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Effective Medium Theory for Elastic Matrix Composites Containing Dispersed Particulates*

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We describe a new, effective medium theory to study the wave propagation and mechanical properties of a composite system with dispersed particulates. One main emphasis here is in formulating the theory and in analyzing the structure of the contribution of the fillers to the elastic response. By constructing the elastic propagator (whose fluid mechanical counterpart is known as the Oseen tensor), we show that an analogy between the theoretical description of the particulate system and of suspension rheology exists when the former corresponds to a high-rigidity solid matrix (or, analogously, when the Poisson ratio is close to 1/2) in steady state. The effective Lamé constants for this case are derived by combining this analogy with the theory developed by Freed and Muthukumar for the rheology of a suspension of spheres. The analogy is also useful in our new prediction of the phenomenon of elastic screening, the possible existence of a cutoff frequency below which elastic waves cannot propagate in the filler system.

KEY WORDS: Particulate filler; composites; Green's function; Oseen tensor; elastic interaction tensor; suspension rheology; Lamé constant; effective medium theory; elastic screening.

1. INTRODUCTION

Two or more materials, when combined, will perform differently, and often more efficiently, than the materials by themselves. A particulate composite is made by dispersing small particles of one material in another and is

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most commonly used in modern technology.[1] The mechanical properties and some phenomenological models of these two-phase composites are summarized well in the book by Nelson.[2]

In this paper, we introduce a new theoretical approach based on the effective medium theory (developed primarily by Freed, Edwards, and Muthukumar, among others, in their studies of suspension[3] and polymer rheology[4]) to determine the elastic moduli of a solid that contains spherical particles of another elastic material.

The conventional approach for studying the mechanical properties of composites is primarily based on the Einstein theory of suspensions.[5] It is possible to evaluate the elastic constants of the composite medium in the limit of infinite dilution,[6] using a calculation of the stored strain energy or of the spatially averaged stress–strain relationship. We believe that the key ingredient of this approach lies in the calculation of the perturbed stress or strain field throughout the arbitrarily loaded matrix, where the perturbations are due to the presence of particles (inclusions) that are immersed in the matrix. In general, the calculation of the perturbed stress or strain field is an extremely laborious technical problem. An analytic solution is available only for a single spherical (or cylindrical) inclusion with a very specific external load.[7]

The first qualitative analogy between the description of elastic media and Einstein's theory of suspension rheology was recognized by Guth and his co-workers[8] four decades after Einstein's studies on suspension. This analogy is still very useful in studying the mechanical properties of the particulate composites, even though the analysis, in principle, is limited to a dilute filler system. On the other hand, the high-filler-concentration range with a random distribution of particulate fillers has recently been studied using percolation theory to calculate effective elastic constants.[9–11] Percolation theory was also adapted for the suspension problem to calculate the shear viscosity of slurries.[12]

Our goal in this paper is to develop a microrheological framework for determining the gross mechanical properties of a composite material in terms of particle shape, interfacial adhesion, volume fraction, and the statistical distribution of fillers, among others. Here we derive the virtually exact, yet formal effective medium equation and also show, in principle, how the elastic constants can be calculated. The detailed analysis requires complicated mathematical details; therefore, by introducing an analogy with the studies of Muthukumar and Freed on suspension rheology,[3] we directly obtain results for effective elastic constants and avoid complicated algebraic details.

The analogy with suspension rheology begins with a derivation of an "elastic interaction tensor," which corresponds to the elastic Green's function in the absence of particulate fillers. We then show that an exact analogy between the elastic and fluid media exists for a high-rigidity solid matrix in steady state. The analogy fails in more general cases, which will be discussed elsewhere.

Since our formulation is general and covers the dynamic response of the filler system, it can also be used in studying wave propagation through random media. Recent studies on sound wave propagation in a porous random media are described in ref. 13. We believe that our new theory is not only complementary, but is also a more powerful technique for studying the mechanical and dynamical response of the composite media.

Section 2 introduces the effective medium equations along with the components Ξ_i of a friction coefficient density due to the embedded particles. The physical interpretation of Ξ_i is given in Section 3 along with methods for their experimental determination. The existence of a long-wavelength contribution to Ξ_i of the appropriate sign is shown to predict the occurrence of a new phenomenon of "elastic screening" in which elastic waves below a cutoff frequency cannot propagate through an unbounded filler system. Calculations presented elsewhere demonstrate the existence of elastic screening at low-filler-volume fractions when no-slip boundary conditions are applicable. Section 4 introduces the dynamical elastic interaction tensor and the hydrodynamic analogy, which permits us to extract the effective elastic constants for the limit of a high-rigidity solid matrix.

2. GENERAL FORMULATION

When homogeneous, elastic media undergo a small deformation, the displacement vector $\mathbf{u}$ is governed by the linearized field equation,[14]

$$\rho \frac{\partial^2}{\partial t^2} \mathbf{u} - (\lambda + \mu)\, \nabla(\nabla \cdot \mathbf{u}) - \mu \nabla^2 \mathbf{u} = \mathbf{F} \tag{1}$$

where ρ is the mass density, λ and μ are elastic coefficients, and $\mathbf{F}$ represents the force density due to all external sources.

With particles present, the equation of motion for the system is nontrivially modified by forces exerted at the particle–elastic medium interface. Stated alternatively, the macroscopic homogeneity of the stress no longer corresponds to microscopic homogeneity as well. It is sufficient for our analysis that an appropriate macroscopically small volume element can be defined such that the stress within it can be viewed as homogeneous. However, the volume element is of sufficient dimensions that the continuum hypothesis is applicable.

We let $\mathbf{R}_\alpha$ be the position vector of a point with angular coordinates Ω_α on the surface of the αth particle. More specifically, Ω_α is the center of a differential element of area $d\Omega_\alpha$ such that $\int d\Omega_\alpha$ gives the surface integral over the particle.

