Improved bounds on the effective elastic and transport properties of fiber-reinforced composites: Effect of polydispersivity in fiber radius

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Improved rigorous bounds on the effective elastic and transport properties of a transversely isotropic fiber-reinforced material composed of oriented, infinitely long, multisized circular cylinders distributed throughout a matrix are computed. Specifically, we evaluate such bounds on the effective axial shear modulus (which includes, by mathematical analogy, the transverse conductivity), effective transverse bulk modulus, and the effective transverse shear modulus. These are generally demonstrated to provide significant improvement over the Hill-Hashin bounds which incorporate only volume-fraction information. Although the upper bounds diverge from the lower bounds when the cylinders are much stiffer than the matrix, the improved lower bounds still yield relatively accurate estimates of the effective properties.

Generally, increasing the degree of polydispersivity in cylinder size increases the effective transverse conductivity (or axial shear modulus) and effective transverse bulk modulus, and decreases (slightly) the effective transverse shear modulus for cases in which the fibers are more conducting or stiffer than the matrix.

I. INTRODUCTION

Rigorous upper and lower bounds on the effective properties of composite materials are useful because they enable one to test the merits of a theory or computer experiment; as successively more microstructural information is incorporated, the bounds become progressively narrower; and one of the bounds can typically provide a relatively accurate estimate of the property, even when the reciprocal bound diverges from it.

For transversely isotropic two-phase composites, Hill* and Hashin4,5 obtained the best possible bounds on the effective axial shear modulus \( \mu_e \), (or, equivalently, effective transverse conductivity \( \sigma_e \),) effective transverse bulk modulus \( k_e \), and effective transverse shear modulus \( G_e \), given the phase properties and volume fractions \( \phi_i \). Although bounds which improve upon these second-order bounds have been available for some time now, their application has been, until recently, very slow because of the difficulty involved in ascertaining the statistical correlation functions involved.

In the last few years, improved bounds on the aforementioned effective properties6-8 have been evaluated for nontrivial model microstructures consisting of random arrays of infinitely long, oriented cylinders in a matrix6-14 by utilizing the analytical representations of the correlation functions derived by Torquato and Stell.15,16 In particular, Torquato and Beasley9,10 and Joslin and Stell11 independently computed improved bounds due to Silnutzer6 and to Milton7,8 for identical overlapping (i.e., randomly centered) cylinders. Joslin and Stell12 evaluated the same bounds for polydispersed overlapping cylinders and showed that the bounds were insensitive to polydispersivity for this microgeometry. Torquato and Lado,13,14 and Smith and Torquato17 calculated these improved bounds for identical nonoverlapping or impenetrable cylinders. This is a useful model of unidirectional fiber composites. For impenetrable cylinders, the effect of polydispersivity in cylinder size on these effective properties has heretofore not been rigorously studied. Unlike the case of overlapping cylinders, the effect of polydispersivity in cylinder size for impenetrable cylinders is generally expected to be significant.

The purpose of this paper is to compute improved bounds on \( \mu_e \) (or \( \sigma_e \),) \( k_e \), and \( G_e \) for composites consisting of random distributions of infinitely long, oriented, impenetrable cylinders with a polydispersivity in size. Such an investigation will enable us to study the effect of polydispersivity on the elastic and transport properties of such unidirectional composites.

II. IMPROVED BOUNDS ON THE EFFECTIVE ELASTIC MODULI

For a two-phase, transversely isotropic fiber-reinforced material, Hill3 and Hashin4,5 derived the best possible bounds on \( \mu_e \), \( k_e \), and \( G_e \), given only the phase volume fractions \( \phi_i \) and \( \phi_2 \), bulk moduli \( K_1 \) and \( K_2 \), and shear moduli...
These are referred to as second-order bounds since they are exact through second order in the difference in phase properties. Silnutzer derived improved third-order bounds on \( p_{C_1}, k_1, \) and \( G_1 \) that depend upon two integrals over certain three-point correlation functions. The Silnutzer bounds were simplified by Milton and are, respectively, given for the axial shear modulus, transverse bulk modulus, and transverse shear modulus by

\[
\mu_1^{(3)} \leq \mu \leq \mu_0^{(3)},
\]

where

\[
\mu_1^{(3)} = \left( \frac{\phi_1 \phi_2 (G_2 - G_1)^2}{\langle G \rangle} \right),
\]

\[
\mu_0^{(3)} = \left( \frac{\phi_1 \phi_2 (1/G_2 - 1/\langle G \rangle)^2}{\langle 1/G \rangle} \right)^{-1},
\]

