

## STOCHASTIC FUNCTIONAL EXPANSION FOR HEAT CONDUCTIVITY OF POLYDISPERSE PERFECTLY DISORDERED SUSPENSIONS

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**Христо Христов, Константин Марков: Применение стохастических функциональных рядов для исследования теплопроводности в полидисперсных совершенно беспорядочных суспензиях.** Применяются функциональные разложения Вольтерра — Винера для исследования переноса тепла в полидисперсном совершенно беспорядочном композите из полидисперсных сферических частиц. Выводятся уравнения для ядер и при помощи вириальности функционального ряда с точечной случайной базисной функцией ставятся замкнутые граничные задачи для двух первых ядер. Показано, что первое ядро совпадает с решением для смещения линейного температурного поля, вносимое одиночной сферой. Полное статистическое решение получено с первым порядком аппроксимации по концентрации частичной фазы и эффективный коэффициент теплопроводности подсчитан с тем же порядком точности. Оказывается, что он совпадает с соответствующим коэффициентом для случая одинаковых сфер, т. е. распределение сфер по размерам не оказывает влияния на значения эффективного коэффициента в первом порядке аппроксимации по концентрации.

Christo Christov, Konstantin Markov. **Stochastic functional expansion for heat conductivity of polydisperse perfectly disordered suspensions.** The problem of random heat conduction through a polydisperse suspension is solved by means of Volterra-Wiener functional expansions. Equations for the kernels of stochastic integrals are derived and using the virial property of the functional expansions with random point basis function, coupled boundary value problems are posed for the first two kernels. The first kernel is shown to coincide with the solution for the perturbation to a linear temperature field due to a single inclusion. The full statistical solution is obtained within the first order of approximation with respect to the concentration of the filler, and the overall heat conductivity is evaluated to the same order. It turns out that this quantity coincides with the quantity for composites of equisized spheres, i. e. it is not affected by the size distribution within the first order of approximation.

### INTRODUCTION

One of the central issues of the modern mechanics of continuous media is the problem of identification of the mechanical behaviour of particulate two-phase materials on the base of the respective properties of the constituents. Although an extensive literature is now available a little progress is made on the way of rigorous treatment of particulate media. This is especially true for the materials of disordered microstructure, where incorporating the knowledge about the statistics of material is based as a rule on quite arbitrary hypothesis on every stage of modelling (see, e.g., [1, 2, 3, 4]).

A new approach to the said problem is originated in authors' works [5, 6, 7, 8]. It is based on the concept that the stochastic fields arising in particulate media can be successfully approximated by truncated Volterra-Wiener functional series with random point basis function. These series turns out to be virial and each higher-order term contributes to the average characteristics quantities proportional to the respective degree of concentration (volume fraction) of inclusions. As the latter is always lesser than unity, a small parameter arises and allows one to obtain asymptotically correct results.

The simplest random point basis function that can be employed is the Poisson random function. Despite its obvious shortcoming connected with the overlapping of spheres the Poisson basis function gives correct results within the first order of approximation. The requirement for non-overlapping is incorporated into the statistics of the basis function in [7, 8] for the case of equi-sized spheres which are "perfectly well distributed" in the matrix. A case of suspension that is not of perfectly disordered type is discussed in [9] acknowledging a kind of clustering and in [10] where are outlined the classes of statistics for which the functional expansion can still be rendered virial and in turn the problem for the different kernels can be decoupled.

Another way of generalization is to consider polydisperse suspensions. This brings into view the notion of marked random point function serving as a basis function with the random radii of spheres playing the role of the mark [11]. For perfectly disordered polydisperse suspensions the series can still be rendered orthogonal (for general consideration see [12]). The latter allows us to extend the results of [7] to polydisperse case and this is the objective of the present paper. The main purpose for solving this problem is to check whether the size distribution influences the results for the overall heat conductivity.

## 1. TRANSPORT COEFFICIENTS OF PARTICULATE MEDIA AND THE MARKED RANDOM POINT FUNCTIONS

### 1.1. Generalized random density function

Consider a system of random points  $\{x_\alpha\}$  dispersed throughout a region  $D \in \mathbf{R}^3$ . As the considerations to follow are not subject to the specific shape of the region  $D$  we assume the latter to occupy the entire space  $\mathbf{R}^3$ . Let us also assume that at each random position  $x_\alpha$  a certain random volume  $V$  is centered which is defined by its characteristic function

$$(1.1) \quad h(x; u) = \begin{cases} 1 & \text{if } x \in V \\ 0 & \text{if } x \notin V, \end{cases}$$

where  $u \in U$  is the random vector of parameters specifying the shape of  $V$  and called hereafter "mark". Respectively  $U$  is the mark space. In general, the mark vector may incorporate other than mere geometrical parameters. For instance, it can consist also of the components of the velocity of the centre of volume in case it is moving in the space. As far as the present paper is concerned with composite materials we consider the case when the mark vector is comprised only by geometrical parameters, and more specifically — by the random radius of the respective sphere centred at the random position  $x_\alpha$ . Therefore the mark space is  $U = \mathbf{R}_+^1 = \{a \mid a > 0\}$ .

The coefficient of heat conductivity of the polydisperse suspension of spheres has the form

$$(1.2) \quad \kappa(\mathbf{x}) = \kappa_m + [\kappa] \sum_{\alpha} h(\mathbf{x} - \mathbf{x}_{\alpha}; a_{\alpha}),$$

where  $\kappa_m$  is the conductivity of the continuous phase (the matrix),  $\kappa_f$  — the conductivity of the particulate phase (the filler), and  $[\kappa] \equiv \kappa_f - \kappa_m$  is the jump of the coefficient.

The function  $\kappa(\mathbf{x})$  can be formally represented by the following Lebesgue integral:

$$(1.3) \quad \kappa(\mathbf{x}) = \kappa_m + \int_{\mathbb{R}^3} \int_{\mathbf{U}} h(\mathbf{x} - \xi; a) \omega(\xi, a) d^3 \xi da,$$

where

$$(1.4) \quad \omega(\mathbf{x}, a) \equiv \omega(\mathbf{r}) = \sum_{\alpha} \delta(\mathbf{r} - \mathbf{r}_{\alpha})$$

is called generalized random density function (see [11, 12]) and  $\mathbf{r} \equiv (x^1, x^2, x^3, a) \in \mathbb{R}^3 \times \mathbf{U}$ . In the above formula  $\delta(\cdot)$  stands for the Dirac delta function of 4-th order.

## 1.2. Description of sets of marked random points

Formulae (1.3) and (1.4) mean that the statistical properties of the function  $\kappa(\mathbf{x})$  are fully defined by the respective characteristics of the generalized random density function  $\omega(\xi; a)$ . In turn, the statistical characteristics of  $\omega(\xi; a)$  are defined by the set of marked random points  $\{\mathbf{x}_{\alpha}, a_{\alpha}\}$ . The way to connect the correlation properties of the random density function to the multi-point probability densities of its generating set of random points is outlined in [13] and generalized to the case of marked points in [12]. A major role is played by the so-called "local" characteristics (see [14])  $F_n$  that define the elementary probabilities

$$(1.5) \quad dP = F_n(\mathbf{x}_1, \dots, \mathbf{x}_n; a_1, \dots, a_n) d^3 \mathbf{x}_1 \dots d^3 \mathbf{x}_n da_1 \dots da_n$$

of simultaneous occurrence in the vicinities of  $n$  geometric positions  $\mathbf{x}_i$  of  $n$  random points each of them with a mark in the vicinity of the value  $a_i$ .

It is convenient to introduce the notation

$$(1.6) \quad F_n = f_n(\mathbf{x}_1, \dots, \mathbf{x}_n; a_1, \dots, a_n) P_n(a_1, \dots, a_n),$$

where  $P_n$  is the multivariate probability density of the mark. This density is to satisfy the usual conditions

$$(1.7) \quad \int_{\mathbf{U}} P_n(a_1, \dots, a_{n-1}, a_n) da_n = P_{n-1}(a_1, \dots, a_{n-1}), \quad P_0 = 1,$$

for  $n = 1, 2, \dots$

Conditions on  $f_n$  are more complicated and can be defined through the so-called (see [14]) "global" characteristics of the random set. One is referred to [12] for details.

It is obvious that there exists an asymmetry between the spatial coordinates  $x_\alpha$  and marks  $a_\alpha$ . Henceforth the variable  $x$  is distinguished only when the said asymmetry is important. Otherwise, the abbreviate notation  $r$  is used.

