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EFFECTIVE STIFFNESS TENSOR OF COMPOSITE MEDIA: II. APPLICATIONS TO ISOTROPIC DISPERSIONS

S. TORQUATO

Department of Civil Engineering and Operations Research and Princeton Materials Institute, Princeton University, Princeton, N.J. 08544, U.S.A.

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ABSTRACT

Accurate approximate relations for the effective elastic moduli of two- and three-dimensional isotropic dispersions are obtained by truncating, after third-order terms, an exact series expansion for the effective stiffness tensor of d-dimensional two-phase composites (obtained in the first paper) that perturbs about certain optimal dispersions. Our third-order approximations of the effective bulk modulus K_e and shear modulus G_e are compared to benchmark data, rigorous bounds and popular self-consistent approximations for a variety of macroscopically isotropic dispersions in both two and three dimensions, for a wide range of phase moduli and volume fractions. Generally, for the cases considered, the third-order approximations are in very good agreement with benchmark data, always lie within rigorous bounds, and are superior to popular self-consistent approximations. © 1998 Elsevier Science Ltd. All rights reserved

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1. INTRODUCTION

In the first part of this series (henceforth referred to as part I), we developed new exact perturbation expansions for the effective stiffness tensor of macroscopically anisotropic composite media consisting of two isotropic phases by introducing an integral equation for the so-called "cavity" strain field. The expansions are not formal but rather the nth-order tensor coefficients are given explicitly in terms of integrals over products of certain tensor fields and a determinant involving the set of n-point probability functions $S_1^{(p)}, S_2^{(p)}, \ldots, S_n^{(p)}$ for phase p that systematically render the integrals absolutely convergent in the infinite-volume limit, i.e., without having to resort to a "renormalization" procedure. The quantity $S_n^{(p)}(\mathbf{x}_1, \ldots, \mathbf{x}_n)$ gives the probability of finding n points with positions $\mathbf{x}_1, \ldots, \mathbf{x}_n$ in phase p (where $p = 1$ or 2). For statistically homogeneous media, $S_n^{(p)}$ depends on the relative displacements $\mathbf{x}_2-\mathbf{x}_1, \ldots, \mathbf{x}_n-\mathbf{x}_1$ and, in particular, $S_1^{(p)}$ is just the volume fraction ϕ_p of phase p.

Another useful feature of the expansions (expressible in powers of the bulk and shear moduli "polarizabilities") is that they converge rapidly for a class of dispersions for all volume fractions, even when the phase moduli differ significantly. The reason for this rapid convergence is that for macroscopically isotropic media, the exact series expressions of part I may be regarded as expansions that perturb about the structures that realize the optimal Hashin–Shtrikman bounds (Hashin and Shtrikman, 1963;

Hashin, 1965). For macroscopically anisotropic media, the series expressions of I may be regarded as expansions that perturb about the structures that realize Willis' (1977) bounds. The structures that realize the Hashin–Shtrikman and Willis bounds have the important feature that one phase is disconnected or dispersed and the other is a continuous matrix phase.

In part I we claimed that the series expression truncated after third-order terms should provide accurate estimates of the effective moduli for a wide range of phase moduli and volume fractions for dispersions in which the inclusions are prevented from forming large clusters. One of the main aims of this sequel is to test this claim quantitatively for a variety of macroscopically isotropic dispersions in both two and three dimensions, and for a wide range of phase contrasts and volume fractions. This will be accomplished by comparing our third-order approximations of the effective bulk modulus K_e and shear modulus G_e to benchmark data, rigorous bounds and popular self-consistent approximations.

In Section 2 we summarize the pertinent results of part I for isotropic composites and, in particular, explicitly give the third-order approximations for the effective bulk and shear moduli in arbitrary space dimension d. The microstructural information incorporated by the approximate formulas include phase volume fractions and parameters that depend on the three-point probability function $S_n^{(p)}$. In Section 3 we investigate the validity of the formulas for ordered and disordered models of isotropic two-dimensional dispersions by comparing the predictions to benchmark data, rigorous bounds and self-consistent approximations for a wide range of conditions. In Section 4 we carry out a similar analysis for ordered and disordered models of isotropic and cubic-symmetric three-dimensional dispersions. Finally, we discuss the results in Section 5.

2. SUMMARY OF RESULTS FOR ELASTIC MODULI OF ISOTROPIC DISPERSIONS

Here we summarize concisely the main results obtained in part I pertaining to expressions for the effective bulk modulus K_e and shear modulus G_e of two-phase isotropic composites. We also remark on the significance of the exact results for the special case of composites consisting of particles of well-defined shape in a matrix.

As in part I, we let p and q denote the "polarized" and "reference" phases, such that $p, q = 1, 2$ but $p \neq q$. It is convenient first to introduce the scalar "polarizabilities" defined by the following relations:

$$\kappa_{pq} = \frac{K_p - K_q}{K_p + \dfrac{2(d-1)}{d} G_q}, \tag{2.1}$$

$$\mu_{pq} = \frac{G_p - G_q}{G_p + \dfrac{G_q[dK_q/2 + (d+1)(d-2)G_q/d]}{K_q + 2G_q}}, \tag{2.2}$$

$$\kappa_{eq} = \frac{K_e - K_q}{K_e + \dfrac{2(d-1)}{d} G_q}, \tag{2.3}$$

$$\mu_{eq} = \frac{G_e - G_q}{G_e + \dfrac{G_q[dK_q/2 + (d+1)(d-2)G_q/d]}{K_q + 2G_q}}. \tag{2.4}$$

The quantities κ_{pq} and μ_{pq} are, respectively, the bulk and shear moduli polarizabilities involving the phase moduli. On the other hand, the parameters κ_{eq} and μ_{eq} are the *effective* bulk and shear moduli polarizabilities, respectively, which depend on both the effective and phase moduli.

2.1. *Bulk modulus*

We showed in Part I that the effective bulk modulus K_e of an isotropic, d-dimensional, two-phase composite can be obtained exactly from the series expansion

$$\phi_p^2 \frac{\kappa_{pq}}{\kappa_{eq}} = \phi_p - \sum_{n=3}^{\infty} C_n^{(p)}, \tag{2.5}$$

where the scalar coefficients $C_n^{(p)}$ are multidimensional integrals over a certain determinant involving the set of n-point probability functions $S_1^{(p)}, \ldots, S_n^{(p)}$. For example, the third-order coefficient is given explicitly by

$$\frac{C_3^{(p)}}{\phi_p} = \frac{(d+2)(d-1)G_q\kappa_{pq}\mu_{pq}}{d(K_q+2G_q)}\phi_q\zeta_p \tag{2.6}$$

where $\zeta_p = 1-\zeta_q$ is a d-dimensional, three-point microstructural parameter defined by

$$\zeta_p = \frac{d^2}{(d-1)\phi_q\phi_p\Omega^2}\iint \frac{d\mathbf{r}}{r^d}\frac{d\mathbf{s}}{s^d}[d(\mathbf{n}\cdot\mathbf{m})^2-1]\left[S_3^{(p)}(\mathbf{r},\mathbf{s}) - \frac{S_2^{(p)}(\mathbf{r})S_2^{(p)}(\mathbf{s})}{\phi_p}\right], \tag{2.7}$$

Ω is the solid angle contained in a d-dimensional sphere and $\mathbf{n} = \mathbf{r}/|\mathbf{r}|$ and $\mathbf{m} = \mathbf{s}/|\mathbf{s}|$ are unit vectors. The parameter ζ_p, which lies in the closed interval [0, 1] for any space dimension d, also arises in rigorous bounds on the effective bulk and shear moduli of two- and three-dimensional composites (Beran and Molyneux, 1966; McCoy, 1970; Milton, 1981, 1982; Milton and Phan-Thien, 1982; Torquato, 1991).

It was demonstrated that series (2.5) can be viewed as an expansion that perturbs around the Hashin–Shtrikman structures. In the family of such structures, one of the phases is a disconnected phase that is dispersed throughout a continuous matrix (Hashin and Shtrikman, 1963; Hashin, 1965; Francfort and Murat 1986). Therefore, expansion (2.5) will 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. Accordingly, we contend that the truncation of this expansion after third-order terms should provide an excellent approximation of the effective bulk modulus K_e of such dispersions for a wide range of conditions. Through third-order terms eqn (2.5) yields

$$\phi_p \frac{\kappa_{pq}}{\kappa_{eq}} = 1 - \frac{(d+2)(d-1)G_q \kappa_{pq} \mu_{pq}}{d(K_q + 2G_q)} \phi_q \zeta_p. \tag{2.8}$$

It is seen that when the three-point parameter ζ_p equals zero, formula (2.8) reduces to the appropriate Hashin–Shtrikman bound, which is exact for the optimal structures corresponding to this bound (Torquato, 1997).

We demonstrate quantitatively in the next sections that relation (2.8) provides an excellent approximation of the effective elastic moduli for a variety of two- and three-dimensional ordered and disordered dispersions. It will be shown that for a dispersed phase that is stiffer than the matrix, relation (2.8) behaves (to an excellent approximation) as a higher-order lower bound on K_e. On the other hand, for a dispersed phase that is more compliant than the matrix, relation (2.8) behaves (to an excellent approximation) as a higher-order upper bound on K_e.

2.2. *Shear modulus*

It was demonstrated in part I that the effective shear modulus G_e of an isotropic, d-dimensional, two-phase composite can be obtained exactly from the series expansion.

$$\phi_p^2 \frac{\mu_{pq}}{\mu_{eq}} = \phi_p - \sum_{n=3}^{\infty} D_n^{(p)}, \tag{2.9}$$

where the scalar coefficients $D_n^{(p)}$ are multidimensional integrals over a certain determinant involving the set of n-point probability functions $S_1^{(p)}, \ldots, S_n^{(p)}$. For example, the third-order coefficient is given explicitly by

$$\frac{D_3^{(p)}}{\phi_p} = \frac{2G_q \kappa_{pq} \mu_{pq}}{d(K_q + 2G_q)} \phi_q \zeta_p + \frac{(d^2-4)G_q(2K_q+3G_q)\mu_{pq}^2}{2d(K_q+2G_q)^2} \phi_q \zeta_p + \frac{1}{2d} \left[\frac{dK_q + (d-2)G_q}{K_q + 2G_q} \right]^2 \mu_{pq}^2 \phi_q \eta_p, \tag{2.10}$$

where η_p is a d-dimensional, three-point microstructural parameter defined by

$$\eta_p = -\frac{(d+2)(5d+6)}{d^2} \zeta_p + \frac{(d+2)^2}{(d-1)\phi_q \phi_p \Omega^2} \iint \frac{\mathrm{d}\mathbf{r}}{r^d} \frac{\mathrm{d}\mathbf{s}}{s^d} [d(d+2)(\mathbf{n}\cdot\mathbf{m})^4 - 3] \times \left[S_3^{(p)}(\mathbf{r}, \mathbf{s}) - \frac{S_2^{(p)}(\mathbf{r}) S_2^{(p)}(\mathbf{s})}{\phi_p} \right], \tag{2.11}$$

where $\mathbf{n} = \mathbf{r}/|\mathbf{r}|$ and $\mathbf{m} = \mathbf{s}/|\mathbf{s}|$ are unit vectors. The parameter η_p, which generally lies in the closed interval [0, 1] for any space dimension d, also arises in rigorous bounds on the effective shear modulus of two- and three-dimensional composites (Beran and Molyneux, 1966; McCoy, 1970; Milton, 1981, 1982; Milton and Phan-Thien, 1982; Torquato, 1991; Gibiansky and Torquato, 1995). We note in passing that the ratio

K_q/G_q for phase q appearing in both eqns (2.6) and (2.10) can be written in terms of the d-dimensional Poisson's ratio v_q for phase q via the expression

$$v_q = \frac{dK_q - 2G_q}{d(d-1)K_q + 2G_q}$$

where v_q lies in the interval $[-1,(d-1)^{-1}]$.

