

## ON THE APPLICATION OF RANDOM-POINT APPROXIMATION FOR IDENTIFICATION OF THE EFFECTIVE DIFFUSIVITY COEFFICIENT OF POLYDISPERSE SPHERICAL SUSPENSION

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**ABSTRACT.** The so-called Random-Point Approximation (RPA) is applied to identifying the effective diffusivity of a polydisperse spherical suspension. The RPA is based on truncated Volterra-Wiener Expansion (VWE) with basis function which is a random point function of perfect-disorder type. The VWE is applied consistently, and the equations for the kernels are derived. The contributions of the different kernels to the overall (effective) modulus are identified. An approximate model based on power-series expansion of the kernels with respect to the volume fraction  $c$  is developed. It is argued that the contribution of the pure binary interaction (two-sphere solution) is negligible for moderately concentrated suspensions. The issues connected with the application of the fourth-order kernel (one-sphere solution) are tackled, by evaluating some new kind of integrals.

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### 1. INTRODUCTION

Estimating the effective transport coefficients of heterogeneous media is of significance for technology, and has attracted vigorous investigation (see the detailed accounts in [1, 2]). The most typical examples of such media are the particulate materials (suspensions) in which the second (particulate) phase is comprised by spherical particles (the *filler*) that are randomly dispersed throughout the continuous phase (the *matrix*). The different transport problems that can be considered for a suspension are the effective electric or heat conductivity, viscosity, and elasticity.

Different methodologies and techniques have been developed during the years to model the behavior of composites, especially their effective transport coefficients. Because of the complexity of the mathematical problems involved, different approximations are still being sought, e.g. limits on the moduli, etc. (see [2] for the current stage of the research).

A rigorous approach to constructing successive approximations to the random fields that describe a mechanical property of a composite (such as viscosity, elasticity, heat diffusivity, etc.) has been proposed in [3]. It is based on the idea to expand the unknown random fields in Volterra–Wiener series, which is a functional expansion (see [4] for the definition of a functional expansion). This functional expansion for random processes was formulated by Wiener in the late 1940s (see [5]). The gist of the Volterra–Wiener expansion (VWE) is that the kernels are non-random functions and only the basis function contains the information about the randomness of the field under consideration. The original VWE employs the Gaussian white noise as a basis function. The expansion can be made orthogonal by using the Hermit polynomials of the white-noise function, and the series is alternatively called ‘Wiener-Hermite Expansion’ (WHE). The WHE was applied to some turbulence problems (see, e.g. [6]) but faced formidable difficulties for nonlinear problems and required frequent renormalization [7]. This shortcoming was explained in [3] by making the observation that the Gaussian white noise has zero third moments (as any normally distributed function). This means that despite the general theorem from [8] that any random function can be expanded into Wiener-Hermite series, the series are practically divergent if the kernels are identified via solving nonlinear hierarchical systems.

The most natural conjecture for a particulate materials is that the centers of the inclusions are represented by a system of random points, while the effect of each inclusion is defined by the mechanical properties and the geometry of the inclusion, i.e., the effect of the latter is deterministic. The superposition of non-random structures that appear in random positions of space or moments of time is called Random Point Function/Process (RPF) (see [9]). This led one of the present authors to suggest that VWE with a RPF basis can open the possibility for rigorous construction of consecutive approximations to the problem of mechanical behavior of the composites. Naturally, the simplest RPF function is the Poisson process, and the proposal in [3] was to use a Poisson basis. Actually, the first paper in which Poisson process was used as basis function was [10], where was demonstrated that the Charlier polynomials are needed to render the functional series orthogonal. However, no applications of the Poisson-Wiener expansion were attempted in [10], and the exclusive relevance of these series for nonlinear problems was argued in [3].

It turned out that the plain Poisson basis function was a very good guess for the basis in modeling the chaotic behavior of nonlinear dynamical systems, e.g. turbulence. It goes beyond the scope of the present paper to give an overview of the research in this direction. We refer the reader to [11] for a comprehensive exposition or to the English language reviews [12, 13]. The application to suspensions/emulsion of the Random-Point Approximation (RPA) was sketched first in [14, 15] (see also [16]) for treating the interaction between the phases in suspensions/emulsions.

The Poisson RPF is not very well suited for composite materials, because if the the random centers are assumed to be completely independent statistically, then the inclusions can consequently overlap. However, the RPA is so well tailored to this kind of problems that the (anti)correlation of the random points needed to prevent the overlapping contributes only to the second order with respect to the volume fraction. This was the reason why in [17] RPF of perfect disorder were introduced, for which the multi-point probability distributions are defined to be zero if the centers of the respective inclusions are closer than the sum of their radii. If the inclusions do not overlap, then the multi-point probability density is equal to unity, which corresponds to Poisson distribution. The effective heat conduction of monodisperse suspension was treated in [17] and shown that the linear contribution of the filler to the effective modulus is equal exactly to Maxwell's formula for the effective electric conductivity. Later on, the elastic problem was solved in [18].

After the generalization of the RPA to marked random point functions was introduced in [19], the most general case of a polydisperse suspension of perfect disorder type became amenable to the VWE method (see [20]). It has been rigorously established in the cited works that the knowledge of the first kernel allows one to obtain the respective effective modulus with a first order of approximation in volume fraction  $c$  of the filler. Respectively, it has been proved that only if one has the the two-sphere solution, then one can rigorously obtain the the second-order approximation in the volume fraction, as the two-sphere solution gives precisely the second-order kernel in the formal Volterra-Wiener expansion.

While, the one-sphere solution was known for many years, the two sphere solution could not be obtained analytically. One of the popular techniques is the two-pole expansion (see, among others [21, 22]), but it has a very slow convergence. The alternative (originated in [23] for the case of constant field at infinity) was to use bi-spherical coordinates. However, the effective transport properties can be obtained, only if the field around two spheres with constant gradient at infinity is found. The latter is a 3D problems, and a Legendre-series method for its solution was developed in [24]. The numerical technique from [24] has been recently implemented in [25], and the actual temperature distribution around two spheres was found when a constant gradient of the field is prescribed at infinity. The magnitude of the contribution due to the interaction is orders of magnitude smaller than the contribution of the perturbation due to a single sphere. The case of very closely situated sphere was interrogated in [26] and even in that case, the two-sphere contribution was of the order of 20% of the one-sphere contribution. This motivates the goal of the present work: to lay the groundwork needed for rigorous identification of the contribution of the first kernel (one-sphere solution) to the second order in the volume fraction expression for the effective heat-conduction modulus.

