

## Exact Expression for the Effective Elastic Tensor of Disordered Composites

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We derive new, exact series expansions for the effective elastic tensor of anisotropic,  $d$ -dimensional, two-phase disordered composites whose  $n$ th-order tensor coefficients are integrals involving  $n$ -point correlation functions that characterize the structure. These series expansions, valid for any structure, perturb about certain optimal dispersions. Third-order truncation of the expansions results in formulas for the elastic moduli of isotropic dispersions that are in very good agreement with benchmark data, always lie within rigorous bounds, and are superior to popular self-consistent approximations. [S0031-9007(97)03646-6]

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Perturbation expansions have been developed for the effective elastic tensor  $\mathbf{C}_e$  of composites for small variations in the phase elastic tensors [1]. Because of the nature of the integral operator, one must contend with conditionally convergent integrals. One resolution to this problem is to carry out a “renormalization” analysis [2]. Drawbacks of such classical expansions are that they are formal and valid only when phase moduli are nearly the same.

In this Letter we develop new, exact series expansions for the effective elastic tensor of macroscopically anisotropic,  $d$ -dimensional, two-phase composite media of arbitrary microstructure by introducing an integral equation for the so-called “cavity” strain field. The expansions are not formal, but rather the  $n$ th-order tensor coefficients are given explicitly in terms of integrals over products of certain tensor fields and a determinant involving  $n$ -point statistical correlation functions that render the integrals absolutely convergent in the infinite-volume limit. Thus, no renormalization analysis is required. Interestingly, the series expressions may be regarded as expansions that perturb about optimal dispersions described below. By truncating the exact expansion after third-order terms, we find approximate relations for the effective moduli of isotropic dispersions that are in very good agreement with benchmark data, always lie within rigorous bounds, and are superior to self-consistent approximations.

Consider a large but finite-sized, ellipsoidal, macroscopically anisotropic composite specimen in arbitrary space dimension  $d$  comprised of two isotropic phases with fourth-order elastic tensors  $\mathbf{C}_1$  and  $\mathbf{C}_2$ . The elastic tensor  $\mathbf{C}_i$  for phase  $i$  can be expressed in terms of the phase bulk and shear moduli  $K_i$  and  $G_i$ , respectively, via  $\mathbf{C}_i = dK_i\mathbf{\Lambda}_h + 2G_i\mathbf{\Lambda}_s$ , where  $\mathbf{\Lambda}_h$  and  $\mathbf{\Lambda}_s$  are the projection tensors [3]. Now let us embed this specimen in an infinite *reference* phase, taken to be phase 1, and subject it to an applied strain field  $\boldsymbol{\epsilon}^0(\mathbf{x})$  at infinity. The effective elastic tensor  $\mathbf{C}_e$  is defined by the averaged Hooke’s law:

$$\langle \boldsymbol{\sigma}(\mathbf{x}) \rangle = \mathbf{C}_e : \langle \boldsymbol{\epsilon}(\mathbf{x}) \rangle, \quad (1)$$

where  $\boldsymbol{\sigma}(\mathbf{x})$  and  $\boldsymbol{\epsilon}(\mathbf{x}) = \frac{1}{2}[\nabla\mathbf{u} + (\nabla\mathbf{u})^T]$  are the local stress and strain tensors, respectively,  $\mathbf{u}(\mathbf{x})$  is the local displacement, angular brackets denote an ensemble aver-

age, and  $:$  denotes a double contraction. The local stress must satisfy the equilibrium requirement

$$\nabla \cdot \boldsymbol{\sigma}(\mathbf{x}) = 0, \quad (2)$$

such that Hooke’s law is obeyed locally, i.e.,

$$\begin{aligned} \boldsymbol{\sigma}(\mathbf{x}) &= \mathbf{C}(\mathbf{x}) : \boldsymbol{\epsilon}(\mathbf{x}) \\ &= \mathbf{C}_1 \boldsymbol{\epsilon}(\mathbf{x}) + \mathbf{p}(\mathbf{x}), \end{aligned} \quad (3)$$

where  $\mathbf{p}(\mathbf{x}) = \chi(\mathbf{x})[\mathbf{C}_2 - \mathbf{C}_1] : \boldsymbol{\epsilon}(\mathbf{x})$  is the induced *polarization* stress tensor and  $\chi(\mathbf{x})$  is the characteristic function of the “polarized” phase 2 (i.e., equal to 1 in phase 2 and zero otherwise).

