mathbf{x}$ (see equation (4.4) of their paper). This is an erroneous expression for $\boldsymbol{\tau}(\mathbf{x})$, because it is inconsistent to neglect the contributions to $\boldsymbol{\tau}(\mathbf{x})$ from the other particles in V' while including the terms $\frac{1}{3}n\langle \mathbf{S}\rangle$ and

$$\frac{1}{4\pi}\int_{V'} n\langle \mathbf{S}\rangle . \nabla'\nabla' \frac{1}{r}\, dV,$$

which arise from the fact that the material within V' is a statistically homogeneous suspension. As a consequence the integral over V' is not absolutely convergent, and

† This result is obtained by using an integral expression for the temperature at a point $\mathbf{x}$ in a particle, similar to the expression (3.1) for the temperature in the matrix. An expression for $\nabla T(\mathbf{x})$ may then be derived by a similar procedure to that employed in the derivation of the expression (3.10) for ∇T in the matrix, and by combining this result with the formula

$$\boldsymbol{\tau}(\mathbf{x}) = (1-\alpha)\, k\nabla T(\mathbf{x}),$$

which is derived from (3.3), we obtain (5.9).

the authors (arbitrarily) choose V' to be a large sphere; for this choice of V', the integral is a constant, independent of the sphere radius.

Willis & Acton use an iterative procedure to obtain an approximate solution to the integral equation (5.10) for $\boldsymbol{\tau}(\mathbf{x})$; for each iteration the integral over the neighbouring sphere and the quantity $\langle \mathbf{S} \rangle$ in (5.10) are both calculated by using the formula for $\boldsymbol{\tau}(\mathbf{x})$ given by the previous iteration. This procedure results in an expression for $\boldsymbol{\tau}(\mathbf{x})$ as a power series in a/r_s, where r_s is the separation distance between the spheres. An expression for the dipole strength of the reference sphere, denoted here by $\mathbf{S}^*(\mathbf{r}_s)$, is then obtained by integrating the formula for $\boldsymbol{\tau}(\mathbf{x})$ over the volume of the reference sphere. This quantity $\mathbf{S}^*(\mathbf{r}_s)$ depends on the shape of the volume V'.

In order to show how Willis & Acton obtain the correct expression for $\langle \mathbf{S} \rangle$ by using the volume-dependent expression for $\mathbf{S}^*(\mathbf{r}_s)$, we first integrate the formula (5.10) for $\boldsymbol{\tau}(\mathbf{x})$ over the volume of the reference sphere (sphere 1), which gives

$$\mathbf{S}^*(\mathbf{r}_s) = \mathbf{S}^0 + \beta\phi\langle \mathbf{S} \rangle - \beta a^3 \int_{V'} n\langle \mathbf{S} \rangle \cdot \nabla'\nabla' \frac{1}{r}\, dV + 4\pi k\beta a^3 \nabla\theta^*(\mathbf{r}_s), \tag{5.11}$$

where

$$\nabla\theta^*(\mathbf{r}_s) = \frac{1}{4\pi k}\int_{V_2} \boldsymbol{\tau}^*(\mathbf{x}') \cdot \nabla'\nabla' \frac{1}{r}\, dV. \tag{5.12}$$

Here $\boldsymbol{\tau}^*(\mathbf{x})$ denotes the solution of the integral equation (5.10), and r denotes distance from the centre of the reference sphere. The expression (5.11) is equivalent to equation (4.19) of Willis & Acton's paper, although in the latter case the quantity $\boldsymbol{\tau}^*(\mathbf{x})$ is replaced by an approximate expression correct to order $(a/r_s)^7$.

Using the averaging procedure adopted by Willis & Acton, we obtain from (5.11) the following expression for the average dipole strength:

$$\langle \mathbf{S} \rangle = \mathbf{S}^0 + \beta\phi\langle \mathbf{S} \rangle + \beta a^3 \int_{V'} \left\{ 4\pi k \langle \nabla\theta^*(\mathbf{r}) \rangle p(\mathbf{r}|\mathbf{0}) - n\langle \mathbf{S} \rangle \cdot \nabla\nabla \frac{1}{r} \right\} dV, \tag{5.13}$$

which is analogous to equation (4.20) of their paper. The quantity $p(\mathbf{r}|\mathbf{0})$ in (5.13) is the usual pair probability function. It is difficult to reconcile this method of averaging with the earlier step of neglecting all but one of the neighbouring spheres in V', for in this case we should expect that the pair probability function in (5.13) would depend on *which* spheres are ignored for each configuration in the ensemble; for example, if we ignore all spheres except the one which is nearest to the reference sphere, then the pair probability function would vanish with increasing r, whereas $p(\mathbf{r}|\mathbf{0}) \to n$ as $|\mathbf{r}| \to \infty$.

