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ON THE ABSORPTION COEFFICIENT OF RANDOM DISPERSIONS

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Михаил Колев, Константин Марков. О КОЭФФИЦИЕНТЕ АБСОРБЦИИ СЛУЧАЙНОЙ ДИСПЕРСИИ СФЕР

Работа посвящена задаче определения эффективного коэффициента абсорбции сред случайной структуры. Вариационные оценки этого коэффициента, недавно предложенные авторами, вычислены явно для случайной суспенсии сфер до порядка „квадрата концентрации" и сравнены с оценками Талбота и Уиллиса. Оказывается, что оценки авторов ўже, однако они, в отличии от оценок Талбота и Уилиса, применимы лишь для концентрации сфер, не превошодящих 0.10.

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The problem of predicting the effective absorption coefficient of random media is discussed. The variational estimates on this coefficient, recently derived by the authors, are explicitly evaluated for random dispersion of spheres to the order "square of concentration". A comparison with the bounds of Talbot and Willis is performed as well. It appears that the proposed bounds are more restrictive but, unlike those of Talbot and Willis, are only applicable for sphere concentrations that do not exceed 0.10.

1. INTRODUCTION

Consider the steady-state equation

$$\Delta\varphi(\mathbf{x}) - k^2(\mathbf{x})\varphi(\mathbf{x}) + K = 0 \tag{1.1}$$

that governs, at the expense of some simplifying assumptions, the concentration

$\varphi(\mathbf{x})$ of a diffusing species (say, irradiation defects), generated at the constant rate K, in a random absorbing (lossy) medium (see [1] for references and more details). The absorption coefficient $k^2(\mathbf{x})$ is a given random field, assumed positive, statistically homogeneous and isotropic. The problem is to evaluate the random field $\varphi(\mathbf{x})$, i.e. all its multipoint correlations, and, in particular, to find the mean defect concentration $\langle\varphi(\mathbf{x})\rangle$; the brackets $\langle\cdot\rangle$ hereafter denote ensemble averaging. The latter value allows to obtain the effective absorption coefficient (sink strength) k^{*2} of the medium, defined by the relation $k^{*2}\langle\varphi(\mathbf{x})\rangle = K$.

Recently the authors have proposed variational estimates on the coefficient k^{*2}, using the technique of truncated functional series and a procedure of Beran's type [2]. We shall recall now these bounds in the particular case of a two-phase medium. Having in mind the application to particulate media and dispersions of spheres in particular, we call one of the constituents, for definiteness sake, filler and denote its absorption coefficient by k_f^2 and its volume fraction — by $c_f = c$; the other constituent is called then matrix and its respective parameters are k_m^2 and $c_m = 1 - c$. Thus the random absorption field of the medium is

$$k^2(\mathbf{x}) = \begin{cases} k_m^2, & \text{if } \mathbf{x} \in \text{matrix}, \\ k_f^2, & \text{if } \mathbf{x} \in \text{filler}, \end{cases}$$

or

$$k^2(\mathbf{x}) = k_m^2 + [k^2]I_f(\mathbf{x}) = \langle k^2\rangle + [k^2]I'_f(\mathbf{x}), \tag{1.2}$$

where $[k^2] = k_f^2 - k_m^2$, $I_f(\mathbf{x})$ is the characteristic function of the region, occupied by the filler, and $I'_f(\mathbf{x}) = I_f(\mathbf{x}) - c$ is its fluctuating part.

The elementary (one-point) bounds on k^{*2} read

$$k_R^2 \leqq k^{*2} \leqq k_V^2, \tag{1.3}$$

where

$$k_V^2 = \langle k^2(\mathbf{x})\rangle = ck_f^2 + (1-c)k_m^2,$$

$$k_R^2 = \frac{1}{\langle\alpha^2(\mathbf{x})\rangle} = (c\alpha_f^2 + (1-c)\alpha_m^2)^{-1};$$

here $\alpha^2(\mathbf{x}) = 1/k^2(\mathbf{x})$ is the compliance field for the medium. The bounds (1.3) are the obvious counterparts of the well-known Voigt and Reuss estimates on the effective conductivity or elastic moduli of a heterogeneous medium.

