Effective viscosity of a periodic suspension

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The effective viscosity of a suspension is defined to be the four-tensor that relates the average deviatoric stress to the average rate of strain. We determine the effective viscosity of an array of spheres centred on the points of a periodic lattice in an incompressible Newtonian fluid. The formulation involves the traction exerted on a single sphere by the fluid, and an integral equation for this traction is derived. For lattices with cubic symmetry the effective viscosity tensor involves just two parameters. They are computed numerically for simple, body-centred and face-centred cubic lattices of spheres with solute concentrations up to 90 % of the close-packing concentration. Asymptotic results for high concentrations are obtained for arbitrary lattice geometries, and found to be in good agreement with the numerical results for cubic lattices. The low-concentration asymptotic expansions of Zuzovsky also agree well with the numerical results.

1. Introduction

We shall calculate the effective viscosity of a suspension of rigid spheres in an incompressible viscous fluid, with the spheres centred at the points of a periodic lattice. For such a special configuration we can obtain results for all values of the volume concentration c, from zero up to the close-packing value c_{max}.

We begin by proving that the average deviatoric stress in the suspension is linearly related to the average rate of strain. The effective viscosity μ^*_{ijkl} is defined to be the four-tensor in this relation. It is determined by the traction of the fluid on a single sphere, and an integral equation for this traction is derived. Its kernel is a periodic Green function, which can be represented as a sum over the reciprocal lattice. The integral equation also involves the angular velocity of the sphere, for which a second equation is obtained.

For a cubic lattice (simple, body-centred or face-centred) the symmetries of the kernel greatly simplify the integral equation. It can be shown that the angular velocity of each sphere is just one half the curl of the average fluid velocity, and the effective viscosity tensor has the form

$$\mu^*_{ijkl} = \mu(1+\beta)\tfrac{1}{2}(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk} - \tfrac{2}{3}\delta_{ij}\delta_{kl}) + \mu(\alpha-\beta)(\delta_{ijkl} - \tfrac{1}{3}\delta_{ij}\delta_{kl}). \tag{1}$$

Here μ is the viscosity of the fluid, δ_{ijkl} is unity if all the subscripts are equal and zero otherwise, and α and β are functions of the concentration and the lattice geometry which can be expressed as integrals of the traction over a single sphere. We use a Galerkin method to solve for the traction, following the procedure used by Zick & Homsy (1982) and Zick (1983). The results for α and β are shown in figures 1–6.

For low concentrations Zuzovsky (1976) and Zuzovsky, Adler & Brenner (1983)

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have obtained asymptotic formulas for α and β to $O(c^{\frac{10}{3}})$. Our numerical results corroborate their formulas and determine their range of validity. Frankel & Acrivos (1967) obtained the leading-order term in the asymptotic expansion for α at high concentrations, but only for a simple cubic lattice. Zuzovsky (1976) corrected their result and also gave the leading-order term for β. We have obtained additional terms in these expansions for a simple cubic lattice and have also calculated the corresponding results for body-centred and face-centred cubic lattices. Our analysis is in fact valid for arbitrary lattice geometries, and a general high-concentration asymptotic formula for μ^{*}_{ijkl} is given. The asymptotic formulas for the three cubic lattices are in good agreement with our numerical results, except for β in the face-centred case, where numerical errors at high concentrations are severe. These results show that the contention by Kapral & Bedeaux (1978) of a singularity at less than close-packing concentrations is incorrect.

A cubic lattice of spheres in a uniform shear flow will be distorted by the flow. The spheres will remain on a lattice, but the lattice will not remain cubic. Therefore our solution for a cubic lattice will apply only at one instant. For unidirectional flows, the lattice will become cubic again after a finite time, and this will be repeated periodically. Then the effective viscosity will be a periodic function of time. In any case, it can be computed by applying the present methods to each lattice through which the configuration passes.

2. Formulation

We consider a set of solid, neutrally buoyant spheres of radius b centred on the points of an infinite three-dimensional lattice $r^{\alpha} = \alpha_1 a_1 + \alpha_2 a_2 + \alpha_3 a_3$, $\alpha \in \mathbb{Z}^3$. The basis vectors a_i determine a unit cell having volume $\tau_0 = |a_1 \cdot (a_2 \times a_3)|$, and the volume concentration of spheres is $c = 4\pi b^3 / 3\tau_0$. We assume that the spheres are immersed in a homogeneous, isotropic Newtonian fluid undergoing slow flow. Then within the region E containing the fluid, the pressure p, viscosity μ, velocity u_i and stress tensor σ_{ij} satisfy the Stokes equations:

$$\frac{\partial u_i}{\partial x_i} = 0 \quad (x_i \in E), \tag{2}$$

$$\sigma_{ij} = -p\delta_{ij} + \mu\left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i}\right) \quad (x_i \in E), \tag{3}$$

$$\frac{\partial \sigma_{ij}}{\partial x_i} = 0 \quad (x_i \in E). \tag{4}$$

We introduce an overall constant shear tensor γ_{ij}, with $\gamma_{ii} = 0$, by requiring that the fluid velocity u_i be the sum of a linear part $\gamma_{ij} x_j$ and a part that is lattice-periodic. More precisely, we impose the condition

$$u_i(x + r^{\alpha}) = u_i(x) + \gamma_{ij} r^{\alpha}_j \quad (x \in E, \quad \alpha \in \mathbb{Z}^3). \tag{5}$$

We impose no-slip boundary conditions on the surfaces of the spheres. The spheres may rotate with some angular velocity ω_i, which, by periodicity, must be the same for each sphere. Let $\mathcal{S}$ denote the sphere centred at the origin and n_l the outward normal to its surface. We can assume that $\mathcal{S}$ is not translating, hence the no-slip boundary condition can be expressed as

$$u_i = \epsilon_{ijk}\omega_j x_k \quad (x_k \in \partial\mathcal{S}). \tag{6}$$

Finally, the assumption of slow flow leads to the vanishing of the torque on each sphere:

$$\int_{\partial \mathscr{S}} \epsilon_{ijk} x_j \sigma_{kl} n_l \, \mathrm{d}A = 0. \tag{7}$$

The force on each sphere is also zero by symmetry.

3. Definition and existence of the effective viscosity

Let S_{ij} and $\partial U_i / \partial x_j$ denote the volume averages of the stress and velocity gradient respectively, over some region $\mathscr{V}$. As shown by Batchelor (1970), they are related by

$$S_{ij} = -\delta_{ij} \frac{1}{V} \int_{\mathscr{V} \cap E} p \, \mathrm{d}V + \mu \left(\frac{\partial U_i}{\partial x_j} + \frac{\partial U_j}{\partial x_i} \right) + \frac{1}{V} \Sigma \int [\sigma_{ik} x_j n_k - \mu (u_i n_j + u_j n_i)] \, \mathrm{d}A. \tag{8}$$

Here V is the volume of the region $\mathscr{V}$, the sum is over the particles in $\mathscr{V}$, and the integral in the summand is over the surface of the summand particle. For the periodic suspension described in §2, with $\mathscr{V}$ a unit cell of the lattice containing $\mathscr{S}$, (8) simplifies to

$$S_{ij} = -\delta_{ij} \frac{1}{V} \int_{\mathscr{V} \cap E} p \, \mathrm{d}V + \mu (\gamma_{ij} + \gamma_{ji}) + \tau_0^{-1} \int_{\partial \mathscr{S}} \sigma_{ik} x_j n_k \, \mathrm{d}A, \tag{9}$$

and the bulk deviatoric stress D_{ij} is therefore given by

$$D_{ij} = \mu (\gamma_{ij} + \gamma_{ji}) + \tau_0^{-1} \int_{\partial \mathscr{S}} (\sigma_{ik} x_j - \tfrac{1}{3} \delta_{ij} \sigma_{mk} x_m) n_k \, \mathrm{d}A. \tag{10}$$

From the linearity of the governing equations and the boundary conditions, the integral in (10) must be linear in the average velocity gradient γ_{ij}:

$$\int_{\partial \mathscr{S}} \sigma_{ik} x_j n_k \, \mathrm{d}A = M_{ijkl} \gamma_{kl}. \tag{11}$$

Since no external couple is acting on the particles, M_{ijkl} must be symmetric in i and j, and for definiteness we may assume that $M_{ijkk} = 0$, since only its contraction with γ_{kl} is significant. By using (11) we can rewrite (10) in the form

$$D_{ij} = 2\mu_{ijkl}^* \gamma_{kl}, \tag{12}$$

where

$$\mu_{ijkl}^* = \tfrac{1}{2}\mu(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk} - \tfrac{2}{3}\delta_{ij}\delta_{kl}) + \tfrac{1}{2}\tau_0^{-1}(M_{ijkl} - \tfrac{1}{3}\delta_{ij} M_{mmkl}). \tag{13}$$

The tensor μ_{ijkl}^* is symmetric and traceless in its first pair of indices and traceless in its last pair of indices. In Appendix A we show that

$$\mu_{ijkl}^* = \mu_{klij}^*, \tag{14}$$

and so μ_{ijkl}^* is symmetric in k and l as well. Thus the average deviatoric stress D_{ij} is linearly related to the symmetric part of γ_{ij}, which is the average rate-of-strain tensor. We are therefore justified in calling μ_{ijkl}^* the effective viscosity of the suspension.

