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ON A STATISTICAL PARAMETER IN THE THEORY OF RANDOM DISPERSIONS OF SPHERES

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Abstract. A two-point statistical parameter which naturally appears in variational bounds in the absorption problem for random media is studied. For random dispersions of nonoverlapping sphere an analytic formula for the parameter is first given through the radial distribution function for the spheres. Analyzing the asymptotic behaviour of the parameter, two kinds of formulae are derived: i) Simple relations between the values of the two-point correlation function and its derivatives at $r = 0$ with the values of radial distribution function and its derivatives at the "touching distance" $r = 2$a. ii) Relations between the moments of the two-point correlation on $(0, \infty)$ and the moments of the radial distribution function. As a simple application, the failure of the well-stirred approximation for sphere fractions higher than 1/8 is finally demonstrated.

1. Introduction

In the theory of random media, when evaluation of their effective macroscopic properties is the aim, the internal random constitution shows up in the final results through certain statistical parameters that incorporate, in an integral form, the multipoint correlations in the media. Presumably the first such parameter appeared in Brown's study [1] of the effective conductivity of weakly inhomogeneous two-phase media. The same Brown's parameter entered later on the well-known variational bounds of Beran [2]. The counterparts of the Beran bounds in the elasticity context and/or Hashin-Shtrikman variational principle involved other and more complicated statistical parameters, see, e.g., the surveys [3,4] for details and references.

If a context, different from conductivity or elasticity, is chosen, different kinds of statistical parameters appear. Consider, for instance, the absorption problem

$$\triangle c(x) - k^2(x)\, c(x) + K = 0,$$

where $c(x)$ is the concentration of a diffusing species absorbed with different rates k_1^2 and k_2^2 in the constituents '1' and '2' respectively of a two-phase random medium (so that $k^2(x)$ is a random field taking the values k_1^2 and k_2^2 depending on whether x lies in '1' or '2'); K is the fixed rate of creation of the species in the bulk of the specimen.

The variational bounds of Beran's type for the effective absorption coefficient of the medium involve, in addition to an integral containing the three-point correlation function in its integrand, the dimensionless two-point statistical parameter (i.e., such in which only two-pair correlation takes part):

$$i_2(p) = p^2 \int_0^\infty r e^{-pr} \gamma_2(r)\, dr, \quad p \in (0, \infty), \tag{1.1}$$

where $\gamma_2(r) = \langle I_1'(0) I_1'(x) \rangle / \eta_1 \eta_2$ is the usual two-point correlation function, $r = |x|$. (The medium is assumed statistically homogeneous and isotropic); the brackets $\langle \cdot \rangle$ signify ensemble averaging. Here $I_1(x)$ is the characteristic function of the region, occupied by the constituent '1' so that $\langle I_1(x) \rangle = \eta_1$, where η_1 is its volume fraction and $\eta_2 = 1 - \eta_1$, $I_1'(x) = I_1(x) - \eta_1$ is the fluctuating part of the field $I_1(x)$. For details we refer the reader to the recent papers [5-7]. Our aim here is to study the parameter (1.1) for random dispersions of equal and nonoverlapping spheres and to extract from its asymptotic behaviour (at $p \to \infty$ and $p \to 0$) certain simple and useful relations and facts concerning this important class of random media.

2. The Evaluation of $i_2(p)$ for Random Dispersions

Hereafter we shall deal with a random dispersion of equal and nonoverlapping spheres. Their centers $\{x_k\}$ form a system of random points, characterized by the usual probability density functions $f_k(y_1, \ldots, y_k)$ [8]. In particular, for the two-point probability density we have $f_2(y_1, y_2) = f_2(r) = n^2 g(r)$, $r = |y_1 - y_2|$, where $g(r)$ is the radial distribution function, $f_1 = n$ is the number density of the spheres, $n = \eta_1 / V_a$, $V_a = \frac{4}{3}\pi a^3$; η_1 is their volume fraction. Then $I_1(x) = \sum_k h_a(x - x_k) = \int h(x-y)\psi(y)\, dy$, where $h_a(x)$ is the characteristic function of a single sphere of radius a, located at the origin, $\psi(x) = \sum_k \delta(x - x_k)$ is the so-called random density field for the dispersion [8], $\delta(x)$ denotes the Dirac delta function and the integration is over the whole R^3. Using the fact that $\langle \psi(y) \rangle = n$, $\langle \psi(y_1)\psi(y_2) \rangle = n\delta(y_1 - y_2) + f_2(y_1, y_2)$, one easily gets the two-point correlation function in the integral form