Let $\mathbf{R}_\alpha^0$ be the position vector of the particle center of mass and $\mathbf{r}_\alpha = \mathbf{R}_\alpha - \mathbf{R}_\alpha^0$ be a radius vector from the particle center to Ω_α. We also define $\boldsymbol{\sigma}_\alpha = \boldsymbol{\sigma}_\alpha(\Omega_\alpha, t)$ as the surface force exerted on the elastic medim at Ω_α. Figure 1 depicts these quantities for spherical particles.

The total force density at $\mathbf{r}$ and t from all the particles is[3]

$$\sum_{\alpha=1}^{N} \int d\Omega_\alpha\, \delta(\mathbf{r} - \mathbf{R}_\alpha)\, \boldsymbol{\sigma}_\alpha(\Omega_\alpha, t) \tag{2}$$

The modified field equation in the presence of the particles becomes

$$\rho \frac{\partial^2}{\partial t^2} \mathbf{u} - (\lambda + \mu)\, \nabla(\nabla \cdot \mathbf{u}) - \mu \nabla^2 \mathbf{u} = \mathbf{F} + \sum_{\alpha=1}^{N} \int d\Omega_\alpha\, \delta(\mathbf{r} - \mathbf{R}_\alpha)\, \boldsymbol{\sigma}_\alpha(\Omega_\alpha, t) \tag{3}$$

In Eqs. (2) and (3), N is the total number of particles in the system. The force $\boldsymbol{\sigma}_\alpha$ is defined to act on the surface of the particle, while $\mathbf{F}$ vanishes both inside the particle and at its surface.

It is important to recognize that the individual forces $\boldsymbol{\sigma}_\alpha(\Omega_\alpha, t)$ need not be represented in any explicit mathematical forms. To do so would require a knowledge of the currently unavailable microscopic forces between the constituent atoms of the particle and those of the matrix and would be beyond the scope of our continuum treatment. Furthermore, the

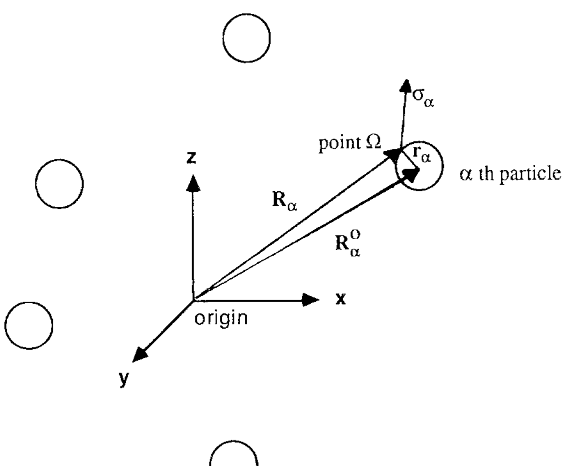


Fig. 1. Microscopic representation of the composite media, notation used in the analysis.

microscopic specifications are unnecessary in the continuum theory because the surface forces $\boldsymbol{\sigma}(\Omega_\alpha, t)$ are established by boundary conditions that are imposed at the surfaces. As the boundary conditions thus represent some *ad hoc* effective specifications on the microscopic forces, they must either be well understood or the problem must be well formulated. In the following analysis, it is assumed that the above requirements do not represent stringent specifications, *as long as the continuum hypothesis is valid.* A subsequent paper presents detailed calculations using the simplest boundary conditions, the no-slip conditions for which $\mathbf{u}(\mathbf{R}_\alpha, t) = 0$ for all α.

The problem is more easily studied in Fourier space. The Fourier transform of Eq. (3) reduces to

$$-\rho\omega^2\mathbf{u}(\mathbf{k}, \omega) + (\lambda + \mu)\,\mathbf{kk}\cdot\mathbf{u}(\mathbf{k}, \omega) + \mu\mathbf{k}^2\mathbf{u}(\mathbf{k}, \omega)$$
$$= \mathbf{F}(\mathbf{k}, \omega) + \sum_{\alpha=1}^{N} \int dt \int d\Omega_\alpha\, \boldsymbol{\sigma}_\alpha(\Omega_\alpha, t) \exp(-i\omega t - i\mathbf{k}\cdot\mathbf{R}_\alpha) \tag{4}$$

We emphasize again that the rightmost term involving $\boldsymbol{\sigma}_\alpha$ contains all of the detailed microscopic dynamics of the filler system. It depends on the boundary conditions, the respective volume fraction of all the phases, and the specific spatial distribution of the particles. Fortunately, it is sufficient for us to determine only the average properties for the entire system. The averaging process is performed over all spatial positions of all particles at some initial time. It differs somewhat from that used in the rheology of suspensions for cases in which the particles are mobile. The particles in the composite are fixed in space, and no specification of the particle velocities is necessary.

The average displacement vector $(\mathbf{v})$ is written as

$$\mathbf{v}(\mathbf{r}, t) \equiv \langle \mathbf{u}(\mathbf{r}, t \mid \{\mathbf{R}_\alpha^0\}) \rangle$$
$$\equiv \int \prod_{\alpha=1}^{N} dR_\alpha^0\, \mathbf{u}(\mathbf{r}, t)\, \Psi(\{\mathbf{R}_\alpha^0\}) \tag{5}$$

where $\Psi(\{\mathbf{R}_\alpha^0\})$ is the initial N particle distribution function (see standard texts on statistical mechanics; e.g., ref. 15) of the particulate fillers, and the dependence of $\mathbf{u}$ on the initial positions of the particles is explicitly denoted.

