# New bounds on the elastic moduli of suspensions of spheres

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We derive rigorous three-point upper and lower bounds on the effective bulk and shear moduli of a two-phase material composed of equisized spheres randomly distributed throughout a matrix. Our approach is analogous to previously derived three-point cluster bounds on the effective conductivity of suspensions of spheres. Our bounds on the effective elastic moduli are then compared to other known three-point bounds for statistically homogeneous and isotropic random materials. For the case of totally impenetrable spheres, the bulk modulus bounds are shown to be equivalent to the Beran–Molyneux bounds, and the shear modulus bounds are compared to the McCoy and Milton–Phan-Thien bounds. For the case of fully penetrable spheres, our bounds are shown to be simple analytical expressions, in contrast to the numerical quadratures required to evaluate the other three-point bounds. © *1995 American Institute of Physics.*

## I. INTRODUCTION

The problem of determining the effective properties of composite media has attracted the attention of some of the luminaries of science[1–3] and continues to be the focus of intense research.[4–13] The effective property of a composite generally depends on the phase properties, phase volume fractions, and the microstructure through an infinite set of correlation functions that statistically characterize it. The major problem is that this set of functions is typically unknown either experimentally or theoretically. Therefore, one usually resorts to obtaining a solution for an idealized geometry (e.g., periodic arrays), finding an approximate solution, or obtaining rigorous bounds on the effective properties for the actual microstructure given limited but nontrivial information about it.[4–13] The latter approach has been found to be fruitful because the bounds can yield useful estimates of the effective properties, even when the reciprocal bound diverges from it in the strong-contrast limit. In the case of the elastic moduli of isotropic two-phase composites, the subject of the present paper, two-point[4,5] and three-point bounds[7,8,10,11] have been derived. By $n$-point bounds we mean bounds that incorporate up to $n$-point correlation function information.

In this work we will derive rigorous three-point upper and lower bounds on the effective bulk modulus $\kappa_e$ and effective shear modulus $\mu_e$ of a class of two-phase composites, namely, suspensions of identical spheres *that interact with one another with an arbitrary potential.* These are referred to as "cluster bounds" since they represent the analog of previously obtained cluster bounds for the effective conductivity of suspensions.[12] Our bounds are evaluated for two extreme situations: totally impenetrable (nonoverlapping) spheres and fully penetrable spheres.

In the case of totally impenetrable spheres, our bounds are evaluated in terms of the known microstructural parameters $\zeta_2$ and $\eta_2$. We find that the cluster bounds on the effective bulk modulus are equal to the Beran–Molyneux bounds (cf. the cluster bounds on effective conductivity[14]). Also, we show that the cluster bounds may or may not be sharper than the McCoy bounds, depending on the volume fraction of inclusions and the elastic moduli of the two phases.

For fully penetrable spheres (i.e., spatially uncorrelated spheres), our cluster bounds can be evaluated exactly analytically, in contrast to previous bounds which require numerical quadratures.[15–18] This is an interesting model because for sphere volume fractions $\phi_2$ in the interval [0.3,0.97] the medium is bicontinuous; i.e., both phases are connected. Thus, a system of randomly overlapping spheres is useful model of consolidated media, such as sandstones and sintered materials.

In Section II we state the variational principles for the effective elastic moduli from which the cluster bounds arise. In Section III we describe the class of model microstructures that will be studied and the associated relevant statistical correlation functions. In Sections IV and V we explicitly calculate the ensemble averages involved in the cluster bounds on the effective bulk and shear modulus, respectively. In Section VI we simplify these two pairs of bounds for the special case of totally impenetrable spheres and compare our results with other bounds on the effective elastic moduli. The same is done for randomly overlapping spheres in Section VII.

## II. VARIATIONAL PRINCIPLES

### A. Principle of minimum potential energy

Consider a trial strain field $\hat{\boldsymbol{\epsilon}}(\mathbf{r})$ at the field point $\mathbf{r}$, i.e., a field that can be written as a symmetrized gradient of displacements, or

$$\hat{\epsilon}_{ij}=\frac{1}{2}\left(\frac{\partial\hat{u}_j}{\partial x_i}+\frac{\partial\hat{u}_i}{\partial x_j}\right). \tag{2.1}$$

We require that the ensemble average $\langle\hat{\boldsymbol{\epsilon}}\rangle$ is equal to the average of the actual strain field $\langle\boldsymbol{\epsilon}\rangle$. Let

$$U(\hat{\boldsymbol{\epsilon}})=\tfrac{1}{2}\langle C_{ijkl}\hat{\epsilon}_{ij}\hat{\epsilon}_{kl}\rangle \tag{2.2}$$

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be the potential energy of the system for the trial strain field, where $C_{ijkl}(\mathbf{r})$ is the local stiffness tensor defined by

$$\tau_{ij}=C_{ijkl}\epsilon_{kl} \tag{2.3}$$

and the Einstein summation convention is employed. Then among all trial strain fields, the field that satisfies the equations

$$\frac{\partial}{\partial x_j}C_{ijkl}\hat{\epsilon}_{kl}=0 \tag{2.4}$$

is the one that uniquely minimizes $U(\hat{\boldsymbol{\epsilon}})$.[6]

For isotropic systems, this implies that

$$\kappa_e\langle\epsilon_{ii}\rangle\langle\epsilon_{kk}\rangle+2\mu_e\langle\tilde{\epsilon}_{ij}\rangle\langle\tilde{\epsilon}_{ij}\rangle\leq\langle\kappa\hat{\epsilon}_{ii}\hat{\epsilon}_{kk}\rangle+2\langle\mu\tilde{\hat{\epsilon}}_{ij}\tilde{\hat{\epsilon}}_{ij}\rangle, \tag{2.5}$$

where $\kappa_e$ is the effective bulk modulus, $\mu_e$ the effective shear modulus, and

$$\tilde{\epsilon}_{ij}=\epsilon_{ij}-\tfrac{1}{3}\epsilon_{kk}\delta_{ij} \tag{2.6}$$

is the deviatoric component of the strain field $\boldsymbol{\epsilon}$.

We now take the trial strain field to be of the form

$$\hat{\boldsymbol{\epsilon}}=\langle\boldsymbol{\epsilon}\rangle+\lambda\boldsymbol{\epsilon}', \tag{2.7}$$

where $\lambda$ is a parameter and $\boldsymbol{\epsilon}'$ is a perturbation term chosen so that $\hat{\boldsymbol{\epsilon}}$ satisfies the requirements of a trial strain field. Setting $\langle\boldsymbol{\epsilon}\rangle=\mathbf{I}$, the unit dyadic, and minimizing over $\lambda$ yields the following rigorous upper bound on the effective bulk modulus:

$$\kappa_e\leq\langle\kappa\rangle-\frac{\langle\kappa\epsilon'_{ii}\rangle^2}{\langle\kappa\epsilon'_{ii}\epsilon'_{kk}\rangle+2\langle\mu\tilde{\epsilon}'_{ij}\tilde{\epsilon}'_{ij}\rangle}. \tag{2.8}$$

Likewise, setting $\langle\epsilon_{ij}\rangle=\delta_{i1}\delta_{j1}-\delta_{i2}\delta_{j2}$ and minimizing over $\lambda$ yields an upper bound on the effective shear modulus:

$$\mu_e\leq\langle\mu\rangle-\frac{\langle\mu(\epsilon'_{11}-\epsilon'_{22})\rangle^2}{\langle\kappa\epsilon'_{ii}\epsilon'_{kk}\rangle+2\langle\mu\tilde{\epsilon}'_{ij}\tilde{\epsilon}'_{ij}\rangle}. \tag{2.9}$$

## B. Principle of minimum complementary energy

We now consider trial stress fields $\hat{\boldsymbol{\tau}}(\mathbf{r})$ that are symmetric and satisfy the equilibrium equations

$$\frac{\partial\hat{\tau}_{ij}}{\partial x_j}=0, \tag{2.10}$$

where $\langle\hat{\boldsymbol{\tau}}\rangle$ is required to equal the average of the actual stress field $\langle\boldsymbol{\tau}\rangle$. Let

$$T(\hat{\boldsymbol{\tau}})=\tfrac{1}{2}\langle S_{ijkl}\hat{\tau}_{ij}\hat{\tau}_{kl}\rangle, \tag{2.11}$$

where $S_{ijkl}$ is the compliance tensor defined by

$$\epsilon_{ij}=S_{ijkl}\tau_{kl}. \tag{2.12}$$

Then among all trial stress fields, the field that is derivable from a displacement field

$$\hat{\tau}_{ij}=C_{ijkl}\frac{1}{2}\left(\frac{\partial\hat{u}_j}{\partial x_i}+\frac{\partial\hat{u}_i}{\partial x_j}\right) \tag{2.13}$$

is the one that uniquely minimizes $T(\hat{\boldsymbol{\tau}})$.[6]

