

## ON A TWO-POINT CORRELATION FUNCTION IN RANDOM DISPERSIONS AND AN APPLICATION

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For a random arrays of identical spheres the “particle-center” correlation  $F^{\text{PC}}(r)$  is considered. A simple integral representation of  $F^{\text{PC}}(r)$  through the radial distribution function is first proposed. As an application, the classical Smoluchowski problem, concerning steady-state diffusion of a species among an array of nonoverlapping and ideally absorbing sinks, is revisited. Using the variational principle of Rubinstein and Torquato, a lower bound on the effective sink strength  $k^{*2}$ , in which  $F^{\text{PC}}(r)$  shows up, is obtained in an elementary manner. It turns out that the bound *coincides* with that derived by Doi and Talbot & Willis, but here neither more complicated “surface” correlations, nor Hashin-Shtrikman’s type principles are invoked. The degeneration of the bound, produced by the 2-D case counterpart of the proposed variational procedure, is demonstrated and discussed.

**Keywords.** random dispersions, correlation functions, effective properties, variational bounds, absorption problem

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### 1 Definitions of the Basic Statistical Characteristics

Consider a dispersion of equal and nonoverlapping spheres of radius  $a$  in  $\mathbb{R}^3$ , whose centers form the random set of points  $\{x_\alpha\}$ . The assumption of statistical isotropy and homogeneity is adopted henceforth. Introduce after Stratonovich [1] the so-called random density field

$$\psi(x) = \sum_{\alpha} \delta(x - x_{\alpha}), \quad (1.1)$$

$\delta(x)$  is the Dirac delta-function. Then, in particular,

$$\langle \psi(x) \rangle = n, \quad F^{\text{cc}}(x) = \langle \psi(x)\psi(0) \rangle = n\delta(x) + n^2g(x), \quad (1.2)$$

where  $n$  is the number density of the spheres, and  $g(x) = g(r)$ ,  $r = |x|$ , is their radial distribution function, see [1]. The brackets  $\langle \cdot \rangle$  signify ensemble averaging. The notation  $F^{\text{cc}}(x)$  in Eq. (1.2) is justified by the interpretation of the quantity  $\langle \psi(x)\psi(0) \rangle$ —this is the “center-center” correlation, in the sense that it obviously gives the probability densities of finding centers of particles both at the origin and at the point  $x$ .

Let

$$I_1(x) = \begin{cases} 1, & \text{if } x \in \mathcal{K}_1, \\ 0, & \text{otherwise,} \end{cases} \quad (1.3)$$

be the characteristic function of the region  $\mathcal{K}_1$ , occupied by the spheres. Then

$$I_1(x) = (h_a * \psi)(x) = \int h_a(x-y)\psi(y) \, dy, \quad I_1'(x) = \int h_a(x-y)\psi'(y) \, dy, \quad (1.4)$$

where  $\psi'(y) = \psi(y) - n$  is the fluctuating part of the field  $\psi(y)$  and  $h_a(y)$  is the characteristic function of a single sphere of radius  $a$ , located at the origin. All integrals hereafter are over the whole  $\mathbb{R}^3$ ; as usual,  $f * g$  denotes convolution.

In turn, the two-point correlation most often used, is

$$F^{\text{pp}}(x) = \langle I_1(0)I_1(x) \rangle, \quad (1.5)$$

whose interpretation is obvious—this is the probability density that two points, separated by the vector  $x$ , when thrown into the medium both fall within the region  $\mathcal{K}_1$ , occupied by the spheres. That is why  $\langle I_1(0)I_1(x) \rangle$  can be called “particle-particle” correlation which explains its notation  $F^{\text{pp}}(x)$  in Eq. (1.5).