The bounds on k^{*2}, announced in [3] and detailed in [4], are already three-point and thus they are always tighter than the elementary ones (1.3). The bounds have the form

$$k_R^2\left\{1 - \frac{[\alpha^2]^2}{\langle\alpha^2\rangle^2}\frac{c(1-c)(I_2^\alpha)^2}{I_2^\alpha + \dfrac{[\alpha^2]}{\langle\alpha^2\rangle}(1-2c)I_3^\alpha}\right\}^{-1}$$

$$\leqq k^{*2} \leqq k_V^2\left(1 - \frac{[k^2]^2}{\langle k^2\rangle^2}\frac{c(1-c)(I_2^k)^2}{I_2^k + \dfrac{[k^2]}{\langle k^2\rangle}(1-2c)I_3^k}\right). \tag{1.4}$$

The following four statistical parameters enter the bounds:

$$(1.5)\qquad I_2^k = \frac{k_V^2}{M_2^k(0)}\int G_V(\mathbf{y})M_2^k(\mathbf{y})\,d^3\mathbf{y},\quad I_2^\alpha = -\frac{1}{M_2^\alpha(0)}\int \Delta G_R(\mathbf{y})M_2^\alpha(\mathbf{y})\,d^3\mathbf{y},$$

$$I_3^k = \frac{k_V^4}{M_3^k(0,0)}\int\int G_V(\mathbf{y}_1)G_V(\mathbf{y}_2)M_3^k(\mathbf{y}_1,\mathbf{y}_2)\,d^3\mathbf{y}_1 d^3\mathbf{y}_2,$$

$$(1.6)$$

$$I_3^\alpha = \frac{1}{M_3^\alpha(0,0)}\int\int \Delta G_R(\mathbf{y}_1)\Delta G_R(\mathbf{y}_2)M_3^\alpha(\mathbf{y}_1,\mathbf{y}_2)\,d^3\mathbf{y}_1 d^3\mathbf{y}_2.$$

Here $G_V(\mathbf{x}) = \dfrac{1}{4\pi|\mathbf{x}|}\exp(-k_V|\mathbf{x}|)$ is the Green function of the operator $\Delta - k_V^2$, i.e.

$$(1.7)\qquad \Delta G_V(\mathbf{x}) - k_V^2 G_V(\mathbf{x}) + \delta(\mathbf{x}) = 0,$$

and similarly for $G_R(\mathbf{x})$ with k_V replaced by k_R;

$$(1.8)\qquad M_2^k(\mathbf{y}) = \langle \delta k^2(\mathbf{0})\delta k^2(\mathbf{y})\rangle,\ M_3^k(\mathbf{y}_1,\mathbf{y}_2) = \langle \delta k^2(\mathbf{0})\delta k^2(\mathbf{y}_1)\delta k^2(\mathbf{y}_2)\rangle$$

are, respectively, the two- and three-point correlation functions for the field $k^2(\mathbf{x})$. The same functions for the compliance field $\alpha^2(\mathbf{x})$ are denoted by $M_2^\alpha(\mathbf{y})$ and $M_3^\alpha(\mathbf{y}_1,\mathbf{y}_2)$. Since the medium is two-phase, we have the well-known relations

$$(1.9)\qquad M_2^k(\mathbf{0}) = c(1-c)[k^2]^2,\quad M_3^k(\mathbf{0},\mathbf{0}) = c(1-c)(1-2c)[k^2]^3,$$

and similarly for $M_2^\alpha(\mathbf{0})$ and $M_3^\alpha(\mathbf{0},\mathbf{0})$. Hereafter the integrals are over the whole space $\mathbb{R}^3$, if the integration domain is not explicitly indicated.

Note that in [4] it was shown, in particular, that the bounds (1.4) are third-order in the weakly-inhomogeneous case. Moreover, the explicit results, obtained in [4] for Miller's cellular media, indicate that the bounds remain useful even when the absorption capabilities of the constituents differ one hundred times.

In this paper we shall consider in detail the evaluation of the statistical parameters (1.5) and (1.6) for random dispersions of nonoverlapping spheres. In Section 2 we briefly summarize the needed in the sequel statistical description of random dispersions. In Section 3 we calculate the parameters I_2^k and I_2^α for the dispersion, given in (1.5), that depend on the two-point correlations. Similar calculations are performed in Section 4 for the parameters I_3^k and I_3^α, see (1.6), but unlike the "two-point" parameters, we are able to give analytical results correct to the asymptotic order c^2 only. In Section 5 we illustrate the performance of the bounds and compare them with those of Talbot and Willis [1].

2. STATISTICAL DESCRIPTION OF RANDOM DISPERSIONS

We consider a random dispersion of spheres, i.e. an unbounded matrix, containing an array of equal and nonoverlapping spherical inhomogeneities, each one of radius a. The medium is thus completely described by the system of random points $\{\mathbf{x}_\alpha\}$ — the centers of the spheres. The statistics of the system $\mathbf{x}_\alpha$ is conveniently

represented by the multipoint distribution densities $f_p(\mathbf{y}_1, \ldots, \mathbf{y}_p)$, or probability density functions. They define the probability dP to simultaneously find a point of the random set $\{\mathbf{x}\}_\alpha$ per each of the infinitesimal volumes $\mathbf{y}_i < \mathbf{y} < \mathbf{y}_i + d\mathbf{y}_i$, $i = 1, \ldots, p$, to be

$$dP = f_p(\mathbf{y}_1, \ldots, \mathbf{y}_p)\, d^3\mathbf{y}_1 \ldots d^3\mathbf{y}_p. \tag{2.1}$$

We assume that the system $\{\mathbf{x}_\alpha\}$ is statistically isotropic and homogeneous; then, in particular, $f_1 = n$ and $f_p = f_p(\mathbf{y}_{2,1}, \ldots, \mathbf{y}_{p,1})$, where $\mathbf{y}_{j,i} = \mathbf{y}_j - \mathbf{y}_i$ and n denotes the number density, i.e. the mean number of points per unit volume.

