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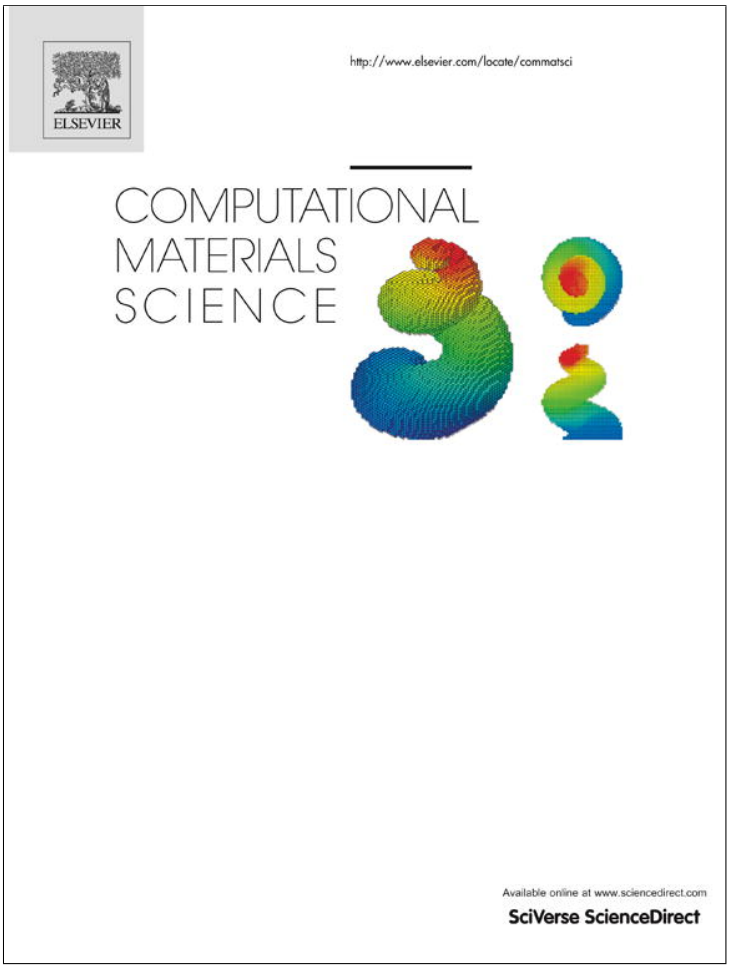

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Effective conductivity of random two-dimensional composites with circular non-overlapping inclusions

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ABSTRACT

We study the effective conductivity of equal unidirectional infinite circular cylinders randomly distributed in a uniform host (disks on the plane). The problem is reduced to a boundary value problem for the two-dimensional Laplace equation. A symbolic–numerical algorithm was proposed in the previous papers to solve the boundary value problem with arbitrary deterministic locations of disks. Application of the Monte Carlo method for the uniform non-overlapping distribution of disks yields the effective conductivity of random composites. The expected value of the effective conductivity is written exactly in the form of a power series in the concentration. This formula is valid for all concentrations.

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1. Introduction

An important area of materials science is the study of effective dielectric, thermal and electrical properties of two phase composites. The conductivity of two-dimensional arrays of disks regularly distributed in the periodicity cell is sufficiently well studied. Rayleigh [26] reduced the problem to an infinite system of linear algebraic equations. McPhedran et al. [15,16] extended Rayleigh's method and obtained approximate formulae for the effective conductivity $\hat{\lambda}$ of the square and hexagonal arrays of disks. Mityushev [18,19,25] obtained an exact formula for the effective conductivity of the square array of disks in the form of power series in Bergman's contrast parameter ρ. One can find other numerical and analytical results devoted to regular arrays in [27] and paper cited therein. A constructive algorithm and approximate analytical formulae for non-overlapping disks were obtained by Berlyand and Mityushev, [8] Mityushev [20,21] and Szczepkowski et al. [28].

Many composites can be considered as materials when inclusions are randomly distributed in the host. Application of the probabilistic methods begun from [5] and were proceeded by many authors (see references in [6,7,17,29]). The homogenization theory of random composites were developed by Golden and Papanicolaou [10] and by Jikov et al. [12]. Constructive results including exact and approximate formulae for the effective conductivity are systematically described by Torquato [29]. This fundamental book contains perhaps all essential results on random heterogeneous media obtained before 2002 year. The statistical analysis of complex three-dimensional fracture structures and advanced numerical tools to solve transport equations are presented by Adler and Thovert [3].

In the present paper, we discuss the effective conductivity of two-dimensional composites with randomly distributed non-overlapping equal disks. This is one of the central problem discussed by Torquato [29] by use of the correlation functions (see, for instance, the three-point approximation (20.83), the four-point bounds (21.41)–(21.46) in [29]). These exact formulae were based on contrast expansions of the effective conductivity, in particular, on the weak-contrast expansion (see (20.1) in [29]). Improved bounds for effective transport properties of random non-percolated composites were developed by means of the security-spheres approach in [2].