A natural statistical quantity, playing an intermediate role between  $F^{\text{cc}}(x)$  and  $F^{\text{pp}}(x)$ , is the “particle-center” correlation, defined as

$$F^{\text{pc}}(x) = \langle I_1(x)\psi(0) \rangle. \quad (1.6)$$

It obviously gives the probability that for a pair of points, separated by the vector  $x$ , one hits a sphere’s center while the other falls into a sphere. In this lecture a more detailed study of this correlation, together with one of its possible applications, will be performed. Of course, the latter correlation is a particular case of the much more general statistical characteristics of two-phase random media, as introduced by Torquato [2], but our aim here will be more specific, namely, the derivation of a simple integral representation of  $F^{\text{pc}}(x)$

by means of the two-point probability density function for the set  $\{x_\alpha\}$  of sphere's centers, see Eq. (2.9) below.

It is natural to represent the above introduced correlations as

$$F^{\text{cc}}(x) = n^2 + \overline{F}^{\text{cc}}(x), \quad F^{\text{pc}}(x) = n\eta_1 + \overline{F}^{\text{pc}}(x), \quad F^{\text{pp}}(x) = \eta_1^2 + \overline{F}^{\text{pp}}(x), \quad (1.7)$$

where  $\eta_1 = \langle I_1(x) \rangle = nV_a$  is the volume fraction of the spheres,  $V_a = \frac{4}{3}\pi a^3$ . As it follows from Eqs. (1.2), (1.4), (1.5) and (1.6),

$$\begin{aligned} \overline{F}^{\text{cc}}(x) &= \langle \psi'(y_1)\psi'(y_2) \rangle = n\delta(x) + n^2\nu_2(x), \quad \overline{F}^{\text{pc}}(x) = \langle I'_1(x)\psi'(0) \rangle \\ &= (h_a * \overline{F}^{\text{cc}})(x) = nh_a(x) + n^2 \int h_a(x-y)\nu_2(y) dy, \end{aligned} \quad (1.8)$$

$$\overline{F}^{\text{pp}}(x) = \langle I'_1(x)I'_1(0) \rangle = (h_a * \overline{F}^{\text{pc}})(x) = (h_a * h_a * \overline{F}^{\text{cc}})(x).$$

Here  $\nu_2(x) = g(x) - 1$  is the so-called binary (or total) correlation function. Assuming there is no long-range order, all  $\nu_2(x)$ ,  $\overline{F}^{\text{cc}}(x)$ ,  $\overline{F}^{\text{pc}}(x)$  and  $\overline{F}^{\text{pp}}(x)$  vanish as  $x \rightarrow \infty$ , since the constants in the right-hand sides of Eqs. (1.7) are just their long-range values.

## 2 A Study of the “Particle-Center” Correlation

Let us split the radial distribution function,  $g(x)$ , as

$$g(x) = g^{\text{ws}}(x) + \tilde{g}(x), \quad (2.1)$$

where

$$g^{\text{ws}}(x) = 1 - h_{2a}(x) = \begin{cases} 0, & \text{if } |x| \leq 2a, \\ 1, & \text{if } |x| > 2a, \end{cases} \quad (2.2)$$

corresponds to the simplest “well-stirred” distribution of spheres;  $\tilde{g}(x)$  is then the “correction” to the latter. In turn, the binary correlation  $\nu_2(x) = g(x) - 1$  is represented as

$$\nu_2(x) = -h_{2a}(x) + \tilde{\nu}_2(x). \quad (2.3)$$

Moreover, one has  $\nu_2(x) = \tilde{\nu}_2(x) = \tilde{g}(x)$ , if  $|x| \geq 2a$ , due to Eqs. (2.1)–(2.3). The nonoverlapping assumption implies that  $g(x) = 0$ , and hence  $\tilde{\nu}_2(x) = \tilde{g}(x) = 0$  as well, if  $|x| \leq 2a$ , as a consequence of the same Eqs. (2.1)–(2.3).

