Int. J. Solids Structures, 1978, Vol. 14, pp. 349–364. Pergamon Press. Printed in Great Britain

THE EFFECTIVE ELASTIC MODULI OF COMPOSITE MATERIALS CONTAINING SPHERICAL INCLUSIONS AT NON-DILUTE CONCENTRATIONS

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(Received 3 January 1977; in revised form 3 October 1977; received for publication 7 November 1977)

Abstract—This paper concerns the determination of the effective elastic moduli of an isotropic composite material containing randomly distributed spheres of the same size at non-dilute concentrations ϕ. By employing the solutions[18, 19] for the elasticity problems of two interacting spheres in the presence of four different applied strains at infinity and a method developed by Batchelor[15, 27] and by Jeffrey[16, 17] for computing bulk quantities which involve conditionally convergent integrals, we evaluate the effective moduli of the composite exactly to order ϕ^2 and thereby extend the Einstein formula.

In the particular case of an incompressible matrix, the expression for the bulk modulus κ, when rearranged, leads to an extension of Taylor's result[20] for the expansion viscosity of an incompressible fluid containing air bubbles.

The present calculations have a wider significance than just to elasticity in that they give a better understanding of the method of normalization[17] for converting a conditionally convergent integral into one that is absolutely convergent. Specifically, when the applied strain is isotropic, two sources of indeterminancy are uncovered. The first arises from the unusual property of $S_{ii}^{(1)}$, the additional dipole of one sphere due to the interaction with a second which is required for the evaluation of the bulk moduli to $O(\phi^2)$, whose trace for $R \gg 1$ is $O(R^{-6})$, when R is the distance between the two sphere centers, whereas, all its other elements are $O(R^{-3})$. This suggests that there may exist a method for calculating the effective bulk modulus which does not require a normalization to lead to an absolutely convergent integral and which gives, apparently, a different result. Secondly, the exact method of normalization is not unambiguous in that two possible ways of normalizing are shown to exist. However, when higher-order particle interactions (especially the three-particle interactions) are taken into account, this indeterminancy is resolved and a unique type of expansion applies.

1. INTRODUCTION

We consider here the elastic behaviour of an idealized composite material consisting of many particles embedded firmly in a continuous matrix. Both the discrete and continuous phases are assumed to be linearly elastic and isotropic with bulk and shear moduli κ_p, μ_p and κ_o, μ_o respectively. We wish to calculate the effective moduli of this composite, κ^* and μ^*, which, in general, will depend on the volume concentration of the inclusions ϕ, the elastic moduli of the two phases and the shape, size and spatial distribution of the particles. The present analysis will be limited to a macroscopically homogeneous and isotropic composite with spherical inclusions of the same size and of random spatial distribution.

Several authors[1]–[14] have examined this problem before. One approach, employed by Hashin and Shtrikman [4] and by Walpole [7], uses a variational theorem to obtain upper and lower bounds for the effective moduli. The corresponding results[4] and [7] are

$$\frac{(\kappa_o+\kappa_l)\phi}{\kappa_o+\kappa_l+(\kappa_p-\kappa_o)(1-\phi)} \le \frac{\kappa^*-\kappa_o}{\kappa_p-\kappa_o} \le \frac{(\kappa_p+\kappa_g)\phi}{\kappa_o+\kappa_g+(\kappa_p-\kappa_o)(1-\phi)}, \tag{1a}$$

$$\frac{(\mu_o+\mu_l)\phi}{\mu_o+\mu_l+(\mu_p-\mu_o)(1-\phi)} \le \frac{\mu^*-\mu_o}{\mu_p-\mu_o} \le \frac{(\mu_o+\mu_g)\phi}{\mu_o+\mu_g+(\mu_p-\mu_o)(1-\phi)} \tag{1b}$$

where if $(\mu_p-\mu_o)(\kappa_p-\kappa_o)\ge 0$, then

$$\kappa_l=\frac{4}{3}\mu_0 \qquad \kappa_g=\frac{4}{3}\mu_p$$

$$\mu_l=\frac{3}{2}\left(\frac{1}{\mu_o}+\frac{10}{9\kappa_o+8\mu_o}\right)^{-1} \quad \mu_g=\frac{3}{2}\left(\frac{1}{\mu_p}+\frac{10}{9\kappa_p+8\mu_p}\right)^{-1},$$

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while if $(\mu_p - \mu_0)(\kappa_p - \kappa_o) \le 0$,

$$\kappa_l = \frac{4}{3}\mu_p \qquad \kappa_g = \frac{4}{3}\mu_0$$

$$\mu_l = \frac{3}{2}\left(\frac{1}{\mu_o} + \frac{10}{9\kappa_p + 8\mu_o}\right)^{-1} \qquad \mu_g = \frac{3}{2}\left(\frac{1}{\mu_p} + \frac{10}{9\kappa_o + 8\mu_p}\right)^{-1}.$$

These inequalities yield satisfactory estimates for the effective moduli when the ratios between the corresponding moduli of the two phases are not too large. In the extreme cases of either empty or rigid inclusions, the bounds diverge even at low concentrations. Surprisingly though, when the two phases have the same shear modulus μ_o the two bounds are equal. In fact, Hill [5] proved that, in that case, the overall bulk modulus of an isotropic mixture depends on the concentration ϕ plus the moduli κ_p and κ_o but not on the shapes of the inclusions, and gave the exact expression for the overall bulk modulus

$$\kappa^* = \kappa_o + \frac{(\kappa_p - \kappa_o)(3\kappa_o + 4\mu_o)\phi}{(3\kappa_p + 4\mu_o) - (3\kappa_p - 3\kappa_o)\phi}. \tag{2}$$

The above result for the effective bulk modulus is also exact for a material that is constructed by filling a body with composite (concentric) spheres of different sizes[13].

Recently, Miller[14] showed that the bounds for the effective modulus κ^* given by (1a) can be further improved by including the shape factor of the cell materials. For a spherical cell shape, he gave

$$\frac{[3\kappa_o + 4\mu_o + 3\kappa_o\phi(\beta^{-1} - 1)]\phi}{3\kappa_p + 4\mu_o + 3\phi(\kappa_o - 2\kappa_p + \kappa_p\beta^{-1}) + 3\phi^2(\kappa_o - \kappa_p)(\beta^{-1} - 1)} \le \frac{\kappa^* - \kappa_o}{\kappa_p - \kappa_o}$$
$$\le \frac{[3\kappa_o + 4\mu_o + 4\phi(\mu_p - \mu_o)]\phi}{3\kappa_p + 4\mu_o + \phi[3(\kappa_o - \kappa_p) + 4(\mu_p - \mu_o)]} \tag{3}$$

for $\beta = (\mu_p/\mu_o)$ and $\kappa_p \ge \kappa_o$. When the volume fraction of the particles is small, the above bounds converge to

$$\kappa^* = \kappa_p + (\kappa_p - \kappa_o)\frac{3\kappa_o + 4\mu_o}{3\kappa_p + 4\mu_o}\phi.$$

A second method, valid at infinite dilution for any ratio of the moduli of the two phases, yields the effective moduli by considering the effect of a single inclusion in an infinite medium under a specified strain. Using this technique, Hashin [3] and others [1, 2] obtained the $O(\phi)$ terms in the expressions for κ^* and μ^*, while Walpole [8] determined in part the $O(\phi^2)$ contribution. Walpole's results can be written as

$$\frac{\kappa^*}{\kappa_o} = 1 + \left(1 + \frac{4\mu_o}{3\kappa_o}\right)\gamma_1\phi + \left(1 + \frac{4\mu_o}{3\kappa_o}\right)\gamma_1^2\phi^2 + O(\phi^3) \tag{4}$$

and

$$\frac{\mu^*}{\mu_o} = 1 + 15(1 - \nu_o)\gamma_2\phi + 30(1 - \nu_o)(4 - 5\nu_o)\gamma_2^2\phi^2 + O(\phi^3) \tag{5}$$

where $\gamma_1 = (3\kappa_p - 3\kappa_o)/(3\kappa_p + 4\mu_o)$, ν_o is Poisson's ratio for the matrix and $\gamma_2 = (\beta - 1)/[2\beta(4 - 5\nu_o) + (7 - 5\nu_o)]$, with $\beta \equiv \mu_p/\mu_o$ being the ratio of the two shear moduli.

To $O(\phi^2)$, these expressions for κ^* and μ^* coincide with the upper bounds given in (1a) and (1b) when the particles are weaker than the matrix ($\kappa_o > \kappa_p$ and $\mu_o > \mu_p$), and with the lower bounds when the converse is true. Unfortunately, Walpole's[8] analysis does not take into account particle-pair interactions within the composite and hence, strictly speaking, (4) and (5) are valid only for a suspension of well-separated spheres. Thus, when the spheres are randomly distributed in the suspension, eqns (4) and (5) are valid only to $O(\phi)$.