Andrianov et al. [1] deduced an approximate analytical formula for the effective conductivity by matching Garnett's formula and the percolation threshold approximation. The "shaking" model in which disks can move randomly inside the periodicity cell according certain uniform distribution was worked out in [8]. The review [30] describes the diversity of jammed configurations and their macroscopic properties. A theory and applications of the two-point correlation functions are presented in [11].

This paper is based on the method of $\mathbb{R}$-linear problem and functional equations applied to doubly periodic problems [23,22]. The same method can be applied to non-periodic problems [20]. However, it is easier to discuss formally periodic problems with arbitrarily distributed disks the number of which is also arbitrary (symbol N below, so one can substitute any number into N). This approach is justified theoretically for composites with invariant distributions of inclusions under translations [10,12]. Practically, we always have finite samples of composites in our disposal.

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The paper is organized as follows. The problem and the main result are presented in Section 2. Following [8,21,28] we describe a constructive algorithm to calculate the effective conductivity of composites with arbitrary located inclusions in Section 3. Section 4 is devoted to computation of the generalized Eisenstein–Rayleigh sums by the Monte Carlo method and to optimization of the simulation parameters. Analytical formulae for the effective conductivity are deduced in Section 5 and compared to the corresponding formulae obtained by Torquato [29]. Concluding remarks and relations to the theory of representative volume elements are presented in Section 6. Appendix contains formulae for Eisenstein's functions and the generalized Eisenstein–Rayleigh sums systematically used in the main text.

2. Random location of disks

Let ω_1 and ω_2 be the fundamental pair of periods on the complex plane $\mathbb{C}$ such that $Im\ \frac{\omega_2}{\omega_1} > 0$. The fundamental parallelogram Q is defined by its vertices $\pm\frac{\omega_1}{2}$ and $\pm\frac{\omega_2}{2}$. Without loss of generality the area of Q can be normalized to one. The points $m_1\omega_1 + m_2\omega_2$ $(m_1, m_2 \in \mathbb{Z})$ generates a doubly periodic lattice $\mathcal{Q}$. Here, $\mathbb{Z}$ stands for the set of integer numbers. Let i denote the imaginary unit. In the case $\omega_1 = \sqrt[4]{\frac{4}{3}}$ and $\omega_2 = \sqrt[4]{\frac{4}{3}}\, e^{\frac{\pi i}{3}}$, the cell Q becomes a rhombus with an angle 60° and the array $\mathcal{Q}$ is called the hexagonal lattice (the equilateral triangular lattice). Further, our numerical computations are performed only for the hexagonal lattice. This assumption does not restrict our investigation since the number of inclusions per cell can be taken arbitrary large, hence, the shape of the cell does not impact on the final result.

Consider N non-overlapping circular disks D_k of radius r with the centers $a_k \in Q$ (see Fig. 1). Let D_0 be the complement of all closure disks $|z - a_k| \leqslant r$ to the domain Q. We study conductivity of the doubly periodic composite when the host $\cup_{m_1,m_2}(D_0 + m_1\omega_1 + m_2\omega_2)$ and the inclusions $D_k + m_1\omega_1 + m_2\omega_2$ are occupied by materials of conductivities λ and λ_1, respectively $(m_1, m_2 \in \mathbb{Z})$. Without loss of generality the conductivity of the host can be normalized to unit, i.e., $\lambda = 1$. Introduce Bergman's contrast parameter [6,7]

$$\rho = \frac{\lambda - 1}{\lambda + 1}, \tag{1}$$

and the concentration of the inclusions $v = N\pi r^2$.

The centers a_k are considered as random variables distributed in such a way that the disks $D_k = \{z \in \mathbb{C} : |z - a_k| < r\}$ generate a set of uniformly distributed non-overlapping disks. Theoretically this distribution can be introduced as the distribution of the variable $\mathbf{a} = (a_1, a_2, \cdots, a_N) \in Q^N$ with the restrictions $|a_m - a_k| > 2r$ for $m \neq k$ $(m, k = 1,2,\ldots,N)$. More formally, consider the probabilistic space (Ω, B, P) where the set of events Ω consists of the locations $\mathbf{a}$ in Q^N, B is the Borel set of all subset of Ω. Let ω be a measurable subset of B. Then $P(\omega)$ is introduced as the volume of ω in $\mathbb{C}^N$. The random variable $\mathbf{a} \in \mathbb{C}^N$ has the probability density function equal to a constant q, if $\mathbf{a}$ belongs to the set $\mathcal{Q} = Q^N \setminus \cup_{m\neq k}\{|a_m - a_k| > 2r\}$. Here, q is taken as the volume of $\mathcal{Q}$. The probability density function is equal to zero, if $(a_1, a_2,\ldots,a_N)$ does not belong to $\mathcal{Q}$. The introduced above distribution of the random value $\mathbf{a}$ is denoted by $\mathcal{U}$. According to [31], $0 \leqslant v \leqslant \frac{\pi}{\sqrt{12}}$ where $\frac{\pi}{\sqrt{12}}$ is the maximal concentration attained for the hexagonal array. It is worth noting that the disks D_k belong to Q in the torus topology when the opposite sides of Q are glued by pairs.