\[
k^{(3)}_1 \leq k \leq k^{(3)}_0,
\]

and

\[
G^{(3)}_1 \leq G \leq G^{(3)}_0,
\]

where

\[
G^{(3)}_1 = \left( \langle G \rangle - \frac{\phi_1 \phi_2 (G_2 - G_1)^2}{\langle G \rangle} \right),
\]

\[
G^{(3)}_0 = \left( \frac{\phi_1 \phi_2 (1/G_2 - 1/\langle G \rangle)^2}{\langle 1/G \rangle} \right)^{-1},
\]

\[
\Theta = 2 \left( \frac{k}{\langle G \rangle} \right)^2 + \left( \frac{k}{\langle G \rangle} \right)^4,
\]

\[
\Xi = 2 \left( \frac{k}{\langle G \rangle} \right)^2 + \left( \frac{G}{\langle G \rangle} \right)^2.
\]

Here we define \( \langle b \rangle = b_1 + b_2, \langle \hat{b} \rangle = b_1 \hat{b}_1 + b_2 \hat{b}_2, \langle b \rangle \hat{b} = b_1 \hat{b}_1 + b_2 \hat{b}_2, \) and \( \langle b \rangle_2 = b_1 \hat{b}_1 + b_2 \hat{b}_2. \) The quantity \( k \) is the transverse bulk modulus of phase 1 for transverse compression without axial extension and may be expressed in terms of the isotropic phase moduli as \( k_1 = K_1 + G_1/3 \) \((i = 1,2).\) The parameters \( \xi_1 \) and \( \xi_2 \) are integrals over the two- and three-point probability functions \( S_3(r) \) and \( S_3(r,s,t), \) and are defined by

\[
\xi_2 = \frac{4}{\pi \phi_1 \phi_2} \int_{0}^{\infty} \int_{0}^{\infty} \int_{0}^{\infty} \int_{0}^{\pi} \frac{d r}{r} \frac{d s}{s} \frac{d t}{t} d \theta \times S_3(r,s,t) - S_1(r)S_2(s) \cos 2\theta,
\]

and

\[
\eta_2 = \frac{16}{\pi \phi_1 \phi_2} \int_{0}^{\infty} \int_{0}^{\infty} \int_{0}^{\infty} \int_{0}^{\pi} \frac{d r}{r} \frac{d s}{s} \frac{d t}{t} d \theta \times S_3(r,s,t) - S_2(r)S_1(s) \cos 4\theta.
\]

The quantities \( S_3(r) \) and \( S_3(r,s,t) \) are, respectively, the probabilities of finding in phase 2 the end points of a line segment of length \( r \) and the vertices of a triangle with sides of length \( r, s, \) and \( t; \) \( \theta \) is the included angle opposite the side of length \( t. \)

For \( \xi_2 = \eta_2 = 0, \) the above third-order bounds coincide and equal the corresponding second-order Hill–Hashin lower bounds for cases in which phase 2 is stiffer than phase 1. For \( \xi_2 = \eta_2 = 1, \) the bounds coincide and equal the corresponding Hill–Hashin upper bounds for instances in which phase 2 is stiffer than phase 1. The third-order bounds always improve upon the second-order bounds of Hill and Hashin, since both \( \xi_1 \) and \( \eta_1 \) lie in the closed interval \([0,1].\) Hashin showed that the problem of determining the effective axial shear modulus \( \mu \) is mathematically equivalent to determining the effective transverse thermal or electrical conductivity \( \sigma. \) Hence, results for \( \mu \) can be immediately translated into equivalent results for \( \sigma. \)

Milton derived fourth-order bounds on the effective axial shear modulus \( \mu \) or transverse conductivity \( \sigma \) of transversely isotropic fiber-reinforced materials that depend upon the phase properties \( \phi, \xi_2, \) and an integral over the four-point probability function \( S_4. \) Milton showed that by utilizing a phase-change theorem, the integral involving \( S_4 \) can be expressed in terms of \( \phi \) and \( \xi_2 \) only. For the case \( G_2 \geq G_1, \) the fourth-order bounds are given by

\[
\mu^{(4)}_1 \leq \mu \leq \mu^{(4)}_0,
\]

where

\[
\mu^{(4)}_1 = G_2 \left( \frac{(G_1 + G_2)(G_1 + \langle G \rangle) - \phi_2 \xi_1 (G_2 - G_1)^2}{(G_1 + G_2)(G_2 + \langle G \rangle) - \phi_2 \xi_1 (G_2 - G_1)^2} \right),
\]

\[
\mu^{(4)}_0 = G_1 \left( \frac{(G_1 + G_2)(G_1 + \langle G \rangle) - \phi_2 \xi_1 (G_2 - G_1)^2}{(G_1 + G_2)(G_1 + \langle G \rangle) - \phi_2 \xi_1 (G_2 - G_1)^2} \right).
\]