As in the case of the bulk modulus, we showed in part I that series (2.9) can be viewed as an expansion that perturbs around the Hashin–Shtrikman structures for the effective shear modulus. This family includes finite-rank laminates (Francfort and Murat, 1986) in which one of the phases is a disconnected phase that is dispersed throughout a continuous matrix. Hence, expansion (2.9) will 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 truncation of expansion (2.9) after third-order terms should provide an excellent approximation of the effective shear modulus G_e of such dispersions for a wide range of conditions. Truncating the series (2.9) after third-order terms yields

$$\phi_p \frac{\mu_{pq}}{\mu_{eq}} = 1 - \frac{2G_q \kappa_{pq}\mu_{pq}}{d(K_q+2G_q)}\phi_q\zeta_p - \frac{(d^2-4)G_q(2K_q+3G_q)\mu_{pq}^2}{2d(K_q+2G_q)^2}\phi_q\zeta_p - \frac{1}{2d}\left[\frac{dK_q+(d-2)G_q}{K_q+2G_q}\right]^2 \mu_{pq}^2\phi_q\eta_p. \quad (2.12)$$

When both three-point parameters ζ_p and η_p equal zero, formula (2.12) reduces to the appropriate Hashin–Shtrikman bound, which is exact for the optimal structures corresponding to this bound (Torquato, 1997). We note that Torquato (1985) derived a formula for the effective conductivity of d-dimensional, isotropic dispersions that is completely analogous to the elastic-moduli expressions (2.8) and (2.12).

In the subsequent sections, we will show that relation (2.12) provides an excellent approximation of the effective elastic moduli for a variety of two-and three-dimensional ordered and disordered dispersions. It will be demonstrated that for a dispersed phase that is stiffer than the matrix, relation (2.12) behaves (to an excellent approximation) as a higher-order lower bound on G_e. On the other hand, for a dispersed phase that is more compliant than the matrix, relation (2.12) behaves (to an excellent approximation) as a higher-order upper bound on G_e.

2.3. *Shear modulus as a function of the space dimension* d

In part I we showed that for any d-dimensional, macroscopically isotropic, n-phase composite possessing non-zero phase moduli, the effective shear modulus G_e is independent of the microstructure and is exactly given by the arithmetic average, i.e.,

$$G_e = \sum_{i=1}^{n} \phi_i G_i, \quad (2.13)$$

in the limit that the space dimension becomes infinite ($d \to \infty$). It was also shown that the effective conductivity behaves in the analogous fashion as $d \to \infty$.

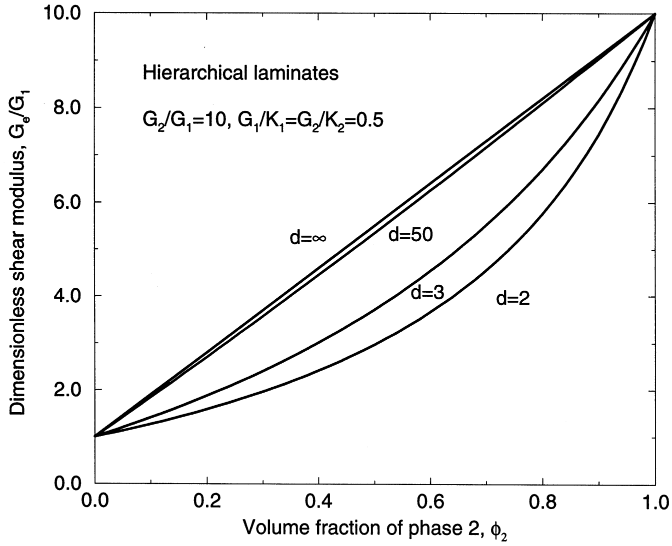

Fig. 1. Dimensionless effective shear modulus G_e/G_1 vs volume fraction of phase 2 ϕ_2 for the hierarchical laminates that realize the Hashin–Shtrikman upper bounds for several values of the space dimension d.

In Fig. 1 we plot the effective shear modulus vs volume fraction for several different values of d for the two-phase hierarchical laminates that realize the Hashin–Shtrikman upper bound. The stiffer, connected phase is phase 2. It is seen that the tendency for the effective shear modulus to approach the arithmetic average as d is made large is already seen in low dimensions, i.e., in going from $d = 2$ to $d = 3$. However, it is also observed that convergence to the arithmetic-mean result ($d = \infty$) is slow as evidenced by the fact that the result for $d = 50$ is still perceptibly different than the former.

2.4. *Remark on exact results for particle dispersions*

An important practical class of composites consists of particles of well-defined shapes (e.g., oriented circular or elliptical cylinders, spheres or ellipsoids) in a matrix. This class of dispersions includes certain fiber-reinforced materials, particulate-reinforced composites, and colloidal dispersions. For such dispersions, Torquato and Stell (1982) have related $S_n^{(2)}$ to multi-dimensional integrals over the infinite set of m-particle distribution functions $g_1, g_2, \ldots, g_{\mathrm{m}}$ $(m \to \infty)$. Roughly speaking, $g_m(\mathbf{r}_1, \ldots, \mathbf{r}_m)$ gives the probability density function associated with finding m particles in a particular spatial configuration $\mathbf{r}_1, \ldots, \mathbf{r}_m$. (The quantity $\mathbf{r}_i$ specifies the center-of-mass and orientation coordinates of a particle.) The g_m are the most basic quantities that characterize the structure of many-particle systems and have been well-studied in the statistical mechanics of liquids and solids (Hansen and McDonald, 1986).

Now consider the exact relations (2.5) and (2.9) for the effective bulk and shear moduli for such dispersions. In light of the discussion above and the fact that the nth term involves the n-point probability function $S_n^{(2)}$, we see that generally the effective moduli will depend upon the infinite set m-particle distribution functions $g_1, g_2, \ldots, g_m$

$(m \to \infty)$. Thus, theoretical expressions for the effective elastic moduli for particle dispersions that incorporate only lower-order information, such as the two-particle function g_2 (see Verberg, *et al.*, 1997), are necessarily approximations over the full range of particle densities. In Appendix A, we show that the three-point parameters ζ_2 and η_2 must depend nontrivially on both g_2 and g_3 for the case of nonoverlapping particle dispersions. For more general interparticle potentials, ζ_2 and η_2 will actually depend on the infinite set $g_1, g_2, \ldots, g_m$ $(m \to \infty)$.

3. TWO-DIMENSIONAL DISPERSIONS

In this section we shall apply the formulas (2.8) and (2.12) for two-dimensional dispersions. Thus, the ensuing results apply to fiber-reinforced materials with transverse isotropy or to isotropic two-phase composites in the form of thin sheets. To emphasize that the bulk modulus is a planar quantity, we shall denote it here by k instead of K which will be reserved for the three-dimensional bulk modulus. In the case of a fiber-reinforced dispersion, the appropriate planar bulk modulus k is the plane-strain bulk modulus which is related to the three-dimensional one by the relation

$$k = K + G/3.$$

If we are dealing with plane-stress elasticity (appropriate for a two-dimensional composite sheet), then the plane-stress bulk modulus k obeys the relation

$$k = \frac{9KG}{3K+4G}.$$

Let phases 1 and 2 denote the matrix and dispersed phases, respectively. For $d = 2$, relations (2.8) and (2.12) give

$$\frac{k_e}{k_1} = \frac{1 + \dfrac{G_1}{k_1}\kappa\phi_2 - \dfrac{2G_1}{k_1+2G_1}\kappa\mu\phi_1\zeta_2}{1 - \kappa\phi_2 - \dfrac{2G_1}{k_1+2G_1}\kappa\mu\phi_1\zeta_2}, \tag{3.1}$$

$$\frac{G_e}{G_1} = \frac{1 + \dfrac{k_1}{k_1+2G_1}\mu\phi_2 - \dfrac{G_1}{k_1+2G_1}\kappa\mu\phi_1\zeta_2 - \dfrac{k_1^2}{(k_1+2G_1)^2}\mu^2\phi_1\eta_2}{1 - \mu\phi_2 - \dfrac{G_1}{k_1+2G_1}\kappa\mu\phi_1\zeta_2 - \dfrac{k_1^2}{(k_1+2G_1)^2},\mu^2\phi_1\eta_2}, \tag{3.2}$$

where

$$\kappa \equiv \kappa_{21} = \frac{k_2 - k_1}{k_2 + G_1}, \tag{3.3}$$

$$\mu \equiv \mu_{21} = \frac{G_2 - G_1}{G_2 + G_1\dfrac{k_1}{k_1+2G_1}}. \tag{3.4}$$

For any isotropic, two-dimensional composite, the three-point parameters ζ_2 and η_2 are defined by the following three-fold integrals:

$$\zeta_2 = \frac{4}{\pi\phi_1\phi_2}\int_0^\infty \frac{\mathrm{d}r}{r}\int_0^\infty \frac{\mathrm{d}s}{s}\int_0^\pi \mathrm{d}\theta\cos(2\theta)\left[S_3^{(2)}(r,s,t) - \frac{S_2^{(2)}(r)S_2^{(2)}(s)}{\phi_2}\right], \quad (3.5)$$

$$\eta_2 = \frac{16}{\pi\phi_1\phi_2}\int_0^\infty \frac{\mathrm{d}r}{r}\int_0^\infty \frac{\mathrm{d}s}{s}\int_0^\pi \mathrm{d}\theta\cos(4\theta)\left[S_3^{(2)}(r,s,t) - \frac{S_2^{(2)}(r)S_2^{(2)}(s)}{\phi_2}\right], \quad (3.6)$$

where θ is the angle opposite the side of the triangle of length t. The parameters ζ_2 and η_2 have also arisen in rigorous bounds on the effective moduli of two-dimensional composites (Silnutzer, 1972; Milton, 1981; Gibiansky and Torquato, 1995) and have been computed for a variety of model dispersions (see the review of Torquato, 1991). In the special case of statistically isotropic, two-dimensional arrays of nonoverlapping particles, Torquato and Lado (1988, 1991) showed that both ζ_2 and η_2 can be expressed exactly in terms of integrals over the two- and three-particle distribution functions, g_2 and g_3, defined in Section 2.3 (see Appendix A for further details).