For isotropic systems, this implies that

$$\frac{\kappa_e^{-1}}{9}\langle\tau_{kk}\rangle^2+\frac{\mu_e^{-1}}{2}\langle\tilde{\tau}_{ij}\rangle\langle\tilde{\tau}_{ij}\rangle\leq\frac{1}{9}\langle\kappa^{-1}\hat{\tau}_{ii}\hat{\tau}_{kk}\rangle+\tfrac{1}{2}\langle\mu^{-1}\tilde{\hat{\tau}}_{ij}\tilde{\hat{\tau}}_{ij}\rangle, \tag{2.14}$$

where $\tilde{\boldsymbol{\tau}}$ is the deviatoric component of the stress field. As before, we now take

$$\hat{\boldsymbol{\tau}}=\langle\boldsymbol{\tau}\rangle+\lambda\boldsymbol{\tau}', \tag{2.15}$$

where $\lambda$ is a parameter and $\boldsymbol{\tau}'$ is a perturbation term chosen so that $\hat{\boldsymbol{\tau}}$ satisfies the requirements of a trial stress field. Setting $\langle\boldsymbol{\epsilon}\rangle=\mathbf{I}$, so that $\langle\boldsymbol{\tau}\rangle=3\kappa_1\mathbf{I}$, and minimizing over $\lambda$ yields a lower bound on the effective bulk modulus:

$$\kappa_e^{-1}\leq\langle\kappa^{-1}\rangle-\frac{\frac{1}{9}\langle\kappa^{-1}\tau'_{ii}\rangle^2}{\frac{1}{9}\langle\kappa^{-1}\tau'_{ii}\tau'_{kk}\rangle+\frac{1}{2}\langle\mu^{-1}\tilde{\tau}'_{ij}\tilde{\tau}'_{ij}\rangle}. \tag{2.16}$$

Likewise, setting $\langle\epsilon_{ij}\rangle=\delta_{i1}\delta_{j1}-\delta_{i2}\delta_{j2}$, so that $\langle\tau_{ij}\rangle=2\mu_1(\delta_{i1}\delta_{j1}-\delta_{i2}\delta_{j2})$, and minimizing over $\lambda$ yields a lower bound on the effective shear modulus:

$$\mu_e^{-1}\leq\langle\mu^{-1}\rangle-\frac{\frac{1}{4}\langle\mu^{-1}(\tau'_{11}-\tau'_{22})\rangle^2}{\frac{1}{9}\langle\kappa^{-1}\tau'_{ii}\tau'_{kk}\rangle+\frac{1}{2}\langle\mu^{-1}\tilde{\tau}'_{ij}\tilde{\tau}'_{ij}\rangle}. \tag{2.17}$$

Choices for $\boldsymbol{\epsilon}'$ and $\boldsymbol{\tau}'$ must be made to obtain lower and upper bounds on the effective elastic moduli. This is shown in Sections IV and V. Before undertaking this, however, we first mathematically describe the model microstructure of the composite material.

# III. MODEL SYSTEM AND MICROSTRUCUTURE CHARACTERIZATION

We now consider systems of volume $V$ with a matrix phase (phase 1) with bulk and shear modulus $\kappa_1$, $\mu_1$, respectively, and $N$ possibly overlapping spherical inclusions (phase 2) of radius $R$ with bulk and shear modulus $\kappa_2$, $\mu_2$, respectivély. The ensembles are assumed to be statistically homogeneous and thus we ultimately take the "thermodynamic limit" $N,V\rightarrow\infty$ such that the number density $N/V$ equals some fixed constant $\rho$. The volume fractions of matrix and spheres are $\phi_1$ and $\phi_2$, respectively. The centers $\mathbf{r}^N=\mathbf{r}_1,\ldots,\mathbf{r}_N$ are randomly positioned and follow a known density function $P_N(\mathbf{r}^N)$. Therefore, if $F(\mathbf{r}^N)$ is a given many-body function, then the ensemble average of $F$ is given by

$$\langle F\rangle=\int\ldots\int d\mathbf{r}^NF(\mathbf{r}^N)P_N(\mathbf{r}^N), \tag{3.1}$$

where $d\mathbf{r}^N=d\mathbf{r}_1\ldots d\mathbf{r}_N$.

The reduced $n$-particle probability density $P_n(\mathbf{r}^n)$ is defined by

$$P_n(\mathbf{r}^n)=\int\ldots\int d\mathbf{r}_{n+1}\ldots d\mathbf{r}_NP_N(\mathbf{r}^N). \tag{3.2}$$

This is the probability of observing molecule $i$ in a volume $d\mathbf{r}_i$ about $\mathbf{r}_i$ for $i=1,\ldots,n$. Therefore, the probability of finding any molecule in a volume $d\mathbf{r}_i$ about $\mathbf{r}_i$ for $i=1,\ldots,n$ is given by

$$\rho_n(\mathbf{r}^n)=[N!/(N-n)!]P_n(\mathbf{r}^n). \tag{3.3}$$

This is called the *generic* $n$-particle probability density function.

Subject to the conditions stated above, the potential of interaction between the spheres is perfectly arbitrary. The spheres may overlap to varying degrees; an example of such an interpenetrable-sphere system is the penetrable-concentric-shell (or cherry-pit) model.[19]

Quantities that statistically describe the microstructure of random heterogeneous materials are now defined; a fuller development is given in Ref. 12. The number density $\rho$ and the reduced density $\eta$ are given by

$$\rho = \lim_{N,V\to\infty} N/V \tag{3.4}$$

and

$$\eta = \frac{4\pi\rho R^3}{3}. \tag{3.5}$$

For partially penetrable spheres the inclusion volume fraction $\phi_2$ can be related to the reduced density. For example, for fully penetrable and totally impenetrable spheres $\phi_2 = 1 - e^{-\eta}$ and $\phi_2 = \eta$, respectively. General expressions which relate $\phi_2$ as a function of $\eta$ for arbitrary $\lambda$ have been obtained for the cherry-pit model.[20]

The distribution function associated with finding phase $i$ at $\mathbf{r}$ and a particular configuration of $q = n-1$ spheres at positions $\mathbf{r}^q$ is defined as

$$G_n^{(i)}(\mathbf{r};\mathbf{r}^q) = \frac{N!}{(N-q)!}\int \dots \int d\mathbf{r}_n \dots d\mathbf{r}_N I^{(i)}(\mathbf{r},\mathbf{r}^N) P_N(\mathbf{r}^N), \tag{3.6}$$

where $I^{(i)}(\mathbf{r})$ is the characteristic function for phase $i$; i.e.,

$$I^{(i)}(\mathbf{r}) = \begin{cases} 1, & \mathbf{r}\in D_i, \\ 0, & \text{otherwise,} \end{cases} \tag{3.7}$$

and $D_i$ is the region of space occupied by phase $i$. We call $G_n^{(i)}(\mathbf{r};\mathbf{r}^q)$ the phase point/$q$-particle function for phase $i$. It is convenient to define another set of point/$q$-particle distribution functions $H_n^{(i)}$ as follows:

$$H_n^{(i)}(\mathbf{r};\mathbf{r}^q) = G_n^{(i)}(\mathbf{r};\mathbf{r}^q) - G_0^{(i)}(\mathbf{r})\rho_q(\mathbf{r}^q), \tag{3.8}$$

so that $H_n^{(i)} \to 0$ as $\mathbf{r}$ moves away from $\mathbf{r}_1,\dots,\mathbf{r}_q$.

In this paper, we are restricting our attention to ensembles of spheres which are statistically homogeneous and isotropic. For such materials, these correlation functions are only dependent on the relative positions of the $n$ points. For example, $\rho_1(\mathbf{r}_1) = \rho$, $G_2^{(i)}(\mathbf{r};\mathbf{r}_1) = G_2^{(i)}(x_1)$ and $G_3^{(i)}(\mathbf{r};\mathbf{r}_1,\mathbf{r}_2) = G_3^{(i)}(x_1,x_2,\hat{\mathbf{x}}_1\cdot\hat{\mathbf{x}}_2)$, where $\mathbf{x}_i = \mathbf{r}-\mathbf{r}_i$, $x_i = |\mathbf{x}_i|$, and $\hat{\mathbf{x}}_i = \mathbf{x}_i/x_i$. It is easily shown that

$$G_2^{(2)}(x) = \rho, \quad x<R \tag{3.9}$$

and

$$H_2^{(2)}(x) = \rho\phi_1, \quad x<R. \tag{3.10}$$

Finally, the radial distribution function $g_2(x)$ and the total correlation function $h(x)$ are defined by

$$g_2(x) = \rho_2(x)/\rho^2 \tag{3.11}$$

and

$$h(x) = g_2(x) - 1. \tag{3.12}$$

## IV. GENERAL CLUSTER BOUNDS ON THE EFFECTIVE BULK MODULUS

Torquato[12] has obtained so-called cluster bounds on the effective conductivity of dispersions by employing trial fields based upon the solution of the single-sphere boundary-value problem. In this and the next section, we shall derive analogous bounds on the effective elastic moduli using such trial fields.