The formulation becomes somewhat easier to use if we multiply both sides of (13) by $2\gamma_{ij}\gamma_{kl}$ and use (11). Then

$$2\mu_{ijkl}^* \gamma_{ij}\gamma_{kl} = \mu\gamma_{ij}(\gamma_{ij} + \gamma_{ji}) + \tau_0^{-1}\gamma_{ij} \int_{\partial \mathscr{S}} x_j \sigma_{ik} n_k \, \mathrm{d}A. \tag{15}$$

Now the effective viscosity μ_{ijkl}^* can be defined as the unique four-tensor that satisfies (15) for all γ_{ij} with $\gamma_{ii} = 0$, and which also satisfies the symmetry conditions

$$\mu_{ijkl}^* = \mu_{jikl}^*, \quad \mu_{iikl}^* = 0, \quad \mu_{ijkl}^* = \mu_{klij}^*. \tag{16}$$

It is interesting to note that (15) is precisely the formula one would get using the energy dissipation method. The right-hand side of (15) is the actual rate of energy dissipation within a lattice cell, while the left-hand side is the rate of energy dissipation that would occur in a homogeneous fluid with viscosity four-tensor μ^*_{ijkl} under the same overall shear γ_{ij}. However, the symmetry condition (14) is necessary to define μ^*_{ijkl} uniquely from this equation, and a proof of this identity apparently requires recourse to the averaged-equation formulation.

4. Integral-equation formulation

The basic unknown quantity in equation (15) for the effective viscosity is the traction f_i exerted by the fluid on the surface of the sphere $\mathscr{S}$. It is given by

$$f_i = \sigma_{ij} n_j \quad (x_i \in \partial \mathscr{S}). \tag{17}$$

In this section we use Green's method and (2)–(7) to derive an integral equation of the first kind for f_i. The derivation follows that of Zick & Homsy (1982).

The fundamental singular solution v_{ik}, τ_{ijk}, q_k, corresponding to unit forces in the k-direction applied to all positions $y + r^\alpha$, satisfies

$$\frac{\partial v_{ik}(x, y)}{\partial x_i} = 0, \tag{18}$$

$$\tau_{ijk}(x, y) = -q_k(x, y) \delta_{ij} + \mu \left(\frac{\partial v_{ik}}{\partial x_j} + \frac{\partial v_{jk}}{\partial x_i} \right), \tag{19}$$

$$\frac{\partial \tau_{ijk}}{\partial x_j} = -\delta_{ik} \sum_\alpha \delta(x - y - r^\alpha). \tag{20}$$

The solution for v_{ik} is (Hasimoto 1959)

$$v_{ik}(x, y) = \frac{1}{4\pi^2 \mu \tau_0} \sum_\alpha{}' \left(\frac{\delta_{ik}}{|S^\alpha|^2} - \frac{S_i^\alpha S_k^\alpha}{|S^\alpha|^4} \right) e^{2\pi i S^\alpha \cdot (x - y)}. \tag{21}$$

Here

$$S^\alpha = \alpha_1 b_1 + \alpha_2 b_2 + \alpha_3 b_3 \quad (\alpha \in \mathbb{Z}^3) \tag{22}$$

is a vector in the reciprocal lattice, which has basis

$$b_1 = \tau_0^{-1}(a_2 \times a_3), \quad b_2 = \tau_0^{-1}(a_3 \times a_1), \quad b_3 = \tau_0^{-1}(a_1 \times a_2). \tag{23}$$

The prime on the sum in (21) means that the index $\alpha = 0$ is to be omitted.

Let T be a unit cell containing the sphere at the origin, and let $E_0 = T - \mathscr{S}$ be the fluid region within T. For $y \in E_0$, we multiply (20) by $u_i(x)$ and integrate over E_0 to get

$$\int_{E_0} u_i(x) \frac{\partial \tau_{ijk}(x, y)}{\partial x_j} \mathrm{d}x = -u_k(y) \quad (y \in E_0). \tag{24}$$

From (2), (3), (18) and (19) it may be shown that

$$\frac{\partial u_i}{\partial x_j} \tau_{ijk} = \frac{\partial v_{ik}}{\partial x_j} \sigma_{ij}. \tag{25}$$

Then, by using (4), (25) and the divergence theorem, we can write (24) in the form†

$$\int_{\partial T - \partial \mathscr{S}} [u_i \tau_{ijk} - v_{ik} \sigma_{ij}] n_j \, \mathrm{d}A(x) = -u_k(y) \quad (y \in E_0). \tag{26}$$

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† $\int_{\partial T - \partial \mathscr{S}}$ should be interpreted as $\int_{\partial T} - \int_{\partial \mathscr{S}}$: the normal n_i points out of both the sphere $\mathscr{S}$ and the unit cell T.

We now proceed to simplify this equation. If f_i is any lattice-periodic function then the integral of $f_i n_i$ over the surface of a unit cell is zero, since for each section of the cell boundary there is a congruent, periodically translated section on which the normal points in the opposite direction. The tensors τ_{ijk}, ν_{ik} and σ_{ij} are all periodic, and the non-periodic part of u_i is simply $\gamma_{ij}x_j$. Thus the integral over ∂T in (26) becomes

$$\int_{\partial T} [u_i \tau_{ijk} - \nu_{ik}\sigma_{ij}] n_j \, dA(\boldsymbol{x}) = \int_{\partial T} \gamma_{il} x_l \tau_{ijk} n_j \, dA(\boldsymbol{x}) \quad (\boldsymbol{y} \in E_0). \tag{27}$$

By using (19), (20), the divergence theorem, the periodicity of ν_{ik} and the fact that $\gamma_{ii} = 0$, we can evaluate this latter integral. The result is

$$\int_{\partial T} [u_i \tau_{ijk} - \nu_{ik}\sigma_{ij}] n_j \, dA(\boldsymbol{x}) = -\gamma_{kl} y_l \quad (\boldsymbol{y} \in E_0). \tag{28}$$

The integral over the surface of the sphere in (26) can also be simplified. Using (6), (20) and the divergence theorem, we can write the first part of it as

$$\int_{\partial \mathscr{S}} u_i \tau_{ijk} n_j \, dA(\boldsymbol{x}) = \int_{\mathscr{S}} [\epsilon_{ilj}\omega_l \tau_{ijk} - \epsilon_{klm}\omega_l x_m \delta(\boldsymbol{x}-\boldsymbol{y})] \, dV \quad (\boldsymbol{y} \in E_0). \tag{29}$$

But $\epsilon_{ilj}\tau_{ijk} = 0$ since τ_{ijk} is symmetric in the indices i and j, and $\delta(\boldsymbol{x}-\boldsymbol{y}) = 0$ because $\boldsymbol{y} \notin \mathscr{S}$. Thus this integral is zero. This fact and (28) enable us to write (26) as

$$-\gamma_{kl} y_l + \int_{\partial \mathscr{S}} \nu_{ik}\sigma_{ij} n_j \, dA(\boldsymbol{x}) = -u_k(\boldsymbol{y}) \quad (\boldsymbol{y} \in E_0). \tag{30}$$

We now let $\boldsymbol{y}$ tend to the boundary of the sphere and use (6) to evaluate the fluid velocity there. Then (30) becomes

$$\int_{\partial \mathscr{S}} \nu_{ik}\sigma_{ij} n_j \, dA(\boldsymbol{x}) = \gamma_{kj} y_j - \epsilon_{kij}\omega_i y_j \quad (\boldsymbol{y} \in \partial \mathscr{S}). \tag{31}$$

This is an integral equation for the traction $f_i = \sigma_{ij} n_j$, but it also involves the angular velocity ω_i. The no-torque condition (7) provides the needed supplementary condition, and we can solve (7) and (31) together for the unknowns f_i and ω_i. These equations are

$$\int_{\partial \mathscr{S}} \nu_{ik}(\boldsymbol{x}, \boldsymbol{y}) f_i(\boldsymbol{x}) \, dA(\boldsymbol{x}) = \gamma_{kj} y_j - \epsilon_{kij}\omega_i y_j \quad (\boldsymbol{y} \in \partial \mathscr{S}). \tag{32}$$

$$\int_{\partial \mathscr{S}} \epsilon_{ijk} x_j f_i(\boldsymbol{x}) \, dA(\boldsymbol{x}) = 0. \tag{33}$$

The homogeneous form of these equations has the solution $f_i(\boldsymbol{x}) = k x_i$ for any constant k. This solution is everywhere normal to the surface of the sphere and represents a uniform pressure. This presents no difficulties, however, for since $\gamma_{ii} = 0$ the solvability condition for (32) is satisfied, and it may also be verified that any uniform pressure in the traction cancels out in (15) for the effective viscosity.