We will require that the two-dimensional approximations (3.1) and (3.2) always lie within the most restrictive three-point upper and lower bounds which are summarized in a compact form in Appendix B. This generally will place restrictions on the range of values of the geometrical parameters ζ_2 and η_2 beyond the normal condition that they must lie in the interval [0, 1]. In some instances, this range of values can only be determined numerically, but in other cases, one can obtain the restrictions analytically. For example, for the two-dimensional effective bulk modulus formula (3.1), it is shown in Appendix B that

$$\zeta_2 \leqslant 0.5$$

in order for eqn (3.1) to lie between the Silnutzer lower bound and the Gibiansky–Torquato upper bound. For the shear modulus, it is usually difficult to obtain similar analytical conditions. However, in the instance of a two-dimensional composite in which both phases are incompressible (i.e., $k_1/G_1 = k_2/G_2 \to \infty$), it is shown in Appendix B that there is no additional restriction on η_2. Thus, for any physically realizable η_2 in this incompressible limit, the prediction of eqn (3.2) always lies within the most restrictive three-point bounds. (As shown below, the expression (3.2) for G_e is independent of ζ_2 for an incompressible composite.) It is noteworthy that for a number of realistic models of two-dimensional dispersions (Torquato, 1991), the parameters ζ_2 and η_2 are such that the estimates from relations (3.1) and (3.2) always lie within the tightest three-point bounds.

In order to validate relations (3.1) and (3.2) as accurate approximations for the effective moduli of two-dimensional dispersions, we will compare them to benchmark model calculations. One such benchmark study was recently carried out by Eischen and Torquato (1993) who obtained comprehensive elastic-moduli simulation data for models of hexagonal arrays of infinitely long, aligned cylinders in a matrix or a thin-plate composite consisting of hexagonal arrays of disks in a matrix. They presented data for eight different phase-material values over a wide range of inclusion volume fractions. To compare the predictions of eqns (3.1) and (3.2) to this data, we also need to know the parameters ζ_2 and η_2 as a function of volume fraction ϕ_2. The evaluations of ζ_2 and η_2 for hexagonal arrays of inclusions were given, respectively, by McPhedran and Milton (1981) and Eischen and Torquato (1993). We will also compare our results rigorous bounds as well as to the self-consistent (SC) approximations for two-dimensional, two-phase isotropic composites (Hill, 1965a) :

$$\phi_1\left[\frac{k_e-k_1}{G_e+k_1}\right]+\phi_2\left[\frac{k_e-k_2}{G_e+k_2}\right]=0, \tag{3.7}$$

$$\phi_1\left[\frac{G_e-G_1}{G_e k_e/(k_e+2G_e)+G_1}\right]+\phi_2\left[\frac{G_e-G_2}{G_e k_e/(k_e+2G_e)+G_2}\right]=0. \tag{3.8}$$

In Fig. 2 we compare the prediction of eqn (3.2) for the effective transverse shear modulus G_e of hexagonal arrays of glass fibers in an epoxy matrix ($G_2/G_1 = 22.5$, $G_1/k_1 = 0.3$, $G_2/k_2 = 0.6$) to the corresponding simulation data of Eischen and Torquato (1993). It is seen that formula (3.2) provides an excellent estimate of the shear modulus for the entire range of volume fractions. The three-point Silnutzer lower bound [given in eqn (B.14)] is very slightly below eqn (3.2) and hence is not shown in the figure. (The prediction of the effective bulk modulus from relation (3.1) is equally accurate but is not shown here.) The two-dimensional SC approximation (3.8) and the three-point Gibiansky–Torquato upper bound [given in eqn (B.14)] in this instance are also included in Fig. 2. It is seen that the SC approximation not only overestimates the effective shear modulus of the glass–epoxy composite for moderate

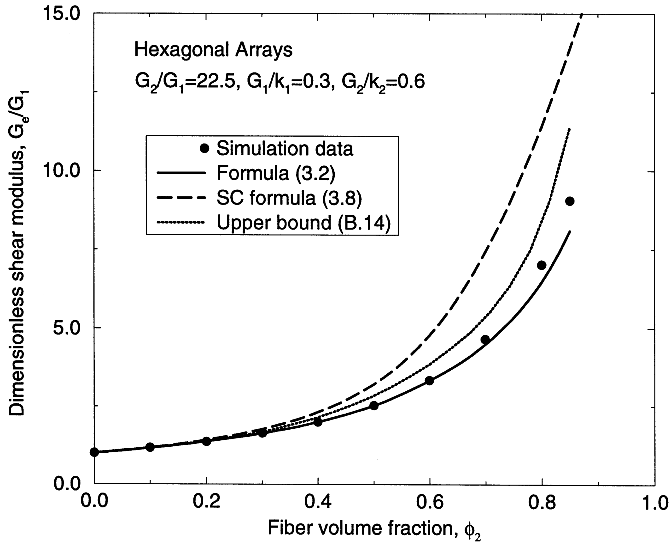

Fig. 2. Dimensionless effective transverse shear modulus G_e/G_1 vs fiber volume fraction ϕ_2 for hexagonal arrays of circular glass fibers in an epoxy matrix. Filled circles are simulation data of Eischen and Torquato (1993), solid curve is our formula (3.2), dashed curve is the SC formula (3.8), and the dotted circle is the Gibiansky and Torquato (1995) three-point upper bound given in eqn (B.14). The parameters ζ_2 and η_2 were taken from McPhedran and Milton (1981) and Eischen and Torquato (1993), respectively.

to high fiber fractions but it begins to violate the upper bound at small values of the fiber volume fraction ϕ_2. However, it is well known that the SC approximations always lie within the lower-order Hashin-Shtrikman bounds.

In what follows, we will examine special cases of relations (3.1) and (3.2) in which one or both of the phases have extreme values, including sheets containing holes, rigid fibers in compressible matrices, and incompressible fiber-reinforced composites. We will compare our predictions to simulation data for hexagonal arrays when it is available in these limiting cases. Moreover, we will apply the formulas (3.1) and (3.2) to random arrays of inclusions for which there is no comprehensive numerical data of the effective moduli.

3.1. *Sheet containing holes*

Consider a sheet (phase 1) containing nonoverlapping holes in which $k_2 = G_2 = 0$. Then relations (3.1) and (3.2) become

$$\frac{k_e}{k_1} = \frac{\phi_1(1-2\zeta_2)}{1+\dfrac{k_1}{G_1}\phi_2 - 2\phi_1\zeta_2}, \tag{3.9}$$

$$\frac{G_e}{G_1} = \frac{\phi_1(1-\zeta_2-\eta_2)}{1+\dfrac{k_1+2G_1}{k_1}\phi_2 - \phi_1(\zeta_2+\eta_2)}. \tag{3.10}$$

Figure 3 compares the prediction of formula (3.9) for the effective bulk modulus

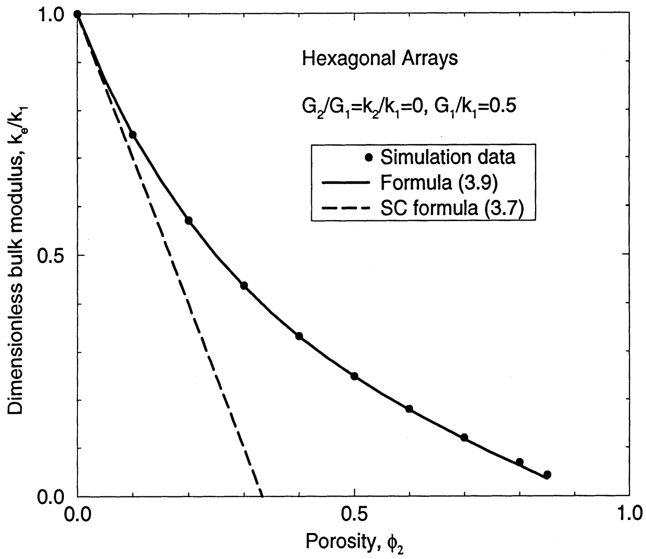

Fig. 3. Dimensionless effective transverse bulk modulus k_e/k_1 vs fiber volume fraction ϕ_2 for hexagonal arrays of circular holes in a compressible sheet. Filled circles are simulation data of Eischen and Torquato (1993), solid curve is our formula (3.9), and the dashed curve is the SC formula (3.7). The values of the parameter ζ_2 were taken from McPhedran and Milton (1981).

to corresponding simulation data of Eischen and Torquato (1993) for hexagonal arrays of circular holes in a matrix for which $G_2/G_1 = k_2/k_1 = 0$ and $G_1/k_1 = 0.5$. The theoretical prediction (3.9) provides an excellent estimate of the effective bulk modulus. On the other hand, the SC approximation (3.7) grossly underestimates the effective bulk modulus for non-dilute hole volume fractions. The three-point upper bound of Gibiansky and Torquato (1995) [see eqns (B.11) or (B.12)] is virtually indistinguishable from eqn (3.9) and of course the associated three-point lower bound is trivially equal to zero.

Cherkaev *et al.* (1992) have shown that the effective dimensionless Young's modulus E_e/E_1 for any sheet containing holes of arbitrary geometry is independent of the Poisson's ratio of the solid phase. It is easily shown that the approximations (3.9) and (3.10) are consistent with this theorem by using the interrelations

$$\frac{4}{E_e} = \frac{1}{k_e} + \frac{1}{G_e}, \quad k_1 = \frac{E_1}{2(1-\nu_1)}, \quad G_1 = \frac{E_1}{2(1+\nu_1)}, \tag{3.11}$$

in conjunction with the previous two expressions to yield

$$\frac{E_e}{E_1} = \frac{\phi_1(1-2\zeta_2)(1-\zeta_2-\eta_2)}{1+2\phi_2-2(1+\phi_2)\zeta_2+(\zeta_2+\eta_2)(2\phi_1\zeta_2-1)}. \tag{3.12}$$

It is seen that eqn (3.12) is indeed independent of the Poisson's ratio of the solid phase.

3.2. *Rigid fibers in compressible matrices*

Consider the instance of a fiber-reinforced composite consisting of rigid fibers, i.e., $G_2/G_1 = k_2/k_1 = \infty$, in a compressible matrix. For such a composite, $\kappa = \mu = 1$ and hence, according to eqns (3.1) and (3.2), the effective bulk and shear moduli are given, respectively, by

$$\frac{k_e}{k_1} = \frac{1+\dfrac{G_1}{k_1}\phi_2 - \dfrac{2G_1}{k_1+2G_1}\phi_1\zeta_2}{1-\phi_2-\dfrac{2G_1}{k_1+2G_1}\phi_1\zeta_2}, \tag{3.13}$$

$$\frac{G_e}{G_1} = \frac{1+\dfrac{k_1}{k_1+2G_1}\phi_2 - \dfrac{G_1}{k_1+2G_1}\phi_1\zeta_2 - \dfrac{k_1^2}{(k_1+2G_1)^2}\phi_1\eta_2}{1-\phi_2-\dfrac{G_1}{k_1+2G_1}\phi_1\zeta_2-\dfrac{k_1^2}{(k_1+2G_1)^2}\phi_1\eta_2}. \tag{3.14}$$

Figure 4 compares relation (3.13) and the SC approximation (3.7) for $d = 2$ to the simulation data of Eischen and Torquato for hexagonal arrays of rigid circular fibers in a compressible matrix in which $G_2/G_1 = k_2/k_1 = \infty$ and $G_1/k_1 = 0.4$. The prediction of eqn (3.13) is remarkably accurate, whereas the SC approximation (3.7) begins to diverge increasingly from the data for values of the fiber volume fraction ϕ_2 larger than 0.5. Silnutzer's three-point lower bound is again virtually indistinguishable from the prediction (3.13) ; the associated upper bound diverges to infinity in this instance.

