

## Increase and Decrease of the Effective Conductivity of Two Phase Composites due to Polydispersity

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We present a two-dimensional mathematical model of a composite material with conducting inclusions (fibers) embedded in a matrix. Our main objective is to study how polydispersity (two different sizes of particles) affects the overall conductivity of the composite. If the conductivity of inclusions is higher than the conductivity of the matrix, then previous studies suggest an increase of the effective conductivity due to polydispersity. We show that for high volume fraction when inclusions are not well-separated and percolation effects play a significant role, polydispersity may result in either an increase or decrease of the effective conductivity. Our proof is based on the method of functional equations and it provides sufficient conditions for both the increase and the decrease of the effective conductivity.

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**KEY WORDS:** Effective conductivity; polydispersity; random composite material; functional equation.

### 1. INTRODUCTION

We consider the problem of determining the effective conductivity of polydispersed fiber composites when inclusions (unidirectional fibers) of conductivity  $\lambda_i$  of two different sizes are randomly embedded in a matrix of conductivity  $\lambda_m$ . A cross-section of such composite is presented in the Fig. 1. The inclusions are modeled by disks of two different radii  $R_l$  (large) and  $R_s$  (small),  $R_l \geq R_s$ .

We introduce a *polydispersity parameter*  $p$ ,  $0 < p < 1$ , which characterizes the relative volume fraction of small disks ( $p = 0$  all disks are large,

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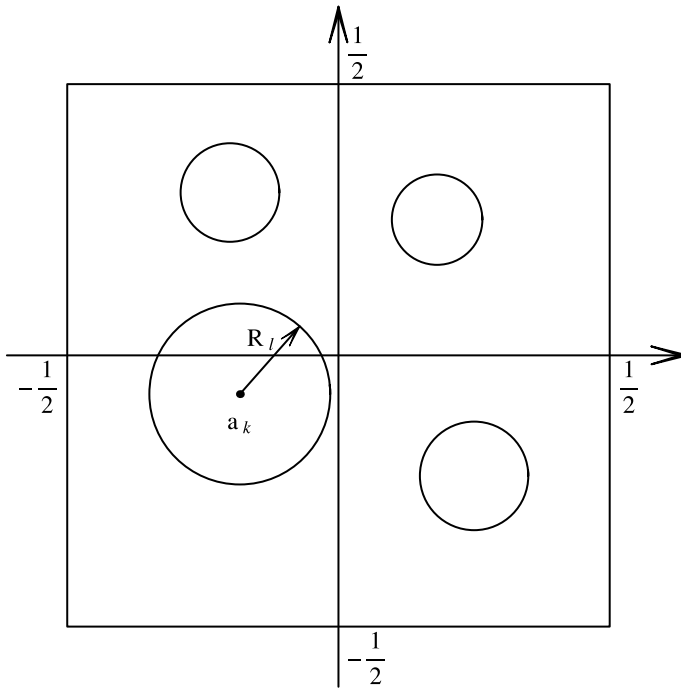


Fig. 1. Bimodal composite.

$p = 1$  all disks are small) for a fixed total volume fraction of disks  $v$ ; that is,  $pv$  is the total volume fraction of small disks. Our main objective is to obtain the analytical dependence of the effective conductivity  $\hat{\lambda}_{\text{poly}}$  on the polydispersity parameter  $p$ . This allows for qualitative and quantitative comparison of the effective conductivities of the poly- and mono-dispersed composites  $\hat{\lambda}_{\text{poly}}(p)$  and  $\hat{\lambda}_{\text{mono}} = \hat{\lambda}_{\text{poly}}(0) = \hat{\lambda}_{\text{poly}}(1)$ .

There is considerable interest in the effect of polydispersity on both the effective conductivity  $\hat{\lambda}$  and the effective viscosity  $\hat{\mu}$  of suspension of rigid inclusions in a fluid (see refs. 7, 11, 14–16, 27). For a Stokes fluid these problems are very similar in nature. The main question here is whether the presence of polydispersity results in an *increase or decrease of the effective conductivity* (for the sake of definiteness we assume hereafter that  $\lambda_i > \lambda_m$ ). Theoretical results on this subject reviewed below point to an increase (see (1) below). This question is of significant practical interest; when polymer/ceramic composites are prepared for capacitors or thermal insulating packages<sup>(14,15)</sup>, should the ceramic powder be monodispersed or polydispersed to achieve desired (e.g. high) dielectric or thermal properties. How significant is the effect of polydispersity? Limited experimental

data<sup>(7,11,14–16,27,28)</sup> are inconclusive and point to an increase<sup>(28)</sup> when the concentration of inclusions is not high. Numerical examples presented in ref. 5 show the decrease (see (3)) when most of the inclusions are large and the total concentration is high. It was suggested in ref. 5 that this decrease is due to percolation effects when small inclusions may not participate in the conducting cluster and their presence effectively reduces the total volume fraction of inclusions. However, this suggestion was not supported by theoretical analysis. In this paper we have developed mathematical models which show that both the increase and decrease occur and provide sufficient conditions for each type of the effective behavior.

Most theoretical works on this subject study polydispersity in the dilute limit case. Thovert *et al.*<sup>(30,31)</sup> (see also ref. 29) applied the techniques of variational bounds<sup>(21)</sup> to estimate  $\hat{\lambda}_{\text{poly}}$  in the low concentration regime in both two and three dimensions. They performed rigorous asymptotic analysis as  $v \rightarrow 0$  and evaluated the terms of order  $O(v)$  and  $O(v^2)$  in the expansion of  $\hat{\lambda}_{\text{poly}}$  in powers of  $v$ , where  $v$  is the total volume fraction of the inclusions. Moreover, they perform partial analysis of the cubic term. The analysis of refs. 29–31 shows that if the conductivity of inclusions is greater than the conductivity of matrix ( $\lambda_i > \lambda_m$ ), then

$$\hat{\lambda}_{\text{poly}} > \hat{\lambda}_{\text{mono}}. \quad (1)$$

The key ingredient of the analysis of refs. 29–31 is an observation that the three- and four-point lower bounds on  $\hat{\lambda}_{\text{poly}}$  and  $\hat{\lambda}_{\text{mono}}$  provide a good approximation of the effective conductivity when the contrast parameter

$$\rho = \frac{\lambda_i - \lambda_m}{\lambda_i + \lambda_m}, \quad (2)$$

introduced by Bergman<sup>(3)</sup>, is not large. Note that formulas for  $\hat{\lambda}_{\text{poly}}$  derived in refs. 29–31 provide good numerical results for  $v < v_{\text{cr}}$  where  $v_{\text{cr}}$  is the percolation threshold.

Robinson and Friedman in ref. 28 considered the problem of the effective conductivity for spherical inclusions of two different sizes in three dimensions. Their derivation of the effective conductivity is based on the Maxwell–Garnett formula. Note that the Maxwell–Garnett formula is an approximation in  $v$  for small  $v$ .<sup>(21)</sup> This formula was derived as an approximation for the dilute limit case when interactions between inclusions are small (see ref. 21, Chapter IX). While it is known that for special geometric arrays the Maxwell–Garnett formula holds for fairly large concentrations (e.g. for periodic structure<sup>(2)</sup>) and other well-separated geometries

(see ref. 21, Section 10.4) in general it only holds to the second order in  $v$  for macroscopically isotropic composites (see ref. 21, p. 192). In particular, the Maxwell–Garnett approximation may not hold for high volume fractions in the presence of pronounced percolation effects, when the conductivity patterns dominate the behavior.

Also for a model of a medium with randomly located identical circular disks (shaking model) it follows from ref. 4 (see formulas (5.1) and (5.4)) that linear and quadratic terms of the Maxwell–Garnett formula do not depend on the random locations and are determined solely by the total volume fraction and material properties. However, it also follows from these formulas that the cubic term in  $v$  depends on the random locations of inclusions. Hence, this model provides an example of a composite, with inclusions that are not well-separated, for which the Maxwell–Garnett formula is incorrect in the third order.

In the present paper we model randomness by introducing shaking parameters which characterise the random locations of the centers of the disks (see Section 4 for precise definition). We propose two models: (i) the “bumping model”, and (ii) “the well-separated model”. The model (i) describes random geometrical arrays when inclusions are highly packed and not well-separated. The key ingredient of this model is the introduction of two shaking parameters  $d_l$  and  $d_s$  (depending on the sizes of the inclusions) for large and small disks, respectively. The only restriction on  $d_l$  and  $d_s$  is that the disks cannot overlap (but can be very close to each other). In the “the well-separated model” the shaking parameter is the same for large and small disks. Model (ii) clearly can be applied in the dilute case and it is not surprising that this model leads to the same conclusion (1). However, the central result of our work is that the model (i) predicts regimes where either (1) or the opposite inequality

$$\hat{\lambda}_{\text{poly}} < \hat{\lambda}_{\text{mono}} \quad (3)$$

holds, depending on the value of the polydispersity parameter  $p$  (the relative volume fractions of large and small disks). This result is consistent with percolation analysis of checkerboard models with cells of two different sizes (L. Berlyand, A. Pisztor, unpublished) and a numerical example in ref. 5.

Our analysis is based on the method of functional equations proposed in refs. 13, 26 and further developed for periodic media in refs. 4, 22 and 25. We generalize this method for polydispersed geometries. The key technical point is to incorporate parameter  $\nu_k$  (defined in (4)) into a series expansion for the solutions of the functional equations. Note that these

expansions are equivalent to successive approximations to functional equations. This observation implies the convergence of the series for the effective conductivity and electric potential by using convergence results for successive approximations of solutions to general functional equations.