In order to calculate $\langle \mathbf{S} \rangle$ correct to $O(\phi)$ with the aid of (5.13) we require an expression for $\langle \nabla\theta^*(\mathbf{r}) \rangle$, correct to $O(1)$. To obtain $\langle \nabla\theta^*(\mathbf{r}) \rangle$ to this accuracy, we substitute for $\boldsymbol{\tau}^*$ in (5.12) the solution to the integral equation

$$\boldsymbol{\tau}^*(\mathbf{x}) = -3\beta \left[k\langle \nabla T \rangle - \frac{1}{4\pi} \sum_{i=1}^{2} \int_{V_i} \boldsymbol{\tau}^*(\mathbf{x}') \cdot \nabla'\nabla' \frac{1}{r}\, dV \right] \tag{5.14}$$

obtained by neglecting the $O(\phi)$ terms in (5.10). In neglecting these terms we have removed the dependence of $\boldsymbol{\tau}^*$ (and hence $\nabla\theta^*(\mathbf{r})$) on V'. It is not difficult to show that the integral equation (5.14) is identical to the equation for $\boldsymbol{\tau}(\mathbf{x})$ for the case of a pair of spheres alone in an infinite matrix with uniform temperature gradient $\langle \nabla T \rangle$ far from the spheres. By integrating (5.14) over the reference sphere, we get

$$4\pi k\beta a^3 \nabla\theta^*(\mathbf{r}) = \mathbf{S}(\mathbf{0}|\mathbf{r}) - \mathbf{S}^0 + O(\phi)$$

and, on substituting in (5.13), we obtain the correct expression (5.7) for $\langle \mathbf{S} \rangle$. Thus Willis & Acton's procedure yields a result which does not depend on V' simply because the quantity $\nabla\theta^*(\mathbf{r})$ is only required to $O(1)$, and to this order of accuracy, $\nabla\theta^*$ is independent of V'.

Although this procedure gives the correct result, it is complicated and it obscures the fact that there *are* significant multi-particle and outer-boundary effects which must be taken into account, even though the final expression for $\langle \mathbf{S} \rangle$ appears to involve only two-sphere interactions.

6. The determination of the effective elastic moduli or the viscosity of a random suspension of spheres

We now turn to the problem of calculating the effective elastic moduli of a random suspension of elastic spheres embedded in an elastic matrix. The problem of determining the effective shear modulus for the case of incompressible particles and an incompressible matrix is mathematically identical to that of determining the effective viscosity of a suspension of spherical droplets immersed in a liquid matrix (Batchelor & Green 1972*b*, § 7) for a given statistical geometry of the particles. Thus the results described here may also be applied to the viscosity problem, although in that case the statistical properties of the particle configuration are affected by the bulk flow. In this section we assume that the relevant statistical properties of the configuration are given.

Our aim is to determine the effective elastic moduli, correct to $O(\phi^2)$. This is the problem studied by Willis & Acton (1976), who obtained approximate formulae for the coefficients of the ϕ^2 terms in the expressions for the elastic moduli, using the procedure described in § 5. In the case of a suspension of rigid particles in an incompressible matrix, a more accurate estimate of the ϕ^2 term for the shear modulus has been obtained by Batchelor & Green (1972*b*) using the renormalization procedure. This latter procedure was also used by Chen & Acrivos (1978) for the determination of the effective modulus of compression of a suspension of elastic spheres and for the determination of the shear modulus for rigid spheres or spherical cavities embedded in an elastic matrix. However, in this case there are a number of possible renormalizing quantities, a point which we shall discuss at the end of this section.

The local stress tensor $\boldsymbol{\sigma}$ is the counterpart of the flux density $\mathbf{F}$, and $\langle \boldsymbol{\sigma} \rangle$ is related to a particle average by an expression of the form

$$\langle \boldsymbol{\sigma} \rangle = \langle \boldsymbol{\sigma}^0 \rangle + n \langle \mathbf{S} \rangle, \tag{6.1}$$

where $\boldsymbol{\sigma}^0(\mathbf{x}) = \boldsymbol{\sigma}(\mathbf{x})$ if $\mathbf{x}$ lies in the matrix, while if $\mathbf{x}$ lies in a particle, $\boldsymbol{\sigma}^0(\mathbf{x})$ denotes the stress which would be obtained at $\mathbf{x}$ if that particle could be replaced by matrix material, with the strain at $\mathbf{x}$ held fixed. Thus the quantity $\langle \boldsymbol{\sigma}^0 \rangle$ is the counterpart of $-k\langle \nabla T \rangle$, i.e. the stress in the matrix material for a uniform strain equal to the average strain in the composite material. The particle dipole strength is here a second-order tensor, given by

$$\mathbf{S} = \int_{V_p} \boldsymbol{\tau}\, dV, \tag{6.2}$$

where

$$\boldsymbol{\tau} = \boldsymbol{\sigma} - \boldsymbol{\sigma}^0 \tag{6.3}$$

is termed the 'polarization stress' (Willis & Acton 1976).