The formal definition of the random variable $\mathbf{a}$ has to be statistically realized for large N to get numerical results. The following two constructive ways are considered in the present paper.

Method I (Sequence location [9,14]). Let the random point a_1 is uniformly distributed in Q, i.e., a_1 belong to a measurable set $G \subset Q$ of the area $|G|$ with the probability $|G|$ (the area of Q holds 1). Next, the random point a_2 is uniformly distributed in $Q\backslash G_1$, where G_1 denote the disk $|z - a_1| \leqslant 2r$. Hence, the distribution of the random point a_2 is conditional and depends on the random point a_1. Next, the random point a_3 is uniformly distributed in $Q\backslash(G_1 \cup G_2)$, where the disk $G_2 = \{z \in \mathbb{C} :| z - a_2 |\leqslant 2r\}$ can overlap the disk G_1 and so on. The last random point a_N is uniformly distributed in $Q \setminus (\cup_{m=1}^{N-1} G_m)$. This joint random variable $(a_1, a_2,\ldots,a_N)$ correctly

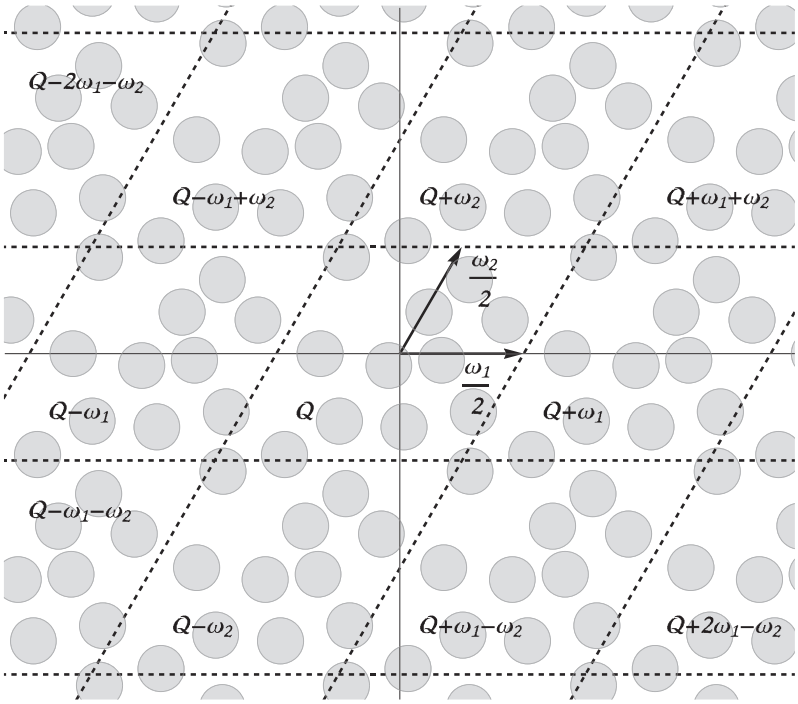

Fig. 1. Doubly periodic composite with inclusions $D_k + m_1\ \omega_1 + m_2\omega_2$ where $m_1, m_2 \in \mathbb{Z}$.

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Table 1
Averaged critical concentration $\langle v\rangle$ for Method I estimated by straightforward simulations for various r.

r	0.025	0.05	0.075	0.1	0.125	0.15	0.175	0.2
$\langle v\rangle$	0.5199	0.5325	0.5372	0.5372	0.5154	0.5443	0.5195	0.5152

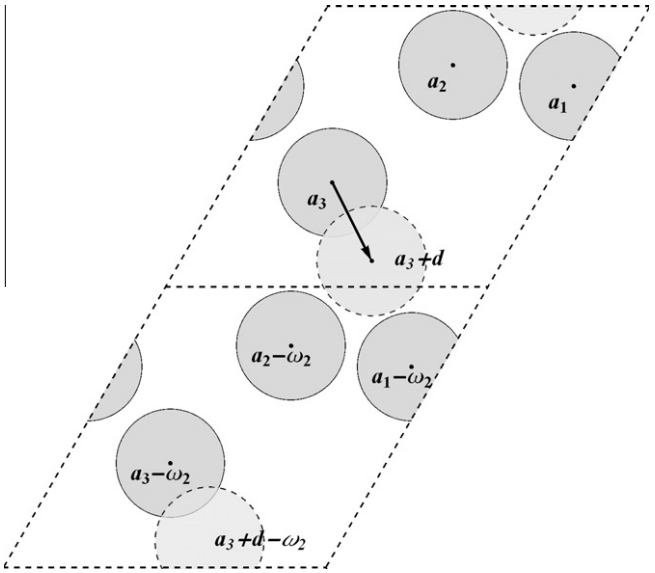

Fig. 2. Illustration of Method II (random walk). The point a_3 moves onto $a_3 - id$ and the disk D_3 goes out of the periodicity cell. However, D_3 lies in Q (by parts) in the torus topology.

