Mean-field approximation to the effective elastic moduli of a solid suspension of spheres

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We study the effective shear and bulk moduli of a solid suspension of spheres with a spherically symmetric elastic profile. A mean-field approximation is derived which corresponds to the Lorentz local field in the theory of dielectrics. Thus the approximate expressions for the effective shear and bulk moduli are the analogs of the Clausius-Mossotti equation for the effective dielectric constant. For the case of uniform spheres the expressions are closely related to the Hashin-Shtrikman bounds. We show that the mean-field expression may be corrected systematically for correlations in the sphere positions on the basis of cluster expansions derived by statistical methods.

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I. INTRODUCTION

The calculation of the effective elastic properties of a solid composite is an important problem of material science. In this article, we study a solid suspension consisting of spheres with a spherically symmetric elastic profile embedded in a uniform and isotropic matrix. We derive mean-field expressions for the effective shear and bulk moduli of the suspension by a method analogous to that used by Lorentz [1] for the derivation of the Clausius-Mossotti formula in the theory of dielectrics [2,3]. For the special case of uniform spheres the mean-field expressions reduce to the Hashin-Shtrikman bounds [4], except when the shear modulus of the spheres is larger than and the bulk modulus is smaller than that of the matrix, or vice versa.

In the statistical theory of dielectrics it is known [5,6] how the Clausius-Mossotti formula may be obtained as an approximation from exact cluster expansions, which have been derived by statistical methods. The cluster expansions provide exact expressions for the effective linear transport properties of suspensions. They allow a systematic calculation of corrections to the mean-field expressions due to correlations in the sphere positions. We show in the following how, in the elastic problem, the mean-field expressions may be obtained from the direct cluster expansion [7], as well as from the renormalized cluster expansion [8]. The latter is the most suitable for the calculation of the correction terms.

II. ELASTIC SUSPENSION

We consider N identical spherical particles of radius a, centered at $\mathbf{R}_1,\ldots,\mathbf{R}_N$, and embedded in an isotropic elastic medium with uniform shear modulus μ_1 and bulk modulus κ_1. The inclusions are assumed to have a spherically symmetric shear modulus $\mu_2(s)$ and bulk modulus $\kappa_2(s)$, so that the elastic moduli in the inclusions are given by

$$\mu(\mathbf{r})=\mu_2(|\mathbf{r}-\mathbf{R}_j|),\quad \kappa(\mathbf{r})=\kappa_2(|\mathbf{r}-\mathbf{R}_j|)\quad \text{for}\,|\mathbf{r}-\mathbf{R}_j|\le a,\quad j=1,\ldots,N\ . \tag{2.1}$$

The linear equation of elastic equilibrium for the displacement field $\mathbf{u}(\mathbf{r})$ can be written as

$$\nabla\cdot\boldsymbol{\sigma}=-\mathbf{F}_0\ , \tag{2.2}$$

where $\boldsymbol{\sigma}(\mathbf{r})$ is the stress tensor and $\mathbf{F}_0(\mathbf{r})$ is an applied force density. The stress tensor is given by the local constitutive equation

$$\boldsymbol{\sigma}=2\mu(\nabla\mathbf{u})^0+\kappa(\nabla\cdot\mathbf{u})\mathbf{1}\ , \tag{2.3}$$

where $\mu(\mathbf{r})$ is the local shear modulus, $\kappa(\mathbf{r})$ is the local bulk modulus, and $(\nabla\mathbf{u})^0$ is the symmetric traceless part of the strain tensor defined by

$$((\nabla u)^0)_{\alpha\beta}=\tfrac{1}{2}(\partial_\alpha u_\beta+\partial_\beta u_\alpha)-\tfrac{1}{3}(\nabla\cdot\mathbf{u})\delta_{\alpha\beta}\ . \tag{2.4}$$

The last term in Eq. (2.3) corresponds to the local pressure

$$p=-\kappa\nabla\cdot\mathbf{u}\ . \tag{2.5}$$

We define the difference functions

$$\tilde{\mu}(\mathbf{r})=\mu(\mathbf{r})-\mu_1,\quad \tilde{\kappa}(\mathbf{r})=\kappa(\mathbf{r})-\kappa_1\ , \tag{2.6}$$

which vanish outside the inclusions, and introduce the stress $\mathbf{s}$ induced by the inclusions as

$$\mathbf{s}=2\tilde{\mu}(\nabla\mathbf{u})^0+\tilde{\kappa}(\nabla\cdot\mathbf{u})\mathbf{1}\ . \tag{2.7}$$

The equilibrium equation (2.2) may then be rewritten as

$$\mu_1\nabla^2\mathbf{u}+\left[\tfrac{1}{3}\mu_1+\kappa_1\right]\nabla(\nabla\cdot\mathbf{u})=-\mathbf{F}_0-\mathbf{F}\ , \tag{2.8}$$

with the induced force density $\mathbf{F}(\mathbf{r})$ given by

$$\mathbf{F}(\mathbf{r})=\sum_{j=1}^{N}\mathbf{F}_j(\mathbf{r})\ , \tag{2.9}$$

where

$$\mathbf{F}_j(\mathbf{r})=\Theta(a-|\mathbf{r}-\mathbf{R}_j|)\nabla\cdot\mathsf{s} \tag{2.10}$$

is the contribution from the jth inclusion. Here $\theta(r)$ is the Heaviside step function. It follows from Eq. (2.2) that $\mathbf{F}$ is the divergence of a tensor field. We may interpret Eq. (2.8) as the equilibrium equation for a uniform medium with moduli μ_1,κ_1 on which the force density $\mathbf{F}_0+\mathbf{F}$ acts locally.