**2. FORMULATION OF THE BOUNDARY VALUE PROBLEM**

Consider a periodic two-dimensional lattice  $\mathcal{Q}$  which is defined by the two fundamental translation vectors 1 and  $i(i^2 = -1)$  in the complex plane  $\mathbf{C} \cong \mathbf{R}^2$  of the complex variable  $z = x + iy$ . The zeroth cell  $\mathcal{Q}_{(0,0)}$  is the unit square  $\{z = x + iy \in \mathbf{C} : -1/2 < x, y < 1/2\}$ . The lattice  $\mathcal{Q}$  consists of the cells  $\mathcal{Q}_{(m_1, m_2)} = \mathcal{Q}_{(0,0)} + m_1 + im_2$ . Assume that the zeroth cell  $\mathcal{Q}_{(0,0)}$  contains mutually nonoverlapping disks  $D_k$  (inclusions) of radii  $r_k$  centered at  $a_k$  that is  $D_k := \{z \in \mathbf{C} : |z - a_k| < r_k\} (k = 1, 2, \dots, N)$ . The radii  $r_k$  take one of the two values  $R_s$  and  $R_l$  ( $R_s \leq R_l$ , small and large disks). Let us introduce a variable

$$v_k = r_k^2 R_l^{-2} \tag{4}$$

which assumes one of the two values 1 and a parameter

$$\chi = (R_s/R_l)^2. \tag{5}$$

The total volume fraction (concentration) of the inclusions is

$$v = N\pi(R_s^2 p + R_l^2(1 - p)), \tag{6}$$

where  $p$  is the relative volume fraction of the disks of radius  $R_s$ . We also introduce the notations for the circumference of the disks  $T_k := \{z \in \mathbf{C} : |z - a_k| = r_k$ , and for the part of the unit square occupied by the matrix  $\Omega := \mathcal{Q}_{(0,0)} \setminus (\cup_{k=1}^N D_k \cup T_k)$ .

We study the conductivity of the doubly periodic composite material, when the domains  $\Omega$  (matrix) and  $D_k$  (inclusions) are occupied by materials of conductivities  $\lambda_m$  and  $\lambda_i$ , respectively (Fig. 1). The potential  $u(x, y)$  satisfies the Laplace equation

$$\Delta u = 0 \text{ in } \cup_{k=1}^N D_k \cup \Omega \tag{7}$$

with the conjugation conditions:

$$u^+ = u^-, \lambda_m \frac{\partial u^+}{\partial n} = \lambda_i \frac{\partial u^-}{\partial n} \text{ on } T_k, k = 1, 2, \dots, N, \tag{8}$$

where  $\frac{\partial}{\partial n}$  is the outward normal derivative and

$$u^+(\xi, \eta) := \lim_{x+iy \rightarrow \xi+i\eta, x+iy \in \Omega} u(x, y),$$

$u^-(\xi, \eta)$  is introduced in a similar way with  $\xi + i\eta \in T_k, k = 1, 2, \dots, N$ . We also impose on  $u(x, y)$  the quasi periodicity conditions which correspond to an external field applied in the  $x$ -direction

$$u(x + 1, y) = u(x, y) + 1, \quad u(x, y + 1) = u(x, y). \tag{9}$$

Our main objective is to find the analytical dependence of the effective conductivity defined as follows (ref. 9, p. 15, ref. 21)

$$\hat{\lambda} = \int_{Q(0,0)} \lambda(x, y) |\nabla u|^2 dx dy = \lambda_m \int_{\Omega} u_x dx dy + \lambda_i \sum_{k=1}^N \int_{D_k} u_x dx dy \tag{10}$$

on parameters  $p$  and  $\chi$  which characterize polydispersity. In Section 3 we compute  $\hat{\lambda}$  for an arbitrary deterministic array of inclusions. In (10)  $\lambda(x, y) = \lambda_m$ , if  $(x, y) \in \Omega$ , and  $\lambda(x, y) = \lambda_i$ , if  $(x, y) \in D_i$ . In Section 4 we consider analogous problems for random locations of  $a_k$  and random distributions of the disk's radii  $r_k$ , and obtain an analogous dependence in this random setting (which is a much harder problem).

It is convenient to rewrite problem (7)–(9) in terms of complex potentials  $\psi(z)$  and  $\psi_k(z)$  which are analytic in  $\Omega$  and  $D_k$ , respectively, and continuous in the closures of  $\Omega$  and  $D_k$ , refs. 1, 4.

$$\psi(z) = \frac{\partial u}{\partial x} - i \frac{\partial u}{\partial y} - 1, \quad z \in \Omega, \quad \psi_k(z) = \frac{\lambda_i + \lambda_m}{2\lambda_m} \left( \frac{\partial u}{\partial x} - i \frac{\partial u}{\partial y} \right), \quad z \in D_k. \tag{11}$$

Straightforward calculations imply from (9) and (11) that

$$\psi(z + 1) = \psi(z) = \psi(z + i). \tag{12}$$

The two real conditions (8) yield (see ref. 26 and Appendices A and B from ref. 4)

$$\begin{aligned} \psi(t) &= \psi_k(t) + \rho \left( \frac{r_k}{t - a_k} \right)^2 \overline{\psi_k(t)} - 1, \\ |t - a_k| &= r_k, \quad k = 1, 2, \dots, N, \end{aligned} \tag{13}$$

where  $\rho = (\lambda_i - \lambda_m)/(\lambda_i + \lambda_m)$ . The Eqs. (12) and (13) form a generalization of the Riemann–Hilbert problem for the unknown functions  $\psi$  and  $\psi_k$  (also known as the  $R$ -linear problem, ref. 26). A conventional way to solve such problems is to reduce them to integral equations and then solve numerically. Following ref. 4 we next show that this problem can be reduced to a system of functional equations. In the remaining part of this section we solve this system analytically in series in  $R_I^2$ . The analytical dependence on  $R_I^2$  has been justified in refs. 22 and 25.

The boundary value problem (13) can be reduced to a system of functional equations

$$\psi_q(z) = \rho \sum_{k=1}^N \sum_{m_1, m_2}^* (W_{m_1 m_2 k} \psi_k)(z) + 1, \quad |z - a_q| \leq r_q, \quad q = 1, 2, \dots, N, \tag{14}$$

where  $\sum_{m_1, m_2}^*$  is taken over all integer  $m_1$  and  $m_2$  except  $m_1 = m_2 = 0$  for  $k = q$ . Operator  $W_{m_1 m_2 k}$  is defined as follows

$$(W_{m_1 m_2 k} \psi_k)(z) = \left( \frac{r_k}{z - a_k - m_1 - im_2} \right)^2 \overline{\psi_k \left( \frac{r_k^2}{t - a_k - m_1 - im_2} + a_k \right)}. \tag{15}$$

The properties of this operator and the reduction of (13) to (14) are outlined in Appendix A.

We seek the solution of functional Eq. (14) in form of power series in  $R_I^2$

$$\psi_m(z) = \sum_{q=0}^{\infty} \psi_m^{(q)}(z) \left( R_I^2 \right)^q, \quad m = 1, 2, \dots, N. \tag{16}$$

Then the coefficients  $\psi_m^{(q)}(z)$  are analytic in  $z$  and depend on  $R_s$  as a parameter. (Expansion in  $R_S^2$  yields the same result.) We next write each function  $\psi_m^{(q)}(z)$  in the form of Taylor series

$$\psi_m^{(q)}(z) = \sum_{n=0}^{\infty} \psi_{nm}^{(q)}(z - a_m)^n, \quad m = 1, 2, \dots, N. \tag{17}$$

We then have

$$\begin{aligned} \sum_{m_1, m_2} (W_{m_1 m_2 m} \psi_m^{(q)})(z) &= \sum_{m_1, m_2} \sum_{n=0}^{\infty} \overline{\psi_{nm}^{(q)}} \frac{r_m^{2(n+1)}}{(z - a_m - m_1 - im_2)^{n+2}} \\ &= \sum_{n=0}^{\infty} \overline{\psi_{nm}^{(q)}} r_m^{2(n+1)} E_{n+2}(z - a_m), \end{aligned} \tag{18}$$

where  $E_n(z)$  is the modified Eisenstein’s function of order  $n$  (for details see (B.2) and (B.7)). Convergence of the series in (18) follows from general results from the method of functional equations<sup>(25,26)</sup>. Substituting (16), (17) in (14) and using (18), (4) we obtain the relation

$$\begin{aligned} \sum_{q=0}^{\infty} \psi_m^{(q)}(z) R_l^{2q} &= \rho \sum_{k=1}^N \sum_{n=0}^{\infty} \sum_{q=0}^{\infty} \psi_{nk}^{(q)} R_l^{2(q+n+1)} v_k^{n+1} E_{n+2}(z - a_k) + 1, \\ |z - a_k| &\leq r_m, \quad m = 1, 2, \dots, N. \end{aligned} \tag{19}$$

Equating the coefficients of like powers of  $R_l^2$  in (19) we arrive at the following theorem in which we determine  $\psi_k^{(q)}(z)$  recursively and thus define the solution  $\psi_k(z)$  of the functional Eq. (14) (compare with Theorem 3.4 from ref. 4):

**Theorem 2.1.** Let  $\psi_k(z)$  be a solution of functional Eq. (14). Then it admits the representation (16), where

$$\begin{aligned} \psi_k^{(0)}(z) &= 1, \\ \psi_k^{(q+1)}(z) &= \rho \left[ \overline{\psi_{0k}^{(q)}} v_k E_2(z - a_k) + \overline{\psi_{1k}^{(q-1)}} v_k^2 E_3(z - a_k) + \dots \right. \\ &\quad \left. + \overline{\psi_{q-1,k}^{(1)}} v_k^q E_{q+1}(z - a_k) \right], \quad q = 0, 1, 2, \dots \end{aligned} \tag{20}$$

Here  $\psi_{lk}^{(q)}$  is the  $l$ th coefficient of the Taylor expansion of  $\psi_k^{(q)}(z)$  (see 17)). The function  $\psi_k(z)$  is represented in the form of the uniformly convergent series (16) in the closure of  $D_k$ .