Let us imagine now that by means of a certain manufacturing process we produce random point systems $\{\mathbf{x}\}_\alpha$ with different number densities n. The statistics of the system $\{\mathbf{x}\}_\alpha$ will then depend on n as a parameter, i.e. $f_p = f_p(\mathbf{Y}_p; n)$, $\mathbf{Y}_p = (\mathbf{y}_1, \ldots, \mathbf{y}_p)$. We shall assume, as usual, that $f_p \sim n^p$, i.e. f_p has the asymptotic order n^p at $n \to 0$, $p = 1, 2, \ldots$ In particular, for the two-point distribution density f_2, which most frequently appears in models and theoretical studies, we have

$$f_2(\mathbf{y}_1, \mathbf{y}_2) = n^2 g(r), \quad g(r) = g_0(r) + O(n), \tag{2.2}$$

$r = |\mathbf{y}_2 - \mathbf{y}_1|$. (The point system $\{\mathbf{x}\}_\alpha$ hereafter will be assumed statistically isotropic as well.) Thus $g_0(r)$ is the zero-density limit of the radial distribution function $g(r)$ for the system $\{\mathbf{x}\}_\alpha$.

A convenient characteristics of the set of random points is the so-called random density field

$$\psi(\mathbf{x}) = \sum_\alpha \delta(\mathbf{x} - \mathbf{x}_\alpha). \tag{2.3}$$

This field was systematically used by Stratonovich [5] in the one-dimensional case when the role of $\mathbf{x}$ is played by the time. The random function $\psi(\mathbf{x})$ is uniquely defined by the random set $\mathbf{x}_\alpha$. The respective formulas [5] read:

$$\begin{aligned}
\langle \psi(\mathbf{y}) \rangle &= f_1(\mathbf{y}) = n, \\
\langle \psi(\mathbf{y}_1)\psi(\mathbf{y}_2) \rangle &= f_1(\mathbf{y}_1)\delta(\mathbf{y}_{1,2}) + f_2(\mathbf{y}_1, \mathbf{y}_2), \\
\langle \psi(\mathbf{y}_1)\psi(\mathbf{y}_2)\psi(\mathbf{y}_3) \rangle &= f_1(\mathbf{y}_1)\delta(\mathbf{y}_{1,2})\delta(\mathbf{y}_{1,3}) \\
&\quad + 3\{\delta(\mathbf{y}_{1,2}) f_2(\mathbf{y}_{1,3})\}_s + f_3(\mathbf{y}_1, \mathbf{y}_2, \mathbf{y}_3),
\end{aligned} \tag{2.4}$$

etc., where $\{\cdot\}_s$ means symmetrization with respect to all different combinations of indices in the braces.

The random absorption field (1.2) of the medium under study has a simple integral representation by means of the field $\psi(\mathbf{x})$, namely

$$k^2(\mathbf{x}) = \langle k^2 \rangle + [k^2] \int h(\mathbf{x} - \mathbf{y}) \psi'(\mathbf{y})\, d^3\mathbf{y}, \tag{2.5}$$

where $\psi'(\mathbf{y}) = \psi(\mathbf{y}) - n$ is the fluctuating part of the field $\psi(\mathbf{y})$, $h(\mathbf{y})$ is the characteristic function of a single sphere of radius a, located at the origin.

3. EVALUATION OF THE PARAMETERS I_2^k AND I_2^α

According to (1.2), (2.4) and (2.5), the parameter I_2^k for the dispersion has the form

$$I_2^k = \frac{k_V^2}{c(1-c)} \int\int\int G_V(\mathbf{y}) h(\mathbf{z}_1) h(\mathbf{y}-\mathbf{z}_2) \langle \psi'(\mathbf{z}_1)\psi'(\mathbf{z}_2) \rangle \, d^3\mathbf{y} d^3\mathbf{z}_1 d^3\mathbf{z}_2$$

$$= \frac{k_V^2}{c(1-c)} \int\int h(\mathbf{z}_1)\chi_V(\mathbf{z}_2) \left[n\delta(\mathbf{z}_1-\mathbf{z}_2) - n^2 R(\mathbf{z}_1-\mathbf{z}_2) \right] d^3\mathbf{z}_1 d^3\mathbf{z}_2 \tag{3.1}$$

$$= \frac{a_0 - a_1 c}{1-c}; \quad R(\mathbf{z}) = 1 - g(\mathbf{z}),$$

with the coefficients

$$a_0 = \frac{k_V^2}{V_a} \int h(\mathbf{z}) \chi_V(\mathbf{z}) \, d^3\mathbf{z}, \tag{3.2}$$

$$a_1 = \frac{k_V^2}{V_a^2} \int\int h(\mathbf{z}_1)\chi_V(\mathbf{z}_2) R(\mathbf{z}_1-\mathbf{z}_2) \, d^3\mathbf{z}_1 d^3\mathbf{z}_2. \tag{3.3}$$