The formal solution to Eq. (2.8) is given by

$$\mathbf{u}(\mathbf{r})=\mathbf{u}_0(\mathbf{r})+\int \mathsf{G}_0(\mathbf{r},\mathbf{r}')\cdot\mathbf{F}(\mathbf{r}')d\mathbf{r}' , \tag{2.11}$$

where $\mathbf{u}_0(\mathbf{r})$ is the solution in the absence of inclusions and $\mathsf{G}_0(\mathbf{r},\mathbf{r}')$ is the Green's function. For an infinite and unbounded medium the Green's function is translationally invariant, so that

$$\mathsf{G}_0(\mathbf{r},\mathbf{r}')=\mathsf{G}_0(\mathbf{r}-\mathbf{r}') . \tag{2.12}$$

By a Fourier transformation, one finds the explicit expression [9]

$$\mathsf{G}_0(\mathbf{r})=\frac{1}{8\pi\mu_1}\left[\frac{\xi+1}{\xi+4}\frac{\mathsf{1}+\hat{\mathbf{r}}\hat{\mathbf{r}}}{r}+\frac{6}{\xi+4}\frac{\mathsf{1}}{r}\right] , \tag{2.13}$$

where ξ is the ratio

$$\xi=3\kappa_1/\mu_1 . \tag{2.14}$$

The first term in Eq. (2.13) is proportional to the Oseen tensor known from low-Reynolds-number hydrodynamics [10].

By substitution of the formal solution (2.11) into Eqs. (2.7) and (2.10), one obtains a self-consistent equation for the induced force density $\mathbf{F}$. The force density exerted by inclusion j on the medium is given by

$$\mathbf{F}_j(\mathbf{r})=\int \mathsf{M}(j;\mathbf{r},\mathbf{r}')\cdot\mathbf{u}_j^a(\mathbf{r}')d\mathbf{r}' , \tag{2.15}$$

where the integral kernel $\mathsf{M}(j;\mathbf{r},\mathbf{r}')$ describes the response of sphere j to an incident field, and $\mathbf{u}_j^a(\mathbf{r})$ is the displacement field acting on sphere j. The latter is given by

$$\mathbf{u}_j^a(\mathbf{r})=\mathbf{u}_0(\mathbf{r})+\sum_{k(\neq j)}\mathbf{u}_k(\mathbf{r}) , \tag{2.16}$$

where $\mathbf{u}_k(\mathbf{r})$ in turn is given by

$$\mathbf{u}_k(\mathbf{r})=\int \mathsf{G}_0(\mathbf{r},\mathbf{r}')\cdot\mathbf{F}_k(\mathbf{r}')d\mathbf{r}' . \tag{2.17}$$

The above equations may be solved by iteration and hence the induced force density $\mathbf{F}(\mathbf{r})$ can be found from the displacement field $\mathbf{u}_0(\mathbf{r})$ for any configuration of particles.

For any configuration $(\mathbf{R}_1,\ldots,\mathbf{R}_N)$ and applied force density $\mathbf{F}_0(\mathbf{r})$, the above equations provide a formal solution for the displacement field and for the induced force density. Assuming that the probability distribution of configurations is known, one can perform an averaging over the positions of the spheres. This leads to a macroscopic equation for the average displacement field and to a constitutive equation for the average force density.

From Eq. (2.8) we find the average equation

$$\mu_1\nabla^2\langle\mathbf{u}\rangle+\left[\tfrac{1}{3}\mu_1+\kappa_1\right]\nabla(\nabla\cdot\langle\mathbf{u}\rangle)=-\mathbf{F}_0-\langle\mathbf{F}\rangle . \tag{2.18}$$

We have assumed that the applied force density $\mathbf{F}_0(\mathbf{r})$ is independent of the configuration of scatterers. The average induced force density $\langle\mathbf{F}(\mathbf{r})\rangle$ may be expressed in terms of the average displacement field $\langle\mathbf{u}(\mathbf{r})\rangle$ by means of a linear integral kernel which has a relatively short range. For a field $\langle\mathbf{u}(\mathbf{r})\rangle$ of slow spatial variation the integral operator may be expressed with a local elastic tensor. For systems which on average are locally uniform and isotropic the average equation in the bulk of the medium takes the form

$$\mu_{\text{eff}}\nabla^2\langle\mathbf{u}\rangle+\left[\tfrac{1}{3}\mu_{\text{eff}}+\kappa_{\text{eff}}\right]\nabla(\nabla\cdot\langle\mathbf{u}\rangle)=-\mathbf{F}_0 \tag{2.19}$$

with effective moduli μ_{eff} and κ_{eff}.