III. EVALUATION OF THE MICROSTRUCTURAL PARAMETERS FOR RANDOM ARRAYS OF POLYDISPERSED CYLINDERS

A. The three-point probability function \( S_3 \)

To evaluate the integrals (12) and (13) for a polydispersed array of infinitely long, oriented, hard cylinders (disks in two dimensions), the three-point probability function \( S_3 \) must be determined. An exact infinite-series representation of the general \( n \) point probability function \( S_n \) has been given by Torquato and Stell for a two-phase system of \( d\)-dimensional identical spheres (phase 2) distributed throughout a matrix (phase 1). The \( S_n \) for random, polydispersed arrays of \( d\)-dimensional spheres can be determined by a straightforward generalization of the explicit expressions of the \( S_n \) derived by Torquato and Stell. For statistically isotropic distributions of impenetrable disks, it has been shown that the infinite series for \( S_n \) terminates with the \( n \)th term; for a system of impenetrable disks having a discrete distribution of \( M \) diameters \( a_i \) \((i = 1,...,M), \) in the case \( n = 3, \) it is given by

\[
S_3(r,s,t) = \frac{4}{\pi \phi_1 \phi_2} \int_{0}^{\infty} \int_{0}^{\infty} \int_{0}^{\infty} \int_{0}^{\pi} \frac{d r}{r} \frac{d s}{s} \frac{d t}{t} d \theta \times S_3(r,s,t) - S_1(r)S_2(s) \cos 2\theta,
\]

\[
S_3(r,s,t) = \frac{16}{\pi \phi_1 \phi_2} \int_{0}^{\infty} \int_{0}^{\infty} \int_{0}^{\infty} \int_{0}^{\pi} \frac{d r}{r} \frac{d s}{s} \frac{d t}{t} d \theta \times S_3(r,s,t) - S_2(r)S_1(s) \cos 4\theta.
\]
\[ S_3 (r_{12}, r_{13}, r_{23}) = S_3^{(1)} (r_{12}, r_{13}, r_{23}) \]
\[ + S_3^{(2)} (r_{12}, r_{13}, r_{23}) \]
\[ + S_3^{(3)} (r_{12}, r_{13}, r_{23}), \]  
(17)

where

\[ S_3^{(1)} = \sum_{i=1}^{M} \rho_i \int m_i (r_{14}) m_i (r_{24}) m_i (r_{34}) \, dr_4, \]  
(18)

\[ S_3^{(2)} = \sum_{i=1}^{M} \sum_{j=1}^{M} \rho_i \rho_j \int m_i (r_{14}) m_i (r_{24}) m_j (r_{35}) \]
\[ \times g_6 (r_{45}) \, dr_4 \, dr_5 + \rho_i \rho_j \int m_i (r_{14}) m_j (r_{35}) \]
\[ \times m_i (r_{24}) g_6 (r_{45}) \, dr_4 \, dr_5 + \rho_i \rho_j \int m_j (r_{14}) \]
\[ \times m_i (r_{24}) m_i (r_{34}) g_6 (r_{45}) \, dr_4 \, dr_5, \]  
(19)

\[ S_3^{(3)} = \sum_{i=1}^{M} \sum_{j=1}^{M} \sum_{k=1}^{M} \rho_i \rho_j \rho_k \int m_i (r_{14}) m_j (r_{25}) \]
\[ \times m_k (r_{36}) g_{ijk} (r_{45} r_{46} r_{56}) \, dr_4 \, dr_5 \, dr_6, \]  
(20)

with

\[ m_i (r) = \begin{cases} 1, & r < a_i, \\
0, & r > a_i. \end{cases} \]  
(21)

Here \( \rho_i \) is the number density of disks (cylinders) of radius \( a_i \), and therefore the disk area fraction (or cylinder volume fraction) \( \phi_i \) is given by the relation

\[ \phi_i = \sum_{i=1}^{M} \rho_i \pi a_i^2. \]  
(22)

\( g_6 \) is the pair (radial) distribution function associated with particles of radii \( a \) and \( a_j \), and \( g_{ijk} \) is the triplet distribution function for particles of radii \( a_i, a_j, \) and \( a_k \), with \( r_{0} = |r - r_i| \). The domain of integration in each of the above integrals in (18)-(20) is the infinite area of the macroscopic sample. Note that \( S_2 \) can be obtained from the expression for \( S_1 \) [Eq. (17)] by letting two of the three points coincide.