In order to calculate the average stress $\langle \boldsymbol{\sigma} \rangle$ (and hence the elastic moduli) correct to $O(\phi^2)$, we require a formula for $\langle \mathbf{S} \rangle$, correct to $O(\phi)$. The starting point for the derivation of this formula is an integral expression for the displacement $\mathbf{u}$ at a point in the matrix, namely

$$u_p(\mathbf{x}) = -\sum_i \int_{V_i} \frac{\partial G_{pm}}{\partial x_k'}(\mathbf{x}-\mathbf{x}')\,\tau_{mk}(\mathbf{x}')\,dV(\mathbf{x}') - \int_\Gamma \{G_{pm}(\mathbf{x}-\mathbf{x}')\,\sigma_{mk}\,n_k - u_m(\mathbf{x}')\,J_{mkp}(\mathbf{x}-\mathbf{x}')\,n_k\}\,dA(\mathbf{x}'). \quad (6.4)$$

The tensor $\mathbf{G}$ is the Green's function for an infinite elastic matrix, defined by (Landau & Lifshitz 1959, p. 30)

$$G_{pm}(x) = \frac{1}{16\pi\mu(1-\nu)}\left[\frac{(3-4\nu)}{|x|}\,\delta_{pm} + \frac{x_p\,x_m}{|x|^3}\right], \quad (6.5)$$

where μ and ν are the modulus of rigidity and Poisson's ratio for the matrix. The quantity $G_{pm}(\mathbf{x})\,F_m$ is the displacement at $\mathbf{x}$ caused by the application of a point force at the origin, and $J_{mkp}\,F_m$ is the stress associated with that displacement field.

On taking the gradient of (6.4) and using the fact that the quantities $\boldsymbol{\sigma}$ and $\mathbf{u}$ which appear in the macroscopic boundary integral may be replaced by their averaged values $\langle \boldsymbol{\sigma} \rangle$ and $\langle \mathbf{u} \rangle$, we obtain an expression for the strain $\mathbf{e}(\mathbf{x})$ which is analogous to the expression (3.7) for the temperature gradient. By using the divergence theorem to convert the macroscopic boundary integral in the expression for $\mathbf{e}(\mathbf{x})$ to a volume integral, we get

$$e_{pq}(\mathbf{x}) = \langle e_{pq} \rangle + \frac{n\eta_1}{\mu}\langle S_{pq} \rangle + \frac{n\eta_2}{\mu}\,\delta_{pq}\langle S_{mm} \rangle + \sum_i \int_{V_i} P_{pqmk}(\mathbf{x}-\mathbf{x}')\,\tau_{mk}'\,dV(\mathbf{x}') - n\int_{V'} P_{pqmk}(\mathbf{x}-\mathbf{x}')\,\langle S_{mk} \rangle\,dV, \quad (6.6)$$

where $$\eta_1 = (4-5\nu)/15(1-\nu), \quad \eta_2 = -1/30(1-\nu) \quad (6.7)$$

and $$P_{pqmk} = \frac{1}{2}\left(\frac{\partial^2 G_{pm}}{\partial x_q'\,\partial x_k'} + \frac{\partial^2 G_{qm}}{\partial x_p'\,\partial x_k'}\right).$$

The result (6.6) is equivalent to the expression (2.7) in Willis & Acton's (1976) paper.

As in the case of the conduction problem (§ 3), we seek an expression relating the dipole strength $\mathbf{S}$ of a sphere in a statistically homogeneous suspension to integrals over the surrounding particles. By taking the average of this expression, with first one and then two spheres held fixed, we obtain an expression for $\langle \mathbf{S} \rangle$, correct to $O(\phi)$.

The required expression for $\mathbf{S}$ is obtained with the aid of a Faxén-type formula for the dipole strength of a sphere placed in an ambient strain field, viz.

$$\mathbf{S} = (\alpha K - \tfrac{2}{3}\beta\mu)\,\mathbf{I}\int_{V_p} \|\mathbf{e}^E\|\,dV + 2\beta\mu\int_{V_p} \mathbf{e}^E dV, \quad (6.8)$$

where $$\alpha = \frac{(K_p-K)}{K}\,\frac{(4\mu+3K)}{(4\mu+3K_p)}, \quad \beta = \left\{\frac{2(4-5\nu)}{15(1-\nu)} - \frac{\mu}{\mu-\mu_p}\right\}^{-1}, \quad (6.9)$$

and V_p is the volume of the particle. Here $\mathbf{e}^E$ denotes the strain tensor at $\mathbf{x}$ in the absence of the particle and $\|\mathbf{e}^E\|$ denotes its trace, μ_p and K_p are the modulus of rigidity and modulus of compression of the particle and K is the modulus of compression of the matrix. The formula (6.8) is derived by using the reciprocal theorem together with the expression for the (uniform) stress in a sphere placed in a uniform strain field (Eshelby 1957); the procedure is analogous to that used in the derivation of the Faxén-type formula (3.14) for the thermal dipole strength, described in the appendix.