In a recent paper by Willis and Acton[12] the effective elastic moduli were also calculated to $O(\phi^2)$ through the use of an integral equation formulation. However, since these authors only used the far field solution for the two-sphere interaction problem, their results are not exact and can be shown to be an approximation to our solution.

In this paper, the coefficient of the ϕ^2 term will be determined for a randomly distributed suspension using a method developed recently by Batchelor and Green[15] and by Jeffrey[16, 17] to relate the macroscopic properties of a composite to its microscopic structure through a probability density function describing the distribution of the inclusions in the medium. To this end, it will be necessary to make use of the solution of certain two-sphere problems presented in Chen's dissertation[18], henceforth referred to as [I], and a companion paper by Chen and Acrivos[19]. For the purpose of comparing them with [4] and [5], the resulting expressions for the effective moduli will then be presented in the form

$$\frac{\kappa^*}{\kappa_o} = 1 + \left(1 + \frac{4\mu_o}{3\kappa_o}\right)\gamma_1\phi + \left(1 + \frac{4\mu_o}{3\kappa_o}\right)\gamma_1^2 H_1\phi^2 + O(\phi^3) \tag{6}$$

and

$$\frac{\mu^*}{\mu_o} = 1 + 15(1-\nu_o)\gamma_2\phi + 30(1-\nu_o)(4-5\nu_o)\gamma_2^2 H_2\phi^2 + O(\phi^3). \tag{7}$$

The coefficient H_1 and H_2 in the above equations depend on the parameter β and the two Poisson's ratios ν_p and ν_o. The function H_1 has been evaluated for different combinations of β, ν_p and ν_o; however, owing to the difficulty of solving the interaction problem for two elastic spheres in a shear strain, H_2 was calculated only for the limiting cases of rigid particles ($\beta = \infty$) and cavities ($\beta = 0$). Physically, the values given for H_1 and H_2 apply to a suspension in which the spheres are randomly distributed. For a suspension containing well-separated spheres, a case studied by Walpole[8], both H_1 and H_2 are equal to unity.

The present results satisfy the Hashin–Shtrikman variational bounds[4], Miller's bounds for the effective bulk modulus[14], and are also in good agreement with Batchelor and Green's[15] calculation of the effective shear modulus for the case of rigid spheres embedded in an incompressible matrix. Their coefficient of the ϕ^2 term in (7) is 5.2 ± 0.3 while ours is 5.01 with a possible error in the third digit. Willis and Acton's[12] corresponding result (155/32) is lower because, as mentioned earlier, these authors used only the far field solution for two-sphere interaction problem.

When the matrix is almost incompressible ($\nu_o \sim 0.5$ and κ_o is large), eqn (6) can be recast in its equivalent form

$$\kappa^* = \kappa_o\left[1 - \left(1 + \frac{4\mu_o}{3\kappa_o}\right)\gamma_1\phi + \left(1 + \frac{4\mu_o}{3\kappa_o}\right)\frac{4\mu_o}{3\kappa_o}\gamma_1^2\left(1 - \frac{1+\nu_o}{2}H\right)\phi^2 + O(\phi^3)\right]^{-1} \tag{8}$$

where H is defined by $H_1 = 1 + (1-2\nu_o)H$. In the particular case of an incompressible matrix, this expression for κ^* becomes

$$\kappa^* = \frac{4\mu_o + 3\kappa_p}{3\phi} - \mu_o\left(\frac{4}{3} - H\right) + O(\phi), \tag{9}$$

which, when the particles are cavities, gives for the effective Lamé constant λ^*

$$\lambda^* = \kappa^* - \frac{2}{3}\mu^* = \frac{4\mu_o}{3\phi} - 2.399\mu_o + O(\phi). \tag{10}$$

The latter extends Taylor's result[20] for the expansion viscosity of an incompressible fluid containing air bubbles.

We will now proceed with the development of the theory.

2. GENERAL EXPRESSION FOR THE EFFECTIVE ELASTIC MODULI TO ORDER ϕ^2

For composite materials satisfying the conditions of macroscopic homogeneity, the bulk stress $\langle\sigma_{ij}\rangle$ and the bulk strain $\langle\varepsilon_{ij}\rangle$ are related by (Russel and Acrivos[21]),

$$\langle\sigma_{ij}\rangle = \lambda_o\langle\varepsilon_{kk}\rangle\delta_{ij} + 2\mu_o\langle\varepsilon_{ij}\rangle + \sum_{ij} \tag{11}$$

where

$$\sum_{ij} = \frac{1}{V}\sum_{n=1}^{N}\int_{A_n}\{x_j\sigma_{ik}n_k - \lambda_o n_k u_k\delta_{ij} - \mu_o(u_in_j + u_jn_i)\}\mathrm{d}A.$$

Here, V is a sample volume whose dimensions are large relative to a, the radius of an individual spherical particle within the composite, but small relative to the characteristic macroscale of the system. Also, N is the number of spheres within V, A_n is the surface of the nth sphere, x_i is the position vector with respect to a fixed origin, n_i is the unit outer normal to A_n, and the brackets $\langle\ \rangle$ denote the volume (or ensemble) average of the enclosed quantity. In deriving (11), the assumption has been made that the matrix is an isotropic and linearly elastic material with Lamé constants λ_o and μ_o, where μ_o is also called the modulus of rigidity or the shear modulus. The relations between the Lamé constants λ and μ, Poisson's ratio ν, Young's modulus E, and the bulk modulus (sometimes called the modulus of compression) κ of an isotropic material can be found in standard texts on the subject, e.g. Sokolnikoff[22]. An isotropic material is characterized by any two of the five parameters listed above. Of these we shall employ primarily κ, μ and ν, using the subscripts o, p and the superscript * to denote the matrix, the inclusions and the composite, respectively. Here and elsewhere in this paper, we adopt the Cartesian tensor subscript notation with repeated subscripts indicating summation.

The effects of the inclusions on the bulk stress are contained in the particle stress Σ_{ij}, which vanishes indentically in the absence of the discrete phase. Now define

$$S_{ij} = \int_{A_{\text{ref}}}\{x_j\sigma_{ik}n_k - \lambda_o u_k n_k\delta_{ij} - \mu_o(u_in_j + u_jn_i)\}\mathrm{d}A, \tag{12}$$

where A_{ref} is the surface of a reference sphere. Equation (11) can then be written as

$$\langle\sigma_{ij}\rangle = \lambda_o\langle\varepsilon_{kk}\rangle\delta_{ij} + 2\mu_o\langle\varepsilon_{ij}\rangle + n\bar{S}_{ij}, \tag{13}$$

where $n = (\phi/V_p)$ is the number density of the inclusions in the composite and V_p denotes the volume of a single sphere. The quantity $\bar{S}_{ij}$ is the average value of S_{ij} over all particles, i.e.

$$\bar{S}_{ij} = \int S_{ij}(C|0)P(C|0)\,\mathrm{d}C,$$

$P(C/0)$ being the probability density function of the configuration C with the reference sphere at the origin†. If $\phi \ll 1$, so that the interaction between particles can be neglected, the average

†The parameter $\bar{S}_{ij}$ introduced above can also be related to the additional strain energy stored in the composite. Specifically, we define W, the average strain energy over the volume V of the composite, by

$$W = \frac{1}{V}\int\frac{1}{2}\sigma_{ij}\varepsilon_{ij}\mathrm{d}V = \frac{1}{2V}\int\sigma_{ij}\frac{\partial u_i}{\partial x_j}\mathrm{d}V,$$

which becomes

$$W = \frac{1}{2}\frac{\partial\langle u_i\rangle}{\partial x_j}\langle\sigma_{ij}\rangle + \frac{1}{2V}\int\frac{\partial(\sigma_{ij}u_i')}{\partial x_j}\mathrm{d}V$$
$$= \frac{1}{2}\langle\varepsilon_{ij}\rangle\langle\sigma_{ij}\rangle + \frac{1}{2}\frac{\partial\langle\sigma_{ij}u_i'\rangle}{\partial x_j},$$

if the displacement is expressed as the sum of the average field and the disturbance, i.e. $u_i = \langle u_i\rangle + u_i'$. However, since the value $\bar{S}_{ij}$ can be approximated by $S_{ij}^{(0)}$, the value of S_{ij} for a single inclusion in an infinite matrix. In this case, (13) becomes

$$\langle\sigma_{ij}\rangle = \lambda_o\langle\varepsilon_{kk}\rangle\delta_{ij} + 2\mu_o\langle\varepsilon_{ij}\rangle + \frac{\phi}{\frac{4}{3}\pi a^3} S_{ij}^{(0)} \tag{14}$$

with an error of $O(\phi^2)$. $S_{ij}^{(0)}$ can be evaluated from the solution for an isotropic tension[3] and that for a simple tension[23] to be

$$S_{ij}^{(0)} = \frac{4}{3}\pi a^3\left(\kappa_o + \frac{4}{3}\mu_o\right)\gamma_1\langle\varepsilon_{kk}\rangle\delta_{ij} + 40\pi a^3\mu_o(1-\nu_o)\gamma_2\left\{\langle\varepsilon_{ij}\rangle - \frac{1}{3}\langle\varepsilon_{kk}\rangle\delta_{ij}\right\}. \tag{15}$$

In obtaining the above from the single particle solution, the applied strain at infinity was set equal to the bulk strain $\langle\varepsilon_{ij}\rangle$.