determines $\mathcal{U}$ if only v does not exceed the critical value $\frac{\pi}{\sqrt{108}} \approx 0.302$ equal to the rarest concentration of disks [31], i.e., such a concentration for which it is impossible to add a disk not overlapped others. The above restriction on v follows from the observation that for any fixed concentration $v > \frac{\pi}{\sqrt{108}}$ there exists a location of $N-1$ disks with sufficiently large number N such that it is impossible to add the N-th disk not overlapped others. In the same time, the distribution $\mathcal{U}$ assumes that exactly N non-overlapping disks belong to Q. Computer simulations yield the critical concentrations from 0.481 to 0.5773. Hence, the distribution $\mathcal{U}$ can be constructively described by this method for concentrations higher than 0.302 (see Table 1).

Method II (Random walk). The second way to describe $\mathcal{U}$ is based on random walks. Put the centers a_k onto the nodes of the hexagonal array. Take a positive number d less than $\min_{k\neq m}|a_k - a_m| - 2r$. Let each a_k moves in a randomly chosen direction $\phi_k \in [0, 2\pi)$ with the step d in the torus topology of Q (see Fig. 2). Then, each center obtains new complex coordinate $a'_k = a_k + de^{i\phi_k}$. This move is repeated with renewed coordinates for each $k = 1,2,\ldots,N$ if $|a'_k - a_m| \geqslant 2r$ (for all $m = k+1, k+2,\ldots,N$). If $|a'_k - a_m| < 2r$ for some m, the point a_k does not move at this step, and we say that it is blocked. After sufficiently large number of the walks the obtained location of the centers can be considered as a statistical realization of the distribution $\mathcal{U}$. This method can be applied for arbitrary concentrations $\left(0 < v < \frac{\pi}{\sqrt{12}}\right)$.

In all computer simulations, we take

$$d = \frac{1}{5}\left(\frac{\omega_1}{\sqrt{N}} - 2r\right). \tag{2}$$

This is the maximal reasonable value of d for which computations are not too frequently blocked for v not closed to $\frac{\pi}{\sqrt{12}}$. Less value of d decrease the velocity of walking, hence, increase the computation time. However for higher concentrations about $\frac{\pi}{\sqrt{12}}$, computations are frequently blocked for any choice of d. To overcome this difficulty we introduce the maximal number of random choices of the angle ϕ_k, denoted below by P.

After a number of experiments we have taken the following parameters to optimize computations in precision and in time. For each fixed N and v we calculate r, d and introduce $P = 1$, 2 or 3. Every center a_k has P attempts to move to a'_k with a randomly chosen direction in each attempt. If it is done for all $k = 1,2,\ldots,N$, we say that a cycle is performed. The minimal number of cycles is taken as 80 in all simulations. More precisely, we begin with the hexagonal array and take the first location of $\mathbf{a} = (a_1, a_2,\ldots,a_N)$ after 80 steps. The second location is taken after 160 steps, the third after 240 and so forth. The total number of locations is denoted by M.

Stochastic principle of symmetry implies that the distribution $\mathcal{U}$ yields composites isotropic in macroscale, i.e., the effective conductivity is expressed by a scalar $\hat{\lambda}$.

3. Effective conductivity of deterministic composites

We study the conductivity of doubly periodic composites, when the domain $D + m_1\omega_1 + im_2\omega_2$ is occupied by material of the normalized unit conductivity. The respective conductivity of inclusions $D_k + m_1\omega_1 + im_2\omega_2$ is denoted by λ. The potentials $u(z)$ and $u_k(z)$ are harmonic in D and D_k ($k = 1,2,\ldots,N$) and satisfies the conjugation conditions

$$u = u_k, \quad \frac{\partial u}{\partial n} = \lambda\frac{\partial u_k}{\partial n} \quad \text{on} \quad |z - a_k| = r, \quad k = 1,2,\ldots,N, \tag{3}$$

where $\partial/\partial n$ is the outward normal derivative. The external field is given by the following conditions

$$u(z + \omega_1) = u(z) + \omega_1, \ u(z + \omega_2) = u(z). \tag{4}$$

Following [23] we introduce the complex potentials

$$\psi_k(z) = \frac{\lambda + 1}{2}\left(\frac{\partial u_k}{\partial x} - i\frac{\partial u_k}{\partial y}\right) \quad |z - a_k| \leqslant r, \quad k = 1,2,\ldots,N. \tag{5}$$

The effective conductivity can be calculated by the formula [23]

$$\hat{\lambda} = 1 + 2\rho v\frac{1}{N}\sum_{k=1}^{N}\psi_k(a_k). \tag{6}$$

Formula (6) corresponds to Rayleigh's formula [26,15,16] for regular arrays when $N = 1$. The Eisenstein–Rayleigh sums and the Eisenstein functions are used for $N > 1$ (see Appendix).