To obtain the cluster bounds on $\kappa_e$, we use Eqs. (2.5) and (2.11) and choose

$$\boldsymbol{\epsilon}'(\mathbf{r};\mathbf{r}^N) = \sum_{i=1}^{N} \boldsymbol{\epsilon}^*(\mathbf{x}_i) - \int d\mathbf{x}_1 \rho(\mathbf{x}_1)\boldsymbol{\epsilon}^*(\mathbf{x}_1) \tag{4.1}$$

and

$$\boldsymbol{\tau}'(\mathbf{r};\mathbf{r}^N) = \sum_{i=1}^{N} \boldsymbol{\tau}^*(\mathbf{x}_i) - \int d\mathbf{x}_1 \rho(\mathbf{x}_1)\boldsymbol{\tau}^*(\mathbf{x}_1), \tag{4.2}$$

where

$$\boldsymbol{\epsilon}^*(x_i) = \begin{cases} -\alpha\mathbf{I}, & x_i<R, \\ \dfrac{\alpha R^3}{r^3}(3\hat{\mathbf{x}}_i\hat{\mathbf{x}}_i - \mathbf{I}), & x_i>R, \end{cases} \tag{4.3}$$

$$\boldsymbol{\tau}^*(\mathbf{x}_i) = \begin{cases} 4\alpha\mu_1\mathbf{I}, & x_i<R, \\ \dfrac{2\alpha\mu_1 R^3}{r^3}(3\hat{\mathbf{x}}_i\hat{\mathbf{x}}_i - \mathbf{I}), & x_i>R, \end{cases} \tag{4.4}$$

$$\mathbf{x}_i = \mathbf{r}-\mathbf{r}_i, \quad x_i = |\mathbf{x}_i|,$$

and

$$\alpha = 3(\kappa_2-\kappa_1)/(3\kappa_2+4\mu_1). \tag{4.5}$$

These fields arise from the solution of the single-sphere boundary value problem, which is described in Appendix A.

The ensembled averaged quantities of Eqs. (2.8) and (2.16) are similar to those derived in Ref. 12. After some algebraic manipulation, which is outlined in Appendix B, they are given by

$$\langle \kappa\epsilon'_{ii}\rangle = -12\pi\alpha(\kappa_2-\kappa_1)I_1, \tag{4.6}$$

$$\langle \kappa\epsilon'_{ii}\epsilon'_{kk}\rangle = \alpha^2[A_1\kappa_1 + B_1(\kappa_2-\kappa_1)], \tag{4.7}$$

$$\langle \mu\tilde{\epsilon}'_{ij}\tilde{\epsilon}'_{ij}\rangle = \alpha^2[C_1\mu_1 + D_1(\mu_2-\mu_1)], \tag{4.8}$$

$$\langle \kappa^{-1}\tau'_{ii}\rangle = 48\pi\alpha\mu_1(\kappa_2^{-1}-\kappa_1^{-1})I_1, \tag{4.9}$$

$$\langle \kappa^{-1}\tau'_{ii}\tau'_{kk}\rangle = 16\mu_1^2\alpha^2[A_1\kappa_1^{-1} + B_1(\kappa_2^{-1}-\kappa_1^{-1})], \tag{4.10}$$

$$\langle \mu^{-1}\tilde{\tau}'_{ij}\tilde{\tau}'_{ij}\rangle = 4\mu_1^2\alpha^2[C_1\mu_1^{-1} + D_1(\mu_2^{-1}-\mu_1^{-1})], \tag{4.11}$$

where

$$A_1 = \langle \epsilon'_{ii}\epsilon'_{kk}\rangle = 9\eta + \frac{81}{2R^6}\eta^2 I_2, \tag{4.12}$$

$$B_1=\langle I^{(2)}\epsilon'_{ii}\epsilon'_{kk}\rangle=\frac{27\eta}{R^3\rho}I_3+\frac{81\eta^2}{2R^6\rho^2}I_4, \tag{4.13}$$

$$C_1=\langle\tilde{\epsilon}'_{ij}\tilde{\epsilon}'_{ij}\rangle=6\eta+27\eta^2 I_5, \tag{4.14}$$

$$D_1=\langle I^{(2)}\tilde{\epsilon}'_{ij}\tilde{\epsilon}'_{ij}\rangle=\frac{18\eta R^3}{\rho}I_6+\frac{27\eta^2}{\rho^2}I_7, \tag{4.15}$$

$$I_1=\int_0^R dr\, r^2 H_2^{(2)}(r), \tag{4.16}$$

$$I_2=\int_0^R dr\, r^2\int_0^R ds s^2\int_{-1}^1 du\, h(t), \tag{4.17}$$

$$I_3=\int_0^R dr\, r^2 G_2^{(2)}(r), \tag{4.18}$$

$$I_4=\int_0^R dr\, r^2\int_0^R ds\, s^2\int_{-1}^1 du\, Q(\mathbf{r},\mathbf{s}), \tag{4.19}$$

$$I_5=\int_R^\infty\frac{dr}{r}\int_R^\infty\frac{ds}{s}\int_{-1}^1 du\, h(t)P_2(u), \tag{4.20}$$

$$I_6=\int_R^\infty dr\,\frac{G_2^{(2)}(r)}{r^4}, \tag{4.21}$$

$$I_7=\int_R^\infty\frac{dr}{r}\int_R^\infty\frac{ds}{s}\int_{-1}^1 du Q(\mathbf{r},\mathbf{s})P_2(u), \tag{4.22}$$

and

$$Q(\mathbf{r},\mathbf{s})=G_3^{(2)}(r,s,u)-\rho G_2^{(2)}(r)-\rho G_2^{(2)}(s)+\rho^2\phi_2. \tag{4.23}$$

In these equations, $P_2(u)$ is the Legendre polynomial of order 2 [not to be confused with the reduced two-particle probability density function of Eq. (3.2)],

$$u=\frac{r^2+s^2-t^2}{2rs}, \tag{4.24}$$

and

$$t=|\mathbf{r}-\mathbf{s}|. \tag{4.25}$$

To summarize, using Eqs. (2.8), (2.16), (3.10), and (4.6)–(4.11), for an isotropic composite system consisting of overlapping equi-sized spheres dispersed throughout a matrix the effective bulk modulus is bounded by

$$\kappa_e\leq\langle\kappa\rangle-\frac{9\eta^2\phi_1^2(\kappa_2-\kappa_1)^2}{A_1\kappa_1+B_1(\kappa_2-\kappa_1)+2[C_1\mu_1+D_1(\mu_2-\mu_1)]} \tag{4.26}$$

and

$$\kappa_e\geq\left(\langle\kappa^{-1}\rangle-\frac{4\eta^2\phi_1^2(\kappa_2^{-1}-\kappa_1^{-1})^2}{\frac{4}{9}[A_1\kappa_1^{-1}+B_1(\kappa_2^{-1}-\kappa_1^{-1})]+\frac{1}{2}[C_1\mu_1^{-1}+D_1(\mu_2^{-1}-\mu_1^{-1})]}\right)^{-1}, \tag{4.27}$$

where $A_1$, $B_1$, $C_1$, and $D_1$ are given by Eqs. (4.12)–(4.15).

## V. GENERAL CLUSTER BOUNDS ON THE EFFECTIVE SHEAR MODULUS

To obtain the cluster bounds on $\mu_e$, we likewise use Eqs. (2.9) and (2.17) and again choose

$$\boldsymbol{\epsilon}'(\mathbf{r};\mathbf{r}^N)=\sum_{i=1}^N\boldsymbol{\epsilon}^*(\mathbf{x}_i)-\int d\mathbf{x}_1\rho(\mathbf{x}_1)\boldsymbol{\epsilon}^*(\mathbf{x}_1) \tag{5.1}$$

and

$$\boldsymbol{\tau}'(\mathbf{r};\mathbf{r}^N)=\sum_{i=1}^N\boldsymbol{\tau}^*(\mathbf{x}_i)-\int d\mathbf{x}_1\rho(\mathbf{x}_1)\boldsymbol{\tau}^*(\mathbf{x}_1), \tag{5.2}$$

where for the strain perturbation field we instead take

$$\epsilon_{ij}^*=c_1(\delta_{i1}\delta_{j1}-\delta_{i2}\delta_{j2}),\quad r<R, \tag{5.3}$$

while for $r>R$ we take

$$\epsilon_{11}^*=\frac{\partial p}{\partial x}x(x^2-y^2)+p(r)(3x^2-y^2)+\frac{\partial q}{\partial x}x+q(r), \tag{5.4}$$

$$\epsilon_{12}^*=\epsilon_{21}^*=\frac{1}{2}(x^2-y^2)\left(\frac{\partial p}{\partial y}x+\frac{\partial p}{\partial x}y\right)+\frac{1}{2}\left(\frac{\partial q}{\partial y}x-\frac{\partial q}{\partial x}y\right), \tag{5.5}$$

$$\epsilon_{13}^*=\epsilon_{31}^*=\frac{1}{2}(x^2-y^2)\left(\frac{\partial p}{\partial z}x+\frac{\partial p}{\partial x}z\right)+p(r)xz+\frac{1}{2}\frac{\partial q}{\partial z}x, \tag{5.6}$$

$$\epsilon_{22}^*=\frac{\partial p}{\partial y}y(x^2-y^2)+p(r)(x^2-3y^2)-\frac{\partial q}{\partial y}y-q(r), \tag{5.7}$$

$$\epsilon_{23}^*=\epsilon_{32}^*=\frac{1}{2}(x^2-y^2)\left(\frac{\partial p}{\partial z}y+\frac{\partial p}{\partial y}z\right)-p(r)yz-\frac{1}{2}\frac{\partial q}{\partial z}y, \tag{5.8}$$

and

$$\epsilon_{33}^*=\frac{\partial p}{\partial z}z(x^2-y^2)+p(r)(x^2-y^2), \tag{5.9}$$

where

$$p(r)=\frac{5c_2}{r^7}+\frac{3\kappa_1+\mu_1}{\mu_1}\frac{c_3}{r^5}, \tag{5.10}$$

$$q(r)=-\frac{2c_2}{r^5}+\frac{2c_3}{r^3}, \tag{5.11}$$

$$c_1=6(\kappa_1+2\mu_1)\gamma, \tag{5.12}$$

$$c_2=-R^5(3\kappa_1+\mu_1)\gamma, \tag{5.13}$$

$$c_3=5R^3\mu_1\gamma, \tag{5.14}$$

and

$$\gamma=\frac{\mu_1-\mu_2}{9\mu_1\kappa_1+6\mu_2\kappa_1+8\mu_1^2+12\mu_1\mu_2}. \tag{5.15}$$