Since the effective moduli are defined by

$$\langle\sigma_{ij}\rangle \equiv \lambda^*\langle\varepsilon_{kk}\rangle\delta_{ij} + 2\mu^*\langle\varepsilon_{ij}\rangle \tag{16}$$

and

$$\kappa^* \equiv \lambda^* + \frac{2}{3}\mu^*, \tag{17}$$

substitution of (15) into (14), followed by a comparison with (16) and (17), leads to the well-known relation for the effective moduli

$$\frac{\mu^*}{\mu_o} = 1 + 15(1-\nu_o)\gamma_2\phi + O(\phi^2) \tag{18a}$$

$$\frac{\lambda^*}{\lambda_o} = 1 + \frac{1}{\lambda_o}\left[\left(\kappa_o + \frac{4}{3}\mu_o\right)\gamma_1 - 10\mu_o(1-\nu_o)\gamma_2\right]\phi + O(\phi^2) \tag{18b}$$

and

$$\frac{\kappa^*}{\kappa_o} = 1 + \left(1 + \frac{4\mu_o}{3\kappa_o}\right)\gamma_1\phi + O(\phi^2). \tag{18c}$$

To extend the above to $O(\phi^2)$, it is necessary to calculate the additional dipole $S_{ij}^{(1)} \equiv S_{ij} - S_{ij}^{(0)}$ on the reference sphere due to the presence of the second particle. This quantity is a function of the relative position of the second particle and of the bulk strain, but, owing to the linearity of the problem, $S_{ij}^{(1)}$ must be linear in $\langle\varepsilon_{ij}\rangle$. Hence its general form becomes

$$S_{ij}^{(1)} = F\langle\varepsilon_{kk}\rangle\delta_{ij} + G\langle\varepsilon_{kl}\rangle\frac{r_k r_l}{R^2}\delta_{ij} + N\langle\varepsilon_{kk}\rangle\frac{r_i r_j}{R^2} + K\left\{\langle\varepsilon_{ij}\rangle - \frac{1}{3}\langle\varepsilon_{kk}\rangle\delta_{ij}\right\} + L\langle\varepsilon_{kl}\rangle\left\{\frac{r_i r_k\delta_{jl} + r_j r_k\delta_{il}}{R^2} - \frac{2}{3}\frac{r_k r_l}{R^2}\delta_{ij}\right\} + M\langle\varepsilon_{kl}\rangle\frac{r_k r_l}{R^2}\left(\frac{r_i r_j}{R^2} - \frac{1}{3}\delta_{ij}\right) \tag{19}$$

orders of magnitude of u_i' and the fluctuating part of σ_{ij} are, respectively, aE and $\mu_o E$ near an inclusion of dimension a and smaller away from the particle, where E is the magnitude of the bulk strain, the second term is negligible compared with the first if, as we have assumed all along this analysis, all bulk quantities within the composite vary appreciably over distances which are much larger than the particle dimension a. Thus we obtain

$$W = \frac{1}{2}\langle\varepsilon_{ij}\rangle\langle\sigma_{ij}\rangle.$$

The additional strain energy can then be calculated as

$$\Delta W = W - W_o = \frac{1}{2}\langle\varepsilon_{ij}\rangle\sum_{ij} = \frac{n}{2}\langle\varepsilon_{ij}\rangle\bar{S}_{ij},$$

where W_o is the strain energy in the absence of inclusions.

where the scalar functions F, G, N, K, L and M depend on the properties of the two individual phases and the dimensionless quantity $\rho = (a/R)$ with $R = |r_i|$ being the distance between the centers of two spheres of radius a. Also the reference particle is at the origin and the second particle is at r_i. The asymptotic forms of these functions for large R, easily determined through the use of method of reflections, are

$$
\begin{aligned}
F &= -\frac{80}{3}\pi a^3\mu_o(1-\nu_o)\gamma_1\gamma_2\rho^3 + \frac{100}{3}\pi a^3\mu_o(1-\nu_o)(5-4\nu_o)\gamma_2^2\rho^3 + F' \\
G &= 40\pi a^3\mu_o(1-\nu_o)\gamma_1\gamma_2\rho^3 + G' \\
N &= 40\pi a^3\mu_o(1-\nu_o)\gamma_1\gamma_2\rho^3 - 100\pi a^3\mu_o(1-\nu_0)(5-4\nu_o)\gamma_2^2\rho^3 + N' \\
K &= -200\pi a^3\mu_o(1-\nu_o)(1-2\nu_o)\gamma_2^2\rho^3 + K' \\
L &= -600\pi a^3\mu_o(1-\nu_o)\nu_o\gamma_2^2\rho^3 + L' \\
M &= 1500\pi a^3\mu_o(1-\nu_o)\gamma_2^2\rho^3 + M'
\end{aligned}
\tag{20}
$$

where F', G', N', K', L' and M' are of $O(\rho^5)$. Following then Jeffrey[15] and [I] we can rewrite (13) as

$$
\langle\sigma_{ij}\rangle = \lambda_o\langle\varepsilon_{kk}\rangle\delta_{ij} + 2\mu_o\langle\varepsilon_{ij}\rangle + nS_{ij}^{(0)} + n\int_{\mathbf{r}}\{S_{ij}^{(1)}(\mathbf{r}|0)P(\mathbf{r}|0) - S_{ij}^{(0)}[\boldsymbol{\epsilon}^{(1)}]P(\mathbf{r})\}\mathrm{d}\mathbf{r} + O(\phi^3), \tag{21}
$$

where $\varepsilon_{ij}^{(1)} \equiv \varepsilon_{ij} - \langle\varepsilon_{ij}\rangle$ denotes the additional strain at the origin due to a single particle being at $\mathbf{r}$ undergoing the applied strain $\langle\varepsilon_{ij}\rangle$ at infinity. The second term in the integral of (21) is needed to render this integral absolutely convergent[17], a choice which, as we shall show in the next section, is unique in spite of two apparently alternate possibilities.

The two functions $P(\mathbf{r}|0)$ and $P(\mathbf{r})$ in (21) refer to the probability density of finding one particle at $\mathbf{r}$ with and without a second particle being at the origin. For a suspension containing randomly distributed particles, $P(\mathbf{r})$ is simply equal to n, the number density of the particles. However, to find $P(\mathbf{r}|0)$, which depends on the microstructure of the suspension, is more difficult. Following earlier work, e.g.[17], we shall assume here that the second particle can take any possible position in the suspension with equal probability except that it cannot overlap with the test sphere. Consequently,

$$
\begin{aligned}
P(\mathbf{r}|0) &= 0 \quad \text{for} \quad R < 2a \\
&= n \quad \text{for} \quad R \geq 2a.
\end{aligned}
$$

For a sphere being at r_i and with the applied strain $\langle\varepsilon_{ij}\rangle$ at infinity, the additional strain $\varepsilon_{ij}^{(1)}$ at the origin can be obtained from Eshelby's result[24]

$$
\varepsilon_{ij}^{(1)} = \frac{1}{8\pi(1-\nu_o)}e_{kl}\frac{\partial^4\omega}{\partial r_i\partial r_j\partial r_k\partial r_l} - \frac{1}{4\pi}\left(e_{ik}\frac{\partial^2\tilde{\omega}}{\partial r_k\partial r_j} + e_{jk}\frac{\partial^2\tilde{\omega}}{\partial r_k\partial r_i}\right) - \frac{\nu_o}{4\pi(1-\nu_o)}e_{kk}\frac{\partial^2\tilde{\omega}}{\partial r_i\partial r_j}, \tag{22}
$$

where

$$
\begin{aligned}
e_{ij} &= -\frac{3(1-\nu_o)}{1+\nu_o}\gamma_1\langle\varepsilon_{kk}\rangle\delta_{ij} - 15(1-\nu_o)\gamma_2\left\{\langle\varepsilon_{ij}\rangle - \frac{1}{3}\langle\varepsilon_{kk}\rangle\delta_{ij}\right\} \\
\tilde{\omega} &= 2\pi a^2 - \frac{2\pi}{3}R^2 \qquad \text{for} \quad R \equiv |r_i| \leq a \\
&= \frac{4\pi a^2}{3}\frac{1}{R} \qquad \text{for} \quad R \geq a \\
\omega &= \pi a^4 + \frac{2}{3}\pi a^2R^2 - \frac{\pi}{15}R^4 \quad \text{for} \quad R \leq a \\
&= \frac{4\pi}{3}a^3R + \frac{4\pi}{15}\frac{a^5}{R} \qquad \text{for} \quad R \geq a.
\end{aligned}
$$