5. Symmetry relations and cubic lattices

For simple, body-centred or face-centred cubic lattices a considerable simplification of the problem is possible. Their symmetries impart a structure to the equations which allow us to determine the angular velocity ω_i explicitly and to reduce to two the number of independent coefficients in the effective viscosity tensor. To show this we define f_i^{rs} to satisfy

$$\mu \tau_0 \int_{\partial \mathscr{S}} \nu_{ik} f_i^{rs} \, dA(\boldsymbol{x}) = \delta_{kr} y_s - \tfrac{1}{3}\delta_{rs} y_k \quad (\boldsymbol{y} \in \partial \mathscr{S}) \tag{34}$$

and
$$\int_{\partial \mathcal{S}} x_i f_i^{rs} \, \mathrm{d}A(x) = 0. \tag{35}$$

The second term on the right-hand side of (34) ensures that the solvability condition for the integral equation is satisfied, and condition (35) pins down the coefficient of the null solution x_i in f_i^{rs}. From (32) and (34) we have, up to a solution of the homogeneous integral equation, that

$$f_i = \mu \tau_0 (\gamma_{rs} - \epsilon_{rls} \omega_l) f_i^{rs}. \tag{36}$$

Consider for a moment an orthogonal linear transformation ξ_{ij} which maps the reciprocal lattice onto itself. Then from (21) we can show that

$$\nu_{ik}(\xi_{pq} x_q, \xi_{pq} y_q) = \xi_{lm} \xi_{kn} \nu_{mn}(x_p, y_p), \tag{37}$$

and by changing variables from x, y to $\xi x, \xi y$ in (34) and (35) we can subsequently show that
$$f_i^{rs}(\xi x) = \xi_{lm} \xi_{rp} \xi_{sq} f_{mn}^{pq}(x). \tag{38}$$

We now specialize to a cubic lattice. With no loss of generality we can assume that the coordinate axes and the principal axes of the lattice coincide. Then the reciprocal lattice is also cubic with its principal axes coincident with the coordinate axes. Thus it is invariant under the reflection transformation $\rho_{ij}^{(t)} = (-1)^{\delta_{ti}} \delta_{ij}$, which changes the sign of the tth coordinate of its argument, and the coordinate permutation transformation $\zeta_{ij}^\sigma = \delta_{\sigma(i)j}$, where σ is any permutation of the symbols $\{1, 2, 3\}$. For these particular orthogonal transformations (38) yields

$$f_i^{rs}(\rho^{(t)} x) = (-1)^{\delta_{ti} + \delta_{tr} + \delta_{ts}} f_i^{rs}(x) \tag{39}$$

and
$$f_i^{rs}(\zeta^\sigma x) = f_{\sigma(i)}^{\sigma(r)\sigma(s)}(x). \tag{40}$$

These identities can be used to simplify (32) and (33). We first solve (33) for ω_i. Substitution of (36) into (33) yields

$$(\gamma_{rs} - \epsilon_{rls} \omega_l) \int_{\partial \mathcal{S}} \epsilon_{ijk} x_j f_i^{rs} \, \mathrm{d}A = 0. \tag{41}$$

However, (39) implies that

$$\int_{\partial \mathcal{S}} \epsilon_{ijk} x_j f_i^{rs} \, \mathrm{d}A = (-1)^{1 + \delta_{tk} + \delta_{tr} + \delta_{ts}} \int_{\partial \mathcal{S}} \epsilon_{ijk} x_j f_i^{rs} \, \mathrm{d}A, \tag{42}$$

and since this must hold for any $t = 1, 2, 3$, the integral is non-zero only if the indices k, r, s are all different. By performing a coordinate permutation σ taking $k \to 1$, $r \to 2$ and $s \to 3$, and using (40), the integrals can be expressed as

$$\int_{\partial \mathcal{S}} \epsilon_{ijk} x_j f_i^{rs} \, \mathrm{d}A = \epsilon_{krs} \int_{\partial \mathcal{S}} \epsilon_{ij1} x_j f_i^{23} \, \mathrm{d}A. \tag{43}$$

Upon substitution of (43) into (41) we obtain

$$(\gamma_{rs} - \epsilon_{rls} \omega_l) \epsilon_{krs} = 0 \quad (k = 1, 2, 3). \tag{44}$$

The solution of this equation for ω_l is

$$\omega_l = -\tfrac{1}{2} \epsilon_{lij} \gamma_{ij}. \tag{45}$$

This is just one-half of the curl of the average velocity, which is the same angular velocity that would be exhibited by a single sphere in an infinite fluid subjected to the shear γ_{ij}. Thus in the case of cubic lattices, each sphere spins as if the other spheres were not there.

The structure of the effective viscosity tensor can now be elucidated. By substituting (45) into (36) we obtain

$$f_i = \tfrac{1}{2}\mu\tau_0(\gamma_{rs}+\gamma_{sr})f_i^{rs}, \tag{46}$$

which upon substitution into (15) yields

$$2\mu_{ijkl}^*\gamma_{ij}\gamma_{kl} = \mu\gamma_{ij}(\gamma_{ij}+\gamma_{ji})+\tfrac{1}{2}\mu\gamma_{ij}(\gamma_{rs}+\gamma_{sr})\int_{\partial\mathscr{S}}x_j f_i^{rs}\,\mathrm{d}A. \tag{47}$$

This can be simplified, for identities (39) and (40) readily yield

$$\int_{\partial\mathscr{S}}x_j f_i^{rs}\,\mathrm{d}A = (-1)^{\delta_{tj}+\delta_{ti}+\delta_{tr}+\delta_{ts}}\int_{\partial\mathscr{S}}x_j f_i^{rs}\,\mathrm{d}A \quad (t=1,2,3) \tag{48}$$

and

$$\int_{\partial\mathscr{S}}x_j f_i^{rs}\,\mathrm{d}A = \int_{\partial\mathscr{S}}x_{\sigma(j)}f_{\sigma(i)}^{\sigma(r)\,\sigma(s)}\,\mathrm{d}A. \tag{49}$$

Identity (48) implies that the integral is zero unless the indices j, i, r, s are equal in pairs, and (49) implies that many of the remaining non-zero integrals are equal. By using (48) and (49) we can write relation (47) in the form

$$2\mu_{ijkl}^*\gamma_{ij}\gamma_{kl} = \mu\gamma_{ij}(\gamma_{ij}+\gamma_{ji})+\tfrac{1}{2}\mu\gamma_{ij}(\gamma_{kl}+\gamma_{lk})\,[\delta_{ijkl}\Gamma_{11}^{11}+(\delta_{ij}\delta_{kl}-\delta_{ijkl})\,\Gamma_{22}^{11}$$
$$+(\delta_{ik}\delta_{jl}-\delta_{ijkl})\,\Gamma_{21}^{12}+(\delta_{il}\delta_{jk}-\delta_{ijkl})\,\Gamma_{12}^{12}], \tag{50}$$

where

$$\Gamma_{ji}^{rs} = \int_{\partial\mathscr{S}}x_j f_i^{rs}\,\mathrm{d}A. \tag{51}$$

The solution of (50) for μ_{ijkl}^* that satisfies the symmetry conditions (16) is

$$\mu_{ijkl}^* = \mu(1+\beta)\tfrac{1}{2}(\delta_{ik}\delta_{jl}+\delta_{il}\delta_{jk}-\tfrac{2}{3}\delta_{ij}\delta_{kl})+\mu(\alpha-\beta)\,(\delta_{ijkl}-\tfrac{1}{3}\delta_{ij}\delta_{kl}), \tag{52}$$

where

$$\alpha = \tfrac{1}{2}(\Gamma_{11}^{11}-\Gamma_{22}^{11}) = \frac{1}{2}\int_{\partial\mathscr{S}}(x_1 f_1^{11}-x_2 f_2^{11})\,\mathrm{d}A, \tag{53}$$

$$\beta = \tfrac{1}{2}(\Gamma_{12}^{12}+\Gamma_{21}^{12}) = \frac{1}{2}\int_{\partial\mathscr{S}}(x_1 f_2^{12}+x_2 f_1^{12})\,\mathrm{d}A. \tag{54}$$

Thus the effective viscosity tensor involves just two parameters when the lattice has cubic symmetry. They can be found from the solutions to two vector integral equations of the set (34), (35), namely those for f_i^{11} and f_i^{12}.

6. Numerical method for cubic lattices

For cubic lattices the two scalars in the effective-viscosity tensor μ_{ijkl}^* are determined by f_i^{11} and f_i^{12}, which satisfy

$$\mu\tau_0\int_{\partial\mathscr{S}}\nu_{ij}(x,y)f_i^{1\nu}(x)\,\mathrm{d}A(x) = \delta_{j1}y_\nu-\tfrac{1}{3}\delta_{1\nu}y_j \quad (y\in\partial\mathscr{S},\ \nu=1,2). \tag{55}$$

$$\int_{\delta\mathscr{S}}x_i f_i^{i\psi}(x)\,\mathrm{d}A(x) = 0. \tag{56}$$

These equations appear to require a numerical solution. The major difficulty in solving (55) is the complexity of the kernel ν_{ij}, given by (21), which precludes any method that requires numerous evaluations of ν_{ij}. We use a Galerkin method, with the basis functions chosen to be analytically integrable against ν_{ij}. This method yields excellent accuracy with relatively few lattice-sum computations required.

The Galerkin method may be outlined as follows. We write $f_i^{1\nu}$ in the form

$$f_i^{1\nu}(x) = \sum_{k,\,m}a_{ik}^{\nu m}\,\phi_k^{(m)}(x), \tag{57}$$

where $\{\phi_k^{(m)}\}$ is a set of basis functions on $\partial\mathcal{S}$ that span all possible solutions for $f_i^{1\nu}$. The basis functions are doubly indexed for later convenience. Substitution of (57) into (55) and integration against a second basis function yields a linear system of infinite rank for the unknown coefficients $a_{ik}^{\nu m}$:

$$\sum_{i=1}^{3}\sum_{k,\,m} A_{ijkl}^{mn}\, a_{ik}^{\nu m} = b_{jl}^{\nu n} \quad (j = 1,2,3;\; l = 1,2,\ldots;\; n = 1,2,\ldots), \tag{58}$$

where

$$A_{ijkl}^{mn} = \int_{\partial\mathcal{S}}\int_{\partial\mathcal{S}} \mu\tau_0\nu_{ij}(x,y)\,\phi_k^{(m)}(x)\,\phi_l^{(n)}(y)\,\mathrm{d}A(x)\,\mathrm{d}A(y) \tag{59}$$

and

$$b_{jl}^{\nu n} = (\delta_{j1}\delta_{\nu s} - \tfrac{1}{3}\delta_{\nu 1}\delta_{js})\int_{\partial\mathcal{S}} y_s\phi_l^{(n)}(y)\,\mathrm{d}A(y). \tag{60}$$

The expansion (57) is truncated and the resulting finite matrix problem is solved, yielding an approximation for the coefficients in (57).