From (20) we obtain explicit formulas

$$\begin{aligned} \psi_m^{(1)}(z) &= \rho \sum_{k=1}^N v_k E_2(z - a_k), \tag{21} \\ \psi_m^{(2)}(z) &= \rho^2 \sum_{k, k_1}^N v_k v_{k_1} \overline{E_2(a_k - a_{k_1})} E_2(z - a_k), \end{aligned}$$



$$\begin{aligned} \psi_m^{(3)}(z) = & \rho^3 \sum_{k,k_1,k_2}^N v_k v_{k_1} v_{k_2} \overline{E_2(a_k - a_{k_1})} E_2(a_{k_1} - a_{k_2}) E_2(z - a_{k_2}) \\ & - 2\rho^2 \sum_{k,k_1}^N v_k^2 v_{k_1} \overline{E_3(a_k - a_{k_1})} E_3(z - a_k) \end{aligned}$$

and so on.

Thus, we have determined the functions  $\psi_k(z)$  in (11) recursively; it is shown in the next section that these functions determine the effective conductivity, so that it is not necessary to define  $\psi(z)$ .

### 3. EFFECTIVE CONDUCTIVITY

The local relation between the flux  $\mathbf{q}$  and the gradient  $\nabla u$  in the unit cell  $Q_{(0,0)}$  is given by Fourier's law in thermal conductivity or Ohm's law in electric problems

$$\mathbf{q} = \begin{cases} \lambda_m \nabla u & \text{in } \Omega \\ \lambda_i \nabla u & \text{in } \cup_{k=1}^N D_k. \end{cases} \tag{22}$$

The effective conductivity tensor in two dimensions

$$\Lambda = \begin{pmatrix} \hat{\lambda}^x & \hat{\lambda}^{xy} \\ \hat{\lambda}^{xy} & \hat{\lambda}^y \end{pmatrix} \tag{23}$$

relates the averaged values over the unit cell of the flux and the gradient

$$\hat{\mathbf{q}} = \Lambda \hat{\nabla} u. \tag{24}$$

In (24) the superscript  $\hat{\phantom{x}}$  stands for the integral over  $Q_{(0,0)}$ . In order to determine  $\Lambda$  it is sufficient to solve two cell problems<sup>(9)</sup> when the external field applied either in the  $x$ - (see (7) and (8)) or  $y$ -direction. Let  $\psi_k(z)$  be the complex potential corresponding to the problem (7) and (8). Then we have<sup>(4,20,25)</sup>

$$\hat{\lambda}^x - i \hat{\lambda}^{xy} = 1 + 2\rho \sum_{k=1}^N \pi r_k^2 \psi_k(a_k). \tag{25}$$

For simplicity we assume that our composite material is macroscopically isotropic. Then the effective tensor  $\Lambda$  is of the form  $\Lambda = \hat{\lambda} I$ , where  $\hat{\lambda}$  is the effective conductivity and  $I$  is the unit tensor. In this case (25) becomes

$$\hat{\lambda} = 1 + \frac{2\rho v}{K_1(\chi)N} \sum_{k=1}^N v_k \psi_k(a_k), \tag{26}$$

where  $v$  is the total volume fraction of inclusions and

$$K_1(\chi) = \chi p + 1 - p. \tag{27}$$

Here  $\chi$  has the form (5). Using (16) and (26) we write  $\hat{\lambda}$  in the form of a series in the total volume fraction  $v$

$$\hat{\lambda} = 1 + 2\rho v \left[ B_0 + B_1 v + B_2 v^2 + \dots \right], \tag{28}$$

where

$$B_q = \frac{1}{\pi^q N^{q+1} K_1^{q+1}(\chi)} \sum_{m=1}^N v_m \psi_m^{(q)}(a_m), \quad q = 0, 1, \dots \tag{29}$$

In order to evaluate  $\psi_m^{(q)}$  we introduce

$$Z_{p_1, p_2, \dots, p_M}^{k_0, k_1, \dots, k_M} = E_{p_1}(a_{k_0} - a_{k_1}) \overline{E_{p_2}(a_{k_1} - a_{k_2})} \dots \mathbf{C}^{M-1} E_{p_M}(a_{k_{M-1}} - a_{k_M}),$$

where  $\mathbf{C}$  is the complex conjugation operator,  $M = 1, 2, 3, \dots$ ;  $k_j$  can assume the values  $1, 2, \dots, N$ ;  $p_j = 1, 2, 3, \dots$ . Let  $P = p_1 + p_2 + \dots + p_M - M + 1$  and introduce

$$X_{p_1, p_2, \dots, p_M} = \frac{1}{K_1(\chi)^P} \sum_{k_0, k_1, \dots, k_M} v_{k_0} v_{k_1}^{p_1-1} \dots v_{k_M}^{p_M-1} Z_{p_1, p_2, \dots, p_M}^{k_0, k_1, \dots, k_M}. \tag{30}$$

In accordance with (20),  $v_{k_0}$  corresponds to  $v_k$  from (26) and each function  $E_n$  from (20) has the coefficient  $v_k^{q-1}$ . Combining (28)–(30) we obtain the coefficients in (28):

$$\begin{aligned} B_0 = 1, \quad B_1 = \frac{\rho}{\pi N^2} X_2, \quad B_2 = \frac{\rho^2}{\pi^2 N^3} X_{22}, \\ B_3 = \frac{1}{\pi^3 N^4} \left[ -2\rho^2 X_{33} + \rho^3 X_{222} \right]. \end{aligned} \tag{31}$$

It is possible to proceed and calculate the next coefficients  $B_q$ ,  $q > 3$ . It follows from Theorem 2.1 that  $B_q$  has the following structure

$$B_q = \sum_{p_1, p_2, \dots, p_M; k_0, k_1, \dots, k_M} \beta_{q, p_1, p_2, \dots, p_M} \frac{v_{k_0} v_{k_1}^{p_1-1} \dots v_{k_M}^{p_M-1}}{K_1(\chi)^P} Z_{p_1, p_2, \dots, p_M}^{k_0, k_1, \dots, k_M}, \tag{32}$$

where the constants  $\beta_{q, p_1, p_2, \dots, p_M}$  depend only on  $\rho$  and  $N$ . For instance,  $B_1$  involves only one term of the type (30) with the coefficient  $\beta_{12} = \frac{\rho}{\pi N^2}$  (see the second equality in (31)).

**Remark 3.1.** For each given  $q, p_1, p_2, \dots, p_M$  the coefficient  $\beta_{q, p_1, p_2, \dots, p_M}$  can be obtained in explicit form. However, for forthcoming analysis of random models a constructive algorithm for  $\beta_{q, p_1, p_2, \dots, p_M}$  which follows from (29) and recursive formulas (20) is sufficient.

From (32) and (30) it follows that  $B_q$  is a sum of  $X_{p_1, p_2, \dots, p_M}$  with coefficients depending only on  $\rho$  and  $N$ . Each term  $X_{p_1, p_2, \dots, p_M}$  is a sum of  $Z_{p_1, p_2, \dots, p_M}^{k_0, k_1, \dots, k_M}$ , which depend only on the locations of the centers  $a_{k_0}, a_{k_1}, \dots, a_{k_M}$ . The coefficients  $v_{k_0} v_{k_1}^{p_1-1} \dots v_{k_M}^{p_M-1} K_1(\chi)^{-P}$  depend only on the radii of the inclusions. This decomposition of  $B_q$  holds for each parameters  $\rho, a_k, R_l, R_s$  for which the inclusions  $D_k$  do not overlap.

#### 4. EFFECTIVE CONDUCTIVITY OF A RANDOM POLYDISPERSED COMPOSITE

In this section we apply the formulas from Section 3 and previous results<sup>(4)</sup> to evaluate the effective conductivity of an isotropic random composite material. The total volume fraction  $v$  is always fixed. We consider two different types of random variables. First, the value (4) is considered as a discrete random variable defined on the set of indices  $k = 1, 2, \dots, N$  which enumerate disks, so

$$v_k = \begin{cases} \chi = (R_s/R_l)^2 & \text{with the probability } p, \\ 1 & \text{with the probability } 1 - p. \end{cases} \tag{33}$$

The notation  $\mathbf{E}[\cdot]$  is used for the expectation of  $v_k$  in the probability space of Bernoulli sequences of length  $N$ . Next we introduce random variables  $a_k$  ( $k = 1, 2, \dots, N$ ), which describe the random position of the centers. These variables  $a_k$  ( $k = 1, 2, \dots, N$ ) are independent and their distribution can be described as follows. Consider the basic periodicity cell  $Q_{(0,0)}$  and suppose that the cell contains  $N = n^2$  inclusions, where  $n$  is an integer number. Partition  $Q_{(0,0)}$  into  $N$  equal subcells so their centers  $b_k$  form a square array in  $Q_{(0,0)}$ . For example, if  $N = 4$  we have  $b_1 = \frac{1}{4}(1 + i)$ ,  $b_2 = \frac{1}{4}(-1 + i)$ ,  $b_3 = \frac{1}{4}(-1 - i)$ ,  $b_4 = \frac{1}{4}(1 - i)$ .