$$\gamma_2(x) = \frac{1}{\eta_1(1-\eta_1)} \iint h_a(x - y') h_a(y'') \langle \psi'(y') \psi'(y'') \rangle\, dy'\, dy''$$

$$= \frac{1}{\eta_1(1-\eta_1)} \left\{ n \int h_a(x-z) h_a(z)\, dz + n^2 \iint h_a(x-y') h_a(y'') \nu_2(y'-y'')\, dy'\, dy'' \right\}, \tag{2.1}$$

where $\nu_2(z) = g(z) - 1$ is the so-called binary correlation function,

Introducing (2.1) into (1.1) allows to evaluate, after some efforts, the parameter $i_2(p)$. The details of the calculations are given, as a matter of fact in [6]. The final result reads

$$i_2(p) = \frac{A(\tau) - \eta_1 B(\tau)}{1 - \eta_1}, \quad A(\tau) = 1 - 3\frac{1+\tau}{\tau^3} e^{-\tau} (\tau \cosh \tau - \sinh \tau),$$

$$B(\tau) = 1 - \frac{36(\tau \cosh \tau - \sinh \tau)^2}{\tau^4} I, \quad I = I(\tau) = \int_1^\infty s e^{-2s\tau} g(s)\, ds, \tag{2.2}$$

where $s = r/2a$ and $\tau = ap$ is dimensionless and I is the statistical parameter, that appeared in Talbot and Willis' [9] bounds on the effective absorption coefficient of the dispersion.

Hence from Eqs. (1.1) and (2.2) it is clear that the Laplace transforms of the functions $r\gamma_2(r)$ and $sg(s)$ are comparatively simply connected. This fact allows us to find a number of useful relations between the two-point correlation and the radial distribution function for a dispersion.

3. Asymptotics of $i_2(p)$ as $p \to \infty$ and its Consequences

Consider first the quantity $e^{2\tau} I$

$$e^{2\tau} I = \int_1^\infty s e^{-2\tau(s-1)} g(s)\, ds. \tag{3.1}$$

As $p \to \infty$, i.e., $\tau = pa \to \infty$, the function $e^{-2\tau(s-1)}$ tends pointwisely to 0, if $s-1 \geq 0$ and equals 1, if $s - 1 = 0$. Therefore only the behaviour of $g(s)$ around $s = 1$ will matter in the limit $\tau \to \infty$. Let

$$\nu_2(s) = g(s) - 1 = g_0 + g_1(s-1) + g_2(s-1)^2 + \cdots, \quad s \geq 1, \tag{3.2}$$

be the Taylor expansion of the binary correlation at the point $s = r/2a = 1$, i.e., $r = 2a$; the coefficients g_N depend in general on the sphere fraction η_1, $g_N = g_N(\eta_1)$. Obviously

$$g_N = \frac{1}{N!}(2a)^N \nu_2^{(N)}(2a), \tag{3.3}$$

so that knowledge of g_N determines immediately the derivatives of the radial distribution function $g(r)$ at the "touching" distance $r = 2a$.

Inserting (3.2) in (3.1) gives

$$e^{2\tau} I = \sum_{N=0}^{\infty} \frac{G_N}{(2\tau)^{N+1}}, \quad G_N = N!\,(g_{N-1} + g_N) \ \text{ at } \ N \geq 2, \tag{3.4}$$

$G_0 = 1 + g_0$, $G_1 = 1 + g_0 + g_1$. Note that (3.4) holds only asymptotically at $\tau = ap \gg 1$, since the binary correlation $\nu_2(r)$ is not obliged in general to be analytical for all $r \geq 2a$—the series (3.2) may converge to $\nu_2(r)$ only in a vicinity of the point $s = 1$.