As is readily verified from (21) and (55), $f_i^{1\nu}$ is an odd function of x. Thus only odd functions $\phi_k^{(m)}$ need be included in the set of basis functions. We choose for the basis functions the polynomials

$$\left.\begin{aligned}
\phi_k^{(1)}(x) &= x_1 x_2^{p_k} x_3^{q_k}, \\
\phi_k^{(2)}(x) &= x_1^{q_k} x_2 x_3^{p_k}, \\
\phi_k^{(3)}(x) &= x_1^{p_k} x_2^{q_k} x_3, \\
\phi_k^{(4)}(x) &= x_1 x_2^{p_k+1} x_3^{q_k+1},
\end{aligned}\right\} \tag{61}$$

where $\{(p_k, q_k), k = 1, 2, \ldots\}$ is the set of all distinct ordered pairs of non-negative even integers. These polynomials can be derived from the spherical-harmonic functions and form a complete basis for the set of odd functions defined on the surface of a sphere.

The above basis functions allow the integrals in (59) to be evaluated analytically. Since the basis functions are odd, substitution of (21) into (59) yields

$$A_{ijkl}^{mn} = \frac{1}{4\pi^2}\sum_{a}{}' \left(\frac{\delta_{ij}}{|S^\alpha|^2} - \frac{S_i^\alpha S_j^\alpha}{|S^\alpha|^4}\right)\int_{\partial\mathcal{S}} \sin(2\pi S^\alpha\cdot x)\,\phi_k^{(m)}(x)\,\mathrm{d}A(x)$$
$$\times\int_{\partial\mathcal{S}} \sin(2\pi S^\alpha\cdot y)\,\phi_l^{(n)}(y)\,\mathrm{d}A(y), \tag{62}$$

and we can show that (Nunan 1983)

$$\int_{\partial\mathcal{S}} \sin(\zeta\cdot x)\,x_1^{p_1} x_2^{p_2} x_3^{p_3}\,\mathrm{d}A(x) = (-1)^{\beta-1}\,4\pi b^{2\beta+1}$$
$$\times\sum_{c_1=0,\,1}^{p_1}\sum_{c_2=0,\,1}^{p_2}\sum_{c_3=0,\,1}^{p_3} \chi_{c_1}^{p_1}\chi_{c_2}^{p_2}\chi_{c_3}^{p_3}\frac{(b\zeta_1)^{c_1}(b\zeta_2)^{c_2}(b\zeta_3)^{c_3}}{|b\zeta|^{\beta+\frac{1}{2}(c_1+c_2+c_3-1)}}\, j_{\beta+\frac{1}{2}(c_1+c_2+c_3-1)}(|b\zeta|). \tag{63}$$

Here $\beta = \frac{1}{2}(p_1+p_2+p_3+1)$, the index c_j begins at 0 or 1 according to whether p_j is even or odd and increases by steps of 2, and

$$\chi_c^p = (-2)^{\frac{1}{2}(c-p)}\frac{p!}{c!\left(\dfrac{p-c}{2}\right)!}. \tag{64}$$

The matrix elements involve double integrals, and thus a product of terms such as (63), but the required calculations are not as computationally difficult as they might seem. Owing to recursive identities for the spherical Bessel functions $j_n(z)$, the matrix

elements corresponding to basis functions of degree less than or equal to a given M can, for cubic lattices, all be computed from the principal lattice sums

$$SS_\kappa^{rst} = \sum_\alpha{}' \frac{z_1^{2r} z_2^{2s} z_3^{2t}}{|z|^{2r+2s+2t+2\kappa+4}} \sin^2(|z|), \tag{65}$$

$$SC_\kappa^{rst} = \sum_\alpha{}' \frac{z_1^{2r} z_2^{2s} z_3^{2t}}{|z|^{2r+2s+2t+2\kappa+3}} \sin(|z|)(\cos(|z|), \tag{66}$$

$$CC_\kappa^{rst} = \sum_\alpha{}' \frac{z_1^{2r} z_2^{2s} z_3^{2t}}{|z|^{2r+2s+2t+2\kappa+2}} \cos^2(|z|), \tag{67}$$

for $r+s+t = M+1, 0 \leqslant r \leqslant s \leqslant t$ and $1 \leqslant \kappa \leqslant M$ (SS_κ^{rst} is also needed for $\kappa = 0$). In (65)–(67), z denotes $2\pi b S^\alpha$. For $M = 11$, which corresponds to 78 basis functions, there are only 684 such principal lattice sums.

For cubic lattices the symmetries of ν_{ij} and the basis polynomials produce considerable structure in the linear system (58). Specifically, the matrix A_{ijkl}^{mn} can be written in block-diagonal form with each block having one fourth the rank of the full matrix, and only one block each is required to compute the coefficients for f_i^{11} and f_i^{12}. The actual systems that must be solved to compute f_i^{11} and f_i^{12} are

$$\begin{bmatrix} A_{11kl}^{11} & A_{12lk}^{12} & A_{12kl}^{12} \\ A_{12kl}^{12} & A_{11kl}^{11} & A_{12lk}^{12} \\ A_{12lk}^{12} & A_{12kl}^{12} & A_{11kl}^{11} \end{bmatrix} \begin{bmatrix} a_{1k}^{11} \\ a_{2k}^{12} \\ a_{3k}^{13} \end{bmatrix} = \frac{1}{3} \begin{bmatrix} 2b_{1l}^{22} \\ -b_{1l}^{22} \\ -b_{1l}^{22} \end{bmatrix} \tag{68}$$

and

$$\begin{bmatrix} A_{11kl}^{22} & A_{12kl}^{12} & A_{31kl}^{42} \\ A_{12lk}^{12} & A_{22kl}^{11} & A_{32kl}^{41} \\ A_{31lk}^{42} & A_{32lk}^{41} & A_{33kl}^{44} \end{bmatrix} \begin{bmatrix} a_{1k}^{22} \\ a_{2k}^{21} \\ a_{3k}^{24} \end{bmatrix} = \begin{bmatrix} b_{1l}^{22} \\ 0 \\ 0 \end{bmatrix} \tag{69}$$

respectively. Each of these matrices is symmetric.

The matrix in (68) is singular, with null vector $a_{ik}^{1m} = \delta_{k1}\delta_{im}$ representing a uniform pressure on the surface of the sphere. There is no physical basis for choosing one pressure over another (condition (56) is merely convenient), but as previously mentioned a uniform pressure does not affect the effective viscosity. We use a least-squares algorithm to obtain a solution of (68), and do not implement (56) in the matrix equations.

The solution of (55) and (56) is not our final goal. That is rather the effective viscosity, which requires the integrals

$$\int_{\partial \mathscr{S}} x_j f_i^{1\nu} \, dA = \sum_{k=1}^\infty a_{ik}^{\nu j} b_{1k}^{22} \quad (\nu = 1, 2). \tag{70}$$

Thus we computed the coefficients in the effective viscosity tensor by solving the linear systems (68) and (69), and then computing

$$\alpha = \tfrac{1}{2} \sum_k b_{1k}^{22} (a_{1k}^{11} - a_{2k}^{12}) \tag{71}$$

and

$$\beta = \tfrac{1}{2} \sum_k b_{2k}^{22} (a_{2k}^{21} + a_{1k}^{22}). \tag{72}$$

Then we substituted these results into the formula for μ_{ijkl}^*, which is

$$\mu_{ijkl}^* = \mu(1+\beta)\tfrac{1}{2}(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk} - \tfrac{2}{3}\delta_{ij}\delta_{kl}) + \mu(\alpha - \beta)(\delta_{ijkl} - \tfrac{1}{3}\delta_{ij}\delta_{kl}). \tag{73}$$

7. Asymptotic form of the effective viscosity tensor

The numerical method just described constituted our primary method of solution for the effective viscosity problem. However, as will be illustrated in §8, the numerical method suffered from poor convergence properties at very high concentrations. An asymptotic analysis for the high concentration situation was therefore undertaken. An outline of the analysis is presented in Appendix B; the results are reported here.