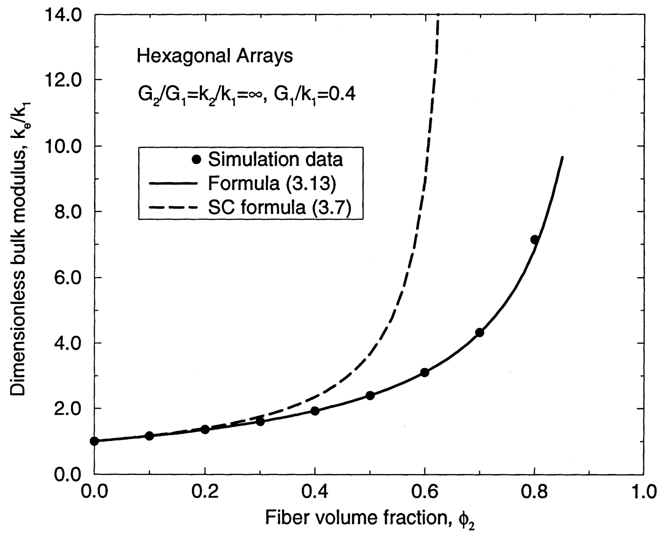

Fig. 4. Dimensionless effective transverse bulk modulus k_e/k_1 vs fiber volume fraction ϕ_2 for hexagonal arrays of circular rigid fibers in a compressible matrix. Filled circles are simulation data of Eischen and Torquato (1993), solid curve is our formula (3.13), and the dashed curve is the SC formula (3.7). The values of the parameter ζ_2 were taken from McPhedran and Milton (1981).

Let us now consider a fiber-reinforced material consisting of rigid, nonoverlapping, circular fibers that are randomly arranged in a compressible matrix ($G_2/G_1 = k_2/k_1 = \infty$, $G_1/k_1 = 0.4$). To our knowledge, exact benchmark calculations of the effective transverse moduli do not exist for this case. However, we can compare our results to rigorous cross-property bounds that utilize effective conductivity measurements to bound the effective elastic moduli (and vice versa). Kim and Torquato (1990) have computed the effective transverse conductivity σ_e of a fiber-reinforced material comprised of an "equilibrium" distribution of oriented, circular cylinders of conductivity σ_2 in a matrix of conductivity σ_1. Roughly speaking, an equilibrium distribution of nonoverlapping particles is the most random distribution subject to the condition of impenetrability. An equilibrium distribution of nonoverlapping cylinders achieves a maximum fiber fraction at the random-close packing value of $\phi_2 \approx 0.82$, which is considerably higher than the maximum value for random sequential addition of cylinders which occurs at the "jammed" state when $\phi_2 \approx 0.55$ (Torquato, 1995). Using the Gibiansky and Torquato (1995) cross-property relations and the effective conductivity data of Kim and Torquato for the superconducting case ($\sigma_2/\sigma_1 = \infty$), we can bound the effective elastic moduli from above. Employing the tabulation of ζ_2 for an equilibrium arrangement of cylinders (Torquato and Lado, 1988; Torquato, 1991), we compare in Fig. 5 the bulk-modulus prediction of (3.13) to the aforementioned cross-property upper bound and to the SC approximation (3.7). The three-point Silnutzer lower bound is very slightly below our approximation (3.13) and hence is not shown. Not surprisingly, the SC formula (3.7) violates the upper bound at a moderate value of the fiber volume fraction.

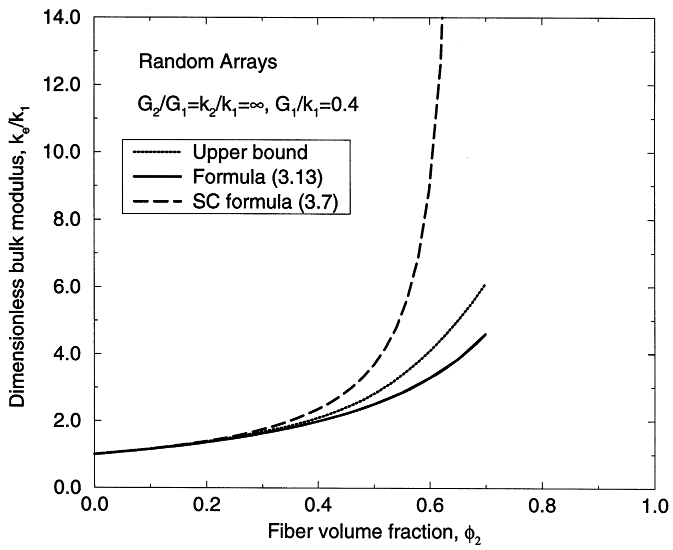

Fig. 5. Dimensionless effective transverse bulk modulus k_e/k_1 vs fiber volume fraction ϕ_2 for random arrays of circular rigid fibers in a compressible matrix. Solid curve is our formula (3.13), the dashed curve is the SC formula (3.7), and the dotted curve is the Gibiansky and Torquato (1995) cross-property upper bound using the conductivity data of Kim and Torquato (1992) as described in the text. The values of the parameter ζ_2 were taken from McPhedran and Milton (1981).

3.3. *Incompressible fiber-reinforced composites*

Let us consider the case of an incompressible fiber-reinforced composite, i.e., $k_1/G_1 = \infty$ and $k_2/G_2 = \infty$. In this limit, relation (3.2) for the effective shear modulus reduces to

$$\frac{G_e}{G_1} = \frac{1+\mu\phi_2-\mu^2-\mu^2\phi_1\eta_2}{1-\mu\phi_2-\mu^2\phi_1\eta_2}, \tag{3.15}$$

where

$$\mu = \frac{G_2-G_1}{G_2+G_1}. \tag{3.16}$$

Now let us in addition consider the limit in which the included phase is rigid ($G_2/G_1 \to \infty$), the most difficult case to treat theoretically. Then for such an incompressible composite we find that eqn (3.15) gives

$$\frac{G_e}{G_1} = \frac{1+\phi_2-\phi_1\eta_2}{1-\phi_2-\phi_1\eta_2}. \tag{3.17}$$

It is seen that in the limit of an incompressible matrix ($k_1/G_1 = \infty$), formula (3.14) becomes identical to relation (3.17).

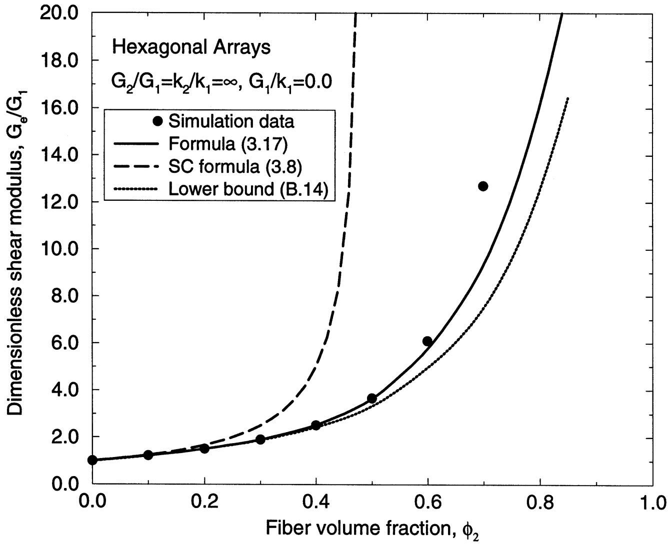

Fig. 6. Dimensionless effective transverse shear modulus G_e/G_1 vs fiber volume fraction ϕ_2 for hexagonal arrays of circular rigid fibers in an incompressible matrix. Filled circles are simulation data of Eischen and Torquato (1993), solid curve is our formula (3.17), dashed curve is the SC formula (3.8), and the dotted curve is Silnutzer's three-point lower bound given in (B.14). The parameters ζ_2 and η_2 were taken from McPhedran and Milton (1981) and Eischen and Torquato (1993), respectively.

Figure 6 compares formula (3.17), the SC approximation and Silnutzer's three-point lower bound to the shear-modulus simulation data of Eischen and Torquato (1993) for the case of an incompressible composite in which rigid circular fibers are arranged in a hexagonal array ($G_2/G_1 = k_2/k_1 = \infty$, $G_1/k_1 = 0$). Here it is seen that the formula (3.17) provides an excellent estimate of the effective shear modulus up to a fiber volume fraction $\phi_2 = 0.6$ but begins to diverge from the data beyond this volume fraction (see discussion in Section 5). The SC approximation diverges from the data at significantly lower volume fractions since it predicts incorrectly a percolation threshold at $\phi_2 = 0.5$, i.e., the rigid phase becomes connected at this critical value. Of course, the true percolation threshold occurs at $\phi_2 = \pi/(2\sqrt{3}) \approx 0.907$.

4. THREE-DIMENSIONAL DISPERSIONS

Here we shall apply the formulas (2.8) and (2.12) for three-dimensional dispersions. Let phases 1 and 2 denote the matrix and dispersed phases, respectively. For $d = 3$, relations (2.8) and (2.12) give

$$\frac{K_e}{K_1} = \frac{1 + \dfrac{4G_1}{3K_1}\kappa\phi_2 - \dfrac{10G_1}{3(K_1+2G_1)}\kappa\mu\phi_1\zeta_2}{1 - \kappa\phi_2 - \dfrac{10G_1}{3(K_1+2G_1)}\kappa\mu\phi_1\zeta_2}, \tag{4.1}$$

$$\frac{G_e}{G_1}=\frac{1+\dfrac{9K_1+8G_1}{6(K_1+2G_1)}\mu\phi_2-\dfrac{2\kappa\mu G_1}{3(K_1+2G_1)}\phi_1\zeta_2-\dfrac{\mu^2}{6}\left\{\left[\dfrac{3K_1+G_1}{K_1+2G_1}\right]^2\phi_1\eta_2+5G_1\left[\dfrac{2K_1+3G_1}{(K_1+2G_1)^2}\right]\phi_1\zeta_2\right\}}{1-\mu\phi_2-\dfrac{2\kappa\mu G_1}{3(K_1+2G_1)}\phi_1\zeta_2-\dfrac{\mu^2}{6}\left\{\left[\dfrac{3K_1+G_1}{K_1+2G_1}\right]^2\phi_1\eta_2+5G_1\left[\dfrac{2K_1+3G_1}{(K_1+2G_1)^2}\right]\phi_1\zeta_2\right\}},\tag{4.2}$$

where

$$\kappa\equiv\kappa_{21}=\frac{K_2-K_1}{K_2+\dfrac{4G_1}{3}},\tag{4.3}$$

$$\mu\equiv\mu_{21}=\frac{G_2-G_1}{G_2+G_1\left[\dfrac{9K_1+8G_1}{6(K_1+2G_1)}\right]}.\tag{4.4}$$

For any isotropic, three-dimensional composite, the three-point parameters ζ_2 and η_2 are defined by the following three-fold integrals:

$$\zeta_2=\frac{9}{2\phi_1\phi_2}\int_0^\infty\frac{\mathrm{d}r}{r}\int_0^\infty\frac{\mathrm{d}s}{s}\int_{-1}^{1}\mathrm{d}(\cos\theta)P_2(\cos\theta)\left[S_3^{(2)}(r,s,t)-\frac{S_2^{(2)}(r)S_2^{(2)}(s)}{\phi_2}\right],\tag{4.5}$$

$$\eta_2=\frac{5\zeta_2}{21}+\frac{150}{7\phi_1\phi_2}\int_0^\infty\frac{\mathrm{d}r}{r}\int_0^\infty\frac{\mathrm{d}s}{s}\int_{-1}^{1}\mathrm{d}(\cos\theta)P_4(\cos\theta)\left[S_3^{(2)}(r,s,t)-\frac{S_2^{(2)}(r)S_2^{(2)}(s)}{\phi_2}\right],\tag{4.6}$$

where P_2 and P_4 are the Legendre polynomials of order 2 and 4, respectively, and θ is the angle opposite the side of the triangle of length t. The parameters ζ_2 and η_2 have also arisen in rigorous bounds on the effective moduli of three-dimensional composites (Beran and Molyneux, 1966; McCoy, 1970; Milton, 1981; Milton and Phan-Thien, 1982) and have been computed for a variety of model dispersions (see the review of Torquato, 1991). In the special case of statistically isotropic, three-dimensional arrays of nonoverlapping particles, it has been demonstrated (Lado and Torquato, 1986; Sen *et al.*, 1989) that both ζ_2 and η_2 can be expressed exactly in terms of integrals over the two- and three-particle distribution functions, g_2 and g_3, defined in Section 2.3 (see Appendix A for further details).