Here $V_a = \frac{4}{3}\pi a^3$ and

$$\chi_V(\mathbf{z}) = (h * G_V)(\mathbf{z}) \tag{3.4}$$

is the Helmholtz potential for a single sphere of radius a, located at the origin. Let us recall that it is the continuous and bounded everywhere solution of the Helmholtz equation $\Delta\chi_V - k_V^2\chi_V + h(\mathbf{z}) = 0$. A simple calculation yields

$$\chi_V(\mathbf{z}) = \frac{1}{k_V^2} \begin{cases} A' \dfrac{a_V \sinh r_V}{r_V \sinh a_V} + 1, & r < a, \\ A'' \dfrac{a_V}{r_V} e^{a_V - r_V}, & r \geqq a, \end{cases} \tag{3.5a}$$

$$A' = -\frac{1+a_V}{a_V} e^{-a_V} \sinh a_V, \quad A'' = \frac{1}{a_V} e^{-a_V}(a_V \cosh a_V - \sinh a_V), \tag{3.5b}$$

where $a_V = ak_V$ and $r_V = rk_V$ are dimensionless, $r = |\mathbf{z}|$.

Using (3.5), we find first the coefficient a_0:

$$a_0 = 1 - F_2(a_V), \tag{3.6}$$

where

$$F_2(x) = 3\frac{1+x}{x^3} e^{-x}(x\cosh x - \sinh x) \tag{3.7}$$

is the function that appeared when calculating the parameters I_2^k and I_2^α for cellular media with spherical shape of the cells, see [3,4].

For the coefficient a_1 we get in turn:

$$a_1 = \frac{k_V^2}{V_a^2} \int h(\mathbf{z}) P(\mathbf{z}) \, d^3\mathbf{z}, \tag{3.8}$$

where P denotes the convolution $P(\mathbf{z}) = (\chi_V * R)(\mathbf{z})$. Due to (1.7), the function P solves the equation

$$\Delta P - k_V^2 P + h * R = 0. \tag{3.9}$$

The assumption of nonoverlapping yields $g(\mathbf{z}) = 0$ and thus $R(\mathbf{z}) = 1 - g(\mathbf{z}) = 1$ at $|\mathbf{z}| \leqq 2a$. That is why $(h * R)(\mathbf{z}) = V_a$ within the sphere $|\mathbf{z}| \leqq a$ and the solution of eqn (3.9) within the same sphere has therefore the form

$$P(\mathbf{z}) = \frac{V_a}{k_V^2}\left(1 + B\frac{a_V \sinh r_V}{r_V \sinh a_V}\right), \quad r = |\mathbf{z}| < a. \tag{3.10}$$

The unknown constant B is found by means of the obvious relation

$$P(\mathbf{0}) = \frac{V_a}{k_V^2}\left(1 + B\frac{a_V}{\sinh a_V}\right) = \int \chi_V(\mathbf{z}) R(\mathbf{z})\, d^3\mathbf{z},$$

or

$$B = \frac{\sinh a_V}{a_V}\left[\frac{k_V^2}{V_a}\int \chi_V(\mathbf{z})(1 - g(\mathbf{z}))\, d^3\mathbf{z} - 1\right]. \tag{3.11}$$

Simple calculations, using (3.5), yield eventually

$$a_1 = 1 - \frac{4a_V^2 e^{2a_V}}{(1 + a_V)^2} F_2^2(a_V) I, \tag{3.12}$$

where

$$I = \int_1^\infty s e^{-2a_V s} g(s)\, ds, \quad s = r/2a, \tag{3.13}$$

is the statistical parameter that appeared in Talbot and Willis' bounds on the effective absorption coefficient k^{*2} [1].

In the simplest two-point statistics — the so-called "well-stirred" case — one has $g(s) = 1$ at $s \geqq 1$, so that

$$I = I^{ws}(a_V) = \frac{1 + 2a_V}{4a_V^2} e^{-2a_V}, \tag{3.14}$$

and thus

$$a_1 = 1 - \frac{1 + 2a_V}{(1 + a_V)^2} F_2^2(a_V). \tag{3.15}$$

Note that Talbot and Willis were able also to evaluate the parameter I in the case when the two-point statistics of the dispersion is governed by the well-known Percus-Yevick approximation:

$$I = I^{py}(a_V) = G(2a_V),$$

$$G(t) = \frac{tL(t)}{12c[L(t) + S(t)e^t]}, \quad L(t) = 12c\left[\left(1 + \frac{1}{2}c\right)t + 1 + 2c\right], \tag{3.16}$$

$$S(t) = (1 - c)^2 t^3 + 6c(1 - c)t^2 + 18c^2 t - 12c(1 + 2c).$$

A simple check shows that

$$I^{py}(a_V) = I^{ws}(a_V) + O(c),$$

as it should be.