With $S_{ij}^{(0)}[\epsilon^{(1)}]$ given by (15) plus (22) and $S_{ij}^{(1)}$ by (19) we then obtain for (21)

$$\langle\sigma_{ij}\rangle = \left(A - \frac{2}{3}B\right)\langle\varepsilon_{kk}\rangle\delta_{ij} + 2B\langle\varepsilon_{ij}\rangle \tag{23}$$

with

$$A = \kappa_o\left[1 + \left(1 + \frac{4\mu_o}{3\kappa_o}\right)\gamma_1\phi + \left(1 + \frac{4\mu_o}{3\kappa_o}\right)\gamma_1{}^2\phi^2\left(1 + \frac{27\int_{2a}^{\infty}\left(F' + \frac{1}{3}G' + \frac{1}{3}N'\right)R^2\mathrm{d}R}{4\pi a^6(3\kappa_o + 4\mu_o)\gamma_1{}^2}\right)\right] \tag{24}$$

and

$$B = \mu_o\left[1 + 15(1 - \nu_o)\gamma_2\phi + 30(1 - \nu_o)(4 - 5\nu_o)\gamma_2{}^2\phi^2\left(1 + \frac{3\int_{2a}^{\infty}\left(K' + \frac{2}{3}L' + \frac{2}{15}M'\right)R^2\mathrm{d}R}{80\pi a^6\mu_o(1 - \nu_o)(4 - 5\nu_o)\gamma_2{}^2}\right)\right]. \tag{25}$$

Comparison of (23) with (16) yields

$$\mu^* = B, \quad \lambda^* = A - \frac{2}{3}B, \quad \text{and} \quad \kappa^* = A. \tag{26}$$

Thus, to find κ^* and μ^*, it is merely necessary that we evaluate the two integrals in (24) and (25) which, as shown below, can be achieved most conveniently by solving the two-sphere elasticity problem for a particular combination of bulk strains and then using (19) to determine the six functions $F, G, \ldots$ from $S_{ij}^{(1)}$ as calculated from these special solutions†.

Specifically, following Batchelor and Green[15], we define a function J as

$$J \equiv K + \frac{2}{3}L + \frac{2}{15}M,$$

and, following their lead, we define

$$I \equiv F + \frac{1}{3}G + \frac{1}{3}N.$$

In view of (20), it can be shown that

$$J = K' + \frac{2}{3}L' + \frac{2}{15}M' \quad \text{and} \quad I = F' + \frac{1}{3}G' + \frac{1}{3}N'$$

Also, although each individual function having a prime is of $O(\rho^5)$, both J and I turn out to be $O(\rho^6)$. As seen from (24) and (25), the calculation of the effective moduli requires that I and J be found. To this end, we fix the particle orientation and let the center of the second sphere be at $r_i = R\delta_{i3}$, so that (19) becomes

$$S_{ij}^{(1)} = F\langle\varepsilon_{kk}\rangle\delta_{ij} + G\langle\varepsilon_{33}\rangle\delta_{ij} + N\langle\varepsilon_{kk}\rangle\delta_{i3}\delta_{j3} + K\left(\langle\varepsilon_{ij}\rangle - \frac{1}{3}\langle\varepsilon_{kk}\rangle\delta_{ij}\right)$$
$$+ L\left(\langle\varepsilon_{i3}\rangle\delta_{j3} + \langle\varepsilon_{j3}\rangle\delta_{i3} - \frac{2}{3}\langle\varepsilon_{33}\rangle\delta_{ij}\right) + M\langle\varepsilon_{33}\rangle\left(\delta_{i3}\delta_{j3} - \frac{1}{3}\delta_{ij}\right).$$

†The ϕ^2 coefficients in (24) and (25) each consists of two parts, of which the first, $1 + (4\mu_o/3\kappa_o)\gamma_1{}^2$ and $30(1 - \nu_o)(4 - 5\nu_o)\gamma_2{}^2$, respectively, are obtained after integrating the last term in (21) from 0 to $2a$. Following Jeffrey [16], we interpret this integral as representing the effects of long-range particle interactions within the suspension which, in fact, were already determined by Walpole[8]. Moreover we can use the present results to help cast some light on the self-consistent scheme particularly in the form proposed by Kerner[2]. Specifically, if Kerner's expressions for the effective moduli are expanded in powers of ϕ, the ϕ^2 coefficients thus obtained are the same as those found here for the long-range contributions. Hence, we may tentatively conclude that, although the self-consistent scheme does account for the long-range interactions within the suspension, it apparently does not do so for the interactions among close particles. Justification for this interpretation of self-consistent schemes has been given recently by Jeffrey[25]. For a further discussion of the possible utility of self-consistent schemes, the reader is referred to Hashin[26].

We can then choose the appropriate bulk strain to solve for the functions I and J, or any combinations of the six functions F, G, N, K, L, M that can produce I and J.

First of all, the choice of the isotropic strain $\langle \varepsilon_{ij} \rangle = \delta_{ij}$ yields

$$S_{ij}^{(1)} = 3F\delta_{ij} + G\delta_{ij} + 3N\delta_{i3}\delta_{j3} + \left(\delta_{i3}\delta_{j3} - \frac{1}{3}\delta_{ij}\right)(2L + M)$$

and, therefore,

$$S_{ii}^{(1)} = 9I \tag{27}$$

from which κ^* can be determined. This case has been studied extensively in [1] for elastic inclusions. Unfortunately, it is not possible to find a single strain that can produce the function J, and it becomes necessary to solve three different problems corresponding to three independent bulk shear strains, before J can be obtained. These strains are:

(1) $\langle \varepsilon_{ij} \rangle = \delta_{i1}\delta_{j1} + \delta_{i2}\delta_{j2} - 2\delta_{i3}\delta_{j3}$, from which we obtain $K + \frac{4}{3}L + \frac{2}{3}M = \frac{1}{3}(S_{11}^{(1)} - S_{33}^{(1)})$ (28)

(2) $\langle \varepsilon_{ij} \rangle = \delta_{i1}\delta_{j2} + \delta_{i2}\delta_{j1}$, from which we obtain $K = S_{12}^{(1)}$ (29)

(3) $\langle \varepsilon_{ij} \rangle = \delta_{i2}\delta_{j3} + \delta_{i3}\delta_{j2}$, from which we obtain $K + L = S_{23}^{(1)}$. (30)

We then have that $J = (1/5) \times$ eqn (28) + $(2/5) \times$ eqn (29) + $(2/5) \times$ eqn (30). The above three cases were also examined in [1] for rigid particles and cavities.

Before proceeding with the calculation of the effective moduli of composite materials, we wish to discuss an alternative method of evaluating the bulk modulus κ^* and to examine briefly the method of normalization which was used to obtain (21) from (13).

3. METHOD OF NORMALIZATION

It is of some interest to point out here that if one adopts the normalization formalism as originally proposed by Batchelor[27], two ambiguities are encountered in the calculation of the effective bulk modulus κ^* which, as we shall show however, can be resolved if the complete formulation is used. The first apparent alternative to (21) arises from the fact that an alternative way of calculating κ^* is, of course, from its definition

$$\langle \sigma_{ii} \rangle = 3\kappa^* \langle \varepsilon_{ii} \rangle, \tag{31}$$

which can be obtained readily from (16). Physically, $\langle \varepsilon_{ii} \rangle$ refers to change of a unit volume of the composite and κ^* is the modulus of compression. Comparison of (14), with $i = j$, and (31) yields the same result as given by (18c).

