Extreme Mechanics Letters 55 (2022) 101818

Contents lists available at ScienceDirect

Extreme Mechanics Letters

journal homepage: www.elsevier.com/locate/eml

The effective shear modulus of a random isotropic suspension of monodisperse rigid n-spheres: From the dilute limit to the percolation threshold

Victor Lefèvre [a], Oscar Lopez-Pamies [b,c,*]

[a] Department of Mechanical Engineering, Northwestern University, Evanston, IL 60208, USA
[b] Department of Civil and Environmental Engineering, University of Illinois, Urbana–Champaign, IL 61801, USA
[c] Département de Mécanique, École Polytechnique, 91128 Palaiseau, France

ARTICLE INFO

Article history:
Received 6 December 2021
Received in revised form 15 April 2022
Accepted 7 June 2022
Available online 15 June 2022

Keywords:
Fillers
Hydrodynamic reinforcement
Homogenization

ABSTRACT

A simple explicit result is introduced for the effective shear modulus of a random isotropic suspension of rigid n-spheres ($n = 3, 2$), each having identical size, firmly embedded in an isotropic incompressible elastic matrix. By construction, the result is in quantitative agreement with all the classical rigorous asymptotic results in the dilute ($c \searrow 0$) and percolation ($c \nearrow p_n$) limits, as well as with new computational results for intermediate values $c \in [0, p_n]$ of the volume fraction of n-spheres. Moreover, as demonstrated by means of iterated homogenization, the proposed result has the added merit of being realizable by a certain class of random isotropic suspension of rigid n-spheres with infinitely many sizes. That the proposed result is descriptive of both isotropic suspensions with monodisperse and with (a specially selected class of) polydisperse rigid n-spheres is nothing more than a manifestation of the richness in behaviors that suspensions of polydisperse rigid n-spheres can exhibit.

1. Introduction

The determination of the effective shear modulus $\overline{\mu}_n$ of a random isotropic suspension of monodisperse rigid n-spheres[1] ($n = 3, 2$) firmly embedded in an isotropic incompressible elastic matrix is a fundamental problem in mechanics that has had a long and rich history since the pioneering work of Einstein [2,3]. In the next few paragraphs, we summarize the various milestones accomplished throughout the years until present times. In Section 2, we introduce the main result of this Letter, to wit, a simple explicit formula for $\overline{\mu}_n$ that is valid from the dilute limit to the percolation threshold. As elaborated in that same section, the key properties of the proposed result for $\overline{\mu}_n$ are that — in addition to being simple and explicit — it is in quantitative agreement with all the rigorous asymptotic and computational results known to date and, furthermore, it is realizable. The details of its realizability are presented in Section 3. We close in Section 4 by recording a few final remarks.

The dilute limit. As alluded to above, the first main result in the literature is famously due to Einstein [2,3], who worked out the exact solution[2] in the asymptotic limit of small volume fraction c of 3-spheres:

$$\overline{\mu}_3 = \mu + \frac{5}{2}\mu\, c + O(c^2); \tag{1}$$

throughout this Letter, μ stands for the shear modulus of the matrix material.

The corresponding solution for the case of 2-spheres can be extracted from the succeeding and also celebrated work of Eshelby [5]:

$$\overline{\mu}_2 = \mu + 2\mu\, c + O(c^2). \tag{2}$$

Later (see, e.g., Chapter 17 in [6]), it was recognized that the dilute solution for $n \geq 2$ dimensions (not just $n = 3$ and 2) can actually be written in the compact form

$$\overline{\mu}_n = \mu + \left(1 + \frac{n}{2}\right)\mu\, c + O(c^2). \tag{3}$$

For reference in the sequel, we note that for the more general case when the n-spheres are not rigid but just incompressible with

* Corresponding author at: Department of Civil and Environmental Engineering, University of Illinois, Urbana–Champaign, IL 61801, USA.
E-mail addresses: victor.lefevre@northwestern.edu (V. Lefèvre), pamies@illinois.edu (O. Lopez-Pamies).

1 In this work, we make use of the terminology usually employed by geometers — and not that employed by topologists — who refer to circles as 2-spheres and spheres as 3-spheres; see, e.g., Section 7.3 in [1].

2 Einstein [2,3] carried out his analysis within the context of Stokes flow, which, as well known, is mathematically equivalent to that of linear elastostatics at a fixed instant in time. Some four decades later, Smallwood [4] transcribed the analysis to linear elastostatics.

https://doi.org/10.1016/j.eml.2022.101818
2352-4316/

shear modulus μ_s, the corresponding effective shear modulus reads

$$\overline{\mu}_n = \mu + \frac{(2+n)(\mu_s - \mu)}{n\mu + 2\mu_s}\mu\, c + O(c^2). \tag{4}$$

Higher-order correction to the dilute limit. Several works of increasing refinement targeted the computation of the correction of $O(c^2)$ in the Einstein formula (1) for 3-spheres. Among these [7–11], the result

$$\overline{\mu}_3 = \mu + \frac{5}{2}\mu\, c + 5.01\mu\, c^2 + O(c^3) \tag{5}$$

due to Chen and Acrivos [11] is the most accurate one, where the coefficient 5.01 is exact to within ±0.2%.

We are not aware of analogous analytical efforts on the computation of the correction of $O(c^2)$ in the result (2) for 2-spheres; see, however, [12] for results on a hexagonal suspension of 2-spheres. Nevertheless, the recent computations of Lefèvre et al. [13] indicate that

$$\overline{\mu}_2 = \mu + 2\mu\, c + 3.64\mu\, c^2 + O(c^3), \tag{6}$$

where the coefficient 3.64 is believed to be accurate to within ±5%.

For the mathematical analysis of the problem, the interested reader is referred to the recent contribution [14] and references therein.