## 2. MEDIUM WITH RANDOM DISCONTINUOUS TRANSPORT COEFFICIENT

We consider the two-phase medium that consists of an array of perfectly disordered spheres of random radii with thermal conductivity  $\varkappa_f$  distributed in an unbounded matrix of conductivity  $\varkappa_m$ . The suspension can be considered as a medium with a discontinuous coefficient of thermal conductivity. We can express the random conductivity coefficient for such a medium in the following way:

$$\varkappa(\mathbf{x}) = \varkappa_m + \llbracket \varkappa \rrbracket \int_{\mathbb{R}^4} h(\mathbf{x} - \boldsymbol{\xi}; a) \omega(\boldsymbol{\xi}; a) d^3 \boldsymbol{\xi} da, \quad h(\mathbf{x}; a) = \begin{cases} 0 & \text{for } |\mathbf{x}| > a, \\ 1 & \text{for } |\mathbf{x}| \leq a, \end{cases} \quad (2.1)$$

where  $h$  is the shape function of a single spherical inclusion, and  $\mathbb{R}^4 = \mathbb{R}^3 \times \mathbb{U}$  is the four-dimensional space which is the cartesian product of the three dimensional configurational (geometric) space and the set  $\mathbb{U}$  of the possible values of the random mark (so-called ‘mark space’). Without losing the generality, we can assume that  $\mathbb{U} = \mathbb{R}_+^1 = \{a | a > 0\}$  because if the set  $\mathbb{U}$  is any finite interval in  $\mathbb{R}$ , then we can augment the probability density of the mark  $P(a)$  with zero outside of the final interval. The randomness shows up through the function

$$\omega(\boldsymbol{\xi}, a) = \sum_{\alpha} \delta(\boldsymbol{\xi} - \boldsymbol{\xi}_{\alpha}) \delta(a - a_{\alpha}),$$

which is a random superposition of Dirac delta functions. Here  $\boldsymbol{\xi}_{\alpha}$  is the random position at which a sphere appears and  $a_{\alpha}$  is a random mark. For the case of suspension, the random mark has clear physical meaning: it is the random radius of the sphere that happens to be centered at the random position  $a_{\alpha}$ . Note that the index  $\alpha$  can range over an infinite set of numbers. Function  $\omega(\mathbf{x}, a)$  is called generalized random density function. Let  $\gamma$  denote the number density of the system of random points, and  $P(a)$  is the probability density of the mark. When  $P(a) = \delta(a - a_0)$ , one gets the case of monodisperse suspension containing spheres of the same radius  $a_0$ .

We observe here that the average volume of a single inclusion is the following mathematical expectation

$$\langle V \rangle \equiv \frac{4\pi}{3} \int_0^{\infty} a^3 P(a) da,$$

and then the concentration of the particulate phase is by definition  $c = \gamma \langle V \rangle$ . One should not confuse the notation for the average volume  $\langle V \rangle$  with the notation  $V_a$  which stands for the geometric domain that is encompassed by a single spherical inclusion of radius  $a$ .

In this section we will consider the equation of the heat diffusion

$$\nabla \cdot [\varkappa(\mathbf{x}) \nabla T(\mathbf{x})] = 0, \quad (2.2)$$

where the coefficient of thermal diffusivity  $\varkappa(\mathbf{x})$  is a random function, given in Eq. (2.1). We will seek a solution of the problem in functional series.

### 3. VOLTERRA-WIENER FUNCTIONAL EXPANSIONS

For the considered here continuum with random modulus, the randomness of the solution is created by the randomness of the coefficient. The natural approach for this kind of problems with defined source of randomness is to consider a functional series with basis function that is related to the random coefficient (the random density function that defines the coefficient). This means that the temperature field can be considered as the following Volterra-Wiener functional series

$$T(\mathbf{x}) = \bar{T}_0 + \int_{\mathbb{R}^4} \bar{T}_1(\mathbf{x} - \boldsymbol{\xi}; a) \omega(\boldsymbol{\xi}; a) d^3 \boldsymbol{\xi} da + \iint_{\mathbb{R}^4} \bar{T}_2(\mathbf{x} - \boldsymbol{\xi}_1, \mathbf{x} - \boldsymbol{\xi}_2; a_1, a_2) \omega(\boldsymbol{\xi}_1; a_1) \omega(\boldsymbol{\xi}_2; a_2) d^3 \boldsymbol{\xi}_1 d^3 \boldsymbol{\xi}_2 da_1 da_2 + \dots, \quad (3.1)$$

where  $T_n$  are non-random kernels.

The direct application of Eq. (3.1) is not very convenient, because of the complex expression for the scalar products of different powers of function  $\omega$ . The application can be streamlined if polynomials of  $\omega$  are used, for which the different terms are orthogonal to each other.

As already above mentioned, to orthogonalize the functionals, one needs to use multivariate stochastic polynomials of the random density function  $\omega$ , in this case: the well-known Charlier polynomials, which were introduced in [10] for the Poisson basis functions. In [19] the Charlier polynomials were generalized for the case of the random density functions of Perfect-Disorder type. In the present context of Perfect Disorder of Spheres of Random Radii, the basis functions are as follow (the first three are presented here)

$$C_\omega^{(0)} \stackrel{\text{def}}{=} 1, \quad (3.2a)$$

$$C_\omega^{(1)}(\mathbf{x}_1; a_1) \stackrel{\text{def}}{=} \omega(\mathbf{x}_1; a_1) - \gamma P(a_1), \quad (3.2b)$$

$$\begin{aligned} C_\omega^{(2)}(\mathbf{x}_1, \mathbf{x}_2; a_1, a_2) &\stackrel{\text{def}}{=} \omega(\mathbf{x}_1; a_1) [\omega(\mathbf{x}_2; a_2) - \delta(\mathbf{x}_1 - \mathbf{x}_2) \delta(a_1 - a_2)] \\ &\quad - \gamma [P(a_2) \omega(\mathbf{x}_1; a_1) + P(a_1) \omega(\mathbf{x}_2; a_2)] + \gamma^2 P(a_1) P(a_2) \\ &= C_\omega^{(1)}(\mathbf{x}_1; a_1) C_\omega^{(1)}(\mathbf{x}_2; a_2) - [C_\omega^{(1)}(\mathbf{x}_1; a_1) + \gamma P(a_1)] \delta(\mathbf{x}_1 - \mathbf{x}_2) \delta(a_1 - a_2), \\ &\quad \dots \dots \end{aligned} \quad (3.2c)$$

We mention here that from Eq. (3.2b) we get  $\omega(\mathbf{x}_1; a_1) = C_\omega^{(1)}(\mathbf{x}_1; a_1) + \gamma P(a_1)$ . Then upon introducing the above expression for  $\omega$  and the concentration in (2.1), we get

an alternative expression for the discontinuous transport coefficients, namely

$$\varkappa(\mathbf{x}) = \langle \varkappa \rangle + \llbracket \varkappa \rrbracket \int_{\mathbb{R}^4} h(\mathbf{x} - \boldsymbol{\xi}; a) C_{\omega}^{(1)}(\boldsymbol{\xi}; a) d^3 \boldsymbol{\xi} da, \quad (3.3)$$

where  $\langle \varkappa \rangle = \varkappa_m + c \llbracket \varkappa \rrbracket$  denotes the average conductivity of the medium. Respectively, the Volterra-Wiener series for the temperature field adopt the form

$$\begin{aligned} T(\mathbf{x}) &= T_0(\mathbf{x}) + \int_{\mathbb{R}^4} T_1(\mathbf{x} - \boldsymbol{\xi}; a) C_{\omega}^{(1)}(\boldsymbol{\xi}; a) d^3 \boldsymbol{\xi} da \\ &+ \iint_{\mathbb{R}^4} T_2(\mathbf{x} - \boldsymbol{\xi}_1, \mathbf{x} - \boldsymbol{\xi}_2; a_1, a_2) C_{\omega}^{(2)}(\boldsymbol{\xi}_1, \boldsymbol{\xi}_2; a_1, a_2) d^3 \boldsymbol{\xi}_1 d^3 \boldsymbol{\xi}_2 da_1 da_2 + \dots, \end{aligned} \quad (3.4)$$

where  $T_0(\mathbf{x}) \equiv \langle T(\mathbf{x}) \rangle$  is the ensemble averaged temperature field. We note here that for finding the effective coefficient, it is enough to consider the case when  $\nabla T_0(\mathbf{x}) = \langle \nabla T(\mathbf{x}) \rangle = \mathbf{G}$ , where  $\mathbf{G}$  is some constant vector gradient at infinity.