In the statistical-mechanical derivation of Eq. (2.19) one considers a probability distribution $W(1,\ldots,N)$ for which the particle centers are localized inside a volume Ω and for which the average density becomes uniform in the thermodynamic limit $N\to\infty$, $\Omega\to\infty$ at constant $n=N/\Omega$. All higher-order distribution functions must become translationally invariant and isotropic. The procedure leads to well-defined statistical expressions for the effective moduli μ_{eff} and κ_{eff}, which are independent of the shape of the volume Ω. We shall return to the statistical theory in Sec. V.

To lowest order in the density, the effective moduli are given by [11–13]

$$\mu_{\text{eff}}=\mu_1+[\mu]\phi\mu_1+O(\phi^2), \quad \kappa_{\text{eff}}=\kappa_1+[\kappa]\phi\kappa_1+O(\phi^2) , \tag{2.20}$$

where $\phi=4\pi na^3/3$ is the volume fraction, and the intrinsic moduli $[\mu]$ and $[\kappa]$ follow from the solution of a single-particle problem [14]. The expressions (2.20) are useful only for very dilute suspensions. At higher volume fractions, correction terms involving elastic interactions between inclusions must be considered.

III. MEAN-FIELD THEORY

In this section we derive expressions for the effective moduli μ_{eff} and κ_{eff} in the mean-field approximation by following the approach first developed by Clausius [15], Mossotti [16], and Lorentz [1,17] in the theory of dielectrics. In the case of dielectrics, the average local electric field acting on a selected particle is expressed in terms of both the average Maxwell field and the average polarization. In the mean-field approximation, the correlations between the spheres, apart from the nonoverlap condition, are neglected. As a result the expression for the effective dielectric constant depends only on the volume fraction occupied by the spheres and contains no further details of the geometry of the microstructure. By following the same approach in the elastic case, we shall find similar expressions for the effective elastic moduli.

We begin by recalling the Lorentz derivation [1,17] of the effective dielectric constant ϵ_{eff} of an isotropic suspension of spherically symmetric polarizable particles embedded in a medium with a uniform dielectric constant

ϵ_1. Lorentz considered a macroscopic sample in an applied electric field. The shape of the sample need not be specified. One imagines a sphere that is centered at a selected particle and is sufficiently large to contain many neighboring particles. Lorentz assumed that the effect of particles outside the sphere on the central particle can be described by the average polarization $\langle \mathbf{P}(\mathbf{r})\rangle$. He showed that, in a region where both the average Maxwell field $\langle \mathbf{E}(\mathbf{r})\rangle$ and the average polarization $\langle \mathbf{P}(\mathbf{r})\rangle$ are slowly varying, the contribution from these particles to the average local field acting on the selected particle is given by

$$\mathbf{E}_L = \langle \mathbf{E}\rangle + \frac{4\pi}{3\epsilon_1}\langle \mathbf{P}\rangle \ . \tag{3.1}$$

The contribution from the particles inside the sphere averages to zero to a good approximation, provided the distribution is isotropic. Thus the average polarization is well approximated by

$$\langle \mathbf{P}\rangle = n\alpha \mathbf{E}_L \ , \tag{3.2}$$

where n is the local density and α is the electric polarizability of a particle. The effective dielectric constant is defined from the equations

$$\langle \mathbf{D}\rangle = \epsilon_1\langle \mathbf{E}\rangle + 4\pi\langle \mathbf{P}\rangle, \quad \langle \mathbf{D}\rangle = \epsilon_{\text{eff}}\langle \mathbf{E}\rangle \ . \tag{3.3}$$

Combining these with Eqs. (3.1) and (3.2) one finds the Clausius-Mossotti (CM) formula

$$\frac{\epsilon_{\text{eff}}-\epsilon_1}{\epsilon_{\text{eff}}+2\epsilon_1} = \frac{4\pi}{3\epsilon_1} n\alpha \ . \tag{3.4}$$

The formula may be cast in the alternative form

$$\epsilon_{\text{eff}} = \epsilon_1 + \frac{[\epsilon]\phi}{1-\frac{1}{3}[\epsilon]\phi}\epsilon_1 \ , \tag{3.5}$$

where $[\epsilon]$ is the intrinsic dielectric constant of a single particle, defined by

$$[\epsilon] = \frac{3\alpha}{\epsilon_1 a^3} \ . \tag{3.6}$$

Spheres with uniform dielectric constant ϵ_2 have a polarizability

$$\alpha = \epsilon_1 \frac{\epsilon_2-\epsilon_1}{\epsilon_2+2\epsilon_1} a^3 \ . \tag{3.7}$$

In that case the CM formula reduces to the Maxwell-Garnett formula [18]. The CM formula yields an excellent approximation to the effective dielectric constant of a suspension of spheres. Computer simulations [19,20] have shown that correction terms are relatively small at least up to a volume fraction $\phi = 0.5$.