Percolation. Because of the mechanical rigidity of the n-spheres, the effective shear modulus $\overline{\mu}_n$ will grow unbounded as the volume fraction c approaches the percolation threshold p_n of the suspension. We thus have the asymptotic result

$$\lim_{c \nearrow p_n} \overline{\mu}_n = +\infty. \tag{7}$$

For the case of 3-spheres, experiments [15] and computations [16, 17] have shown that

$$p_3 \approx 0.64. \tag{8}$$

For the case of 2-spheres, on the other hand, computations [16, 17] have shown that

$$p_2 \approx 0.90. \tag{9}$$

Here, it is important to emphasize that the percolation thresholds (8) and (9) are roughly upper bounds. Indeed, depending on the construction process of the suspensions, somewhat lower values may already lead to percolation.

Computational results. Advances over the past two decades in computational resources and methods for solving the underlying elastostatics equations have allowed to generate numerical approximations for $\overline{\mu}_n$ for finite volume fractions c of n-spheres beyond the dilute limit. The basic idea consists in approximating the random isotropic suspensions of interest here as periodic suspensions where the periodically repeated unit cell contains a sufficiently large number of randomly distributed n-spheres so that the homogenized elastic response is isotropic to within a high degree of accuracy [18]. Finite-element (FE) results (based on hybrid finite elements to deal with the incompressibility of the matrix material) for 3-spheres have been presented — to various degrees of accuracy — in [19,20] up to $c = 0.40$ and in [21–23] up to $c = 0.55$. For the case of 2-spheres, results up to $c = 0.35$ have been presented in [24] and up to $c = 0.50$ in [25].

In two recent contributions, Lefèvre et al. [13] and Lefèvre [26] have provided the most comprehensive and accurate set of computational results yet for suspensions with both 3- and 2-spheres. In a nutshell, they examined tens of thousands of realizations for unit cells containing up to 960 randomly distributed 3-spheres

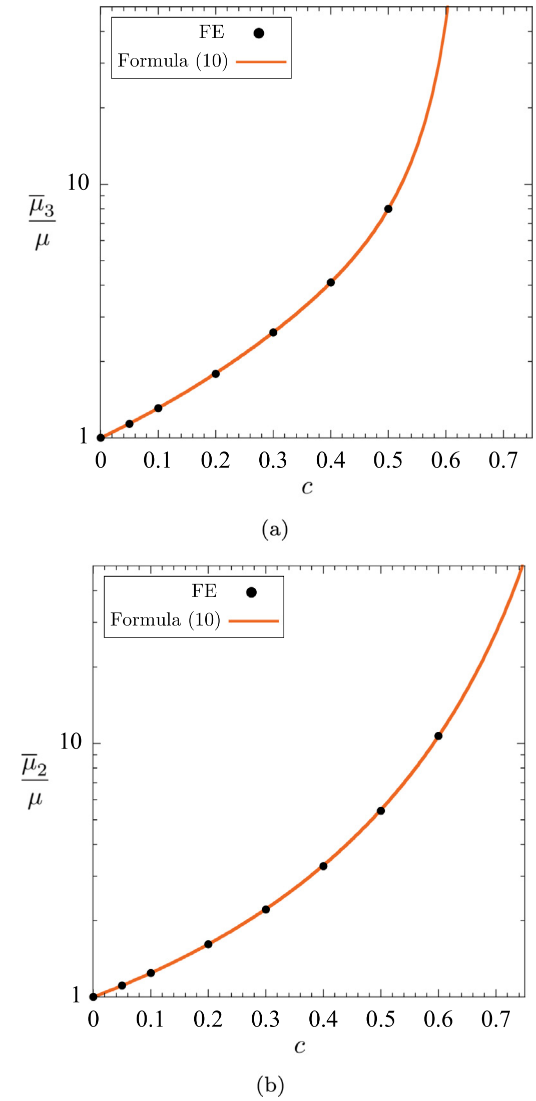

Fig. 1. FE results from [13,26] for the effective shear modulus $\overline{\mu}_n$ of random isotropic suspensions of monodisperse rigid n-spheres in an isotropic incompressible elastic matrix. The results are shown (in semi-log scale) normalized by the shear modulus of the matrix μ as a function of the volume fraction c of 3-spheres in part (a) and of 2-spheres in part (b). For direct comparison, the proposed formula (10) is also included (solid lines) in the figures.

with volume fractions in the range $c \in [0, 0.50]$ and randomly distributed 2-spheres with volume fractions in the range $c \in [0, 0.60]$ for several fixed values of the minimum distance d between the n-spheres (more on this in Section 4.1 below). They then filtered out the realizations that did not exhibit an isotropic elastic response to within a stringent tolerance and averaged the ones that did for each volume fraction c and minimum inter-n-sphere distance d that they considered. Out of those, as expected [27], the maximum difference between any two realizations with the same c and d was less than 2%. Their results for $d = 0+$ are presented in Fig. 1(a) for the case of 3-spheres and in Fig. 1(b) for that of 2-spheres.

2. The main result and its properties

In this section, we show that the explicit formula Eq. (10) given in Box I is in quantitative agreement with all the analytical and computational results outlined above and hence that it provides an accurate description for the effective shear modulus of

$$\overline{\mu}_n=\frac{\mu}{\left[\left(1+\alpha_n\left(\beta_n+\left(\frac{c}{p_n}\right)^{n-1}\right)\left(\frac{c}{p_n}\right)^2\right)\left(1-\frac{c}{p_n}\right)\right]^{\frac{(2+n)p_n}{2}}}\quad\text{with}\quad\begin{cases}\begin{cases}\alpha_3=0.635\\ \beta_3=0.027\end{cases}\\ \\ \begin{cases}\alpha_2=-0.299\\ \beta_2=0.796\end{cases}\end{cases},\qquad n=3,2. \tag{10}$$

Box I.

a random isotropic suspension of monodisperse rigid n-spheres, this for any volume fraction $c\in[0,p_n]$ from the dilute limit to the percolation threshold p_n.