The average field $\mathbf{v}$ is much simpler both in structure and to calculate than $\mathbf{u}$. The average of Eq. (4) produces a formal expression for an effective medium theory of solid composites,

$$[-\rho\omega^2 + \Xi_1(\omega)]\,\mathbf{v}(\mathbf{k}, \omega)$$
$$+ [\lambda + \mu + \Xi_2(\mathbf{k}, \omega)]\,\mathbf{kk}\cdot\mathbf{v}(\mathbf{k}, \omega)$$
$$+ [\mu + \Xi_3(\mathbf{k}, \omega)]\,k^2\mathbf{v}(\mathbf{k}, \omega) = \boldsymbol{F}(\mathbf{k}, \omega) \tag{6}$$

The quantities Ξ_1, Ξ_2, and Ξ_3 are defined from Eqs. (4)–(6) by

$$\left\langle \int dt \int \sum_{\alpha=1} d\Omega_\alpha \exp(-i\mathbf{k}\cdot\mathbf{R}_\alpha - i\omega t)\, \boldsymbol{\sigma}_\alpha(\Omega_\alpha, t) \right\rangle$$

$$\equiv [\Xi_1(\omega)\, \boldsymbol{\delta} + \Xi_2(\mathbf{k}, \omega)\, \mathbf{kk} + \Xi_3(\mathbf{k}, \omega)\, k^2 \boldsymbol{\delta}] \cdot \mathbf{v} \tag{7}$$

where $\boldsymbol{\delta}$ is the unit tensor.

Some brief comments on Eq. (7) are in order. The left-hand side must be proportional to $\mathbf{v}$ in the region of linear elasticity, and the proportionality constant must be a second-rank tensor. Since the average is performed over the statistical distribution of fillers, the resulting second-rank tensor can only depend on $\mathbf{k}$. This implies that this tensor is a linear combination of $\boldsymbol{\delta}$ and $\mathbf{kk}$ in the form written in Eq. (7).

However, it still remains to determine Ξ_1, Ξ_2, and Ξ_3. If we can calculate these quantities, the problem is completely solved; however, the exact calculation of these quantities is virtually impossible. The major purpose of this paper is to investigate the properties, physical interpretation, and methods for experimental determination of these quantities without introducing tremendous algebraic details. Specific computations of new results will be provided in a subsequent paper for the steady-state limit.

All of the relevant information pertaining to the volume fraction, particle distribution, and boundary conditions is conveyed into the formulation by Ξ_1, Ξ_2, and Ξ_3. We propose an approach for their separable, experimental determination in the next section.

3. PHYSICAL MEANING OF Ξ_1, Ξ_2, AND Ξ_3

The steady-state ($\omega = 0$) and low-wavevector (i.e., $k \to 0$) limit enables Eq. (6) to be reduced to

$$\Xi_1(0)\, \mathbf{v}(\mathbf{k}, 0) + [\lambda + \mu + \Xi_2(0, 0)]\, \mathbf{kk} \cdot \mathbf{v}(\mathbf{k}, 0)$$

$$+ [\mu + \Xi_3(0, 0)]\, k^2 \mathbf{v}(\mathbf{k}, 0) = \mathbf{F}(\mathbf{k}, 0) \tag{8}$$

Alternatively, the real-space form of Eq. (8) is

$$\Xi_1(0)\, \mathbf{v} - [\lambda + \mu + \Xi_2(0, 0)]\, \nabla(\nabla \cdot \mathbf{v}) - [\mu + \Xi_3(0, 0)]\, \nabla^2 \mathbf{v} = \mathbf{F} \tag{9}$$

In this zero-frequency limit, the real-space representation of the effective field equation (8) becomes

$$-(\lambda^* + \mu^*)\, \nabla(\nabla \cdot \mathbf{v}) - \mu^* \nabla^2 \mathbf{v} = \mathbf{F} \tag{10}$$

where λ^* and μ^* are the effective composition-dependent Lamé coefficients for the composite material. By comparing Eqs. (9) and (10) [setting $\Xi_1(0)=0$ for simplicity], we have

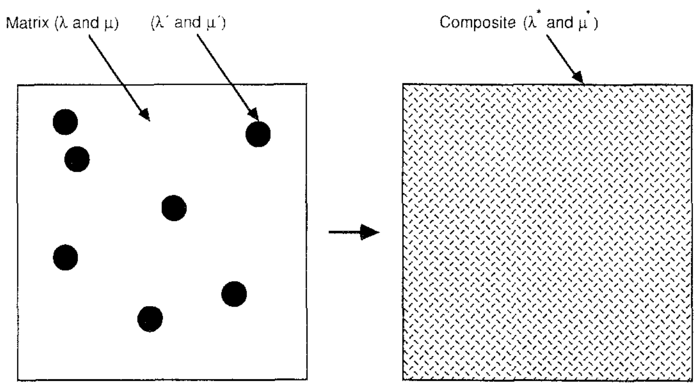
$$\mu^* = \mu + \Xi_3(0, 0), \qquad \lambda^* = \lambda + [\Xi_2(0, 0) - \Xi_3(0, 0)] \tag{11}$$

Figure 2 illustrates the effective medium concept.

Therefore, if we can calculate Ξ_2 and Ξ_3 to obtain effective Lamé constants from Eq. (11), we can determine any other set of effective elastic properties. For example, the relationships

$$E = \frac{\mu(3\lambda + 2\mu)}{\lambda + \mu}, \qquad v = \frac{\lambda}{2(\lambda + \mu)} \tag{12}$$

define the Young's modulus E and Poisson ratio v. A future paper gives a general scheme for calculating Ξ_2 and Ξ_3 as expansions in filler volume fraction ϕ. Consequently, we believe this method to be more general and systematic than any previous theory.[1,2] One point deserves special emphasis: Ξ_1 is, in general, nonvanishing for the system. The term originates as a *screening of elastic interaction due to the fixed particle's presence.* The existence of randomly distributed fillers damps or "screens" the elastic waves in the medium. A similar concept that is useful in the fluid mechanics of porous media is known as Darcy's law. A study of Eq. (6) for finite frequencies also enables us to derive information about the influence of the particles on sound wave propagation and attenuation. These two cases are now discussed separately.



(A) (B)

Fig. 2. Notation used for the elastic constants. (A) A system that may represent the actual microscopic configuration of some region in the composite. The force dynamics for this system is given by Eq. (4). (B) An averaged system with the same effective macroscopic characteristics as for (A). The dynamics for (B) are described by either Eq. (8), or its equivalent, Eq. (10). In Eq. (10), we have introduced the quantities λ^* and μ^*, effective elastic constants.