Let us note that it was shown in [17, 20] that the functional series are *virial* in the sense that the integral containing the  $n$ -th kernel contributes to the averaged characteristics a quantity of order  $\gamma^n$ , where  $\gamma$  gives us the above defined number density. Actually, after acknowledging the connection of  $\gamma$  to  $c$ , we can reformulate the property of ‘viriality’ and to claim that each term contributes a quantity of order of  $c^n$ . Since  $c < 1$ , the virial property of the functional series ensures their convergence. Albeit the latter may turn out to be slow for very concentrated suspensions, the viriality is indispensable property, because it provides for a rigorous perturbation methodology.

Combining (3.3) and (3.4), we find the expression for the heat flux as a random function within the adopted order of approximation  $O(\gamma^3)$ :

$$\begin{aligned} \mathbf{Q} \stackrel{\text{def}}{=} \varkappa(\mathbf{x}) \nabla T(\mathbf{x}) &= \langle \varkappa \rangle \nabla T_0(\mathbf{x}) + \int_{\mathbb{R}^4} \nabla_x T_1(\mathbf{x} - \boldsymbol{\xi}_1; a_1) C_{\omega}^{(1)}(\boldsymbol{\xi}_1; a_1) d^3 \boldsymbol{\xi}_1 da_1 \\ &+ \langle \varkappa \rangle \iint_{\mathbb{R}^4} \nabla_x T_2(\mathbf{x} - \boldsymbol{\xi}_1, \mathbf{x} - \boldsymbol{\xi}_2; a_1, a_2) C_{\omega}^{(2)}(\boldsymbol{\xi}_1, \boldsymbol{\xi}_2; a_1, a_2) d^3 \boldsymbol{\xi}_1 d^3 \boldsymbol{\xi}_2 da_1 da_2 \\ &+ \llbracket \varkappa \rrbracket \int_{\mathbb{R}^4} \nabla_x T_0(\mathbf{x}) h(\mathbf{x} - \boldsymbol{\xi}_1; a_1) C_{\omega}^{(1)}(\boldsymbol{\xi}_1; a_1) d^3 \boldsymbol{\xi}_1 da_1 \\ &+ \llbracket \varkappa \rrbracket \iint_{\mathbb{R}^4} h(\mathbf{x} - \boldsymbol{\xi}_1; a_1) \nabla_x T_1(\mathbf{x} - \boldsymbol{\xi}_2; a_2) C_{\omega}^{(1)}(\boldsymbol{\xi}_1; a_1) C_{\omega}^{(1)}(\boldsymbol{\xi}_2; a_2) d^3 \boldsymbol{\xi}_1 d^3 \boldsymbol{\xi}_2 da_1 da_2 \\ &+ \llbracket \varkappa \rrbracket \iiint_{\mathbb{R}^4} h(\mathbf{x} - \boldsymbol{\xi}_1; a_1) \nabla_x T_2(\mathbf{x} - \boldsymbol{\xi}_2, \mathbf{x} - \boldsymbol{\xi}_3; a_2, a_3) \\ &\quad \times C_{\omega}^{(1)}(\boldsymbol{\xi}_1; a_1) C_{\omega}^{(2)}(\boldsymbol{\xi}_2, \boldsymbol{\xi}_3; a_2, a_3) d^3 \boldsymbol{\xi}_1 d^3 \boldsymbol{\xi}_2 d^3 \boldsymbol{\xi}_3 da_1 da_2 da_3 + \dots. \end{aligned} \quad (3.5)$$

In the last integral the notation  $\nabla_x$  stands for the gradient of  $T_2$  with respect to  $\mathbf{x}$ , which is obtained from the gradients with respect to its two spatial vector arguments.

In order to obtain an expression for the averaged heat flux, we need the expressions for the average values of the products developed in the above cited works. For

the first couple of them we have

$$\langle C_\omega^{(1)}(\boldsymbol{\xi}_1; a_1)C_\omega^{(1)}(\boldsymbol{\xi}_2; a_2) \rangle = \gamma P(a_1)\delta(\boldsymbol{\xi}_1 - \boldsymbol{\xi}_2)\delta(a_1 - a_2) + \gamma^2 P(a_1)P(a_2)(Q_{12} - 1), \quad (3.6a)$$

$$\begin{aligned} \langle C_\omega^{(1)}(\boldsymbol{\xi}_1; a_1)C_\omega^{(1)}(\boldsymbol{\xi}_2; a_2)C_\omega^{(1)}(\boldsymbol{\xi}_3; a_3) \rangle &= \gamma P(a_1)\delta(\boldsymbol{\xi}_1 - \boldsymbol{\xi}_2)\delta(\boldsymbol{\xi}_1 - \boldsymbol{\xi}_3)\delta(a_1 - a_2)\delta(a_1 - a_3) \\ &+ \gamma^2 [P(a_3)P(a_1)\delta(\boldsymbol{\xi}_1 - \boldsymbol{\xi}_2)\delta(a_1 - a_2)(Q_{31} - 1) \\ &+ P(a_1)P(a_2)\delta(\boldsymbol{\xi}_2 - \boldsymbol{\xi}_3)\delta(a_2 - a_3)(Q_{12} - 1) \\ &+ P(a_2)P(a_3)\delta(\boldsymbol{\xi}_3 - \boldsymbol{\xi}_1)\delta(a_3 - a_1)(Q_{23} - 1)] + O(\gamma^3), \end{aligned} \quad (3.6b)$$

$$\begin{aligned} \langle C_\omega^{(1)}(\boldsymbol{\xi}_3; a_3)C^{(2)}(\boldsymbol{\xi}_1, \boldsymbol{\xi}_2; a_1, a_2) \rangle &= \gamma^2 [P(a_3)P(a_1)\delta(\boldsymbol{\xi}_1 - \boldsymbol{\xi}_2)\delta(a_1 - a_2)(Q_{31} - 1) \\ &+ P(a_1)P(a_2)\delta(\boldsymbol{\xi}_2 - \boldsymbol{\xi}_3)\delta(a_2 - a_3)(Q_{12} - 1) \\ &+ P(a_2)P(a_3)\delta(\boldsymbol{\xi}_3 - \boldsymbol{\xi}_1)\delta(a_3 - a_1)(Q_{23} - 1)] + O(\gamma^3), \end{aligned} \quad (3.6c)$$