The three-dimensional analog of (17) through two-body terms (i.e., up to \( S_2 \)) was utilized by Thovert et al.,\(^{18} \) but was not stated explicitly by them. Torquato\(^2 \) has given a general discussion on correlation functions for polydisperse inclusions, and Lu and Torquato\(^{19} \) have extended the formalism of Torquato\(^20 \) to obtain the so-called general n-point distribution function \( H_n \) for polydispersed suspensions, which contains result (17) as a special case.

**B. The microstructural parameter \( \xi_2 \)**

It is clear from (17)-(20) and (12) that fivefold, sevenfold, and ninefold integrations must be carried out in order to evaluate the microstructural parameter \( \xi_2 \). Torquato and Lado\(^{13} \) have greatly simplified these “cluster” integrals by expanding the angle-dependent terms in Chebyshev polynomials and using the orthogonality properties of these circular harmonics. We shall follow this procedure to compute \( \xi_2 \) for impenetrable disks with a polydispersivity in disk diameter.

We shall first exploit an important property of \( \xi_2 \) for distributions of d-dimensional spheres that only recently has fully come to light; namely, the low-volume-fraction expansion of \( \xi_2 \) provides a remarkably accurate approximation to the exact \( \xi_2 \) for a wide range of \( \phi_2 \), with the term linear in \( \phi_2 \) being the dominant one\(^{11} \) (see also Ref. 2). This property, also true for the parameter \( \eta_3 \), has recently been employed by Thovert et al.,\(^{18} \) Torquato\(^{20} \) and Torquato and Lado\(^{14} \) in related problems. Such behavior has been verified by the computer simulation study of Miller and Torquato\(^{21} \) for impenetrable spheres for virtually the entire volume-fraction range. It is also supported by the simulations of Smith and Torquato\(^7 \) for the nonequilibrium, random-sequential addition of hard disks. This implies that the calculation of \( \xi_2 \) in the superposition approximation by Torquato and Lado\(^{13} \) (although accurate and approximately linear for \( \phi_2 \leq 0.45 \)) overestimates \( \xi_2 \) for large \( \phi_2 \) (i.e., \( \phi_2 > 0.45 \)). The fact that \( \xi_2 \) is approximately linear for a wide range of \( \phi_2 \) makes our task considerably easier since we only need to compute \( \xi_2 \) to leading order in \( \phi_2 \), a result obtainable analytically. Thus our strategy is to evaluate (12) through first order in \( \phi_2 \), implying that we need to substitute only the first two terms of (17) (i.e., up to two-body terms). After simplification, we find that

\[ \xi_2 = \frac{2\phi_2}{\pi \phi_1} b_2, \]  
(23)

where

\[ b_2 = \frac{\pi \rho^2}{\phi_2^2} \int_{0}^{\infty} \int_{r_0}^{\infty} \int_{r_0}^{\infty} da \, db \, a^2 b^2 f(a) f(b) \]
\[ \times \int_{r_0}^{\infty} \int_{r_0}^{\infty} \int_{r_0}^{\infty} dr \, a^2 b^2 \Delta_2 \left( \frac{a}{b} \right) + O(\phi_2), \]  
(24)

where

\[ \Delta_2 \left( \frac{b}{a} \right) = 2 \int_{r_0}^{\infty} \frac{a^2 r \, dr}{(r^2 - b^2)^2} = \frac{1}{1 + 2a/b}. \]  
(25)

Note that the term \( \pi \rho a^2 \) above will lead to a factor of \( \phi_2 \) when integrated over \( a \). When (26) is integrated over both \( a \) and \( b \), a factor of \( \phi_2 \) will arise, making the first term in (26)
of order \( \phi_3 \) rather than of order \( \phi_2^{-1} \) as it may appear. For discrete distributions of \( M \) different disk radii, \( f(a) \) takes the form

\[
f(a) = \sum_{i=1}^{M} \frac{\rho_i}{\rho} \delta(a - a_i)
\]

where \( \delta(a) \) is the Dirac \( \delta \) function. For a monodispersed distribution of disks, \( M = 1 \) and \( a = b \), (26) yields the \( \mathcal{O}(\phi_2) \) result of Torquato and Lado\(^{13}\) for equisized disks:

\[
\xi_2 = \phi_2^2/3 + \mathcal{O}(\phi_2^2).
\]