3.1. $\Xi_1 = 0$ (No Interparticle Elastic Screening)

In this case, Eq. (6) becomes

$$-\rho\omega^2\mathbf{v}(\mathbf{k}, \omega) + [\lambda + \mu + \Xi_2(0, 0)]\, \mathbf{kk} \cdot \mathbf{v}(\mathbf{k}, \omega) + [\mu + \Xi_3(0, 0)]\, k^2\mathbf{v}(\mathbf{k}, \omega) = \mathbf{F} \tag{13}$$

The general dispersion relation for wave propagation is $\omega = \pm ck$. The transverse and longitudinal waves propagate by different mechanisms, and their components can be separated from Eq. (13) through their respective dependences on $\mathbf{v}_\perp = \mathbf{v} - \hat{\mathbf{k}}(\hat{\mathbf{k}} \cdot \mathbf{v})$ and $\mathbf{v}_{11} = \hat{\mathbf{k}}(\hat{\mathbf{k}} \cdot \mathbf{v})$. Here, $\hat{\mathbf{k}} = \mathbf{k}/k$ is a unit vector in the k direction.

The longitudinal wave equation is

$$(-\rho\omega^2 + \rho c_\parallel^2 k^2)(\mathbf{k} \cdot \mathbf{v}) = \mathbf{k} \cdot \mathbf{F}$$

$$c_\parallel^2 = \frac{\lambda + 2\mu + \Xi_2(0, 0) + \Xi_3(0, 0)}{\rho} \tag{14}$$

with $c_\parallel$ the longitudinal sound velocity in the averaged composite.

On the other hand, the transverse wave equation follows from Eq. (13) as

$$(-\rho\omega^2 + \rho c_\perp^2 k^2)[\mathbf{v} - \hat{\mathbf{k}}(\hat{\mathbf{k}} \cdot \mathbf{v})] = \mathbf{F} - \hat{\mathbf{k}}\hat{\mathbf{k}} \cdot \mathbf{F}$$

$$c_\perp^2 = \frac{\mu + \Xi_3(0, 0)}{\rho} \tag{15}$$

The two dispersion relations (14) and (15) enable the experimental determination of Ξ_2 and Ξ_3 as shown in Figs. 3A and 3B.

2.2. Elastic Screening Occurs ($\Xi_1 \neq 0$)

Screening effects on the hydrodynamics of concentrated particle systems have been much studied in recent years.[3,4,16,17] However, a convenient way for the direct experimental measurement of hydrodynamic screening has not yet been developed. We propose that the analogous effects for the solid composite could be determined by acoustics. As the approach is new, its consequences are now carefully considered.

First, consider the frequency versus wave number response (Fig. 3C). As $k \to 0$ and $\omega \to 0$, Ξ_1 is written as

$$\Xi_1 = \Xi_1^0 + \omega^2 \Xi_1^1 \tag{16}$$

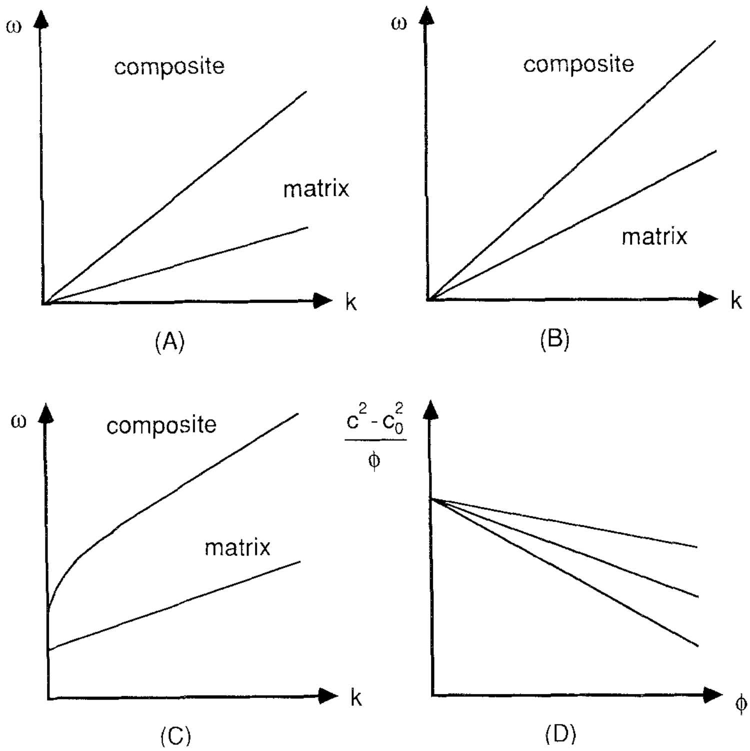


Fig. 3. Predicted low frequency limiting behavior for sound wave propagation in particulate-filled composites. With actual experimental data of the type depicted, we interpret the dispersion relations in the following ways. Following are explanations of the *gedanken* experiments performed and their results: (A) Results from studies of transverse waves in dilute systems where nearly zero screening exists. This gives us Ξ_3. (B) Similar studies of the longitudinal waves, yielding $\{\Xi_2 + \Xi_3\}$. (C) The behavior of longitudinal waves at higher concentrations, where screening occurs, and is used to obtain Ξ_1. (D) The results from a series of related composites; that is, where they all contain the same type of particles, interfacial bonding, and matrix material, but differ in concentration and also the degree of dispersion. The lines of differing slopes connect composites having the same normalized particle spatial distribution function. The limiting low concentration intercept is a function of the interfacial boundary conditions. The result of a common intercept (as shown) would not be a general one unless the interface is controlled. The first and the second order coefficients for concentration dependencies of the composites effective elastic constants [see Eqs. (21) and (22)] can be obtained directly from this type of data.