Remark 1 (*The Dilute Limit*)**.** In the limit as the volume fraction $c\searrow 0$, the effective shear modulus $(10)_1$ reduces asymptotically to

$$\overline{\mu}_n=\mu+\left(1+\frac{n}{2}\right)\mu\, c+\frac{(2+n)(2+2p_n+np_n-4\alpha_n\beta_n)}{8p_n}\mu\, c^2+O(c^3). \tag{11}$$

Thus, the formula (10) agrees identically with the exact dilute result (3).

Remark 2 (*Higher-order Correction to the Dilute Limit*)**.** For the case of 3-spheres when the percolation threshold is given by (8), the asymptotic result (11) specializes to

$$\overline{\mu}_3=\mu+\frac{5}{2}\mu\, c+5.01\mu\, c^2+O(c^3). \tag{12}$$

On the other hand, for the case of 2-spheres when the percolation threshold is given by (9), the result (11) specializes to

$$\overline{\mu}_2=\mu+2\mu\, c+3.64\mu\, c^2+O(c^3). \tag{13}$$

Thus, the formula (10) also agrees identically with the higher-order asymptotic results (5) and (6).

Remark 3 (*Percolation*)**.** It is trivial to verify that the effective shear modulus (10) grows unbounded as the volume fraction c of n-spheres approaches the percolation threshold p_n, and hence that it satisfies the asymptotic result (7).

Remark 4 (*Comparison with Computational Results*)**.** Fig. 1 shows comparisons between the formula (10) and the computational results in [13,26]. It is plain that both sets of results are in good quantitative agreement for all volume fractions c for which the computational results are available.

Remark 5 (*Connection with the Classical Differential Scheme*)**.** The formula (10) can be thought of as a generalization of the classical differential-scheme (*DS*) result [28–30]

$$\overline{\mu}_{DS_n}=\frac{\mu}{(1-c)^{\frac{2+n}{2}}} \tag{14}$$

for the effective shear modulus of an isotropic suspension of n-spheres with infinitely many sizes in that the volume fraction c is re-scaled by the percolation threshold p_n, $c\mapsto c/p_n$. A re-scaling of this type, which can be traced back to the work of Eilers [31], has been used *heuristically* — and, in particular, disconnected from the differential scheme [28] — by countless authors in an attempt to account for percolation in the analogous problem of the determination of the viscosity of suspensions of rigid 3-spheres in a Newtonian fluid; see, e.g., the review articles [32,33] and references therein. Among the numerous heuristic formulas that have been proposed with the re-scaling $c\mapsto c/p_3$ in the fluids mechanics literature, it is worth noting that the one proposed by Krieger and Dougherty [34]

$$\overline{\mu}_{KD}=\frac{\mu}{\left(1-\frac{c}{p_3}\right)^{\frac{5}{2}p_3}} \tag{15}$$

falls squarely within the functional form of the effective shear modulus $(10)_1$. Indeed, the Krieger–Dougherty formula (15) can be viewed as a special case of the formula $(10)_1$ for the choice of coefficient $\alpha_3=0$ when $n=3$.

Remark 6 (*Realizability*)**.** The formula (10) is not "just" a formula that happens to be in agreement with the above-summarized analytical and computational results, but has also the merit to be realizable. Precisely, as elaborated in the next section, the formula (10) can be shown to be the *exact homogenization solution* for the effective shear modulus of a certain class of random isotropic suspensions of rigid n-spheres with *infinitely many sizes*. As a result, it is guaranteed to satisfy all pertinent physical and mathematical restrictions (e.g., bounds) on the elastic response of isotropic suspensions of rigid n-spheres. That the effective shear modulus (10) is descriptive of both isotropic suspensions with monodisperse and with (a specially selected class of) polydisperse rigid n-spheres is nothing more than a manifestation of the richness in behaviors that suspensions of polydisperse rigid n-spheres can exhibit.

3. Realizability

It follows immediately from use of the Eshelby solution (4) in the generalized differential scheme originally introduced by Norris [35], and later made rigorous by Avellaneda [36], that the first-order nonlinear ordinary differential equation (ODE)

$$(1-\phi_1(t)-\phi_2(t))\frac{\mathrm{d}\widehat{\mu}_n}{\mathrm{d}t}(t)=\left[(1-\phi_2(t))\frac{\mathrm{d}\phi_1}{\mathrm{d}t}(t)+\phi_1(t)\times\frac{\mathrm{d}\phi_2}{\mathrm{d}t}(t)\right]\frac{(2+n)(\mu-\widehat{\mu}_n(t))}{n\widehat{\mu}_n(t)+2\mu}\widehat{\mu}_n(t)+\left[(1-\phi_1(t))\frac{\mathrm{d}\phi_2}{\mathrm{d}t}(t)+\phi_2(t)\frac{\mathrm{d}\phi_1}{\mathrm{d}t}(t)\right]\frac{2+n}{2}\widehat{\mu}_n(t)\qquad t\in(0,1], \tag{16}$$

with initial condition

$$\widehat{\mu}_n(0)=\mu, \tag{17}$$

defines the effective shear modulus

$$\overline{\mu}_n=\widehat{\mu}_n(1) \tag{18}$$

for a large class of random isotropic suspensions of n-spheres of infinitely many sizes firmly embedded in an isotropic incompressible elastic matrix with shear modulus μ. In Eq. (16), $\phi_1(t)$ and $\phi_2(t)$ stand for non-negative continuous functions of choice subject to the constraints that $\phi_1(t)+\phi_2(t)\leq 1$, the combinations $\phi_1(t)/(1-\phi_1(t)-\phi_2(t))$ and $\phi_2(t)/(1-\phi_1(t)-\phi_2(t))$