Again, as we did for $d=2$, we will require that the three-dimensional approximations (4.1) and (4.2) always lie within the most restrictive three-point upper and lower bounds which are summarized in a compact form in Appendix B. This generally implies that the intervals in which the geometrical parameters ζ_2 and η_2 lie will be more restrictive than the interval [0, 1]. For the three-dimensional effective bulk modulus formula (4.1), it is shown in Appendix B that when $G_1/K_1 \leqslant 0.75$ then

$$\zeta_2 \leqslant 0.6 + \frac{8}{15}\frac{G_1}{K_1},$$

in order for (4.1) to lie between the Beran–Molyneux bounds. However, if $G_1/K_1 > 0.75$, then there is no additional restriction on ζ_2. We note that the parameter ζ_2 is less restricted in three dimensions than it is in two dimensions. For the shear modulus, it is very difficult to obtain similar analytical conditions. However, in the instance of a three-dimensional composite in which both phases are incompressible (i.e., $K_1/G_1 = K_2/G_2 \to \infty$), it is shown in Appendix B that

$$\frac{1+\eta_2}{2-3\eta_2} \geqslant \frac{1-\frac{11}{16}\zeta_2-\frac{5}{16}\eta_2}{2(1-\zeta_2)(1+\frac{5}{16}\zeta_2-\frac{21}{16}\eta_2)}.$$

We emphasize that for a number of realistic models of three-dimensional dispersions (Torquato, 1991), the parameters ζ_2 and η_2 are such that the estimates from relations (4.1) and (4.2) always lie within the tightest three-point bounds.

In order to validate the accuracy of approximations (4.1) and (4.2) for the effective moduli of three-dimensional dispersions, we will compare them to rigorous bounds and SC approximations for ordered and disordered model microstructures. The SC approximations for the effective bulk and shear moduli of three-dimensional, two-phase composites (Hill, 1965b; Budiansky, 1965) are, respectively, given by

$$\phi_1\left[\frac{K_e-K_1}{4G_e/3+K_1}\right]+\phi_2\left[\frac{K_e-K_2}{4G_e/3+K_2}\right]=0, \tag{4.7}$$

$$\phi_1\left[\frac{G_e-G_1}{G_e(9K_e+8G_e)/(6K_e+12G_e)+G_1}\right]+\phi_2\left[\frac{G_e-G_2}{G_e(9K_e+8G_e)/(6K_e+12G_e)+G_2}\right]=0. \tag{4.8}$$

In the special case of cubic lattices of rigid spheres in compressible matrices, we will compare our bulk modulus prediction (4.1) to the numerical data of Nunan and Keller (1984). Our result eqn (4.1) for the effective bulk modulus K_e applies not only to isotropic composites but to composites with cubic symmetry as well.

We begin by considering a random dispersion of identical glass spheres in an epoxy matrix such that $G_2/G_1 = 22.5$, $K_2/K_1 = 10.0$ and $G_1/K_1 = 0.33$. The nonoverlapping spheres are in an equilibrium arrangement and thus the system has a maximum volume fraction at random-close packing when $\phi_2 \approx 0.644$ (Rintoul and Torquato, 1996). We are not aware of precise and comprehensive numerical calculations for the effective moduli of such a composite. However, we can compare our results to rigorous

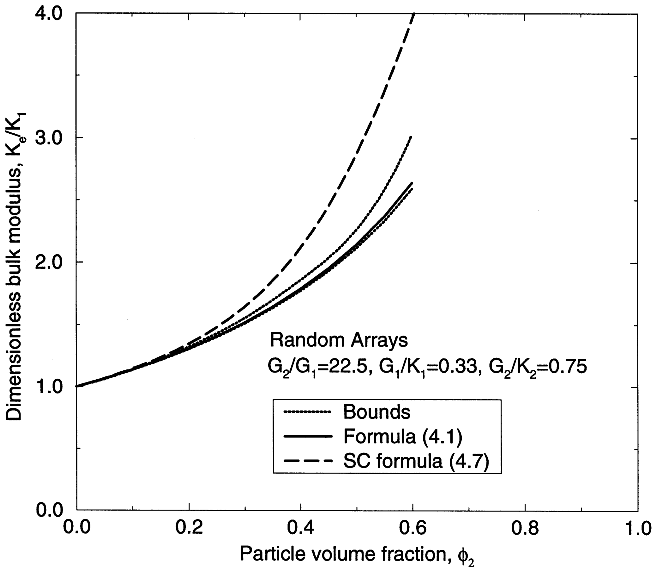

Fig. 7. Dimensionless effective bulk modulus K_e/K_1 vs particle volume fraction, ϕ_2 for random arrays of glass spheres in a epoxy matrix. Solid curve is our formula (4.1), the dashed curve is the SC formula (4.7), and the dotted curves are the Beran and Molyneux (1966) lower bound eqn (B.6) and Gibiansky and Torquato (1996) cross-property upper bound [which uses the conductivity data of Kim and Torquato (1991)]. The values of the parameters ζ_2 were taken from the table in Torquato (1991).

bounds since the parameters ζ_2 and η_2 have been computed for this random model for various values of ϕ_2 (Torquato and Lado, 1986; Sen *et al.*, 1989; Torquato, 1991). This is illustrated in Fig. 7 for the case of the effective bulk modulus. It is seen that the approximation (4.1) lies between the very narrow bound widths. The upper bound shown is obtained from the cross-property bounds of Gibiansky and Torquato (1996) utilizing effective conductivity data for this model in the case of superconducting spheres as found by Kim and Torquato (1991). The other bound is the three-point Beran–Molyneux lower bound given by eqns (B.6) or (B.7) with $d = 3$. The SC approximation (4.7) again is seen to overestimate the effective modulus for this glass–epoxy composite and indeed violates the upperbound.

In the ensuing discussion, we will examine special cases of relations (4.1) and (4.2) in which one or both of the phases have extreme values, solid containing cavities, cavities (bubbles) in incompressible matrices (liquids), rigid inclusions in compressible matrices, and incompressible particulate composites.

4.1. *Solid containing nonoverlapping cavities*

Consider a situation in which the dispersed phase consists of nonoverlapping cavities ($K_2 = G_2 = 0$). Then relations (4.1) and (4.2) for the effective bulk and shear moduli, respectively, reduce to

$$\frac{K_e}{K_1} = \frac{1-\phi_2-\dfrac{15K_1}{9K_1+8G_1}\phi_1\zeta_2}{1+\dfrac{3K_1}{4G_1}\phi_2-\dfrac{15K_1}{9K_1+8G_1}\phi_1\zeta_2}, \tag{4.9}$$

$$\frac{G_e}{G_1} = \frac{\left\{1-\phi_2-\dfrac{3K}{(9K_1+8G_1)}\phi_1\zeta_2-6\left[\dfrac{3K_1+G_1}{9K_1+8G_1}\right]^2\phi_1\eta_2 - \dfrac{30G_1(2K_1+3G_1)}{(9K_1+8G_1)^2}\phi_1\zeta_2\right\}}{\left\{1+\dfrac{6(K_1+2G_1)}{(9K_1+8G_1)}\phi_2-\dfrac{3K_1}{(9K_1+8G_1)}\phi_1\zeta_2-6\left[\dfrac{3K_1+G_1}{9K_1+8G_1}\right]^2\phi_1\eta_2 - \dfrac{30G_1(2K_1+3G_1)}{(9K_1+8G_1)^2}\phi_1\zeta_2\right\}}. \tag{4.10}$$

4.2. *Cavities* (*bubbles*) *in an incompressible matrix* (*liquid*)

Expressions for cavities in an incompressible matrix can be obtained from the cavity relations (4.9) and (4.10) by taking the limit that $K_1/G_1 \to \infty$. This process leads to

$$\frac{K_e}{G_1} = \frac{4\phi_1}{3\phi_2}\left(1-\frac{5}{3}\phi_1\zeta_2\right), \tag{4.11}$$

$$\frac{G_e}{G_1} = \frac{1-\phi_2-\dfrac{1}{3}\phi_1\zeta_2-\dfrac{2}{3}\phi_1\eta_2}{1-\dfrac{2}{3}\phi_2-\dfrac{1}{3}\phi_1\zeta_2-\dfrac{2}{3}\phi_1\eta_2}. \tag{4.12}$$

Since the effective Lamé constant $\lambda_e = K_e - 2G_e/3$, then we also have from relations (4.11) and (4.12) that

$$\frac{\lambda_e}{G_1} = \frac{4\phi_1}{3\phi_2}\left(1-\frac{5}{3}\phi_1\zeta_2\right) - \frac{2\phi_1}{3}\left[\frac{1-\dfrac{1}{3}\zeta_2-\dfrac{2}{3}\eta_2}{1-\dfrac{2}{3}\phi_2-\dfrac{1}{3}\phi_1\zeta_2-\dfrac{2}{3}\phi_1\eta_2}\right]. \tag{4.13}$$

Interestingly, a composite consisting of spherical cavities in an incompressible matrix of shear modulus G_1 is exactly equivalent to an incompressible liquid of shear viscosity μ containing air bubbles. The analogs of the shear modulus, bulk modulus and Lamé constant in the liquid problem are the shear viscosity, bulk viscosity and expansion viscosity, respectively. Let us denote the effective expansion viscosity by ξ_e. Thus, eqn (4.13) interpreted in this fashion is an expression for the dimensionless effective bulk viscosity λ_e/G_1. Neglecting interactions between bubbles ($\phi_2 \ll 1$), Taylor (1954) found that the effective expansion viscosity is given by

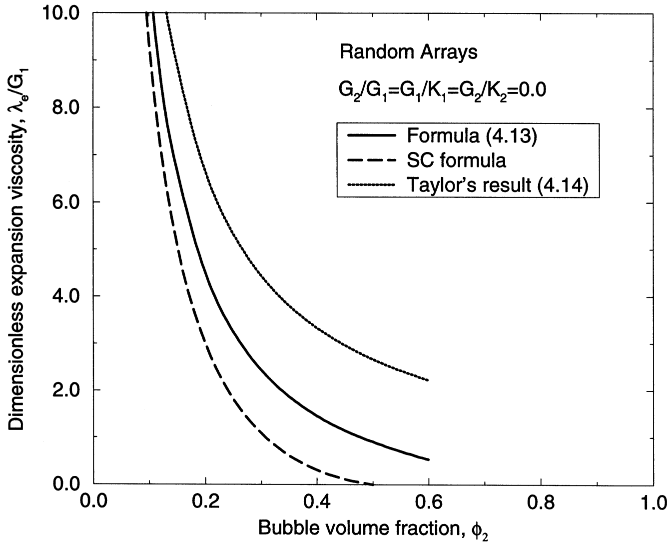

Fig. 8. Dimensionless effective expansion viscosity λ_e/G_1 vs bubble volume fraction ϕ_2 for randomly arranged spherical bubbles in an incompressible matrix. Solid curve is our formula (4.13), the dashed curve is the SC result, and the dotted curve is Taylor's result (4.14). The parameters ζ_2 and η_2 were taken from the tables in Torquato (1991).

$$\frac{\lambda_e}{G_1} = \frac{4}{3\phi_2}. \tag{4.14}$$

Therefore, expression (4.13) corrects Taylor's result by accounting for interactions between the bubbles at non-dilute concentrations. From Fig. 8, it is seen that Taylor's result eqn (4.14) overestimates the effective bulk viscosity at non-dilute concentrations. On the other hand, the SC approximation generally underestimates the effective bulk viscosity, especially at high bubble concentrations where it predicts a spurious percolation threshold of $\phi_2 = 0.5$.