Note that the parameter I for the Percus-Yevick (PY) approximation is analytically known due to Wertheim [10] and hence the coefficients G_N can be easily found. In turn, using (3.3) and (3.4), one can obtain the values of the PY radial distribution function and its derivatives at $r = 2a$, in particular,

$$g_0 = g(2a) = \frac{2+\eta_1}{2(1-\eta_1)^2} - 1\,, \quad g_1 = -\frac{9}{2}\,\frac{\eta_1(1+\eta_1)}{(1-\eta_1)^3}\,,$$

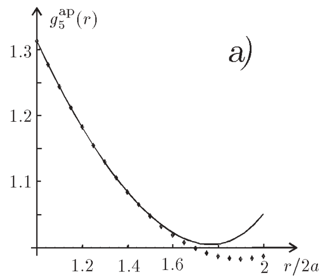
$$g_2 = \frac{3\eta_1(1+2\eta_1)^2}{2(1-\eta_1)^4}\,, \quad g_3 = \frac{\eta_1(1+2\eta_1)^2}{2(1-\eta_1)^4}\,,$$

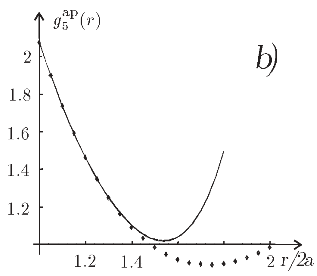
$$g_4 = -\frac{3\eta_1^2(2+\eta_1)^3}{4(1-\eta_1)^6}\,, \quad g_5 = \frac{3\eta_1^2(8+5\eta_1+5\eta_1^2)}{20(1-\eta_1)^7}\,, \tag{3.5}$$

etc. The coefficients g_N at $N \geq 6$ can be also found analytically, using a symbolic algebra package, but their form will be more and more complicated with k increasing.

Note that the first of these values, i.e. $g(2a)$, was pointed out by Lebowitz [11].

An obvious application of the formulae (3.5) consists in an approximate evaluation of the PY function $g(r)$ in a vicinity of the point $r = 2a$. To this end, truncate the series (3.2), say, after the term $g_5(s-1)^5$, use the values of g_k at $k \leq 5$, see Eq. (3.5), and denote the result by $g_5^{\rm ap}(r)$. The function $g_5^{\rm ap}(r)$ is plotted in Fig. 1 for the values $8na^3 = \frac{\pi}{6}\eta_1 = 0.2$, 0.5 and 1, i.e., for sphere fractions $\eta_1 \approx 0.105$, $\eta_1 \approx 0.262$ and $\eta_1 \approx 0.523$; the dots correspond to the numerical solution of the PY equation, due to Throop and Bearman [12]. Obviously, the higher the sphere fraction, the smaller is the region where the approximation $g_5^{\rm ap}(r)$ is useful. Nevertheless, the latter provides a very good fit to the numerical data in the region $2a \leq r \leq 3a$, if $\eta_1 \approx 0.105$, and in the region $2a \leq r \leq 2.5a$, if $\eta_1 \approx 0.524$.





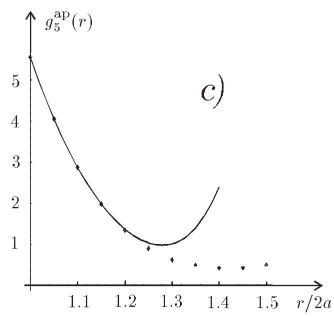


Fig. 1. Plots of the approximation g_5^{ap}. The dots correspond to the Throop and Bearman [12] numerical solution for the PY function. Sphere fractions:
a) $\eta_1 = \pi/20 \approx 0.105$;
b) $\eta_1 = \pi/12 \approx 0.262$;
c) $\eta_1 = \pi/6 \approx 0.523$.

In the well-stirred case $g(r) = 1$ at $r \geq 2a$, so that $g_N = 0$, $\forall N$, and (3.4) yields

$$I = I^{\rm ws} = \frac{1+2\tau}{4\tau^2}\, e^{-2\tau}, \quad \tau \in (0, \infty), \tag{3.6}$$

which can be directly obtained from (3.1) by elementary integration.

Assume that $i_2(p)$ admits the expansion

$$i_2(p) = 1 + \frac{C_1}{p} + \frac{C_2}{p^2} + \cdots + \varepsilon(\tau), \quad \tau = ap \gg 1, \tag{3.7}$$

$C_0 = 1$. Then

$$\gamma_2^{(N)}(0) = \frac{C_N}{N+1} = \frac{1}{N+1} \lim_{p\to\infty} p^N \left[i_2(p) - \sum_{j=0}^{N-1} \frac{C_j}{p^j} \right], \tag{3.8}$$

$N = 0, 1, \ldots$, which easily follows from the definition (1.1) of $i_2(p)$ and the well-known properties of the Laplace transform. Hereafter $\varepsilon(\tau)$ denotes terms that decrease exponentially as $\tau \to \infty$.