$$\mathcal{E}(\gamma_n;c)=\frac{2\gamma_n}{2\gamma_n-n\,c}\ln\left[\frac{(2+n)c}{2(c+\gamma_n)+(n\,c-2\gamma_n)\left[\left(1+\alpha_n\left(\beta_n+\left(\frac{c}{p_n}\right)^{n-1}\right)\left(\frac{c}{p_n}\right)^2\right)\left(1-\frac{c}{p_n}\right)\right]^{-\frac{(2+n)p_n}{2}}}\right]+$$
$$\ln\left[\frac{1-c-\gamma_n}{\left(1+\alpha_n\left(\beta_n+\left(\frac{c}{p_n}\right)^{n-1}\right)\left(\frac{c}{p_n}\right)^2\right)^{p_n}\left(1-\frac{c}{p_n}\right)^{p_n}}\right]=0. \tag{19}$$

Box II.

are monotonically increasing functions of t, $\phi_1(0)=\phi_2(0)=0$, and $\phi_2(1)=c$, where, again, c stands for the volume fraction of n-spheres in the suspension. From a computational point of view, we remark that the ODE (16) needs to be solved from the initial condition (17) at $t=0$ up to $t=1$, as its solution then $\widehat{\mu}_n(1)$ defines the effective shear modulus (18) of the suspension.

The specific choice of functions $\phi_1(t)$ and $\phi_2(t)$ defines the type of suspension being considered, that is, the specific sizes and spatial distributions of the n-spheres. For instance, taking $\phi_1(t)=0$ and $\phi_2(t)=c\,t$ leads to the classical *DS* result (14). It is not difficult to show that the result (10) proposed in this work belongs to the more general family of "radial" construction paths

$$\phi_1(t)=\gamma_n(c)\,t \qquad \text{and} \qquad \phi_2(t)=c\,t,$$

where $\gamma_n(c)$ is not zero but rather defined implicitly as the smallest positive root of the nonlinear algebraic equation (19) given in Box II. In this last expression, we have omitted the argument c in γ_n for simplicity and recall that the percolation threshold p_n is given by (8) and (9) for 3- and 2-spheres. More importantly, the coefficients α_n and β_n in (19) are arbitrary so long as they lead to real solutions for γ_n in the range $0\le\gamma_n\le 1-c$. Different choices of α_n and β_n lead to different spatial distributions of n-spheres and hence to different solutions for the effective shear modulus $\overline{\mu}_n$ of the resulting suspension. For instance, the choice $\alpha_3=0$ leads to the Krieger–Dougherty formula (15) thereby demonstrating that this heuristically-derived classical result is in fact realizable by a certain class of random isotropic suspension of rigid 3-spheres with infinitely many sizes. More importantly, the specific values $(10)_2$ for α_n and β_n are the ones that happen to best fit the FE results in Fig. 1, at the same time that they also lead to the exact asymptotic results (12), (13), and (7).

While Eq. (19) does not admit an explicit solution for γ_n, it is a simple matter to solve it numerically. Fig. 2 provides plots of such solutions when α_n and β_n take the values $(10)_2$. One point worth remarking from them for both 3- and 2-spheres is that the value of γ_n at percolation, when $c=p_n$, is given by $\gamma_n=1-p_n$.

We close this section by emphasizing that even though the formula (10) is exact for the effective shear modulus of the class of random isotropic suspensions of n-spheres with *infinitely many sizes* defined by the ODE (16) with (17)–(19), it is also asymptotically exact in the dilute (up to $O(c^2)$) and percolation limits for the effective shear modulus of the class of *monodisperse* suspensions of interest here. Although quantitatively accurate for intermediate values of the volume fraction c of n-spheres, the formula (10) is not expected to be exact beyond those asymptotic limits for the monodisperse suspensions.

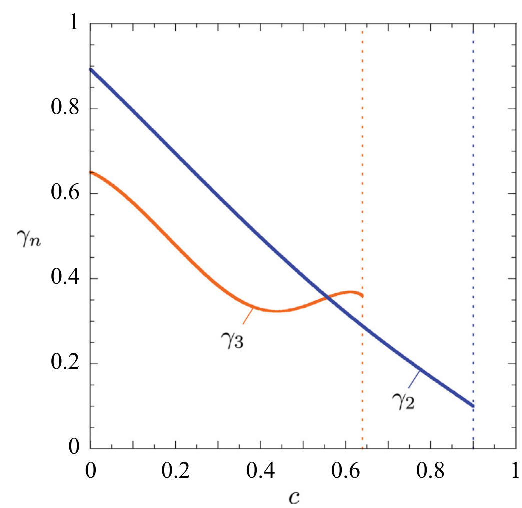

Fig. 2. Plots of the variable γ_n defined by the nonlinear algebraic equation (19), with percolation thresholds (8), (9), and coefficients $(10)_2$, as a function of the volume fraction c of n-spheres.