Along with  $F_n$  the so-called (see [13], [14]) correlation functions  $G_n$  of the probability distribution are frequently used to describe a set of random points. For these functions we have the relation (see [12])

$$(1.8) \quad G_k(x_1, \dots, x_k; a_1, \dots, a_k) = \sum_{k_1 + \dots + k_p = k} \{F_1^{k_1} \dots F_p^{k_p}\}_{\text{sym}} \\ \times \frac{k!(-1)^{(k_1 + \dots + k_p - 1)} (k_1 + \dots + k_p - 1)}{k_1!(1!)^{k_1} \dots k_p!(p!)^{k_p}},$$

where  $F_p^{k_p}$  stands for the product  $\prod_{i=1}^{k_p} F_p(r_{i_1}, \dots, r_{i_p})$  and does not mean the respective power.

### 1.3. Defining the statistics of marked random point function through the respective properties of the set of marked random points

The relationship between the moments of the generalized random density function  $\omega$  and the multipoint probability densities  $F_n$  of the generating set of random points stems from the connection between the characteristic and generating functionals. The general formulae can be found in [12]. The first couple of them read

$$(1.9) \quad \langle \omega(r) \rangle = F_1(r) \\ \langle \omega(r_1) \omega(r_2) \rangle = F_1(r_1) \delta(r_1 - r_2) + F_2(r_1, r_2) \\ \langle \omega(r_1) \omega(r_2) \omega(r_3) \rangle = F_1(r_1) \delta(r_1 - r_2) \delta(r_1 - r_3) \\ + 3\{\delta(r_1 - r_3) F_2(r_1, r_2)\}_{\text{sym}} + F_3(r_1, r_2, r_3),$$

where  $\{\cdot\}_{\text{sym}}$  denotes a symmetrization with respect to all non-trivial combinations of arguments.

It is interesting to note that functions  $F_n$  are related to the so-called factorial moments ([15], [16]). Let us denote

$$(1.10) \quad \Delta^{(n)}[\omega] = \omega(r_1) [\omega(r_2) - \delta(r_1 - r_2)] \dots [\omega(r_n) - \delta(r_1 - r_n)] \\ - \dots - \delta(r_{n-1} - r_n).$$

For the average value of the above expression one has (see [11], [12])

$$(1.11) \quad \langle \Delta^{(n)}[\omega] \rangle = F_n(r_1, \dots, r_n).$$

#### 1.4. The notion of perfect disorder

The marked random functions in general form too rich a class and cover virtually all practically important area of heterogeneous media. If our aim is to reach certain more tangible results we must narrow this class of functions, moreover that usually a detailed information is available neither for the locations, nor for the radii of particles. A reasonable simplification seems then to be the assumption of Perfect Disorder (PD), originally proposed by Kröner [18] as a model of random media whose properties at different points are statistically independent. This notion proved useful and was later on discussed and modified by a number of authors. For a suspension of equi-sized spheres the definition of PD is presented in [7] and the respective random field is named Perfect Disorder of Spheres (PDS). For marked random volumes the definition is generalized in [12] arriving to the Perfect Disorder of Marked Volumes (PDMV). For the particular case under consideration, the conditions for PD can be specified as follows:

(i) The marks  $a_i$  of an arbitrary sample of  $n$  random points are mutually statistically independent, regardless to the specific geometric positions  $\mathbf{x}_i$  at which they are situated:

$$(1.12) \quad P_n(a_1, \dots, a_n) = P(a_1) \dots P(a_n),$$

where  $P(a)$  is the probability density of a mark.

(ii) The space positions  $\mathbf{x}_i$  at which occur  $n$  arbitrary random points are independent, i.e.:

$$(1.13) \quad f_n(\mathbf{x}_1, \dots, \mathbf{x}_n; a_1, \dots, a_n) = \gamma^n,$$

where  $\gamma$  is the number of points per unit volume.

Note that the system  $\{\mathbf{x}_i\}$  with the properties (i), (ii) is just the compound Poisson system studied in detail in [19] when the independent coordinate is the time. If we are to consider, however, a system of random volumes, then one more requirement is to be imposed, namely:

(iii) The probability to find  $n$  random points at  $n$  prescribed spatial locations  $\mathbf{x}_i$ ,  $i = 1, \dots, n$ , is equal to zero if at least two of the respective random volumes that are centered at these points overlap each other.

The marked random system with properties (i)—(iii) is named (see [11]) Perfect Disorder of Marked Spheres of Random Radii (PDSRR). The above requirements can be expressed in the following more concise way:

$$(1.14) \quad F_n(\mathbf{r}_1, \dots, \mathbf{r}_n) = \gamma^n P(a_1) \dots P(a_n) Q_{12} \dots Q_{1n} Q_{23} \dots Q_{2n} \dots Q_{n-1,n},$$

where

$$(1.15) \quad Q_{ik} \equiv Q(\mathbf{x}_i, \mathbf{x}_k; a_i, a_k) = \begin{cases} 0 & \text{if } |\mathbf{x}_i - \mathbf{x}_k| \leq a_i + a_k \\ 1 & \text{if } |\mathbf{x}_i - \mathbf{x}_k| > a_i + a_k. \end{cases}$$

Obviously, when the spheres centered at points  $\mathbf{x}_i$  do not overlap, then (1.13) holds. Otherwise  $F_n = 0$  which means that the probability to find such a combination of spheres and spatial positions of their centres is equal to zero. Application of functions  $F_2$  is discussed in [2], [20].

Note that the PDS system is related to the Poisson system (PDP or Perfect Disorder of Points) in the same manner as the PDSRR system is related to the PDMP (Perfect Disorder of Marked Points or Compound Poisson) System. Both PDS and PDSRR systems take into account the finite size of the spheres (Fig. 1).

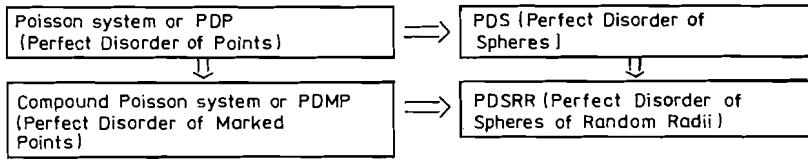


Fig. 1. Varioys modifications of the definition of Perfect Disorder. The Horizontal arrows indicate a generalization to systems of non-overlapping volumes; vertical arrows — to systems with marks

## 2. VOLTERRA-WIENER FUNCTIONAL EXPANSION

### 2.1. Generalized Charlier polynomials

It was Ogura [21] who found out that the functional expansion with Poisson basis function can be rendered orthogonal by means of Charlier polynomials. Applicability of these polynomials was extended to PDS basis functions in [7] under certain fully natural restrictions on kernels. In [12] generalized Charlier polynomials of the random density functions of Perfect-Disorder type are introduced. For the PDSRR basis function they read

$$\begin{aligned}
 (2.1) \quad C_{\omega}^{(0)} &= 1, \quad C_{\omega}^{(1)} = \omega(\mathbf{r}_1) - \gamma P(a_1), \\
 C_{\omega}^{(2)}(\mathbf{r}_1, \mathbf{r}_2) &= \omega(\mathbf{r}_1) [\omega(\mathbf{r}_2) - \delta(\mathbf{r}_1 - \mathbf{r}_2)] \\
 &\quad - \gamma [P(a_2) \omega(\mathbf{r}_1) + P(a_1) \omega(\mathbf{r}_2)] + \gamma^2 P(a_1) P(a_2), \\
 &\quad \dots \dots \dots \\
 C_{\omega}^{(n)}(\mathbf{r}_1, \dots, \mathbf{r}_n) &= \Delta_{\omega}^{(2)}(\mathbf{r}_1, \dots, \mathbf{r}_n) \\
 &\quad - \gamma \binom{n}{n-1} \{ \Delta_{\omega}^{(n-1)}(\mathbf{r}_1, \dots, \mathbf{r}_{n-1}) P(a_n) \}_{\text{sym}} \\
 &\quad \dots \dots \dots \\
 &\quad + (-\gamma)^{n-2} \binom{n}{2} \{ \Delta_{\omega}^{(2)}(\mathbf{r}_1, \mathbf{r}_2) P(a_3) \dots P(a_n) \}_{\text{sym}} \\
 &\quad + (-\gamma)^{n-1} \binom{n}{1} \{ \omega(\mathbf{r}_1) P(a_2) \dots P(a_n) \}_{\text{sym}} + (-\gamma)^n P(a_1) \dots P(a_n).
 \end{aligned}$$