To find the coefficients C_j in the expansion (3.7), note first that

$$A(\tau) = 1 - \frac{3}{2}\left(\frac{1}{\tau} - \frac{1}{\tau^3}\right) + \varepsilon(\tau), \quad \tau \gg 1, \tag{3.9}$$

and rewrite next the coefficient B, see (2.2), in the form

$$B(\tau) = 1 - F(\tau)(e^{2\tau} I),$$

$$F(\tau) = \frac{36(\tau\cosh\tau - \sinh\tau)^2}{\tau^4}\, e^{-2\tau} = 9\,\frac{(\tau-1)^2}{\tau^4} + \varepsilon(\tau), \quad \tau \gg 1. \tag{3.10}$$

The asymptotic expansion of $e^{2\tau} I$ is given in (3.4). Combining the latter with (3.9) and (3.10) and inserting the result into the formula (1.1) for the statistical parameter $i_2(p)$ gives after some algebra

$$i_2(p) = 1 - \frac{3}{2(1-\eta_1)}\frac{1}{\tau} + \frac{3(1+3\eta_1+3g_0\eta_1)}{2(1-\eta_1)}\frac{1}{\tau^3} + \sum_{N=4}^{\infty} \frac{T_N}{\tau^N} + \varepsilon(\tau), \quad \tau \gg 1, \tag{3.11}$$

with the coefficients

$$T_N = \frac{9\eta_1}{2^{N-2}(1-\eta_1)}\Big(G_{N-3} - 4G_{N-4} + 4G_{N-5}\Big), \tag{3.12}$$

$N = 4, 5, \ldots$, assuming $G_j = 0$ at $j < 0$. Using in turn the formula for G_N, see (3.4), gives

$$T_N = \frac{9\eta_1}{2^{N-2}(1-\eta_1)}\Big((N-3)!\, g_{N-3} + (N-7)(N-4)!\, g_{N-4}$$
$$-4(N-5)(N-5)!\, g_{N-5} + 4(N-5)!\, g_{N-6}\Big). \tag{3.13}$$

From (3.8) and (3.11) one finds, first of all,

$$\gamma_2'(0) = -\frac{3}{4(1-\eta_1)a}, \quad \gamma_2''(0) = 0. \tag{3.14}$$

The first of Eqs (3.14) is a simple consequence of Debye *et al.* formula [13], which connects the specific surface of a two-phase material with $\gamma_2'(0)$. Thus for any dispersion of *nonoverlapping* spheres $\gamma_2'(0)$ is not sensitive to the sphere statistics depending, at a fixed radius a, on the sphere fraction η_1 only. A stronger and more curious fact is embodied into the second relation of (3.14), namely, the vanishing at the origin of the second derivative of the correlation function for such dispersion whatever be the sphere statistics. The assumed spherical shape of the particles is not important here; the fact that $\gamma_2''(0) = 0$ is essentially connected with the assumption of nonoverlapping. Indeed, consider a statistically isotropic dispersion of particles of fixed shape, whose location and orientation are both random but not interconnected statistically; the particles should not overlap whatever their orientations at fixed locations. An averaging with respect to orientation first (which is possible, due to the statistical independence of the latter), leads just to a dispersion of nonoverlapping spheres. Each one is obtained through rotation of the particle, centered at the same location; the rotation represents simply the averaging with respect to all possible orientations of the particle. Note that the fact that $\gamma_2''(0) = 0$ for a dispersion of nonoverlapping particles was first noticed by Kirste and Porod [14] using different and more complicated geometrical arguments; they also assumed that there are no corner points on the particle's surfaces. This assumption is not necessary, as easily seen from the foregoing reasoning. The results of Kirste and Porod were rederived and extended by Frisch and Stillinger [15] who expanded directly the two-point correlation function $\gamma_2(r)$ at $r = 0$ starting, as a matter of fact, with its integral representation (2.1).