Thus the needed statistical parameter I_2^k for the dispersion is

$$I_2^k = \frac{a_0 - a_1 c}{1 - c} = \varphi_2(a_V), \tag{3.17}$$

where a_0, a_1 are explicitly given in (3.6), (3.12) respectively. Hence φ_2 is a known function of the dimensionless parameter a_V, depending on the radial distribution function $g(r)$ for the dispersion through the statistical parameter I.

The evaluation of the second statistical parameter I_2^α, as given in (1.5), is now straightforward. Keeping in mind (1.7), we get immediately

$$I_2^\alpha = -\frac{1}{M_2^\alpha(0)} \int \Delta G_R(\mathbf{y}) M_2^\alpha(\mathbf{y})\, d^3\mathbf{y}$$

$$= 1 - \frac{k_R^2}{M_2^\alpha(0)} \int G_R(\mathbf{y}) M_2^\alpha(\mathbf{y})\, d^3\mathbf{y},$$

so that

$$I_2^\alpha = 1 - \varphi_2(a_R), \tag{3.18}$$

where $\varphi_2(a_R)$ is the function, defined in (3.17), in which a_V should be replaced everywhere by a_R.

4. EVALUATION OF THE PARAMETERS I_3^k AND I_3^α

Unlike I_2^k and I_2^α, we are able to evaluate the three-point parameters I_3^k and I_3^α to the order c^2 only. The reason is that the three-point probability density f_3 will enter the needed moments, so that the only way to obtain analytical results is to neglect it, assuming $f_3 \sim c^3$, see Section 2. Thus all formulae hereafter are correct to the order $O(c^2)$ only.

According to (1.9), (2.4) and (2.5), the parameter I_3^k for the dispersion has the form

$$I_3^k = \frac{k_V^4}{c(1-c)(1-2c)} \iiint \left[\int G_V(\mathbf{y}_1) h(\mathbf{y}_1 - \mathbf{z}_1)\, d^3\mathbf{y}_1 \right]$$

$$\times \left[\int G_V(\mathbf{y}_2) h(\mathbf{y}_2 - \mathbf{z}_2)\, d^3\mathbf{y}_2 \right] h(\mathbf{z}_3) \langle \psi'(\mathbf{z}_1)\psi'(\mathbf{z}_2)\psi'(\mathbf{z}_3) \rangle\, d^3\mathbf{z}_1 d^3\mathbf{z}_2 d^3\mathbf{z}_3$$

$$= \frac{k_V^4}{c(1-c)(1-2c)} \iiint \chi_V(\mathbf{z}_1)\chi_V(\mathbf{z}_2) h(\mathbf{z}_3) \tag{4.1}$$

$$\times \left[n\delta(\mathbf{z}_{1,2})\delta(\mathbf{z}_{1,3}) - n^2 3 \{ \delta(\mathbf{z}_{1,2}) R_0(\mathbf{z}_{2,3}) \}_s \right] d^3\mathbf{z}_1 d^3\mathbf{z}_2 d^3\mathbf{z}_3$$

$$= \frac{b_0 - b_1 c}{(1-c)(1-2c)}; \quad R_0(\mathbf{z}) = 1 - g_0(\mathbf{z}),$$

with the coefficients

$$b_0 = \frac{k_V^4}{V_a}\int h(\mathbf{z})\chi_V^2(\mathbf{z})\,d^3\mathbf{z}, \tag{4.2}$$

$$b_1 = 2J_1 + J_2, \tag{4.3}$$

$$J_1 = \frac{k_V^4}{V_a^2}\int\int h(\mathbf{z}_1)\chi_V(\mathbf{z}_1)\chi_V(\mathbf{z}_2)R_0(\mathbf{z}_1-\mathbf{z}_2)\,d^3\mathbf{z}_1 d^3\mathbf{z}_2, \tag{4.4}$$

$$J_2 = \frac{k_V^4}{V_a^2}\int\int h(\mathbf{z}_1)\chi_V^2(\mathbf{z}_2)R_0(\mathbf{z}_1-\mathbf{z}_2)\,d^3\mathbf{z}_1 d^3\mathbf{z}_2, \tag{4.5}$$

$g_0(\mathbf{z})$ is the zero-density limit of the radial distribution function $g(\mathbf{z})$ for the dispersion, see (2.2).