To calculate the $O(\phi^2)$ term from the above definition leads, however, to some difficulties. First of all, following a standard procedure (see Ref.[17]), we rewrite (13) as

$$\langle \sigma_{ij} \rangle = \lambda_o \langle \varepsilon_{kk} \rangle \delta_{ij} + 2\mu_o \langle \varepsilon_{ij} \rangle + nS_{ij}^{(0)} + n \int S_{ij}^{(1)}(C|0)P(C|0)\mathrm{d}C \tag{32}$$

and then approximate $S_{ij}^{(1)}(C|0)$ by $S_{ij}^{(1)}(\mathbf{r}|0)$ which is the additional dipole due to the presence of a second sphere at $\mathbf{r}$. Then, if we neglect the effect of all the other spheres in C on $S_{ij}^{(1)}$, we obtain

$$\langle \sigma_{ij} \rangle = \lambda_o \langle \varepsilon_{kk} \rangle \delta_{ij} + 2\mu_o \langle \varepsilon_{ij} \rangle + nS_{ij}^{(0)} + n \int S_{ij}^{(1)}(\mathbf{r}|0)P(\mathbf{r}|0)\mathrm{d}\mathbf{r}. \tag{33}$$

However, since for larger $R \equiv |\mathbf{r}|$, $S_{ij}^{(1)}(\mathbf{r}|0)$ varies as R^{-3} and $P(\mathbf{r}|0) = n$, the integral in (33) is only conditionally convergent and gives no meaningful results unless the mode of integration is justified. One way of modifying such a conditionally convergent integral, originally proposed by Batchelor[27], is to subtract from (33) a term which averages to zero and has the same asymptotic behavior as $S_{ij}^{(1)}$ (r|0) at infinity. This is the so-called method of normalization.

On the other hand, when the trace of (33) is considered one obtains

$$\langle\sigma_{ii}\rangle = 3\kappa_o\langle\varepsilon_{ii}\rangle + nS_{ii}^{(0)} + n\int S_{ii}^{(1)}(\mathbf{r}|0)P(\mathbf{r}|0)d\mathbf{r},$$

in which the integral is absolutely convergent because $S_{ii}^{(1)}(\mathbf{r}|0)$ is only R^{-6} for large R when the applied strain is isotropic. On physical grounds it should not matter, of course, whether the full tensor of only its trace is used for the purpose of calculating κ^* and, the results should always be the same as they are for the $O(\phi)$ term. It would appear though that the method which retains the full tensors and uses the normalization technique gives an extra term which comes from the excluded volume effect or the multiparticle interactions. This apparent contradiction will now be resolved.

As remarked above, the expansion for $\bar{S}_{ij}$, i.e.

$$\begin{aligned}\bar{S}_{ij} &= \bar{S}_{ij}^{(0)} + \bar{S}_{ij}^{(1)} + \bar{S}_{ij}^{(2)} + \ldots\ldots \\ &= S_{ij}^{(0)} + \int S_{ij}^{(1)}(\mathbf{r}|0)P(\mathbf{r}|0)d\mathbf{r} + \int S_{ij}^{(2)}(\mathbf{r}_1\mathbf{r}_2|0)P(\mathbf{r}_1\mathbf{r}_2|0)d\mathbf{r}_1d\mathbf{r}_2 + \ldots\ldots \qquad (35)\end{aligned}$$

contains, in general, conditionally convergent integrals, which must be recast into absolutely convergent ones, e.g. through use of the normalization technique, before they can be evaluated. For the purpose of calculating the effective bulk modulus, however, we can employ the trace of (35), i.e.

$$\bar{S}_{ii} = S_{ii}^{(0)} + \int S_{ii}^{(1)}(\mathbf{r}|0)P(\mathbf{r}|0)d\mathbf{r} + \int S_{ii}^{(2)}(\mathbf{r}_1\mathbf{r}_2|0)P(\mathbf{r}_1\mathbf{r}_2|0)d\mathbf{r}_1d\mathbf{r}_2 + \ldots\ldots$$

in which $\bar{S}_{ii}^{(1)}$ is absolutely convergent. Nevertheless, it can be shown, see [I], that the next term $\bar{S}_{ii}^{(2)}$ and, therefore all the rest in the above expression, are only conditionally convergent, and hence, in order to evaluate $\bar{S}_{ii}$, a normalization is still needed. Thus, an extra term has to be subtracted from (34) to give the correct result.

Having resolved the first ambiguity, we now turn to the second. A way of implementing the normalization to the present problem is to follow Batchelor[27] and Jeffrey[16] and rewrite (32) as

$$\langle\sigma_{ij}\rangle = \lambda_o\langle\varepsilon_{kk}\rangle\delta_{ij} + 2\mu_o\langle\varepsilon_{ij}\rangle + nS_{ij}^{(0)} + n\int [S_{ij}^{(1)}(\mathbf{r}|0)P(\mathbf{r}|0) - X\cdot\boldsymbol{\epsilon}^{(1)}(\mathbf{r})P(\mathbf{r})]d\mathbf{r} + o(\phi^2), \qquad (36)$$

where X has to be determined so that the integral will be absolutely convergent. For most types of applied strains, X can be uniquely determined; however, as shown in [I] there are two possible choices for X when the applied strain at infinity is isotropic, i.e. $\langle\varepsilon_{ij}\rangle = \delta_{ij}$. One of them is $X = 40\pi a^3(1-\nu_o)\mu_o\gamma_2$ and the other is the fourth order tensor

$$X_{ijkl} = \frac{4}{3}\pi a^3\left(\kappa_o + \frac{4}{3}\mu_o\right)\gamma_1\delta_{ij}\delta_{kl} + 20\pi a^3(1-\nu_o)\mu_o\gamma_2\left(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk} - \frac{2}{3}\delta_{ij}\delta_{kl}\right).$$

Further analysis shows, however, that since the normalization has to be applied as well to the higher order terms of (35), one has to adopt the scheme developed by Jeffrey[17] and identify the term $X\cdot\boldsymbol{\epsilon}^{(1)}$ with $S_{ij}^{(0)}(\boldsymbol{\epsilon}^{(1)})$. This uniquely determines X to be the fourth order tensor given above and thereby leads directly to eqns (21). It should be noted that the results given in this section have been made the basis of a more general discussion by Jeffrey[25] of the possible normalization methods which have so far appeared in the literature.

Having then satisfied ourselves regarding the validity and uniqueness of (21), we shall proceed now directly from (24) and (25) and utilize the solutions for the two-particle interaction problems given in [I] and [19] to calculate the effective moduli.

4. RESULTS AND DISCUSSIONS

In [1] and in the companion paper [19], the four different two-sphere problems corresponding to the four different applied strains mentioned in Section 2 were solved by the use of the "multipole expansion" technique. Accordingly, the solution of each problem was expanded in a series of spherical harmonic functions with respect to the two centers of the spheres, and a set of equations relating the coefficients of these spherical harmonics was obtained by applying the boundary conditions of continuity of traction and displacement on the surface of the spheres. To solve for these coefficients as functions of the separation distance R between the two spheres, each of them was further expanded into a power series of the parameter $\rho = a/R$, and a recurrence formula was thereby obtained for calculating this new set of coefficients.

The first of the four problems that was solved was for the system of two elastic particles under the applied isotropic strain $\langle \varepsilon_{ij} \rangle = \delta_{ij}$ at infinity. The effective bulk modulus, as computed from (27) and (24), can then be expressed as

$$\frac{\kappa^*}{\kappa_o} = 1 + \left(1 + \frac{4\mu_o}{3\kappa_o}\right)\gamma_1\phi + \left(1 + \frac{4\mu_o}{3\kappa_o}\right)\gamma_1{}^2\phi^2[1 + (1 - 2\nu_o)H] + O(\phi^3) \tag{37}$$

where $H(\beta, \nu_p, \nu_0) = \sum_{m=6}^{\infty} \sum_{n=1}^{m-3} 2^{(3-m)}(n+1)/(m-3)C_{n(m-n-2)}$ and C_{nm} are given by the recurrence formula presented in [1]. The first few terms in the series are

$$\begin{aligned} H(\beta, \nu_p, \nu_o) = {} & \frac{5(\beta-1)}{4[2\beta(4-5\nu_o)+(7-5\nu_o)]} + \frac{21(\beta-1)}{16[2\beta(11-14\nu_o)+(13-7\nu_o)]} \\ & + \frac{25(\beta-1)^2(2-\nu_0)}{32[2\beta(4-5\nu_o)+(7-5\nu_o)]^2} + \frac{9(\beta-1)}{32[2\beta(7-9\nu_o)+(7-3\nu_o)]} \\ & + \frac{(\beta-1)^2}{256[2\beta(4-5\nu_o)+(7-5\nu_o)]}\left[\frac{105(11-4\nu_o)}{2\beta(11-14\nu_o)+(13-7\nu_o)} \right. \\ & \left. - \frac{135}{2\beta(4-5\nu_o)+(7-5\nu_o)}\right] + \frac{1}{384}\left[\frac{250(2-\nu_o)^2(\beta-1)^3}{[2\beta(4-5\nu_o)+(7-5\nu_o)]^3} \right. \\ & + \frac{165(\beta-1)}{4\beta(17-22\nu_o)+(31-11\nu_o)} \\ & \left. + \frac{25(\beta-1)^2[\beta(1+\nu_p)(1-2\nu_o)-(1-2\nu_p)(1+\nu_o)]}{[2\beta(4-5\nu_o)+(7-5\nu_o)]^2[\beta(1+\nu_p)+(1-2\nu_p)]}\right] \\ & + \ldots\ldots \end{aligned} \tag{38}$$