The solution of (2) and (3) can be expressed by the integral relation [4]

$$\mathbf{u}(\mathbf{x}) = \mathbf{u}^0(\mathbf{x}) + \int d\mathbf{x}' \nabla \mathbf{g}(\mathbf{x}, \mathbf{x}') : \mathbf{p}(\mathbf{x}'), \quad (4)$$

where, in component form,

$$g_{ij}(\mathbf{r}) = \begin{cases} \frac{1}{2\Omega G_1} \ln\left(\frac{1}{r}\right) \delta_{ij} + b_1 n_i n_j, & d = 2, \\ a_1 \frac{\delta_{ij}}{r^{d-2}} + b_1 \frac{n_i n_j}{r^{d-2}}, & d \geq 3, \end{cases} \quad (5)$$

is the infinite-space,  $d$ -dimensional Green’s function,  $\Omega$  is the total solid angle contained in a  $d$ -dimensional sphere,  $\mathbf{r} = \mathbf{x} - \mathbf{x}'$ ,  $\mathbf{n} = \mathbf{r}/|\mathbf{r}|$ , and  $a_1$  and  $b_1$  are parameters that depend on phase 1 moduli [5].

Now to obtain the strain, one must differentiate (4); however, because of the singular nature of the integral one cannot simply differentiate under the integral sign. Excluding a spherical region or cavity from the origin in (4) and integrating by parts yields

$$\boldsymbol{\epsilon}(\mathbf{x}) = \boldsymbol{\epsilon}^0(\mathbf{x}) + \int d\mathbf{x}' \mathbf{G}(\mathbf{r}) : \mathbf{p}(\mathbf{x}'), \quad (6)$$

where

$$\mathbf{G}(\mathbf{r}) = -\mathbf{A}\delta(\mathbf{r}) + \mathbf{H}(\mathbf{r}). \quad (7)$$

Here  $\mathbf{A}$  is the constant fourth-order tensor

$$\begin{aligned} \mathbf{A} &= \frac{\mathbf{\Lambda}_h}{dK_1 + 2(d-1)G_1} \\ &+ \frac{d(K_1 + 2G_1)\mathbf{\Lambda}_s}{G_1(d+2)[dK_1 + 2(d-1)G_1]}, \end{aligned} \quad (8)$$

that arises because of the exclusion of the spherical cavity in (4),  $\delta(\mathbf{r})$  is the Dirac delta function, and  $\mathbf{H}(\mathbf{r})$  is the symmetrized double gradient  $\nabla\nabla\mathbf{g}(\mathbf{r})$  [4].

We depart substantially from previous treatments by introducing an integral equation [6] for the “cavity” strain field  $\mathbf{f}$  by substituting (7) into expression (6), i.e.,

$$\mathbf{f}(\mathbf{x}) = \boldsymbol{\varepsilon}^0(\mathbf{x}) + \int d\mathbf{x}' \mathbf{H}(\mathbf{x} - \mathbf{x}') : \mathbf{p}(\mathbf{x}), \quad (9)$$

which is related to the usual strain by the expression

$$\mathbf{f}(\mathbf{x}) = \{\mathbf{I} + \mathbf{A} : [\mathbf{C}(\mathbf{x}) - \mathbf{C}_1]\} : \boldsymbol{\varepsilon}(\mathbf{x}). \quad (10)$$

The polarization and cavity strain field are related via

$$\mathbf{p}(\mathbf{x}) = \mathcal{L}(\mathbf{x}) : \mathbf{f}(\mathbf{x}), \quad (11)$$

$$\mathcal{L}(\mathbf{x}) = \{\mathbf{C}(\mathbf{x}) - \mathbf{C}_1\} \{\mathbf{I} + \mathbf{A} : [\mathbf{C}(\mathbf{x}) - \mathbf{C}_1]\}^{-1}. \quad (12)$$

The effective tensor  $\mathbf{L}_e$  is given by the averaged relation

$$\langle \mathbf{p}(\mathbf{x}) \rangle = \mathbf{L}_e : \langle \mathbf{f}(\mathbf{x}) \rangle, \quad (13)$$

where  $\mathbf{L}_e$  is related to the effective elastic tensor  $\mathbf{C}_e$  via

$$\mathbf{L}_e = \{\mathbf{C}_e - \mathbf{C}_1\} \{\mathbf{I} + \mathbf{A} : [\mathbf{C}_e - \mathbf{C}_1]\}^{-1}. \quad (14)$$