The small parameter in the asymptotic analysis is

$$\epsilon = 1 - \left(\frac{c}{c_{\max}}\right)^{\frac{1}{3}}. \tag{74}$$

The final result for the equation defining the effective viscosity tensor is

$$\mu_{ijkl}^{*}\gamma_{ij}\gamma_{kl} = \mu c_{\max}\gamma_{ij}\gamma_{kl}\{(\tfrac{9}{16}\epsilon^{-1} + \tfrac{21}{80}\ln\epsilon^{-1})\sum_{\hat{n}}n_{i}n_{j}n_{k}n_{l}$$

$$+ \tfrac{3}{4}[\delta_{ik}\sum_{\hat{n}}n_{j}n_{l} - \epsilon_{ipq}\epsilon_{krs}A_{rp}^{-1}\sum_{\hat{n}}n_{l}n_{s}\sum_{\hat{n}}n_{j}n_{q}]\ln\epsilon^{-1} + O(1)\}. \tag{75}$$

Here the sums are over all unit vectors $\hat{n}$ in the direction of a nearest-neighbour sphere, with respect to a single reference sphere, and the matrix A_{ij} is defined by

$$A_{ij} = \sum_{\hat{n}}(\delta_{ij} - n_{i}n_{j}). \tag{76}$$

If each sphere in the lattice has more than two nearest-neighbour spheres then **A** is invertible. If each sphere has only two nearest-neighbour spheres, in directions $\pm\hat{n}$, then **A** is singular with codimension 1 and null vector $\hat{n}$. For such two-nearest-neighbour lattices $\mathbf{A}^{-1}$ should be replaced in (75) by $(\mathbf{B}^{T}\mathbf{B})^{-1}\mathbf{A}$, where **B** is the 4×3 matrix consisting of **A** augmented by $\hat{n}^{T}$.

When (75) is solved for μ_{ijkl}^{*}, the term in brackets must be made symmetric and traceless in the subscript pairs i, j and k, l (by adding terms of the form $c_{ij}\delta_{kl}$ or $\delta_{ij}c_{kl}$), and symmetric between these pairs (by adding a term of the form c_{ijkl}, where $c_{ijkl} = -c_{klij}$), in order to satisfy the defining conditions (16). The precise way to do this will depend upon the particular lattice being considered. A number of special cases have been examined for illustration, and the resulting effect viscosity tensors are as follows.

(I) Two-nearest-neighbour lattices, with incident direction $\pm\hat{n}$:

$$\mu_{ijkl}^{*} = \mu c_{\max}(\tfrac{9}{8}\epsilon^{-1} + \tfrac{81}{40}\ln\epsilon^{-1})(n_{i}n_{j} - \tfrac{1}{3}\delta_{ij})(n_{k}n_{l} - \tfrac{1}{3}\delta_{kl}) + O(1). \tag{77}$$

(II) Cubic lattices:

$$\mu_{ijkl}^{*} = \mu c_{\max}[(\tfrac{9}{16}\epsilon^{-1} + \tfrac{21}{80}\ln\epsilon^{-1})\sum_{\hat{n}}(n_{i}n_{j} - \tfrac{1}{3}\delta_{ij})(n_{k}n_{l} - \tfrac{1}{3}\delta_{kl})$$

$$+ \tfrac{1}{8}[\sum_{\hat{n}}1](\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk} - \tfrac{2}{3}\delta_{ij}\delta_{kl})\ln\epsilon^{-1}] + O(1). \tag{78}$$

(III) Tetragonal lattice (orthogonal basis $\mathbf{a}_{i}$, with $|\mathbf{a}_{1}| = |\mathbf{a}_{2}| < |\mathbf{a}_{3}|$):

$$\mu_{ijkl}^{*} = \mu c_{\max}[(\tfrac{9}{16}\epsilon^{-1} + \tfrac{21}{80}\ln\epsilon^{-1})\sum_{\hat{n}}(n_{i}n_{j} - \tfrac{1}{3}\delta_{ij})(n_{k}n_{l} - \tfrac{1}{3}\delta_{kl})$$

$$+ \tfrac{3}{16}[B_{ik}B_{jl} + B_{il}B_{jk} - \tfrac{2}{3}\delta_{ij}B_{kl}^{2} - \tfrac{2}{3}\delta_{kl}B_{ij}^{2} + \tfrac{2}{9}\delta_{ij}\delta_{kl}B_{pp}^{2}]\ln\epsilon^{-1}] + O(1), \tag{79}$$

where
$$B_{ij} = \sum_{\hat{n}}n_{i}n_{j} \quad \left(\hat{n} = \pm\frac{\mathbf{a}_{1}}{|\mathbf{a}_{1}|}, \pm\frac{\mathbf{a}_{2}}{|\mathbf{a}_{2}|}\right).$$

(IV) Hexagonal lattice: Same as for tetragonal lattice, but with the factor of $\frac{3}{16}$ in the second line of (79) replaced by $\frac{1}{8}$.

Equation (78) for cubic lattices can be written in the form (52), from which asymptotic formulas for α and β are easily obtained. They will be presented in §8, where numerical estimates of the leading non-singular terms in the asymptotic expansions will also be given.

8. Numerical results and discussion

The Galerkin method used to solve (55) and (56) involves approximating $f_i^{1\nu}$ by a finite linear combination of basis polynomials. For any given calculation all basis polynomials of degree less than or equal to a specified M were used. The number of basis functions for a given M is $\frac{1}{2}(M+1)(M+2)$, but because not all basis functions contribute to each $f_i^{1\nu}$, the actual number of unknowns is $\frac{3}{8}(M+1)(M+3)$ for f_i^{11} and $\frac{1}{8}(M+1)(3M+5)$ for f_i^{12}. By using increasing values of M while keeping other parameters of the problem fixed† we obtained a sequence of results converging to the exact values of α and β. Table 1 illustrates the convergence of the solutions as a function of M for various values of c. These results are typical.

For low concentrations convergence is very rapid, and precise results can be obtained with just a few basis functions. Indeed, for concentrations less than 0.06 only 3 basis functions ($M = 1$) give results which have four decimal places of precision, and for concentrations less than 0.24 a similar degree of precision is obtained with just 21 basis functions ($M = 5$).

For higher concentrations the convergence rate is not as good. As c increases, the traction on the surface of a sphere becomes more and more peaked near points which are closest to other spheres, and more basis polynomials are needed to represent it accurately. However, the matrices increase in size with the number of basis functions, and their condition numbers increase accordingly. The effect of the inadequacy of the basis functions in representing the traction is magnified due to these high-condition numbers. This is particularly noticeable in the last column of table 1, where the computed value of α for $M = 13$ is actually less than that for $M = 11$. The typical behaviour was for α to increase with M, and such anomalies were never seen at lower concentrations.

The inadequacy of a necessarily finite number of basis functions in representing the traction at high concentrations was the major source of numerical error. There was strong evidence of a downward bias in the high-concentration computations, for all three cubic lattices. This was because the basis polynomials could represent only a smoothed approximation to the traction at high concentrations, and so the computed estimates of the viscosity coefficients were sometimes too low. The face-centred cubic lattice results were the most adversely affected, because each sphere in a face-centred cubic lattice has more nearest neighbours than spheres in either a simple or body-centred cubic lattice. Thus the traction on the sphere surface is more complicated, and less well representable by the basis polynomials, for face-centred cubic lattices than in the other two cases.

The truncation error in the principal lattice sums (65)–(67), as measured by the variation in the computed coefficients α and β as the summation range was increased, was insignificant except for high concentrations and high values of M. Cancellation

† Since the matrix for a given M contains all the elements necessary to form the matrix for a smaller M, computing a sequence of results requires only slightly more effort than computing one result.

$M \backslash c$	0.02	0.06	0.12	0.24	0.32	0.40	0.46	0.49
1	0.053803	0.18650	0.46468	1.4679	2.6563	4.3606	5.8289	6.5165
3	0.053804	0.18653	0.46552	1.5022	2.8638	5.3206	8.4031	10.479
5	—	0.18654	0.46579	1.5224	3.0151	6.2231	11.397	15.353
7	—	—	0.46580	1.5228	3.0239	6.3882	12.859	19.388
9	—	—	—	1.5228	3.0256	6.4440	13.674	22.260
11	—	—	—	—	3.0257	6.4541	14.022	24.499
13	—	—	—	—	—	6.4541	14.037	24.256

TABLE 1. Convergence of α for simple cubic lattices

and roundoff errors in the principal lattice sums were deemed insignificant in all cases, as the result did converge with increasing summation range.

The speed of the algorithm depended upon the type of lattice, the number of basis polynomials, and the summation range. The computation of the principal lattice sums consumed the preponderance of CPU time, and the complexity of this computation increased as the cube of the summation range and as the square of M for a given lattice. The sums contained twice as many points for a body-centred lattice as for a simple cubic lattice, and four times as many for a face-centred lattice. One of the longer runs typically required several minutes of CPU time on a VAX-11/780, while a low-concentration run required only a few CPU seconds.

As previously mentioned, the numerical results for low concentrations are both very precise and quick to compute. The question remains as to whether the results are correct. Zuzovsky (1976) obtained low concentration asymptotic results for what we have identified as the effective viscosity tensor. His results are

$$\alpha = \tfrac{5}{2}c[1 - (1 - 60b)c + 12ac^{\frac{5}{3}} + O(c^{\frac{7}{3}})]^{-1}, \tag{80}$$

$$\beta = \tfrac{5}{2}c[1 - (1 + 40b)c - 8ac^{\frac{5}{3}} + O(c^{\frac{7}{3}})]^{-1}, \tag{81}$$

where a and b depend on the lattice geometry as follows:

	SC	BCC	FCC
a	0.2857	−0.0897	−0.0685
b	−0.04655	0.01432	0.01271

A comparison of our computed results with (80) and (81) is illustrated in table 2. The results agree to four decimal places for concentrations of 0.04 or less, with the degree of agreement slowly dropping off as the concentration increases. Zuzovsky's formulas thus confirm that our program works correctly and produces accurate results. We can then turn the tables and use our results to check his. The values in table 2 and similar comparisons for the other lattices indicate that (80) and (81) are accurate to within 0.2 % for concentrations up to approximately 25 % that of close packing, and to within 5 % for concentrations up to approximately 50 % that of close packing.