The probability distribution of  $a_k$ ,  $k=1, 2, \dots, N$  is defined as follows. In each subcell introduce the “shaking disk”  $G_k := \{z \in \mathbf{C} : |z - b_k| < d_k\}$  of radius  $d_k$  centered at  $b_k$ . The location of the point  $a_k$  (center of the disk  $D_k$ ) is randomly chosen within the disk  $G_k$  with the density  $(\pi d_k)^{-1} = |G_k|^{-1}$  of the equiprobable distribution. Since we assume that each disk (inclusion)  $D_k$  lies in a subcell of size length  $\frac{1}{\sqrt{N}}$  the shaking parameter  $d_k$  must satisfy the natural condition  $2(d_k + r_k) < \frac{1}{\sqrt{N}}$ , where  $r_k = \sqrt{v_k} R_1$  is the random variable defined by the random variable  $v_k$  from (33) (Fig. 2). Hence, we have

$$d_k + r_k < \frac{1}{2\sqrt{N}}. \quad (34)$$

It will be clear that in the two models described below inequality (34) represents the non-overlapping of the disks  $D_k$  which models impenetrable inclusions. We will consider two models of polydispersity: well-separated model in which all shaking parameters  $d_k$  are identical and the bumping model in which shaking parameters  $d_k$  are determined by the sizes of the inclusions.

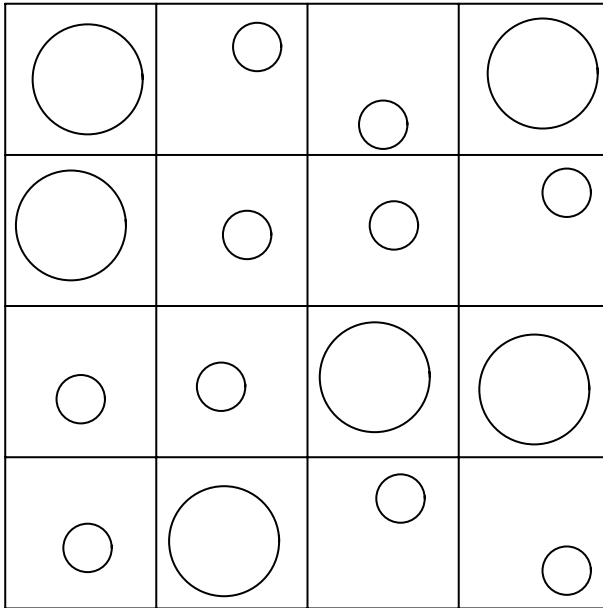


Fig. 2. Shaking of inclusions within small squares.

Below we consider functions  $f(a_1, a_2, \dots, a_N; v_1, v_2, \dots, v_N)$ , where  $v_1, v_2, \dots, v_N$  are independent random variables defined above. The random variables  $a_1, a_2, \dots, a_N$  are mutually independent, but may depend on  $v_1, v_2, \dots, v_N$ . Denote by

$$F(v_1, v_2, \dots, v_N) = \langle f(a_1, a_2, \dots, a_N; v_1, v_2, \dots, v_N) \rangle$$

the expectation of  $f$  as a function of the random variables  $a_1, a_2, \dots, a_N$  ( $v_k, k=1, 2, \dots, N$  are parameters). More precisely,  $\langle \cdot \rangle$  means the expectation in the probability space defined as a product of the probability spaces of each  $a_k$ . Further, one can calculate the expectation

$$\mathbf{E}[F(v_1, v_2, \dots, v_N)] = \mathbf{E}[\langle f(a_1, a_2, \dots, a_N; v_1, v_2, \dots, v_N) \rangle] \quad (35)$$

with respect to random variables  $v_1, v_2, \dots, v_N$ . Note that it is possible to define a probability space which corresponds to a joint distribution of all  $a_k$  and  $v_k$  and introduce a corresponding “double” expectation in all of these random variables. This double expectation leads to the same result as consecutive expectations (35). This model is a generalization of the polydispersed case of the analogous shaking model introduced in ref. 4 for monodispersed random composites where only the expectation  $\langle \cdot \rangle$  was used.

Since our objective is to provide comparison between poly- and mono-dispersed composites, we review briefly the results of ref. 4 using notations from this paper. If the disks are identical, then  $\chi = 1$  in (5) and (28) becomes

$$\hat{\lambda} = 1 + 2\rho v \left[ 1 + A_1 v + A_2 v^2 + \dots \right], \quad (36)$$

where  $A_q = B_q|_{\chi=1}$ . The effective conductivity  $\hat{\lambda}$  has been calculated in ref. 4. In particular the following representation has been obtained

$$A_q = \sum_{p_1, p_2, \dots, p_M; k_0, k_1, \dots, k_M} \beta_{q, p_1, p_2, \dots, p_M} Z_{p_1, p_2, \dots, p_M}^{k_0, k_1, \dots, k_M}, \quad (37)$$

where  $\beta_{q, p_1, p_2, \dots, p_M}$  are defined by (29), (32) (and are not random). Note that the coefficients  $\beta_{q, p_1, p_2, \dots, p_M}$  and  $Z_{p_1, p_2, \dots, p_M}^{k_0, k_1, \dots, k_M}$  are the same in (32) (polydisperse) and (37) (monodisperse). The following results has been established in ref. 4.

**Theorem 4.1.** Suppose that the centers  $a_k$  are equiprobably located in the shaking disks  $G_k$  of the equal radii ( $d_1 = d_2 = \dots = d_N = d$ ). Then

$\langle Z_{p_1, p_2, \dots, p_M}^{k_0, k_1, \dots, k_M} \rangle$  is represented in the form of the power series in  $d^2$

$$\langle Z_{p_1, p_2, \dots, p_M}^{k_0, k_1, \dots, k_M} \rangle = \sum_{s=0}^{\infty} \gamma_{p_1, p_2, \dots, p_M}^{k_0, k_1, \dots, k_M}(s) d^{2s}. \tag{38}$$

Moreover, the following inequality holds

$$\beta_{q, p_1, p_2, \dots, p_M} \gamma_{p_1, p_2, \dots, p_M}^{k_0, k_1, \dots, k_M}(s) \geq 0 \tag{39}$$

for all  $q, p_1, p_2, \dots, p_M; k_0, k_1, \dots, k_M;$  and  $s = 0, 1, 2, \dots$

**Remark 4.2.** The coefficients  $\gamma_{p_1, p_2, \dots, p_M}^{k_0, k_1, \dots, k_M}(s)$  are defined in ref. 4 with  $\langle Z_{p_1, p_2, \dots, p_M}^{k_0, k_1, \dots, k_M} \rangle = \pi^2 d^4 J_{p_1, p_2, \dots, p_M}^*$ . More precisely, (38) were presented in<sup>(4)</sup> in the form (see Appendix D from ref. 4 and, in particular, Lemma 6.2 for details)

$$\langle Z_{p_1, p_2, \dots, p_M}^{k_0, k_1, \dots, k_M} \rangle = \sum_{s_0, \dots, s_M} \Gamma_{p_1, p_2, \dots, p_M}^{k_0, k_1, \dots, k_M}(s_0, \dots, s_M) d^{2(s_0 + \dots + s_M - 1)}. \tag{40}$$

**Remark 4.3.** In ref. 4 (39) was proved for  $s = 1, 2, \dots$ . Since  $s = 0$  is necessary for polydispersed case, we prove it in Appendix C.

**Remark 4.4.** Inequality (39) is very useful. For example, by combining (36)–(39) the following extremal property of the regular array was proven:<sup>(4)</sup>

$$\hat{\lambda}(d=0) < \hat{\lambda}(d > 0). \tag{41}$$

According to (35) we define the effective conductivity of a random polydispersed composite  $\hat{\lambda}_{\text{poly}}$  by averaging  $\hat{\lambda}$  (see (10)) over random locations and random radii

$$\hat{\lambda}_{\text{poly}} = \mathbf{E}[\langle \hat{\lambda} \rangle]. \tag{42}$$

Taking the expectation  $\langle \cdot \rangle$  and  $\mathbf{E}[\cdot]$  in (28) we obtain (see the first formula (31) for  $B_0$ )

$$\hat{\lambda}_{\text{poly}} = 1 + 2\rho v \left( 1 + \mathbf{E}[\langle B_1 \rangle] v + \mathbf{E}[\langle B_2 \rangle] v^2 + \dots \right), \tag{43}$$

where  $0 < v < v_{\text{max}}$  and  $v_{\text{max}} = \frac{\pi}{4}(p\chi + 1 - p)$  corresponds to touching inclusions. Below we investigate the dependence of  $\hat{\lambda}_{\text{poly}}$  on  $p$  for fixed  $\chi = (R_s/R_l)^2$ . Then in this case we have  $v_{\text{max}} = \frac{\pi\chi}{4}$ .