It is desired to find an explicit expression for the effective moduli  $\mathbf{L}_e$  (or  $\mathbf{C}_e$ ) using the solution of (9) which we rewrite in schematic operator notation as

$$\mathbf{f} = \boldsymbol{\varepsilon}^0 + \mathbf{H}\mathbf{p}, \quad (15)$$

where for a general operator  $\mathbf{\Gamma}$ ,  $\mathbf{\Gamma}\mathbf{p} \equiv \int d2 \mathbf{\Gamma}(1,2) : \mathbf{p}(2)$ , and 1 and 2 represent  $\mathbf{x}$  and  $\mathbf{x}'$ , respectively. Multiplying (15) from the left by  $\mathcal{L}(\mathbf{x})$  yields an integral equation for the polarization  $\mathbf{p}$  having the solution

$$\mathbf{p} = \mathbf{T}\boldsymbol{\varepsilon}^0, \quad (16)$$

where  $\mathbf{T} = \mathcal{L}[\mathbf{I} - \mathcal{L}\mathbf{H}]^{-1}$ . Ensemble averaging (16) yields

$$\langle \mathbf{p} \rangle = \langle \mathbf{T} \rangle \boldsymbol{\varepsilon}^0. \quad (17)$$

It is seen that the operator  $\langle \mathbf{T} \rangle$  generally involves products of the tensor  $\mathbf{H}$  which decays to zero like  $r^{-d}$  for large  $r$ . Thus,  $\langle \mathbf{T} \rangle$  at best involves conditionally convergent integrals and hence must be dependent upon the shape of the ellipsoidal composite specimen.

Given the nonlocal nature of the relation (17), the remaining strategy is clear. One must eliminate the applied field  $\boldsymbol{\varepsilon}^0$  in favor of the appropriate average field in order to get a local, shape-independent relation. Thus, we invert (17) and average (15) to obtain

$$\langle \mathbf{f} \rangle = \mathbf{X} \langle \mathbf{p} \rangle, \quad (18)$$

where  $\mathbf{X} = \langle \mathbf{T} \rangle^{-1} + \mathbf{H}$ . Comparing (18) to (14) and letting  $\mathbf{U}(\mathbf{r}) = \mathbf{L} : \mathbf{H}(\mathbf{r})$  [where  $\mathbf{L}$  is given by (14) with  $\mathbf{C}_e$  replaced by  $\mathbf{C}_2$ ] yields the desired exact expression

$$\phi_2^2 \mathbf{L} : (\mathbf{L}_e)^{-1} = \phi_2 \mathbf{I} - \sum_{n=2}^{\infty} \mathbf{B}_n, \quad (19)$$

where  $\phi_2$  is the volume fraction of phase 2,  $\mathbf{I}$  is the fourth-order identity tensor, and

$$\mathbf{B}_2 = \int d2 \mathbf{U}(1,2) [S_2(1,2) - \phi_2^2], \quad (20)$$

$$\mathbf{B}_n = \left(\frac{-1}{\phi_2}\right)^{n-2} \int d2 \cdots \int dn \mathbf{U}(1,2) : \mathbf{U}(2,3) \cdots : \mathbf{U}(n-1,n) \Delta_n, \quad n \geq 3, \quad (21)$$

$\Delta_n$  is the position-dependent determinant

$$\Delta_n = \begin{vmatrix} S_2(1,2) & S_1(2) & \cdots & 0 \\ S_3(1,2,3) & S_2(2,3) & \cdots & 0 \\ \vdots & \vdots & \ddots & \vdots \\ S_n(1, \dots, n) & S_{n-1}(2, \dots, n) & \cdots & S_2(n-1, n) \end{vmatrix} \quad (22)$$

and  $S_n(\mathbf{x}_1, \dots, \mathbf{x}_n) = \langle \chi(\mathbf{x}_1) \cdots \chi(\mathbf{x}_n) \rangle$  is the  $n$ -point correlation function [7] for phase 2. Note that the  $n$ -point tensor coefficient  $\mathbf{B}_n$  involves the parameters

$$\kappa = \frac{K_2 - K_1}{K_2 + \frac{2(d-1)}{d} G_1}, \quad (23)$$

$$\mu = \frac{G_2 - G_1}{G_2 + \frac{G_1[dK_1/2 + (d+1)(d-2)G_1/d]}{K_1 + 2G_1}}.$$

In analogy with dielectric theory, we refer to  $\kappa$  and  $\mu$  as the *bulk and shear moduli polarizabilities*, respectively.