For PDMP basis functions the generalized Charlier polynomials are centered random variables that are orthogonal in stochastic sense. If random volumes are involved this ceases to be true. For the sake of convenience we give here the

explicit expressions for the second and third moments of the first-order generalized Charlier polynomials of the PDSRR basis function  $\omega$ :

$$(2.2) \quad \begin{aligned} M^{(2)}(\mathbf{r}_1, \mathbf{r}_2) &\equiv \langle C_{\omega}^{(1)}(\mathbf{r}_1) C_{\omega}^{(1)}(\mathbf{r}_2) \rangle \\ &= \gamma P(a_1) \delta(\mathbf{r}_1 - \mathbf{r}_2) + \gamma^2 P(a_1) P(a_2) (Q_{12} - 1) \end{aligned}$$

and

$$(2.3) \quad \begin{aligned} M^{(3)}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3) &\equiv \langle C_{\omega}^{(1)}(\mathbf{r}_1) C_{\omega}^{(1)}(\mathbf{r}_2) C_{\omega}^{(1)}(\mathbf{r}_3) \rangle \\ &= 3\gamma^2 \{P(a_1) P(a_3) \delta(\mathbf{r}_1 - \mathbf{r}_2) (Q_{13} - 1)\}_{\text{sym}} \\ &\quad + \gamma^3 P(a_1) P(a_2) P(a_3) [Q_{12} Q_{23} Q_{31} - Q_{12} - Q_{23} - Q_{31} + 2], \end{aligned}$$

where  $Q_{ij}$  are defined in (1.15).

An essential property of the generalized Charlier polynomials is the decomposition of a multivariate polynomial into a product of first-order polynomials when the arguments are not equal

$$(2.4) \quad C_{\omega}^{(n)}(\mathbf{r}_1, \dots, \mathbf{r}_n) = C_{\omega}^{(n)}(\mathbf{r}_1) \dots C_{\omega}^{(n)}(\mathbf{r}_n), \quad \mathbf{r}_i \neq \dot{\mathbf{r}}_j.$$

For the PDSRR basis functions (2.4) is true when the arguments are well separated, i. e. when

$$(2.5) \quad |\mathbf{x}_i - \mathbf{x}_j| > a_i + a_j.$$

General formulae for the third and second moments of multivariate polynomials are obtained in [12] making use of (2.2) — (2.5), but they are too lengthy to be presented here. Rather we just cite the first couple of expressions for those moments:

$$(2.6) \quad \begin{aligned} \langle C_{\omega}^{(2)}(\mathbf{r}_1, \mathbf{r}_2) C_{\omega}^{(2)}(\mathbf{s}_1, \mathbf{s}_2) \rangle &= M^{(2)}(\mathbf{r}_1, \mathbf{s}_1) M^{(2)}(\mathbf{r}_2, \mathbf{s}_2) \\ &\quad + M^{(2)}(\mathbf{r}_1, \mathbf{s}_2) M^{(2)}(\mathbf{r}_2, \mathbf{s}_1), \end{aligned}$$

$$(2.7) \quad \begin{aligned} \langle C_{\omega}^{(3)}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3) C_{\omega}^{(3)}(\mathbf{s}_1, \mathbf{s}_2, \mathbf{s}_3) \rangle \\ &= 6 \{M^{(2)}(\mathbf{r}_1, \mathbf{s}_1) M^{(2)}(\mathbf{r}_2, \mathbf{s}_2) M^{(2)}(\mathbf{r}_3, \mathbf{s}_3)\}_{\text{sym}}, \end{aligned}$$

$$(2.8) \quad \begin{aligned} \langle C_{\omega}^{(1)}(\mathbf{r}_1) C_{\omega}^{(1)}(\mathbf{s}_1) C_{\omega}^{(2)}(\mathbf{t}_1, \mathbf{t}_2) \rangle &= M^{(2)}(\mathbf{t}_1, \mathbf{r}_1) M^{(2)}(\mathbf{t}_2, \mathbf{s}_1) \\ &\quad + M^{(2)}(\mathbf{t}_1, \mathbf{s}_1) M^{(2)}(\mathbf{t}_2, \mathbf{r}_1), \end{aligned}$$

$$(2.9) \quad \begin{aligned} \langle C_{\omega}^{(1)}(\mathbf{r}_1) C_{\omega}^{(2)}(\mathbf{s}_1, \mathbf{s}_2) C_{\omega}^{(2)}(\mathbf{t}_1, \mathbf{t}_2) \rangle \\ &= 4 \{M^{(3)}(\mathbf{t}_1, \mathbf{s}_1, \mathbf{r}_1) M^{(2)}(\mathbf{t}_2, \mathbf{s}_2)\}_{\text{sym}}, \end{aligned}$$

$$(2.10) \quad \begin{aligned} \langle C_{\omega}^{(2)}(\mathbf{r}_1, \mathbf{r}_2) C_{\omega}^{(2)}(\mathbf{s}_1, \mathbf{s}_2) C_{\omega}^{(2)}(\mathbf{t}_1, \mathbf{t}_2) \rangle \\ &= 4 \{M^{(3)}(\mathbf{t}_1, \mathbf{s}_1, \mathbf{r}_1) M^{(3)}(\mathbf{t}_2, \mathbf{s}_2, \mathbf{r}_2)\}_{\text{sym}} \\ &\quad + 8 \{M^{(2)}(\mathbf{t}_1, \mathbf{s}_2) M^{(2)}(\mathbf{t}_2, \mathbf{r}_1) M^{(2)}(\mathbf{s}_1, \mathbf{r}_2)\}_{\text{sym}}, \end{aligned}$$

$$(2.11) \quad \langle C_{\omega}^{(1)}(r_1) C_{\omega}^{(2)}(s_1, s_2) C_{\omega}^{(3)}(t_1, t_2, t_3) \rangle \\ = 6 \{ M^{(2)}(t_1, r_1) M^{(2)}(t_2, s_1) M^{(2)}(t_3, s_2) \}_{\text{sym}},$$

$$(2.12) \quad \langle C_{\omega}^{(1)}(r_1) C_{\omega}^{(3)}(s_1, s_2, s_3) C_{\omega}^{(3)}(t_1, t_2, t_3) \rangle \\ = 18 \{ M^{(3)}(t_1, s_1, r_1) M^{(2)}(t_2, s_2) M^{(2)}(t_3, s_3) \}_{\text{sym}},$$

$$(2.13) \quad \langle C_{\omega}^{(2)}(r_1, r_2) C_{\omega}^{(3)}(s_1, s_2, s_3) C_{\omega}^{(3)}(t_1, t_2, t_3) \rangle \\ = 36 \{ M^{(3)}(t_1, s_1, r_1) M^{(3)}(t_2, s_2, r_2) M^{(2)}(t_3, s_3) \}_{\text{sym}} \\ + 36 \{ M^{(2)}(t_1, s_2) M^{(2)}(t_2, s_3) M^{(2)}(s_1, r_2) M^{(2)}(t_3, r_1) \}_{\text{sym}},$$

$$(2.14) \quad \langle C_{\omega}^{(3)}(r_1, r_2, r_3) C_{\omega}^{(3)}(s_1, s_2, s_3) C_{\omega}^{(3)}(t_1, t_2, t_3) \rangle \\ = 36 \{ M^{(3)}(t_1, r_1, s_1) M^{(3)}(t_2, r_2, s_2) M^{(3)}(t_3, r_3, s_3) \}_{\text{sym}} \\ + 432 \{ M^{(3)}(t_1, s_2, r_3) M^{(2)}(t_2, s_3) M^{(2)}(t_3, r_1) M^{(2)}(s_1, r_2) \}_{\text{sym}},$$

where  $\mathbf{s} = (y^1, y^2, y^3, b)$ ,  $\mathbf{t} = (z^1, z^2, z^3, c)$  are vectors of the type of vector  $\mathbf{r}$  (see (1.4)).