We follow the same approach in the case of elastic suspensions. First of all we must find the equivalent of the polarization. The multipole moments of the force density of a selected sphere j are defined by [21]

$$\mu_j^{(n)} = \frac{1}{(n-1)!}\int \mathbf{F}_j(\mathbf{r})(\mathbf{r}-\mathbf{R}_j)^{n-1} d\mathbf{r} \ , \tag{3.8}$$

where $\mathbf{a}^n$ indicates the direct tensor product of n vectors $\mathbf{a}$. The moment for $n=1$ vanishes, since the inclusion can exert no force, and the moment for $n=2$ is symmetric, since the inclusion can exert no torque. The corresponding force multipole density of order n is defined by

$$\mathsf{F}^{(n)} = \sum_{j=1}^{N} \mu_j^{(n)}\delta(\mathbf{r}-\mathbf{R}_j) \ . \tag{3.9}$$

The average induced forced density may be effectively replaced by the multipole expansion

$$\langle \mathbf{F}(\mathbf{r})\rangle = \sum_{n=2}^{\infty} (-1)^{n-1}\nabla^{n-1} \colon \langle \mathsf{F}^{(n)}(\mathbf{r})\rangle \ . \tag{3.10}$$

In situations with slow spatial variation it is sufficient to consider only the dipolar term $\langle \mathsf{F}^{(2)}(\mathbf{r})\rangle$. This is the analogue of the electric polarization in the dielectric case. We separate the symmetric second rank tensor into two parts:

$$\langle \mathsf{F}^{(2)}\rangle = \langle (\mathsf{F}^{(2)})^0\rangle + \tfrac{1}{3}\mathbf{1}\,\mathrm{Tr}\langle \mathsf{F}^{(2)}\rangle \ . \tag{3.11}$$

The average stress in the suspension may be approximated by

$$\langle \sigma\rangle = 2\mu_1(\nabla\langle \mathbf{u}\rangle)^0 - \langle (\mathsf{F}^{(2)})^0\rangle + \mathbf{1}(\kappa_1\nabla\cdot\langle \mathbf{u}\rangle - \tfrac{1}{3}\mathrm{Tr}\langle \mathsf{F}^{(2)}\rangle) \ . \tag{3.12}$$

Thus the first term in Eq. (3.11) contributes to the average shear stress, and the second term contributes to the average pressure.

Next we consider the response of an isolated inclusion centered at $\mathbf{R}$ to an almost uniform acting displacement field. The relevant moments are

$$(\mu^{(2)})^0 = -\frac{8\pi a^3}{3}[\mu]\mu_1(\nabla \mathbf{u})^0|_{\mathbf{R}}, \qquad \mathrm{Tr}\mu^{(2)} = -4\pi a^3[\kappa]\kappa_1\nabla\cdot\mathbf{u}|_{\mathbf{R}} \ . \tag{3.13}$$

At low density the acting field may be replaced by the average field. This leads to the low-density expressions (2.20). At higher density one must consider corrections to the local field. Again we draw a large Lorentz sphere around the selected inclusion and calculate the contribution from the particles outside the sphere in the continuum approximation. The influence of the average second-rank force density $\langle \mathsf{F}^{(2)}\rangle$ is mediated by the Green's function given in Eq. (2.13). The second term in this expression is just a Coulomb propagator. We consider first the part of the displacement field caused by this part of the propagator. Denoting the field by $\mathbf{u}_C(\mathbf{r})$ we obtain

$$\langle \mathbf{u}_C(\mathbf{r})\rangle = \frac{3}{4\pi\mu_1}\frac{1}{\xi+4}\int \frac{1}{|\mathbf{r}-\mathbf{r}'|}\langle \mathbf{F}(\mathbf{r}')\rangle d\mathbf{r}' \ . \tag{3.14}$$

Substituting Eq. (3.10) and neglecting the higher-order multipole densities we find

$$\langle \mathbf{u}_C(\mathbf{r})\rangle = \frac{3}{4\pi\mu_1}\frac{1}{\xi+4}\int \frac{-1}{|\mathbf{r}-\mathbf{r}'|}\nabla'\cdot\langle \mathsf{F}^{(2)}(\mathbf{r}')\rangle d\mathbf{r}' \ . \tag{3.15}$$

This means that each of the Cartesian components of

$\langle \mathbf{u}_C(\mathbf{r})\rangle$ may be regarded as the electrostatic potential generated by a charge density given by the corresponding component of $-\nabla\cdot\langle \mathsf{F}^{(2)}(\mathbf{r})\rangle$. Recalling that in electrostatics $-\nabla\cdot\mathbf{P}$ acts like a charge density we can write down the Lorentz field corresponding to Eq. (3.15), namely,

$$(\nabla \mathbf{u}_C)_L=\nabla\langle \mathbf{u}_C\rangle-\frac{1}{\mu_1}\frac{1}{\xi+4}\langle \mathsf{F}^{(2)}\rangle \ . \tag{3.16}$$