Since the quantity within the brackets of (20) and the determinant  $\Delta_n$  in (21) identically vanish at the specimen boundary [8], because of the asymptotic properties of the  $S_n$  [7], the integrals in (19) are independent of the shape of the macroscopic ellipsoid (i.e., absolutely convergent), and hence any convenient shape may be used in the infinite-volume limit. Interestingly, we can show [4] that the series expressions (19) may be regarded as expansions that perturb about the optimal structures [9–11] that realize the isotropic Hashin-Shtrikman (HS) bounds [9] and Willis' anisotropic bounds [12]. For simplicity, we discuss the isotropic instance below.

The isotropic case can be obtained from (19) after appropriate contractions [4], with the results

$$\phi_2^2 \frac{\kappa}{\kappa_e} = \phi_2 - \sum_{n=3}^{\infty} C_n, \quad (24)$$

$$\phi_2^2 \frac{\mu}{\mu_e} = \phi_2 - \sum_{n=3}^{\infty} D_n, \quad (25)$$

where the scalars  $C_n = \mathbf{\Lambda}_h \dot{\dot{\dot{\dot{\mathbf{B}}}}}_n$  and  $D_n = \frac{2}{(d+2)(d-1)} \times \mathbf{\Lambda}_s \dot{\dot{\dot{\dot{\mathbf{B}}}}}_n$ , the symbol  $\dot{\dot{\dot{\dot{\mathbf{B}}}}}$  denotes the quadruple dot product, and

$$\kappa_e = \frac{K_e - K_1}{K_e + \frac{2(d-1)}{d} G_1}, \quad (26)$$

$$\mu_e = \frac{G_e - G_1}{G_e + \frac{G_1[dK_1/2 + (d+1)(d-2)G_1/d]}{K_1 + 2G_1}},$$

are the effective polarizabilities. In contrast to classical perturbation expansions, the expansions (24) and (25) are *nonclassical* in the sense that the expansion parameters are the polarizabilities  $\kappa$  and  $\mu$  and, for certain structures, can converge rapidly for any phase contrast.

To understand the physical meaning of the expansion (24) and (25), it is helpful to consider the structures for which the  $n$ -point parameters  $C_n$  or  $D_n$  vanish. For  $d = 2$  and  $d = 3$ , expression (24) with  $C_n = 0$  for all  $n$  is recognized to coincide with the isotropic HS bounds on the effective bulk modulus  $K_e$  [8], and hence is exact for the assemblages of coated circles ( $d = 2$ ) and coated spheres ( $d = 3$ ) that realize the bounds [9] as well as certain finite-rank laminates [10]. Similarly, for  $d = 2$  and  $d = 3$ , expression (25) with  $D_n = 0$  for all  $n$  equals the isotropic HS bounds on the effective shear modulus  $G_e$  [13] and hence is exact for certain finite-rank laminates in both two and three dimensions [10]. Note that for either the coated-inclusion assemblages or finite-rank laminates, one of the phases is always a disconnected, dispersed phase in a connected matrix phase (except trivially when  $\phi_2 = 1$ ).

Therefore, expansions (24) and (25) should converge rapidly for any values of the phase moduli for dispersions

in which the inclusions, taken to be the polarized phase, are prevented from forming large clusters. Consequently, we contend that the first few terms of this expansion will provide an excellent approximation of the effective bulk modulus  $K_e$  and shear modulus  $G_e$  of such dispersions.