Torquato and Lado actually evaluated \( \xi_2 \), exactly through \( \mathcal{O}(\phi_2^3) \), but found that the linear term was by far the dominant one. The effects of polydispersity are greatest for suspensions of fibers with widely separated radii, i.e., \( b/a \rightarrow 0 \). For a bidispersed suspension with widely separated fiber radii (\( M = 2 \)) and a polydispersed suspension of \( M \) different (\( M \rightarrow \infty \)) and widely separated fiber radii, (26) yields, respectively,

\[
\xi_2 = \phi_2^2 + \mathcal{O}(\phi_2^3),
\]

\[
\xi_2 = \phi_2^2/2 + \mathcal{O}(\phi_2^2).
\]

Note that (31) is identical to that found in three dimensions by Thouheit et al.\(^{18}\) for a polydispersion of impenetrable spheres with widely separated sizes. Further, observe that (30) lies exactly midway between the monodispersed result (29) and (31).

Following Torquato and Lado,\(^{14}\) we shall apply these low-density expansions for \( \xi_2 \) in the fiber volume-fraction range \( 0 < \phi_2 < 0.7 \). The volume fraction \( \phi_2 = 0.7 \) corresponds to about 87% of the maximum random-close-packing value for monodispersions\(^{22}\). Thus, for polydispersions (which have a larger close-packing fraction), the linear results may be applied beyond \( \phi_2 = 0.7 \); however, this shall not be done here.

In Fig. 1, we plot the parameter \( \xi_2 \) as given in Eq. (26) for the three hard-cylinder cases noted above in (29)–(31), i.e., the monodispersed case, the bidispersion with widely separated cylinder sizes, and the polydispersed case for widely separated cylinder sizes. For purposes of comparison, Fig. 1 also includes \( \xi_2 \) as calculated for symmetric-cell materials\(^{23,24}\) and identical “fully penetrable” or “overlapping” cylinders\(^{9,11}\) (i.e., spatially uncorrelated cylinders), each of which involves no approximation.

It is of interest to note that the effect of polydispersivity is to increase \( \xi_2 \) relative to equisized disks. One might initially expect the converse to be true since \( \xi_2 \) would then be approaching \( \xi_2 = 0 \), the value corresponding to the well-known polydispersed composite-cylinder assemblage of Hashin\(^{4,5}\) for the transverse conductivity (axial shear modulus) and transverse bulk modulus in the cases of more conducting or stiffer inner cylinders. These assemblages realize the second-order bounds. As noted by Torquato,\(^2\) however, because the average separation distance between the inner cylinders in the composite-cylinder assemblage is larger than in the polydispersed, equilibrium hard-cylinder model examined here, the former construction inhibits clustering, and therefore \( \xi_2 \) for the present model should increase, rather than decrease, relative to the monodispersed result. This implies that the lower bounds on \( \mu_\perp \) (or \( \sigma_\perp \)) and \( k_\perp \), which turn out to yield good estimates of the effective properties in these instances,\(^2,13,14,25,26\) will increase with increasing polydispersivity.

As seen in Fig. 1, the symmetric cell material result for \( \xi_2 = \phi_2^2 \) appears to be an upper bound on \( \xi_2 \) for cylinders in all the geometries studied, be they overlapping or totally impenetrable equisized disks, or polydispersed disks; i.e.,

\[
\xi_2 < \phi_2.
\]

This was first noted by Torquato and Lado,\(^{12}\) but has yet to be rigorously proven. Torquato\(^3\) has conjectured (32) to be true for a class of distributions of \( d \)-dimensional spheres. If shown to be rigorously true, then \( \xi_2 \) would be restricted to the closed interval \([0, \phi_2]\) for this class of geometries of cylinders rather than the wider interval \([0, 1] \) which generally applies.

C. The microstructural parameter \( \eta_2 \)

As was done in the previous section, \( \eta_2 \) is evaluated for polydispersed hard cylinders by first simplifying the cluster integrals which result after substituting (17)–(20) into (13). Again, using the circular-harmonics-expansion technique of Torquato and Lado,\(^{13}\) the integral (13) can be significantly simplified (after considerable algebra). As in the case of \( \xi_2 \), the linear term of the low-volume-fraction expansion of \( \eta_2 \) should serve as a very good approximation to \( \eta_2 \) for a wide range of \( \phi_2 \). Thus, we need only substitute the first two terms of (17) into (13) to obtain