According to (3.12), the statistics of the dispersion, that is, the radial distribution function, shows up only in the derivatives $\gamma_2^{(N)}$ at $N \geq 3$. Indeed, from (3.12) and (3.13) it follows

$$\gamma_2'''(0) = \frac{3(1 + 3\eta_1 + 3g_0\eta_1)}{8(1-\eta_1)a^3}\,, \quad \gamma_2^{(4)}(0) = \frac{9(g_1 - 3(1+g_0))\eta_1}{20(1-\eta_1)a^4}\,,$$

$$\gamma_2^{(5)}(0) = \frac{3(g_2 - g_1)\eta_1}{8(1-\eta_1)a^5}\,, \quad \gamma_2^{(6)}(0) = \frac{9(3g_3 - g_2 - 2g_1 + 2(1+g_0))\eta_1}{56(1-\eta_1)a^6}\,, \tag{3.15}$$

and, in general,

$$\gamma_2^{(N)}(0) = \frac{T_N}{(N+1)a^N}\,, \quad N = 7, 8, \ldots\,, \tag{3.16}$$

where T_N is expressed in (3.13) by the coefficients g_{N-3}, g_{N-4}, g_{N-5} and g_{N-6}, connected with the local behaviour of the binary correlation $\nu_2(r)$ at the "touching" distance $r = 2a$. Note that the first of expressions (3.15)—the value of $\gamma_2'''(0)$—coincides with that given by Kirste and Porod [14] and Frisch and Stillinger [15].

In the well-stirred case all g_N vanish. From (3.13), (3.15) and (3.16) one finds the needed values of $\gamma_2'''(0)$, $\gamma_2^{(4)}(0)$ and $\gamma_2^{(6)}(0)$; all the rest of the derivatives $\gamma_2^{(N)}(0) = 0$ at $N = 4$ and $N \geq 7$ vanish in this case. Thus in a certain vicinity of the origin the two-point correlation function of the well-stirred dispersion is the polynomial

$$\gamma_2(r) = 1 - \frac{3}{4(1-\eta_1)}\,\frac{r}{a} + \frac{1+3\eta_1}{16(1-\eta_1)}\left(\frac{r}{a}\right)^3$$

$$-\frac{9\eta_1}{160(1-\eta_1)}\left(\frac{r}{a}\right)^4+\frac{\eta_1}{2240(1-\eta_1)}\left(\frac{r}{a}\right)^6. \tag{3.17}$$

Note that the function $\gamma_2(r)$ should vanish at $r = 4a$ in the well-stirred case under study. The polynomial (3.17) does not possess this property which means that $\gamma_2(r)$ is not analytical on the whole semiaxis $(0,\infty)$ and hence (3.17) holds in a certain vicinity of the origin $r = 0$. Indeed, a direct analytical computation, details of which will be reported elsewhere, shows that (3.17) holds only at $r \leq 2a$. In the point $r = 2a$, $\gamma_2^{(4)}(r)$ is discontinuous. It is to be mentioned that the above computation allows us to claim that the general formula for $\gamma_2^{(4)}(0)$, given by Frisch and Stillinger [15], is not correct, since in the well-stirred case it does not yield the respective value in (3.17).

4. Asymptotics of $i_2(p)$ as $p \longrightarrow 0$ and its Consequences

Note immediately that at small $p \ll 1$:

$$i_2(p)=\theta_1\tau^2-\theta_2\tau^3+\cdots=\tau^2\sum_{N=0}^{\infty}\frac{(-1)^N}{N!}\theta_{n+1}\tau^N, \tag{4.1}$$

$$\theta_N=\int_0^{\infty}t^N\gamma_2(r/a)\,dt, \quad t=r/a, \tag{4.2}$$

so that θ_N are the moments of the correlation function $\gamma_2(r)$ on the semiaxis $(0,\infty)$. To connect these moments with the appropriate moments of the binary correlation note first that

$$I=I(\tau)=\int_1^{\infty}se^{-2\tau s}g(s)\,ds=\frac{1+2\tau}{4\tau^2}e^{-2\tau}+\int_1^{\infty}se^{-2\tau s}\nu_2(s)\,ds, \tag{4.3}$$

using the definition $\nu_2(s) = g(s) - 1$ of the binary correlation, $s = r/2a$. The first term in the right side of (4.3) is just the parameter $I = I^{\mathrm{ws}}$ in the well-stirred case, already known, see (3.6).