**4.1. Well-Separated Model (Identical Shaking Parameters)**

In the present subsection we assume that all shaking parameters are identical, that is

$$d_1 = d_2 = \dots = d_N = d, \tag{44}$$

where

$$d + R_l < \frac{1}{2\sqrt{N}}. \tag{45}$$

We calculate  $\hat{\lambda}$  using (43) and (44). In order to calculate  $\mathbf{E}[\langle B_q \rangle]$  we use (32) and (27)

$$\mathbf{E}[\langle B_q \rangle] = \sum_{p_1, p_2, \dots, p_M; k_0, k_1, \dots, k_M} \beta_{q, p_1, p_2, \dots, p_M} \mathbf{E} \left[ \frac{v_{k_0} v_{k_1}^{p_1-1} \dots v_{k_M}^{p_M-1}}{K_1(\chi)^P} \right] \langle Z_{p_1, p_2, \dots, p_M}^{k_0, k_1, \dots, k_M} \rangle, \tag{46}$$

taking into account the following properties: (i)  $\mathbf{E}[\langle \beta_{q, p_1, p_2, \dots, p_M} \rangle] = \beta_{q, p_1, p_2, \dots, p_M}$ , since  $\beta_{q, p_1, p_2, \dots, p_M}$  does not depend on  $v_k$  and  $a_k$ ; (ii) the values  $Z_{p_1, p_2, \dots, p_M}^{k_0, k_1, \dots, k_M}$  do not depend on the random values  $v_k$ ,  $k = 1, 2, \dots, N$ , hence  $\mathbf{E}[Z_{p_1, p_2, \dots, p_M}^{k_0, k_1, \dots, k_M}] = Z_{p_1, p_2, \dots, p_M}^{k_0, k_1, \dots, k_M}$  and therefore  $\mathbf{E}[\langle Z_{p_1, p_2, \dots, p_M}^{k_0, k_1, \dots, k_M} \rangle] = \langle Z_{p_1, p_2, \dots, p_M}^{k_0, k_1, \dots, k_M} \rangle$ ; (iii)  $\langle v_{k_0} v_{k_1}^{p_1-1} \dots v_{k_M}^{p_M-1} \rangle = v_{k_0} v_{k_1}^{p_1-1} \dots v_{k_M}^{p_M-1}$ , since the random variables  $v_k$  do not depend on  $a_k$ .

Recall that for the monodispersed case we have<sup>(4)</sup>

$$\hat{\lambda}_{\text{mono}} = \langle \hat{\lambda} \rangle = 1 + 2\rho v \left( 1 + \langle A_1 \rangle v + \langle A_2 \rangle v^2 + \dots \right), \tag{47}$$

where

$$\langle A_q \rangle = \sum_{p_1, p_2, \dots, p_M; k_0, k_1, \dots, k_M} \beta_{q, p_1, p_2, \dots, p_M} \langle Z_{p_1, p_2, \dots, p_M}^{k_0, k_1, \dots, k_M} \rangle \tag{48}$$

and  $0 < v < v_{\text{max}}$ ,  $v_{\text{max}} = \pi/4$  corresponds to touching inclusions. In order to compare  $\hat{\lambda}_{\text{mono}}$  and  $\hat{\lambda}_{\text{poly}}$  defined by (43) and (47), respectively, we fix

the values of the concentration  $v$  and the shaking parameter  $d$  in both formulas. Then it is sufficient to compare corresponding coefficients given by (46) and (48) which differ only by the factor

$$w_{p_1, p_2, \dots, p_M} := \frac{\mathbf{E} \left[ v_{k_0} v_{k_1}^{p_1-1} \dots v_{k_M}^{p_M-1} \right]}{K_1(\chi)^P}. \tag{49}$$

For simplicity we first compute (49) assuming that all subscripts  $k_0, k_1, \dots, k_M$  in (49) are different, then the corresponding random variables  $v_{k_0}, v_{k_1}^{p_1-1}, \dots, v_{k_M}^{p_M-1}$  are also different and therefore independent. Hence, (49) implies

$$w_{p_1, p_2, \dots, p_M} = \frac{K_{p_1-1}(\chi) K_{p_2-1}(\chi) \dots K_{p_M-1}(\chi)}{K_1^{p_1-1}(\chi) K_1^{p_2-1}(\chi) \dots K_1^{p_M-1}(\chi)}, \tag{50}$$

where the following moments are introduced (see (5))

$$\mathbf{E}[v_k^q] = K_q(\chi) = \chi^q p + 1 - p, \quad q = 1, 2, \dots \tag{51}$$

If in (49)  $k_i = k_j$  for some  $i \neq j$ , we also arrive at an analogous formula (49) which requires more cumbersome notations.

We now consider the function

$$f(\chi) := \frac{K_j(\chi)}{[K_1(\chi)]^j} = \frac{\chi^j p + 1 - p}{(\chi p + 1 - p)^j}, \quad j = 1, 2, \dots \tag{52}$$

From (5) it follows that  $\chi \in [0, 1]$  and therefore  $f(\chi)$  decreases on the interval  $[0, 1]$  from  $(1 - p)^{-j+1}$  to 1. Since  $w_{p_1, p_2, \dots, p_M}$  is obtained by multiplication of the factors of the form (52), we have

$$w_{p_1, p_2, \dots, p_M} \geq 1. \tag{53}$$

Moreover, equality in (53) is attained in two cases (i)  $\chi = 1$  (when  $p = 0$ ) that is no polydispersity present; (ii)  $p_1 = p_2 = \dots = p_M = 2$  (see (50)). It follows from inequality (53) that  $\mathbf{E}[\langle B_q \rangle] \geq \langle A_q \rangle$ . This yields the following

**Theorem 4.5.** Let  $\hat{\lambda}_{\text{mono}}$  defined by (47), be the effective conductivity of a macroscopically isotropic monodispersed composite and  $\hat{\lambda}_{\text{poly}}$



defined by (42) (see also (43)), be the effective conductivity of a macroscopically isotropic polydispersed composite. Then for any fixed  $d$ , satisfying the non-overlapping condition (34), the following inequality holds

$$\hat{\lambda}_{\text{mono}} < \hat{\lambda}_{\text{poly}} \tag{54}$$

for any relative concentration  $p$  of the small inclusions and for any fixed total volume fraction  $v$ .

**Remark 4.6.** Theorem 4.5 was proved for a fixed value of the shaking parameter  $d$ . However, it can be easily generalized to the case when the shaking parameters  $d_k$ ,  $k = 1, 2, \dots, N$ , are i. i. d.'s in the interval  $[d_{\min}, d_{\max}]$ . This is the most general case of identical shaking, since  $d_k$  do not depend on radii of the disks unlike in the bumping model (see the next section). This model describes the physical situation when inclusions are well-separated (not too close to touching) and, in particular, it applies to the dilute case.

#### 4.2. Bumping Model (Two Different Shaking Parameters)

In this section we introduce a model which describes highly packed composites with inclusions of two different sizes. In such composites inclusions are often not well-separated and percolation patterns dominate the effective behavior. That is why no approach based on the Maxwell–Garnett formula (and its refinements) will work here.

We now present a heuristic motivation for our model. In high concentration regime where inclusions are close to each other and there is not much room for them to move around, a large inclusion would bump into another one after relatively small change in the location. However, a small inclusion is likely to have more room to move around before it bumps into a neighbouring inclusion. Thus, while in a low concentration (more generally well-separated) composite it is reasonable to introduce identical shaking parameters, as we did it in the previous section, for highly packed random composites it is natural to introduce two different shaking parameters  $d_s, d_l$  for small and large inclusions, respectively, such that

$$d_l < d < d_s, \tag{55}$$

where  $d$  is the shaking parameter for the monodispersed model introduced in the beginning of the section (at the same total volume fraction). In other words the inequality (55) models the geometric observation

that small inclusions are more mobile than the large ones. Note that the validity of this model for highly packed composites is supported by the percolation analysis of a random checkerboard with squares of two different sizes (L. Berlyand, A. Pisztor, unpublished). As we will see below our model leads to  $S$ -shape dependence of the effective conductivity on the polydispersity parameter (for a fixed total volume fraction), that is (1) holds for  $p$  close to one, (3) holds for small  $p$  (see Figure 3). Also (3) agrees with the numerical example from ref. 5, in which the decrease in the effective conductivity was observed for a mixture of large and small particles when  $p$  is small.

In L. Berlyand, A. Pisztor (unpublished) geometric patterns of small and large inclusions embedded in a matrix are modeled. No particular physical properties were considered and the connectivity of such patterns was the main objective. For identical inclusions randomly embedded in a matrix, random checkerboard models of black (inclusions) and white (matrix) squares were successfully used in the study of both the connectivity patterns and physical properties such as conductivity, viscosity, elastic modulus etc. (see refs. 6, 9 and 12)

It was proposed in L. Berlyand, A. Pisztor (unpublished) to use a random checkerboard with black squares of two different sizes to model a polydispersed medium. It was shown that the density of the infinite cluster of black squares can either increase or decrease depending on the relative

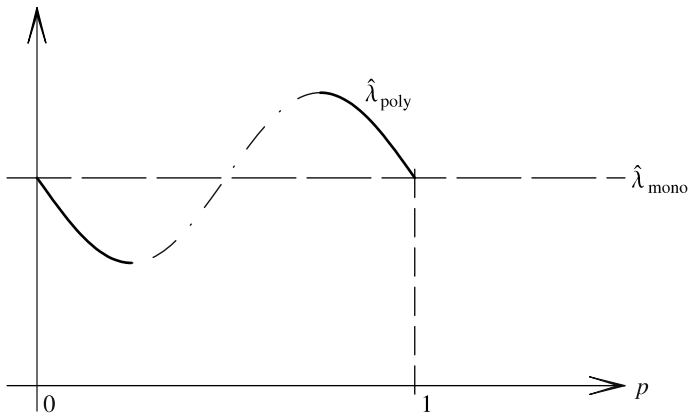


Fig. 3. Dependence of the effective conductivity  $\hat{\lambda}_{\text{mono}}$  on the relative volume fraction of the small inclusions  $p$  for monodispersed composites, broken line; and for polydispersed composites  $\hat{\lambda}_{\text{poly}}$ , solid and dot-dashed line, in the case of high concentrations of inclusions. Solid line is rigorously justified, dot-dashed is supported by numerical results,  $\chi = (R_s/R_l)^2$  is fixed.

volume fraction of small and large black squares (total volume fraction of all black squares is fixed, when it is above the percolation threshold). This conclusion supports our main result, Theorem 4.7.