For high concentrations, we compare the numerical results with our asymptotic results for α and β, obtained by writing (78) in the form (52). These formulas contain only singular terms in the small parameter $\epsilon = 1 - (c/c_{\max})^{\frac{1}{3}}$. To improve them, we assume that the leading non-singular terms are 1 and $\epsilon \ln \epsilon^{-1}$. Thus, for example, we write the difference between α and the singular asymptotic terms for a simple cubic lattice in the form

$$\alpha - (\tfrac{3}{16}\pi\epsilon^{-1} + \tfrac{27}{80}\pi \ln \epsilon^{-1}) = C + D\epsilon \ln \epsilon^{-1} + O(\epsilon), \tag{82}$$

c	α (comp.)	α (asymp.)	β (comp.)	β (asymp.)
0.005	0.012735	0.012735	0.012451	0.012450
0.01	0.025941	0.025943	0.024813	0.024812
0.02	0.053804	0.053810	0.049320	0.049316
0.04	0.11567	0.11570	0.097696	0.097677
0.08	0.26755	0.26756	0.19337	0.19323
0.12	0.46580	0.46517	0.28995	0.28938
0.16	0.72502	0.72100	0.39009	0.38830
0.20	1.0666	1.0506	0.49665	0.49209
0.24	1.5228	1.4726	0.61306	0.60299
0.28	2.1459	2.0068	0.74379	0.72355

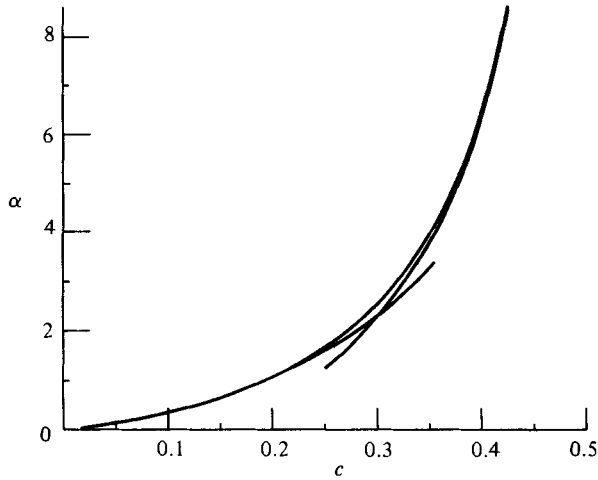TABLE 2. Numerical and asymptotic results: low concentrations, simple cubic lattices

FIGURE 1. α for a simple cubic lattice versus concentration c, computed numerically and also from the low- and high-concentration expansions.
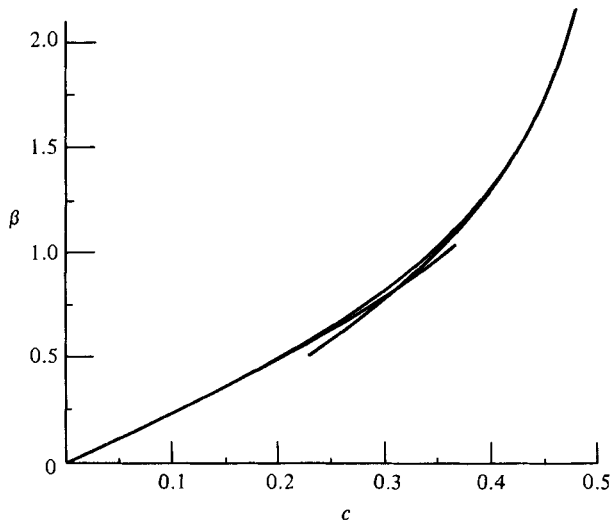

FIGURE 2. β for a simple cubic lattice versus concentration c, computed numerically and also from the low- and high-concentration expansions.
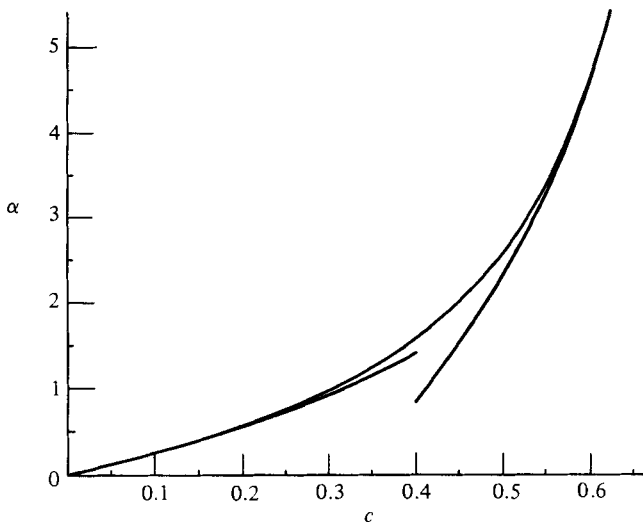
FIGURE 3. α for a body-centred cubic lattice versus concentration c, computed numerically and also from the low- and high-concentration expansions.
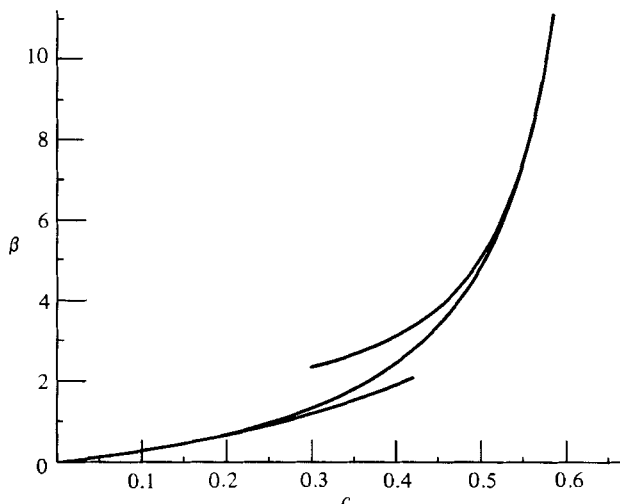

FIGURE 4. β for a body-centred cubic lattice versus concentration c, computed numerically and also from the low- and high-concentration expansions.

where C and D are constants to be determined. By plotting the computed value of the left side of (82) against $\epsilon \ln \epsilon^{-1}$, we determine C and D from the intercept and slope of a line drawn through the plotted data for small ϵ. We do the same for all other α and β.

Unfortunately, it is precisely for small ϵ that the numerical results are least precise. Moreover, in computing the difference between the numerical and asymptotic values, we may lose significant digits. For a body-centred or face-centred cubic lattice, the graphs for α exhibit definite linear behaviour as ϵ becomes small, and the parameters C and D of the limiting line can be determined with confidence. But in the other cases our estimates of C and D are not as reliable, and in the case of β for a face-centred cubic lattice no estimate of C and D can be made.
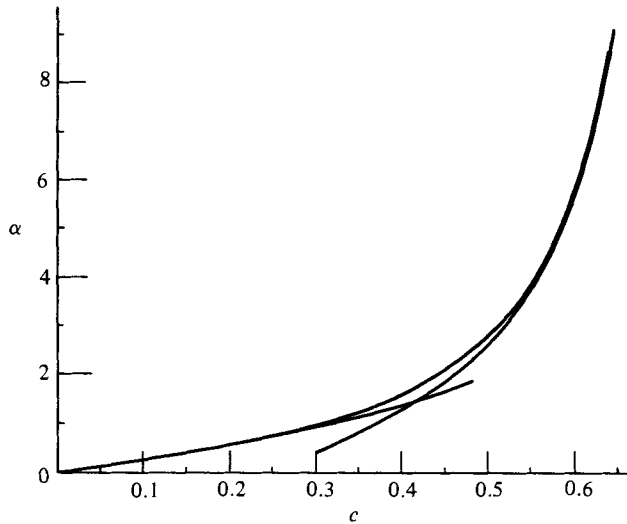
FIGURE 5. α for a face-centred cubic lattice versus concentration c, computed numerically and also from the low- and high-concentration expansions.
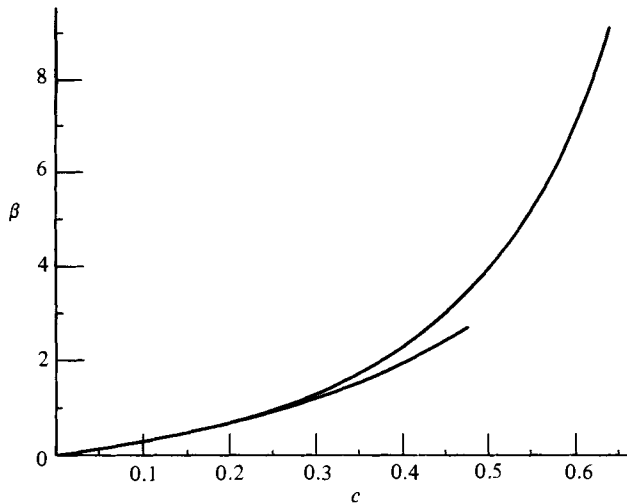

FIGURE 6. β for a face-centred cubic lattice versus concentration c, computed numerically and also from the low-concentration expansion.

The high concentration asymptotic formulas for the effective viscosity coefficients α and β, including our estimates of the constant terms and coefficients of $\epsilon \ln \epsilon^{-1}$, are as follows.