Expand next $I(\tau)$, as given in (4.3), around $\tau = 0$:

$$\begin{aligned}I(\tau)&=\frac{1+2\tau}{4\tau^2}\left(1-2\tau+\frac{(2\tau)^2}{2!}-\cdots\right)+\left(m_1-2\tau m_2+\frac{(2\tau)^2}{2!}m_3-\cdots\right)\\&=\frac{1}{4\tau^2}\left(1+\sum_{N=2}^{\infty}\frac{(-1)^{N-1}(1-Nm_{N-1})}{N\,(N-2)!}(2\tau)^N\right)\\&=\frac{1}{4\tau^2}\left(1-2(1-2m_1)\tau^2+\frac{8}{3}(1-3m_2)\tau^3-2(1-4m_3)\tau^4+\cdots\right),\end{aligned} \tag{4.4}$$

where

$$m_l=\int_1^{\infty}s^l\,\nu_2(s)\,ds, \quad s=r/2a, \tag{4.5}$$

$l = 0, 1, \ldots$, are the moments of the binary correlation on the semiaxis $(1,\infty)$.

Insert the series (4.4) for I into the formula (1.1) for the statistical parameter $i_2(p)$, and use that

$$A(\tau) = 12\tau^2 \sum_{N=0}^{\infty} \frac{(-1)^N (N+1)(N+4)}{(N+5)!} (2\tau)^N = \tau^2 \left(\frac{2}{5} - \frac{1}{3}\tau + \frac{6}{35}\tau^2 - \cdots \right),$$

$$F(\tau)e^{2\tau} = 9 \sum_{N=0}^{\infty} \frac{(N+1)(2N+5)\, 2^{2(N+3)}}{(2(N+3))!} (2\tau)^N = 4\tau^2 \left(1 + \frac{1}{5}\tau^2 + \frac{3}{175}\tau^4 + \cdots \right),$$

see (2.2) and (3.10). Then

$$i_2(p) = \tau^2 \left[\frac{2/5 - \eta_1(9/5 - 4m_1)}{5(1-\eta_1)} - \frac{1 - 8\eta_1(1-3m_2)}{3(1-\eta_1)}\tau + \cdots \right],$$

which, when compared to (4.1), gives the interconnection between the moments θ_N of the two-point correlation $\gamma_2(r)$ and the moments m_l of the binary correlation $\nu_2(r)$ for a dispersion of nonoverlapping spheres. In particular,

$$\theta_1 = \frac{2 - \eta_1(9 - 20m_1)}{5(1-\eta_1)}, \quad \theta_2 = \frac{1 - 8\eta_1(1-3m_2)}{3(1-\eta_1)}, \quad \text{etc.} \tag{4.6}$$

The formulae (4.6) are very convenient, if the binary correlation is given analytically. For instance, in the well stirred case $\nu_2(r) = 0$ at $r \geq 2a$, so that all the moments m_l vanish and hence, in particular,

$$\theta_1 = \frac{2 - 9\eta_1}{5(1-\eta_1)}, \quad \theta_2 = \frac{1 - 8\eta_1}{3(1-\eta_1)}, \tag{4.7}$$

in this case.

Note that for any statistically homogeneous and isotropic random medium the moments θ_1 and θ_2 should be nonnegative. (As a matter of fact, this follows from the Bochner-Khinchine theorem which states that the two-point correlation function $\gamma_2(r)$ should be positive-definite for such media [16,17].) An elementary proof of this fact consists in introducing the random fields

$$\chi(x) = \int \frac{1}{4\pi|x-y|} I_1'(y)\, dy, \quad \phi(x) = \int I_1'(x-y)\, dy,$$

and noting that $\langle |\nabla\chi|^2 \rangle \geq 0$ and $\langle \phi^2 \rangle \geq 0$.

The nonnegativeness of θ_1 and θ_2 imposes, through Eq. (4.6), restrictions on the moments m_1 and m_2 of the binary correlation for any realistic dispersion of spheres, namely,

$$m_1 \geq \frac{9\eta_1 - 2}{20\eta_1}, \quad m_2 \geq \frac{8\eta_1 - 1}{24\eta_1}. \tag{4.8}$$

Hence the well-stirred approximation, for which $m_1 = m_2 = 0$, is realistic only at $\eta_1 \leq 1/8$—something conjectured by Willis [18], who noticed that a certain well-known scheme of mechanics of composites in the wave propagation context yields unrealistic predictions for this approximation, if $\eta_1 > 1/8$.

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