The dipole strength of sphere j in a statistically homogeneous suspension is given by an expression of the form (6.8), where

$$e^E_{pq}(\mathbf{x}) = \langle e_{pq}\rangle + \frac{n\eta_1}{\mu}\langle S_{pq}\rangle + \frac{n\eta_2}{\mu}\delta_{pq}\langle S_{mm}\rangle + \sum_{\substack{i\\ i\neq j}} \int_{V_i} P_{pqmk}(\mathbf{x}-\mathbf{x}')\,\tau'_{mk}\,dV(\mathbf{x}') - n\int_{V'} P_{pqmk}(\mathbf{x}-\mathbf{x}')\langle S_{mk}\rangle\,dV. \quad (6.10)$$

The quantity $\mathbf{e}^E(\mathbf{x})$ may be thought of as the strain tensor which would be obtained at $\mathbf{x}$ if the reference sphere were replaced by matrix material with the stress in the surrounding particles held fixed.

On expanding $\mathbf{e}^E(\mathbf{x})$ in (6.8) in a Taylor series about the centre of the reference sphere, which we take as the origin, and using the fact that $\nabla^4\mathbf{u}^E = \mathbf{0}$ and $\nabla^2\|\mathbf{e}^E\| = 0$, we get

$$\mathbf{S}^j = \tfrac{4}{3}\pi a^3[(\alpha K - \tfrac{2}{3}\beta\mu)\,\|\mathbf{e}^E(\mathbf{0})\|\,\mathbf{I} + 2\beta\mu(\mathbf{e}^E(\mathbf{0}) + \tfrac{1}{10}a^2\nabla^2\mathbf{e}^E(\mathbf{0}))], \quad (6.11)$$

where a is the radius of the particle. From the expression (6.4) for the displacement, we find that $\nabla^2\mathbf{e}^E$ is given by

$$\nabla^2 e^E_{pq}(\mathbf{0}) = \sum_{\substack{i\\ i\neq j}} \int_{V_i} \nabla^2 P_{pqmk}(\mathbf{x})\,\tau_{mk}\,dV(\mathbf{x}). \quad (6.12)$$

Note that there are no macroscopic boundary terms in this expression, for the terms in the integral over Γ drop off like $1/r^4$ and the integral may thus be neglected in the limit as V' becomes infinite.

By combining (6.10) and (6.12) with the Faxén-type formula (6.11), we obtain an expression, analogous to (3.15), relating the dipole strength of the reference sphere to integrals over the surrounding spheres. However, as this expression is rather cumbersome we shall work with each of (6.10), (6.11) and (6.12) separately. Taking the average of (6.11), we get

$$\langle\mathbf{S}\rangle = \tfrac{4}{3}\pi a^3[(\alpha K - \tfrac{2}{3}\beta\mu)\,\|\langle\mathbf{e}^E(\mathbf{0})\rangle\|\,\mathbf{I} + 2\beta\mu(\langle\mathbf{e}^E(\mathbf{0})\rangle + \tfrac{1}{10}a^2\langle\nabla^2\mathbf{e}^E(\mathbf{0})\rangle)], \quad (6.13)$$

and on combining the expressions obtained by averaging (6.10) and (6.12) first with one sphere fixed at the origin and then with another fixed at $\mathbf{r}$, we obtain

$$\langle e^E_{pq}(\mathbf{0})\rangle = \langle e_{pq}\rangle + \frac{n\eta_1}{\mu}\langle S_{pq}\rangle + \frac{n\eta_2}{\mu}\delta_{pq}\langle S_{mm}\rangle + \int_{r=2a}^{\infty} [\{\langle e^E_{pq}(\mathbf{0}|\mathbf{r})\rangle - \langle e_{pq}\rangle\}p(\mathbf{r}|\mathbf{0}) - n\,P_{pqmn}(\mathbf{r})\langle S_{mn}\rangle]\,dV + O(\phi^2) \quad (6.14)$$

and
$$\langle\nabla^2 e^E_{pq}(\mathbf{0})\rangle = \int_{r=2a}^{\infty} \{\nabla^2 e^E(\mathbf{0}|\mathbf{r})_{pq}\rangle\, p(\mathbf{r}|\mathbf{0})\,dV(\mathbf{r}) + O(\phi^2). \quad (6.15)$$