Accordingly, simplification of (24) and (25) through third-order terms gives

$$\phi_2 \frac{\kappa}{\kappa_e} = 1 - \frac{(d+2)(d-1)G_1\kappa\mu}{d(K_1+2G_1)} \zeta_2 \phi_1, \quad (27)$$

$$\begin{aligned} \phi_2 \frac{\mu}{\mu_e} = & 1 - \frac{2G_1\kappa\mu}{d(K_1+2G_1)} \zeta_2 \phi_1 \\ & + \frac{(d^2-4)G_1(2K_1+3G_1)\mu^2}{2d(K_1+2G_1)^2} \zeta_2 \phi_1 \\ & + \frac{1}{2d} \left[ \frac{dK_1+(d-2)G_1}{K_1+2G_1} \right]^2 \mu^2 \eta_2 \phi_1, \end{aligned} \quad (28)$$

where  $\zeta_2$  and  $\eta_2$  are three-point microstructural parameters

$$\zeta_2 = \frac{d^2}{(d-1)\phi_1\phi_2\Omega^2} \int \int \frac{d\mathbf{r}}{r^d} \frac{ds}{s^d} [d(\mathbf{n} \cdot \mathbf{m})^2 - 1] \left[ S_3(\mathbf{r}, \mathbf{s}) - \frac{S_2(r)S_2(s)}{\phi_2} \right], \quad (29)$$

$$\eta_2 = -\frac{(d+2)(5d+6)}{d^2} \zeta_2 + \frac{(d+2)^2}{(d-1)\phi_1\phi_2\Omega^2} \int \int \frac{d\mathbf{r}}{r^d} \frac{ds}{s^d} [d(d+2)(\mathbf{n} \cdot \mathbf{m})^4 - 3] \left[ S_3(\mathbf{r}, \mathbf{s}) - \frac{S_2(r)S_2(s)}{\phi_2} \right], \quad (30)$$

and  $\mathbf{n} = \mathbf{r}/|\mathbf{r}|$  and  $\mathbf{m} = \mathbf{s}/|\mathbf{s}|$  are unit vectors. The three-point parameters  $\zeta_2$  and  $\eta_2$  (which always lie in the interval  $[0, 1)$ ) also arise in rigorous three-point bounds on the effective elastic moduli [7,14,15,16].

We have compared the third-order approximations (27) and (28) for  $K_e$  and  $G_e$  to benchmark data, rigorous bounds, and popular self-consistent (SC) approximations [17] for a variety of 2D and 3D isotropic dispersions. In Fig. 1 we compare the prediction of (28) with  $d = 2$  for the effective transverse shear modulus  $G_e$  of oriented, hexagonal arrays of glass fibers (phase 2) in an epoxy matrix to the corresponding simulation data of Eischen and Torquato [18]. Formula (28) provides an excellent estimate of  $G_e$  for all volume fractions. The three-point Silnutzer [14,15] lower bound (not shown) is very slightly below (28) and the three-point Gibiansky-Torquato [16] upper bound is somewhat above (28). Note that the SC formula begins to violate the upper bound at small  $\phi_2$ .

Consider now a fiber-reinforced material consisting of oriented, superrigid, circular fibers that are randomly arranged in a compressible matrix. We are not aware of comprehensive benchmark data for this case. However, we can compare our results to the rigorous cross-property bounds [20] that use effective conductivity measurements [21] to bound the effective elastic moduli (and vice versa). Figure 2 shows that formula (27) with  $d = 2$  for the effective transverse bulk modulus  $K_e$  is below the sharp cross-property upper bound and slightly above the three-point Silnutzer lower bound (not shown). Not

surprisingly, the SC formula violates the upper bound at moderate  $\phi_2$ .

Figure 3 compares relation (27) with  $d = 3$  and the SC formula [17] to the numerical data of Nunan and Keller [22] for superrigid face-centered cubic arrays of spheres in a compressible matrix. The prediction of (27), applicable to cubic symmetric composites, is remarkably accurate,

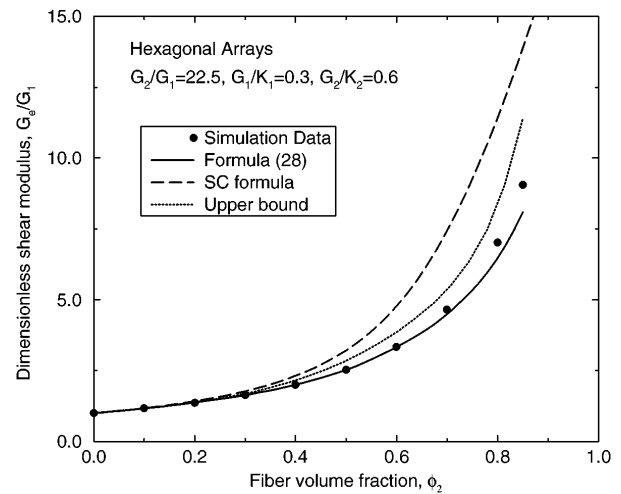


FIG. 1. Dimensionless effective transverse shear modulus  $G_e/G_1$  vs fiber volume fraction  $\phi_2$  for hexagonal arrays of circular glass fibers in an epoxy matrix. Simulation data are from Ref. [18], and parameters  $\zeta_2$  and  $\eta_2$ , for use in (28) and bounds, are from Refs. [19] and [18], respectively.