In order to compare  $\hat{\lambda}_{\text{mono}}$  and  $\hat{\lambda}_{\text{poly}}$  we compare the expectation  $\mathbf{E}[\cdot]$  of the corresponding coefficients (32) and (37). Recall that  $\mathbf{E}[\cdot]$  corresponds to subsequent averaging over random locations of the centers  $a_k$  (shaking parameters) followed by averaging over random sizes  $v_k$  of the disks  $D_k$ .

We first calculate  $\langle B_q \rangle$ . This amounts to replacement of  $d^{2(s_0+\dots+s_M)}$  in (40) by

$$\frac{v_{k_0} v_{k_1}^{p_1-1} \dots v_{k_M}^{p_M-1}}{K_1(\chi)^P} d_{k_0} d_{k_1}^{p_1-1} \dots d_{k_M}^{p_M-1}, \quad k_j = 1, 2, \dots, N; \quad M = 1, 2, 3, \dots \tag{56}$$

Next, we take the expectation  $\mathbf{E}[\cdot]$  in (56)

$$\mathbf{E} \left[ \frac{v_{k_0} v_{k_1}^{p_1-1} \dots v_{k_M}^{p_M-1}}{K_1(\chi)^P} d_{k_0} d_{k_1}^{p_1-1} \dots d_{k_M}^{p_M-1} \right]. \tag{57}$$

Since each index  $k_0, k_1, \dots, k_M$  takes one of the values  $1, 2, \dots, N$  and  $M$  is any natural number, there may be repeated indices among  $k_0, k_1, \dots, k_M$ . We select the repeated indices and using the independence of the random variables  $v_k$  ( $d_k(v_k)$ ) decompose (57) into a product of terms in the following form

$$\mathbf{E} \left[ \frac{v_k^j}{K_1^j(\chi)} d_k^j \right] = \frac{p\chi^j d_s^j + (1-p)d_l^j}{(p\chi + (1-p))^j}. \tag{58}$$

Therefore, comparison of  $\hat{\lambda}_{\text{poly}}$  and  $\hat{\lambda}_{\text{mono}}$  amounts to comparison of the terms

$$\omega(d_s, d_l) = \frac{p\chi^j d_s^j + (1-p)d_l^j}{(p\chi + (1-p))^j} \quad \text{and} \quad \omega(d, d) = d^j. \tag{59}$$

We do this for two limit cases: for sufficiently small  $p$  ( $p \sim 0$ , i.e., almost all disks are large) and for  $p$  closed to 1 ( $p \sim 1$ , i.e., almost all disks are small) for fixed  $\chi$ . Due to (55) we obtain

$$\omega(d_s, d_l) = d_l^j < d^j \text{ as } p \sim 0; \quad \omega(d_s, d_l) = d_s^j > d^j \text{ as } p \sim 1. \tag{60}$$

In order to compare  $\hat{\lambda}_{\text{poly}}$  and  $\hat{\lambda}_{\text{mono}}$  for the same total concentration  $v$ , we first fix all parameters corresponding to the monodispersed composite, i.e., either the radius  $R$  of the disks, or the number of the subcells (disks) per unit cell,  $N$ . These values depend on each other through the relation  $v = N\pi R^2$ . For the polydisperse composite we use the same  $v$  and  $N$  and choose the relative volume fraction  $p$  ( $0 \leq p \leq 1$ ) and the parameter  $\chi = (R_s/R_l)^2$ . The radii  $R_s$  and  $R_l$  are then determined by solving the system of Eqs. (5) and (6).

Secondly, we chose the following shaking parameters

$$d = \frac{1}{2\sqrt{N}} - R, \quad (61)$$

$$d_s = \frac{1}{2\sqrt{N}} - R_s, \quad d_l = \frac{1}{2\sqrt{N}} - R_l. \quad (62)$$

These relations mean that we take the maximum possible shaking parameters within the framework of the non-overlapping model. Our parameters are consistent in the following sense. If  $p$  goes to 0 or 1, or if  $\chi \rightarrow 1$ , then a polydispersed composite becomes a monodispersed one.

In Theorem 4.7 below, we compare  $\hat{\lambda}_{\text{poly}}$  with the constant  $\hat{\lambda}_{\text{mono}}$  for any  $p$  and  $\chi$ . This theorem follows from the comparison of (59) and (60) established above. Fig. 3 illustrates this comparison for a fixed  $\chi$  and shaking parameters (61) and (62).

**Theorem 4.7.** Let  $\hat{\lambda}_{\text{mono}}$  defined by (47), be the effective conductivity of a macroscopically isotropic monodispersed composite and  $\hat{\lambda}_{\text{poly}}$  defined by (42), be the effective conductivity of a macroscopically isotropic polydispersed composite (see also (43)). Then for a fixed  $v$  (same total volume fraction for mono- and poly-dispersed composites) and any shaking parameters  $d_s, d_l, d$  satisfying the conditions (34) and (55), the following inequalities hold

$$\hat{\lambda}_{\text{poly}} < \hat{\lambda}_{\text{mono}} \quad \text{for } p \sim 0; \quad \hat{\lambda}_{\text{poly}} > \hat{\lambda}_{\text{mono}} \quad \text{for } p \sim 1. \quad (63)$$

## 5. DISCUSSION

We prove that  $\hat{\lambda}_{\text{poly}}$  has different behavior depending on the conditions on the mobility of inclusions (55) and (44). Condition (55) arises from natural restrictions that at high concentrations disks bump into each

other, since they are hard and cannot penetrate. This condition implies behavior displayed in Fig. 3 which also agrees with percolation analysis of checkerboard models with squares of two different sizes (L. Berlyand, A. Pisztor, unpublished) and numerical example.<sup>(5)</sup>

For low concentration regime it is natural that the probability of bumping is close to zero. Therefore, the shaking parameters should be equal as in (44). This leads to the behavior described in Fig. 4 and agrees with rigorous asymptotical analysis for small concentration<sup>(29-31)</sup>.

The same curve (see Fig. 4) was obtained by Robinson and Friedman<sup>(28)</sup> using a clever two step application of the Maxwell–Garnett formula. However, their analysis<sup>(28)</sup> tacitly uses the assumption of small concentration, because the Maxwell–Garnett formula is derived for small concentration only. In particular, no use of this formula is capable of capturing percolation effects. Note that for concentrations above the precolation threshold  $v_{cr}$  connectivity patterns (percolation patterns) dominate behavior of the effective conductivity while for  $v < v_{cr}$  the percolation effects are non-essential and behavior presented in Fig. 4, though rigorously justified only for small concentrations  $v \ll 1$ , is expected to be valid for  $v < v_{cr}$ .

We remark that the shaking model has a restriction that each disk is confined in its own subcell which is why the number of disks in the basic cell  $Q_{(0,0)}$  is always the square of an integer. This restriction is introduced in order to obtain an analytical solution (series in which all coefficients are evaluated recursively) which is only possible for very few specific models. A prominent example of a model which can be treated analytically is

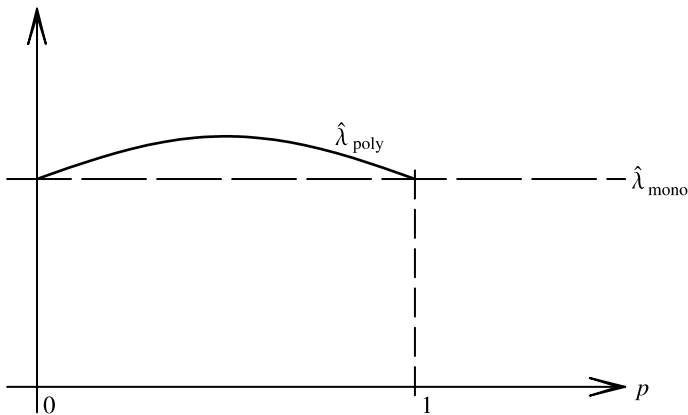


Fig. 4. Dependence of the effective conductivity  $\hat{\lambda}$  on the relative volume fraction of the small inclusions  $p$  for monodispersed composites (broken line) and for polydispersed composites (solid line) in dilute case.

the Keller-Dykhne random checkerboard<sup>(8,17)</sup> where effective conductivity is given by a simple formula. It is known that qualitative behavior of the effective properties predicted by such restricted models is usually valid for much wider class of physical problems.