From Eqs. (1.7) and (2.3) it now follows

$$\overline{F}^{\text{pc}}(x) = \overline{F}^{\text{pc}}_{\text{ws}}(x) + \tilde{F}^{\text{pc}}(x), \quad (2.4)$$

where

$$\begin{aligned}\overline{F}_{\text{ws}}^{\text{pc}}(x) &= nh_a(x) - n^2(h_a * h_{2a})(x) \\ &= n\eta_2 h_a(x) - \frac{n\eta_1}{16\rho} (3 - \rho)^2 (\rho^2 + 6\rho - 3) [h_{3a}(x) - h_a(x)],\end{aligned}\quad (2.5)$$

$$\widetilde{F}^{\text{pc}}(x) = n^2 \int h_a(x - y) \nu_2(y) dy, \quad \rho = |x|/a. \quad (2.6)$$

To represent  $\widetilde{F}^{\text{pc}}(x)$  as a simple one-tuple integral, containing the binary correlation  $\nu_2(x)$ , write down the latter as

$$\nu_2(y) = \int_{2a}^{\infty} \nu_2(\xi) \frac{\partial}{\partial \xi} h_\xi(y) d\xi, \quad (2.7)$$

which follows from the obvious formula

$$\frac{\partial h_\xi(y)}{\partial \xi} = \delta(|y| - \xi). \quad (2.8)$$

Then, by virtue of Eqs. (2.6) and (2.7),

$$\begin{aligned}\widetilde{F}^{\text{pc}}(x) &= n^2 \int_2^{\infty} d\mu \nu_2(\mu) \frac{\partial}{\partial \mu} (h_a * h_\xi)(r) \\ &= \frac{3n\eta_1}{4\rho} \int_{\max\{2, \rho-1\}}^{\rho+1} [1 - (\mu - \rho)^2] \mu \nu_2(\mu) d\mu,\end{aligned}\quad (2.9)$$

having changed the order of integration and recalling the formula for the common volume of  $(h_a * h_\xi)(r)$  of two spheres of radii  $a$  and  $\xi$ , with centers separated by  $r$ ;  $\mu = \xi/a$ .

The formula (2.9) is just the needed simple integral representation of  $\widetilde{F}^{\text{pc}}(x)$  by means of the binary correlation function for the set  $\{x_\alpha\}$  of sphere's centers. Its derivation has been inspired here by the reasoning of the recent paper [3], devoted to the "particle-particle" correlation  $F^{\text{pp}}(x)$ . A detailed study of the rest of the two-point correlation functions for the dispersion, including the interfacial ones, and of their integral representations similar to (2.9), can be found in [4].

The obtained formula (2.9) is useful, for example, when statistical quantities

$$\theta_U^{\text{pc}} = \int_0^{\infty} U(\rho) \overline{F}^{\text{pc}}(\rho) d\rho \quad (2.10)$$

are of interest, for a given function  $U(\rho)$ . Indeed,  $\theta_U^{\text{pc}} = \theta_{U,\text{ws}}^{\text{pc}} + \tilde{\theta}_U^{\text{pc}}$ , where the term  $\theta_{U,\text{ws}}^{\text{pc}}$  corresponds to the well-stirred distribution and can be immediately found, see (2.5); the ‘‘correction’’  $\tilde{F}^{\text{pc}}(\rho)$  generates  $\theta_U^{\text{pc}}$ . To evaluate the latter, it suffices to apply (2.9) and change afterwards the order of integration:

$$\begin{aligned}\tilde{\theta}_U^{\text{pc}} &= \int_0^\infty U(\rho) \tilde{F}^{\text{pc}}(\rho) \, d\rho = n\eta_1 \int_2^\infty H_U^{\text{pc}}(\mu) \mu \nu_2(\mu) \, d\mu, \\ H_U^{\text{pc}}(\mu) &= \frac{3}{4} \int_{\mu-1}^{\mu+1} \frac{U(\rho)}{\rho} [1 - (\mu - \rho)^2] \, d\rho.\end{aligned}\tag{2.11}$$

(A similar idea was used by Drugan and Willis [5] when deriving a formula of the type (2.13) below for the ‘‘particle-particle’’ correlation.)