An expression for the average dipole strength of an elastic sphere in a statistically homogeneous suspension is obtained by substituting the expressions (6.14) and (6.15) for $\langle \mathbf{e}^E \rangle$ and $\langle \nabla^2 \mathbf{e}^E \rangle$ in (6.13), which gives

$$\begin{aligned}\langle S_{pq} \rangle = S^0_{pq} + \phi \left\{ \delta_{pq} \langle S_{mm} \rangle \left[\eta_1 \left(\frac{\alpha K}{\mu} - \tfrac{2}{3}\beta \right) + \frac{3\alpha K}{\mu} \eta_2 \right] + 2\beta\eta_1 \langle S_{pq} \rangle \right\} \\ + \int_{r=2a}^{\infty} \{ [\langle S_{pq}(\mathbf{0}|\mathbf{r}) \rangle - S^0_{pq}] p(\mathbf{r}|\mathbf{0}) - \phi [(\alpha K - \tfrac{2}{3}\beta\mu)\, \delta_{pq} P_{kkmn}(\mathbf{r}) \langle S_{mn} \rangle \\ + 2\beta\mu\, P_{pqmn}(\mathbf{r}) \langle S_{mn} \rangle] \}\, dV, \end{aligned} \tag{6.16}$$

where
$$S^0_{pq} = \tfrac{4}{3}\pi a^3 [(\alpha K - \tfrac{2}{3}\beta\mu) \langle e_{mm} \rangle \delta_{pq} + 2\beta\mu \langle e_{pq} \rangle] \tag{6.17}$$

is the average dipole strength in the limit as $\phi \to 0$.

As in § 5, we can show that

$$\langle \mathbf{S}(\mathbf{0}|\mathbf{r}) \rangle = \mathbf{S}(\mathbf{0}|\mathbf{r}) + O(\phi), \tag{6.18}$$

where $\mathbf{S}(\mathbf{0}|\mathbf{r})$ denotes the dipole strength of one of a pair of spheres separated by $\mathbf{r}$ and alone in an infinite matrix with an ambient uniform strain $\langle \mathbf{e} \rangle$. On substituting (6.18) in (6.16), and replacing $\langle \mathbf{S} \rangle$ by $\mathbf{S}^0$ in the integrand, we get

$$\begin{aligned}\langle S_{pq} \rangle = S^0_{pq} + \phi \left\{ \delta_{pq} S^0_{mm} \left[\eta_1 \left(\frac{\alpha K}{\mu} - \tfrac{2}{3}\beta \right) + \frac{3\alpha K}{\mu} \eta_2 \right] + 2\beta\eta_1 S^0_{pq} \right\} \\ + \int_{r=2a}^{\infty} \{ [S_{pq}(\mathbf{0}|\mathbf{r}) - S^0_{pq}] p(\mathbf{r}|\mathbf{0}) - \phi [(\alpha K - \tfrac{2}{3}\beta\mu)\, \delta_{pq} P_{kkmn}(\mathbf{r}) S^0_{mn} \\ + 2\beta\mu\, P_{pqmn}(\mathbf{r}) S^0_{mn}] \}\, dV + O(\phi^2), \end{aligned} \tag{6.19}$$

thus $\langle \mathbf{S} \rangle$ may be calculated with the aid of solutions to the two-sphere problem. In order to obtain expressions for the effective elastic moduli from (6.19) we must consider the general form of $\mathbf{S}(\mathbf{0}|\mathbf{r})$.

The elasticity equations and the boundary conditions for the two-sphere problem are linear, hence $\mathbf{S}(\mathbf{0}|\mathbf{r})$ is linear in $\langle \mathbf{e} \rangle$. From symmetry considerations it can be shown that the deviatoric part of $\mathbf{S}(\mathbf{0}|\mathbf{r})$ is given by an expression of the form

$$\begin{aligned}\mathbf{S}^D(\mathbf{0}|\mathbf{r}) / \tfrac{8}{3}\pi a^3 \beta\mu = (1+A) \langle \mathbf{e}^D \rangle + B[\hat{\mathbf{r}}(\hat{\mathbf{r}} \,.\, \langle \mathbf{e}^D \rangle) + (\hat{\mathbf{r}} \,.\, \langle \mathbf{e}^D \rangle)\, \hat{\mathbf{r}} - \tfrac{2}{3} \hat{\mathbf{r}} \,.\, \langle \mathbf{e}^D \rangle \,.\, \hat{\mathbf{r}} \mathbf{I}] \\ + C \hat{\mathbf{r}} \,.\, \langle \mathbf{e}^D \rangle \,.\, \hat{\mathbf{r}} + D \| \langle \mathbf{e} \rangle \| \,(\hat{\mathbf{r}}\hat{\mathbf{r}} - \tfrac{1}{3}\mathbf{I}), \end{aligned} \tag{6.20}$$