**APPENDIX A**

In the present section we outline the main idea of reduction of the conjugation condition (13) to the functional Eq. (14). For simplicity we consider one inclusion  $D_1 := \{|z| < r\}$  in the unit cell. Then (13) becomes

$$\psi(t) = \psi_1(t) + \rho \left(\frac{r}{t}\right)^2 \overline{\psi_1(t)} - 1, \quad t \in \partial D_1 = L. \tag{A.1}$$

Introduce the operator

$$(Pf)(z) = \frac{1}{2\pi i} \int_L f(t) E_1(t-z) dt = \frac{1}{2\pi i} \int_L f(t) \sum_{m_1, m_2} \frac{dt}{(t - m_1 - im_2 - z)}, \tag{A.2}$$

where  $f$  is a Hölder continuous function on  $L$ ,  $z \in D_1$ ,  $E_1$  is the Eisenstein function described in Appendix B. We use the Cauchy formula

$$\frac{1}{2\pi i} \int_L \frac{f(t)}{t-z} dt = \begin{cases} f(z), & z \in D_1, \\ 0, & z \in D_1^-, \end{cases} \tag{A.3}$$

where  $f$  is analytic in  $D_1$  and  $D_1^-$  is the complement of  $D_1$  to the extended complex plane. If  $f$  is analytic in  $D_1^-$ , we have<sup>(10)</sup>

$$\frac{1}{2\pi i} \int_L \frac{f(t)}{t-z} dt = \begin{cases} f(\infty), & z \in D_1, \\ -f(z) + f(\infty), & z \in D_1^-. \end{cases} \tag{A.4}$$

We apply the operator  $P$  to LHS of (A.1).

$$\begin{aligned} P\psi(z) &= \frac{1}{2\pi i} \int_L \sum_{m_1, m_2} \frac{\psi(t)}{t - m_1 - im_2 - z} dt \\ &= \frac{1}{2\pi i} \sum_{m_1, m_2} \int_{L+m_1+im_2} \frac{\psi(t)}{t-z} dt + c_0. \end{aligned} \tag{A.5}$$

Here we use double periodicity of  $\psi(z)$ . The latter integral is equal to a constant by (A.4), since it is calculated over the curve  $\sum_{m_1, m_2} (L + m_1 +$

$im_2$ ), out of which  $\psi(z)$  is analytic. It is possible to prove that  $c_0=0$  (for details see ref. 25).

Note that the operator (15) can be described as follows. The inversion transformation of the extended complex plane with respect to  $T_k = \partial D_k$  is defined in a standard way

$$z_{(k)}^* = \frac{r_k^2}{z - a_k} + a_k. \tag{A.6}$$

Then  $\overline{\psi(z_{(k)}^*)}$  is analytic outside of  $T_k$  (see ref. 1). The factor  $\left(\frac{r}{z-a_k}\right)^2$  in (15) for  $m_1=m_2=0$  appears because of the same factor in (13). The shifts by  $m_1 + im_2$  are used to obtain doubly periodic functions.

Next we apply  $P$  to the RHS of (A.1). First, observe that  $P(\psi_1 - 1)(z) = \psi_1(z) - 1$ . Further, applying  $P$  to  $(r/t)^2 \overline{\psi_1(t)}$  and using (A.3) and (A.4) we obtain

$$P \left[ \left(\frac{r}{t}\right)^2 \overline{\psi_1\left(\frac{r^2}{t}\right)} \right] (z) = \frac{1}{2\pi i} \int_L \left(\frac{r}{t}\right)^2 \overline{\psi_1\left(\frac{r^2}{t}\right)} \frac{dt}{t-z} + \sum'_{m_1, m_2} \frac{1}{2\pi i} \int_L \left(\frac{r}{t}\right)^2 \overline{\psi_1\left(\frac{r^2}{t}\right)} \frac{dt}{t - m_1 - im_2 - z}, \tag{A.7}$$

where  $\sum'_{m_1, m_2}$  means summation over all integers  $m_1, m_2$  except  $m_1 = m_2 = 0$ . Taking into account (A.3) and (A.4) we observe that (A.7) implies

$$P \left[ \left(\frac{r}{t}\right)^2 \overline{\psi_1\left(\frac{r^2}{t}\right)} \right] (z) = \sum'_{m_1, m_2} \left(\frac{r}{t - m_1 - im_2}\right)^2 \overline{\psi_1\left(\frac{r^2}{t - m_1 - im_2}\right)}, \tag{A.8}$$

Here we took into account that  $\left(\frac{r}{t}\right)^2 \overline{\psi_1\left(\frac{r^2}{t}\right)}$  is analytic in  $D_1^-$  and that  $\left(\frac{r}{t - m_1 - im_2}\right)^2 \overline{\psi_1\left(\frac{r^2}{t - m_1 - im_2}\right)}$  is analytic in  $D_1$  for  $(m_1, m_2) \neq (0, 0)$ . The relations (A.7) and (A.8) yield the functional Eq. (14) for  $N = 1$ .

**Remark 1.1.** We derive functional equations (14) ( $N = 1$ ) from (A.1) formally. The interchanging of the integral and the sum  $\sum_{m_1, m_2}$  can be justified using properties of the Eisenstein function  $E_1(z)$ . Generalization to the case  $N > 1$  is straightforward.

**APPENDIX B. EISENSTEIN'S SERIES**

In the present and previous papers we exploit the theory of elliptic functions. This theory can be viewed as a generalization of the meromorphic functions on the complex plane to the meromorphic functions on the torus (doubly periodic meromorphic functions).

Any function analytic on the extended complex plane  $\mathbb{C} \cup \{\infty\}$  except  $z=0$  can be expanded in the Laurent series

$$\phi(z) = \sum_{n=0}^{\infty} \phi_n z^{-n}. \tag{B.1}$$

In order to obtain a periodic analog of (B.1) one has to replace  $z^{-n}$  by appropriate basic functions periodic with respect to lattice  $\mathcal{Q}$ . The periodic analog of  $z^{-n}$  is given by the series first introduced by Eisenstein<sup>(32)</sup>

$$E_n(z) := \sum_{m_1, m_2} (z - m_1 - im_2)^{-n}. \tag{B.2}$$

Here,  $m_1$  and  $m_2$  run over all integer numbers. The series (B.2) converges almost uniformly and absolutely for  $n \geq 3$ . For  $n=1$  and  $n=2$  the Eisenstein method of summation should be used:

$$E_n(z) = \lim_{M_2 \rightarrow \infty} \sum_{m_2=-M_2}^{M_2} \lim_{M_1 \rightarrow \infty} \sum_{m_1=-M_1}^{M_1} (z - m_1 - im_2)^{-n}. \tag{B.3}$$

We use Eisenstein series instead of the Weierstrass functions because they provide a periodic analog of Laurent series and also enable us to use convenient computational formulas (see (B.10)) for  $E_n(z)$ . It is possible to use the Weierstrass functions  $\zeta$ ,  $\mathcal{P}$  and their derivatives, but we prefer to deal a periodic form of Laurent series

$$\phi(z) = \sum_{n=1}^{\infty} \phi_n E_n(z) + \phi_0. \tag{B.4}$$

Any function  $\phi$  analytic in a multiply connected domain  $D = \mathbb{C} \cup \{\infty\} \setminus \bigcup_{k=1}^N (D_k \cup \partial D_k)$  is represented in the form<sup>(26)</sup>

$$\phi(z) = \sum_{k=1}^N [\overline{\phi_k(z_{(k)}^*)} + A_k \ln(z - a_k)], \tag{B.5}$$



where  $A_k$  are real constants,  $\phi_k(z)$  is analytic in the disk  $D_k$   $z_{(k)}^*$  stands for the inversion (A.6). Since  $\overline{\phi_k(z_{(k)}^*)}$  is analytic out of  $D_k$  (see Appendix A), the series (B.1) for this function can be used.

Any function  $\phi$  analytic in a multiply connected domain  $D$  on the torus  $\mathcal{Q}$  is represented in the form<sup>(24)</sup>

$$\phi(z) = \sum_{k=1}^N [\phi_k(z) + A_k(\ln \sigma(z - a_k) + a_k \zeta(z - a_k))], \tag{B.6}$$

where  $\phi_k(z)$  are analytic out of  $D_k$  and doubly periodic;  $\sigma$  and  $\zeta$  are the Weierstrass functions.

On the plane we use the series (B.1) for functions  $\phi$  in (B.5). On the torus we use (B.4) for functions  $\phi$  in (B.6), which is why we deal with series involving the Eisenstein functions in the main body of this paper. Note that the effective conductivity is determined by the derivatives of the complex potential (B.6) and  $(\ln \sigma(z))' = \zeta(z) = E_1(z) + S_2 z$ ,  $-\zeta'(z) = \mathcal{P}(z) = E_2(z) - S_2$  (see refs. 18 and 32, where  $S_2 = \pi$  is established for the square lattice).

In the formula (19) for a disk  $D_m$  ( $m = 1, 2, \dots, N$ ) and later in the paper we introduce the function

$$E_l^{(m)}(z - a_k) = \begin{cases} E_l(z - a_k) & \text{if } k \neq m, \\ E_l(z - a_m) - (z - a_m)^{-l} & \text{if } k = m, \end{cases} \tag{B.7}$$

where  $l = 2, 3, \dots$ . For simplicity we write  $E_l$  instead of  $E_l^{(m)}$ . In ref. 4 we use another symbol  $\sigma_l(z - a_m)$  for  $E_l(z - a_m) - (z - a_m)^{-l}$ . The use of designation (B.7) simplifies the final formula for the effective conductivity. For instance, (B.7) implies

$$E_l(0) := S_l, \quad l = 2, 3, \dots, \tag{B.8}$$

where

$$S_l = \sum'_{m_1, m_2} (m_1 + im_2)^{-l} \tag{B.9}$$

are the Eisenstein–Rayleigh lattice sums.  $\sum_{m_1, m_2}$  in (B.9) stands for summation over all integer numbers  $m_1, m_2$  except  $m_1 = m_2 = 0$ . The sums (B.9) converge slowly. Efficient fast formulas for  $S_l$  are proposed in ref. 23.

Note that for the square array  $S_2 = \pi, S_4 = 3.15121, S_8 = 3.93885, S_{4l+1} = S_{4l+2} = S_{4l+3} = 0$  ( $l = 1, 2, \dots$ );  $S_l$  are non-negative numbers<sup>(19,20)</sup>.