The kernels  $H_U^{\text{pc}}(\mu)$  in (2.11) become extremely simple when  $U(\rho) = \rho^k$ ,  $k = 0, 1, \dots$ . In this case  $H_U^{\text{pc}}(\mu) = H_k^{\text{pc}}(\mu)$  are polynomials whose explicit evaluation is straightforward. In particular,  $H_1^{\text{pc}}(\mu) = 1$ , so that

$$\theta_1^{\text{pc}} = \int_0^\infty \rho \bar{F}^{\text{pc}}(\rho) \, d\rho = n \left( \frac{5 - 19\eta_1}{10} + \eta_1 m_1 \right), \quad m_1 = \int_2^\infty \mu \nu_2(\mu) \, d\mu, \tag{2.12}$$

see (2.5). It is noted that Eq. (2.12) is fully similar to the formula, recently derived by Markov and Willis [3], for the ‘‘particle-particle’’ correlation, namely,

$$\theta_1^{\text{pp}} = \int_0^\infty \rho \bar{F}^{\text{pp}}(\rho) \, d\rho = \eta_1 \left( \frac{2 - 9\eta_1}{5} + \eta_1 m_1 \right), \tag{2.13}$$

with the same  $m_1$ , as defined in Eq. (2.12).

### 3 Absorption Problem and Rubinstein-Torquato’s Principle

As a simple application of the ‘‘particle-center’’ correlation and of its representation (2.4), (2.5) and (2.9), consider a dispersion of ideal and nonoverlapping spherical sinks (the phase ‘1’), immersed into an unbounded matrix. Let a species (defects) be generated at the rate  $K$  within the matrix (phase ‘2’), occupying the region  $\mathcal{K}_2$ , and absorbed by the sinks in the region  $\mathcal{K}_1 = \mathbb{R}^3 \setminus \mathcal{K}_2$ . In the steady-state limit the concentration of the defects  $c(x)$  is governed by the well-known equations:

$$\Delta c(x) + K = 0, \quad x \in \mathcal{K}_2, \quad c(x) \Big|_{\partial\mathcal{K}_2} = 0. \tag{3.1}$$

The creation of defects is *exactly* compensated by their removal from the sinks:

$$k^{*2} \langle c(x) \rangle = K(1 - \eta_1). \tag{3.2}$$

The rate constant  $k^{*2}$  is just the effective absorption coefficient (the sink strength) of the medium. Its evaluation and bounding for special kinds of random constitution and, above all, for random dispersion of spheres, has been the subject of numerous works, starting with the classical studies of Smoluchowski (1912), see, e.g. [6, 7, 8, 9, 10], and the references therein. (Note that we have added the factor  $1 - \eta_1$  in (3.2), due to the fact that in the case under study defects are created *only* within the phase '2', see [11] for a discussion.)

We shall confine the analysis to variational bounding of the sink strength  $k^{*2}$ , taking into account the foregoing two-point statistical characteristics. Recall to this end the variational principle of Rubinstein and Torquato (R-T) [9].

Let  $\mathcal{A}$  be the class of statistically homogeneous trial fields such that

$$\mathcal{A} = \left\{ u(x) \mid \Delta u(x) + K = 0, \quad x \in \mathcal{K}_2 \right\}. \quad (3.3)$$

Then

$$k^{*2} \geq \frac{K^2(1 - \eta_1)}{\langle I_2(x) |\nabla u(x)|^2 \rangle}. \quad (3.4)$$

The equality sign in (3.4) is achieved, if  $u(x) = c(x)$  is the actual field that solves the problem (3.1).

Since  $\langle I_2(x) |\nabla u(x)|^2 \rangle \leq \langle |\nabla u(x)|^2 \rangle$ , another bound follows from (3.4):

$$k^{*2} \geq \frac{K^2(1 - \eta_1)}{\langle |\nabla u(x)|^2 \rangle}, \quad (3.5)$$

see [9]. Though weaker than (3.4), the evaluation of the bound (3.5) is simpler, because it obviously employs a smaller amount of statistical information.