\[
\eta_2 \approx \phi_2.
\]
\[ \eta_2 = \frac{8 \phi_2}{\pi \phi_1} c_2, \]  
where  
\[ c_2 = \frac{\pi^2 \rho^2}{\phi_2^2} \int_0^\infty \int_0^\infty da \, db \, a^2 b^2 f(a) f(b) \times \int_a^b \, dr \, r g_2(r) W(r), \]  
\[ W(r) = \frac{\pi a^2}{(r^2 - b^2)^2} \left( 1 - \frac{3a^2}{(r^2 - b^2)^2} \right) - \frac{9}{2} \frac{a^2 b^2}{(r^2 - b^2)^2} + \frac{9}{4} \frac{a^4}{(r^2 - b^2)^2} + \frac{9}{(r^2 - b^2)^3} + \frac{15}{2} \frac{a^4 b^4}{(r^2 - b^2)^3}. \]  
Relation (33) is the polydispersed generalization of the monodispersed result first obtained by Torquato and Lado,\(^{14}\) the three-body term (not given here) was also derived by them for monodispersions.

Combination of (25), (33), (34), and (35) yields through first order in \( \phi_2 \) that  
\[ \eta_2 = \frac{\pi^2 \rho^2}{\phi_2^2} \int_0^\infty \int_0^\infty da \, db \, a^2 b^2 f(a) f(b) \Delta_n \left( \frac{a}{b} \right) + O(\phi_1^3), \]  
where  
\[ \Delta_n \left( \frac{b}{a} \right) = 8 \int_a^b \, r W(r) \, dr \]  
\[ = 8 \left( \frac{1}{2(1 + 7b/a)} - \frac{3}{4(1 + 2b/a)^2} \right) \]  
\[ - \frac{3}{4} \frac{(b/a)^2}{(1 + 2b/a)^3} + \frac{3}{8} \frac{1}{(1 + 2b/a)^3} \]  
\[ + \frac{9}{8} \frac{(b/a)^2}{(1 + 2b/a)^4} + \frac{3}{4} \frac{(b/a)^4}{(1 + 2b/a)^5}. \]  
Figure 2 compares the functions \( \Delta_\zeta \) and \( \Delta_\eta \) as a function of the cylinder size ratio \( a/b \).

Employing Eq. (28), one can then examine the behavior of \( \eta_2 \) for the three cases noted above for \( \zeta_2 \), namely, the monodispersed result, and the widely spaced bidispersed and polydispersed suspensions. For these three cases, one finds, respectively,  
\[ \eta_2 = 8 \phi_2 + O(\phi_1^2), \]  
\[ \eta_2 = 12 \phi_2 + O(\phi_1^3), \]  
\[ \eta_2 = 14 \phi_2 + O(\phi_1^3). \]  
As before, the result (38) recovers the result of Torquato and Lado\(^{14}\) for a random distribution of equisized hard disks. For the aforementioned polydispersed microgeometries, \( \eta_2 \) (unlike \( \zeta_2 \)) decreases as the degree of polydispersivity increases, although the effect is much smaller than the increase observed for \( \zeta_2 \). The polydispersed result (40) is identical to that found by Thovert et al.\(^{18}\) for the analogous three-dimensional case, and again we note that the result (39) for a bidispersed suspension of fibers with widely spaced radii lies exactly midway between (38) and (40).

In Fig. 3, we plot the preceding three results as well as the result for symmetric-cell materials\(^{23,24}\) and random arrays of identical overlapping cylinders.\(^{10,12}\)

We note that Torquato\(^{2}\) has conjectured that for a class of distributions of \( d \)-dimensional spheres (e.g., symmetric-
cell materials with spherical or cylindrical cells, overlapping and impenetrable disks or spheres),

\[ \eta_2 > \zeta_2. \]  

(41)

He based this conjecture on observations for known results. He also noted that if (32) is true, then \( \eta_2 \) for \( d \)-dimensional spheres will lie in the more restricted closed interval \([\zeta_2, \theta_2]\) rather than the interval \([0,1]\) which generally applies.

Note that the results above differ from the analogous results for hard spheres in two ways. First, for hard cylinders, increasing polydispersivity decreases \( \eta_2 \) from \( \eta_2 = 56/81 \phi_2 \) for monodispersions [Eq. (38)] to \( \eta_2 = 0.5 \phi_2 \) for polydispersions with widely spaced cylinder radii [Eq. (40)], while for hard spheres, \( \eta_2 \) increases from the monodispersed result \( \eta_2 = 0.48274 \phi_2 \) to \( \eta_2 = 0.5 \phi_2 \) for polydispersed hard spheres with widely spaced radii. Second, the relative difference between the polydispersed and monodispersed values is considerably greater for hard cylinders than for hard spheres. This will lead to a significant difference in the behavior of the effective shear modulus for dispersions of hard spheres and the effective transverse shear modulus for suspensions of oriented hard cylinders.