It will be convenient to separate this into the relevant tensor parts. The symmetric traceless part is

$$((\nabla \mathbf{u}_C)_L)^0=\langle(\nabla \mathbf{u}_C)^0\rangle-\frac{1}{\mu_1}\frac{1}{\xi+4}\langle(\mathsf{F}^{(2)})^0\rangle \ , \tag{3.17}$$

and the trace part is

$$\mathrm{Tr}(\nabla \mathbf{u}_C)_L=\langle\nabla\cdot\mathbf{u}_C\rangle-\frac{1}{\mu_1}\frac{1}{\xi+4}\mathrm{Tr}\langle \mathsf{F}^{(2)}\rangle \ . \tag{3.18}$$

Next we consider the displacement field caused by the first part of the Green's function in Eq. (2.13). We denote this Oseen part as $\mathbf{u}_{\mathrm{Os}}(\mathbf{r})$ and obtain

$$\langle \mathbf{u}_{\mathrm{Os}}(\mathbf{r})\rangle=\frac{\xi+1}{\xi+4}\int \mathsf{T}(\mathbf{r}-\mathbf{r}')\cdot\langle \mathbf{F}(\mathbf{r}')\rangle d\mathbf{r}' \ , \tag{3.19}$$

where the propagator is given by the Oseen tensor

$$\mathbf{T}(\mathbf{r})=\frac{1}{8\pi\mu_1}\frac{\mathbf{1}+\hat{\mathbf{r}}\hat{\mathbf{r}}}{r} \ . \tag{3.20}$$

By comparison with Eq. (2.11) we have

$$\langle \mathbf{u}\rangle=\mathbf{u}_0+\langle \mathbf{u}_C\rangle+\langle \mathbf{u}_{\mathrm{Os}}\rangle \ . \tag{3.21}$$

The Lorentz field corresponding to the Oseen propagator has been studied by one of us [21]. Neglecting again the higher-order multipole densities we obtain from Eq. (6.4) of Ref. [21] the Lorentz field

$$(\nabla \mathbf{u}_{\mathrm{Os}})_L=\nabla\langle \mathbf{u}_{\mathrm{Os}}\rangle-\frac{1}{5\mu_1}\frac{\xi+1}{\xi+4}[\langle \mathsf{F}^{(2)}\rangle-\tfrac{1}{3}\mathbf{1}\,\mathrm{Tr}\langle \mathsf{F}^{(2)}\rangle] \ . \tag{3.22}$$

The symmetric traceless part of this equation is

$$((\nabla \mathbf{u}_{\mathrm{Os}})_L)^0=\langle(\nabla \mathbf{u}_{\mathrm{Os}})^0\rangle-\frac{1}{5\mu_1}\frac{\xi+1}{\xi+4}\langle(\mathsf{F}^{(2)})^0\rangle \ . \tag{3.23}$$

The trace part vanishes identically. We define the local pure strain in the mean-field approximation as

$$\mathsf{E}_L=(\nabla \mathbf{u}_0)^0+((\nabla \mathbf{u}_C)_L)^0+((\nabla \mathbf{u}_{\mathrm{Os}})_L)^0 \ . \tag{3.24}$$

Altogether we find from Eqs. (3.18), (3.21), and (3.23)

$$\mathsf{E}_L=(\nabla\langle \mathbf{u}\rangle)^0-\frac{1}{5\mu_1}\frac{\xi+6}{\xi+4}\langle(\mathsf{F}^{(2)})^0\rangle \ . \tag{3.25}$$

Similarly we define the local pressure in mean-field approximation:

$$P_L=-\kappa_1\nabla\cdot\mathbf{u}_0-\kappa_1\mathrm{Tr}(\nabla \mathbf{u}_C)_L \ . \tag{3.26}$$

From Eqs. (3.18) and (3.21) we find

$$P_L=-\kappa_1\nabla\cdot\langle \mathbf{u}\rangle+\frac{1}{3}\frac{\xi}{\xi+4}\mathrm{Tr}\langle \mathsf{F}^{(2)}\rangle \ . \tag{3.27}$$

This last equation is in agreement with Eq. (6.9) of Ref. [21].