Consider the trial fields

$$u(x) = -\frac{K}{\mu} \int G_0(x-y) [I_1(y) - \mu] dy + K \lambda V_a \int H_0(x-y) \psi(y) dy, \quad (3.6)$$

$$G_0(x) = \frac{1}{4\pi|x|}, \quad H_0(x) = \begin{cases} G_0(a), & \text{if } |x| < a, \\ G_0(x), & \text{if } |x| \geq a, \end{cases}$$

with adjustable constants  $\lambda, \mu$ . Since  $\Delta G_0(x) + \delta(x) = 0$ , it is easily seen that  $\Delta u(x) = K$ , if  $x \in \mathcal{K}_2$ , and therefore the fields  $u(x)$  in (3.6)<sub>1</sub> are admissible.

Consider now the quantity of central importance  $\mathcal{U} = \langle |\nabla u(x)|^2 \rangle / K^2$ . For the latter to be finite, and hence to produce a nontrivial lower bound (3.5), it is necessary that the integrand in (3.6)<sub>1</sub> have a zero mean value when  $x \rightarrow \infty$ :

$$-\frac{1}{\mu} (\eta_1 - \mu) + \lambda n V_a = 0, \quad \text{i.e.} \quad \lambda = \frac{\eta_1 - \mu}{\eta_1 \mu}. \quad (3.7)$$

Integration by parts yields

$$\begin{aligned} \mathcal{U} = & \frac{1}{\mu^2} \int G_0(x) \left[ \overline{F}^{\text{PP}}(x) + (\eta_1 - \mu)^2 \right] dx - \frac{2\lambda V_a}{\mu} \int H_0(x) \\ & \times \left[ \overline{F}^{\text{PC}}(x) + n(\eta_1 - \mu) \right] dx + \lambda^2 V_a^2 \int \chi(x) \left[ \overline{F}^{\text{CC}}(x) + n^2 \right] dx, \end{aligned} \quad (3.8)$$

see Eqs. (1.2), (1.5), (1.6), (1.7). In Eq. (3.8),  $\chi(x)$  is the convolution

$$\chi(x) = \int \nabla_y H_0(x-y) \cdot \nabla_y H_0(y) dy = \begin{cases} G_0(a)(1 - \rho/4), & |x| < 2a, \\ G_0(x), & |x| \geq 2a. \end{cases} \quad (3.9)$$

Eq. (3.9) simply follows if the Fourier transform techniques is employed.

A comment, concerning the class (3.6), is warranted before starting calculations. In the random case under study the realizations of the random medium occupy the *whole*  $\mathbb{R}^3$ , and therefore *no* boundary conditions are imposed on the Green function  $G(x)$  for the Laplace operator. This makes the choice of this function ambiguous, because  $G_0(x) = 1/(4\pi|x|)$  is *as good* for our purposes (to have in (3.6)<sub>1</sub> an admissible trial field) as  $G(x) = G_0(x) + \text{const}$ , and there were *no* reasons at that stage of the analysis to take this constant zero, as it was implicitly done in (3.6)<sub>2</sub>. The quantity  $\langle \nabla u(x)|^2 \rangle$ , and hence the bound (3.5), do not feel the presence of this constant, since it enters under the nabla operator. However, to perform the integration by parts that leads us to Eq. (3.8), it is necessary to employ the Green function that *decays* at infinity (i.e. far from the source), and this is exactly the reason which explains the choice of this function in (3.6)<sub>2</sub>, and *fixes* the value of the additive constant as zero in the proposed procedure. It is to be pointed out immediately that the situation in 2-D is drastically different from 3-D in the sense that among the multitude of possible Green functions  $-\ln|x|/(2\pi) + \text{const}$ , *no one* decays at infinity. This, in particular, makes impossible the appropriate integration by parts in the double integrals that appear in this case in the quantity  $\langle \nabla u(x)|^2 \rangle$ , cf. Eqs. (3.6) and (3.8).