The values of H were then obtained by summing the infinite series using a computer. Fifty or more terms of the series were calculated. We then used the partial sums of these terms, S_m, plotted vs $1/m$ to extrapolate to $1/m = 0$, and estimated the sum of the infinite series. For a few values of ν_o, we plotted the logarithm of each term in (38) vs ln m and obtained the corresponding asymptotic form of the terms of large m, which was then used to sum the rest of the series. The results, from both methods differed at most in the second significant figure and affected the calculated $H_1 = 1 + (1 - 2\nu_o)H$, only in the third significant figure. The values of H are presented in tables in Chen's dissertation [18] for different combinations of β, ν_p and ν_o. It has been shown that the effect of ν_p, Poisson's ratio of the inclusions, on κ^* is rather insignificant. Thus the most important variables that determine H are ν_0, Poisson's ratio of the matrix, and β, the ratio of the shear moduli of the two phases. Figure 1 shows that, for $\beta > 10^2$ or $\beta < 10^{-2}$, H can be approximated by using the corresponding solutions for rigid particles or cavities (see [1]) with a maximum error of about 1% for H_1 which is the ratio of the coefficient of the ϕ^2 term calculated from the present analysis to that of Walpole's [8] as given by (4).

In [1], the other three different boundary value problems mentioned previously were also solved to yield three relations between K, L and M, i.e. eqns (28)–(30), for inclusions that are either rigid particles or cavities. These solutions can then be used to evaluate the effective shear modulus for these two limiting cases. Specifically, when the inclusions are rigid, $\beta = \mu_p/\mu_o = \infty$

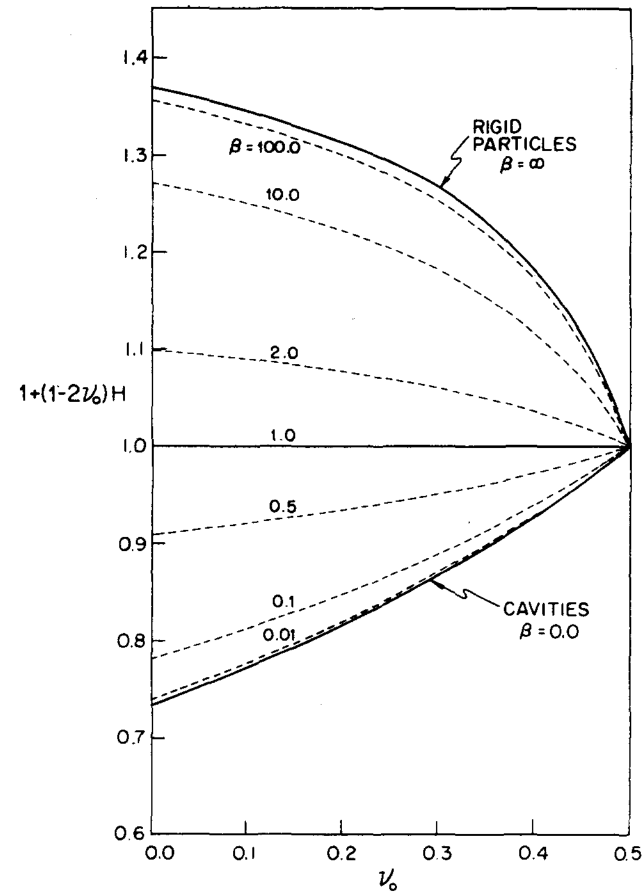

Fig. 1. Ratio of the coefficients for the ϕ^2 term of the effective bulk modulus calculated from the present work to that of Walpole's for $\nu_p = 0.25$ as a function of ν_o and β.

and $\gamma_2 = [1/2(4-5\nu_o)]$. In this case, we have

$$K + \frac{2}{3}L + \frac{2}{15}M = \frac{4\pi a^3 \mu_o(1-\nu_o)}{5(4-5\nu_o)} \sum_{m=6}^{\infty} \left(-\frac{1}{3}C^{I}_{1m} + 2B^{II}_{1m} + 2B^{III}_{1m}\right)\rho^m$$

and therefore, from (24) and (26),

$$\frac{\mu^*}{\mu_o} = 1 + \frac{15(1-\nu_o)}{2(4-5\nu_o)}\phi + \frac{15(1-\nu_o)}{2(4-5\nu_o)}\left[1 + \frac{3}{25}\sum_{m=6}^{\infty}\frac{1}{m-3}\left(-\frac{1}{3}C^{I}_{1m} + 2B^{II}_{1m} + 2B^{III}_{1m}\right)\left(\frac{1}{2}\right)^{m-3}\right]\phi^2 + O(\phi^3). \tag{39}$$

On the other hand, when the inclusions are cavities, $\beta = 0$ and $\gamma_2 = \frac{-1}{7-5\nu_o}$. Consequently,

$$K + \frac{2}{3}L + \frac{2}{15}M = \frac{8\pi a^3 \mu_o(1-\nu_o)}{5(7-5\nu_o)} \sum_{m=6}^{\infty} \left(-\frac{1}{3}C^{I}_{1m} + 2B^{II}_{1m} + 2B^{III}_{1m}\right)\rho^m$$

and

$$\frac{\mu^*}{\mu_o} = 1 - \frac{15(1-\nu_o)}{7-5\nu_o}\phi + \frac{30(1-\nu_o)(4-5\nu_o)}{(7-5\nu_o)^2}\left[1 + \frac{3(7-5\nu_o)}{50(4-5\nu_o)} \times \sum_{m=6}^{\infty}\frac{1}{m-3}\left(-\frac{1}{3}C^{I}_{1m} + 2B^{II}_{1m} + 2B^{III}_{1m}\right)\left(\frac{1}{2}\right)^{m-3}\right]\phi^2 + O(\phi^3). \tag{40}$$

The superscripts I, II and III in the above equations denote the solutions from the three systems (28)–(30) respectively. The summation of the infinite series in eqns (39) and (40) was carried out using the computer to calculate the terms up to $m = 80$ and the sums were obtained by plotting the partial sum S_m vs $1/m$ and extrapolating to $1/m = 0$. The final results are believed to have a maximum deviation of $\pm 0.5\%$. In some of the calculations, the sum of the first eighty terms gave a good enough approximation and no extrapolation was performed. The results for different values of Poisson's ratio ν_o for the matrix are plotted in Fig. 2 for rigid particles and cavities, respectively.

In a recent paper by Willis and Acton[12] the effective elastic moduli were also calculated to $O(\phi^2)$ through the use of an integral equation formulation. These authors, however, made use only of the far field solution, $O[(a/R)^6]$, for the two-sphere interaction problem and so their expression for the $O(\phi^2)$ coefficient contains only the first ($m = 6$) term of the infinite series in (37), (39) and (40).

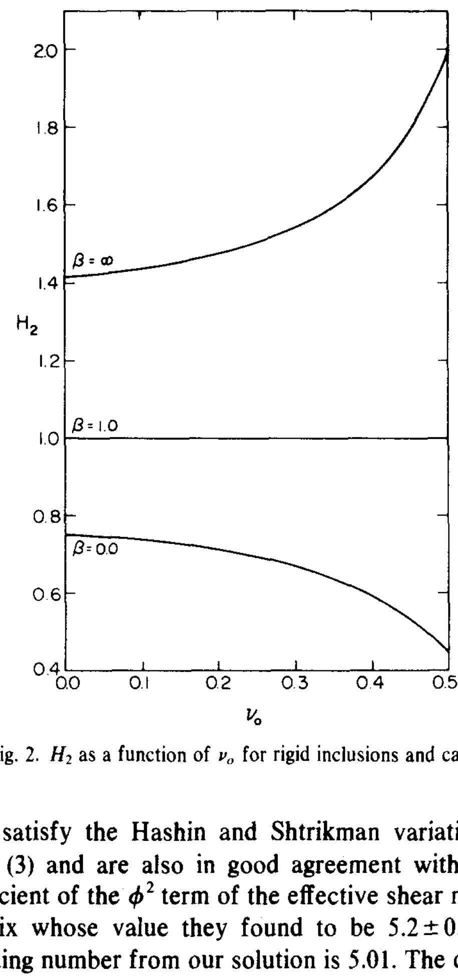

Fig. 2. H_2 as a function of ν_o for rigid inclusions and cavities.