Theorem 1 (*8,21,28*). *The effective conductivity $\hat{\lambda}$ of macroscopically isotropic composite has the form*

$$\hat{\lambda} = 1 + 2\rho v + 2\rho v\sum_{k=1}^{\infty}A_k v^k. \tag{7}$$

Few first coefficients A_k can be calculated by the following formulae

$$\begin{aligned}
A_1 &= \frac{\rho}{\pi}e_2, \quad A_2 = \frac{\rho^2}{\pi^2}e_{22}, \ A_3 = \frac{1}{\pi^3}[-2\rho^2 e_{33} + \rho^3 e_{222}],\\
A_4 &= \frac{1}{\pi^4}[3\rho^2 e_{44} - 2\rho^3(e_{332} + e_{233}) + \rho^4 e_{2222}],\\
A_5 &= \frac{1}{\pi^5}[-4\rho^2 e_{55} + 3\rho^3(e_{442} + 2e_{343} + e_{244})\\
&\quad - 2\rho^4(e_{3322} + e_{2332} + e_{2233}) + \rho^5 e_{22222}],\\
A_6 &= \frac{1}{\pi^6}[5\rho^2 e_{66} - 4\rho^3(e_{255} + 3e_{354} + 3e_{453} + e_{552})\\
&\quad + \rho^4(3e_{2244} + 6e_{2343} + 4e_{3333} + 3e_{2442} + 6e_{3432} + 3e_{4422})\\
&\quad - 2\rho^5(e_{22233} + e_{22332} + e_{23322} + e_{33222}) + \rho^6 e_{222222}],
\end{aligned} \tag{8}$$

where $e_{m_1\ldots m_q}$ has the form (33).

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A constructive algorithm to symbolically calculate the next A_k ($k > 6$) is described in [8,21,28]. It will be seen later that approximations of (7) up to $O(v^7)$ give good results for all concentrations $v < \frac{\pi}{\sqrt{12}}$. So, below only coefficients A_k ($k \leqslant 5$) are evaluated. Formulae (8) are valid for arbitrary location of a_k ($k = 1,2,\ldots,N$).

Consider the normalized effective conductivity as a function $\hat{\lambda} = \hat{\lambda}(\rho)$ of ρ. It follows from Keller's identity [13] that

$$\hat{\lambda}(\rho)\hat{\lambda}(-\rho) = 1. \tag{9}$$

Substitute (7) and (8) into (9) and compare the coefficients on v^m ($m = 0,1,\ldots,6$). Equations obtained by the coefficients on v^0, v^1 and v^3 are identities. The coefficients on v^2, v^4, v^5 and v^6 yield

$$\begin{aligned} &e_2 = \pi,\ e_{222} = 2\pi e_{22} - \pi^3,\ e_{233} + e_{332} \\ &\quad = 2\pi e_{33},\ e_{244} + e_{343} + e_{442} = 2\pi e_{44}. \end{aligned} \tag{10}$$

This implies that the coefficient A_3, A_4 and A_5 from (8) can be simplified and the normalized effective conductivity $\hat{\lambda}$ can be calculated up to $O(v^7)$ by formula

$$\begin{aligned} \hat{\lambda} &= 1 + 2\rho v + 2\rho^2 v^2 - 2\rho^4 v^4 + 4\rho^6 v^6 \\ &\quad + \frac{2\rho^3 v^3}{\pi^2}\Big[(1 + 2\rho v - 4\rho^3 v^3)e_{22} + \frac{\rho^3 v^3}{\pi^2} e_{22}^2 - \frac{2v}{\pi}(1 + 2\rho v)e_{33} \\ &\quad + \frac{3v^2}{\pi^2}(1 + 2\rho v)e_{44} + \frac{\rho^2 v^2}{\pi^2}(1 + 2\rho v)e_{2222} - \frac{4v^3}{\pi^3} e_{55} \\ &\quad - \frac{2\rho^2 v^3}{\pi^3}(e_{2332} + 2Re\ e_{3322})\Big] + O(v^7). \end{aligned} \tag{11}$$

The real part *Re* of (11) arises in the later formula after simplifications based on Lemma 1.

In the present paper, formula (11) is considered with random locations of the centers described in Section 2. Denote the mathematical expectation of the random function $f(\mathbf{a})$ by $\mathbf{E}[f(\mathbf{a})]$ where the random variable $\mathbf{a}$ obeys the distribution $\mathcal{U}$. The expected effective conductivity of the considered random composite is obtained by the averaged formula (11)

$$\mathbf{E}[\hat{\lambda}] = 1 + 2\rho v + 2\rho^2 v^2 - 2\rho^4 v^4 + 4\rho^6 v^6 + \mu(v,\rho) + O(v^7), \tag{12}$$

where

$$\begin{aligned} \mu(v,\rho) = \frac{2\rho^3 v^3}{\pi^2}\Big[&(1 + 2\rho v - 4\rho^3 v^3)\mathbf{E}[e_{22}] + \frac{\rho^3 v^3}{\pi^2}\mathbf{E}[e_{22}]^2 \\ &- \frac{2v}{\pi}(1 + 2\rho v)\mathbf{E}[e_{33}] + \frac{3v^2}{\pi^2}(1 + 2\rho v)\mathbf{E}[e_{44}] \\ &+ \frac{\rho^2 v^2}{\pi^2}(1 + 2\rho v)\mathbf{E}[e_{2222}] - \frac{4v^3}{\pi^3}\mathbf{E}[e_{55}] - \frac{2\rho^2 v^3}{\pi^3}(\mathbf{E}[e_{2332}] \\ &+ 2Re\ \mathbf{E}[e_{3322}])\Big]. \end{aligned} \tag{13}$$