Finally we calculate the average force dipole density by replacing the acting field in Eq. (3.13) by the local field in analogy to Eq. (3.2). For the symmetric traceless part this yields

$$\langle(\mathsf{F}^{(2)})^0\rangle=-2\phi[\mu]\mu_1\mathsf{E}_L \tag{3.28}$$

and for the trace part

$$\mathrm{Tr}\langle \mathsf{F}^{(2)}\rangle=3\phi[\kappa]P_L \ . \tag{3.29}$$

Substituting Eq. (3.25) into Eq. (3.28) and solving for the force density tensor we find

$$\langle(\mathsf{F}^{(2)})^0\rangle=-\frac{2[\mu]\phi\mu_1}{1-\frac{2}{5}\frac{\xi+6}{\xi+4}[\mu]\phi}(\nabla\langle \mathbf{u}\rangle)^0 \ . \tag{3.30}$$

Substituting Eq. (3.27) into Eq. (3.29) we find

$$\mathrm{Tr}\langle \mathsf{F}^{(2)}\rangle=-\frac{3[\kappa]\phi\kappa_1}{1-\frac{\xi}{\xi+4}[\kappa]\phi}\nabla\cdot\langle \mathbf{u}\rangle \ . \tag{3.31}$$

These are the desired constitutive equations in mean-field approximation. Substituting in Eq. (3.12) we find the effective shear modulus

$$\mu_{\mathrm{eff}}=\mu_1+\frac{[\mu]\phi}{1-\frac{2}{5}\frac{\xi+6}{\xi+4}[\mu]\phi}\mu_1 \tag{3.32}$$

and the effective bulk modulus

$$\kappa_{\mathrm{eff}}=\kappa_1+\frac{[\kappa]\phi}{1-\frac{\xi}{\xi+4}[\kappa]\phi}\kappa_1 \ . \tag{3.33}$$

There is a clear resemblance to the CM formula (3.5) for the effective dielectric constant. In the incompressible limit, where κ_1 and ξ tend to infinity, the expression (3.32) for the effective shear modulus becomes

$$\mu_{\mathrm{eff}}=\mu_1+\frac{[\mu]\phi}{1-\frac{2}{5}[\mu]\phi}\mu_1 \quad (\kappa_1\rightarrow\infty) \ , \tag{3.34}$$

which is closely similar to Saitô's expression for the effective shear viscosity of a fluid suspension of hard spheres [22]. Saitô's derivation was based on a local field argument similar to that of Lorentz for dielectric suspensions [21].

IV. UNIFORM SPHERES

For uniform spheres the effective elastic moduli in mean-field approximation, as given by Eqs. (3.32) and (3.33), were obtained earlier by Weng [23] on the basis of the Mori-Tanaka method [24]. The method is explained particularly clearly by Benveniste [25] and Christensen [26]. There is no obvious relation to the concept of the effective local field. An expression for the effective elastic tensor of a system of uniform ellipsoids proposed by Markov [27] reduces to Eqs. (3.32) and (3.33) for the case of spheres. For this case Markov refers to an earlier result by Levin. The mean-field expressions (3.32) and (3.33) are closely related to the so-called Hashin-Shtrikman (HS) bounds [4]. In this section, we investigate the relation.

For a uniform sphere with shear modulus μ_2 and bulk modulus κ_2 the intrinsic shear modulus is given by [13,14]

$$[\mu]=5(\xi+4)\frac{\mu_2-\mu_1}{(2\xi+12)\mu_2+(3\xi+8)\mu_1}\ , \qquad (4.1)$$

and the intrinsic bulk modulus is given by [13,14]

$$[\kappa]=(\xi+4)\frac{\kappa_2-\kappa_1}{\xi\kappa_2+4\kappa_1}\ . \qquad (4.2)$$

Following Walpole [28], the Hashin-Shtrikman upper and lower bounds can be presented in the following form. We introduce

$$\mu^{U*}=\frac{3}{2}\left[\frac{1}{\mu^g}+\frac{10}{9\kappa^g+8\mu^g}\right]^{-1},$$
$$\mu^{L*}=\frac{3}{2}\left[\frac{1}{\mu^l}+\frac{10}{9\kappa^l+8\kappa^l}\right]^{-1}, \qquad (4.3)$$
$$\kappa^{U*}=\frac{4}{3}\mu^g,\quad \kappa^{L*}=\frac{4}{3}\mu^l\ ,$$

where

$$\mu^g=\max(\mu_1,\mu_2),\quad \mu^l=\min(\mu_1,\mu_2)\ ,$$
$$\kappa^g=\max(\kappa_1,\kappa_2),\quad \kappa^l=\min(\kappa_1,\kappa_2)\ . \qquad (4.4)$$

Then the upper (U) and lower (L) bounds on $\mu_{\rm eff},\kappa_{\rm eff}$ are

$$\mu_{\rm HS}^{U(L)}=\mu_1+\phi(\mu_2-\mu_1)\left[1+\frac{(\mu_2-\mu_1)(1-\phi)}{\mu_1+\mu^{U(L)*}}\right]^{-1}, \qquad (4.5)$$

$$\kappa_{\rm HS}^{U(L)}=\kappa_1+\phi(\kappa_2-\kappa_1)\left[1+\frac{(\kappa_2-\kappa_1)(1-\phi)}{\kappa_1+\kappa^{U(L)*}}\right]^{-1}. \qquad (4.6)$$

We consider first the bulk modulus. After some algebra one finds

$$\kappa_{\rm eff}=\kappa_{\rm HS}^{L}\quad \text{for } \mu_1<\mu_2\ ,$$
$$\kappa_{\rm eff}=\kappa_{\rm HS}^{U}\quad \text{for } \mu_1>\mu_2\ . \qquad (4.7)$$