and similarly the isotropic part of $\mathbf{S}(\mathbf{0}|\mathbf{r})$ may be written in the form

$$\| \mathbf{S}(\mathbf{0}|\mathbf{r}) \| / 4\pi a^3 \alpha K = (1+E) \| \langle \mathbf{e} \rangle \| + F(\hat{\mathbf{r}} \,.\, \langle \mathbf{e}^D \rangle \,.\, \hat{\mathbf{r}}), \tag{6.21}$$

where $\hat{\mathbf{r}} = \mathbf{r}/r$ and $\langle \mathbf{e}^D \rangle = \langle \mathbf{e} \rangle - \tfrac{1}{3} \| \langle \mathbf{e} \rangle \| \mathbf{I}$. The quantities A, B, C, D, E and F depend only on the scalar variables r/a, K/μ, μ_p/μ and K_p/μ. In the limit as $r \to \infty$, $\mathbf{S}(\mathbf{0}|\mathbf{r}) \to \mathbf{S}^0$, and by comparing (6.20) and (6.21) with the expression (6.17) for $\mathbf{S}^0$ it can be seen that each of the quantities A, B, C, D, E and F approaches zero as r becomes large. The expressions (6.20) and (6.21) are equivalent to the expression (19) for $\mathbf{S}(\mathbf{0}|\mathbf{r})$ given by Chen & Acrivos (1978); the scalar functions F, G, K, L, M and N which appear there may be related to the quantities A, B, C, D, E and F used here by simply equating the coefficients of like terms in the expressions for $\mathbf{S}(\mathbf{0}|\mathbf{r})$. For the case of rigid particles and a rigid matrix, the quantities A, B and C are equal to the functions K, L and M defined by Batchelor & Green (1972a) for the analogous problem of two spheres immersed in a linear flow field.

If the pair-probability function has the isotropic form

$$p(\mathbf{r}|\mathbf{0}) = nq(r) \tag{6.22}$$

we may simplify the integral in the expression (6.19) for $\langle S_{pq} \rangle$ by first performing the integration with respect to the direction of $\mathbf{r}$. (We are free to use any order of integration since the integral in (6.19) converges.) In this case the renormalizing term vanishes, and we get, using (6.20) and (6.21),

$$\int_{r\,\text{const}} [\mathbf{S}(\mathbf{0}|\mathbf{r}) - \mathbf{S}^0]\, dA = \frac{(4\pi)^2 a^3 r^2}{3} [2\beta\mu G(r) \langle \mathbf{e}^D \rangle + \alpha K E(r) \| \langle \mathbf{e} \rangle \| \mathbf{I}], \tag{6.23}$$

where

$$G(r) = A(r) + \tfrac{2}{3} B(r) + \tfrac{2}{15} C(r). \tag{6.24}$$

On substituting (6.22) and (6.23) in the expression (6.19) for the average dipole strength, we find

$$\langle \mathbf{S} \rangle = \mathbf{S}^0 + \phi \left\{ \|\mathbf{S}^0\| \mathbf{I} \left[\eta_1 \left(\frac{\alpha K}{\mu} - \tfrac{2}{3}\beta \right) + \frac{3\alpha K}{\mu} \eta_2 \right] + 2\beta\eta_1 \mathbf{S}^0 \right\}$$
$$+ \langle \mathbf{e}^D \rangle\, 8\pi\phi\beta\mu \int_{r=2a}^{\infty} G(r)\, q(r)\, r^2 dr + \|\langle \mathbf{e} \rangle\| \mathbf{I}\, 4\pi\phi\alpha K \int_{r=2a}^{\infty} E(r)\, q(r)\, r^2 dr.$$

On substituting this expression for $\langle \mathbf{S} \rangle$ in the formula (6.1) for the average stress in the suspension, we obtain an expression of the form

$$\langle \boldsymbol{\sigma} \rangle = K^* \|\langle \mathbf{e} \rangle\| \mathbf{I} + 2\mu^* \langle \mathbf{e}^D \rangle, \tag{6.25}$$

where the effective modulus of compression K^* and the effective shear modulus μ^* are given by

$$\frac{K^*}{K} = 1 + \alpha\phi + \phi^2 \left\{ \frac{\alpha^2(1-2\nu)}{2(1-\nu)} + \frac{3\alpha}{a^3} \int_{r=2a}^{\infty} E(r)\, q(r)\, r^2 dr \right\} \tag{6.26}$$

and

$$\frac{\mu^*}{\mu} = 1 + \beta\phi + \phi^2 \left\{ \frac{2\beta^2(4-5\nu)}{15(1-\nu)} + \frac{3\beta}{a^3} \int_{r=2a}^{\infty} G(r)\, q(r)\, r^2 dr \right\}. \tag{6.27}$$

Willis & Acton's (1976) expressions (5.20) and (5.21) for the effective moduli have the same form as the expressions derived here, although in their case the functions E and G are replaced in the integrals by far-field forms correct to $O(a^7/r^7)$.