## 2.2. Wiener G-functionals

Consider the functionals of the random density function  $\omega$ :

$$(2.15) \quad G^n[\omega] = \int_{\mathbf{R}^3} \dots \int_{\mathbf{U}} \dots \int K^{(n)}(\mathbf{x} - \mathbf{x}_1, \dots, \mathbf{x} - \mathbf{x}_n; a_1, \dots, a_n) \\ \times C_{\omega}^{(n)}(\mathbf{x}_1, \dots, \mathbf{x}_n; a_1, \dots, a_n) d^3x_1 \dots d^3x_n da_1 \dots da_n.$$

Each of these functionals is a homogeneous in the strict sense stochastic function of the variable  $\omega$ . We call them Wiener  $G$ -functionals though the original meaning of this coinage refers to Wiener-Hermite functionals [22]. A Wiener  $G$ -functional can be rendered centered stochastic variable upon assuming that (see [7])

$$(2.16) \quad K^{(n)}(\mathbf{x}_1, \dots, \mathbf{x}_n; a_1, \dots, a_n) = 0 \text{ for } |\mathbf{x}_i - \mathbf{x}_k| \leq a_i + a_k.$$

This requirement is completely natural for the case under consideration and leads to no loss of generality since in a system of non-overlapping spheres one never has a pair of points  $\mathbf{x}_i, \mathbf{x}_k$  situated closer than  $a_i + a_k$ . Then being reminded of (1.15) after averaging of (2.1) and multiplying by (2.16) one gets

$$(2.17) \quad \langle G^{(n)}[\omega] \rangle = 0, \quad n = 1, 2, \dots$$

Another important corollary of the assumption (2.16) is that the functionals (2.15) become orthogonal in stochastic sense. In order to prove that we invoke (2.4) which indeed holds due to the fact that (2.16) yields (2.5). Then for two different sets of functionals we have:



$$(2.18) \quad \langle G_F^{(n)}[\omega] G_H^{(m)}[\omega] \rangle = 0 \text{ for } n \neq m,$$

$$\begin{aligned} \langle G_F^{(n)} G_H^{(n)} \rangle &= \int_{\mathbf{R}} \dots \int_{\mathbf{U}} \dots \int K_F^{(n)}(\mathbf{x} - \mathbf{x}_1, \dots, \mathbf{x} - \mathbf{x}_n; a_1, \dots, a_n) \\ &\times K_H^{(n)}(\mathbf{y} - \mathbf{y}_1, \dots, \mathbf{y} - \mathbf{y}_n; b_1, \dots, b_n) \{M^{(2)}(\mathbf{r}_{i_1}, \mathbf{s}_{j_1}) \dots M^{(2)}(\mathbf{r}_{i_n}, \mathbf{s}_{j_n})\}_{\text{sym}} \\ &\times d^3 \mathbf{x}_1 \dots d^3 \mathbf{x}_n d^3 \mathbf{y}_1 \dots d^3 \mathbf{y}_n da_1 \dots da_n db_1 \dots db_n, \end{aligned}$$

where the notation of (1.4) is adopted.

In a similar manner are derived formulae for the third moments of Wiener  $G$ -functionals [12].

### 2.3. The functional expansion

Having the functionals (2.7) one can expand a random point function into functional series with respect to random point function  $\omega$ :

$$(2.19) \quad F(\mathbf{x}) = \sum_{n=0}^{\infty} G_F^{(n)}[\omega] \equiv G_{\omega}^{(0)} + \sum_{n=1}^{\infty} \int_{\mathbf{R}} \dots \int_{\mathbf{U}} \dots \int K_F^{(n)}(\mathbf{x} - \mathbf{x}_1, \dots, \mathbf{x} - \mathbf{x}_n) \\ \times C_{\omega}^{(n)}(\mathbf{x}_1, \dots, \mathbf{x}_n; a_1, \dots, a_n) d^3 \mathbf{x}_1 \dots d^3 \mathbf{x}_n da_1 \dots da_n.$$

This expansion contains all the statistical information about the random function  $F(\mathbf{x})$  in the sense that one can calculate its various characteristics such as correlation functions, spectra, etc. For instance, the two-point correlation function is given by

$$(2.20) \quad \begin{aligned} \langle F(\mathbf{x}) H(\mathbf{y}) \rangle &= G_F^{(0)} G_H^{(0)} + \int_{\mathbf{R}} \int_{\mathbf{U}} K_F^{(1)}(\mathbf{x} - \mathbf{x}_1; a_1) K_H^{(1)}(\mathbf{y} - \mathbf{y}_1; b_1) \\ &\times M^{(2)}(\mathbf{x}_1, \mathbf{y}_1; a_1, b_1) d^3 \mathbf{x}_1 d^3 \mathbf{y}_1 da_1 db_1 \\ &+ \int_{\mathbf{R}} \int_{\mathbf{U}} K_F^{(2)}(\mathbf{x} - \mathbf{x}_1, \mathbf{x} - \mathbf{x}_2; a_1, a_2) K_H^{(2)}(\mathbf{y} - \mathbf{y}_1, \mathbf{y} - \mathbf{y}_2; b_1, b_2) \\ &\times [M^{(2)}(\mathbf{x}_1, \mathbf{y}_1; a_1, b_1) M^{(2)}(\mathbf{x}_2, \mathbf{y}_2; b_2, a_2) \\ &+ M^{(2)}(\mathbf{x}_1, \mathbf{y}_2; a_1, b_2) M^{(2)}(\mathbf{x}_2, \mathbf{y}_1; a_2, b_1)] \\ &\times d^3 \mathbf{x}_1 d^3 \mathbf{x}_2 d^3 \mathbf{y}_1 d^3 \mathbf{y}_2 da_1 da_2 db_1 db_2 + \dots \end{aligned}$$

Taking a more close look at formulae (2.2) and (2.3) one sees that  $M^{(2)}$  and  $M^{(3)}$  contain terms proportional to the powers of the number of points per unit volume  $\gamma$ . The least power is  $\gamma$  which means that the respective second and third moments of the Wiener  $G$ -functionals are also power series with respect to  $\gamma$  with  $\gamma^n$  being the least power, where  $n$  is the highest order of the functionals involved in the particular moment. All this means that the Volterra-Wiener functional

expansion with random point basis function of perfect disorder type is virial. The viriality can be still in place for certain weaker conditions than the perfect disorder [10], but it goes beyond the scope of present work to discuss the respective technique.

The virial property is of crucial importance when particulate media are investigated because it allows one to obtain asymptotically correct results with respect to any priory prescribed degree of concentration  $c = \gamma V$  ( $V$  being the mean volume of an inclusion).

As pointed out in [23] the functional series are in a sense Fourier expansions provided that the integration is replaced by ensemble averaging. This determines the way of identification of kernels (Fourier coefficients for the case under consideration). These coefficients can be recovered after multiplying the respective expressions by specially devised functionals that form an orthogonal set which is bi-orthogonal to the set of  $G$ -functionals. In Poisson case (PDMP basis function) the said functionals are simply the Charlier polynomials. In cases when the non-overlapping is required (PDSRR basis function) we corroborate further the idea of [7] and introduce the polynomials

$$\begin{aligned}
 (2.21) \quad \tilde{C}_\omega^{(1)} &= C_\omega^{(1)}(\mathbf{0}; a_0), \\
 \tilde{C}_\omega^{(2)} &= C_\omega^{(2)}(\mathbf{0}, \mathbf{z}_1; a_0, a_1) Q(\mathbf{z}_1; a_0 + a_1), \\
 \tilde{C}_\omega^{(3)} &= C_\omega^{(3)}(\mathbf{0}, \mathbf{z}_1, \mathbf{z}_2; a_0, a_1, a_2) Q_{01} Q_{12} Q_{20}, \\
 &\dots \dots \dots \\
 \tilde{C}_\omega^{(n)} &= C_\omega^{(n)}(\mathbf{0}, \mathbf{z}_1, \dots, \mathbf{z}_{n-1}; a_0, \dots, a_{n-1}) \prod_{i \neq j=0}^{n-1} Q_{ij}.
 \end{aligned}$$