Thus the mean-field approximation coincides with one of the Hashin-Shtrikman bounds. The same situation obtains for the effective dielectric constant. The situation is more complicated for the effective shear modulus. Here we find

$$\mu_{\rm eff}=\mu_{\rm HS}^{L}\quad \text{for } \mu_1<\mu_2,\quad \kappa_1<\kappa_2,$$
$$\mu_{\rm eff}=\mu_{\rm HS}^{U}\quad \text{for } \mu_1>\mu_2,\quad \kappa_1>\kappa_2\ . \qquad (4.8)$$

However, in mixed cases where $\mu_1<\mu_2$, $\kappa_1>\kappa_2$ or $\mu_1>\mu_2$, $\kappa_1<\kappa_2$, the mean-field approximation lies between the two Hashin-Shtrikman bounds.

For the special case $\mu_1=\mu_2$, the upper and lower Hashin-Shtrikman bounds for the bulk modulus coincide, so that for that case the bounds give the exact value. It may be cast in the form

$$\frac{1}{3\kappa_{\rm eff}+4\mu}=\frac{1-\phi}{3\kappa_1+4\mu}+\frac{\phi}{3\kappa_2+4\mu}\ , \qquad (4.9)$$

where $\mu=\mu_1=\mu_2$. This remarkable result was first proved in greater generality by Hill [29].

V. CLUSTER EXPANSION

In this section we return to the general case of inclusions with a spherically symmetric elastic profile, as specified in Eq. (2.1). We show that expressions (3.32) and (3.33), obtained in the mean-field approximation, may be systematically corrected for correlations in the positions of the spheres. We can utilize exact cluster expansions which have been derived for the calculation of the linear transport properties of suspensions. In these expansions the mean-field expressions are obtained as a first approximation.

A cluster expansion for the calculation of the effective dielectric constant has been developed by Felderhof, Ford, and Cohen [7]. They have shown that the CM formula (3.4) may be obtained from this expansion by a summation of the so-called virtual-overlap integrals [5]. The cluster expansion was applied to elastic suspensions by Jones and Schmitz [9]. They studied the first two terms in the density expansion of the effective elastic moduli [13]. In particular they evaluated the first virtual-overlap integral. We show here how expressions (3.32) and (3.33) may be obtained by a summation of a geometric series of virtual-overlap integrals.

The elastic moduli may be combined into an effective elastic tensor $\mathsf{C}^{\rm eff}$ by

$$\mathsf{C}^{\rm eff}=2\mu_{\rm eff}\mathsf{P}+3\kappa_{\rm eff}\mathsf{Q}\ , \qquad (5.1)$$

where P and Q are invariant fourth-rank tensors which are orthogonal projectors in the space of symmetric second-rank tensors:

$$P_{\alpha\beta\gamma\delta}=\tfrac{1}{2}(\delta_{\alpha\gamma}\delta_{\beta\delta}+\delta_{\alpha\delta}\delta_{\beta\gamma})-\tfrac{1}{3}\delta_{\alpha\beta}\delta_{\gamma\delta}\ ,\quad Q_{\alpha\beta\gamma\delta}=\tfrac{1}{3}\delta_{\alpha\beta}\delta_{\gamma\delta}\ . \qquad (5.2)$$

The one-body contribution to $\mathsf{C}^{\rm eff}$ is given by

$$\mathsf{C}^{\rm eff}_{\text{one-body}}=2[\mu]\phi\mu_1\mathsf{P}+3[\kappa]\phi\kappa_1\mathsf{Q}\ . \qquad (5.3)$$

The calculation of the two-body virtual-overlap contribution $\mathsf{C}^{\rm eff}$ is based on the theorem

$$\int_{S_{2a}}\mathsf{G}(\mathbf{r}-\mathbf{R})d\mathbf{R}=-\frac{1}{5\mu_1}\frac{\xi+6}{\xi+4}\mathsf{P}-\frac{1}{\mu_1(\xi+4)}\mathsf{Q}\quad\text{for } r<2a\ , \qquad (5.4)$$

where the integration is over a sphere of radius $2a$ and where the tensor Green's function $\mathsf{G}(\mathbf{r})$ is given by

$$\mathsf{G}_{\alpha\beta\gamma\delta}(\mathbf{r})=\tfrac{1}{4}(\partial_\alpha\partial_\gamma G_{0\beta\delta}+\partial_\alpha\partial_\delta G_{0\beta\gamma}+\partial_\beta\partial_\gamma G_{0\alpha\delta}+\partial_\beta\partial_\delta G_{0\alpha\gamma})\ . \qquad (5.5)$$