Simple cubic lattices:

$$\alpha = \tfrac{3}{16}\pi\epsilon^{-1} + \tfrac{27}{80}\pi \ln \epsilon^{-1} + 3.1 + 0.25\epsilon \ln \epsilon^{-1} + O(\epsilon), \tag{83}$$

$$\beta = \tfrac{1}{4}\pi \ln \epsilon^{-1} + 0.63 + 0.0\epsilon \ln \epsilon^{-1} + O(\epsilon). \tag{84}$$

Body-centred cubic lattices:

$$\alpha = \tfrac{1}{4}\sqrt{3}\,\pi \ln \epsilon^{-1} - 1.73 + 12.3\epsilon \ln \epsilon^{-1} + O(\epsilon), \tag{85}$$

$$\beta = \tfrac{1}{8}\sqrt{3}\,\pi\epsilon^{-1} + \tfrac{37}{120}\sqrt{3}\,\pi \ln \epsilon^{-1} + 12.8 - 35\epsilon \ln \epsilon^{-1} + O(\epsilon). \tag{86}$$

Face-centred cubic lattices:

$$\alpha = \tfrac{3}{32}\sqrt{2}\,\pi\epsilon^{-1} + \tfrac{87}{160}\sqrt{2}\,\pi\ln\epsilon^{-1} + 9.7 - 15.5\epsilon\ln\epsilon^{-1} + O(\epsilon), \qquad (87)$$

$$\beta = \tfrac{3}{16}\sqrt{2}\,\pi\epsilon^{-1} + \tfrac{47}{80}\sqrt{2}\,\pi\ln\epsilon^{-1} + O(1). \qquad (88)$$

In (83)–(88) $\epsilon = 1 - (c/c_{\max})^{\frac{1}{3}}$, where $c_{\max} = \tfrac{1}{6}\pi$ for a simple cubic lattice, $\tfrac{1}{8}\sqrt{3}\,\pi$ for a body-centred cubic lattice, and $\tfrac{1}{6}\sqrt{2}\,\pi$ for a face-centred lattice.

Figures 1–6 show our computed results plotted against the volume concentration of the spheres, and also show graphs of Zuzovsky's low-concentration asymptotic formulas and our high-concentration asymptotic formulas.

Appendix A. A symmetry relation for μ_{ijkl}^{*}

In this appendix we prove the identity $\mu_{ijkl}^{*} = \mu_{klij}^{*}$, where μ_{ijkl}^{*} is the tensor appearing in (12). The proof is a modification of that of Hinch (1972), who proved a related result for a single particle in a fluid undergoing Stokes flow.

We begin with the reciprocal theorem of Lorentz (1896), which states that for any two solutions (u_i, σ_{ij}) and (u_i', σ_{ij}') of the Stokes equations (2)–(4) in a volume bounded by a surface Γ with unit normal $\boldsymbol{n}$,

$$\int_{\Gamma} u_i \sigma_{ij}' n_j \, \mathrm{d}A = \int_{\Gamma} u_i' \sigma_{ij} n_j \, \mathrm{d}A. \qquad (\text{A }1)$$

Let (u_i, σ_{ij}) and (u_i', σ_{ij}') be the two solutions of (2)–(7) corresponding to γ_{ij} and γ_{ij}' respectively. Let T be any unit lattice cell which completely encloses the sphere at the origin. Taking Γ to be the boundary of the fluid region within T, the first integral in (A 1) becomes

$$\int_{\partial T - \partial \mathscr{S}} u_i \sigma_{ij}' n_j \, \mathrm{d}A = \int_{\partial T} u_i \sigma_{ik}' n_k \, \mathrm{d}A - \int_{\partial \mathscr{S}} \epsilon_{ikl} \omega_k x_l \sigma_{ij}' n_j \, \mathrm{d}A. \qquad (\text{A }2)$$

Only the non-periodic part of $u_i \sigma_{ij}'$ contributes to the integral over the cell boundary in (A 2). Moreover, the no-torque condition (7) implies that the last integral in (A 2) is zero. Thus we have

$$\int_{\partial T - \partial \mathscr{S}} u_i \sigma_{ij}' n_j \, \mathrm{d}A = \int_{\partial T} \gamma_{ij} x_j \sigma_{ik}' n_k \, \mathrm{d}A. \qquad (\text{A }3)$$

By using (3)–(6) and the divergence theorem we can then show that

$$\int_{\partial T - \partial \mathscr{S}} u_i \sigma_{ij}' n_j \, \mathrm{d}A = \mu \tau_0 \gamma_{ij} (\gamma_{ij}' + \gamma_{ji}') + \int_{\partial \mathscr{S}} \gamma_{ij} x_j \sigma_{ik}' n_k \, \mathrm{d}A, \qquad (\text{A }4)$$

which, using (11), may be written as

$$\int_{\partial T - \partial \mathscr{S}} u_i \sigma_{ij}' n_j \, \mathrm{d}A = \mu \tau_0 \gamma_{ij} (\gamma_{ij}' + \gamma_{ji}') + \gamma_{ij} M_{ijkl} \gamma_{kl}'. \qquad (\text{A }5)$$

Similarly,

$$\int_{\partial T - \partial \mathscr{S}} u_i' \sigma_{ij} n_j \, \mathrm{d}A = \mu \tau_0 \gamma_{ij}' (\gamma_{ij} + \gamma_{ji}) + \gamma_{ij}' M_{ijkl} \gamma_{kl}. \qquad (\text{A }6)$$

The reciprocal theorem and (13) then imply

$$\gamma_{ij}\mu^*_{ijkl}\gamma'_{kl} = \gamma'_{ij}\mu^*_{ijkl}\gamma_{kl}. \tag{A 7}$$

But the choices of γ_{ij} and γ'_{ij} were arbitrary, up to tracelessness, and μ^*_{ijkl} is traceless in its first and last pairs of indices. Thus

$$\mu^*_{ijkl} = \mu^*_{klij}. \tag{A 8}$$

Appendix B. Asymptotic analysis

We consider two spheres, each of radius b and with their centres a distance $2L$ apart. Let $\hat{n}$ denote a unit vector along the line connecting their centres. We define a set of cylindrical coordinates $(\bar{r}, \theta, \bar{z})$ with its origin at the midpoint between the spheres and its $\bar{z}$-axis in the $\hat{n}$-direction. Let $\hat{m}$ be a unit vector in the $\theta = 0$ direction and let $\hat{p} = \hat{n} \times \hat{m}$. For convenience we assume that the origin is translating with a velocity equal to the average velocity of the spheres.

We write the fluid velocity in cylindrical coordinates as $\boldsymbol{u} = \bar{u}\hat{r} + \bar{v}\hat{\theta} + \bar{w}\hat{z}$. We define the parameter

$$\epsilon = 1 - \frac{b}{L}, \tag{B 1}$$

which is small when the two spheres are nearly touching, and make the following scalings:

$$\left.\begin{array}{ll}
\bar{z} = \epsilon L z, & z = \epsilon^{-1}L^{-1}\bar{z}, \\[4pt]
\bar{r} = \epsilon^{\frac{1}{2}}Lr, & r = \epsilon^{-\frac{1}{2}}l^{-1}\bar{r}, \\[4pt]
\bar{u} = Lu, & u = L^{-1}\bar{u}, \\[4pt]
\bar{v} = Lv, & v = L^{-1}\bar{v}, \\[4pt]
\bar{w} = \epsilon^{\frac{1}{2}}Lw, & w = \epsilon^{-\frac{1}{2}}L^{-1}\bar{w}, \\[4pt]
\bar{p} = \mu\epsilon^{-\frac{3}{2}}p, & p = \mu^{-1}\epsilon^{\frac{3}{2}}\bar{p}.
\end{array}\right\} \tag{B 2}$$

Equations (2)–(4) can then be written as

$$u_r + r^{-1}u + r^{-1}v_\theta + w_z = 0, \tag{B 3}$$

$$-p_r + u_{zz} + \epsilon[r^{-2}u_{\theta\theta} - r^{-1}v_{r\theta} - r^{-2}v_\theta - w_{rz}] = 0, \tag{B 4}$$

$$-r^{-1}p_\theta + v_{zz} + \epsilon[r^{-1}u_{r\theta} + r^{-2}u_\theta + v_{rr} + r^{-1}v_r - r^{-2}v - r^{-1}w_{\theta z}] = 0, \tag{B 5}$$

$$-p_z + \epsilon[-u_{rz} - r^{-1}u_z - r^{-1}v_{\theta z}] + \epsilon^2[w_{rr} + r^{-1}w_r + r^{-2}w_{\theta\theta}] = 0. \tag{B 6}$$

The boundary condition (5) can be written in the scaled coordinates as

$$u \pm \tfrac{1}{2}\epsilon(h^2-1)u_z + O(\epsilon^2) = \pm(\boldsymbol{\omega}\cdot\hat{m} + p_i\gamma_{ij}n_j)\sin\theta \pm (-\boldsymbol{\omega}\cdot\hat{p} + m_i\gamma_{ij}n_j)\cos\theta$$
$$- \epsilon z[-\boldsymbol{\omega}\cdot\hat{m}\sin\theta + \boldsymbol{\omega}\cdot\hat{p}\cos\theta], \tag{B 7}$$

$$v \pm \tfrac{1}{2}\epsilon(h^2-1)v_z + O(\epsilon^2) = \pm(\boldsymbol{\omega}\cdot\hat{p} - m_i\gamma_{ij}n_j)\sin\theta \pm (\boldsymbol{\omega}\cdot\hat{m} + p_i\gamma_{ij}n_j)\cos\theta$$
$$- \epsilon^{\frac{1}{2}}\boldsymbol{\omega}\cdot\hat{n}r + \epsilon z[\boldsymbol{\omega}\cdot\hat{p}\sin\theta + \boldsymbol{\omega}\cdot\hat{m}\cos\theta], \tag{B 8}$$

$$w \pm \tfrac{1}{2}\epsilon(h^2-1)w_z + O(\epsilon^2) = -\boldsymbol{\omega}\cdot\hat{m}r\sin\theta + \boldsymbol{\omega}\cdot\hat{p}r\cos\theta \pm \epsilon^{-\frac{1}{2}}n_i\gamma_{ij}n_j \tag{B 9}$$

on $z = \pm h$, where $h = 1 + \tfrac{1}{2}r^2$.