Here, unlike in fluid mechanics, the linear term in ω vanishes due to time-reversal symmetry. The Ξ_1^0 and Ξ_1^1 in Eq. (16) are independent of k and ω. Therefore, Eqs. (6) and (16) imply

$$\begin{aligned}\Xi_1^0\mathbf{v}(\mathbf{k},\omega)-(\rho-\Xi_1^1)\,\omega^2\mathbf{v}(\mathbf{k},\omega)\\+[\lambda+\mu+\Xi_2(0,0)]\,\mathbf{kk}\cdot\mathbf{v}(\mathbf{k},\omega)\\+[\mu+\Xi_3(0,0)]\,k^2\mathbf{v}(\mathbf{k},\omega)=\mathbf{F}(\mathbf{k},\omega)\end{aligned} \tag{17}$$

The longitudinal wave follows from Eq. (17) as the solution to

$$[\Xi_1^0-(\rho-\Xi_1^1)\,\omega^2+(\lambda+2\mu+\Xi_2+\Xi_3)\,k^2](\mathbf{k}\cdot\mathbf{v})=\mathbf{k}\cdot\mathbf{F} \tag{18}$$

thereby providing the general dispersion relation for the frequency of longitudinal waves as

$$\omega^2=\frac{\Xi_1^0+(\lambda+2\mu+\Xi_2+\Xi_3)\,k^2}{\rho-\Xi_1^1} \tag{19}$$

Ξ alters the longitudinal sound velocity, i.e., the curvature in Eq. (19). In addition, screening makes the frequency approach a nonzero minimum frequency or intercept of

$$\omega\to\left(\frac{\Xi_1^0}{\rho-\Xi_1^1}\right)^{1/2}\qquad\text{as}\quad k\to 0 \tag{20}$$

thereby allowing us to measure Ξ_1. The transverse wave can be similarly analyzed. A subsequent paper provides an explicit computation of Ξ_1^0 for dilute fillers.

Note that $\Xi_1^0<0$ for certain ranges of μ and λ.[20] In that case, unless $\rho-\Xi_1^1<0$, Eq. (20) implies that ω is purely imaginary. Hence, the waves are damped for $k^2<-\Xi_1^0/(\lambda+2\mu+\Xi_2+\Xi_3)$.

In addition, we may study how concentration influences the speed of sound (Fig. 3D). As an example, assume that the elastic behavior of the composite is given by

$$\mu^*=\mu(1+K_1\phi+K_2\phi^2) \tag{21}$$

Equation (17) then implies that a measurement of transverse sound speed enables K_1 and K_2 to be obtained as

$$\frac{c^2-c_0^2}{\phi}=K_1+K_2\phi \tag{22}$$

where c is the sound velocity of composite and c_0 is the sound velocity in the absence of filler. Therefore, plotting $(c^2-c_0^2)/\phi$ against ϕ, as shown in

Fig. 3D, provides the elasticity coefficients K_1 and K_2 (Einstein's and Huggins' coefficients) as intercept and slope.

The intercept gives K_1, and we believe that it is primarily dependent on interfacial characteristics (through boundary conditions such as no-slip). The slope yields K_2, and we speculate that it depends, in part, on the statistical distribution of particles.

There exists a remarkable contrast in the dispersion relation between the cases of Sections 2.1 and 2.2. In the former case the absence of screening causes ω to vanish as k approaches zero (acoustic phonon), while in the latter case with screening ω remains finite (nonzero) as k approaches zero (optical phonon). Our dilute, steady-state, no-slip computations in a subsequent paper, however, find that $\Xi_1(0)$ is always nonzero, and that question should be investigated for other choices of boundary conditions.

4. CALCULATION OF Ξ_1, Ξ_2, AND Ξ_3, AND THE ELASTIC INTERACTION TENSOR

A full-scale effective medium theory for calculating Ξ_1, Ξ_2, and Ξ_3 is developed elsewhere. Here, instead, we show how some results may be obtained by studying the analogy and differences between the theory of fluid mechanics and elastic media for particle systems.

The field equation for an elastic medium in Eq. (1) resembles the linearized Navier–Stokes equation of fluid mechanics for the fluid velocity $\mathbf{v}_f$,

$$\rho \frac{\partial}{\partial t} \mathbf{v}_f - \eta \nabla^2 \mathbf{v}_f + \nabla p = \mathbf{F} \tag{23}$$

with the incompressibility constraint

$$\nabla \cdot \mathbf{v}_f = 0 \tag{24}$$

Here, p is the pressure and η is the shear viscosity of the fluid.

The analogy consists of interpreting the displacement vector $\mathbf{u}$ as a velocity disturbance $\mathbf{v}_f$ and in making the correspondences $\eta \to \mu$ and $p \to -(\lambda + \mu) \nabla \cdot \mathbf{u}$. However, the underlying physics is somewhat different in both cases, as follows:

1. Equation (23) has a first-order time derivative which is characteristic of a dissipative mechanism, while Eq. (1) involves a second-order derivative as in wave propagation.

2. The relation $p \to -(\lambda + \mu) \nabla \cdot \mathbf{u}$ is only a mathematically convenient analogy. The dependence on pressure in fluid mechanics arises directly due to external forces.

3. Incompressibility, $\nabla \cdot \mathbf{u} = 0$ as in Eq. (24), is relatively rare in solids, but generally holds for Newtonian fluids at low frequencies.

In order to introduce the mathematical analogy between the description of the elastic medium and the suspension rheology, it is important to study the fundamental solution to the field equation (1) for homogeneous elasticity when there is a point source, i.e., when $\mathbf{F}(\mathbf{r}, t) = \mathbf{A}\delta(\mathbf{r} - \mathbf{r}_0)\,\delta(t - t_0)$. The general solution in this case is written as

$$\mathbf{u}_0(\mathbf{r}, t) = \mathsf{G}(\mathbf{r} - \mathbf{r}_0; t - t_0) \cdot \mathbf{A} \tag{25}$$

where G is called the dynamic elastic interaction tensor. This is now derived.