4. Final comments

4.1. *Suspensions with a minimum distance d between the n-spheres*

The result (10) applies to isotropic suspensions where the n-spheres are not allowed to overlap, but other than that their locations are not subject to any separation constraint. In particular, the minimum distance d between any two n-spheres could be vanishingly small. Isotropic suspensions where the n-spheres are still randomly distributed but the minimum distance d between them is restricted to be larger than a certain imposed threshold — this is straightforward to accomplish *in silico*, while

Table 1
Values of the coefficients α_2 and β_2 in the effective shear modulus $(10)_1$ for random isotropic suspensions of 2-spheres of radius R with minimum distance d between them.

d/R	α_2	β_2
0+	−0.299	0.796
0.01	−0.278	0.820
0.02	−0.208	1.051
0.05	−0.055	3.424
0.10	0.144	−0.966
0.20	0.309	−0.129

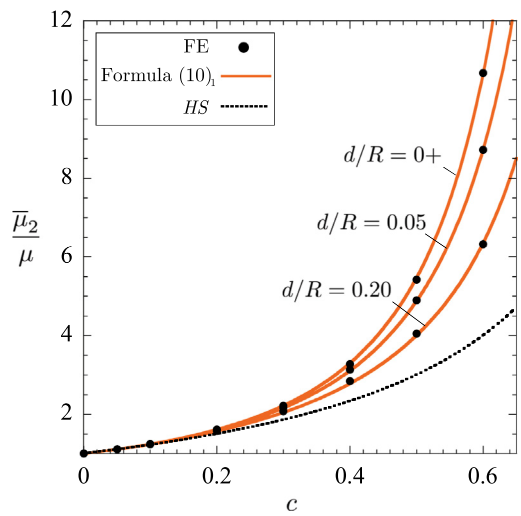

Fig. 3. Comparison between the formula (10)$_1$, with the coefficients α_2 and β_2 given in Table 1, and the computational results from [26] for the effective shear modulus of random isotropic suspensions of monodisperse rigid 2-spheres wherein the minimum distance d between the 2-spheres is a fraction of their radius R. The results are shown normalized by the shear modulus of the matrix μ for $d/R = 0+$, 0.05, and 0.20 as functions of the volume fraction c of 2-spheres. For reference, the Hashin–Shtrikman (HS) lower bound (23) is also included in the figure.

in practice chemical treatments on the surfaces of the n-spheres, for instance, may have the same effect — may feature a softer elastic response, especially at larger volume fractions c of n-spheres [13,26]. It turns out that the formula (10)$_1$ can also provide an accurate description for the effective shear moduli of those suspensions by appropriately calibrating the value of its coefficients α_n and β_n in terms of the minimum inter-n-sphere distance d.

By way of an example, Table 1 presents the calibrated values of α_2 and β_2 for the isotropic suspensions investigated computationally in [13,26] featuring a minimum distance between the 2-spheres of 1%, 2%, 5%, 10%, and 20% of their radius R; the values (10)$_2$ for α_2 and β_2 corresponding to $d/R = 0+$ are also included in the table for direct comparison. Interestingly enough, straightforward calculations show that the formula (10)$_1$ evaluated at all the pairs of coefficients α_2 and β_2 listed in Table 1 for $d/R = 0.01, 0.02, 0.05, 0.10, 0.20$ remains solution of (16) with (17)–(19) and hence realizable.

Fig. 3 confronts the formula (10)$_1$, with the values given in Table 1 for α_2 and β_2, to the computational results in [26] for suspensions with minimum inter-2-sphere distances $d/R = 0.05$ and 0.20; the results for $d/R = 0+$ are also included for direct comparison. Three observations are in order. First, the formula (10)$_1$ is indeed in good quantitative agreement with the computational results. Second, the effective shear modulus of suspensions of monodisperse 2-spheres is largely insensitive to the minimum distance between the 2-spheres up to a volume fraction of around $c = 0.20$. For larger volume fractions, suspensions with larger minimum inter-2-sphere distances exhibit a sizably smaller effective shear modulus. Nevertheless, the effect of a minimum inter-2-sphere distance $d/R > 0$ is already present at $O(c^2)$. Indeed, the evaluation of the asymptotic expression (11) at the values given in Table 1 for α_2 and β_2 yields $\overline{\mu}_2 = \mu + 2\mu c + 3.64\mu c^2$, $\overline{\mu}_2 = \mu + 2\mu c + 3.53\mu c^2$, $\overline{\mu}_2 = \mu + 2\mu c + 3.20\mu c^2$ for $d/R = 0+, 0.05, 0.20$, respectively. As expected on physical grounds, the correction of $O(c^2)$ decreases with increasing minimum inter-2-sphere distance. Third, the softer behavior afforded by larger minimum inter-2-sphere distances is akin to that found in suspensions of polydisperse n-spheres [19,20,37]. As a reference result to aid in gauging this softening, the Hashin–Shtrikman (HS) lower bound (23) is included in Fig. 3.

4.2. Suspensions with polydisperse n-spheres

It is also instructive to compare the result (10) for isotropic suspensions of monodisperse n-spheres with two results for isotropic suspensions of n-spheres with infinitely many sizes that are widely utilized in the literature. Those are the DS result (14) already referenced above and the result originally introduced by Christensen and Lo [38], and later derived by alternative means and shown to be realizable by Avellaneda [36], for the so-called differential coated n-sphere (DCS) assemblage. For 3- and 2-spheres, the latter reads

$$\overline{\mu}_{DCS_3} = \mu + \frac{35c}{7 - 15c + 8c^{10/3} + \sqrt{q_3}}\,\mu \tag{20}$$

with

$$q_3 = 49 + 14c - 1175c^2 + 2352c^{8/3} - 1288c^{10/3} - 16c^{13/3} + 64c^{20/3}$$

and

$$\overline{\mu}_{DCS_2} = \mu + \frac{4c}{1 - 2c + c^4 + \sqrt{q_2}}\,\mu \tag{21}$$

with

$$q_2 = 1 - 12c^2 + 24c^3 - 14c^4 + c^8,$$

respectively.