The argument given by Jones and Schmitz [13] for the validity of Eq. (5.4) is not quite sufficient, since it applies only for $\mathbf{r}$ at the origin. However, the integral on the left is constant throughout the sphere [30,31]. The two-body virtual-overlap contribution is found to be [13]

$$\mathsf{C}^{\text{eff}}_{\text{two-body}}=\frac{4}{5}\frac{\xi+6}{\xi+4}[\mu]^2\phi^2\mu_1\mathsf{P}+3\frac{\xi}{\xi+4}[\kappa]^2\phi^2\kappa_1\mathsf{Q}\ . \tag{5.6}$$

The constant tensor on the right of Eq. (5.4) occurs repeatedly in the virtual-overlap integrals. This leads to a geometric series which is easily summed by use of the projector properties of the tensors P and Q. The whole geometric series of virtual overlap integrals may be constructed from the first two terms given in Eqs. (5.3) and (5.6). Summing the series we find the virtual-overlap approximation

$$\mathsf{C}^{\text{eff}}_{\text{ov}}=2\mu^{\text{MF}}_{\text{eff}}\mathsf{P}+3\kappa^{\text{MF}}_{\text{eff}}\mathsf{Q}\ , \tag{5.7}$$

where $\mu^{\text{MF}}_{\text{eff}}$ and $\kappa^{\text{MF}}_{\text{eff}}$ are given by Eqs. (3.32) and (3.33).

It has been shown by Cichocki and Felderof [8] that alternatively one may formulate a so-called renormalized cluster expansion. In this formulation large classes of averaged multiple scattering processes are resummed effectively. The final result of the expansion may be expressed in terms of a wave-vector-dependent susceptibility tensor $\boldsymbol{\chi}(\mathbf{q})$. From isotropy it follows that the tensor has the form

$$\boldsymbol{\chi}(\mathbf{q})=\chi_L(q)\hat{\mathbf{q}}\hat{\mathbf{q}}+\chi_T(q)(\mathbf{1}-\hat{\mathbf{q}}\hat{\mathbf{q}})\ . \tag{5.8}$$

The effective shear modulus is given by the limiting value

$$\mu_{\text{eff}}=\mu_1-\lim_{q\to 0}\chi_T(q)/q^2 \tag{5.9}$$

and the effective bulk modulus is given by

$$\kappa_{\text{eff}}=\kappa_1-\lim_{q\to 0}[\chi_L(q)-\tfrac{1}{3}\chi_T(q)]/q^2\ . \tag{5.10}$$

The susceptibility tensor is given by the exact expression [32]

$$\boldsymbol{\chi}(\mathbf{q})=n(\mathbf{q}|\mathbf{M}[\mathbf{I}-n\mathbf{R}(\mathbf{q})\mathbf{M}]^{-1}|\mathbf{q}) \tag{5.11}$$

with the notation

$$(\mathbf{q}|A|\mathbf{q}')=\int e^{-i\mathbf{q}\cdot\mathbf{r}}A(\mathbf{r},\mathbf{r}')e^{i\mathbf{q}'\cdot\mathbf{r}'}d\mathbf{r}d\mathbf{r}'\ . \tag{5.12}$$

The operator $\mathbf{M}$ in Eq. (5.11) is the response kernel of a single inclusion, defined in Eq. (2.15). The inclusion may be taken to be centered at the origin. The recurrence operator $\mathbf{R}(\mathbf{q})$ has been expressed as a cluster expansion

$$\mathbf{R}(\mathbf{q})=\sum_{s=2}^{\infty}\mathbf{R}_s(\mathbf{q})\ , \tag{5.13}$$

where a term $\mathbf{R}_s(\mathbf{q})$ involves an s-body elastic problem and integration over s-body correlation functions. The recurrence operator may be written as a sum of a virtual-overlap contribution and a nonoverlap contribution [33]

$$\mathbf{R}(\mathbf{q})=\mathbf{R}_{\text{ov}}(\mathbf{q})+\mathbf{R}_{\text{no}}(\mathbf{q})\ . \tag{5.14}$$

We have shown that the mean-field expressions (3.32) and (3.33) result if the recurrence operator $\mathbf{R}(\mathbf{q})$ is approximated by its overlap part $\mathbf{R}_{\text{ov}}(\mathbf{q})$. The analysis follows the lines given earlier for the case of effective viscosity [34]. The details will be published elsewhere.

The geometric series structure is built into the expression (5.11). Hence, this expression is the most suitable starting point for a calculation of correction terms to the mean-field approximation. Such a calculation involves the solution of the two-sphere problem, the three-sphere problem, etc., and its average over the appropriate correlation functions.

VI. DISCUSSION

We have found mean-field expressions for the effective elastic moduli of a suspension of spheres, with a spherically symmetric elastic profile, that are embedded in a uniform and isotropic matrix. We have indicated how the correction terms may be evaluated by relating the calculation to existing cluster expansions for the effective linear properties of suspensions. It will be of interest to study the correction terms in analogy to the theory of dielectrics.

The theory presented here may be extended in several directions. For example, one might consider oriented ellipsoids or a random distribution of parallel cylinders. The theory applies directly to coated spheres. It will be of interest to study the effective elastic properties of such systems.

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