For the stress perturbation field, we take

$$\tau_{ij}^*=\begin{cases}2c_4[\delta_{i1}\delta_{j1}-\delta_{i2}\delta_{j2}], & r<R,\\ (\kappa_1-\frac{2}{3}\mu_1)\epsilon_{kk}\delta_{ij}+2\mu_1\epsilon_{ij}^*, & r>R,\end{cases} \tag{5.16}$$

where

$$c_4=\mu_2-\mu_1+\mu_2c_1=-\mu_1(9\kappa_1+8\mu_1)\gamma. \tag{5.17}$$

As in the previous section, these perturbation fields also arise from the solution of the single-sphere boundary value problem discussed in Appendix A.

As in the previous section, we now simplify the ensemble averages of Eq. (2.9) and (2.17). We find that

$$\langle\mu(\epsilon_{11}'-\epsilon_{22}')\rangle=8\pi c_1(\mu_2-\mu_1)I_1, \tag{5.18}$$

$$\langle\kappa\epsilon_{ii}'\epsilon_{kk}'\rangle=A_2\kappa_1+B_2(\kappa_2-\kappa_1), \tag{5.19}$$

$$\langle\mu\tilde{\epsilon}_{ij}'\tilde{\epsilon}_{ij}'\rangle=C_2\mu_1+D_2(\mu_2-\mu_1), \tag{5.20}$$

$$\langle\mu^{-1}(\tau_{11}'-\tau_{22}')\rangle=16\pi c_4(\mu_2^{-1}-\mu_1^{-1})I_1, \tag{5.21}$$

$$\langle\kappa^{-1}\tau_{ii}'\tau_{kk}'\rangle=A_3\kappa_1^{-1}+B_3(\kappa_2^{-1}-\kappa_1^{-1}), \tag{5.22}$$

$$\langle\mu^{-1}\tilde{\tau}_{ij}'\tilde{\tau}_{ij}'\rangle=C_3\mu_1^{-1}+D_3(\mu_2^{-1}-\mu_1^{-1}), \tag{5.23}$$

where

$$A_2=\frac{48c_3^2}{5R^6}\eta+\frac{216c_3^2}{5R^6}\eta^2I_5, \tag{5.24}$$

$$B_2=\frac{144c_3^2}{5R^3\rho}\eta I_6+\frac{216c_3^2}{5R^6\rho^2}\eta^2I_7, \tag{5.25}$$

$$\begin{aligned}C_2=&\left[2c_1^2+48\left(\frac{c_2^2}{R^{10}}+\frac{2c_2c_3(3\kappa_1+\mu_1)}{5\mu_1R^8}\right.\right.\\&\left.\left.+\frac{c_3^2(27\kappa_1^2+24\kappa_1\mu_1+16\mu_1^2)}{60\mu_1^2R^6}\right)\right]\eta\\&+\left(\frac{9c_1^2}{R^6}I_2+\frac{18c_3^2(9\kappa_1^2+48\kappa_1\mu_1+92\mu_1^2)}{35\mu_1^2R^6}I_5\right.\\&\left.+\frac{72}{7R^6}I_{10}\right)\eta^2-\frac{A_2}{3},\end{aligned} \tag{5.26}$$

$$\begin{aligned}D_2=&\left(\frac{6c_1^2}{R^3\rho}I_3+\frac{48}{R^3\rho}I_8\right)\eta+\left(\frac{9c_1^2}{R^6\rho^2}I_4\right.\\&+\frac{18c_3^2(9\kappa_1^2+48\kappa_1\mu_1+92\mu_1^2)}{35\mu_1^2R^6\rho^2}I_7\\&\left.+\frac{72}{7R^6\rho^2}I_{11}\right)\eta^2-\frac{B_2}{3},\end{aligned} \tag{5.27}$$

$$A_3=9\kappa_1^2A_2, \tag{5.28}$$

$$B_3=9\kappa_1^2B_2, \tag{5.29}$$

$$\begin{aligned}C_3=&\left[8c_4^2+192\left(\frac{c_2^2\mu_1^2}{R^{10}}+\frac{2c_2c_3\mu_1(3\kappa_1+\mu_1)}{5R^8}\right.\right.\\&\left.\left.+\frac{c_3^2(3\kappa_1^2+2\kappa_1\mu_1+\mu_1^2)}{5R^6}\right)\right]\eta\\&+\left(\frac{36}{R^6}c_4^2I_2+\frac{576c_3^2(9\kappa_1^2+6\kappa_1\mu_1+8\mu_1^2)}{35R^6}I_5\right.\\&\left.+\frac{288\mu_1^2}{7R^6}I_{10}\right)\eta^2-\frac{A_3}{3},\end{aligned} \tag{5.30}$$

and

$$\begin{aligned}D_3=&\left(\frac{24}{R^3\rho}c_4^2I_3+\frac{192}{5R^3\rho}I_9\right)\eta+\left(\frac{36}{R^6\rho^2}c_4^2I_4\right.\\&\left.+\frac{576c_3^2(9\kappa_1^2+6\kappa_1\mu_1+8\mu_1^2)}{35R^6\rho^2}I_7+\frac{288\mu_1^2}{7R^6\rho^2}I_{11}\right)\\&\times\eta^2-\frac{B_3}{3}.\end{aligned} \tag{5.31}$$

In these equations $I_1$ through $I_7$ are given by Eqs. (4.16)–(4.22),

$$\begin{aligned}I_8=&\int_R^\infty drG_2^{(2)}(r)\left(\frac{7c_2^2}{r^8}+\frac{2c_2c_3(3\kappa_1+\mu_1)}{\mu_1r^6}\right.\\&\left.+\frac{c_3^2(27\kappa_1^2+24\kappa_1\mu_1+16\mu_1^2)}{20\mu_1^2r^4}\right),\end{aligned} \tag{5.32}$$

$$\begin{aligned}I_9=&\int_R^\infty drG_2^{(2)}(r)\left(\frac{35c_2^2\mu_1^2}{r^8}+\frac{10c_2c_3\mu_1(3\kappa_1+\mu_1)}{r^6}\right.\\&\left.+\frac{3c_3^2(3\kappa_1^2+2\kappa_1\mu_1+\mu_1^2)}{r^4}\right),\end{aligned} \tag{5.33}$$

$$\begin{aligned}I_{10}=&\int_R^\infty\frac{dr}{r^3}\left(7c_2+\frac{3\kappa_1+\mu_1}{\mu_1}c_3r^2\right)\int_R^\infty\frac{ds}{s^3}\\&\times\left(7c_2+\frac{3\kappa_1+\mu_1}{\mu_1}c_3s^2\right)\int_{-1}^1duh(t)P_4(u),\end{aligned} \tag{5.34}$$

and

$$I_{11}=\int_R^\infty \frac{dr}{r^3}\left(7c_2+\frac{3\kappa_1+\mu_1}{\mu_1}c_3r^2\right)\int_R^\infty \frac{ds}{s^3}$$

$$\times\left(7c_2+\frac{3\kappa_1+\mu_1}{\mu_1}c_3s^2\right)\int_{-1}^1 du Q(\mathbf{r},\mathbf{s})P_4(u). \quad (5.35)$$

In summary, using Eqs. (2.9), (2.17), (3.10), and (5.18)–(5.23) for an isotropic composite system consisting of overlapping equi-sized spheres dispersed throughout a matrix, the effective shear modulus is bounded by

$$\mu_e\leq\langle\mu\rangle-\frac{4c_1^2\eta^2\phi_1^2(\mu_2-\mu_1)^2}{A_2\kappa_1+B_2(\kappa_2-\kappa_1)+2[C_2\mu_1+D_2(\mu_2-\mu_1)]} \quad (5.36)$$

and

$$\mu_e\geq\left(\langle\mu^{-1}\rangle-\frac{4c_4^2\eta^2\phi_1^2(\mu_2^{-1}-\mu_1^{-1})^2}{\frac{1}{9}[A_3\kappa_1^{-1}+B_3(\kappa_2^{-1}-\kappa_1^{-1})]+\frac{1}{2}[C_3\mu_1^{-1}+D_3(\mu_2^{-1}-\mu_1^{-1})]}\right)^{-1}. \quad (5.37)$$