Taking the space-time Fourier transform of Eq. (1) produces

$$-\rho\omega^2\mathbf{u} + (\lambda + \mu)\,\mathbf{k}(\mathbf{k} \cdot \mathbf{u}) + \mu k^2\mathbf{u} = \mathbf{F} \tag{26}$$

Multiplying Eq. (26) by $\mathbf{k} \cdot$ yields

$$-\rho\omega^2(\mathbf{k} \cdot \mathbf{u}) + (\lambda + \mu)\,k^2(\mathbf{k} \cdot \mathbf{u}) + \mu k^2(\mathbf{k} \cdot \mathbf{u}) = \mathbf{k} \cdot \mathbf{F} \tag{27}$$

while rearrangement yields

$$\mathbf{k} \cdot \mathbf{u} = \frac{\mathbf{k} \cdot \mathbf{F}}{-\rho\omega^2 + (\lambda + 2\mu)\,k^2} \tag{28}$$

Therefore, Eqs. (26) and (28) combine as

$$\mathbf{u}(\mathbf{k}, \omega) = \mathsf{G}(\mathbf{k}, \omega) \cdot \mathbf{F}(\mathbf{k}, \omega) \tag{29}$$

where the explicit solution for G emerges as

$$\begin{aligned} \mathsf{G} &= \frac{1}{-\rho\omega^2 + \mu k^2}\left[\boldsymbol{\delta} - \frac{(\lambda + \mu)\,\hat{\mathbf{k}}\hat{\mathbf{k}}}{-\rho\omega^2 + (\lambda + 2\mu)\,k^2}\right] \\ &= \frac{\boldsymbol{\delta} - \hat{\mathbf{k}}\hat{\mathbf{k}}}{-\rho\omega^2 + \mu k^2} + \frac{\hat{\mathbf{k}}\hat{\mathbf{k}}}{-\rho\omega^2 + (\lambda + 2\mu)\,k^2} \end{aligned} \tag{30}$$

The first term in Eq. (30) is associated with a transverse elastic wave having a propagation velocity c_t of $c_t^2 = \mu/\rho$. The second term on the right-hand side in Eq. (30) describes the longitudinal elastic wave with velocity

$$c_l^2 = (\lambda + 2\mu)/\rho = E(1 - \nu)/\rho(1 + \nu)(1 - 2\nu)$$

Both of these relations are presented in standard texts on elasticity.[14]

Given the definitions of transverse and longitudinal wave velocities, G may be rewritten as

$$\mathsf{G} = \frac{\delta - \hat{\mathbf{k}}\hat{\mathbf{k}}}{\rho(-\omega^2 + c_t^2 k^2)} + \frac{\hat{\mathbf{k}}\hat{\mathbf{k}}}{\rho(-\omega^2 + c_l^2 k^2)} \tag{31}$$

It is of interest to express Eq. (29) in its real-space form

$$\mathbf{u}(\mathbf{r}, t) = \int d^3\bar{r} \int d\bar{t}\, \mathsf{G}(\mathbf{r} - \bar{\mathbf{r}}; t - \bar{t}) \cdot \mathbf{F}(\bar{\mathbf{r}}, \bar{t}) \tag{32}$$

For mathematical simplicity, we consider the steady-state limit ($\omega = 0$) of the elastic interaction tensor (G_s),

$$\begin{aligned} \mathsf{G}_s &= \frac{\delta - \hat{\mathbf{k}}\hat{\mathbf{k}}}{\rho c_t^2 k^2} + \frac{\hat{\mathbf{k}}\hat{\mathbf{k}}}{\rho c_l^2 k^2} \\ &= \frac{\delta - \hat{\mathbf{k}}\hat{\mathbf{k}}}{\mu k^2} + \frac{\hat{\mathbf{k}}\hat{\mathbf{k}}}{(\lambda + 2\mu)\, k^2} \end{aligned} \tag{33}$$

The real-space representation of G_s is obtained by taking the inverse Fourier transform in the Appendix. The result is found to be

$$\begin{aligned} \mathsf{G}_s(\mathbf{r} - \mathbf{r}') &\equiv \iiint \frac{d^3k}{(2\pi)^3} \exp[-i\mathbf{k} \cdot (\mathbf{r} - \mathbf{r}')]\, \mathsf{G}_s(k) \\ &= \frac{\delta + \hat{\mathbf{R}}\hat{\mathbf{R}}}{8\pi\mu R} + \frac{\delta - \hat{\mathbf{R}}\hat{\mathbf{R}}}{8\pi(\lambda + 2\mu)\, R} \end{aligned} \tag{34}$$

where $\mathbf{R} \equiv \mathbf{r} - \mathbf{r}'$ and $\hat{\mathbf{R}} = \mathbf{R}/R$. Thus, when $\lambda \to \infty$ (or $\nu \to 1/2$), G_s becomes identical to the Oseen tensor in fluid mechanics, where upon Eqs. (33) and (34) reduce to

$$\mathsf{G}_s(k) = \frac{1}{\mu k^2} (\delta - \hat{\mathbf{k}}\hat{\mathbf{k}}), \qquad \lambda \to \infty \tag{35}$$

$$\mathsf{G}_s(\mathbf{R}) = \frac{1}{8\pi\mu R} (\delta + \hat{\mathbf{R}}\hat{\mathbf{R}}), \qquad \lambda \to \infty \tag{36}$$

Here we only treat the $\nu \to 1/2$ case to establish the hydrodynamic analogy and to show how results for the suspension rheology can be used for filler elasticity. Elsewhere we give new results for $\nu \neq 1/2$. When $\nu = 1/2$, we may readily determine Ξ_2 and Ξ_3 of Eq. (7) by using the calculations of Freed and Muthukumar[3] (with slight modification) for spheres, which imply

$$\lim_{\substack{k \to 0 \\ \omega \to 0}} \left\langle \sum_{\alpha=1}^{N} \int dt \int d\Omega_\alpha \, \boldsymbol{\sigma}_\alpha(\Omega_\alpha, t) \exp[-i\mathbf{k} \cdot \mathbf{R}_\alpha - i\omega t] \right\rangle$$
$$= \Xi_1(0)\, \delta + \frac{c}{2} \left[-3\mu v_s k^2 \delta + \frac{\mu}{3} v_s (24k^2 \delta - 4\hat{\mathbf{k}}\hat{\mathbf{k}}) \right] + O(c^2) \quad (37)$$

where c is the number concentration of the particulate filler and $v_s = 4\pi a^3/3$ is the volume of a filler sphere.