4.3. *Rigid inclusions in compressible matrices*

Consider the instance of a dispersion consisting of rigid inclusions, i.e., $G_2/G_1 = K_2/K_1 = \infty$, in a compressible matrix. For such a composite, $\kappa = \mu = 1$ and hence the effective bulk and shear moduli are given respectively by

$$\frac{K_e}{K_1} = \frac{1 + \dfrac{4G_1}{3K_1}\phi_2 - \dfrac{10G_1}{3(K_1+2G_1)}\phi_1\zeta_2}{1 - \phi_2 - \dfrac{10G_1}{3(K_1+2G_1)}\phi_1\zeta_2}, \tag{4.15}$$

$$\frac{G_e}{G_1} = \frac{1+\dfrac{9K_1+8G_1}{6(K_1+2G_1)}\phi_2-\dfrac{2G_1}{3(K_1+2G_1)}\phi_1\zeta_2-\dfrac{1}{6}\left\{\left[\dfrac{3K_1+G_1}{K_1+2G_1}\right]^2\phi_1\eta_2+5G_1\left[\dfrac{2K_1+3G_1}{(K_1+2G_1)^2}\right]\phi_1\zeta_2\right\}}{1-\phi_2-\dfrac{2G_1}{3(K_1+2G_1)}\phi_1\zeta_2-\dfrac{1}{6}\left\{\left[\dfrac{3K_1+G_1}{K_1+2G_1}\right]^2\phi_1\eta_2+5G_1\left[\dfrac{2K_1+3G_1}{(K_1+2G_1)^2}\right]\phi_1\zeta_2\right\}}. \tag{4.16}$$

Cubic-symmetric composites, such as cubic lattices of spheres, serve as useful benchmark models since the special symmetry enables one to solve for the effective moduli essentially exactly. Thus, using the tabulation of ζ_2 for cubic lattices obtained by McPhedran and Milton (1981), we will compare the predictions of (4.1) to the numerical results of Nunan and Keller (1984) for the effective bulk moduli of rigid cubic arrays of spheres in compressible matrices. The effective stiffness tensor **C** of such a composite is expressible in component form as

$$C_{ijkl} = (\lambda_1+G_1\gamma)\delta_{ij}\delta_{kl}+G_1(1+\beta)(\delta_{ik}\delta_{jl}+\delta_{il}\delta_{jk})+2G_1(\alpha-\beta)\delta_{ijkl}. \tag{4.17}$$

Here $\lambda_1 = K_1-2G_1/3$ is the Lamé constant, δ_{ij} is the Kronecker delta, δ_{ijkl} is one if all the subscripts are equal and zero otherwise, and α, β, and γ are functions of the inclusion volume fraction tabulated by Nunan and Keller (1984). (Note that the factor 2 in the last term of (4.17) is missing in the corresponding formula in Nunan and Keller, 1984.) As follows from eqn (4.17), the effective bulk modulus of such a composite (in terms of the functions α and γ) is given by

$$K_e = K_1+G_1(\gamma+2\alpha/3). \tag{4.18}$$

Figure 9 compares relation (4.15) and the SC approximation (4.7) for $d = 3$ to the numerical data of Nunan and Keller for face-centered cubic arrays of rigid spheres in a compressible matrix in which $G_2/G_1 = K_2/K_1 = \infty$ and $G_1/K_1 = 0.46$. The prediction of eqn (4.15) is remarkably accurate, whereas the SC approximation (4.7) begins to diverge increasingly from the data for values of the fiber volume fraction ϕ_2 larger than 0.3. The Beran–Molyneux three-point lower bound is virtually indistinguishable from the prediction (4.15); the associated upper bound diverges to infinity in this instance.

Figure 10 compares the predictions of relation (4.15) and the SC approximation (4.7) for $d = 3$ to rigorous bounds for random arrays of rigid spheres in a compressible matrix in which $G_2/G_1 = K_2/K_1 = \infty$ and $G_1/K_1 = 0.46$. The upper bound shown is obtained from the cross-property bounds of Gibiansky and Torquato (1996) utilizing effective conductivity data for this model in the case of superconducting spheres as found by Kim and Torquato (1991). The other bound is the three-point Beran–Molyneux lower bound given in eqn (B.6). Whereas the expression (4.15) lies within the narrow bounds, the SC approximation again is seen to overestimate significantly the effective modulus for non-dilute concentrations.

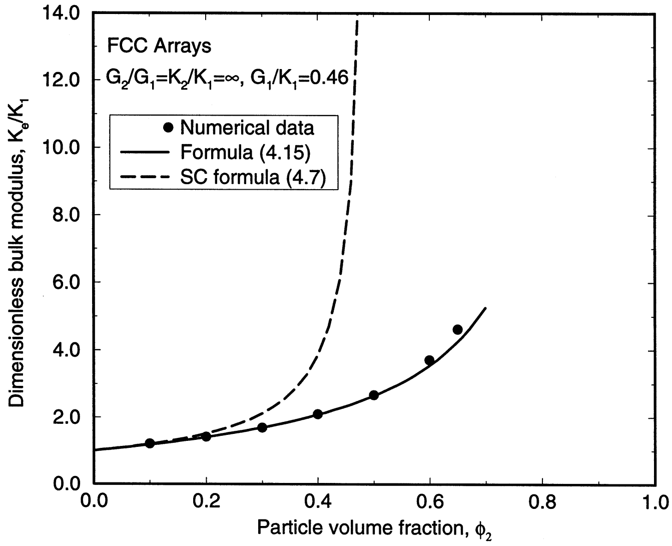

Fig. 9. Dimensionless effective bulk modulus K_e/K_1 vs particle volume fraction ϕ_2 for face-centered cubic arrays of rigid spheres in a compressible matrix. Filled circles are numerical data of Nunan and Keller (1984), solid curve is our formula (4.15), and the dashed curve is the SC formula (4.7). The values of the parameters ζ_2 were taken from McPhedran and Milton (1981).

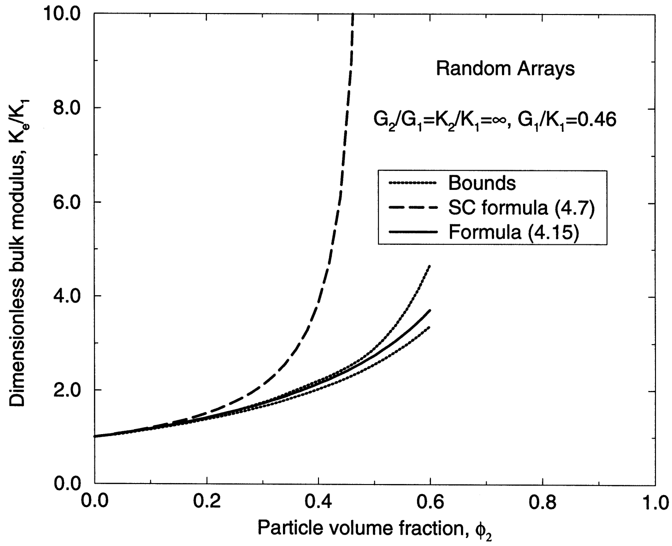

Fig. 10. Dimensionless effective bulk modulus K_e/K_1 vs particle volume fraction, ϕ_2 for random arrays of rigid spheres in a compressible matrix. Solid curve is our formula (4.15), the dashed curve is the SC formula (4.7), and the dotted curves are the Beran–Molyneux lower bound eqn (B.6) and Gibiansky and Torquato (1996) cross-property upper bound [which uses the conductivity data of Kim and Torquato (1991)]. The values of the parameters ζ_2 were taken from the table in Torquato (1991).

4.4. *Incompressible isotropic dispersions*

Let us consider the case of an incompressible isotropic dispersion, i.e., $K_1/G_1 \to \infty$ and $K_2/G_2 \to \infty$. In this limit, relation (4.2) for the effective shear modulus reduces to

$$\frac{G_e}{G_1} = \frac{1 + \frac{3}{2}\mu\phi_2 - \frac{3}{2}\mu^2\phi_1\eta_2}{1 - \mu\phi_2 - \frac{3}{2}\mu^2\phi_1\eta_2}, \tag{4.19}$$

where

$$\mu = \frac{G_2 - G_1}{G_2 + \frac{3}{2}G_1}. \tag{4.20}$$

If we now allow the included phase of this incompressible composite to be rigid ($G_2/G_1 \to \infty$), then relation (4.19) yields the expression

$$\frac{G_e}{G_1} = \frac{1 + \frac{3}{2}\phi_2 - \frac{3}{2}\phi_1\eta_2}{1 - \phi_2 - \frac{3}{2}\phi_1\eta_2}. \tag{4.21}$$

As noted earlier, this is the most difficult case to treat theoretically, especially at high particle concentrations. Note that in the limit of an incompressible matrix ($K_1/G_1 = \infty$), formula (4.16) becomes identical to relation (4.21). Interestingly, the determination of the effective shear modulus of such a dispersion is exactly equivalent to finding the effective viscosity of the dispersion in the infinite-frequency limit.

In Fig. 11 we plot the formula (4.19), the three-dimensional SC approximation (4.8) and the Milton–Phan-Thien (1982) three-point lower bound for the case of an incompressible composite containing a random array of rigid spheres ($G_2/G_1 = K_2/K_1 = \infty$, $G_1/K_1 = 0$) in an equilibrium arrangement. The SC approximation diverges appreciably from the formula (4.19) and indeed predicts a low percolation threshold of $\phi_2 = 0.4$. In actuality, an equilibrium dispersion of mutually impenetrable spheres is expected to percolate at the random close-packing value of $\phi_2 \approx 0.644$ (Rintoul and Torquato, 1996). Based on the prediction of our two-dimensional counterpart for such a special incompressible composite (see Fig. 6), we expect that formula (4.19) will estimate G_e well for low to moderately high sphere volume fractions but will significantly underestimate it near the random close-packing fraction of 0.644, even though it is appreciably above the Milton–Phan-Thien (1982) three-point lower bound in this high volume-fraction range (see discussion in Section 5).