## VI. CALCULATION OF BOUNDS FOR TOTALLY IMPENETRABLE SPHERES

Beasley and Torquato[14] showed that the three-point cluster bounds on the effective conductivity obtained by Torquato[12] and the three-point perturbation bounds obtained by Beran[21] were in fact equivalent for systems of totally impenetrable spheres. We now establish connections between the three-point cluster bounds and three-point perturbation bounds on the effective elastic moduli. We first compute the two sets of bounds in Eq. (4.26), (4.27), (5.36), and (5.37) for systems of totally impenetrable spheres. We will then show that the bounds on $\kappa_e$ are equivalent to the three-point Beran–Molyneux bounds[7] for this particular system. Also, we compare the cluster bounds on $\mu_e$ to the three-point bounds obtained by McCoy[8,10] and by Milton and Phan-Thien.[11]

### A. Calculation of the $I_n$ for totally impenetrable spheres

To calculate these ten integrals, we use the results the expressions for $G_n^{(i)}$ for totally impenetrable spheres obtained by Torquato.[12] In showing the equivalence of the Torquato and Beran bounds on effective conductivity for totally impenetrable spheres, Beasley and Torquato[14] calculated the first six integrals and found that

$$I_2=-2R^6/9, \quad (6.1)$$

$$I_3=\rho R^3/3, \quad (6.2)$$

$$I_4=\tfrac{2}{9}\rho^2R^6(-2+\phi_2), \quad (6.3)$$

$$I_5=-2/9, \quad (6.4)$$

$$I_6=\rho\phi_2\Omega/R^3, \quad (6.5)$$

and

$$I_7=\tfrac{2}{9}\rho^2\Lambda\phi_2, \quad (6.6)$$

where

$$\Omega=R^3\int_{2R}^\infty dr\,\frac{r^2}{(r^2-R^2)^3}\,g_2(r), \quad (6.7)$$

and

$$\Lambda=\frac{9}{32\pi^2}\sum_{l=2}^\infty l(l-1)R^{2l-4}\int\int d\mathbf{r}\,d\mathbf{s}[g_3(r,s,t)-g_2(r)g_2(s)]\frac{P_l(u)}{r^{l+1}s^{l+1}}. \quad (6.8)$$

Here $P_l$ is the $l$th Legendre polynomial and $g_3$ is the three-particle distribution function, which is related to the function $\rho_3$ defined in Eq. (3.3) by [cf. Eq. (3.11)]

$$g_3(r,s,t)=\rho_3(r,s,t)/\rho^3. \quad (6.9)$$

To calculate $I_8$ and $I_9$, we calculate the following integral:

$$\begin{aligned}I&=\int_R^\infty drG_2^{(2)}(r)\left(\frac{aR^{10}}{r^8}+\frac{bR^8}{r^6}+\frac{cR^6}{r^4}\right)\\&=2\pi\rho^2\int_R^\infty dr\left(\frac{aR^{10}}{r^8}+\frac{bR^8}{r^6}+\frac{cR^6}{r^4}\right)\int_0^R dss^2\int_{-1}^1 dug_2(t)\\&=2\pi\rho^2\int_0^\infty dttg_2(t)\int_0^R dss\int_{|t-s|}^{t+s}dre(r)\\&\quad\times\left(\frac{aR^{10}}{r^9}+\frac{bR^8}{r^7}+\frac{cR^6}{r^5}\right).\end{aligned} \quad (6.10)$$

This calculation was done by substituting Eq. (18) of Ref. 14, using the change of variable in Eq. (4.24), and changing the order of integration.

For impenetrable spheres, $g(r)=0$ for $r<2R$. Using this, we find that

$$I=\rho\eta R^3\int_{2R}^\infty dtt^2g_2(t)\left(\frac{aR^{13}(7t^4+14t^2R^2+3R^4)}{7(t^2-R^2)^7}+\frac{bR^{11}(5t^2+3R^2)}{5(t^2-R^2)^5}+\frac{cR^9}{(t^2-R^2)^3}\right). \quad (6.11)$$

[Not surprisingly, we obtain Eq. (6.5) after appropriate substitution of $a$, $b$, and $c$.] We conclude, using Eqs. (5.13), (5.14), (5.32) and (5.33), that

$$I_8=\frac{75}{14}\,\rho\eta R^3(3\kappa_1+\mu_1)^2\gamma^2\Upsilon+\frac{5}{28}\,\rho\eta R^3$$
$$\times(9\kappa_1^2+48\kappa_1\mu_1+92\mu_1^2)\gamma^2\Omega \tag{6.12}$$

and

$$I_9=\tfrac{375}{14}\,\rho\eta R^3(3\kappa_1+\mu_1)^2\mu_1^2\gamma^2\Upsilon+\tfrac{50}{7}\,\rho\eta R^3$$
$$\times(9\kappa_1^2+6\kappa_1\mu_1+8\mu_1^2)\mu_1^2\gamma^2\Omega, \tag{6.13}$$

where

$$\Upsilon=\frac{1}{2\pi}\int_{2R}^{\infty}dr r^2 g_2(r)W_2(r) \tag{6.14}$$

and $W_2$ is given in Eq. (35) of Ref. 22.

In a similar fashion, we find that

$$I_{10}=-\tfrac{8}{9}(3\kappa_1+\mu_1)^2\gamma^2R^6. \tag{6.15}$$

Finally, after a spherical harmonics expansion, $I_{11}$ is identical to an intermediate expression of Ref. 22 except for a trivial factor, and so

$$I_{11}=25\rho^2\eta R^6(3\kappa_1+\mu_1)^2\gamma^2\Psi, \tag{6.16}$$

where $\Psi$ is Eq. (40) of Ref. 22 after substitution of Eq. (57) of that reference.

## B. Equivalence of bounds on $\kappa_e$

The Beran–Molyneux bounds on $\kappa_e$ are

$$\kappa_e\leqslant\langle\kappa\rangle-\frac{3\phi_1\phi_2(\kappa_2-\kappa_1)^2}{3\langle\tilde{\kappa}\rangle+4\langle\mu\rangle_\zeta} \tag{6.17}$$

and

$$\kappa_e\geqslant\left[\langle\kappa^{-1}\rangle-\frac{4\phi_1\phi_2(\kappa_2^{-1}-\kappa_1^{-1})^2}{4\langle\tilde{\kappa}^{-1}\rangle+3\langle\mu^{-1}\rangle_\zeta}\right]^{-1}, \tag{6.18}$$

where

$$\langle b\rangle_\zeta=b_1\zeta_1+b_2\zeta_2 \tag{6.19}$$

for any property $b$. In this equation $\zeta_2=1-\zeta_1$ is a three-point microstructural parameter defined by

$$\zeta_2=\frac{9}{16\pi^2\phi_1\phi_2}\int\int d\mathbf{r}\,d\mathbf{s}[S_3(r,s,t)$$
$$-S_2(r)S_2(s)/\phi_2]\frac{P_2(u)}{r^3s^3}, \tag{6.20}$$

where again $P_2(u)$ is the second-order Legendre polynomial. The $n$-point probability functions $S_n$ give the probabilities of finding $n$ points in the particle phase. For totally impenetrable spheres, $S_3$ is expressible as the sum of a six-fold integral over $g_2$ and a nine-fold integral over $g_3$. Lado and Torquato[23] simplified $\zeta_2$ for this system and found that

$$\zeta_2=(3\Omega\phi_2+\Lambda\phi_2^2)/\phi_1, \tag{6.21}$$

where $\Omega$ and $\Lambda$ were defined in Eqs. (6.7) and (6.8).

From the results of the previous subsection, the constants in the bounds (4.26) and (4.27) are

$$A_1=9\phi_1\phi_2, \tag{6.22}$$

$$B_1=9\phi_1^2\phi_2, \tag{6.23}$$

$$C_1=6\phi_1\phi_2, \tag{6.24}$$

and

$$D_1=18\phi_2^2\Omega+6\phi_2^3\Lambda \tag{6.25}$$

for a system of totally impenetrable spheres. Substituting these expressions into Eqs. (4.26) and (4.27) shows that these bounds are indeed equivalent to the Beran–Molyneux bounds of Eqs. (6.17) and (6.18). The trial fields used in the Beran–Molyneux bounds are based on small contrast expansions.