In the limit as the volume fraction $c \searrow 0$, the effective shear moduli (14) and (20)–(21) reduce asymptotically to

$$\begin{cases} \overline{\mu}_{DS_3} = \mu + \dfrac{5}{2}\mu\, c + 4.38\mu\, c^2 + O(c^3) \\ \overline{\mu}_{DS_2} = \mu + 2\mu\, c + 3.00\mu\, c^2 + O(c^3) \end{cases}$$

and

$$\begin{cases} \overline{\mu}_{DCS_3} = \mu + \dfrac{5}{2}\mu\, c + 2.50\mu\, c^2 + O(c^3) \\ \overline{\mu}_{DCS_2} = \mu + 2\mu\, c + 2.00\mu\, c^2 + O(c^3) \end{cases}. \tag{22}$$

As expected on physical grounds, the coefficients of $O(c^2)$ in both of these results are smaller than the ones in (12) and (13) and, by the same token, in (5) and (6). The quantitative difference is particularly significant for the DCS result (20)–(21), which happens to coincide identically up to and including $O(c^2)$ with the Hashin–Shtrikman (HS) *lower* bound [39,40]

$$\overline{\mu}_{HS_n} = \frac{2 + nc}{2(1-c)}\mu = \mu + \left(1 + \frac{n}{2}\right)\mu \sum_{r=1}^{\infty} c^r. \tag{23}$$

Another significant difference is that the effective moduli (14) and (20)–(21) only percolate at $c = 1$, rather than percolating at $c = p_n$. This is because these results correspond, again, to suspensions wherein the n-spheres are of infinitely many different sizes and spatially distributed in ways in which they can occupy the entire volume of the suspension at hand.

Having pointed to the differences in the dilute and percolation limits, we now turn to comparing the result (10) with (14) and (20)–(21) for volume fractions of n-spheres in the entire range $c \in [0, 1]$. Fig. 4 presents such a comparison for the case of 3-spheres, while Fig. 5 presents the corresponding comparison for 2-spheres. For better visualization of the quantitative differences, parts (b) show the results in the entire range of volume fractions $c \in [0, 1]$, while parts (a) zoom in the range $c \in [0, 0.4]$.

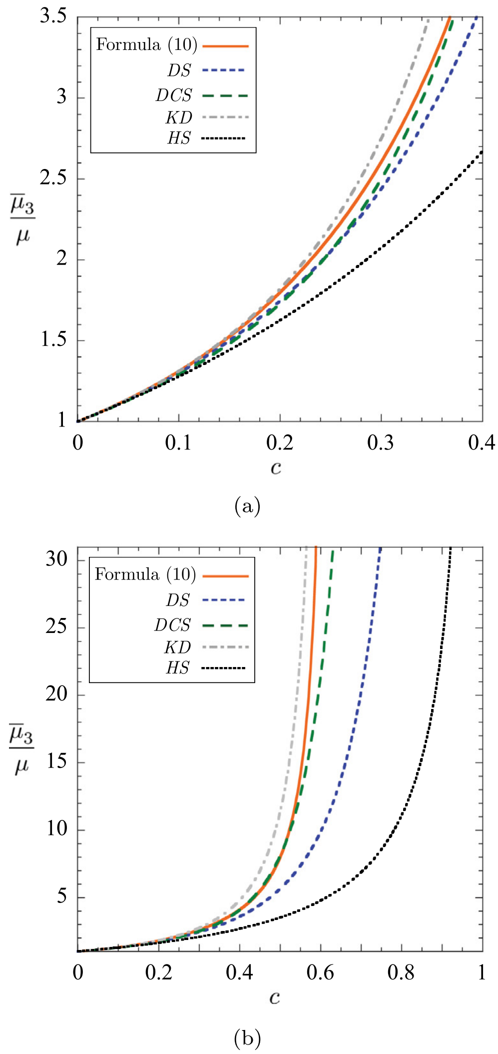

Fig. 4. Comparison of the effective shear modulus (10) for random isotropic suspensions of monodisperse rigid 3-spheres with the corresponding classical differential-scheme (*DS*) result (14) and the differential-coated-sphere (*DCS*) result (20) for suspensions with 3-spheres of infinitely many sizes. The results are shown normalized by the shear modulus of the matrix μ as a function of the volume fraction c of 3-spheres. Part (a) zooms in the small-to-moderate range of c, while part (b) shows the entire range of volume fractions. For further comparison, the Krieger–Dougherty (*KD*) formula (15) and the Hashin–Shtrikman (*HS*) bound (23) are also included in the figures.