Therefore, comparing Eq. (37) with Eq. (7) provides the identifications

$$\Xi_2(0, 0) = -\frac{2}{3} c\mu v_s + O(c^2)$$
$$\Xi_3(0, 0) = \frac{5}{2} c\mu v_s + O(c^2) \quad (38)$$

Hence, substituting Eq. (38) into Eq. (11) yields the effective Lamé coefficients to order ϕ as

$$\mu^* = \mu + \frac{5}{2} c\mu v_s = \mu(1 + 2.5\phi) \quad (39)$$

$$\lambda^* = \lambda + \left[-\frac{2}{3} c\mu v_s - \frac{5}{2} c\mu v_s \right] = \lambda - \frac{19}{6} \mu\phi \quad (40)$$

Using Eq. (12), we find for the Young's modulus and Poisson ratio for $\lambda \gg \mu$

$$\nu = \frac{1}{2} (1 - \mu/\lambda) + O[(\mu/\lambda)^2] \quad (41)$$

$$E = 2\mu(1 + \nu) + O(\mu/\lambda) \quad (42)$$

while the effective Young's modulus E^* and the Poisson ratio ν^* are found from Eqs. (39)–(42) to be

$$\nu^* = \nu - \frac{5}{4} (1 - 2\nu)\, \phi \quad (43)$$

$$E^* = E \left(1 + \frac{5}{4} \frac{1 + 4\nu}{1 + \nu}\, \phi \right) \quad (44)$$

The dilute-concentration limit with no-slip boundary conditions at the surface of particulate filler corresponds exactly to the limit considered by Muthukumar and Freed[3] for the suspension problem.

5. SUMMARY AND CONCLUSION

This paper demonstrates the general, yet formal structure of an effective medium equation for particulate filled systems. To complete the analysis, it is necessary to calculate Ξ_1, Ξ_2, and Ξ_3 for a given choice of boundary conditions and statistical distribution of the particulates. Exact calculation of the Ξ_i ($i = 1, 2, 3$) is an impossible task; however, it is possible to develop a systematic method for the approximate calculation of these quantities. A subsequent paper utilizes concentration expansions to derive new results for the Lamé constants for general v at low filler volume fractions. We show here how the Ξ_i are, in principle, extracted from experimental measurements such as wave propagation in the composite medium and the mechanical properties of the filler system.

The Ξ_i are determined here for the simplest physial situation (i.e., no-slip boundary conditions, dilute filler concentration, steady state, and $\lambda \to \infty$) by making an analogy between the particulate problem and the fluid mechanical suspension problem. The analogy is established by using an "elastic interaction tensor," which is analogous to the Oseen tensor in hydrodynamics. This elastic interaction tensor is shown to become structurally identical to the Oseen tensor, enabling us to adapt the results of Freed and Muthukumar[3] to evaluating Ξ_i.

Although the $\phi \to 0$, $\omega \to 0$, $\lambda \to \infty$ limit produces results identical to Einstein's theory of suspension, the importance of the current work should not be underestimated. Several important contributions from the new formulation are as follows.

1. Even though Einstein's original idea of energy dissipation was adapted by various authors to calculate the effective Lamé constant, the derivations in the literature do not lend themselves to systematic generalization of arbitrary ω, ϕ, and λ. The present paper, however, presents the most general mathematical structure for the dilute filler system by introducing modern field-theoretic techniques, which permit some of these extensions to be made.

2. It is possible to capitalize upon a vast amount of calculations obtained by Freed and Muthukumar by making an analogy in the study of the particulate system when $\lambda \gg \mu$. For example (see Fig. 3D), the filler system analogy of Huggins' coefficient,[18] of hydrodynamic screening,[3,4,16,17] and of the effect of the statistical distribution of

particulates[18,19] can all be obtained through analogy with the Freed–Muthukumar theory. These results and their comparison with experimental data will be presented elsewhere.

3. Most importantly, elsewhere[20] we show how a modification of the techniques of Freed and Muthukumar enables us to treat a dilute particulate system with an arbitrary value of λ. This extension requires substantial algebra, so this subsequent paper provides a further description of the explicit computations necessary to evaluate the Ξ_i, whereas here we focus on their definition, interpretation, and extraction from experimental data.

4. Further extensions are, in principle, possible to calculations for $\omega \neq 0$ and/or for higher filler concentrations. These interesting, but technically much more involved, computations are left for future work.

APPENDIX. REAL-SPACE REPRESENTATION OF ELASTIC INTERACTION TENSOR

To avoid added mathematical complications, we limit the analysis to the real-space representation of the elastic interaction tensor in a steady state [see Eq. (33)]. The evaluation of the Fourier inverse of $\mathbf{G}_s$ requires evaluation of the integrals

$$\begin{aligned}\iiint d^3k \exp(-i\mathbf{k}\cdot\mathbf{R})\, f_1(k^2)\,\boldsymbol{\delta} &= 2\pi \int_0^\infty k^2\,dk \int_{-1}^{1} d\mu \exp(-ikR\mu)\, f_1(k^2)\,\boldsymbol{\delta} \\ &= 2\pi \int_0^\infty k^2\,dk\, I_0(kR)\, f_1(k^2)\,\boldsymbol{\delta} \qquad \text{(A.1)}\end{aligned}$$

where

$$I_0(kR) \equiv \int_{-1}^{1} d\mu \exp(-ikR\mu) = \frac{2\sin(kR)}{kR}$$

and evaluation of

$$\begin{aligned}\iiint d^3k \exp(-i\mathbf{k}\cdot\mathbf{R})\, f_2(k^2)\,\hat{\mathbf{k}}\hat{\mathbf{k}} = \int_0^\infty k^2\,dk \int_{-1}^{1} d\mu \int_0^{2\pi} d\phi \exp(-ikR\mu)\, f_2(k^2)\,\hat{\mathbf{k}}\hat{\mathbf{k}} \qquad \text{(A.2)}\end{aligned}$$