## C. Effective shear modulus bounds

We now consider the bounds on the effective shear modulus for suspensions of totally impenetrable spheres. Previous three-point bounds on $\mu_e$ have been written in the form

$$\mu_e\leqslant\langle\mu\rangle-\frac{6\phi_1\phi_2(\mu_2-\mu_1)^2}{6\langle\tilde{\mu}\rangle+\Theta} \tag{6.26}$$

and

$$\mu_e^{-1}\geqslant\left(\langle\mu^{-1}\rangle-\frac{\phi_1\phi_2(\mu_2^{-1}-\mu_1^{-1})^2}{\langle\tilde{\mu}^{-1}\rangle+6\Xi}\right)^{-1}, \tag{6.27}$$

as shown in Refs. 10 and 11. In these bounds,

$$\langle b\rangle_\eta=b_1\eta_1+b_2\eta_2 \tag{6.28}$$

as above, where $\eta_2=1-\eta_1$ is another three-point microstructural parameter (not to be confused with the reduced density $\eta$) defined by

$$\eta_2=\frac{5\zeta_2}{21}+\frac{150}{56\pi^2\phi_1\phi_2}\int\int d\mathbf{r}\,d\mathbf{s}[S_3(r,s,t)$$
$$-S_2(r)S_2(s)/\phi_2]\frac{P_4(u)}{r^3s^3}, \tag{6.29}$$

where $P_4(u)$ is the fourth-order Legendre polynomial. Sen, Lado, and Torquato[22] simplified $\eta_2$ for systems of totally impenetrable spheres in a manner analogous to the simplification of $\zeta_2$ and found that

$$\eta_2=\phi_2^2(\Upsilon+\phi_2\Psi), \tag{6.30}$$

where $\Upsilon$ and $\Psi$ were defined in Eqs. (6.14) and (6.16), respectively.

From Eqs. (6.1)–(6.6), (6.11), (6.15), and (6.16), we see that the constants defined by Eqs. (5.24)–(5.31) are given by

$$A_2=240\mu_1^2\gamma^2\phi_1\phi_2, \tag{6.31}$$

$$B_2=240\mu_1^2\gamma^2\phi_1\phi_2\zeta_2, \tag{6.32}$$

$$C_2=60(3\kappa_1^2+8\kappa_1\mu_1+8\mu_1^2)\gamma^2\phi_1\phi_2, \tag{6.33}$$

$$D_2=72(\kappa_1+2\mu_1)^2\gamma^2\phi_1^2\phi_2+60(2\kappa_1+3\mu_1)\mu_1\gamma^2\phi_1$$
$$\times\phi_2\zeta_2+12(3\kappa_1+\mu_1)^2\gamma^2\phi_1\phi_2\eta_2, \tag{6.34}$$

$$A_3=2160\kappa_1^2\mu_1^2\gamma^2\phi_1\phi_2, \tag{6.35}$$

$$B_3=2160\kappa_1^2\mu_1^2\gamma^2\phi_1\phi_2\zeta_2, \tag{6.36}$$

$$C_3=40\mu_1^2(27\kappa_1^2+48\kappa_1\mu_1+32\mu_1^2)\gamma^2\phi_1\phi_2, \tag{6.37}$$

and

$$D_3=8\mu_1^2(9\kappa_1+8\mu_1)^2\gamma^2\phi_1^2\phi_2+240\mu_1^3(2\kappa_1+3\mu_1)\mu_1\gamma^2 \times\phi_1\phi_2\zeta_2+48\mu_1^2(3\kappa_1+\mu_1)^2\gamma^2\phi_1\phi_2\eta_2. \tag{6.38}$$

After substitution into Eqs. (5.36) and (5.37), we find that the present bounds on $\mu_e$ can also be represented in the form of Eqs. (6.26) and (6.27), where we replace $\Theta$ and $\Xi$ with

$$\Theta_1=\frac{10\mu_1^2\langle\kappa\rangle_\zeta+5\mu_1(2\kappa_1+3\mu_1)\langle\mu\rangle_\zeta+(3\kappa_1+\mu_1)^2\langle\mu\rangle_\eta}{(\kappa_1+2\mu_1)^2} \tag{6.39}$$

and

$$\Xi_1=\frac{10\kappa_1^2\langle\kappa^{-1}\rangle_\zeta+5\mu_1(2\kappa_1+3\mu_1)\langle\mu^{-1}\rangle_\zeta+(3\kappa_1+\mu_1)^2\langle\mu^{-1}\rangle_\eta}{(9\kappa_1+8\mu_1)^2}. \tag{6.40}$$

For systems which satisfy $\kappa_1>\kappa_2$ and $\mu_1>\mu_2$,

$$\Theta_1\leqslant\Theta_{HS}\equiv\frac{\mu_1(9\kappa_1+8\mu_1)}{\kappa_1+2\mu_1}, \tag{6.41}$$

and so the upper bound on $\mu_e$ is more restrictive than the Hashin–Shtrikman upper bound, in which $\Theta$ in Eq. (6.26) is replaced by $\Theta_{HS}$.[4] Likewise, for systems which satisfy $\kappa_1<\kappa_2$ and $\mu_1<\mu_2$,

$$\Xi_1\leqslant\Xi_{HS}\equiv\frac{\kappa_1+2\mu_1}{\mu_1(9\kappa_1+8\mu_1)}, \tag{6.42}$$

and so the lower bound on $\mu_e$ is more restrictive than the Hashin–Shtrikman lower bound.

At a given volume fraction $\phi_2$, the cluster upper bound will be more restrictive than the McCoy upper bound for some choices of the elastic moduli and less restrictive for others; which is determined by the values of $\zeta_2$ and $\eta_2$ at volume fraction $\phi_2$ and the larger of $\kappa_2/\kappa_1$ and $\mu_2/\mu_1$. This property is also true of the lower bound. However, both bounds will not be as restrictive as the Milton–Phan-Thien bounds, which are based on small-contrast trial fields.

We now calculate the cluster bounds for two different infinite-contrast cases, which are the most difficult to treat theoretically. We first consider a system of spherical voids (i.e., $\kappa_2=\mu_2=0$). For this special case, the cluster bound is identical to the McCoy bound. The four upper bounds are shown in Figure 1, using the values of $\zeta_2$ obtained by simulation in Ref. 15 and the first-order approximation to $\eta_2$ in Ref. 16. As we see, the cluster bound is a substantial improvement upon the Hashin–Shtrikman bound and almost identical to the Milton–Phan-Thien bound, although the numerical difference between the two bounds is slight.

The second infinite-contrast case which we now describe is an incompressible composite with impenetrable rigid spherical inclusions (i.e., $\kappa_1=\kappa_2=\infty$ and $\mu_2\gg\mu_1$). For such a system, the only possible nontrivial bound is the lower bound on the effective shear modulus; the determination of $\mu_e$ for such systems is called the *Einstein problem*. The four lower bounds are shown in Figure 2. Again, the cluster bound for this case is identical to the McCoy bound, and hence a substantial improvement on the Hashin–Shtrikman bound. The Milton–Phan-Thien bound is once more only slightly more restrictive than the cluster bound.

The precise reasons why the Milton–Phan-Thien bounds on the shear modulus are slightly better than the corresponding cluster bounds in these two infinite-contrast cases are difficult to ascertain. By contrast, recall that the cluster bounds for the bulk modulus are identical to the Beran-Molyneux bounds, which (as in the case of Milton–Phan-Thien bounds) are based on small-contrast trial fields.

## VII. CALCULATION OF BOUNDS FOR FULLY PENETRABLE SPHERES

In the case of fully penetrable spheres, the cluster bounds (4.26), (4.27), (5.36), and (5.37) can be evaluated analytically. This is in contrast to the other three-point bounds, which require numerical integration to determine the parameters $\zeta_2$ and $\eta_2$ at a given volume fraction. In the special case of a system of spherical voids, we find simple analytical bounds which are nearly identical to the other three-point bounds and also give an improvement for sufficiently large $\phi_2$. We also compare the cluster the sets of

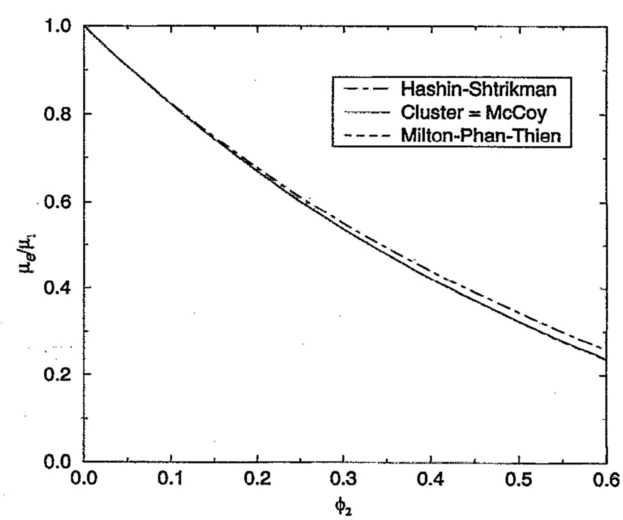


FIG. 1. The four upper bounds on $\mu_e$ for totally impenetrable spherical voids. The cluster bound for this system is identical to the McCoy bound, a considerable improvement on the Hashin–Shtrikman bound, and nearly identical to the Milton–Phan-Thien bound.

bounds in the Einstein limit. Finally, we examine the relation of the cluster bounds on the elastic moduli to the cluster bounds on effective conductivity.