Then we obtain for the kernels:

$$\begin{aligned}
 (2.22) \quad \langle F(\mathbf{x}) \tilde{C}_\omega^{(1)} \rangle &= \langle G_F^{(1)}[\omega] \tilde{C}_\omega^{(1)} \rangle = \gamma P(a_0) K_F^{(1)}(\mathbf{x}; a_0) \\
 &- \gamma^2 P(a_0) \int_{\mathbf{R}} \int_{\mathbf{U}} K_F^{(1)}(\mathbf{x} - \xi; a) [1 - Q(\xi; a + a_0)] P(a) d^3 \xi da,
 \end{aligned}$$

$$\begin{aligned}
 (2.23) \quad \langle F(\mathbf{x}) \tilde{C}_\omega^{(2)} \rangle &= \langle G_F^{(2)}[\omega] \tilde{C}_\omega^{(2)} \rangle = 2\gamma^2 P(a_0) P(a_1) K_F^{(2)} \\
 &- 2\gamma^3 P(a_0) P(a_1) \int_{\mathbf{R}} \int_{\mathbf{U}} \{ K_F^{(2)}(\mathbf{x}, \mathbf{x} - \xi; a_0, b) [1 - Q(\xi; b + a_1)] \\
 &+ K_F^{(2)}(\mathbf{x} - \mathbf{z}_1, \mathbf{x} - \xi; a_1, b) [1 - Q(\xi; b + a_0)] \} P(b) d^3 \xi db \\
 &+ 2\gamma^4 P(a_0) P(a_1) \int_{\mathbf{R}} \int_{\mathbf{U}} \int_{\mathbf{U}} K_F^{(2)}(\mathbf{x} - \xi_1; \mathbf{x} - \xi_2; a, b) P(a) P(b) \\
 &\times [Q(\xi_1; a + a_0) - 1] [Q(\xi_2; b + a_1) - 1] d^3 \xi_1 d^3 \xi_2 da db,
 \end{aligned}$$

$$(2.24) \quad \langle F(\mathbf{x}) \bar{C}_\omega^{(n)} \rangle = \langle G_F^{(n)}[\omega] \bar{C}_\omega^{(n)} \rangle \\ = n! P(a_0) \dots P(a_{n-1}) \gamma^n K^{(n)}(\mathbf{x}, \mathbf{x}-z_1, \dots, \mathbf{x}-z_{n-1}; a_0 \dots, a_{n-1}) + o(\gamma^{(n+1)}).$$

Considering the left-hand sides of the above equalities as known functions one obtains a complicated set of integral equations for kernels which offers a little hope for analytic solution. Fortunately, the virial property of Volterra-Wiener expansions provides the means to asymptotically decouple the equations and to obtain explicit integral equations for the kernels.

### 3. APPLICATION TO THERMAL CONDUCTIVITY OF SUSPENSIONS

For mere simplicity we shall consider here only the problem of heat conduction through a random particulate medium. This problem is a simple representative of the wide class of similar transport phenomena conveniently tabulated by Batchelor [2, table 1]. The application to other transport phenomena is straightforward [6, 8].

#### 3.1. Posing the problem

Consider the two-phase medium that consists of an array of perfectly disordered spheres of random radii with thermal conductivity  $\kappa_f$  distributed in an unbounded matrix of conductivity  $\kappa_m$ . The random conductivity coefficient for such a medium has the form (1.3). It is convenient to rewrite the coefficient as follows:

$$(3.1) \quad \kappa(\mathbf{x}) = \langle \kappa \rangle + [\kappa] \int_{\mathbf{R}^3} \int_{\mathbf{U}} h(\mathbf{x}-\xi; a) C_\omega^{(1)}(\xi; a) d^3\xi da \equiv \langle \kappa \rangle + [\kappa] G_h^{(1)}[\omega],$$

where

$$\langle \kappa \rangle = \kappa_m + c[\kappa] = c\kappa_f + (1-c)\kappa_m$$

is the average conductivity of the medium.

Let us assume that the average temperature gradient is constant

$$(3.2) \quad \langle \nabla T \rangle = \mathbf{G} = \text{const},$$

where  $T=T(\mathbf{x})$  is the random temperature field.

The very form of the conductivity coefficient (3.1) hints the idea to seek the temperature field as Volterra-Wiener expansion with  $\omega(\mathbf{x}; a)$  as a basis function

$$(3.3) \quad T(\mathbf{x}) = T_0(\mathbf{x}) + \int_{\mathbf{R}^3} \int_{\mathbf{U}} T_1(\mathbf{x}-\xi; a) C_\omega^{(1)}(\xi; a) d^3\xi da + \dots \\ + \int_{\mathbf{R}^3} \dots \int_{\mathbf{U}} \dots \int_{\mathbf{U}} T_n(\mathbf{x}-\xi_1, \dots, \mathbf{x}-\xi_n; a_1, \dots, a_n) C_\omega^{(n)} d^3\xi_1 \dots d^3\xi_n da_1 \dots da_n \\ + \dots = \sum_{n=0}^{\infty} G_T^{(n)}[\omega].$$

One should be reminded that the kernels  $T_n$  for  $n \geq 2$  are supposed to satisfy the condition (2.16) and symmetry conditions that stem from the symmetry of Charlier polynomials.

The heat flux is given by

$$\begin{aligned}
 (3.4) \quad \kappa(\mathbf{x}) \nabla T(\mathbf{x}) &= \langle \kappa \rangle \sum_{n=0}^{\infty} G_{\nabla T}^{(n)}[\omega] + [\kappa] \sum_{n=0}^{\infty} G_h^{(1)}[\omega] G_{\nabla T}^{(n)}[\omega] \\
 &= \langle \kappa \rangle \{ \nabla T_0 + \int_{\mathbb{R}^3} \int_{\mathbf{U}} \nabla T_1(\mathbf{x}-\xi; a) C_{\omega}^{(1)}(d^3\xi da + \dots) \} \\
 &\quad + [\kappa] \{ \nabla T_0 \int_{\mathbb{R}^3} \int_{\mathbf{U}} h(\mathbf{x}-\xi; a) C_{\omega}^{(1)}(\xi; a) d^3\xi da \\
 &\quad + \int_{\mathbb{R}^3} \int_{\mathbf{U}} \int_{\mathbf{U}} h(\mathbf{x}-\xi; a) \nabla T_1(\mathbf{x}-\xi_1; a_1) C_{\omega}^{(1)}(\xi; a) C_{\omega}^{(1)}(\xi_1; a_1) d^3\xi d^3\xi_1 da da_1 + \dots \},
 \end{aligned}$$

and its average value is simply the following

$$(3.5) \quad \langle \kappa \nabla T \rangle = \langle \kappa \rangle \nabla T_0 + [\kappa] \langle G_h^{(1)}[\omega] G_{\nabla T}^{(1)}[\omega] \rangle = \kappa^* G,$$

since the reminder vanishes due to orthogonality of the functionals. Here  $\kappa^*$  stands for the overall heat conductivity of the medium. Making use of the appropriate properties of Charlier polynomials we have

$$\begin{aligned}
 (3.6) \quad \kappa^* G &= \langle \kappa \rangle G + [\kappa] \int_{\mathbb{R}^3} \int_{\mathbf{U}} \int_{\mathbf{U}} h(\mathbf{x}; a) \nabla T_1(\mathbf{y}; b) \{ \gamma P(a) \delta(\mathbf{x}-\mathbf{y}) \delta(a-b) \\
 &\quad - \gamma^2 P(a) P(b) [1 - Q(\mathbf{x}, \mathbf{y}; a, b)] \} d^3\mathbf{x} d^3\mathbf{y} da db,
 \end{aligned}$$

i. e. the overall conductivity  $\kappa^*$  is determined by  $\nabla T_1$  only. The latter, however, depends on the whole set of kernels, since the temperature satisfies the equation of heat conduction

$$(3.7) \quad \nabla \cdot [\kappa(\mathbf{x}) \nabla T(\mathbf{x})] = 0$$

with the boundary condition (3.2) and requirement of ergodicity.

### 3.2. Hierarchy of governing equations for kernels

Taking the mere average of (3.7) we obtain

$$\begin{aligned}
 (3.8) \quad \nabla \cdot \{ \langle \kappa \rangle G + [\kappa] \int_{\mathbb{R}^3} \int_{\mathbf{U}} \int_{\mathbf{U}} h(\mathbf{x}; a) \nabla T_1(\mathbf{y}; b) \{ \gamma P(a) \delta(\mathbf{x}-\mathbf{y}) \delta(a-b) \\
 + \gamma^2 P(a) P(b) [Q(\mathbf{x}, \mathbf{y}; a, b) - 1] \} d^3\mathbf{x} d^3\mathbf{y} da db \} = 0,
 \end{aligned}$$

which is trivially satisfied since all of the terms enclosed into brackets are constants.