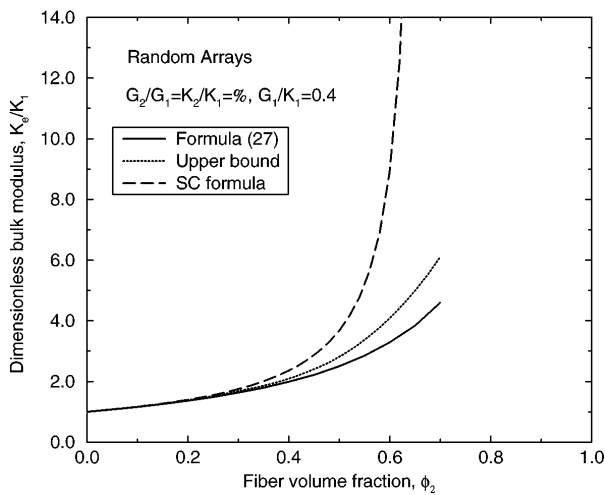


FIG. 2. Dimensionless effective transverse bulk modulus  $K_e/K_1$  vs fiber volume fraction  $\phi_2$  for random arrays of circular superrigid fibers in a compressible matrix. Cross-property upper bound uses conductivity data of Ref. [20] and  $\zeta_2$  values, for use in (27), are from Ref. [19].

whereas the SC formula begins to diverge increasingly from the data for values of the particle volume fraction  $\phi_2 > 0.3$ . The Beran-Molyneux three-point lower bound is virtually indistinguishable from the prediction (27).

To summarize, by careful treatment of the integral equation for the cavity strain field, we are able to find an explicit, exact expression for the effective elastic tensor of arbitrary,  $d$ -dimensional, two-phase composites. This exact representation is powerful, enabling one to prove a variety of rigorous results [4]. For example, we can show [4] that the effective shear modulus  $G_e$  is independent of the microstructure and is exactly equal to the arithmetic average ( $G_1\phi_1 + G_2\phi_2$ ) in the limit that the space dimension  $d \rightarrow \infty$ . Our choice of excluding a spherical cavity in the singular integrals resulted in

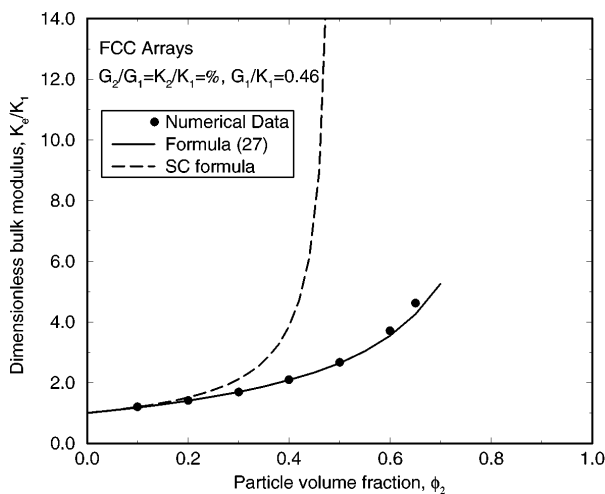


FIG. 3. Dimensionless effective bulk modulus  $K_e/K_1$  vs particle volume fraction  $\phi_2$  for face-centered cubic arrays of superrigid spheres in a compressible matrix. Numerical data are from Ref. [21] and  $\zeta_2$  values, for use in (27), are from Ref. [19].

an expansion that perturbs about optimal dispersions and suggested accurate approximations for the moduli of a class of isotropic dispersions. SC formulas did not provide good moduli estimates for dispersions and indeed violated bounds. Finally, note that the procedure outlined here can be used to find exact representations of other physical properties that are defined by local linear constitutive laws.

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