For fully penetrable spheres we have[12]

$$\eta = -\ln \phi_1, \tag{7.1}$$

$$G_2^{(2)}(x) = \begin{cases} \rho, & x<R, \\ \rho\phi_2, & x>R, \end{cases} \tag{7.2}$$

$$h(x)=0, \tag{7.3}$$

and

$$Q(\mathbf{x}_1,\mathbf{x}_2) = \begin{cases} -\rho^2\phi_1, & x_1,x_2<R, \\ 0, & \text{otherwise.} \end{cases} \tag{7.4}$$

Using these relations in Eqs. (4.26), (4.27), (5.36), and (5.37) yields

$$\kappa_e \le \langle\kappa\rangle - \frac{3\eta\phi_1^2(\kappa_1-\kappa_2)^2}{3[\kappa_2-\eta\phi_1(\kappa_2-\kappa_1)]+4\langle\mu\rangle}, \tag{7.5}$$

and

$$\kappa_e \ge \left(\langle\kappa^{-1}\rangle - \frac{4\eta\phi_1^2(\kappa_1^{-1}-\kappa_2^{-1})^2}{4[\kappa_2^{-1}-\eta\phi_1(\kappa_2^{-1}-\kappa_1^{-1})]+3\langle\mu^{-1}\rangle}\right)^{-1}, \tag{7.6}$$

$$\mu_e \le \langle\mu\rangle - \frac{6\eta\phi_1^2(\mu_1-\mu_2)^2}{6[\mu_2-\eta\phi_1(\mu_2-\mu_1)]+\Theta_2}, \tag{7.7}$$

and

$$\mu_e \ge \left(\langle\mu^{-1}\rangle - \frac{\eta\phi_1^2(\mu_1^{-1}-\mu_2^{-1})^2}{\mu_2^{-1}-\eta\phi_1(\mu_2^{-1}-\mu_1^{-1})+6\Xi_2}\right)^{-1}, \tag{7.8}$$

where

$$\Theta_2 = \frac{10\langle\kappa\rangle\mu_1^2+\langle\mu\rangle(9\kappa_1^2+16\kappa_1\mu_1+16\mu_1^2)}{(\kappa_1+2\mu_1)^2}. \tag{7.9}$$

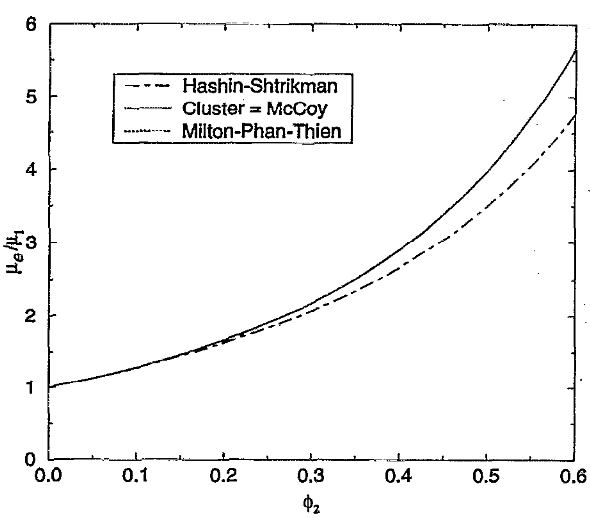


FIG. 2. The four lower bounds on $\mu_e$ for the Einstein problem with totally impenetrable inclusions. The cluster bound is identical to the McCoy bound, a considerable improvement on the Hashin–Shtrikman bound, and nearly identical to the Milton–Phan-Thien bound.

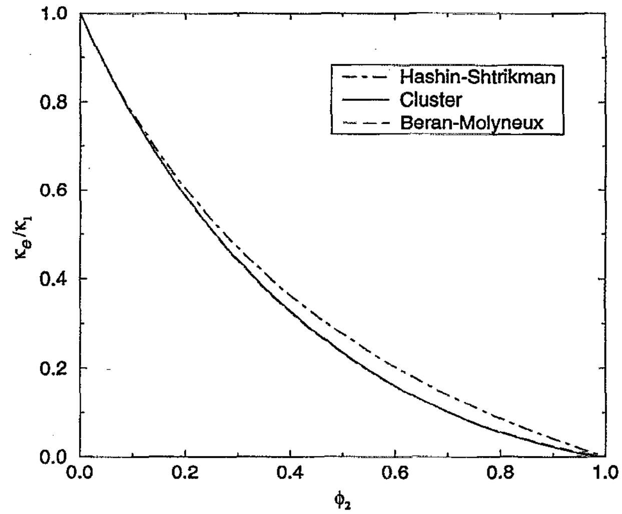


FIG. 3. The upper bound on $\kappa_e$ for a system of fully penetrable spherical voids with $\nu_1=0.3$. The cluster and Beran–Molyneux bounds are nearly identical, crossing around $\phi_2=0.60$.

and

$$\Xi_2 = \frac{10\langle\kappa^{-1}\rangle\kappa_1^2+\langle\mu^{-1}\rangle(9\kappa_1^2+16\kappa_1\mu_1+16\mu_1^2)}{(9\kappa_1+8\mu_1)^2}. \tag{7.10}$$

As expected, for small $\phi_2$ these bounds collapse to the well-known dilute limits.[9]

### A. Spherical voids

If the spherical inclusions are actually voids, then the lower bounds trivially become zero, while the upper bounds reduce to

$$\frac{\kappa_e}{\kappa_1} \le \frac{4f\phi_1}{3\eta+4f} \tag{7.11}$$

and

$$\frac{\mu_e}{\mu_1} \le \frac{(9+8f)\phi_1}{6\eta(1+2f)+(9+8f)}, \tag{7.12}$$

where $f=\mu_1/\kappa_1=(3-6\nu_1)/(2+2\nu_1)$ and $\nu_i$ is Poisson's ratio for phase $i$.

Interestingly, these bounds are nearly identical to the other three-point bounds, as indicated in Fig. 3. This observation holds for all possible values of $\nu_1$, including a matrix with a negative Poisson's ratio.[24] (The values of $\zeta_2$ and $\eta_2$ obtained from numerical integration are found in Refs. 17 and 18, respectively.) For small $\phi_2$ the previous three-point bounds are marginally better than the above cluster bounds, while the cluster bounds are marginally better than the Beran–Molyneux, McCoy, and Milton–Phan-Thien bounds for sufficiently large $\phi_2$.

### B. Einstein problem

We now reconsider the Einstein problem, allowing the particles to be fully penetrable. For this system, the cluster lower bound on $\mu_e$ reduces to

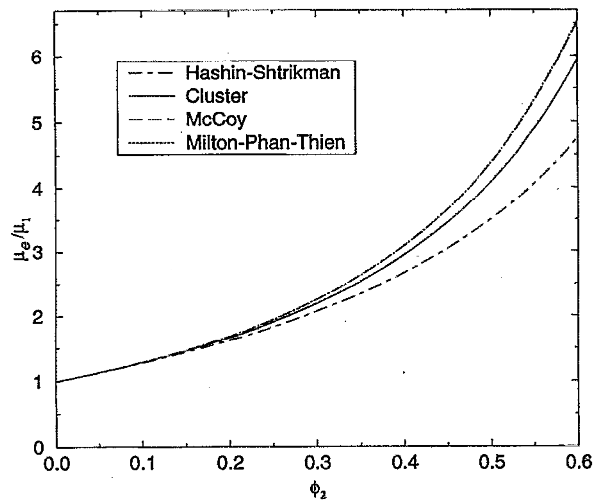


FIG. 4. The four lower bounds on $\mu_e$ for the Einstein problem with fully penetrable inclusions. The cluster bound in this case is not as sharp as the McCoy and Milton–Phan-Thien bounds.

$$\frac{\mu_e}{\mu_1} \geqslant \frac{3\eta+2}{2\phi_1}. \tag{7.13}$$

This bound is not as sharp as the McCoy and Milton–Phan-Thien bounds, as shown in Figure 4. However, the present bound was obtained purely by analytical means, while evaluation of the other bounds requires numerical integrations.[16]

### C. Cross-property relations

We now consider a system of fully penetrable spheres in which the matrix and spheres also have electrical conductivities $\sigma_1$ and $\sigma_2$, respectively. Torquato[12] derived the following bounds for the effective conductivity $\sigma_e$ of this system:

$$\sigma_e \leqslant \langle\sigma\rangle - \frac{\eta\phi_1^2(\sigma_2-\sigma_1)^2}{3\sigma_1+(\sigma_2-\sigma_1)(1+2\phi_2-\eta\phi_1)} \tag{7.14}$$

and

$$\sigma_e \geqslant \left(\langle\sigma^{-1}\rangle - \frac{2\eta\phi_1^2(\sigma_2^{-1}-\sigma_1^{-1})^2}{3\sigma_1^{-1}+(\sigma_2^{-1}-\sigma_1^{-1})(2+\phi_2-2\eta\phi_1)}\right)^{-1}. \tag{7.15}$$

It has been shown[25,26] that if $\kappa_2/\kappa_1 \leqslant \sigma_2/\sigma_1$ and the phase Poisson's ratios are non-negative, then

$$\kappa_e/\kappa_1 \leqslant \sigma_e/\sigma_1. \tag{7.16}$$

Likewise, if $\mu_2/\mu_1 \leqslant \sigma_2/\sigma_1$ and the phase Poisson's ratios are non-negative, then

$$\mu_e/\mu_1 \leqslant \sigma_e/\sigma_1. \tag{7.17}$$

It is straightforward to show that Eq. (7.16) remains true if $\kappa_e$ and $\sigma_e$ are replaced by the bounds in Eqs. (7.5), (7.14) and Eqs. (7.6), (7.15) if $\kappa_2/\kappa_1 = \sigma_2/\sigma_1$ and the phase Poisson's ratios are non-negative, and becomes an equality if the phase Poisson's ratios are both equal to zero.