5. DISCUSSION

Approximate relations for the effective bulk and shear moduli of two-phase, d-dimensional isotropic dispersions are obtained by truncating the exact series expan-

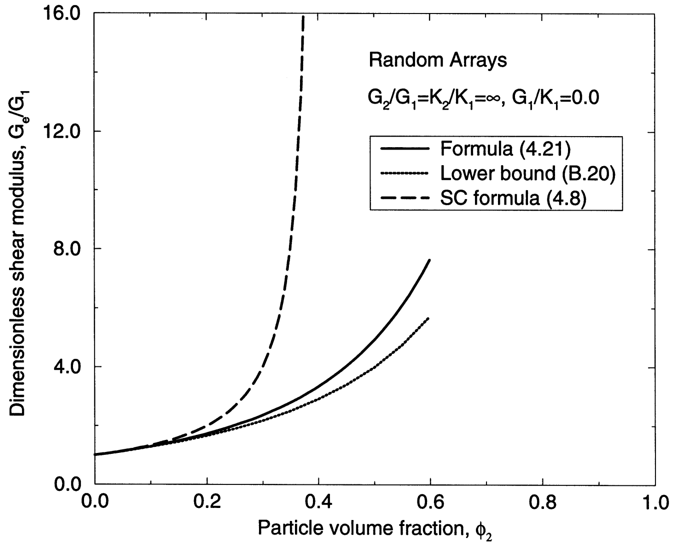

Fig. 11. Dimensionless effective shear modulus G_e/G_1 vs particle volume fraction ϕ_2 for random arrays of rigid spheres in an incompressible matrix. Solid curve is our formula (4.21), dashed curve is the SC formula (4.8), and dotted curve is the Milton and Phan-Thien (1982) three-point lower bound given in eqn (B.20). The values of the parameters η_2 were taken from the table in Torquato (1991).

sions for these effective parameters (obtained in part I) after third-order terms. The approximate expressions (2.8) and (2.12) incorporate volume fraction information as well as three-point information via the parameters ζ_2 and η_2. Since the exact series expressions of part I perturb about the optimal Hashin–Shtrikman structures, it was claimed that the aforementioned truncated expression should provide accurate estimates of the effective moduli for a wide range of phase moduli and volume fractions for dispersions in which the inclusions are prevented from forming large clusters. We tested the predictions of the third-order approximations for a variety of two- and three-dimensional dispersions. Generally, for the cases considered, the third-order approximations were found to be in very good agreement with benchmark data, always lay within rigorous three-point or cross-property bounds, and were superior to popular self-consistent approximations. When the dispersed phase was stiffer than the matrix, the third-order formulas mimicked (to an excellent approximation) the behavior of higher-order lower bounds on the effective moduli. On the other hand, when the dispersed phase was more compliant than the matrix, the third-order formulas mimicked (to an excellent approximation) the behavior of higher-order upper bounds on the effective moduli.

The self-consistent formulas in many cases not only provided poor approximations of the effective moduli of dispersions but violated rigorous bounds that incorporate higher-order microstructural information. Despite this fact, SC approximations continue to be applied to estimate the effective moduli of dispersions just because they are simple to use. Why do self-consistent formulas provide poor estimates of the effective moduli of dispersions, especially at significant phase contrast? The reason is clear when one understands the class of structures for which self-consistent formulas are indeed exact. Milton (1984) showed that this class, roughly speaking, consists of granular aggregates such that phase 1 grains and phase 2 grains of comparable size are well separated with self-similarity on many length scales. This class of hierarchical composites possesses a special topological symmetry, i.e., the morphology of phase 1 at volume fraction ϕ_1 is identical (statistically) to the morphology of phase 2 when its volume fraction equals ϕ_1. (Clearly, dispersions do not possess this topological symmetry.) For low contrast and volume fractions, the self-consistent structures will have effective moduli that are close in value to single-scale dispersions; but as the contrast and volume fraction are made large, it is clear that they will have moduli that differ significantly from single-scale dispersions. Accordingly, we want to emphasize that we are not denigrating self-consistent models, but rather are pointing out that they are not appropriate property estimates for dispersions. Indeed, self-consistent formulas will be superior to our third-order approximations for media in which the phases are topologically equivalent (e.g., some biconnected two-phase composites).

Finally, we observe that near the percolation threshold of the dispersed phase (i.e., large ϕ_2) of composites consisting rigid inclusions ($G_2/G_1 \to \infty$) in incompressible matrices, the third-order relations (3.17) and (4.21) do not provide sharp estimates the effective shear modulus G_e. The fact that the third-order relations underestimate G_e in this most difficult regime is not surprising given that the actual dispersions begin to deviate appreciably from structures that perturb about the optimal Hashin–Shtrikman dispersions.

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APPENDIX A: ζ_2 AND η_2 FOR STATISTICALLY ISOTROPIC DISPERSIONS

Consider a statistically isotropic, *d*-dimensional array of nonoverlapping particles of a well-defined shape with a particle indicator function

$$m(\mathbf{x}) = \begin{cases} 1, & \mathbf{x} \text{ in particle region,} \\ 0, & \text{otherwise,} \end{cases} \tag{A.1}$$

where $\mathbf{x}$ is measured from the centroid of the particle. The particles generally may possess a size distribution. Denote by $\mathbf{r}_i$ the center-of-mass and orientation of the *i*th particle. Let the region of space occupied by the particles be phase 2 and define $S_3(\mathbf{x}_1, \mathbf{x}_2, \mathbf{x}_3) \equiv S_3^{(2)}(\mathbf{x}_1, \mathbf{x}_2, \mathbf{x}_3)$ to be the three-point probability function of phase 2, i.e., the probability of simultaneously finding three points in phase 2 at positions $\mathbf{x}_1$, $\mathbf{x}_3$, and $\mathbf{x}_3$, respectively. It is clear that the probability of finding three points in the particle phase can be written as the sum of three different probabilities, i.e.,

$$S_3(r, s, t) = s^{(1)}(r, s, t) + s^{(2)}(r, s, t) + s^{(3)}(r, s, t), \tag{A.2}$$

where $s^{(1)}$ is the probability that all three points fall in one particle, $s^{(2)}$ is the probability that one of the three points falls in one particle and the remaining two points fall in another particle, and $s^{(3)}$ is the probability that each point falls in three different particles. Moreover, because of statistical isotropy, S_3 depends on the distances $r \equiv |\mathbf{x}_2 - \mathbf{x}_1|$, $s \equiv |\mathbf{x}_3 - \mathbf{x}_1|$, and $t \equiv |\mathbf{x}_3 - \mathbf{x}_2|$.

The functions $s^{(i)}$ can be related to multidimensional integrals over the two- and three-particle functions g_2 and g_3. (Recall that the *n*-particle distribution function $g_n(\mathbf{r}_1, \ldots, \mathbf{r}_n)$ characterizes the probability density of finding *n* particles with configuration $\mathbf{r}_1, \ldots, \mathbf{r}_n$). For example, for identical, nonoverlapping spheres of radius *a* and number density ρ, Torquato and Stell (1985) found that

$$s^{(1)}(r, s, t) = \rho \int m(x_{14}) m(x_{24}) m(x_{34})\, \mathrm{d}\mathbf{r}_4, \tag{A.3}$$

$$s^{(2)}(r, s, t) = \rho^2 \int m(x_{14}) m(x_{24}) m(x_{35}) g_2(r_{45})\, \mathrm{d}\mathbf{r}_4\, \mathrm{d}\mathbf{r}_5 + \rho^2 \int m(x_{14}) m(x_{34}) m(x_{25}) g_2(r_{45})\, \mathrm{d}\mathbf{r}_4\, \mathrm{d}\mathbf{r}_5 + \rho^2 \int m(x_{15}) m(x_{24}) m(x_{34}) g_2(r_{45})\, \mathrm{d}\mathbf{r}_4\, \mathrm{d}\mathbf{r}_5,$$

$$s^{(3)}(r, s, t) = \rho^3 \int m(x_{14}) m(x_{25}) m(x_{36}) g_3(r_{45}, r_{46}, r_{56})\, \mathrm{d}\mathbf{r}_4\, \mathrm{d}\mathbf{r}_5, \quad \text{(A.4)} \tag{A.5}$$

where $x_{ij} = |\mathbf{x}_i - \mathbf{r}_j|$, $r_{ij} = |\mathbf{r}_i - \mathbf{r}_j|$ and

$$m(r) = \begin{cases} 1, & r \leqslant a, \\ 0, & r > a. \end{cases} \tag{A.6}$$

Substitution of the representation (A.2) for $S_3(r, s, t)$ into the integrals (2.7) and (2.11) that define ζ_2 and η_2, respectively, results in complex multidimensional integrals over g_2 and g_3. However, these can be greatly simplified by expanding orientation-dependent terms of the integrand in orthogonal polynomials, yielding the expressions

$$\zeta_2 = \int_0^\infty \mathrm{d}x g_2(x) f_\zeta(x) + \int_0^\infty \mathrm{d}z \int_0^\infty \mathrm{d}y \int_{|\mathbf{z}-\mathbf{y}|}^{z+y} \mathrm{d}x g_3(x, y, z) h_\zeta(x, y, z), \tag{A.7}$$

$$\eta_2 = \int_0^\infty \mathrm{d}x g_2(x) f_\eta(x) + \int_0^\infty \mathrm{d}z \int_0^\infty \mathrm{d}y \int_{|\mathbf{z}-\mathbf{y}|}^{z+y} \mathrm{d}x g_3(x, y, z) h_\eta(x, y, z). \tag{A.8}$$

Here $f_\zeta(r)$, $f_\eta(r)$, $h_\zeta(r, s, t)$, and $h_\eta(r, s, t)$ are functions that depend on the space dimension d; they were explicitly given for $d = 2$ in the case of identical circular disks by Torquato and Lado (1988, 1991) and for $d = 3$ in the case of identical spheres by Lado and Torquato (1986) and Sen *et al.* (1989). With straightforward modification, similar results can be obtained for nonoverlapping particles of non-spherical shape.

The analysis above applies to nonoverlapping particles, i.e., a hard-particle potential. For more general interparticle potentials (e.g., soft repulsive potentials), S_3 and, hence, the three-point parameters ζ_2 and η_2 will generally depend on the infinite set $g_1, g_2, \ldots, g_m$ $(m \to \infty)$.

APPENDIX B: 3-POINT ESTIMATES AND THE Y-TRANSFORMATION

The purpose of this appendix is twofold. First, it summarizes existing three-point bounds on the effective plastic moduli, focusing primarily on the most restrictive bounds. These bounds are expressed in a compact form using the so-called Y-transformation (see Milton, 1991; Cherkaev and Gibiansky, 1992) as first described by Gibiansky and Torquato (1995). Second, we express our three-point approximations using the Y-transformation. This makes it much simpler to find the range of the geometrical parameters ζ_2 and η_2 for which the approximations always lie within the most restrictive bounds. We first define the Y-transformation and then use it to express the bounds and approximations in a compact form.