$$\begin{aligned} &\langle C_\omega^{(2)}(\boldsymbol{\xi}_1, \boldsymbol{\xi}_2; a_1, a_2)C_\omega^{(2)}(\boldsymbol{\xi}_3, \boldsymbol{\xi}_4; a_3, a_4) \rangle \\ &= (Q_{12} - 1)P(a_1)P(a_2)\delta(\boldsymbol{\xi}_1 - \boldsymbol{\xi}_3)\delta(\boldsymbol{\xi}_2 - \boldsymbol{\xi}_4)\delta(a_1 - a_3)\delta(a_2 - a_4) \\ &+ (Q_{12} - 1)P(a_1)P(a_2)\delta(\boldsymbol{\xi}_1 - \boldsymbol{\xi}_4)\delta(\boldsymbol{\xi}_2 - \boldsymbol{\xi}_3)\delta(a_1 - a_4)\delta(a_2 - a_3) + O(\gamma^3), \end{aligned} \quad (3.6d)$$

$$\begin{aligned} &\langle C_\omega^{(1)}(\boldsymbol{\xi}_1; a_1)C_\omega^{(1)}(\boldsymbol{\xi}_2; a_2)C_\omega^{(2)}(\boldsymbol{\xi}_3, \boldsymbol{\xi}_4; a_3, a_4) \rangle \\ &= (Q_{12} - 1)P(a_1)P(a_2)\delta(\boldsymbol{\xi}_1 - \boldsymbol{\xi}_3)\delta(\boldsymbol{\xi}_2 - \boldsymbol{\xi}_4)\delta(a_1 - a_3)\delta(a_2 - a_4) \\ &+ (Q_{12} - 1)P(a_1)P(a_2)\delta(\boldsymbol{\xi}_1 - \boldsymbol{\xi}_4)\delta(\boldsymbol{\xi}_2 - \boldsymbol{\xi}_3)\delta(a_1 - a_4)\delta(a_2 - a_3) + O(\gamma^3). \end{aligned} \quad (3.6e)$$

Eq. (3.6e) is obtained from Eq. (3.6d) after acknowledging the connection between  $C^{(2)}$  and  $C^{(1)}$  from Eq. (3.2c), orthogonality of  $C^{(2)}$  and  $C^{(1)}$  and the fact that  $C^{(2)}$  is a centered random function.

Functions  $Q_{ik}$ , which are involved in the above formula were introduced in [20] to forbid the spheres from intersecting, namely

$$Q_{ik} \stackrel{\text{def}}{=} Q(\boldsymbol{\xi}_i, \boldsymbol{\xi}_k; a_i, a_k) = \begin{cases} 0 & \text{for } |\boldsymbol{\xi}_i - \boldsymbol{\xi}_k| \leq a_i + a_k, \\ 1 & \text{for } |\boldsymbol{\xi}_i - \boldsymbol{\xi}_k| > a_i + a_k. \end{cases} \quad (3.7)$$

The above expression for  $Q$  ensures that when the centers of two spheres are farther than the sum of their radii, they are statistically independent, and the basis function behave as a Poissonian random function (as sometimes called ‘perfectly random function’). Random functions of this type were called in [20] ‘Random Functions of Perfect Disorder Type.’ Note that  $Q_{ik}$  is symmetric with respect to interchanging the arguments in the first and second pairs.

#### 4. GOVERNING EQUATION FOR KERNELS

The basic problem we are faced with when employing functional expansions is the identification of the respective kernels. We follow [20] which is a less accessible

publication. We mention that the gist of the method can be found also in [17], but for the limiting case of monodisperse suspensions. First, we assume that the average gradient of the temperature fields is constant, namely  $\nabla T_0(\mathbf{x}) = \mathbf{G} = \text{const}$ . Taking the average of Eq. (2.2) with Eq. (3.5) acknowledged, gives

$$\nabla \cdot \left\{ \langle \varkappa \rangle \mathbf{G} + [\varkappa] \int_{\mathbb{R}^4} h(\mathbf{x}; a) \nabla T_1(\mathbf{y}; b) [\gamma P(a) \delta(\mathbf{x} - \mathbf{y}) \delta(a - b) + \gamma^2 P(a) P(b) (Q_{12}(\mathbf{x}, \mathbf{y}; a, b) - 1)] d^3 \mathbf{x} d^3 \mathbf{y} da db \right\} = 0,$$

which is trivially satisfied since all of the terms enclosed between the braces are constant. At this junction, in order to simplify the treatment of the double integral, we define an auxiliary function as follows

$$S(\mathbf{y}; a, b) \stackrel{\text{def}}{=} \frac{1}{V_a} \int_{\mathbb{R}^3} h(\mathbf{x}) [Q_{12}(\mathbf{x}, \mathbf{y}; a, b) - 1] d^3 \mathbf{x}. \quad (4.1)$$

The meaning of this function is that when used as a kernel in an integral, it parametrizes the effect of the spheres being restricted from overlapping. We will call  $S$  in what follows the ‘interaction function.’ As it will become evident later in the text, the statistical average of the interaction function plays an important role. For this reason we introduce a special notation for it

$$\mathfrak{S}_P(\mathbf{y}; b) = \frac{V_a}{\langle V \rangle} \int_0^\infty S(\mathbf{y}; a, b) P(a) da, \quad (4.2)$$

where the superscript  $P$  refers to the fact that the average is taken with respect to the probability distribution  $P(b)$ . Note that  $\mathfrak{S}_P$  is introduced mostly for simplifying the notation in the general formulas. The integration over  $b$  is not an obvious procedure, and very often, it is better deferred to the stage where the integration with respect to  $\mathbf{y}$  has already been performed.

In addition we introduce the ‘volume average’ of the gradient of  $T_1$

$$\overline{\nabla T_1}^{V_a} \stackrel{\text{def}}{=} \frac{1}{V_a} \int_{\mathbb{R}^3} h(\mathbf{x}; a) \nabla T_1(\mathbf{x}; a) d^3 \mathbf{x} = \frac{1}{V_a} \iiint_{V_a} \nabla T_1(\mathbf{x}; a) d^3 \mathbf{x} \quad (4.3)$$

over the volume of a single sphere. In some cases one may find useful to use the divergence theorem and to render the above integral to a surface integral, but this goes beyond the scope of the present work. In a similar fashion we define

$$\overline{\nabla T_1}^{(V,P)} = \frac{1}{\langle V \rangle} \int_0^\infty V_a \overline{\nabla T_1}^{V_a} P(a) da, \quad (4.4)$$

where the meaning is that the average is taken over all different volumes of sphere whose radii are distributed with probability  $P$ . Then the first equation for the kernel can be rewritten as (note that  $\nabla T_0(\mathbf{x}) \equiv \mathbf{G}$ ,  $c = \gamma \langle V \rangle$ )

$$\begin{aligned} \nabla \cdot (\langle \varkappa(\mathbf{x}) T(\mathbf{x}) \rangle) = 0, \quad \langle \varkappa(\mathbf{x}) T(\mathbf{x}) \rangle = \langle \varkappa \rangle \mathbf{G} + [\varkappa] c \overline{\nabla T_1}^{(V,P)} \\ + [\varkappa] c^2 \frac{1}{\langle V \rangle} \int_{\mathbb{R}^4} \nabla T_1(\mathbf{y}; b) \mathfrak{S}_P(\mathbf{y}; b) P(b) d^3 \mathbf{y} db. \end{aligned} \quad (4.5a)$$