Similarly, Eqs. (7.7), (7.14) and Eqs. (7.8), (7.15) satisfy Eq. (7.17) if $\mu_2/\mu_1 = \sigma_2/\sigma_1$ and the phase Poisson's ratios are non-negative.

## VIII. CONCLUSIONS

In this article rigorous bounds on the effective elastic moduli of suspensions of spheres of variable penetrability have been derived and shown to depend on three-point microstructural information. Our method is analogous to previously derived cluster bounds on the effective conductivity of suspensions of spheres. The general bounds due to Beran and Molyneux, McCoy, and Milton and Phan-Thien have been compared to the cluster bounds. For the special case of totally impenetrable spheres, the cluster bounds on the effective bulk modulus and the Beran–Molyneux bounds are shown to be identical, while the cluster bounds on the effective shear modulus are somewhat different than the McCoy and Milton–Phan-Thien bounds. The cluster bounds, however, are easier to compute when the spheres are allowed to overlap, providing an improvement for the special case of fully penetrable spherical voids with sufficiently high volume fraction of voids.

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## APPENDIX A: SOLUTION OF THE SINGLE SPHERE BOUNDARY-VALUE PROBLEM

Consider a matrix containing a single spherical inclusion with radius $R$ centered at the origin, with an applied strain field $\langle\epsilon\rangle$ at infinity. We divide the microscopic strain into the average strain and the perturbation caused by the spherical inclusion; i.e.,

$$\epsilon = \langle\epsilon\rangle + \epsilon^*. \tag{A1}$$

We now take $\langle\epsilon\rangle = \mathbf{I}$, the unit dyadic. The solution of the elasticity equations for this case is well documented.[27] Since the strain is at infinity is spherically symmetric, the displacement is only dependent on radius and so the equilibrium conditions of Eq. (2.10) reduce to

$$\frac{\partial^2 u}{\partial r^2} + \frac{2}{r}\frac{\partial u}{\partial r} - \frac{2}{r^2}u = 0, \tag{A2}$$

which, after satisfying the boundary condition and continuity along the interface, has solution

$$u(r) = k_1 r + \frac{k_2}{r^2}, \tag{A3}$$

where

$$k_1 = \begin{cases} \dfrac{3\kappa_1+4\mu_1}{3\kappa_2+4\mu_1}, & r<R, \\ 1, \quad r>R, \end{cases} \tag{A4}$$

$$k_2=\begin{cases} 0, & r<R, \\ 3R^3\dfrac{\kappa_1-\kappa_2}{3\kappa_2+4\mu_1}, & r>R. \end{cases} \tag{A5}$$

Differentiation of this displacement yields the strain perturbation field given in Eq. (4.3).

To obtain the stress perturbation field, we notice, since $\phi_2=0$ for a single inclusion,

$$\begin{aligned}\tau^*_{ij}&=\tau_{ij}-\langle\tau_{ij}\rangle=(\kappa-\tfrac{2}{3}\mu)\delta_{ij}\epsilon_{kk}+2\mu\epsilon_{ij}\\ &\quad-\langle(\kappa-\tfrac{2}{3}\mu)\delta_{ij}\epsilon_{kk}+2\mu\epsilon_{ij}\rangle\\ &=(\kappa-\tfrac{2}{3}\mu)\delta_{ij}[\langle\epsilon_{kk}\rangle+\epsilon^*_{kk}]+2\mu[\langle\epsilon_{ij}\rangle+\epsilon^*_{ij}]\\ &\quad-(\kappa_1-\tfrac{2}{3}\mu_1)\delta_{ij}\langle\epsilon_{kk}\rangle-2\mu_1\langle\epsilon_{ij}\rangle.\end{aligned} \tag{A6}$$

Setting $\langle\epsilon_{ij}\rangle=\delta_{ij}$ yields Eq. (4.4).

If we instead take $\langle\epsilon_{ij}\rangle=\delta_{i1}\delta_{j1}-\delta_{i2}\delta_{j2}$, the solution of the boundary-value problem is considerably more involved.[9] In this case, there is no spherical symmetry to reduce the complexity of the equilibrium equations. If $\mathbf{r}=(x,y,z)$ and $r=|\mathbf{r}|$, then, under the assumption of isotropy, Christensen's solution for the displacement vector has components

$$u_1=p(r)x(x^2-y^2)+[q(r)+1]x, \tag{A7}$$

$$u_2=p(r)y(x^2-y^2)-[q(r)+1]y, \tag{A8}$$

$$u_3=p(r)z(x^2-y^2), \tag{A9}$$

where $p(r)$ and $q(r)$ are given by Eqs. (5.10) and (5.11). (This is similar to the form of the velocity of the flow due to a sphere embedded in a pure straining motion in fluid permeability theory.[28]) Differentiation of these displacements yields Eqs. (5.3)–(5.9). The stress perturbation field for this case; i.e., Eq. (5.16), is derived using Eq. (A6).

## APPENDIX B: SIMPLIFICATION OF ENSEMBLE AVERAGES

We now present how the ensemble averages Eqs. (4.6)–(4.11) and (5.18)–(5.23) are simplified. To begin we take the strain perturbation field $\epsilon'$ used in Section IV. To calculate $B_1$, combination of Eq. (3.1), (3.6), and (4.1) gives

$$\begin{aligned}B_1&=\int d\mathbf{x}_1 G_2^{(2)}(\mathbf{x}_1)\epsilon^*_{ii}(\mathbf{x}_1)\epsilon^*_{kk}(\mathbf{x}_1)\\ &\quad+\int\int d\mathbf{x}_1\, d\mathbf{x}_2 Q(\mathbf{x}_1,\mathbf{x}_2)\epsilon^*_{ii}(\mathbf{x}_1)\epsilon^*_{kk}(\mathbf{x}_2).\end{aligned} \tag{B1}$$

Let $\mathbf{r}=\mathbf{x}_1$ and $\mathbf{s}=\mathbf{x}_2$ have spherical coordinates $(r,\theta_r,\phi_r)$ and $(s,\theta_s,\phi_s)$, respectively. Then the first integral of Eq. (B1) is easily obtained using Eq. (4.3). We calculate the second integral following Lado and Torquato[23] by first expanding $Q$ in Legendre polynomials; i.e.,

$$Q(\mathbf{r},\mathbf{s})=\sum_{n=0}^{\infty} D_n(r,s)P_n(u_{rs}), \tag{B2}$$

where

$$D_n(r,s)=\frac{2n+1}{2}\int_{-1}^{1} du_{rs}Q(\mathbf{r},\mathbf{s})P_n(u_{rs}) \tag{B3}$$

and

$$u_{rs}=\frac{\mathbf{r}\cdot\mathbf{s}}{r\cdot s}. \tag{B4}$$

The second integral can now be calculated by using the addition theorem[29]

$$\begin{aligned}P_n(u_{rs})&=P_n(u_r)P_n(u_s)+2\sum_{m=1}^{n}\frac{(n-m)!}{(n+m)!}P_n^m\\ &\quad\times(u_r)P_n^m(u_s)\cos m(\phi_r-\phi_s),\end{aligned} \tag{B5}$$

where $u_r=\cos\theta_r$ and $u_s=\cos\theta_s$, and the orthogonality properties of the Legendre polynomials.

The ensemble average $A_1$ may be obtained from Eq. (B1) by replacing $G_2^{(2)}$ and $Q$ by $\rho$ and $\rho^2 h$, respectively.

To calculate $D_1$, we proceed as above and find that

$$\begin{aligned}D_1&=\int d\mathbf{x}_1 G_2^{(2)}(\mathbf{x}_1)\epsilon^*_{ij}(\mathbf{x}_1)\epsilon^*_{ij}(\mathbf{x}_1)+\int\int d\mathbf{x}_1\, d\mathbf{x}_2 Q(\mathbf{x}_1,\mathbf{x}_2)\\ &\quad\times\epsilon^*_{ij}(\mathbf{x}_1)\epsilon^*_{ij}(\mathbf{x}_2)-\tfrac{1}{3}\langle I^{(2)}\epsilon'_{kk}\epsilon'_{ll}\rangle.\end{aligned} \tag{B6}$$

$D_1$ can now be calculated using Eq. (4.3). $C_1$ can be obtained from $D_1$ by again replacing $G_2^{(2)}$ and $Q$ by $\rho$ and $\rho^2 h$, respectively, and replacing the final term with $-\frac{1}{3}\langle\epsilon'_{kk}\epsilon'_{ll}\rangle$.

Finally, the ensemble averages of Section V may be derived as above by using Eqs. (5.3)–(5.9) and Eq. (5.16) instead of Eq. (4.3). These computations, although entirely elementary, are somewhat lengthy due to the complicated form of the solution of the single sphere boundary value problem.


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