The right-hand sides of the boundary conditions can be divided into three parts

corresponding to the relative normal, tangential and rotational motion of the spheres. We solve (B 3)–(B 6) for each part separately. For a normal motion of the spheres, v is identically zero and p, u and w do not depend on θ. We express these latter variables as $\epsilon^{\frac{1}{2}} n_i \gamma_{ij} n_j$ times a regular power series in ϵ, and perform a standard asymptotic analysis. The resulting differential equations can be integrated, with the boundary conditions determining the constants of integration, until a last second-order differential equation involving the pressure is obtained. This is solved with the following regularity conditions: (i) p_r is regular at $r = 0$, and (ii) $p \to 0$ as $r \to \infty$. The first condition is necessary so that the fluid velocity will be regular at $r = 0$, and the second condition is required so that this inner solution for the pressure can match with a (regular in ϵ) outer solution. A similar analysis is performed for the relative tangential and rotational motion problems as well. The combined results are

$$p = -\tfrac{3}{4}\epsilon^{-\frac{1}{2}} n_i \gamma_{ij} n_j h^{-2} + \epsilon^{\frac{1}{2}} n_i \gamma_{ij} n_j [(3h^{-3} - \tfrac{9}{2}h^{-4}) z^2 - \tfrac{27}{20}h^{-1} + \tfrac{21}{10}h^{-2} - \tfrac{3}{4}h^{-3})]$$

$$- \epsilon[(p_i \gamma_{ij} n_j + \boldsymbol{\omega} \cdot \hat{\boldsymbol{m}}) \sin\theta + (m_i \gamma_{ij} n_j - \boldsymbol{\omega} \cdot \hat{\boldsymbol{p}}) \cos\theta] h^{-2} rz + O(\epsilon^{\frac{3}{2}}), \quad \text{(B 10)}$$

$$u = \epsilon^{-\frac{1}{2}} n_i \gamma_{ij} n_j \tfrac{3}{4} h^{-3} r(z^2 - h^2) + [(p_i \gamma_{ij} n_j + \boldsymbol{\omega} \cdot \hat{\boldsymbol{m}}) \sin\theta$$

$$+ (m_i \gamma_{ij} n_j - \boldsymbol{\omega} \cdot \hat{\boldsymbol{p}}) \cos\theta] h^{-1} z + \epsilon^{\frac{1}{2}} n_i \gamma_{ij} n_j [(-\tfrac{3}{2}h^{-4} r + 3h^{-5} r)(z^4 - h^4)$$

$$+ (\tfrac{27}{40}h^{-2} r - \tfrac{18}{5}h^{-3} r + \tfrac{9}{8}h^{-4} r)(z^2 - h^2) - \tfrac{3}{4} r(1 - h^2)] + O(\epsilon), \quad \text{(B 11)}$$

$$v = [(-m_i \gamma_{ij} n_j + \boldsymbol{\omega} \cdot \hat{\boldsymbol{p}}) \sin\theta + (p_i \gamma_{ij} n_j + \boldsymbol{\omega} \cdot \hat{\boldsymbol{m}}) \cos\theta] h^{-1} z - \boldsymbol{\omega} \cdot \hat{\boldsymbol{n}} r \epsilon^{\frac{1}{2}} + O(\epsilon), \quad \text{(B 12)}$$

$$w = \epsilon^{-\frac{1}{2}} n_i \gamma_{ij} n_j [(h^{-3} - \tfrac{3}{2}h^{-4}) z^3 + \tfrac{3}{2} h^{-2} z] + [(p_i \gamma_{ij} n_j + \boldsymbol{\omega} \cdot \hat{\boldsymbol{m}}) \sin\theta$$

$$+ (m_i \gamma_{ij} n_j - \boldsymbol{\omega} \cdot \hat{\boldsymbol{p}}) \cos\theta] \tfrac{1}{2}(h^{-2} z^2 - 1) r - [\boldsymbol{\omega} \cdot \hat{\boldsymbol{m}} \sin\theta - \boldsymbol{\omega} \cdot \hat{\boldsymbol{p}} \cos\theta] r + O(\epsilon^{\frac{1}{2}}).$$
$$\text{(B 13)}$$

By using (3), (17) and (B 10)–(B 13), a formula for the traction on one of the spheres can be obtained. For the sphere centred at $(\bar{r} = 0, \bar{z} = -L)$ it is

$$f_i = \mu b^{-1} L \{[\epsilon^{-\frac{3}{2}} n_i \gamma_{ij} n_j (-\tfrac{3}{4} h^{-2} r) + \epsilon^{-1} [(p_i \gamma_{ij} n_j + \boldsymbol{\omega} \cdot \hat{\boldsymbol{m}}) \sin\theta$$

$$+ (m_i \gamma_{ij} n_j - \boldsymbol{\omega} \cdot \hat{\boldsymbol{p}}) \cos\theta] h^{-1} + \epsilon^{-\frac{1}{2}} n_i \gamma_{ij} n_j (\tfrac{3}{2} h^{-1} r + \tfrac{3}{5} h^{-2} r - \tfrac{3}{2} h^{-3} r) + O(1)] \hat{\boldsymbol{r}}$$

$$+ [\epsilon^{-1} [(-m_i \gamma_{ij} n_j + \boldsymbol{\omega} \cdot \hat{\boldsymbol{p}}) \sin\theta + (p_i \gamma_{ij} n_j + \boldsymbol{\omega} \cdot \hat{\boldsymbol{m}}) \cos\theta] h^{-1} + O(1)] \hat{\boldsymbol{\theta}}$$

$$+ [\epsilon^{-2} n_i \gamma_{ij} n_j \tfrac{3}{4} h^{-2} + \epsilon^{-1} n_i \gamma_{ij} n_j (\tfrac{3}{5} h^{-1} + \tfrac{9}{2} h^{-2} + \tfrac{3}{4} h^{-3})$$

$$- \epsilon^{-\frac{1}{2}} [(p_i \gamma_{ij} n_j + \boldsymbol{\omega} \cdot \hat{\boldsymbol{m}}) \sin\theta + (m_i \gamma_{ij} n_j - \boldsymbol{\omega} \cdot \hat{\boldsymbol{p}}) \cos\theta] 2h^{-1} r + O(1)] \hat{\boldsymbol{z}}\}. \quad \text{(B 14)}$$

The dominant contribution to the integral in (15) at high concentrations is from the traction near those points which are closest to neighbouring spheres. By multiplying (B 14) by x_j and integrating over just that half of the sphere facing its neighbour, and summing the result over all unit vectors $\hat{\boldsymbol{n}}$ in the direction of a nearest-neighbour sphere, we obtain

$$\int_{\partial \mathscr{S}} x_j f_i \, \mathrm{d}A = \pi \mu L^3 \sum_{\hat{\boldsymbol{n}}} [n_i n_j n_k \gamma_{kl} n_l (\tfrac{3}{2}\epsilon^{-1} + \tfrac{27}{10} \ln \epsilon^{-1}) - \tfrac{3}{2} \delta_{ij} n_k \gamma_{kl} n_l \ln \epsilon^{-1}$$

$$+ 2(\delta_{ik} - n_i n_k) \gamma_{kl} n_l n_j \ln \epsilon^{-1} - 2\epsilon_{ikl} \omega_k n_l n_j \ln \epsilon^{-1}] + O(1). \quad \text{(B 15)}$$

The identity $n_i n_j + m_i m_j + p_i p_j = \delta_{ij}$ has been used to remove any explicit dependence of (B 15) on $\hat{\boldsymbol{m}}$ or $\hat{\boldsymbol{p}}$.

By substituting (B 15) into (7) we obtain an algebraic equation for ω_k. It is

$$A_{pk} \omega_k = \epsilon_{pji} \gamma_{il} \sum_{\hat{\boldsymbol{n}}} n_j n_l, \quad \text{(B 16)}$$

where $\boldsymbol{A}$ is given by (76). If there exist only two nearest-neighbour spheres then $\boldsymbol{A}$ is singular and the component of ω_k along the line through their centres is free. However, it is easily verified that such a free component does not contribute to (B 15).

The final equation for the effective viscosity is obtained by substituting $A_{pk}^{-1}\epsilon_{pji}\gamma_{il}\Sigma n_j n_l$ for ω_k in (B 15), and then substituting the result into (15). After reordering the subscripts, and using the fact that $\tau_0^{-1}L^3 = 3c_{\mathrm{max}}/4\pi$, we obtain (75).

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