Following the procedure outlined in [3] and developed in [17, 19, 20], we multiply Eqs. (2.2), (3.5) by the first-order Charlier polynomial  $C_\omega^{(1)}(\mathbf{0}; a)$ , and obtain within the order  $O(\gamma^3)$  the following equation for the nonrandom kernel  $T_1(\mathbf{x}; a)$

$$\begin{aligned} & \nabla \cdot \left\{ \langle \chi \rangle cP(a)T_1(\mathbf{x}; a) \right. \\ & + \langle \chi \rangle c^2P(a) \frac{1}{\langle V \rangle} \int_{\mathbb{R}^4} \nabla T_1(\mathbf{x} - \boldsymbol{\xi}_1; a_1)[Q(\mathbf{0}, \boldsymbol{\xi}_1; a, a_1) - 1]P(a_1)d^3\boldsymbol{\xi}_1 da_1 \\ & + [\chi] \mathbf{G}[cP(a)h(\mathbf{x}; a) \\ & + c^2P(a) \frac{1}{\langle V \rangle} \int_{\mathbb{R}^4} h(\mathbf{x} - \boldsymbol{\xi}_1; a_1)[Q(\mathbf{0}, \boldsymbol{\xi}_1; a, a_1) - 1]P(a_1) d^3\boldsymbol{\xi}_1 da_1] \\ & + [\chi] cP(a)h(\mathbf{x}; a)\nabla T_1(\mathbf{x}; a) \\ & + [\chi] c^2P(a) \frac{1}{\langle V \rangle} \int_{\mathbb{R}^4} h(\mathbf{x} - \boldsymbol{\xi}_1; a_1)\nabla T_1(\mathbf{x} - \boldsymbol{\xi}_1; a_1)[Q(\boldsymbol{\xi}_1, \boldsymbol{\xi}_1; a_1, a_1) - 1]P(a_1)d^3\boldsymbol{\xi}_1 da_1 \\ & + c^2[\chi] P(a)[\nabla T_1(\mathbf{x}; a) \frac{1}{\langle V \rangle} \int_{\mathbb{R}^4} h(\mathbf{x} - \boldsymbol{\xi}_1; a_1)[Q(\boldsymbol{\xi}_1, \mathbf{0}; a_1, a) - 1]P(a_1)d^3\boldsymbol{\xi}_1 da_1 \\ & + c^2[\chi] P(a)h(\mathbf{x}; a) \frac{1}{\langle V \rangle} \int_{\mathbb{R}^4} \nabla T_1(\mathbf{x} - \boldsymbol{\xi}_2; a_2)[Q(\mathbf{0}, \boldsymbol{\xi}_2; a, a_2) - 1]P(a_2)d^3\boldsymbol{\xi}_2 da_2 \\ & \left. + 2c^2[\chi] \frac{1}{\langle V \rangle} \int_{\mathbb{R}^4} h(\mathbf{x} - \mathbf{y}; b)\nabla T_2(\mathbf{x} - \mathbf{y}, \mathbf{x}; b, a)P(b)d^3\mathbf{y} db \right\} + O(\gamma^3) = 0. \end{aligned}$$

Before proceeding further, we manipulate the integral on the fourth line of the above equation taking into account the property of function  $Q(\boldsymbol{\xi}_1, \boldsymbol{\xi}_1; a_1, a_1) = 0$ , namely

$$\begin{aligned} & \int_{\mathbb{R}^4} h(\mathbf{x} - \boldsymbol{\xi}_1; a_1)\nabla T_1(\mathbf{x} - \boldsymbol{\xi}_1; a_1)[Q(\boldsymbol{\xi}_1, \boldsymbol{\xi}_1; a_1, a_1) - 1]P(a_1)d^3\boldsymbol{\xi}_1 da_1 \\ & = - \int_{\mathbb{R}^4} h(\mathbf{x} - \boldsymbol{\xi}_1; a_1)\nabla T_1(\mathbf{x} - \boldsymbol{\xi}_1; a_1)P(a_1)d^3\boldsymbol{\xi}_1 da_1 \\ & = - \int_0^\infty \left[ \iiint_{V_{a_1}} \nabla_1 T_1(\mathbf{y}; a_1)d^3\mathbf{y} \right] P(a_1) dda_1 = - \int_0^\infty \overline{\nabla T_1}^{V_a} P(a) da, \end{aligned}$$

where the localization properties of function  $h$  are taken into account. Now, we are equipped to derive the most convenient form of the equation for the kernel  $T_1$ :

$$\begin{aligned} & \nabla \cdot \left\{ cP(a)[\langle \chi \rangle T_1(\mathbf{x}; a) + [\chi] h(\mathbf{x}; a)\nabla T_0(\mathbf{x}; a) + [\chi] h(\mathbf{x}; a)\nabla T_1(\mathbf{x}; a)] \right. \\ & + c^2P(a)[\langle \chi \rangle + [\chi] h(\mathbf{x}; a)] \frac{1}{\langle V \rangle} \int_{\mathbb{R}^4} \nabla T_1(\mathbf{x} - \boldsymbol{\xi}_1; a_1)[Q(\mathbf{0}, \boldsymbol{\xi}_1; a, a_1) - 1]P(a_1) d^3\boldsymbol{\xi}_1 da_1 \\ & + 2[\chi] c^2P(a)\nabla T_0(\mathbf{x}; a)\mathfrak{S}_P(\mathbf{x}; a) - [\chi] c^2P(a)\overline{\nabla T_1}^{(V,P)} \\ & \left. + 2c^2[\chi] P(a) \frac{1}{\langle V \rangle} \int_{\mathbb{R}^4} h(\mathbf{x} - \mathbf{y}; b)\nabla T_2(\mathbf{x} - \mathbf{y}, \mathbf{x}; b, a)P(b) d^3\mathbf{y} db \right\} = 0. \quad (4.5b) \end{aligned}$$

Following the general procedure, we multiply Eqs. (2.2), (3.5) by the second-order Charlier polynomial  $C_\omega^{(2)}(\mathbf{0}, \mathbf{z}; a, b)$  to obtain

$$c^2 \nabla \cdot \left( 2 \{ \varkappa_m + \llbracket \varkappa \rrbracket [h(\mathbf{x}; a) + h(\mathbf{x} - \mathbf{z}; b)] \} \nabla T_2(\mathbf{x}, \mathbf{x} - \mathbf{z}; a, b) \right. \\ \left. + \llbracket \varkappa \rrbracket [h(\mathbf{x}; a) \nabla T_1(\mathbf{x} - \mathbf{z}; b) + h(\mathbf{x} - \mathbf{z}; b) \nabla T_1(\mathbf{x}; a)] \right) = 0. \quad (4.5c)$$

**4.1. Asymptotic Uncoupling of the Hierarchy.** As we should have expected, the method of functional expansion – as all the other methods – leads to an infinite hierarchy of equations. What makes the difference, however, is that the expansion under consideration is virial and the higher-order functionals contribute to the averaged characteristics quantity proportional to the respective power of  $\gamma$  (or which is the same – to  $c$ ). Respectively the  $n$ -th order kernel appears in the hierarchy always multiplied by  $\gamma^n$  (or  $c^n$ ). As far as  $c < 1$  one can effectively disconjugate the hierarchy developing the kernels in to power series with respect to the concentration  $c$ .

$$T_1 = T_{10} + cT_{11} + c^2T_{12} + \dots, \quad T_2 = T_{20} + cT_{21} + c^2T_{22} + \dots .$$