Now, elementary calculations, using Eqs. (2.5), (3.9), (2.12) and (2.13), give eventually

$$\mathcal{U}/a^2 = F(\mu) + m_1, \quad F(\mu) = \frac{1 - \eta_1}{3\eta_1} - \frac{2\eta_1}{15} \frac{1}{\mu} + \frac{\eta_1}{15} \frac{1}{\mu^2}. \quad (3.10)$$

As it now follows from (3.10)<sub>1</sub>, the best bound (3.5) corresponds obviously to  $\mu = \mu_0$  that minimizes  $F(\mu)$ , i.e. to  $\mu = 1$ . Hence this bound is

$$k^{*2} a^2 \geq \frac{3\eta_1(1 - \eta_1)}{1 - 5\eta_1 - \eta_1^2/5 + 3\eta_1 m_1} = 3\eta_1 + o(\eta_1). \quad (3.11)$$

As indicated, the bound (3.11) is *exact* in the dilute limit  $\eta_1 \ll 1$ , since it reproduces in this case the well-known Smoluchowski result.

The bound (3.11) coincides with that derived by Talbot and Willis [8] by means of an ingenious variational procedure of Hashin-Shtrikman's type. In this paper [8] the appropriate Green function was defined *unambiguously*, starting with the finite body case and only then passing to the "unbounded" limit. Later on it was noticed by Talbot (unpublished manuscript) that (3.11), for a dispersion of nonoverlapping spheres, coincides with the earlier proposed bound of Doi [7] (the authors of [8] apparently were not aware of Doi's paper). The variational procedure, used by Doi, was clarified later on by Rubinstein and Torquato [9] and put within the frame of their variational principle (3.3), (3.4). The fact that the original Doi's result can be recast in the elegant Talbot and Willis' form (3.11) was noticed in passing also by Beasley and Torquato [12], who apparently were not aware of the paper [8]. Due to all these reasons it seems proper to call (3.11) Doi-Talbot-Willis (DTW) bound.

It is important to point out that the appearance of the Doi result (in the form (3.11)) out of the class (3.6) is however fully natural. The explanation follows from the fact that Doi [7] has employed the trial fields

$$u(x) = K \int G_0(x-y) (I_2(y) - \xi |\nabla I_1(y)|) dy, \quad (3.12)$$

with  $\xi$  uniquely defined from the condition that the integrand should possess zero mean value. Since  $|\nabla I_1(y)|$  is a  $\delta$ -function, concentrated on the surfaces of the spheres, one has

$$\begin{aligned} \int G_0(x-y) |\nabla I_1(y)| dy &= \int G_0(x-y) \int \frac{\partial}{\partial b} h_b(y-z) \psi(z) dz dy \Big|_{b=a} \\ &= \int \frac{\partial}{\partial b} \varphi_b(x-z) \Big|_{b=a} \psi(z) dz = 4\pi a^2 \int H_0(x-z) \psi(z) dz, \end{aligned} \quad (3.13)$$

having taken Eqs. (1.1) and (2.8) into account. In Eq. (3.13)

$$\varphi_b(x) = (G_0 * h_b)(x) = \begin{cases} (3b^2 - r^2)/6, & \text{if } r < b, \\ b^3/(3r), & \text{if } r \geq b, \end{cases} \quad (3.14)$$

is the well-known harmonic potential of a sphere of radius  $b$ . We have also employed in (3.13) the obvious identity  $\partial \varphi_b(x) / \partial b \Big|_{b=a} = 4\pi a^2 H_0(x)$ , with the *same* function  $H_0(x)$  as in (3.6)<sub>2</sub>. The latter immediately implies that the classes (3.6) and (3.12) coincide, which explains the appearance of the Doi bound (in its form (3.11)) here. In our derivation we have employed, however,



the “particle-center” correlation  $F^{PC}(r)$  which is simpler than the interfacial correlations. Also, the herein proposed variational procedure, based on the R-T principle (3.3), (3.4), is considerably simpler than the one employed by Talbot and Willis [8].