We proceed by representing $\hat{\mathbf{k}}\hat{\mathbf{k}}$ in the coordinate system of Fig. 4. Use of Fig. 4 implies

$$\hat{\mathbf{k}} = \hat{\mathbf{R}}\mu + \hat{\mathbf{x}}vc + \hat{\mathbf{y}}vs$$

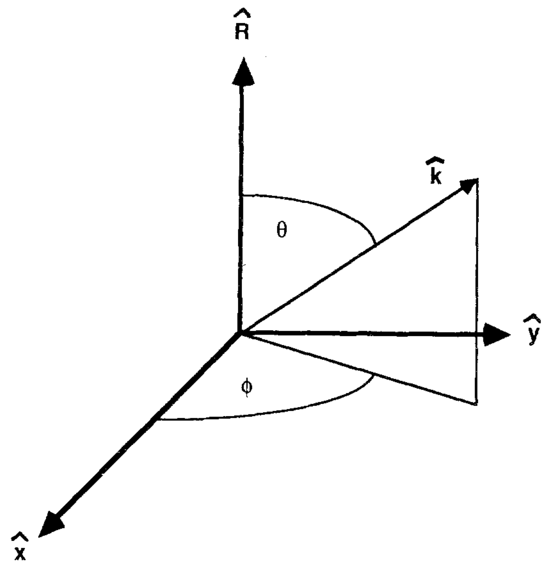


Fig. 4. Notation used for the coordinate system. From this figure, we define the following: $\hat{\mathbf{R}} = \mathbf{R}/R$; $\mathbf{R} = \mathbf{r} - \mathbf{r}'$; $\mu = \cos\theta$; $\nu = \sin\theta$; $c = \cos\phi$; $s = \sin\phi$.

and consequently we have

$$\hat{\mathbf{k}}\hat{\mathbf{k}} = \hat{\mathbf{R}}\hat{\mathbf{R}}\mu^2 + \hat{\mathbf{x}}\hat{\mathbf{x}}\nu^2 c^2 + \hat{\mathbf{y}}\hat{\mathbf{y}}\nu^2 s^2 + \text{cross term} \tag{A.3}$$

Therefore, integrating Eq. (A.3) makes the cross terms vanish and leaves

$$\int_0^{2\pi} d\phi\, \hat{\mathbf{k}}\hat{\mathbf{k}} = 2\pi\left[\hat{\mathbf{R}}\hat{\mathbf{R}}\mu^2 + \frac{1}{2}(\hat{\mathbf{x}}\hat{\mathbf{x}} + \hat{\mathbf{y}}\hat{\mathbf{y}})\,\nu^2\right]$$
$$= \pi[(\delta - \hat{\mathbf{R}}\hat{\mathbf{R}}) - (\delta - 3\hat{\mathbf{R}}\hat{\mathbf{R}})\,\mu^2] \tag{A.4}$$

Substituting (A.4) into (A.2) gives

$$\iiint d^3k \exp(-i\mathbf{k}\cdot\mathbf{R})\, f_2(k^2)\, \hat{\mathbf{k}}\hat{\mathbf{k}}$$
$$= \pi\int_0^\infty k^2\,dk \int_{-1}^{1} d\mu \exp(-ikR\mu)\, f_2(k^2)[(\delta - \hat{\mathbf{R}}\hat{\mathbf{R}}) - (\delta - 3\hat{\mathbf{R}}\hat{\mathbf{R}})\,\mu^2]$$
$$= \pi\int_0^\infty k^2\,dk\, f_2(k^2)\left[I_0(kR)(\delta - \hat{\mathbf{R}}\hat{\mathbf{R}}) + \frac{\partial^2 I_0(kR)}{\partial(kR)^2}(\delta - 3\hat{\mathbf{R}}\hat{\mathbf{R}})\right]$$
$$= \pi\int_0^\infty k^2\,dk\, f_2(k^2)\frac{\sin(kR)}{kR}(\delta - \hat{\mathbf{R}}\hat{\mathbf{R}}) \tag{A.5}$$

Combining Eqs. (33), (A.1), and (A.5) produces the final real-space form of the elastic propagator given in Eq. (34),

$$
\begin{aligned}
\mathrm{G}_s(R) &= \iiint \frac{d^3k}{(2\pi)^3}\left[\frac{1}{\mu k^2}(\delta - \hat{\mathbf{k}}\hat{\mathbf{k}}) + \frac{\hat{\mathbf{k}}\hat{\mathbf{k}}}{(\lambda+2\mu)\,k^2}\right]\exp(-i\mathbf{k}\cdot\mathbf{R}) \\
&= \frac{4\pi}{(2\pi)^3}\int_0^\infty k^2\,dk\,\frac{1}{\mu k^2}\frac{\sin(kR)}{kR}\left[\delta - \frac{1}{2}(\delta - \hat{\mathbf{R}}\hat{\mathbf{R}})\right] \\
&\quad + \frac{(2\pi)}{(2\pi)^2}\int_0^\infty k^2\,dk\,\frac{\hat{\mathbf{k}}\hat{\mathbf{k}}}{(\lambda+2\mu)\,k^2} \\
&= \frac{\pi^2}{(2\pi)^3\,\mu R}(\delta + \hat{\mathbf{R}}\hat{\mathbf{R}}) + \frac{\pi^2}{(2\pi)^3\,(\lambda+2\mu)\,R}(\delta - \hat{\mathbf{R}}\hat{\mathbf{R}}) \\
&= \frac{1}{8\pi\mu R}(\delta + \hat{\mathbf{R}}\hat{\mathbf{R}}) + \frac{1}{8\pi(\lambda+2\mu)\,R}(\delta - \hat{\mathbf{R}}\hat{\mathbf{R}})
\end{aligned}
$$

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