In order to display the gist of the method it is enough to consider the first two kernels. So we discard terms proportional to  $\gamma^n, n \geq 3$ , and combining the terms of the like power we arrive at the following systems of equations (be noted that the expression  $\langle \varkappa \rangle = \varkappa_m + c\llbracket \varkappa \rrbracket$  is already taken into account and the higher order terms in  $c$  are neglected):

$$\nabla \cdot \{ \varkappa_m \nabla T_{10}(\mathbf{x}; a) + \llbracket \varkappa \rrbracket h(\mathbf{x}; a) [\mathbf{G} + \nabla T_{10}(\mathbf{x}; a)] \} = 0, \quad (4.6a)$$

$$\nabla \cdot \left( 2 \{ \varkappa_m + \llbracket \varkappa \rrbracket [h(\mathbf{x}; a) + h(\mathbf{x} - \mathbf{y}; b)] \} \nabla T_{20}(\mathbf{x}, \mathbf{x} - \mathbf{z}; a, b) \right. \quad (4.6b)$$

$$\left. + \llbracket \varkappa \rrbracket [h(\mathbf{x}; a) \nabla T_{10}(\mathbf{x} - \mathbf{z}; b) + h(\mathbf{x} - \mathbf{y}; b) \nabla T_{10}(\mathbf{x}; a)] \right) = 0,$$

$$\nabla \cdot \left\{ [\varkappa_m T_{11}(\mathbf{x}; a) + \llbracket \varkappa \rrbracket h(\mathbf{x}; a) \nabla T_{11}(\mathbf{x}; a)] + 2\llbracket \varkappa \rrbracket \nabla T_0(\mathbf{x}) \mathfrak{S}_P(\mathbf{x}; a) \right. \quad (4.6c)$$

$$\left. + [\varkappa_m + \llbracket \varkappa \rrbracket h(\mathbf{x}; a)] \frac{1}{\langle V \rangle} \int_{\mathbb{R}^4} \nabla T_{10}(\mathbf{x} - \boldsymbol{\xi}_1; a_1) [Q(\mathbf{0}, \boldsymbol{\xi}_1; a, a_1) - 1] P(a_1) d^3 \boldsymbol{\xi}_1 da_1 \right. \\ \left. - \llbracket \varkappa \rrbracket \overline{\nabla T_1}^{(V,P)} + 2\llbracket \varkappa \rrbracket \frac{1}{\langle V \rangle} \int_{\mathbb{R}^4} h(\mathbf{x} - \mathbf{y}; b) \nabla T_2(\mathbf{x} - \mathbf{y}, \mathbf{x}; b, a) P(b) d^3 \mathbf{y} db \right\} = 0.$$

The above set are equation of discontinuous coefficients and their solutions have derivatives that are generalized functions.

**4.2. First-order Solution.** In the first order, only the function  $T_{10}$  contributes. This function is a solution of Eq. (4.6a) and is nothing else, but the perturbation to a linear temperature field due to the presence of single sphere of radius  $a$ . It has the

following well-known form

$$T_{10}(\mathbf{x}; a) = \begin{cases} -\beta \mathbf{G} \cdot \mathbf{x} & \text{for } |\mathbf{x}| \leq a, \\ -\beta \frac{a^3}{|\mathbf{x}|^3} \mathbf{G} \cdot \mathbf{x} & \text{for } |\mathbf{x}| > a, \end{cases} \quad \beta = \frac{[\varkappa]}{\varkappa_f + 2\varkappa_m}. \quad (4.7)$$

From this expression we can find the volume average (see Eq. (4.3)) of  $T_{10}$

$$\overline{\nabla T_{10}}^{V_a} = \frac{1}{V_a} \int_{\mathbb{R}^3} h(\mathbf{x}; a) \nabla T_{10}(\mathbf{x}; a) d^3 \mathbf{x} = \frac{1}{V_a} \iiint_{V_a} (-\beta \mathbf{G}) d^3 \mathbf{x} = -\beta \mathbf{G}. \quad (4.8)$$

**4.3. Solution for the Second Kernel.** The physical meaning of the solution to the second equation becomes clearer if the following notation is introduced

$$T^{(2)}(\mathbf{x}, \mathbf{x} - \mathbf{z}; a, b) = 2T_{20}(\mathbf{x}, \mathbf{x} - \mathbf{z}; a, b) + T_{10}(\mathbf{x}; a) + T_{10}(\mathbf{x} - \mathbf{z}; b),$$

and the respective equation is derived from Eq. (4.6a) and (4.6b)

$$\nabla \cdot \left( \{ \varkappa_m + [\varkappa] [h(\mathbf{x}; a) + h(\mathbf{x} - \mathbf{z}; b)] \} [\mathbf{G} + \nabla T^{(2)}(\mathbf{x}, \mathbf{x} - \mathbf{z}; a, b)] \right) = 0.$$

The solution to the last equation is merely the perturbation to a linear temperature field due to the presence of two spheres of radii  $a$  and  $b$  respectively, first of the spheres centered in the origin and the second – in the point  $\mathbf{z}$ . Now it becomes transparent that  $T_{20}$  gives the additional perturbation, the perturbation due to the binary interaction between their fields, to the sum of the perturbation due to the individual presence of each sphere. This additional perturbation is introduced as “pure” perturbation in [25], and shown to be orders of magnitude smaller than  $T_{10}$  for moderate and large distances between the spheres. In [26], it was shown that  $T_{20}$  about 20% of  $T_{10}$  even for spheres that almost touch each other.

**4.4. Equation for  $T_{11}$ .** Following the argument from the previous section, we neglect the kernel  $T_{20}$  in Eq. (4.6c) to obtain

$$\begin{aligned} \nabla \cdot \left\{ [\varkappa_m + [\varkappa] h(\mathbf{x}; a)] \nabla T_{11}(\mathbf{x}; a) + 2[\varkappa] \nabla T_0(\mathbf{x}) \mathfrak{S}_P(\mathbf{x}; a) - [\varkappa] \overline{\nabla T_{10}}^{(V,P)} \right. \\ \left. + [\varkappa_m + [\varkappa] h(\mathbf{x}; a)] \frac{1}{\langle V \rangle} \int_{\mathbb{R}^4} \nabla T_{10}(\mathbf{x} - \boldsymbol{\xi}_1; a_1) [Q(\mathbf{0}, \boldsymbol{\xi}_1; a, a_1) - 1] P(a_1) d^3 \boldsymbol{\xi}_1 da_1 \right\} = 0. \end{aligned} \quad (4.9)$$

The derivation of this equation is one of the main results of the present work. Acknowledging Eq. (4.8) shows that  $-\beta \mathbf{G}$  will act as a source term for  $T_{11}$ . In its virtue of an equation with discontinuous coefficient and discontinuous forcing terms, Eq. (4.9) contains the jump conditions on the flux related to the function  $T_{11}$ . In order to find the actual solution one needs to evaluate also the last integral in Eq. (4.9), which is a formidable task on its own, and will be treated elsewhere.