For the rest of kernels we apply the scheme for identification outlined in (2.21) and obtain for  $T_1$ :

$$\begin{aligned}
 (3.9) \quad & \nabla \cdot \{ \langle \mathbf{x} \rangle [\gamma \nabla T_1(\mathbf{x}; a) + \gamma^2 \iint \nabla T_1(\mathbf{x} - \mathbf{y}; b) (Q_{0y} - 1) P(b) d^3 y db \\
 & + [\mathbf{x}] \mathbf{G} [\gamma h(\mathbf{x}; a) + \gamma^2 \iint h(\mathbf{x} - \mathbf{y}; b) (Q_{0y} - 1) P(b) d^3 y db \\
 & + [\mathbf{x}] [\gamma h(\mathbf{x}; a) \nabla T_1(\mathbf{x}; a) + \gamma^2 \iint h(\mathbf{x} - \mathbf{y}; b) \nabla T_1(\mathbf{x} - \mathbf{y}; b) \\
 & \quad \times (Q_{0y} - 1) P(b) d^3 y db \\
 & + \gamma^2 [\mathbf{x}] \iint [\nabla T_1(\mathbf{x}; a) h(\mathbf{x} - \mathbf{y}; b) + \nabla T_1(\mathbf{x} - \mathbf{y}; b) h(\mathbf{x}; a)] \\
 & \quad \times (Q_{0y} - 1) P(b) d^3 y db \\
 & + \gamma^3 [\mathbf{x}] \iiint h(\mathbf{x} - \mathbf{y}_1; b_1) \nabla T_1(\mathbf{x} - \mathbf{y}_2; b_2) [Q_{y_1 y_2} Q_{y_2 0} Q_{0 y_1} \\
 & \quad - Q_{y_1 y_2} - Q_{y_2 0} - Q_{0 y_1} + 2] P(b_1) P(b_2) d^3 y_1 d^3 y_2 db_1 db_2 \\
 & \quad + 2\gamma^2 [\mathbf{x}] \iint h(\mathbf{x} - \mathbf{y}; b) \nabla T_2(\mathbf{x} - \mathbf{y}, \mathbf{x}; b, a) P(b) d^3 y db \\
 & + 2\gamma^3 [\mathbf{x}] \iiint h(\mathbf{x} - \mathbf{y}_1; b_1) [\nabla T_2(\mathbf{x} - \mathbf{y}_2; \mathbf{x}; b_2; a) (Q_{y_1 y_2} - 1) \\
 & \quad + \nabla T_2(\mathbf{x} - \mathbf{y}_1, \mathbf{x} - \mathbf{y}_2; b_1, b_2) (Q_{y_2 0} - 1)] P(b_1) P(b_2) d^3 y_1 d^3 y_2 db_1 db_2 \\
 & + 2\gamma^4 [\mathbf{x}] \int \dots \int h(\mathbf{x} - \mathbf{y}_1; b_1) \nabla T_2(\mathbf{x} - \mathbf{y}_2, \mathbf{x} - \mathbf{y}_3; b_2, b_3) (Q_{y_2 0} - 1) \\
 & \quad \times (Q_{y_1 y_3} - 1) P(b_1) P(b_2) P(b_3) d^3 y_1 d^3 y_2 d^3 y_3 db_1 db_2 db_3 \} = 0 .
 \end{aligned}$$

Respectively, for  $T_2$  we have

$$\begin{aligned}
 (3.10) \quad & Q_{0z} \nabla \cdot (2\gamma^2 \langle \mathbf{x} \rangle \nabla T_2(\mathbf{x}, \mathbf{x} - \mathbf{z}; a, b) + 2\gamma^3 \langle \mathbf{x} \rangle \iint P(c) d^3 \xi dc \\
 & \times [\nabla T_2(\mathbf{x} - \xi, \mathbf{x} - \mathbf{z}; c, b) (Q_{0\xi} - 1) + \nabla T_2(\mathbf{x}, \mathbf{x} - \xi; a, c) (Q_{\xi z} - 1)] \\
 & + 2\gamma^4 \langle \mathbf{x} \rangle \iiint \nabla T_2(\mathbf{x} - \xi_1, \mathbf{x} - \xi_2; c_1, c_2) (Q_{0\xi_1} - 1) (Q_{\xi_1 \xi_2} - 1) \\
 & \quad \times P(c_1) P(c_2) d^3 \xi_1 d^3 \xi_2 dc_1 dc_2 \\
 & + \gamma^2 [\mathbf{x}] [h(\mathbf{x}; a) \nabla T_1(\mathbf{x} - \mathbf{z}; b) + h(\mathbf{x} - \mathbf{z}; b) \nabla T_1(\mathbf{x}; a)] \\
 & + \gamma^3 [\mathbf{x}] \iint \{ [\nabla T_1(\mathbf{x} - \mathbf{z}; b) h(\mathbf{x} - \xi; c) + \nabla T_1(\mathbf{x} - \xi; c) h(\mathbf{x} - \mathbf{z}; b) (Q_{0\xi} - 1) \\
 & + [\nabla T_1(\mathbf{x}; a) h(\mathbf{x} - \xi; c) + \nabla T_1(\mathbf{x} - \xi; c) h(\mathbf{x}; a) (Q_{\xi z} - 1) P(c) d^3 \xi dc \\
 & + \gamma^4 [\mathbf{x}] \iiint h(\mathbf{x} - \xi_1; c_1) \nabla T_1(\mathbf{x} - \xi_2; c_2) [(Q_{0\xi_1} - 1) (Q_{\xi_2 z} - 1) \\
 & \quad + (Q_{0\xi_2} - 1) (Q_{\xi_1 z} - 1)] P(c_1) P(c_2) d^3 \xi_1 d^3 \xi_2 dc_1 dc_2 \\
 & + 2\gamma^2 [\mathbf{x}] [h(\mathbf{x}; a) + h(\mathbf{x} - \mathbf{z}; b)] \nabla T_2(\mathbf{x}, \mathbf{x} - \mathbf{z}; a, b) \\
 & + \gamma^3 [\mathbf{x}] \iint h(\mathbf{x} - \xi; c) [\nabla T_2(\mathbf{x} - \xi, \mathbf{x} - \mathbf{z}; c, b) + \nabla T_2(\mathbf{x} - \xi, \mathbf{x}; c, a)]
 \end{aligned}$$