4. Computation of Eisenstein–Rayleigh sums

The method of Monte Carlo is used to calculate the effective conductivity (12) through the expected values $\mathbf{E}[\,\cdot\,]$ from the right hand part of (13). More precisely, a set of the centers $\mathbf{a}$ satisfying the distribution $\mathcal{U}$ is numerically simulated and the corresponding sum is computed. This is one numerical experiment in the method of Monte Carlo. Such computations are performed M times and the mean value of the results is taken as an approximation of the corresponding expected value. The statistically estimated value $\mathbf{E}[X]$ of a random value X is denoted by $\langle X\rangle$.

4.1. Computation of e_2

In order to numerically apply the method of Monte Carlo it is worth to numerically check the relation following from the first Eq. (10)

$$\mathbf{E}[e_2] = \pi. \tag{14}$$

We need to estimate the number N of inclusions per cell and the number of the realizations M of the random value $\mathbf{a}$ to investigate the question, for which N and M the statistical average $\langle e_2\rangle$ is closed to the theoretical value (14). Here, $\langle e_2\rangle$ is equal to the mean value of $(e_2)_m$ calculated for $\mathbf{a}$ generated in the mth numerical experiment, more precisely:

$$\langle e_2\rangle = \frac{1}{M}\sum_{m=1}^{M}(e_2)_m. \tag{15}$$

Further, it is assumed that the approximation $\langle X\rangle \approx \mathbf{E}[X]$ is valid, if the respective approximation does not exceed 2%, i.e.,

$$\left|\frac{\langle X\rangle - \mathbf{E}[X]}{\mathbf{E}[X]}\right| < 0.02. \tag{16}$$

It is necessary also to perform such computations for the random value $\mathbf{a}$ simulated by the both methods described in Section 2. It is clear that Method I realizes $\mathcal{U}$ for small v and Method II for large v. It is important to justify numerically that these methods are matched for moderate v when the number of inclusions per periodicity cell N or the number of the numerical experiments M is sufficiently large. We have to numerically determine which N and M can be taken as sufficiently large to match the both methods, i.e., to compare the sums computed by the first (sequence locations) $\langle e_2\rangle_I$ and by the second (random walk) methods $\langle e_2\rangle_{II}$.

The results for $N = 64$ and $1 < M < 6000$ are presented in Fig. 3. Computations for $v = 0.4$ and $M = 6000$ yield

$$\begin{aligned} &\langle e_2\rangle_I \approx 3.14035 - 0.00156569i, \\ &\langle e_2\rangle_{II} \approx 3.13979 - 0.00505772i, \end{aligned} \tag{17}$$

with the respective error 0.0011. The imaginary parts also show the absolute errors of computations. Therefore, $\langle e_2\rangle_I$ and $\langle e_2\rangle_{II}$ have almost the same computed values and further are not separated. One can see that $\langle e_2\rangle$ does not depend on v. The result $\langle e_2\rangle \approx \pi$ with error 2% by (16) is achieved for $N = 64$ and $M > 1000$ for all not small v. This observation suggests that the expected values in (12) can be statistically estimated with the same N and M.

Remark. The declared error refers to the concentrations greater than 0.05. The average deviation of the Esenstein–Rayleigh sums for v less than 0.05 is very high since the distances between the centers are small. The singularity of the Eisenstein functions at the point $z = 0$ implies that their oscillations near zero are high. Fortunately, the Clausius–Mossotti approximation

$$\lambda_e \approx \frac{1 + \rho v}{1 - \rho v}$$

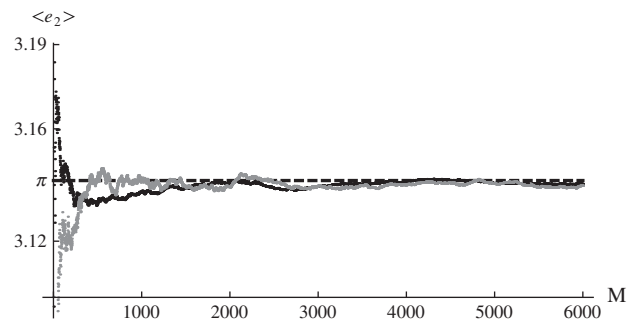

Fig. 3. Sums computed by the first $\langle e_2\rangle_I$ and by the second methods $\langle e_2\rangle_{II}$ for $N = 64$, $v = 0.4$ and $P = 2$.