Using (3.5), we find first the coefficient b_0:

$$b_0 = 1 - 2F_2(a_V) + F_3(a_V), \tag{4.6}$$

where

$$F_3(x) = \frac{3}{2}\frac{(1+x)^2}{x^3}e^{-2x}(\sinh x\cosh x - x) \tag{4.7}$$

is the function that appeared when evaluating the three-point statistical parameters I_3^k and I_3^α for a cellular medium, see [3,4], and $F_2(x)$ is defined in (3.7).

Let us evaluate next the coefficient b_1 in (4.3). To this end we first recast the integral J_1 as

$$J_1 = \frac{k_V^4}{V_a^2}\int h(\mathbf{z})\chi_V(\mathbf{z})P_0(\mathbf{z})\,d^3\mathbf{z}, \tag{4.8}$$

where $P_0(\mathbf{z}) = (\chi_V * R_0)(\mathbf{z})$ is the convolution, similar to that used in Section 3. Keeping in mind (3.5), (3.10) and (3.11), we find straightforwardly that

$$J_1 = 1 - F_2(a_V) + \frac{3}{4}\frac{a_V\cosh a_V - \sinh a_V}{a_V^5(1+a_V)}e^{a_V}\left[F_3(a_V) - F_2(a_V)\right]I, \tag{4.9}$$

where I is the statistical parameter of Talbot and Willis, see (3.13), corresponding to the zero-density limit $g_0(\mathbf{z})$ of the radial distribution function.

In the particular case of a well-stirred dispersion we have, due to (3.14),

$$J_1 = 1 - F_2(a_V) + \frac{1+2a_V}{(1+a_V)^2}F_2(a_V)\left[F_3(a_V) - F_2(a_V)\right]. \tag{4.10}$$

The evaluation of the second integral J_2, entering the expression for the coefficient b_1, is more complicated. We first recast its definition (4.4) as

$$J_2 = \frac{k_V^4}{V_a^2}\int \chi_V^2(\mathbf{z})F_0(\mathbf{z})\,d^3\mathbf{z}, \tag{4.11}$$

where

$$F_0(\mathbf{z}) = \int h(\mathbf{z}-\mathbf{y})R_0(\mathbf{y})\,d^3\mathbf{y} = V_a - \int h(\mathbf{z}-\mathbf{y})g_0(\mathbf{y})\,d^3\mathbf{y}. \tag{4.12}$$

Let $h_A(\mathbf{y})$ be the characteristic function of a sphere of radius A located at the origin. Following [6], we denote

$$d_A h_A(\mathbf{y}) = h_{A+dA}(\mathbf{y}) - h_A(\mathbf{y}) = \begin{cases} 1, & \text{if } A < |\mathbf{y}| < A + dA, \\ 0, & \text{otherwise.} \end{cases}$$

It is easily seen that

$$g_0(\mathbf{y}) = \int_{2a}^{\infty} g_0(A) d_A h_A(\mathbf{y}),$$

which is inserted into (4.12):

$$F_0(\mathbf{z}) = V_a - \int_{2a}^{\infty} g_0(A) \left[\int h(\mathbf{z}-\mathbf{y}) \frac{h_{A+dA}(\mathbf{y}) - h_A(\mathbf{y})}{dA} d^3\mathbf{y} \right] dA$$

(4.13)

$$= V_a - \int_{2a}^{\infty} g_0(A) \left[\frac{d}{dA} F^A(\mathbf{z}) \right] dA, \quad F^A(\mathbf{z}) = (h * h_A)(\mathbf{z}).$$

We introduce, in turn, (4.13) into (4.11) and integrate by parts:

$$J_2 = \frac{k_V^4}{V_a^2} \left\{ V_a \int \chi_V^2(\mathbf{z})\, d^3\mathbf{z} - \int_{2a}^{\infty} g_0(A) \left[\frac{d}{dA} \int \chi_V^2(\mathbf{z}) F^A(\mathbf{z})\, d^3\mathbf{z} \right] dA \right\}$$

(4.14)

$$= \frac{k_V^4}{V_a^2} \left\{ \int_{2a}^{\infty} g_0'(A) \int \left[\chi_V^2(\mathbf{z}) F^A(\mathbf{z})\, d^3\mathbf{z} \right] dA + g_0(2a) \int \chi_V^2(\mathbf{z}) F^{2a}(\mathbf{z})\, d^3\mathbf{z} \right\},$$

having used the facts that $F_\infty(\mathbf{z}) = V_a$ and $g_0(\infty) = 1$.