$$\begin{aligned}
& \times [(Q_{0\xi} - 1) + (Q_{\xi z} - 1)] P(c) d^3 \xi dc \\
& + 2\gamma^3 [\kappa] [h(\mathbf{x}; a) + h(\mathbf{x} - \mathbf{z}; b)] \iint [\nabla T_2(\mathbf{x} - \xi, \mathbf{x} - \mathbf{z}; c, b) (Q_{0\xi} - 1) \\
& \quad + \nabla T_2(\mathbf{x} - \xi, \mathbf{x}; c, a) (Q_{\xi z} - 1)] P(c) d^3 \xi dc \\
& + 2\gamma^3 [\kappa] \nabla T_2(\mathbf{x}, \mathbf{x} - \mathbf{z}; a, b) \iint h(\mathbf{x} - \xi; c) (Q_{0\xi} + Q_{\xi z} - 2) P(c) d^3 \xi dc \\
& \quad + 2\gamma^4 [\kappa] \iint h(\mathbf{x} - \xi; c) (Q_{0\xi} + Q_{\xi z} - 2) P(c) d^3 \xi dc \iint P(c) d^3 \xi dc \\
& \times [\nabla T_2(\mathbf{x} - \xi, \mathbf{x} - \mathbf{z}; c, b) (Q_{0\xi} - 1) + \nabla T_2(\mathbf{x} - \xi, \mathbf{x}; c, a) (Q_{\xi z} - 1)] \\
& \quad + 2\gamma^4 [\kappa] [h(\mathbf{x}; a) + h(\mathbf{x} - \mathbf{z}; b)] \iiint \nabla T_2(\mathbf{x} - \xi_1, \mathbf{x} - \xi_2; c_1, c_2), \\
& \quad \times (Q_{0\xi_1} - 1) (Q_{\xi_2 z} - 1) P(c_1) P(c_2) d^3 \xi_1 d^3 \xi_2 dc_1 dc_2 \\
& + 2\gamma^4 [\kappa] \iiint h(\mathbf{x} - \xi_1; c_1) [\nabla T_2(\mathbf{x} - \xi_2; \mathbf{x} - \mathbf{z}; c_2, b) + \nabla T_2(\mathbf{x} - \xi_2, \mathbf{x}; c_2, a)] \\
& \quad \times (Q_{\xi_1} - 1) (Q_{\xi_2 0} - 1) (Q_{\xi_1} - 1) P(c_1) P(c_2) d^3 \xi_1 d^3 \xi_2 dc_1 dc_2 \\
& + 2\gamma^4 [\kappa] \iiint h(\mathbf{x} - \xi_1; c_1) \nabla T_2(\mathbf{x} - \xi_1; \mathbf{x} - \xi_2; c_1, c_2) [(Q_{0\xi_1} - 1) (Q_{\xi_2 z} - 1) \\
& \quad + (Q_{\xi_2 0} - 1) (Q_{\xi_1 z} - 1)] P(c_1) P(c_2) d^3 \xi_1 d^3 \xi_2 dc_1 dc_2 \\
& + 2\gamma^4 [\kappa] \iiint h(\mathbf{x} - \xi_1; c_1) (Q_{\xi_1 \xi_2} - 1) [(Q_{0\xi_1} + Q_{0\xi_2} - 2) \nabla T_2(\mathbf{x} - \xi_2, \mathbf{x} - \mathbf{z}; c_2, b) \\
& \quad + (Q_{\xi_1 z} + Q_{\xi_2 z} - 2) \nabla T_2(\mathbf{x} - \xi_2, \mathbf{x}; c_2, a)] P(c_1) P(c_2) \\
& \quad \times d^3 \xi_1 d^3 \xi_2 dc_1 dc_2 \\
& \quad + 2\gamma^5 [\kappa] \iiint h(\mathbf{x} - \xi; c_1) \nabla T_2(\mathbf{x} - \xi_2, \mathbf{x} - \xi_3; c_2, c_3) \\
& \times [(Q_{0\xi_1} - 1) (Q_{0\xi_2} - 1) (Q_{\xi_2 z} - 1) + (Q_{0\xi_1} - 1) (Q_{\xi_1 \xi_2} - 1) (Q_{\xi_3 z} - 1) \\
& \quad + (Q_{\xi_1 \xi_2} - 1) (Q_{0\xi_2} - 1) (Q_{\xi_3 z} - 1) + (Q_{\xi_1 z} - 1) (Q_{0\xi_2} - 1) (Q_{\xi_1 \xi_3} - 1) \\
& \quad + (Q_{\xi_1 \xi_2} - 1) (Q_{\xi_2 z} - 1) (Q_{0\xi_3} - 1) \\
& \quad + (Q_{\xi_1 \xi_3} - 1) (Q_{\xi_2 z} - 1) (Q_{0\xi_1} - 1) (Q_{0\xi_3} - 1) \\
& \quad + (Q_{\xi_1 \xi_3} - 1) (Q_{\xi_2 z} - 1) (Q_{z\xi_1} - 1) (Q_{z\xi_3} - 1)] \\
& \quad \times P(c_1) P(c_2) P(c_3) d^3 \xi_1 d^3 \xi_2 d^3 \xi_3 dc_1 dc_2 dc_3 \\
& + 6\gamma^3 [\kappa] \iint h(\mathbf{x} - \xi; c) \nabla T_3(\mathbf{x} - \xi, \mathbf{x} - \mathbf{z}, \mathbf{x}; c, b, a) P(c) d^3 \xi dc \\
& + 6\gamma^4 [\kappa] \iiint h(\mathbf{x} - \xi_1; c_1) [\nabla T_3(\mathbf{x} - \xi_1, \mathbf{x} - \xi_2, \mathbf{x} - \mathbf{z}; c_1, c_2, b) (Q_{0\xi_2} - 1) \\
& \quad + \nabla T_3(\mathbf{x} - \xi_2, \mathbf{x}, \mathbf{x} - \mathbf{z}; c_2, a, b) (Q_{z\xi_3} - 1) + (Q_{\xi_1 \xi_2} - 1) \\
& \quad \times \nabla T_3(\mathbf{x} - \xi_2, \mathbf{x}, \mathbf{x} - \mathbf{z}; c_2, a, b)] P(c_1) P(c_2) d^3 \xi_1 d^3 \xi_2 dc_1 dc_2 \\
& + 6\gamma^5 [\kappa] \int \dots \int h(\mathbf{x} - \xi_1; c_1) [\nabla T_3(\mathbf{x} - \xi_1, \mathbf{x} - \xi_2, \mathbf{x} - \xi_3; c_1, c_2, c_3)
\end{aligned}$$

$$\begin{aligned}
& \times (Q_{0\xi_2} - 1) (Q_{z\xi_3} - 1) + \nabla T_3 (\mathbf{x} - \xi_2, \mathbf{x} - \xi_3, \mathbf{x} - \mathbf{z}; c_2, c_3, b) \\
& \times (Q_{0\xi_3} - 1) (Q_{\xi_1\xi_2} - 1) + \nabla T_3 (\mathbf{x} - \xi_2, \mathbf{x} - \xi_3, \mathbf{x}; c_2, c_3, a) \\
& \times (Q_{z\xi_3} - 1) (Q_{\xi_1\xi_2} - 1) P(c_1) P(c_2) P(c_3) d^3\xi_1 d^3\xi_2 d^3\xi_3 dc_1 dc_2 dc_3 \\
& + 6\gamma^6 [\chi] \int \dots \int h(\mathbf{x} - \xi_1; c_1) \nabla T_3 (\mathbf{x} - \xi_2, \mathbf{x} - \xi_3, \mathbf{x} - \xi_4; c_2, c_3, c_4) \\
& \times (Q_{0\xi_2} - 1) (Q_{z\xi_3} - 1) (Q_{\xi_1\xi_4} - 1) P(c_1) P(c_2) P(c_3) P(c_4) \\
& \times d^3\xi_1 d^3\xi_2 d^3\xi_3 d^3\xi_4 dc_1 dc_2 dc_3 dc_4 = 0.
\end{aligned}$$

Proceeding further one can obtain equations for the higher-order kernels. The equation for the  $n$ -th kernel  $T_n$  contains also the kernels  $T_{n-1}$  and  $T_{n+1}$ , i. e. the governing system for the kernels is an infinite hierarchy of conjugated equations.

### 3.3. Asymptotic uncoupling of the hierarchy

As should have been expected, the method of functional expansions — as all other methods — leads to an infinite hierarchy of equations. Which makes the difference, however, is that the expansion under consideration is virial and the higher-order functionals contribute to the averaged characteristics quantities 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 into power series with respect to the concentration  $c$ , namely,

$$(3.11) \quad T_1 = T_{10} + c T_{11} + c^2 T_{12} + \dots, \quad T_2 = T_{20} + c T_{21} + c^2 T_{22} + \dots$$

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 to the following systems of equations (be noted there is already taken into account that  $\langle \chi \rangle = \chi_m + c[\chi]$ ):

$$(3.12a) \quad \nabla \cdot \{ \chi_m \nabla T_{10}(\mathbf{x}; a) + [\chi] h(\mathbf{x}; a) [G + \nabla T_{10}(\mathbf{x}; a)] \} = 0,$$

$$(3.12b) \quad \nabla \cdot (2 \{ \chi_m + [\chi] [h(\mathbf{x}; a) + h(\mathbf{x} - \mathbf{z}; b)] \} \nabla T_{20}(\mathbf{x}, \mathbf{x} - \mathbf{z}; a, b) \\ + [\chi] [h(\mathbf{x}; a) \nabla T_{10}(\mathbf{x} - \mathbf{z}; b) + h(\mathbf{x} - \mathbf{z}; b) \nabla T_{10}(\mathbf{x}; a)]) = 0.$$

It is interesting to discuss here the interpretation of the equations of the above set. They are equations of discontinuous coefficients and their solutions have derivatives that are generalized functions. The first equation (3.12a) possesses a solution which is nothing else, but the perturbation to a linear temperature field due to the presence of a single sphere of radius  $a$ . The physical meaning of the solution to the second equation becomes more clear if the following notation is introduced

$$(3.13) \quad T^{(2)}(\mathbf{x}, \mathbf{x} - \mathbf{z}; a, b) = 2T_{20}(\mathbf{x}, \mathbf{x} - \mathbf{z}; a, b) + T_{10}(\mathbf{x}; a) + T_{10}(\mathbf{x} - \mathbf{z}; b),$$

and the respective equation is derived from (3.12a) and (3.12b)

$$(3.14) \quad \nabla \cdot (\{ \chi_m + [\chi] [h(\mathbf{x}; a) + h(\mathbf{x} - \mathbf{z}; b)] \} [G + \nabla T^{(2)}(\mathbf{x}, \mathbf{x} - \mathbf{z}; a, b)]) = 0.$$

The solution to the latter is just the perturbation to a linear temperature field due to the presence of two spheres of radii  $a$  and  $b$  respectively, first of the sphere centered in the origin, the second — in the point  $z$ . Now it becomes transparent that  $T_{20}$  gives the additional perturbation to the single perturbances of each sphere due to binar interaction of their fields.