### 5. THE INTERACTION FUNCTION

Both terms in the integrand of the integral Eq. (4.1) that defines the interaction functions are nonzero only inside the supports of the two step functions:  $h(\mathbf{x}; a)$  and  $[Q(\mathbf{0}, \mathbf{y}; a, b) - 1]$ , i.e., inside the spheres of radius  $a$ ,  $a + b$ , respectively. The center of the first sphere is considered to be in the origin of the coordinate system, where the position vector of the center of the second sphere is  $\mathbf{y}$ . The integral is, in fact, the volume of the bodies defined by the intersections of the two spheres (see Fig. 1-(a)), Expressions for the volume in the region under consideration when the

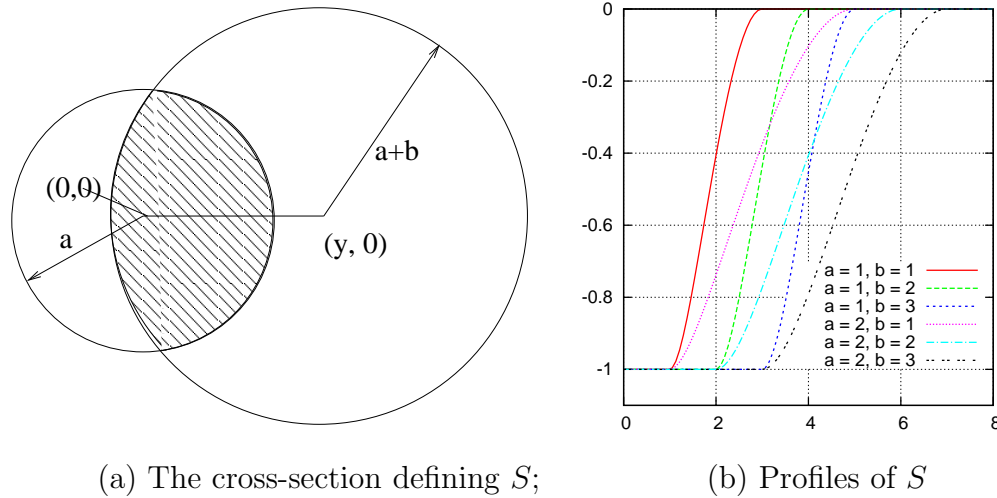


FIGURE 1. Function  $S(y)$  for several sets of the parameters.

spheres intersect (namely when  $b < y \leq 2a + b$ ) can be found in [27]. The simple geometric consideration reveals that the function  $S$  in the three dimensional case is function only of the modulus of its spatial variable  $y \stackrel{\text{def}}{=} |\mathbf{y}|$ . We can consider three different situations

- (i) For  $y \geq 2a + b$  the spheres have no common points, and then  $S(y; a, b) = 0$ .
- (ii) For  $b < y \leq 2a + b$ , the integrand is nonzero and equal to the volume of the body formed by the intersection of the two spheres.
- (iii) For  $y \leq b$  the smaller sphere of radius  $a$  lies entirely in the bigger one, and hence the volume of the intersection is simply the volume of the smaller sphere.

Finally,

$$S(\mathbf{y}; a, b) = \begin{cases} 0 & y = |\mathbf{y}| \geq 2a + b, \\ -\frac{(2a + b - y)^2[-3b^2 + 2(2a + b)y + y^2]}{16ya^3} & b < y = |\mathbf{y}| \leq 2a + b, \\ -1 & y = |\mathbf{y}| \leq b. \end{cases} \quad (5.1)$$

The above expression for the function  $S$  is visualized in Fig. 1-(b) for a couple of different values of the parameters  $a$  and  $b$ .

## 6. THE EFFECTIVE MODULUS

The effective modulus can be computed by evaluating the average flux from Eq. (4.5a) after the solutions for kernels have already been solved.

**6.1. First Order of Approximation.** In order to elucidate the application of RPA to identifying the effective transport coefficient, we outline the procedure for the first-order approximation. First, we observe that  $\nabla[\beta\mathbf{G}\cdot\xi] \equiv \nabla(\beta\mathbf{G})\cdot\xi + \beta\mathbf{G} = \beta\mathbf{G}$ . Then,

$$\begin{aligned} \llbracket\kappa\rrbracket \gamma \int_0^\infty \left[ \iiint_{V_a} \nabla T_{10}(\xi; a) d^3\xi \right] P(a) da &= -\llbracket\kappa\rrbracket \gamma \int_0^\infty \left[ \iiint_{V_a} \beta\mathbf{G} d^3\xi \right] P(a) da \\ &= -\frac{4\pi}{3} \llbracket\kappa\rrbracket \gamma \beta\mathbf{G} \int_0^\infty a^3 P(a) da = -\llbracket\kappa\rrbracket \beta\gamma \langle V \rangle = -\llbracket\kappa\rrbracket \beta c. \end{aligned}$$

The first line of Eq. (4.5a) gives within the order  $O(c^2)$

$$\begin{aligned} \kappa_{\text{eff}}\mathbf{G} \equiv \langle \kappa(\mathbf{x})\nabla T(\mathbf{x}) \rangle &= \langle \kappa \rangle \mathbf{G} - \llbracket\kappa\rrbracket \beta c \mathbf{G} = [\kappa_m + c\llbracket\kappa\rrbracket (1 - \beta)] \mathbf{G} \\ &= \left( \kappa_m + c\llbracket\kappa\rrbracket \frac{3\kappa_m}{2\kappa_m + \kappa_f} \right) \mathbf{G} = \kappa_m (1 + 3c\beta) \mathbf{G}, \quad (6.1) \end{aligned}$$

where the expression for  $\beta$ , Eq. (4.7), is acknowledged. The effective coefficient is obtained from the above equation after dividing the two sides by  $\mathbf{G}$ . This is the celebrated Maxwell's result for the effective modulus. As shown above, a double integral involving  $h$  and  $Q_{12}$  has to be evaluated, in order to find the contribution of the first kernel  $T_1$  to the second-order approximation of the effective modulus.

**6.2. Contribution of  $T_{10}$  to the second order.** Now, let us consider the second integral in Eq. (4.5a) which will contribute to second order approximation of the volume fraction:

$$\begin{aligned} \Phi_1 &= \llbracket\kappa\rrbracket \gamma^2 \int_{\mathbb{R}^4} \nabla T_1(\mathbf{y}; b) \mathfrak{S}_P(\mathbf{y}; a) P(a) d^3\mathbf{y} da \\ &= \llbracket\kappa\rrbracket \frac{c^2}{\langle V \rangle} \int_0^\infty \int_0^\infty \Psi(a, b) P(a) P(b) da db, \\ \text{where } \Psi(a, b) &= \frac{1}{\langle V \rangle} \int_{\mathbb{R}^3} V_a S(\mathbf{y}; a, b) \nabla T_1(\mathbf{y}; b) d^3\mathbf{y}. \quad (6.2) \end{aligned}$$

The above function is evaluated in the Appendix C, so we find that

$$\Phi_1 = \llbracket\kappa\rrbracket \frac{c^2}{\langle V \rangle^2} \int_{V_a} \int_{V_b} \beta\mathbf{G} \left( \frac{4\pi}{3} \right)^2 a^3 b^3 P(a) P(b) da db = \llbracket\kappa\rrbracket c^2 \beta \mathbf{G},$$

because

$$\int_0^\infty \int_0^\infty \left( \frac{4\pi}{3} \right)^2 a^3 b^3 P(a) P(b) da db = \langle V \rangle^2.$$