For the case of rigid particles in an incompressible matrix our expression for the shear modulus reduces to

$$\frac{\mu^*}{\mu} = 1 + \tfrac{5}{2}\phi + \phi^2 \left\{ \tfrac{5}{2} + \frac{15}{2a^3} \int_{r=2a}^{\infty} G(r)\, q(r)\, r^2 dr \right\}, \tag{6.28}$$

which is identical to Batchelor & Green's (1972*b*) expression (5.6) for the effective viscosity of a suspension of rigid spherical particles in a pure straining motion, which was obtained using the renormalization procedure.

Our expressions for the effective moduli are also in agreement with Chen & Acrivos's (1978) formulae (equations (24), (25) and (26) of that paper).† In attempting to

† For the case of a uniform pair distribution, viz. $q(r) = n$ for $r > 2a$, Chen & Acrivos have evaluated the coefficient of ϕ^2 in the expression for K^* over a range of μ_p/μ values; their results for the shear modulus μ^* are less complete for they have considered only the cases of rigid particles and spherical cavities. For elastic particles, Willis & Acton's (1976) approximate expression for μ^* may be used.

calculate the effective modulus of compression by the renormalization technique Chen & Acrivos encountered some difficulties in selecting the renormalizing quantity, for in this problem it is possible to obtain three different convergent expressions for K^*. These difficulties arise from the fact that the authors were concerned with the case of a pure compression, viz.

$$\langle \mathbf{e} \rangle = -e\,\mathbf{I}.$$

In order to show how it is possible to obtain more than one expression for K^*, we consider the renormalizing term

$$\phi[(\alpha K - \tfrac{2}{3}\beta\mu)\,\delta_{pq} P_{kkmn}(\mathbf{r})\,S^0_{mn} + 2\beta\mu P_{pqmn}(\mathbf{r})\,S^0_{mn}], \tag{6.29}$$

which appears in the integrand in the expression (6.19) for $\langle S_{pq}\rangle$. In the case of pure compression, the expression (6.17) for the dipole strength of an isolated sphere reduces to

$$\mathbf{S}^0 = -4\pi a^3 \alpha K e\,\mathbf{I}$$

and hence the renormalizing term (6.29) becomes

$$-4\pi a^3 \phi\alpha K e[(\alpha K - \tfrac{2}{3}\beta\mu)\,\delta_{pq} P_{kkmm} + 2\beta\mu P_{pqmm}].$$

Since only the isotropic part of $\langle \mathbf{S}\rangle$ is required in order to calculate K^* (see the expressions (6.1) and (6.25) for $\langle \boldsymbol{\sigma}\rangle$), we take the trace of the above expression, and obtain

$$-12\pi a^3 \phi \alpha^2 K^2 e P_{kkmm}.$$

This term is identically zero, since $P_{kkmm} \equiv 0$. Thus the renormalizing term in the expression for $\|\langle \mathbf{S}\rangle\|$ vanishes in the case of pure compression, and from (6.19) we get

$$\|\mathbf{S}\| = \|\mathbf{S}^0\|\,(1+\phi\zeta) + \int_{r=2a}^{\infty} \{\|\mathbf{S}(\mathbf{0}\,|\,\mathbf{r})\| - \|\mathbf{S}^0\|\}\,p(\mathbf{r}\,|\,\mathbf{0})\,dV, \tag{6.30}$$

where
$$\zeta = 2\beta\eta_1 + 3\left[\left(\frac{\alpha K}{\mu} - \tfrac{2}{3}\beta\right)\eta_1 + \frac{3\alpha K}{\mu}\,\eta_2\right].$$

Although the renormalizing factor has vanished, the above integral converges, since

$$S_{qq}(\mathbf{0}\,|\,\mathbf{r}) - S^0_{qq} = \text{constant} \times P_{qqpp}(\mathbf{r}) + O(r^{-4}) = O(r^{-4}) \quad \text{as} \quad r \to \infty.$$

If instead of using the expression (6.19) for $\langle \mathbf{S}\rangle$ we use the incorrect equation

$$\langle \mathbf{S}\rangle = \mathbf{S}^0 + \int (\mathbf{S}(\mathbf{0}\,|\,\mathbf{r}) - \mathbf{S}^0)\,p(\mathbf{r}\,|\,\mathbf{0})\,dV,$$

based on the assumption that only pair interactions are important [cf. (2.1)], then on taking the trace of this expression, we get

$$\|\langle \mathbf{S}\rangle\| = \|\mathbf{S}^0\| + \int (\|\mathbf{S}(\mathbf{0}\,|\,\mathbf{r})\| - \|\mathbf{S}^0\|)\,p(\mathbf{r}\,|\,\mathbf{0})\,dV. \tag{6.31}$$

Although this expression converges, it can be seen on comparing it with the result (6.30) (which takes into account the contribution from the macroscopic boundary) that the term $\phi\zeta\|\mathbf{S}^0\|$ is missing.