All of the existing three-point bounds as well as the three-point approximations (2.8) and (2.12) can be written in terms of the function F that depends on five variables defined as follows:

$$F(a_1, a_2, \phi_1, \phi_2, y) = \phi_1 a_1 + \phi_2 a_2 - \frac{\phi_1 \phi_2 (a_1 - a_2)^2}{\phi_2 a_1 + \phi_1 a_2 + y}, \tag{B.1}$$

where a represents any phase property. Let us now introduce the Y-transformation that is an inverse to the function F as a function of its fifth variable y, i.e.,

$$y(a_1, a_2, \phi_1, \phi_2, a_e) = -\phi_2 a_1 - \phi_1 a_2 + \frac{\phi_1 \phi_2 (a_1 - a_2)^2}{\phi_1 a_1 + \phi_2 a_2 - a_e}. \tag{B.2}$$

For brevity we sometimes omit the first four arguments of this function and write it as $y_a(a_e) = y(a_1, a_2, \phi_1, \phi_2, a_e)$. One can easily check that the bounds

$$F(a_1, a_2, \phi_1, \phi_2, y_1) \leqslant a_e \leqslant F(a_1, a_2, \phi_1, \phi_2, y_2) \tag{B.3}$$

are equivalent to the following bounds in terms of the Y-transformations:

$$y_1 \leqslant y_a(a_e) \leqslant y_2. \tag{B.4}$$

Moreover, we introduce the following shorthand notation:

$$\langle a \rangle = \phi_1 a_1 + \phi_2 a_2, \quad \langle a \rangle_\zeta = \zeta_1 a_1 + \zeta_2 a_2, \quad \langle a \rangle_\eta = \eta_1 a_1 + \eta_2 a_2. \tag{B.5}$$

Three-point bounds on the effective bulk modulus K_e were derived by Beran and Molyneux (1966) for $d = 3$ and by Silnutzer for $d = 2$. These bounds were subsequently simplified by Milton (1981, 1982). The d-dimensional Beran-type bounds on the effective bulk modulus K_e that incorporate volume fractions ϕ_1, ϕ_2 and geometrical parameters ζ_1, ζ_2 can be written (Gibiansky and Torquato, 1995) in the form

$$F\left(K_1, K_2, \phi_1, \phi_2, \frac{2(d-1)\langle G^{-1}\rangle_\zeta^{-1}}{d}\right) \leqslant K_e \leqslant F\left(K_1, K_2, \phi_1, \phi_2, \frac{2(d-1)\langle G\rangle_\zeta}{d}\right). \quad \text{(B.6)}$$

By using the Y-transformation we can rewrite eqn (B.6) as follows:

$$\frac{2(d-1)\langle G^{-1}\rangle_\zeta^{-1}}{d} \leqslant y_K(K_e) \leqslant \frac{2(d-1)\langle G\rangle_\zeta}{d}. \quad \text{(B.7)}$$

The inequalities (B.7) can be rewritten as

$$F\left(\frac{2(d-1)G_1}{d}, \quad \frac{2(d-1)G_2}{d}, \quad \zeta_1, \zeta_2, 0\right) \leqslant y_K(K_e) \leqslant F\left(\frac{2(d-1)G_1}{d}, \quad \frac{2(d-1)G_2}{d}, \quad \zeta_1, \zeta_2, \infty\right) \quad \text{(B.8)}$$

or, again by using the Y-transformation, by

$$0 \leqslant z_K(K_e) \leqslant \infty, \quad \text{(B.9)}$$

where

$$z_K(K_e) \equiv y\left(\frac{2(d-1)G_1}{d}, \quad \frac{2(d-1)G_2}{d}, \quad \zeta_1, \zeta_2, y_K(K_e)\right). \quad \text{(B.10)}$$

In the two-dimensional case, Gibianksy and Torquato (1995) obtained the following improved upper bound:

$$y_K(K_e) \leqslant F(G_1, G_2, \zeta_1, \zeta_2, K_{\max}), \quad (d = 2), \quad \text{(B.11)}$$

or, equivalently,

$$z_K(K_e) \leqslant K_{\max}, \quad (d = 2), \quad \text{(B.12)}$$

where $z_K(K_e) = y(G_1, G_2, \zeta_1, \zeta_2, y(K_e))$ and $K_{\max}$ is the maximal phase bulk modulus. Thus, the most restrictive bounds in two dimensions are

$$0 \leqslant z_K(K_e) \leqslant K_{\max}. \quad \text{(B.13)}$$

For the two-dimensional effective shear modulus, Gibiansky and Torquato (1995) showed that the most restrictive three-point bounds can be presented in the form

$$[2\langle K^{-1}\rangle_\zeta + \langle G^{-1}\rangle_\eta]^{-1} \leqslant y_G(G_e) \leqslant A^{-1}, \quad (d = 2), \quad \text{(B.14)}$$

where

$$A = \begin{cases} y_{1*}, & \text{if} \quad t \in [-K_{\max}^{-1}, G_{\max}^{-1}], \\ y_{2*}, & \text{if} \quad t \leqslant -K_{\max}^{-1}, \\ y_{3*}, & \text{if} \quad t \geqslant G_{\max}^{-1} \end{cases} . \quad \text{(B.15)}$$

$$y_{1*} = \langle 2K^{-1}\rangle_\zeta + \langle G^{-1}\rangle_\eta - \frac{[\sqrt{2\zeta_1\zeta_2(K_1^{-1} - K_2^{-1})^2} + \sqrt{\eta_1\eta_2(G_1^{-1} - G_2^{-1})^2}]^2}{\eta_1 G_2^{-1} + \eta_2 G_1^{-1} + 2\zeta_1 K_2^{-1} + 2\zeta_2 K_1^{-1}} \quad \text{(B.16)}$$

$$y_{2*} = \left\langle \frac{1}{G^{-1}+K_{\max}^{-1}} \right\rangle_\eta^{-1} + \frac{1}{K_{\max}}, \quad y_{3*} = 2\left\langle \frac{1}{K^{-1}+G_{\max}^{-1}} \right\rangle_\eta^{-1} - \frac{1}{G_{\max}}. \tag{B.17}$$

$$t = \frac{\sqrt{2\zeta_1\zeta_2(K_1^{-1}-K_2^{-1})^2}(\eta_1 G_2^{-1}+\eta_2 G_1^{-1}) - \sqrt{\eta_1\eta_2(G_1^{-1}-G_2^{-1})^2}(\zeta_1 K_2^{-1}+\zeta_2 K_1^{-1})}{\sqrt{2\zeta_1\zeta_2(K_1^{-1}-K_2^{-1})^2}+\sqrt{\eta_1\eta_2(G_1^{-1}-G_2^{-1})^2}}, \tag{B.18}$$

where $G_{\max}$ is the maximal phase shear modulus. The lower bound in eqn (B.14) is due to Silnutzer (1972) and the upper bound in eqn (B.14) is due to Gibiansky and Torquato (1995). For the special case of an incompressible composite ($K_1/G_1 = K_2/G_2 = \infty$), eqn (B.14) can be recast as

$$0 \leqslant z_G(G_e) \leqslant \infty, \tag{B.19}$$

where $z_G(G_e) = F(G_1, G_2, \eta_1, \eta_2, y_G(G_e))$.

Three-point bounds on the effective shear modulus of a three-dimensional composite were obtained by McCoy (1970) and improved upon by Milton and Phan-Thien (1982). The latter bounds are expressible as

$$\Xi \leqslant y_G(G_e) \leqslant \Theta, \quad (d=3), \tag{B.20}$$

where

$$\Xi = \frac{\left\langle \frac{128}{K} + \frac{99}{G} \right\rangle_\zeta + 45\left\langle \frac{1}{G} \right\rangle_\eta}{30\left\langle \frac{1}{G} \right\rangle_\zeta \left\langle \frac{6}{K} - \frac{1}{G} \right\rangle_\zeta + 6\left\langle \frac{1}{G} \right\rangle_\eta \left\langle \frac{2}{K} + \frac{21}{G} \right\rangle_\zeta}, \tag{B.21}$$

$$\Theta = \frac{3\langle G\rangle_\eta \langle 6K+7G\rangle_\zeta - 5\langle G\rangle_\zeta^2}{6\langle 2K-G\rangle_\zeta + 30\langle G\rangle_\eta}. \tag{B.22}$$

Whereas ζ_2 can lie anywhere in the interval [0, 1], Milton and Phan-Thien showed that for $d = 3$, η_2 lies in the smaller interval $[5\zeta_2/21, (16+5\zeta_2)/21]$.

In order to compare our approximations to the aforementioned three-point bounds, we now rewrite them using the Y-transformation. The d-dimensional effective bulk modulus expression (2.8) can be rewritten as

$$\tilde{z}_K(K_e) = \frac{-(d-1)G_1\{[4+2d-2d^2]G_1 + d[(d+2)\zeta_2 - d]K_1\}}{d\{[2d-(d+2)(d-1)\zeta_2]G_1 + dK_1\}}, \tag{B.23}$$

where $\tilde{z}_K(K_e)$ is just the corresponding z-function defined by eqn (B.10). The expression (2.12) for the effective shear modulus is too complicated to be put in such a form for general values of the phase moduli. However, in the special case of an incompressible composite ($K_1/G_1 = K_2/G_2 = \infty$), it can be recast as

$$\tilde{y}_G(G_e) = F\left(\frac{dG_1}{2}, \quad \frac{dG_2}{2}, \quad \eta_1, \eta_2, \tilde{z}_G(G_e)\right), \tag{B.24}$$

where

$$\tilde{z}_G(G_e) = \frac{dG_1[d-2\eta_2]}{2[2-d\eta_2]}. \tag{B.25}$$

Let us first consider the effective bulk modulus approximation (B.23). For relation (B.23) to lie within the bounds, it must satisfy the inequalities (B.13) for $d = 2$ and inequalities (B.9) for

$d \geqslant 3$. In the former case, one can easily check that this implies that the parameter ζ_2 must be bounded from above according to the inequalities

$$\zeta_2 \leqslant 0.5, \quad (d=2). \tag{B.26}$$

In the instance of three dimensions, a similar analysis yields

$$\zeta_2 \leqslant 0.6 + \frac{8}{15}\frac{G_1}{K_1}, \quad \text{for} \quad 0 \leqslant G_1/K_1 \leqslant 0.75, \quad (d=3), \tag{B.27}$$

$$\zeta_2 \leqslant 1, \quad \text{for} \quad G_1/K_1 > 0.75, \quad (d=3). \tag{B.28}$$

Thus, for $G_1/K_1 > 0.75$, there is no additional restriction and ζ_2. The most restrictive case is when phase 1 is compressible ($G_1/K_1 = 0$), where it is seen that $\zeta_2 \leqslant 0.6$ and hence we conclude that the three-dimensional approximation for K_e is less constrained than its two-dimensional counterpart for which $\zeta_2 \leqslant 0.5$.

We see that in the case of two-dimensional, incompressible composites, the approximation (B.25) always lies within the bounds eqn (B.19) and hence any realizable value of η_2 will yield an estimate of G_e that does not violate the three-point bounds for $d = 2$. For three-dimensional, incompressible composites, the approximation (B.24) always satisfies the upper bound of eqn (B.20). In order for eqn (B.24) to satisfy the lower bound of eqn (B.20) in this incompressible limit, it is required that

$$\frac{1+\eta_2}{2-3\eta_2} \geqslant \frac{1-\frac{11}{16}\zeta_2-\frac{5}{16}\eta_2}{2(1-\zeta_2)(1+\frac{5}{16}\zeta_2-\frac{21}{16}\eta_2)}, \quad \left(d=3, \quad \frac{K_1}{G_1}=\frac{K_2}{G_2}=\infty\right). \tag{B.29}$$