The present results satisfy the Hashin and Shtrikman variational bounds (1a) and (1b), Miller's bounds for κ^* (3) and are also in good agreement with Batchelor and Green's[15] calculation for the coefficient of the ϕ^2 term of the effective shear modulus for rigid particles in an incompressible matrix whose value they found to be 5.2 ± 0.3 for randomly distributed spheres. The corresponding number from our solution is 5.01. The difference is probably due to the fact that Batchelor and Green[15] did not treat their interaction term exactly. The exact calculation for the interaction between two spheres is rather tedious to perform because, as seen in Fig. 3, the quantity $\hat{J} = [(4-5\nu_o)J/20\pi a^3(1-\nu_o)\mu_o]$ as a function of the separation distance between the spheres changes very rapidly when the two spheres are almost in contact with each other. In fact, a more detailed calculation of $\hat{K}$, $\hat{K}+\hat{L}$ and $\hat{K}+(4/3)\hat{L}+(2/3)\hat{M}$ for different values of (a/R) shows that this rapid change occurs in the term $\hat{K}+\hat{L}$, where the caret denotes the appropriate quantity divided by $[20\pi a^3(1-\nu_o)\mu_o/(4-5\nu_o)]$. Thus a series having eighty terms gives convergent values for $\hat{K}+(4/3)\hat{L}+(2/3)\hat{M}$, $\hat{K}+\hat{L}$ when $(R/a) \geq 2.10$. For $(R/a) \leq 2.05$, the series for $\hat{K}$ also converges but for $\hat{K}+(4/3)\hat{L}+(2/3)\hat{M}$ and $\hat{K}+\hat{L}$, the sum corresponding to an infinite number of terms can only be obtained by extrapolation.

Comparison of our results for these functions with the exact values can be made when two spheres are in contact, i.e. when $(R/a) = 2.0$. Using Wakiya's[29] solution, Batchelor and

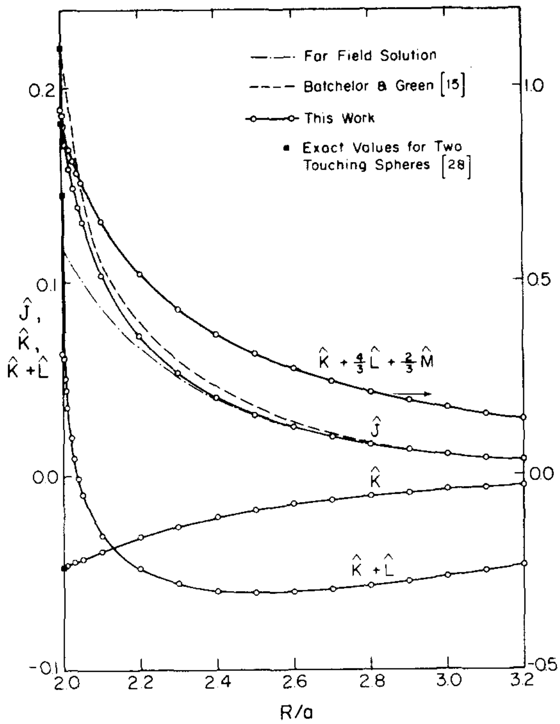

Fig. 3. Behavior of $\hat{K}$, $\hat{K}+\hat{L}$, $\hat{J}$ and $\hat{K}+(4/3)\hat{L}+(2/3)\hat{M}$ as functions of the separation distance between the spheres.

Green[28] showed that, in that case,

$$\hat{K} = -0.0472$$
$$\hat{K}+\hat{L} = 0.1456$$
$$\hat{K}+\frac{4}{3}\hat{L}+\frac{2}{3}\hat{M} = 0.9104$$
$$\hat{J} = 0.2214$$

whereas, our calculations give

$$\hat{K} = -0.04724$$
$$\hat{K}+\hat{L} = 0.0630$$
$$\hat{K}+\frac{4}{3}\hat{L}+\frac{2}{3}\hat{M} = 0.9135$$
$$\hat{J} = 0.1890.$$

Thus, the results for $\hat{K}+(4/3)\hat{L}+(2/3)\hat{M}$ are accurate to three significant figures in the range $2.0 \le (R/a) \le 2.05$. On the other hand, the errors involved in the calculation of $\hat{K}+\hat{L}$ are not certain owing to the rapid change of the function as (R/a) approaches 2.0; it is believed though that, in the range $2.0025 \le (R/a) \le 2.05$, the results are still accurate to three significant figures but, as $(R/a) \to 2.0$, the accuracy decreases rapidly to only one significant figure ($\hat{K}+\hat{L} \sim 0.1$).

Using the exact values at $(R/a) = 2.0$, a curve fit for ε, defined as $\varepsilon = (R/a) - 2.0$, in the range $0 \le \varepsilon \le 0.05$ gives

$$\begin{aligned} \hat{J} &= 0.2214 + 0.20\,(\ln\varepsilon)^{-1} - 0.55\varepsilon + o(\varepsilon) \\ \hat{K} &= -0.04724 + 0.0878\,\varepsilon + o(\varepsilon) \\ \hat{K}+\hat{L} &= 0.1456 + 0.51\,(\ln\varepsilon)^{-1} + 0.30\,\varepsilon + o(\varepsilon) \\ \hat{K}+\frac{4}{3}\hat{L}+\frac{2}{3}\hat{M} &= 0.9104 - 3.50\,\varepsilon + o(\varepsilon) \end{aligned} \tag{41}$$

which are depicted in Fig. 4 for $\varepsilon \to 0$. As shown in [1], the form of (41) can be justified rigorously using "lubriction theory".

Although the present numerical calculation for the function J loses accuracy when the particles almost touch, this does not affect the accuracy of the computed coefficient of the $O(\phi^2)$ term in the effective shear modulus which, being dependent only on the integral $\int_{2a}^{\infty} J(R) R^2 dR$, is very insensitive to the exact shape of the function J when $(R/a) = 2$.

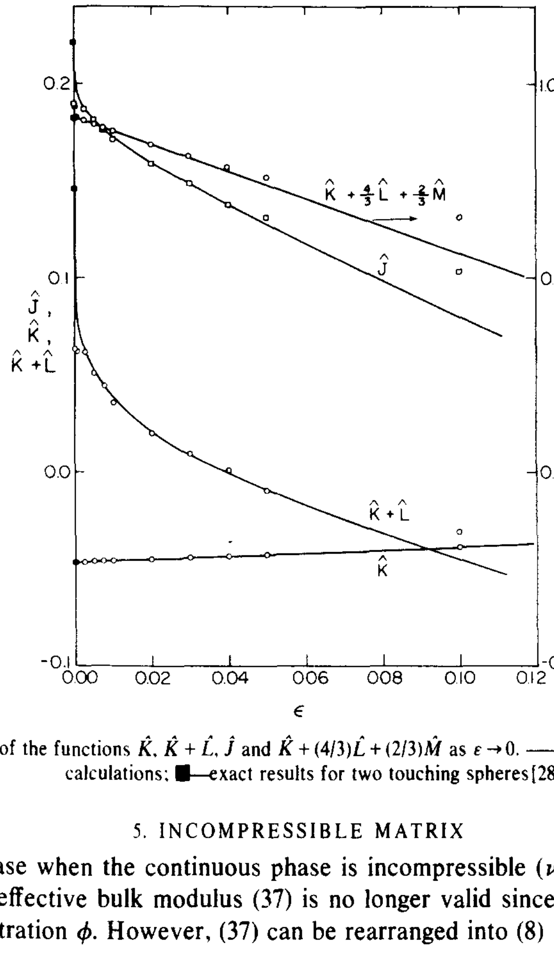

Fig. 4. Behavior of the functions $\hat{K}$, $\hat{K}+\hat{L}$, $\hat{J}$ and $\hat{K}+(4/3)\hat{L}+(2/3)\hat{M}$ as $\varepsilon \to 0$. ——, eqn (41); ○—present calculations; ■—exact results for two touching spheres [28].