Sums (B.7) can be represented as follows<sup>(32)</sup>

$$E_l(z - a_k) = \begin{cases} \frac{1}{(z - a_k)^l} + (-1)^l \sum_{s=0}^{\infty} C_{l+s-1}^s S_{l+s} (z - a_k)^s & \text{if } k \neq m, \\ (-1)^l \sum_{s=0}^{\infty} C_{l+s-1}^s S_{l+s} (z - a_k)^s & \text{if } k = m, \end{cases} \tag{B.10}$$

where  $C_q^s = \frac{q!}{s!(q-s)!}$ .

### APPENDIX C. AN INEQUALITY FOR COEFFICIENTS

In order to complete the proof of Theorem 4.1 we have to prove the inequality (39) for  $s = 0$ . To establish this inequality it is sufficient to show that all corresponding terms in each approximation of  $\psi_m(0)$  are non-negative. Note, that  $s = 0$  means that all shaking parameters are zeros, that is  $a_k$  form a periodic square lattice. Then  $N = 1$  and (20) becomes

$$\psi^{(0)}(z) = 1, \quad \psi^{(q+1)}(z) = \rho \left[ \overline{\psi_0^{(q)}} E_2(z) + \overline{\psi_1^{(q-1)}} E_3(z) + \dots + \overline{\psi_q^{(0)}} E_{q+2}(z) \right], \tag{C.1}$$

$q = 0, 1, 2, \dots$

Here we omit subscript  $k$  in  $\psi(z)$  and assume the inclusion is centered at zero,  $a_1 = 0$ . Substitute  $z = 0$  into (C.1). Using (B.8) we have

$$\psi_0^{(q+1)} = \psi^{(q+1)}(0) = \rho \left[ \overline{\psi_0^{(q)}} S_2 + \overline{\psi_1^{(q-2)}} S_4 + \dots + \overline{\psi_q^{(0)}} S_{q+2} \right]. \tag{C.2}$$

Calculate the derivatives

$$\psi_l^{(q+1)} = \frac{1}{l!} \frac{d^l \psi^{(q+1)}}{dz^l}(0) = \rho (-1)^l \left[ \overline{\psi_0^{(q)}} \frac{(l+1)!}{2!} S_{l+2} + \overline{\psi_1^{(q-1)}} \frac{(l+2)!}{3!} \times S_{l+3} + \dots + \overline{\psi_q^{(0)}} \frac{(q+l+1)!}{(q+1)!} S_{l+q+2} \right]. \tag{C.3}$$

Here we use the relation for the derivatives of the Eisenstein series<sup>(32)</sup>

$$E_q^{(l)}(z) = (-1)^l q(q+1) \cdot (q+l-1) E_q(z). \tag{C.4}$$

It follows from (C.2) and (C.3) that  $\psi_0^{(q)}$  has the following form

$$\psi_0^{(q)} = \pi^q N^{q+1} \sum_{p_1, p_2, \dots, p_M} \beta_{q, p_1, p_2, \dots, p_M} Z_{p_1, p_2, \dots, p_M}, \tag{C.5}$$

where

$$Z_{p_1, p_2, \dots, p_M} = S_{p_1} S_{p_2} \cdots S_{p_M} \geq 0. \tag{C.6}$$

Therefore,  $\psi_0^{(q)}$  is a sum of the products (C.6) with the coefficients  $\pi^q N^{q+1} \beta_{q, p_1, p_2, \dots, p_M}$ . We have to prove that

$$\beta_{q, p_1, p_2, \dots, p_M} \geq 0 \tag{C.7}$$

for all  $q, p_1, p_2, \dots, p_M$ .

We now explain that (C.7) implies (39) for  $s = 0$ . First, we compare the terms in (C.5), (C.6) written in the case  $s = 0$  ( $N = 1$ ) with the general terms discussed in Sections 3 and 4. In this case  $Z_{p_1, p_2, \dots, p_M}^{k_0, k_1, \dots, k_M}$  from (30) becomes (indices  $k_0, k_1, \dots, k_M$  are omitted)

$$Z_{p_1, p_2, \dots, p_M} = E_{p_1}(0) E_{p_2}(0) \cdots E_{p_M}(0)$$

that gives (C.6) (see also (B.8)). Then (30) yields

$$X_{p_1, p_2, \dots, p_M} = Z_{p_1, p_2, \dots, p_M}.$$

The representation (32) and the relation (29) for  $N = 1$  imply (C.5). The term (39) for  $s = 0$  can be considered as (38) with  $d = 0$ . Then we obtain

$$Z_{p_1, p_2, \dots, p_M} = \gamma_{p_1, p_2, \dots, p_M}(0).$$

It follows from (C.6) that  $\gamma_{p_1, p_2, \dots, p_M}$  are non-negative for all  $p_1, p_2, \dots, p_M$  and the required inequality (39) for  $s = 0$  is reduced to (C.7).

We now prove (C.7) by induction in  $q$ . Although we only need  $\psi^{(q)}(0) \geq 0$  it is convenient to show

$$\frac{d^l \psi^{(q)}}{dz^l}(0) \geq 0, \quad l = 0, 2, 4, \dots \tag{C.8}$$

Since  $\psi^{(0)}(z) = 1$  and  $\psi^{(1)}(z) = \rho E_2(z) = \sum_{m=0}^{\infty} (m+1)! S_{m+2} z^m$ , (C.8) holds for  $q=0$  and  $q=1$ . Assume now that  $q$  is even and all coefficients in  $\frac{d^l \psi^{(q)}}{dz^l}(0)$  are non-negative ( $l=0, 2, 4, \dots$ ).  $\psi^{(q+1)}(0)$  is given by (C.2). Combining this with the basis of the induction we see that all coefficients of  $\psi^{(q+1)}(0)$  are non-negative. It also follows from (C.3) that all coefficients of  $\frac{d^l \psi^{(q+1)}}{dz^l}(0)$  are non-negative ( $l=0, 2, 4, \dots$ ).

The proof of Theorem 4.1 is completed.

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## REFERENCES

1. M. Ablowitz and A. Fokas, *Complex Variables* (Cambridge University Press, 1997).
2. V. L. Berdichevskij, *Variational Principles of Continuum Mechanics* (Moscow, Nauka, 1983).
3. D. J. Bergman and K. J. Dunn, *Phys. Rev. Ser. B* **45**:13262–13271 (1992).
4. L. Berlyand and V. Mityushev, *J. Stat. Phys.* **102**(N1/2):115–145 (2001).
5. L. Berlyand and A. Kolpakov, *Arch. Rational Mech. Anal.* **159**:179–227 (2001).
6. L. Berlyand and K. Golden, *Phys. Rev. B* **50**:2114–2117 (1994).
7. Ch. Chang, R. L. Powell, *J. Rheol* **38**:85–98 (1994).
8. A. Dykhne, *JEPT* **32**:63–65 (1971).
9. V. V. Jikov, S. M. Kozlov, and O. A. Olejnik, *Homogenization of Differential Operators and Integral Functionals* (Berlin etc., Springer-Verlag, 1994).
10. F. D. Gakhov, *Boundary Value Problems* (Pergamon Press, Oxford, 1964).
11. H. Goto and H. Kuno, *J. Rheol* **28**:197–205 (1984).
12. A. Fannjiang and G. Papanicolaou, *J. Stat. Phys.* **88**:1033–1076 (1997).
13. G. M. Golusin, *Matem. Sbornik* **42**:191–198 (1935).
14. R. F. Hill, in *Proc. of Technical Program of SMTA National Symposium, "Emerging Packing Technologies."* Research Triangle Park, North Carolina, Nov. 18–21.
15. R. F. Hill and P. H. Supancic, *J. Am. Cer. Soc.* **85**:851–857 (2002).
16. D. J. Jeffrey and A. Acrivos, *AIChE J.* **22**:417–432 (1976).
17. J. B. Keller, *J. Math. Phys.* **5**:548–549 (1964).
18. Lord Rayleigh, *Phil. Mag.* **34**:481–502 (1892).
19. R. C. McPhedran and D. R. McKenzie, *Proc. Roy. Soc. Lond. Ser. A* **359**:45–52 (1978).
20. R. C. McPhedran, *Proc. Roy. Soc. Lond. Ser. A* **408**:31–43 (1986).
21. G. W. Milton, *The Theory of Composites* (Cambridge University Press, 2002).
22. V. Mityushev, *Demonstratio Math.* **30**:63–70 (1997).
23. V. Mityushev and P. M. Adler, *ZAMM* **82**:335–345 (2002).
24. V. Mityushev and P. M. Adler, *ZAMP* **53**:486–517 (2002).
25. V. Mityushev, *Appl. Math. Optim.* **44**:17–31 (2001).

26. V. Mityushev and S. V. Rogosin, *Constructive Methods for Linear and Nonlinear Boundary Value Problems of the Analytic Functions Theory* (Chapman&Hall/CRC, Boca Raton, 2000).
27. A. J. Poslinski, M. E. Ryan, R. K. Gupta, S. G. Seshadri, and F. J. Frechette, *J. Rheology* **32**:751–771 (1988).
28. D. A. Robinson and S. F. Friedman, *Water Res. Research* **37**(N 1):33–40 (2001).
29. S. Torquato, *Random Heterogeneous Materials*, (Springer, NY etc., 2002).
30. J. F. Thovert and A. Acrivos, *Chem. Eng. Comm.* **82**:177–191 (1989).
31. J. F. Thovert, I. C. Kim, S. Torquato, and A. Acrivos, *J. Appl. Phys.* **67**:6088–6098 (1990).
32. A. Weil, *Elliptic Functions According to Eisenstein and Kronecker* (Springer-Verlag, Berlin etc., 1976).