#### 4 A Discussion of the 2-D Case

In the 2-D case the spheres are replaced by an array of identical parallel circular cylinders. Equivalently, we can consider a “perforated” plane, in which the circular perforations are randomly and nonoverlappingly positioned. The R-T variational principle and the bound (3.5) remain unchanged. The only difference is that the 3-D Green function in (3.6) should be replaced by its 2-D counterpart  $G(x) = -\ln|x|/(2\pi) + \text{constant}$ . But the convolution integral (3.9) becomes then *divergent*, since its integrand behaves as  $1/|x|$  when  $|x| \rightarrow \infty$ . As a result, it can be easily shown that the counterpart of our procedure (Section 3), that has lead us to the DTW bound (3.11), *degenerates* in the 2-D case, due to the aforementioned asymptotic behaviour of the appropriate Green’s function in infinity. In this sense the situation is fully similar to the Talbot and Willis’ procedure [8] which also fails in 2-D, due to convergence difficulties, as noticed by the authors (private communication).

It is noted that Doi’s procedure, corresponding to the trial fields (3.12) coupled with the R-T principle, fails in 2-D as well. The reason is obvious, if one writes down the denominator of (3.5) in the form

$$\langle |\nabla u(x)|^2 \rangle = K^2 \int \Gamma(x) [F^{PP}(x) - 2\xi F^{SP}(x) + \xi^2 F^{SS}(x)] dx,$$

where  $F^{SP}(x) = \langle I_1(0) |\nabla I_1(x)| \rangle$  and  $F^{SS}(x) = \langle |\nabla I_1(0)| |\nabla I_1(x)| \rangle$  are the interfacial correlations [7, 9], and  $\Gamma(x)$  is the convolution integral

$$\Gamma(x) = \int \nabla_y G(x-y) \cdot \nabla_y G(y) dy$$

which, similarly to  $\chi(x)$ , see Eq. (3.9), is *divergent* in the 2-D case under study.

The failure of all the above mentioned variational procedures in 2-D does not mean however that the homogenization of the problem (3.1) is impossible in this case. The failures should be attributed to the procedures themselves, and presumably, to the fact that in 2-D the interactions between sinks become much stronger than in 3-D and hence trial fields, that somewhat more essentially incorporate these interactions, must be employed.

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## References

1. R. L. Stratonovich, *Topics in Theory of Random Noises*, vol. 1, Gordon and Breach, New York, 1963.
2. S. Torquato, *J. Stat. Phys.* **45** (1986) 843.
3. K. Z. Markov and J. R. Willis, *Mathematical Models and Methods in Applied Sciences* **8** (1998) 359.
4. K. Z. Markov, On the two-point correlation functions in random arrays of nonoverlapping spheres, *Annuaire (Godishnik) Univ. Sofia, Fac. Mat. Inf., Livre 2, Mat. Appl. et Informatique* **91/1997**, 151–175.
5. W. J. Drugan and J. R. Willis, *J. Mech. Phys. Solids*, **44** (1996) 497–524.
6. B. U. Felderhof and J. M. Deutch, *J. Chem. Phys.* **64** (1976) 4551.
7. M. Doi, *J. Phys. Soc. Japan* **40** (1976) 567.
8. D. R. S. Talbot and J. R. Willis, *Proc. R. Soc. London A* **370** (1980) 351.
9. J. Rubinstein and S. Torquato, *J. Chem. Phys.* **88** (1988) 6372.
10. S. Torquato and J. Rubinstein, *J. Chem. Phys.* **90** (1989) 1644.
11. P. M. Richards and S. Torquato, *J. Chem. Phys.* **87** (1987) 4612.
12. J. D. Beasley and S. Torquato, *Phys. Fluids A* **1** (1989) 199.