A parameter \( \eta_2 \) which decreases as the degree of polydispersivity increases can have the effect of lowering the third-order bounds (7) on the transverse shear modulus \( G_t \) relative to the monodisperse result. [Note that unlike second-order bounds on \( \mu_c \) (\( \sigma_e \)) and \( k_c \), the second-order bounds on \( G_c \) are not achieved by composite-cylinder assemblies.4] In the case of perfectly rigid cylinders in an incompressible matrix, the results (38)–(40) and bounds (7) imply that \( G_c \) exactly, through third order in \( (G_c - G_t) \), must decrease as the polydispersivity increases for the hard-cylinder model.

IV. CALCULATION OF IMPROVED BOUNDS ON THE ELASTIC MODULI OF RANDOM ARRAYS OF POLYDISPERSED CYLINDERS

Here we employ the results (26) and (36) to compute the bounds presented in Eqs. (1)–(11) for suspensions of long, oriented, impenetrable cylinders with a polydispersivity in cylinder radius. We will examine cases in which the fiber stiffness (conductivity) is greater that that of the matrix in the range of fiber volume fractions \( 0 < \phi_2 < 0.7 \). It should be noted that for the cases presented here, the lower bounds will provide a relatively accurate estimate of the effective property,2,13,14,25,26 even when the bounds are not tight. This has recently been confirmed by exact determinations of the effective transverse conductivity (or effective axial shear modulus) for equilibrium distributions of hard cylinders obtained by Kim and Torquato.27 This is due to the fact that the stiffer phase (fibers) can never form large connected clusters in the range of volume fractions considered here. Equilibrium distributions of cylinders do not form interparticle contacts until the random-close-packing volume fraction22 (\( \phi_2 \approx 0.81 \)) is reached.

In Fig. 4, we present third- and fourth-order bounds on the scaled effective transverse conductivity \( \sigma_e / \sigma_1 \) as calculated from Eqs. (1) and (14) as a function of the fiber volume fraction \( \phi_2 \) for a composite with a conductivity ratio

\[ \sigma_2 / \sigma_1 = 10 \]  

for a distribution of polydispersed impenetrable cylinders with widely spaced radii, i.e., with \( \zeta_2 \) given by (31). In these bounds we have replaced \( \mu_c \), \( \sigma_t \), and \( G_t \), with \( \sigma_c \), \( \sigma_1 \), and \( \sigma_2 \), respectively. Also included in the figure are the second-order upper and lower bounds derived by Hashin4,5. Observe that the third-order Slinmiser bounds are

\[ \sigma_e / \sigma_1 = 50 \]  

for a distribution of polydispersed impenetrable cylinders with widely spaced radii [cf. Eq. (31)] at \( \sigma_2 / \sigma_1 = 10 \); dotted curve, second-order bounds; (see Refs. 4 and 5) dashed curve, third-order bounds; (see Ref. 6) and solid curve, fourth-order bounds (see Ref. 8). These results can be immediately translated to equivalent results for the effective axial shear modulus (see Ref. 5).
considerably more restrictive than the second-order Hashin bounds, and the fourth-order Milton bounds are a further improvement over the third-order bounds. The fourth-order lower bound provides an accurate estimate of the effective transverse conductivity (or, equivalently, axial shear modulus) for the range of fiber volume fractions reported.

Figure 5 presents the same bounds for the case $\sigma_2/\sigma_1 = 50$. As expected, the bounds are wider than in the previous case, but as noted earlier, $\sigma_2$ is still accurately estimated by the fourth-order lower bound.

The case of superconducting fibers ($\sigma_2/\sigma_1 = \infty$) (or superrigid fibers in the case of the axial shear modulus) is presented in Fig. 6. Here, we present only the fourth-order lower bounds since the upper bounds diverge to infinity, but include the three cases (29)–(31) corresponding to monodispersed impenetrable cylinders, bidispersed impenetrable cylinders with widely spaced radii, and polydispersed impenetrable cylinders with widely spaced radii, respectively. The effect of polydispersivity is to increase the effective transverse conductivity or effective axial shear modulus.