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gives excellent results for dilute composites. Actually, $\langle e_2 \rangle$ is taken here equal to π and the higher order Esenstein–Rayleigh sums are not used in this approximation.

4.2. Computation of Esenstein–Rayleigh sums of higher orders

It is established in the previous section that computations of $\langle e_2 \rangle$ are satisfactory for $N = 64$ and $M > 1000$. Now, we compute $\langle e_{22} \rangle$, $\langle e_{33} \rangle$, $\langle e_{44} \rangle$, $\langle e_{55} \rangle$, $\langle e_{2222} \rangle$, $\langle e_{2332} \rangle$ and $\langle Re\ e_{3322} \rangle$ to optimize the parameters N, M and P. First, put $N = 64$ and investigate the dependence of computations on M. Consider the following cases: (a) $v = 0.2$, $P = 1$, (b) $v = 0.45$, $P = 2$ and (c) $v = 0.7$, $P = 3$. The results are summarized in Tables 2–7 and Figs. 4–9. One can observe that errors do not exceed 2% for $M = 1500$.

We now proceed to investigate the dependence of computations on N. Consider the following cases: (a) $N = 4$, 9, 25, 36, 49, 64, 100 and (a.1) $P = 1$, $v = 0.2$, (a.2) $P = 2$, $v = 0.45$, (a.3) $P = 3$, $v = 0.7$; (b) $N = 64{,}100$ and (b.1) $P = 2{,}3$, $v = 0.2$, (b.2) $P = 1{,}3$, $v = 0.45$, (b.3) $P = 1{,}2$, $v = 0.7$ with M corresponding to N in accordance with Table 2.

The results are presented in Figs. 7–9 and Tables 3 and 4. One can see that the error 3% is achieved for $N = 64$ and the corresponding M from Table 2. The worst computations take place for e_{44} for high v (see Fig. 9) when the error slightly exceed 2%.

It follows from computations that the optimal parameter P depends on the concentration v as follows: $P = 1$ for $v \leqslant 0.3$, $P = 2$ for $0.3 < v \leqslant 0.6$, $P = 3$ for $v \geqslant 0.65$.

Actually, the computations were performed for wide range of the parameters to select the optimal values. In particular, we checked the case $P = 10$ and other formulae for d (the coefficient $\frac{1}{5}$ is optimal in (2)) for all concentrations.

5. Effective conductivity

The results of computations for $\langle e_{22} \rangle$, $\langle e_{33} \rangle$, $\langle e_{44} \rangle$, $\langle e_{55} \rangle$, $\langle e_{2332} \rangle$, $\langle e_{3322} \rangle$ and $\langle\ e_{2222} \rangle$ with the optimal parameters found in the previous section are gathered in Tables 8 and 9. These values are computed with formulae (36) and (38).

The value $\mu(v, \rho)$ is computed by (13) at the points (v, ρ). The structure of the coefficients A_k described in Theorem 3 and (12) and (13) imply that $\mu(v, \rho)$ has the form

$$\mu(v,\rho) = \rho^3 v^3 (b_1(\rho) + b_2(\rho)v + b_3(\rho)v^2 + b_4(\rho)v^3), \tag{18}$$

where $b_j(\rho)$ are third power polynomials in ρ. Coefficients of these polynomials are computed by the method of least squares. The final formula for the effective conductivity (12) and (13) has the form

$$\begin{aligned}\hat{\lambda}(v,\rho) = {} & 1 + 2v\rho + 2v^2\rho^2 + 4.9843v^3\rho^3 - 6.829v^4\rho^3 \\ & + 4.2139v^5\rho^3 - 0.3462v^6\rho^3 - 0.0688v^3\rho^4 \\ & + 7.3652v^4\rho^4 - 12.4218v^5\rho^4 + 7.0868v^6\rho^4 \\ & - 0.1463v^3\rho^5 + 6.3079v^4\rho^5 - 10.4599v^5\rho^5 \\ & + 6.7108v^6\rho^5 - 0.7996v^3\rho^6 + 4.517v^4\rho^6 \\ & - 7.8602v^5\rho^6 + 5.9897v^6\rho^6. \end{aligned} \tag{19}$$

Fig. 10 demonstrate the results of computations with (19).

Consider the limit case of perfectly conducting inclusions when λ tends to infinity. Then (1) implies that $\rho = 1$. In this case, the effective conductivity tends to infinity as the concentration v tends to the maximal value $\frac{\pi}{\sqrt{12}}$ achieved for the hexagonal array. This physical effect can be observed in formula (19) by substitution $\rho = 1$ and application of Padé approximations. We have

Table 2
Choice of M for each N.

N	4	9	16	25	36	49	64	81	100
M	6000	5000	4000	3000	2500	2000	1500	1500	1500

Table 3
Computed values $\langle e_{22} \rangle$ for various parameters.