Let

$$\mu(\lambda, a_V) = \frac{k_V^4}{V_a^2} \int \chi_V^2(\mathbf{z}) F^{\lambda a}(\mathbf{z})\, d^3\mathbf{z}; \quad \lambda = \frac{A}{a} \geqq 2. \tag{4.15}$$

Simple algebra, based on the analytical form (3.5) of the Helmholtz potential $\chi_V(\mathbf{z})$, yields

$$\mu(\lambda, a_V) = 1 - 2F_2(a_V) + F_3(a_V) + F_4(a_V, \lambda) + F_5(a_V, \lambda), \tag{4.16}$$

where

$$F_4(x, y) = \frac{3}{2} \frac{(x\cosh x - \sinh x)^2}{x^3} \left(e^{-2x} - e^{-2(y-1)x} \right),$$

$$F_5(x, y) = \frac{3}{128} \left(\frac{x\cosh x - \sinh x}{x^3} \right)^2 \Big\{ \left[(12(y^3 + y^2 - y) + 52)x^3 \right.$$

$$- 6(y+1)^2 x^2 + 6(y-1)x + 3 \left] e^{-2(y-1)x} \right.$$

(4.17)

$$- \left[12(y-1)^2(y+1)x^3 - 6(y-1)^2 x^2 + 6(y+1)x + 3 \right] e^{-2(y+1)x}$$

$$\left. + 24(y-1)^2(y+1)^2 x^4 \left[Ei(-2(y-1)x) - Ei(-2(y+1)x) \right] \right\}$$

and F_2 and F_3 are the functions, defined in (3.7) and (4.7), respectively. As usual

$$Ei(-t) = \int_{-\infty}^{-t} \frac{e^s}{s}\, ds$$

denotes the integral exponent.

Thus

$$J_2 = a\int_2^\infty g_0'(\lambda a)\mu(\lambda, a_V)\, d\lambda + g_0(2a)\mu(2, a_V). \tag{4.18}$$

In the particular case of a well-stirred dispersion

$$J_2 = b_0 + \frac{3}{128}\left(\frac{a_V\cosh a_V - \sinh a_V}{a_V^3}\right)^2$$

$$\times\Big\{\big[172a_V^3 - 54a_V^2 + 6a_V + 3\big]e^{-2a_V} - \big[36a_V^3 - 6a_V^2 + 18a_V + 3\big]e^{-6a_V} \tag{4.19}$$

$$+216a_V^4\big[Ei(-2a_V) - Ei(-6a_V)\big]\Big\}.$$

Eventually, the needed parameter I_3^k is

$$I_3^k = \frac{b_0 - b_1 c}{(1-c)(1-2c)} = \varphi_3(a_V), \tag{4.20}$$

where a_0, a_1 are explicitly given in (4.6), (4.3), etc., respectively. Hence φ_3 is a known function of the dimensionless parameter a_V, depending on the zero-density limit $g_0(r)$ of the radial distribution function for the dispersion through the integrals I and J_2, see (3.13) and (4.18) respectively.

The evaluation of the statistical parameter I_3^α is already easy. From its definition $(1.6)_2$ and eqn (1.7) (with k_V^2 replaced by k_R^2) we have

$$I_3^\alpha = 1 - \frac{2k_R^2}{M_3^\alpha(\mathbf{0},0)}\int G_R(\mathbf{y})M_3^\alpha(\mathbf{0},\mathbf{y})\, d^3\mathbf{y}$$

$$+\frac{k_R^4}{M_3^\alpha(\mathbf{0},0)}\iint G_R(\mathbf{y}_1)G_R(\mathbf{y}_2)M_3^\alpha(\mathbf{y}_1,\mathbf{y}_2)\, d^3\mathbf{y}_1 d^3\mathbf{y}_2. \tag{4.21}$$

But

$$M_3^\alpha(\mathbf{0},\mathbf{y}) = [\alpha]^3\left\langle I_f'^2(0)I_f'(\mathbf{y})\right\rangle = (1-2c)[\alpha]^3\left\langle I_f'(0)I_f'(\mathbf{y})\right\rangle = (1-2c)[\alpha]M_2^\alpha(\mathbf{y}),$$

since $I_f'^2(0) = (I_f(0) - c)^2 = (1-2c)I_f(0) + c^2$. (Note that $I_f^2(\mathbf{x}) = I_f(\mathbf{x})$.) Hence

$$\frac{k_R^2}{M_3^\alpha(\mathbf{0},0)}\int G_R(\mathbf{y})M_3^\alpha(\mathbf{0},\mathbf{y})\, d^3y = \frac{k_R^2}{M_2^\alpha(0)}\int G_R(\mathbf{y})M_2^\alpha(\mathbf{y})\, d^3\mathbf{y} = \varphi_2(a_R),$$

see (3.17), so that

$$I_3^\alpha = 1 - 2\varphi_2(a_R) + \varphi_3(a_R), \tag{4.22}$$

because the last term in the r.-h. side of (4.21) is immediately recognized as the function φ_3 from (4.20) in which a_V is to be replaced everywhere by a_R.

5. COMPARISON WITH THE BOUNDS OF TALBOT AND WILLIS

The results of Sections 3 and 4 allow us to evaluate the bounds (1.4) for the dispersion to the order c^2. Indeed the relations (3.17) and (3.18) give us the values of the two-point statistical parameters I_2^k and I_2^σ for an arbitrary radial distribution function $g(r)$. In turn, eqns (4.20) and (4.22) provide the values of the three-point statistical parameters I_3^k and I_3^σ, again for an arbitrary radial distribution function $g(r)$, but to the order c^2 only.