The terms proportional to  $\gamma^2$  give the following equation for  $T_{11}$ :

$$(3.15) \quad \nabla \cdot (\{\kappa_m + [\kappa] h(\mathbf{x}; a)\} \nabla T_{11}(\mathbf{x}; a) + [\kappa] \nabla T_{10}(\mathbf{x}; a) - [\kappa] \frac{1}{V} \iint \left[ \nabla T_{10}(\mathbf{x}; a) h(\mathbf{x}-\mathbf{y}; b) + \nabla T_{10}(\mathbf{x}-\mathbf{y}; b) h(\mathbf{x}; a) \right] P(b) d^3y db - \frac{2}{V} \left\{ \kappa_m + [\kappa] h(\mathbf{x}; a) \right\} \iint \nabla T_{20}(\mathbf{x}-\mathbf{y}; \mathbf{x}; b, a) P(b) d^3y db) = 0,$$

where  $V$  stands for the mean volume of particles

$$(3.16) \quad V = \iint h(\mathbf{x}; a) P(a) d^3x da, \quad c = \gamma V.$$

When deriving (3.15) eqs. (3.12a) and (3.12b) are acknowledged.

Now the advantages of the virial expansion become transparent: eq. (3.12a) contains only one unknown function  $T_{10}$ ; after it is solved eq. (3.12b) becomes closed for  $T_{20}$ ; after the two quantities  $T_{10}$  and  $T_{20}$  are known the eq. (3.15) becomes closed for  $T_{11}$  etc. This property of the virial expansion is present for all other higher-order kernels and equations.

### 3.4. First-order solution

Under the provisions of ergodicity we have

$$(3.17) \quad T_{10} \rightarrow 0, \quad |\mathbf{x}| \rightarrow \infty,$$

and the solution to the discontinuous-coefficient equation (3.12a) is

$$(3.18) \quad T_{10}(\mathbf{x}; a) = \beta G \cdot \left\{ -\mathbf{x} h(\mathbf{x}; a) + a^3 \nabla \frac{1}{|\mathbf{x}|} [1 - h(\mathbf{x}; a)] \right\}, \quad \beta = \frac{[\kappa]}{\kappa_f + 2\kappa_m}.$$

Making use of the properties of the characteristic function one has

$$(3.19) \quad h(\mathbf{x}; a) \nabla T_{10}(\mathbf{x}; a) = -\beta G h(\mathbf{x}; a),$$

and hence

$$(3.20) \quad \iint h(\mathbf{x}; a) \nabla T_{10}(\mathbf{x}; a) P(a) d^3x da = -\beta G V.$$

Then the overall heat conduction modulus is calculated from the formula:

$$(3.21) \quad \kappa^* G \equiv \langle \kappa \nabla T \rangle = \langle \kappa \rangle G + c[\kappa] \frac{1}{V} \iint h(\mathbf{x}; a) \nabla T_{10} P(a) d^3x da \\ = \langle \kappa \rangle G - c[\kappa] \beta = [\kappa_m + c[\kappa] (1 - \beta)] G$$



$$= \left\{ \kappa_m + \frac{3 \kappa_m [\chi]}{\kappa_f + 2\kappa_m} c \right\} G = \kappa_m (1 + 3\beta c) G ,$$

i.e.

$$(3.22) \quad \kappa^* = \kappa_m (1 + 3\beta c) + o(c) ,$$

which is the linear part of the famous Maxwell [24] formula for the effective electric permeativity of two-phase materials. It is well seen that the probability density of size distribution does not show up in the final result which means that the linear approximation to the effective conductivity is not affected by the size distribution.

#### 4. CONCLUSION

The generalization of the method of stochastic functional expansions to the case of marked random point basis functions appears to be a crucial point when modelling the real composite materials whose inclusions can have random shapes. The simplest situation in which the random centres are attributed with random parameters is the polydisperse suspension of spheres. The method of functional expansion with marked random point basis function (namely: PDSRR field or Perfect Disorder of Spheres of Random Radii) provides the rigorous basis for treating the polydisperse systems, since the random point functions of Perfect-Disorder type are natural approximations to variety of physical fields. The technique developed enables one to derive closed equations for kernels that are asymptotically correct. The first-order solution shows that the overall conductivity does not depend on the probability distribution of spheres radii.

#### REFERENCES

1. Beran M. *Statistical Continuum Theories*. New York, John Wiley, 1968.
2. Batchelor G. K. Transport properties of two-phase materials with random structure. — *Ann. Rev. Fluid Mech.*, **6**, 1974, 227—255.
3. Christensen R. M. *Mechanics of Composite Materials*. New York, John Wiley, 1979.
4. MacCoy J. J. Macroscopic response of continua with random microstructure. — In: *Progress in Solid Mechanics* (Nemat-Nasse S. ed.) Vol. 6, Part 1. Oxford — New York, Pergamon Press, 1981.
5. Christov C. I. Poisson-Wiener expansion in non-linear stochastic systems. — *Ann. de l'Univ. de Sofia, Fac. Math. Mec.*, **75**, Livre 2 — Mecanique, 1981, 143—165.
6. Christov C. I. Toward a statistical theory of emulsions. — In: *Proc. 13th Spring Conf. Union Bulg. Math.*, Sunny Beach, 1984, 473—481.
7. Christov C. I., Markov K. Z. Stochastic functional expansion for random media with perfectly disordered constitution. — *SIAM J. Appl. Math.*, **45**, 1985, 289—311.
8. Christov C. I., Markov S. Z. Stochastic functional expansion in elasticity of heterogeneous solids. — *Int. J. Solids Struct.*, **21**, 1985, 1197—1211.
9. Markov K.Z. An application of Volterra-Wiener series in mechanics of composite materials. — *BAS TPM*, **15**, 1984, No 1, 41—59.
10. Markov K. Z. Application of Volterra-Wiener series for bounding the overall conductivity of heterogeneous media. I General procedure. II Suspensions of equi-sized spheres. — *SIAM J. Appl. Math.*, **47**, 1987.
11. Christov C. I. A further development of the concept of random density function with application to Volterra-Wiener expansions. — *C. R. Acad. Bulg.*, **38**, 1985, 35—38.
12. Christov C. I. Outline of the method of stochastic functional expansions with marked random point basis function (in preparation).
13. Кузнецов П. П., Стратонович Р. Л. К математической теории коррелированных случайных точек. — *Изв. АН СССР, сер. Матем.*, **20**, 1956, 167—178.

14. Большаков И. А., Ракошиц В. С. Прикладная теория случайных потоков. М., Советское радио, 1978.
15. Moyal J. E. The general theory of stochastic population processes. — *Acta Math.*, **108**, 1962, 1—31.
16. Ramakrishnan A. Stochastic processes relating to particles distribution in a continuous infinity of states. — *Proc. Cambr. Phil. Soc.*, **46**, 1951, 595—602.
17. Vere-Jones D. Some applications of probability generating functionals to the study of input-output streams. — *J. Roy. Stat. Soc.*, **B30**, 1968, 321—333.
18. Kröner E. Elastic moduli of perfectly disordered materials. — *J. Mech. Phys. Solids.*, **15**, 1967, 319—329.
19. Snyder D. Random point processes. New York, John Wiley, 1975.
20. Batchelor G. K., Green J. T. The hydrodynamic interaction of two small freely-moving spheres in a linear flow field. — *J. Fluid Mech.*, **56**, 1972, 375—400.
21. Ogura H. Orthogonal functionals of the Poisson process. — *IEEE Trans. Inf. Theory*, **18**, 1972, 473—481.
22. Schetzen M. The Volterra and Wiener theories of nonlinear systems. New York, John Wiley, 1980.
23. Cameron R. H., Martin W. T. The orthogonal development of non-linear functional into series of Fourier-Hermite functionals. — *Ann. Math.*, **48**, 1947, 385—392.
24. Maxwell J. Elasticity and Magnetism. New York, Dover, 1954.

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