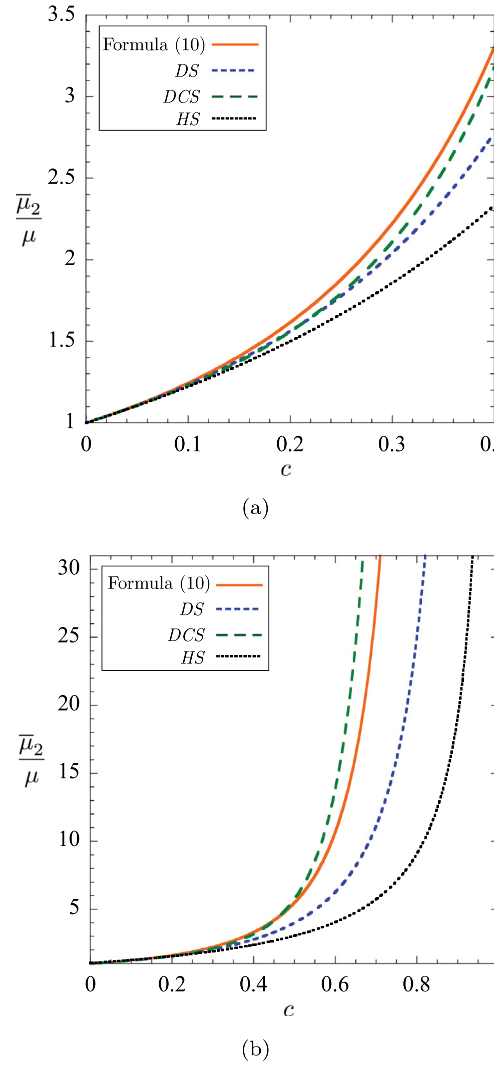

Fig. 5. Comparison of the effective shear modulus (10) for random isotropic suspensions of monodisperse rigid 2-spheres with the corresponding classical differential-scheme (*DS*) result (14) and the differential-coated-sphere (*DCS*) result (21) for suspensions with 2-spheres of infinitely many sizes. The results are shown normalized by the shear modulus of the matrix μ as a function of the volume fraction c of 2-spheres. Part (a) zooms in the small-to-moderate range of c, while part (b) shows the entire range of volume fractions. For further comparison, the Hashin–Shtrikman (*HS*) bound (23) is also included in the figures.

The *DS* result (14) remains within 10% of the formula (10), bounding it from below, up to a volume fraction of around $c = 0.38$ for the case of 3-spheres and of $c = 0.33$ for 2-spheres, indicating that the size polydispersity of the n-spheres does not have a significant effect on the elastic response of the suspensions for small and moderate c. For larger volume fractions, consistent with its larger percolation threshold at $c = 1$, the *DS* result is increasingly softer.

The behavior of the *DCS* result (20)–(21) is more complex. Despite its much softer asymptotic behavior (22) and larger percolation threshold at $c = 1$, it remains within 10% of the formula (10) up to a volume fraction of around $c = 0.55$ for the case of 3-spheres and of $c = 0.53$ for 2-spheres. It does so by intersecting with (10) twice, for 3-spheres, first at around $c = 0.41$ and then at around $c = 0.51$, while for 2-spheres the intersections occur at around $c = 0.46$ and $c = 0.86$.

4.3. Use in comparison-medium methods

Since the pioneering work of Talbot and Willis [41], linear [42–46] and nonlinear [19,24,37,47–49] comparison-medium methods have repeatedly proven extremely powerful to construct approximations for the homogenized response of the nonlinear mechanical and physical properties of composite materials from corresponding homogenization solutions for linear properties. The result (10) proposed here provides a new such homogenization solution of ample practical relevance for use in these methods.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

Support for this work by the National Science Foundation, USA through the Grants CMMI–1661853 and DMREF–1922371 is gratefully acknowledged. V.L. would also like to acknowledge support through the computational resources and staff contributions provided for the Quest high performance computing facility at Northwestern University which is jointly supported by the Office of the Provost, the Office for Research, and Northwestern University Information Technology.