In order to select the correct renormalizing quantity, Chen & Acrivos use Jeffrey's (1974) group expansion for the dipole strength to show that only one renormalizing quantity gives a convergent expression for the three-sphere term (of order ϕ^3). The method presented here is free of such complications, for the renormalizing quantity in the expression for $\langle \mathbf{S} \rangle$ arises naturally from a consideration of the macroscopic-boundary term in the integral expression (6.4) for the displacement.

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Appendix. The derivation of the Faxén-type formula (3.14) for the dipole strength

The required formula for the dipole strength of a sphere placed in an ambient field T^E is obtained with the aid of Green's Second Identity (Protter & Weinberger 1967) for the case of a single particle in an infinite matrix, viz.

$$\oint_A (T_1 \mathbf{F}_2 - T_2 \mathbf{F}_1) \cdot \hat{\mathbf{n}} dA = \oint_{A_p} (T_2 \mathbf{F}_1 - T_1 \mathbf{F}_2) \cdot \hat{\mathbf{n}} dA, \tag{A 1}$$

where T_1 and T_2 are two temperature fields and $\mathbf{F}_1$ and $\mathbf{F}_2$ the corresponding flux densities. Here A_p denotes the surface of the particle, and A denotes any closed surface enclosing A_p. For our purposes it proves convenient to choose A as a large sphere, centred on the particle.

Let T_I denote the temperature field for a sphere placed in the linear ambient field $\mathbf{G} \cdot \mathbf{x}$, and let T_{II} and $\mathbf{S}_{\mathrm{II}}$ denote the temperature field and dipole strength of a sphere placed in the ambient field T^E. Our aim is to derive an expression for $\mathbf{S}_{\mathrm{II}}$. On substituting

$$T_1(\mathbf{x}) = T_{\mathrm{I}}(\mathbf{x}) - \mathbf{G} \cdot \mathbf{x}$$

and

$$T_2(\mathbf{x}) = T_{\mathrm{II}}(\mathbf{x}) - T^E(\mathbf{x})$$

in the Green's identity (A 1), we find that the integral over the large sphere A vanishes as the sphere becomes infinite (since T_1 and T_2 are $O(r^{-2})$) and hence the identity (A 1) yields

$$\oint_{A_p} (T_2 \mathbf{F}_1 - T_1 \mathbf{F}_2) \cdot \hat{\mathbf{n}} dA = 0. \tag{A 2}$$

Applying the divergence theorem to this surface integral, we get

$$\oint_{V_p} (\nabla T_2 \cdot \mathbf{F}_1 - \nabla T_1 \cdot \mathbf{F}_2) dV = 0,$$

and on using the fact that

$$\mathbf{F}_1 = -\alpha k \nabla T_{\mathrm{I}} + k\mathbf{G}, \quad \mathbf{F}_2 = -\alpha k \nabla T_{\mathrm{II}} + k \nabla T^E$$

within the particle, we obtain

$$\int_{V_p} \boldsymbol{\tau}_{\mathrm{II}} \cdot \mathbf{G} dV - \int_{V_p} \boldsymbol{\tau}_{\mathrm{I}} \cdot \nabla T^E dV = 0,$$

where $\boldsymbol{\tau} = (1-\alpha) k \nabla T$. Hence the dipole strength $\mathbf{S}_{\mathrm{II}}$ is given by

$$\mathbf{S}_{\mathrm{II}} \cdot \mathbf{G} = \int_{V_p} \boldsymbol{\tau}_{\mathrm{I}} \cdot \nabla T^E dV. \tag{A 3}$$

From the expression for T_{I} (given in Jeffrey's 1973 paper) we find

$$\boldsymbol{\tau}_{\mathrm{I}}(=(1-\alpha)\,k\nabla T_{\mathrm{I}}) = -\frac{3k(\alpha-1)}{(\alpha+2)}\,\mathbf{G},$$

at points in the particle. On substituting this result into (A 3) and using the fact that

$$\int_{V_p} \nabla T^E dV = \tfrac{4}{3}\pi a^3 \nabla T^E(\mathbf{x}_0),$$

we obtain the desired result

$$\mathbf{S}_{\mathrm{II}} = -4\pi a^3 \frac{(\alpha-1)}{(\alpha+2)}\,k\nabla T^E(\mathbf{x}_0), \tag{A 4}$$

where $\mathbf{x}_0$ denotes the centre of the sphere.

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