In Fig. 7, we present the fourth-order bounds on the scaled effective axial shear modulus $\mu_\alpha/G_\alpha$ [Eqs. (15) and (16)] as a function of the fiber volume fraction $\phi_2$ for a glass-epoxy composite for which $G_2/G_1 = 22$ for three microgeometries of hard cylinders: monodispersed, bidispersed with widely spaced cylinder radii, and polydispersed with widely spaced radii [Eqs. (29)–(31)]. Also included are the corresponding second-order bounds. The fourth-

**Figure 5.** Fourth-order lower bounds on the scaled effective transverse conductivity $\sigma_2/\sigma_1$ for three microgeometries of hard cylinders, at $\sigma_2/\sigma_1 = \infty$; dotted curve, monodispersed hard cylinders using (29); dashed curve, bidispersed hard cylinders with widely spaced cylinder radii using (30); solid curve, polydispersed hard cylinders with widely spaced cylinder radii using (31). Also included is the Hashin (see Refs. 4 and 5) two-point bounds (dot-dashed curve).

**Figure 6.** Fourth-order lower bounds on the scaled effective transverse conductivity $\sigma_2/\sigma_1$ for three microgeometries of hard cylinders, at $\sigma_2/\sigma_1 = \infty$; dotted curve, monodispersed hard cylinders using (29); dashed curve, bidispersed hard cylinders with widely spaced cylinder radii using (30); solid curve, polydispersed hard cylinders with widely spaced cylinder radii using (31).

**Figure 7.** Fourth-order bounds on the scaled effective transverse conductivity $\sigma_2/\sigma_1$ for three microgeometries of hard cylinders, at $\sigma_2/\sigma_1 = \infty$; dotted curve, monodispersed hard cylinders using (29); dashed curve, bidispersed hard cylinders with widely spaced cylinder radii using (30); solid curve, polydispersed hard cylinders with widely spaced cylinder radii using (31).

**Figure 8.** Third-order bounds (see Ref. 6) on the scaled effective transverse bulk modulus $k_\alpha/k_1$ vs the cylinder volume fraction $\phi_2$ for glass-epoxy composites for which $G_2/G_1 = 22$, $G_1/k_1 = 0.21$, and $G_2/k_2 = 0.46$; dotted curve, monodispersed hard cylinders using (29); dashed curve, bidispersed hard cylinders with widely spaced cylinder radii using (30); solid curve, polydispersed hard cylinders with widely spaced cylinder radii using (31). Also included are the Hashin (see Refs. 4 and 5) two-point bounds (dot-dashed curve).
order bounds are considerably tighter than the second-order bounds and will yield good estimates of $\mu _c$ over a wide range of fiber volume fractions $\phi _f$.

In Fig. 8, we compare the third-order Silnutzer bounds$^9$ on the scaled effective transverse bulk modulus $k_e/k_1$ for a glass-epoxy composite with $G_e/G_1 = 22$, $G_1/K_1 = 0.21$, and $G_2/K_2 = 0.46$ for three microgeometries: equisized hard cylinders, bidispersed hard cylinders with widely spaced radii, and polydispersed hard cylinders with widely spaced radii. The corresponding second-order bounds$^3,4$ are presented for comparison. Note that the effect of polydispersivity is to shift both upper and lower bounds upward.

Figure 9 presents third-order bounds$^9$ on $G_e/G_1$ with the same material properties as in Fig. 8 for equisized hard disks, bidispersed hard disks with widely spaced radii, and polydispersed hard disks with widely spaced radii. Included for comparison are the corresponding second-order bounds$^3,4$. In the case of $G_e$, it can be seen that the effect of polydispersivity is to decrease the effective property.

As is clear from the results presented here, the effect of polydispersivity in cylinder size can either increase or decrease the effective property. For dispersions of impenetrable cylinders in general, an increase in polydispersivity will increase the effective axial shear modulus $\mu _c$ (or effective bulk modulus $k_c$) and the effective transverse bulk modulus $k_e$ relative to the monodispersed case. In the case of the effective transverse shear modulus $G_e$, however, polydispersivity acts to decrease the effective property relative to the monodisperse case. This behavior is in contrast to that noted for three dimensions, i.e., dispersions of hard spheres. Thovert et al. found that the effect of increasing the polydispersivity of a distribution of hard spheres increased the effective transverse shear modulus $G_e$ relative to monodispersions in most cases. However, they noted that in certain instances when the difference in phase moduli $G_1$ and $G_2$ was small, the polydispersivity may lead to a decrease in the effective property $G_e$. This difference between two and three dimensions is attributed to the different behavior of $q z$ for cylinders relative to spheres noted in the previous section.

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