Let us recall that in [1] Talbot and Willis derived bounds on the effective absorption coefficient k^{*2} for a dispersion of spheres, using an original variational principle of Hashin–Shtrikman's type. Their bounds have the form

$$\frac{k^{*2}}{k_0^2} = 1 + \frac{\lambda\delta + \mu\gamma}{\alpha\delta + \beta\gamma} \tag{5.1a}$$

with the coefficients

$$\alpha = \frac{3c(a_f\cosh a_f - \sinh a_f)}{(1-c)a_f^3}, \quad \beta = \frac{a^2k_0^2}{k_f^2}\langle k^2\rangle,$$

$$\gamma = e^{-ak_0}(\cosh a_f + \frac{k_0}{k_f}\sinh a_f) - \frac{3c(k_f^2 - k_0^2)(a_f\cosh a_f - \sinh a_f)\eta}{k_0^2a_f^3(1-c)} + \theta,$$

$$\eta = (1 + ak_0)e^{-ak_0} - \frac{12cI}{ak_0}(ak_0\cosh ak_0 - \sinh ak_0), \tag{5.1b}$$

$$\theta = \frac{12cI}{ak_0}\left[\cosh ak_f \sinh ak_0 - \frac{k_0}{k_f}\sinh ak_f\cosh ak_0\right],$$

$$\delta = \frac{k_0^2}{k_f^2}(a_f^2 - a_m^2)\eta, \quad \lambda = \frac{3c(k_f^2 - k_0^2)(a_f\cosh a_f - \sinh a_f)}{(1-c)k_0^2a_f^3},$$

$$\mu = c\frac{k_m^2}{k_f^2}(a_f^2 - (ak_0)^2) + (1-c)(a_m^2 - (ak_0)^2), \quad a_f = ak_f,\ a_m = ak_m.$$

(Our notations differ a bit from the original ones used in [1].)

Upon inserting $k_0 = \min(k_m, k_f)$ in (5.1) one obtains a lower bound on k^{*2} and, similarly, inserting $k_0 = \max(k_m, k_f)$ — an upper one. In (5.1b) I is the statistical parameter, defined in (3.13), which carries information about the two-point statistics of the dispersion. In this sense the bounds (5.1) are two-point and therefore should be expected to be less restrictive than ours (1.4) which are three-point.

It is to be pointed out, however, that Talbot and Willis' bounds (5.1) are useful for all values $c \in (0,1)$ of the sphere volume fraction while the bounds (1.4) have been calculated in the foregoing analysis only for dilute fractions — to the

order c^2 — and thus may be expected to provide useful results for values of c not exceeding $0.10 - 0.15$. The numerical calculations confirm these expectations. The c^2-bounds (1.4) are closer to the exact values of k^{*2} and more restrictive than the Talbot and Willis estimates (5.1) only at sphere fractions c not exceeding 0.1. This is illustrated in Tables 1 and 2 for a well-stirred dispersion of spheres in the two cases $k_f^2/k_m^2 = 10$ and $k_f^2/k_m^2 = 0.1$ respectively (at $a_m = 1$). The exact values are found by means of the numerical procedure, developed in [7] which employs the techniques of the factorial functional series [8] and allows to obtain explicitly the full statistical solution of eqn (1.1) to the order c^2 for the dispersion and, in particular, the effective absorption coefficient k^{*2} to the same order. The results for other values of a_m ($a_m = 10$ and $a_m = 0.1$) are similar and therefore they are not shown here.

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Table 1

c	TW-lower	KM-lower	exact	KM-upper	TW-upper
0.0	1	1	1	1	1
0.02	1.071	1.071	1.071	1.072	1.089
0.04	1.147	1.147	1.147	1.147	1.183
0.06	1.229	1.230	1.230	1.231	1.281
0.08	1.317	1.318	1.318	1.319	1.384
0.10	1.413	1.414	1.415	1.417	1.492

Estimates on the effective absorption coefficient k^{*2} for a well-stirred dispersion at $a_m = 1$, $k_f^2/k_m^2 = 10$: KM — the bounds (1.4); TW — the bounds (5.1) of Talbot and Willis

Table 2

c	TW-lower	KM-lower	exact	KM-upper	TW-upper
0.0	1	1	1	1	1
0.02	0.977	0.978	0.979	0.979	0.979
0.04	0.955	0.957	0.958	0.958	0.958
0.06	0.934	0.937	0.937	0.937	0.937
0.08	0.914	0.917	0.917	0.917	0.917
0.10	0.894	0.896	0.896	0.897	0.897

The same as in Table 1 at $k_f^2/k_m^2 = 0.1$

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