5. INCOMPRESSIBLE MATRIX

In the special case when the continuous phase is incompressible ($\nu_o = 0.5$ and $\kappa_o = \infty$), the expression for the effective bulk modulus (37) is no longer valid since the series diverges for any volume concentration ϕ. However, (37) can be rearranged into (8)

$$\kappa^* = \kappa_o\left[1 - \left(1 + \frac{4\mu_o}{3\kappa_o}\right)\gamma_1\phi + \left(1 + \frac{4\mu_o}{3\kappa_o}\right)\frac{4\mu_o}{3\kappa_o}\gamma_1^2\left(1 - \frac{1+\nu_o}{2}H\right)\phi^2 + O(\phi^3)\right]^{-1}$$

which is equivalent to (37) when $\phi \ll 1$, but is more useful when $\nu_o \sim 0.5$ and κ_o is very large. Specifically, in the limit as $\kappa_o \to \infty$ and $\nu_o \to 0.5$, we have that $\kappa_o^{-1}\gamma_1 \to -3/(3\kappa_p + 4\mu_o)$ and

$$\kappa^* = \left[\frac{3}{3\kappa_p + 4\mu_o}\phi + \frac{3\mu_o}{(3\kappa_p + 4\mu_o)^2}(4 - 3H)\phi^2 + O(\phi^3)\right]^{-1},$$

which can be immediately recast into (9).

It is observed that when the particles are also incompressible ($\kappa_p = \infty$), (9) gives $\kappa^* = \infty$ since the composite should be incompressible as well. When the two phases have the same shear modulus, we have $H = 0$ and therefore, from (9),

$$\kappa^* = \frac{3\kappa_p + 4\mu_o}{3\phi} - \frac{4}{3}\mu_o + O(\phi),$$

which is again in agreement with (3), Hill's equation for $\kappa_o = \infty$. For the particular case of cavities in an incompressible material, (9) becomes

$$\kappa^* = \frac{4\mu_o}{3\phi} - 1.733\mu_o + O(\phi). \tag{42}$$

Since $\kappa^* = \lambda^* + (2/3)\mu^*$ and $\mu^* = \mu_o + O(\phi)$, we can also obtain for the effective Lamé constant

$$\lambda^* = \frac{4\mu_o}{3\phi} - 2.399\mu_o + O(\phi). \tag{43}$$

It is to this last case that the analogy between solid mechanics and fluid mechanics applies. In fluid mechanics, the stress-rate of strain relation is given as

$$\sigma_{ij} = -p\delta_{ij} + \mu'\varepsilon_{kk}\delta_{ij} + 2\mu\varepsilon_{ij}$$

where p is the pressure, μ the shear viscosity and μ' the second coefficient of viscosity (or expansion viscosity) of the fluid. Lamé's constant λ is therefore analogous to μ' and the counterpart of the bulk modulus κ is the bulk viscosity defined as $\zeta = \mu' + (2/3)\mu$. Now, a composite consisting of cavities in an incompressible material is equivalent to an incompressible fluid containing air bubbles. Taylor[20] studied this case and obtained for the second coefficient of viscosity

$$\mu'^* = \frac{4\mu}{3\phi} + O(1). \tag{44}$$

Later Davies[30] showed that the singularity $\mu'^* \to \infty$ when $\phi \to 0$, can be eliminated by introducing compressibility in the surrounding fluid. However, it should be pointed out that Davies' results does not correspond to any of our findings because the analogy between solid mechanics and fluid mechanics is valid only in the limiting case when the fluid is incompressible. At any rate it is clear that (42) extends Taylor's results by taking into account two-particle interactions.

As concerns the effective shear modulus, eqn (7) is valid when $\nu_o = 0.5$. However, the analogy between the effective shear modulus of a composite and the effective viscosity of a suspension will hold only when the concentration ϕ is very small and the interactions between particles are negligible because when particle interactions have to be considered, the statistical properties of particle arrangement in the two cases are different. Thus, in the fluid suspension case, the bulk motion will greatly affect the probability density function $P(\mathbf{r}|0)$, whereas in the solid composite the infinitesimal strain applied has negligible effect on the arrangement of the particles. Thus the analogy between fluid and solid mechanics does not hold in general for (7).

Acknowledgements—This work was supported in part by N.S.F. grants GK-36515X and GK-43608. The authors would also like to thank Dr. D. J. Jeffrey for his useful comments on this work.

REFERENCES

1. J. M. Dewey, The elastic constants of materials loaded with non-rigid fillers. *J. Appl. Phys.* **18**, 578 (1947).
2. E. H. Kerner, The elastic and thermal-elastic properties of composite media. *Proc. Phys. Soc.* B69, 808 (1956).
3. Z. Hashin, The moduli of an elastic solid containing spherical particles of another elastic material. IUTAM *Non-Homogeneity in Elasticity and Plasticity* Symposium, Warsaw. (Edited by W. Olszak). p. 463 (1959).
4. Z. Hashin and S. Shtrikman, A variational approach to the theory of the elastic behavior of multiphase materials. *J. Mech. Phys. Solids* **11**, 127 (1963).
5. R. Hill, Elastic properties of reinforced solids: some theoretical principles. *J. Mech. Phys. Solids* **11**, 357 (1963).
6. Z. Hashin, Theory of mechanical behavior of heterogeneous media. *Appl. Mech. Rev.* **17**, 1 (1964).
7. L. J. Walpole, On bounds for the overall elastic moduli of inhomogeneous system, I. *J. Mech. Phys. Solids* **14**, 151 (1966).
8. L. J. Walpole, The elastic behavior of a suspension of spherical particles. *Q. J. Mech. Appl. Math.* **25**, 153 (1972).
9. J. C. Smith, Correction and extension of Van Der Poel's method for calculating the shear modulus of a particulate composite. *J. Res.* **78A**, 355 (1974).
10. J. C. Smith, Simplification of Van Der Poel's formula for the shear modulus of a particulate composite. *J. Res.* **79A**, 419 (1975).
11. S. Boucher, Differential scheme of approximation to compute effective moduli. *Revue M.* **22**, N1 (1975).

12. J. R. Willis and J. R. Acton, The overall elastic moduli of a dilute suspension of spheres. *Q. J. Mech. Appl. Math.* **29**, 163 (1976).
13. Z. Hashin, The elastic moduli of heterogeneous materials. *J. Appl. Mech.* **29**, 143 (1962).
14. M. N. Miller, Bounds for effective bulk modulus of heterogeneous materials. *J. Math. Phys.* **10**, 2005 (1969).
15. G. K. Batchelor and J. T. Green, The determination of the bulk stress in a suspension of spherical particles to order c^2. *J. Fluid Mech.* **56**, 401 (1972).
16. D. J. Jeffrey, Conduction through a random suspension of spheres. *Proc. R. Soc. Lond.* **A335**, 355 (1973).
17. D. J. Jeffrey, Group expansion for the bulk properties of a statistically homogeneous random suspensions. *Proc. R. Soc. Lond.* **A338**, 503 (1974).
18. H. S. Chen, (referred to as [I] in the text) "The Effective Properties of Composite Materials and Suspension". Ph.D. Dissertation, Stanford University (1977).
19. H. S. Chen and A. Acrivos, The solution of the equations of linear elasticity for an infinite region containing two spherical inclusions. *Int. J. Solids Structures* **14**, 331–348 (1978).
20. G. I. Taylor, The two coefficients of viscosity for an incompressible fluid containing air bubbles. *Proc. R. Soc. Lond.* **A226**, 34 (1954).
21. W. B. Russel and A. Acrivos, On the effective moduli of composite materials: slender rigid inclusions at dilute concentrations. *J. Appl. Math. & Phys. (ZAMP)* **23**, 434 (1972).
22. I. S. Sokolnikoff, *Mathematical Theory of Elasticity*, p. 71. McGraw-Hill, New York (1956).
23. J. N. Goodier, Concentration of stress around spherical and cylindrical inclusions and flaws. *Trans ASME* **55**, 39 (1933).
24. J. D. Eshelby, Elastic inclusions and inhomogeneities, In *Progress in Solid Mechanics* **2** (Edited by I. N. Sneddon and R. Hill), p. 89. Interscience, New York (1961).
25. D. J. Jeffrey, The physical significance of non-convergent integrals in expressions for effective transport properties. *Proc. 2nd Int. Conf. Cont. Models Discrete Sys.*, Montreal (1977).
26. Z. Hashin, Assessment of the self consistent scheme approximation: conductivity of particular composites. *J. Composite Mat.* **2**, 284 (1968).
27. G. K. Batchelor, Sedimension in a dilute dispersion of spheres. *J. Fluid Mech.* **52**, 245 (1972).
28. G. K. Batchelor and J. T. Green, The hydrodynamic interaction of two small freely-moving spheres in a linear flow field. *J. Fluid Mech.* **56**, 375 (1972).
29. S. Wakiya, Slow motion in shear flow of a doublet of two spheres in contact. *J. Phys. Soc. Japan* **31**, 1581 (1971).
30. R. O. Davies, A note on Sir Geoffrey Taylor's paper. *Proc. R. Soc. Lond.* **A226**, 39 (1954).