References

[1] H.S.M. Coxeter, Regular Polytopes, Dover, 1973.
[2] A. Einstein, Eine neue bestimmung der moleküldimensionen. [A new determination of molecular dimensions], Ann. Physics 324 (1906) 289–306.
[3] A. Einstein, Berichtigung zu meiner Arbeit: Eine neue bestimmung der moleküldimensionen. [Correction: A new determination of molecular dimensions], Ann. Physics 339 (1911) 591–592.
[4] H.M. Smallwood, Limiting law of the reinforcement of rubber, J. Appl. Phys. 15 (1944) 758–766.
[5] J.D. Eshelby, The determination of the elastic field of an ellipsoidal inclusion and related problems, Proc. R. Soc. Lond. A 241 (1957) 376–396.
[6] S. Torquato, Random Heterogeneous Materials: Microstructure and Macroscopic Properties, Springer, 2002.
[7] J.M. Peterson, M. Fixman, Viscosity of polymer solutions, J. Chem. Phys. 39 (1963) 2516–2523.
[8] G.D. Batchelor, J.T. Green, The determination of the bulk stress in a suspension of spherical particles to order c^2, J. Fluid Mech. 56 (1972) 401–427.
[9] J.R. Willis, J.R. Acton, The overall elastic moduli of a dilute suspension of spheres, Quart. J. Mech. Appl. Math. 29 (1976) 163–177.
[10] J.R. Willis, Relationships between derivations of the overall properties of composites by perturbation expansions and variational principles, in: S. Nemat-Nasser (Ed.), Variational Methods in Mechanics of Solids, Oxford, Pergamon, 1980, pp. 59–66.
[11] H.-S. Chen, A. Acrivos, The effective elastic moduli of composite materials containing spherical inclusions at non-dilute concentrations, Int. J. Solids Struct. 14 (1978) 349–364.
[12] Y. Kantor, D.J. Bergman, Elastostatic resonances — a new approach to the calculation of the effective elastic constants of composites, J. Mech. Phys. Solids 30 (1982) 355–376.
[13] V. Lefèvre, G.A. Francfort, O. Lopez-Pamies, The curious case of 2D isotropic incompressible Neo-Hookean composites, J. Elasticity (2022) (in press).
[14] M. Duerinckx, A. Gloria, On Einstein's effective viscosity formula, 2021, arXiv:2008.03837.
[15] G.D. Scott, Packing of spheres: Packing of equal spheres, Nature 188 (1960) 908–909.
[16] B.D. Lubachevsky, F.H. Stillinger, E.N. Pinson, Disks vs. spheres: Contrasting properties of random packings, J. Stat. Phys. 64 (1991) 501–523.
[17] S. Torquato, F.H. Stillinger, Jammed hard-particle packings: From Kepler to Bernal and beyond, Rev. Modern Phys. 82 (2010) 2633–2672.
[18] A.A. Gusev, Representative volume element size for elastic composites: A numerical study, J. Mech. Phys. Solids 45 (1997) 1449–1459.
[19] O. Lopez-Pamies, T. Goudarzi, K. Danas, The nonlinear elastic response of suspensions of rigid inclusions in rubber: II — A simple explicit approximation for finite concentration suspensions, J. Mech. Phys. Solids 61 (2013) 19–37.
[20] S.A. Spinelli, V. Lefèvre, O. Lopez-Pamies, Dielectric elastomer composites: A general closed-form solution in the small-deformation limit, J. Mech. Phys. Solids 83 (2015) 263–284.
[21] A. Gusev, Controlled accuracy finite element estimates for the effective stiffness of composites with spherical inclusions, Int. J. Solids Struct. 80 (2016) 227–236.
[22] F. de Francqueville, P. Gilormini, J. Diani, Representative volume elements for the simulation of isotropic composites highly filled with monosized spheres, Int. J. Solids Struct. 158 (2019) 277–286.
[23] B. Shrimali, W.J. Parnell, O. Lopez-Pamies, A simple explicit model constructed from a homogenization solution for the large-strain mechanical response of elastomeric syntactic foams, Int. J. Non-Linear Mech. 126 (2020) 103548.
[24] V. Lefèvre, K. Danas, O. Lopez-Pamies, A general result for the magnetoelastic response of isotropic suspensions of iron and ferrofluid particles in rubber, with applications to spherical and cylindrical specimens, J. Mech. Phys. Solids 107 (2017) 343–364.
[25] Lopez. Jimenez, F, Modeling of soft composites under three-dimensional loading, Composites B 59 (2014) 173–180.
[26] V. Lefèvre, The nonlinear elastic response of random isotropic suspensions of rigid n-spheres in Gaussian elastomers, 2022, (in preparation).
[27] G.C. Papanicolaou, S.R.S. Varadhan, Boundary value problems with rapidly oscillating random coefficients, Colloq. Math. Soc. János Bolyai 27 (1981) 835–873.
[28] D.A.G. Bruggeman, Berechnung verschiedener physikalischer konstanten von heterogenen substanzen. I. Dielektrizitätskonstanten und leitfähigkeiten der mischkörper aus isotropen substanzen [Calculation of various physical constants in heterogeneous substances. I. Dielectric constants and conductivity of composites from isotropic substances], Ann. Physics 416 (1935) 636–664.
[29] H.C. Brinkman, The viscosity of concentrated suspensions and solutions, J. Chem. Phys. 20 (1952) 571–573.
[30] R. Roscoe, The viscosity of suspensions of rigid spheres, Br. J. Appl. Phys. 3 (1952) 267–269.
[31] H. Eilers, Die viskosität von emulsionen hochviskoser stoffe als funktion der konzentration. [The viscosity of emulsions of highly viscous substances as a function of concentration], Kolloidn. Zh. 97 (1941) 313–321.
[32] R. Rutgers, Relative viscosity and concentration, Rheol. Acta 2 (1962) 305–348.
[33] J.J. Stickel, R.L. Powell, Fluid mechanics and rheology of dense suspensions, Annu. Rev. Fluid Mech. 37 (2005) 129–149.
[34] I.M. Krieger, T.J. Dougherty, A mechanism for non-Newtonian flow in suspensions of rigid spheres, Trans. Soc. Rheol. 3 (1959) 137–152.
[35] A. Norris, A differential scheme for the effective moduli of composites, Mech. Mater. 4 (1985) 1–16.
[36] M. Avellaneda, Iterated homogenization, differential effective medium theory and applications, Commun. Pur. Appl. Math. 40 (1987) 527–554.
[37] V. Lefèvre, O. Lopez-Pamies, Nonlinear electroelastic deformations of dielectric elastomer composites: I — Ideal elastic dielectrics, J. Mech. Phys. Solids 99 (2017) 409–437.
[38] R.M. Christensen, K.M. Lo, Solutions for effective shear properties in three-phase sphere and cylinder models, J. Mech. Phys. Solids 27 (1979) 315–330.
[39] Z. Hashin, S. Shtrikman, A variational approach to the theory of the elastic behaviour of multiphase materials, J. Mech. Phys. Solids 11 (1963) 127–140.
[40] Z. Hashin, On elastic behaviour of fibre reinforced materials of arbitrary transverse phase geometry, J. Mech. Phys. Solids 13 (1965) 119–134.
[41] D.R.S. Talbot, J.R. Willis, Variational principles for inhomogeneous nonlinear media, IMA J. Appl. Math. 35 (1985) 39–54.
[42] Ponte. Castañeda, P, The effective mechanical properties of nonlinear isotropic composites, J. Mech. Phys. Solids 39 (1991) 45–71.
[43] J.R. Willis, On methods for bounding the overall properties of nonlinear composites, J. Mech. Phys. Solids 39 (1991) 73–86.
[44] D.R.S. Talbot, J.R. Willis, Some simple explicit bounds for the overall behaviour of nonlinear composites, Int. J. Solids Struct. 29 (1992) 1981–1987.
[45] P. Suquet, Overall potentials and extremal surfaces of power law or ideally plastic composites, J. Mech. Phys. Solids 41 (1993) 981–1002.
[46] Ponte. Castañeda, P, Second-order homogenization estimates for nonlinear composites incorporating field fluctuations: I—theory, J. Mech. Phys. Solids 50 (2002) 737–757.
[47] D.R.S. Talbot, J.R. Willis, Upper and lower bounds for the overall properties of a nonlinear composite dielectric. I. Random microgeometry, 447, 1994, pp. 365–384,
[48] G. deBotton, G. Shmuel, A new variational estimate for the effective response of hyperelastic composites, J. Mech. Phys. Solids 58 (2010) 466–483.
[49] V. Lefèvre, O. Lopez-Pamies, Nonlinear electroelastic deformations of dielectric elastomer composites: II — Non-Gaussian elastic dielectrics, J. Mech. Phys. Solids 99 (2017) 438–470.