**6.3. Contribution of  $T_{11}$  to the second order.** The second-order contribution is connected with the volume-average term (see Eq. (4.3)) that enters Eq. (4.5a). Since the term is proportional to the volume fraction  $c$ , then, in order to evaluate the second-order contribution we need just the knowledge about the  $T_{11}$  inside the sphere. Then

$$\Phi_2 = \llbracket \varkappa \rrbracket c^2 \int_0^\infty \overline{\nabla T_{11}}^{V_a} da = \llbracket \varkappa \rrbracket c^2 \int_0^\infty \left[ \frac{1}{\langle V \rangle} \iiint_{V_a} \nabla T_{11}(\mathbf{x}; a) d^3 \mathbf{x} \right] da. \quad (6.3)$$

Thus we have closed the description on the conceptual level: the solution of Eq. (4.9) for  $T_{11}$  must be used in the last formula in order to find the contribution  $\Phi_2$ , and thus the full contribution of the first kernel (as represented by the functions  $T_{10}$  and  $T_{11}$ ) to the second-order approximation of the effective coefficient.

## 7. CONCLUSION

In the present paper, the method Volterra–Wiener functional expansion (VWE) with random point function (RPF) basis is applied to the problem of identification of the effective heat conduction modulus of a polydisperse spherical suspension. A special attention is paid to the role of the two-point probability distributions of the system of random points generated by the randomly distributed centers of spherical inclusions. We limit ourselves to the case of Random Point Functions of Perfect Disorder type, when the normalized two-point correlations are either zero when the points are close than the sum of the respective radii, or equal to one (Poisson distribution of the generating system of random points) when the points are far enough. The description requires the evaluation of some quadratures of the shape function and the two-point correlations, which is done in the paper. The viriality of the VWE is made use, and a perturbation expansion with respect to the powers of the volume fraction is used to derive the governing equations for the first- and second- order kernels. The quadratures that give the contribution to the effective modulus are derived, and some of them are solved. Thus a self-consistent description of the problem is achieved.

## ACKNOWLEDGEMENT

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## APPENDIX. THE INTEGRAL INVOLVING $T$ AND $S$

Being reminded that  $S$  is a function of  $y = |\mathbf{y}|$  only, and that it has a finite support, we can introduce spherical coordinates  $(y, \phi, \theta)$

$$\mathbf{y} = y \sin \theta \cos \phi \mathbf{i} + y \sin \theta \sin \phi \mathbf{j} + y \cos \theta \mathbf{k},$$

where  $\mathbf{i}, \mathbf{j}, \mathbf{k}$  are the unit Cartesian vectors. The ranges of the coordinates are  $-\infty < y < +\infty$ ,  $0 \leq \phi < 2\pi$  and  $0 \leq \theta \leq \pi$  and the Jacobian ( $J$ ) is  $y^2 \sin \theta$ . We compute the gradient of the temperature field  $T_{10}$  from Eq. (4.7) as follows

$$\nabla T_{10}(\mathbf{y}; b) = \begin{cases} -\beta \mathbf{G} & \text{for } |\mathbf{y}| \leq b, \\ \beta \frac{a^3}{y^3} \left[ -\mathbf{G} + 3 \frac{\mathbf{y}(\mathbf{G} \cdot \mathbf{y})}{y^2} \right] & \text{for } |\mathbf{y}| > b. \end{cases}$$

By using appropriate expression for  $S$ , we can formulate  $\Psi(a, b)$  in the following way

$$\begin{aligned} \Psi(a, b) = & \int_0^\pi \int_0^{2\pi} \int_0^b \beta \mathbf{G} \frac{4\pi}{3} a^3 y^2 \sin \theta \, dy \, d\phi \, d\theta \\ & + \int_0^\pi \int_0^{2\pi} \int_b^{2a+b} \beta \frac{a^3}{y^3} \left[ -\mathbf{G} + 3 \frac{\mathbf{y}(\mathbf{G} \cdot \mathbf{y})}{y^2} \right] S(y; a, b) y^2 \sin \theta \, dy \, d\phi \, d\theta. \end{aligned}$$

Now, the second volume integral from the above equality can be rewritten as follows

$$- \int_b^{2a+b} \beta \frac{a^3}{y^3} \left\{ \int_0^\pi \int_0^{2\pi} \left[ -\mathbf{G} + 3 \frac{\mathbf{y}(\mathbf{G} \cdot \mathbf{y})}{y^2} \right] \sin \theta \, d\phi \, d\theta \right\} S(y; a, b) \, dy.$$

In terms of the spherical coordinates we get

$$\begin{aligned} \mathbf{y}(\mathbf{G} \cdot \mathbf{y}) = & y^2 [G_1 \sin \theta \cos \phi + G_2 \sin \theta \sin \phi + G_3 \cos \theta] (\sin \theta \cos \phi \mathbf{i} + \sin \theta \sin \phi \mathbf{j} + \cos \theta \mathbf{k}), \\ \int_0^\pi \int_0^{2\pi} \left[ -\mathbf{G} + 3 \frac{\mathbf{y}(\mathbf{G} \cdot \mathbf{y})}{y^2} \right] \sin \theta \, d\phi \, d\theta = & - \int_0^\pi \int_0^{2\pi} \mathbf{G} \sin \theta \, d\phi \, d\theta + 3I, \\ I = & \int_0^\pi \int_0^{2\pi} \left\{ \mathbf{i} [G_1 \sin^3 \theta \cos^2 \phi + G_2 \sin^3 \theta \sin \phi \cos \phi + G_3 \cos \theta \sin^2 \theta \cos \phi] \right. \\ & + \mathbf{j} [G_1 \sin^3 \theta \cos \phi \sin \phi + G_2 \sin^3 \theta \sin^2 \phi + G_3 \cos \theta \sin^2 \theta \sin \phi] \\ & \left. + \mathbf{k} [G_1 \sin^2 \theta \cos \theta \cos \phi + G_2 \sin^2 \theta \cos \theta \sin \phi + G_3 \sin \theta \cos^2 \theta] \right\} d\phi \, d\theta. \quad (\text{A.1}) \end{aligned}$$

In the above formula most of the integrals are zero, except the following

$$\int_0^{2\pi} \cos^2 \phi \, d\phi = \int_0^\pi \sin^2 \phi \, d\phi = \pi, \quad \int_0^\pi \sin \theta \cos^2 \theta \, d\theta = \frac{2}{3}, \quad \int_0^\pi \sin^3 \theta \, d\theta = \frac{4}{3},$$

which gives  $I = \pi \frac{4}{3} G_1 \mathbf{i} + \pi \frac{4}{3} G_2 \mathbf{j} + 2\pi \frac{2}{3} G_3 \mathbf{k} = \frac{4\pi}{3} \mathbf{G}$ . This means that the value of the integral in Eq. (A.1) is zero. Thus we get for the function  $\Psi$  only the contribution from the first integral, namely

$$\Psi(a, b) = \int_0^\pi \int_0^{2\pi} \int_0^b \beta \mathbf{G} \frac{4\pi}{3} a^3 y^2 \sin \theta \, dy \, d\phi \, d\theta = \beta \mathbf{G} \left( \frac{4\pi}{3} \right)^2 a^3 b^3.$$

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