N	$P = 1$, $v = 0.2$	$P = 2$, $v = 0.45$	$P = 3$, $v = 0.7$
4	16.8903	10.7185	9.8935
9	17.7393	11.4631	9.9885
16	18.0750	11.5222	10.0216
25	18.1191	11.5276	10.0440
36	18.1236	11.5276	10.0485
49	18.1481	11.5290	10.0608
64	18.2159	11.5527	10.0566
81	18.1874	11.5531	10.0508
100	18.1954	11.5249	10.0599

Table 4
Computed values $\langle e_{44} \rangle$ for various parameters.

N	$P = 1$, $v = 0.2$	$P = 2$, $v = 0.45$	$P = 3$, $v = 0.7$
4	64.5058	7.5137	0.3422
9	72.4801	7.9055	0.7710
16	75.1789	7.8997	0.9270
25	76.0096	7.9246	0.9837
36	76.7844	7.9559	1.0265
49	76.8793	7.9656	1.0569
64	76.7296	7.9377	1.0751
81	77.3498	7.9647	1.0853
100	77.5732	7.9885	1.0939

Table 5
Computed values $\langle e_{22} \rangle$, $\langle\ e_{33} \rangle$, $\langle e_{2222} \rangle$, $Re\ (\langle e_{55} \rangle)$, $\langle e_{2332} \rangle$, $\langle e_{44} \rangle$, $\langle e_{55} \rangle$ for $P = 1$, $v = 0.2$.

	$N = 64$	$N = 100$	Respective error
$\langle e_{22} \rangle$	18.21590	18.19538	0.00113
$\langle e_{33} \rangle$	−23.25810	−23.36730	0.00467
$\langle e_{222} \rangle$	576.09200	575.24150	0.00148
$Re\ (\langle e_{3322} \rangle)$	−573.46100	−576.06877	0.00453
$\langle e_{2332} \rangle$	−591.86700	−592.80760	0.00159
$\langle e_{44} \rangle$	76.72960	77.57321	0.01087
$\langle e_{55} \rangle$	−252.174000	−256.51035	0.01691

Table 6
Computed values $\langle e_{22} \rangle$, $\langle e_{33} \rangle$, $\langle e_{2222} \rangle$, $Re\ (\langle e_{55} \rangle)$, $\langle e_{2332} \rangle$, $\langle e_{44} \rangle$, $\langle e_{55} \rangle$ for $P = 2$, $v = 0.45$.

	$N = 64$	$N = 100$	Respective error
$\langle e_{22} \rangle$	11.55270	11.52493	0.00241
$\langle e_{33} \rangle$	−3.49796	−3.50618	0.00234
$\langle e_{222} \rangle$	161.76800	161.13483	0.00393
$Re\ (\langle e_{3322} \rangle)$	−41.88310	−41.85512	0.00067
$\langle e_{2332} \rangle$	−48.51970	−48.40682	0.00233
$\langle e_{44} \rangle$	7.93768	7.98846	0.00636
$\langle e_{55} \rangle$	−15.78050	−15.77928	0.00008

Table 7
Computed values $\langle e_{22} \rangle$, $\langle\ e_{33} \rangle$, $\langle e_{2222} \rangle$, $Re\ (\langle\ e_{55} \rangle)$, $\langle e_{2332} \rangle$, $\langle\ e_{44} \rangle$, $\langle e_{55} \rangle$ for $P = 3$, $v = 0.7$.

	$N = 64$	$N = 100$	Respective error
$\langle e_{22} \rangle$	10.05659	10.05990	0.00033
$\langle e_{33} \rangle$	−0.40234	−0.40832	0.01464
$\langle e_{222} \rangle$	103.82047	103.93200	0.00107
$Re\ (\langle e_{3322} \rangle)$	−3.64061	−3.68913	0.01315
$\langle e_{2332} \rangle$	−5.11471	−5.17244	0.01116
$\langle e_{44} \rangle$	1.07512	1.09393	0.01719
$\langle e_{55} \rangle$	−3.85177	−3.85965	0.00204

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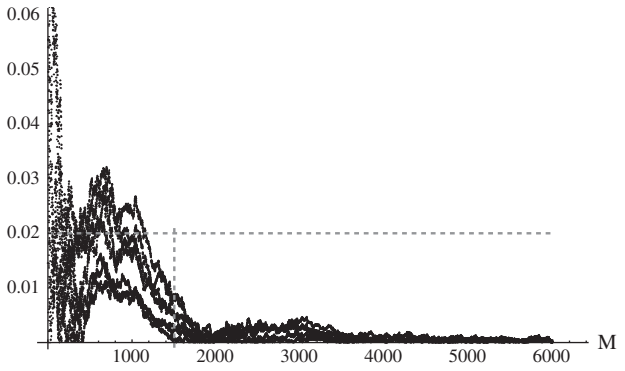

Fig. 4. Computational error in $\langle e_p \rangle$ when p runs over the set 22, 33, 44, 55, 2222, 2332, 3322 in case (a).

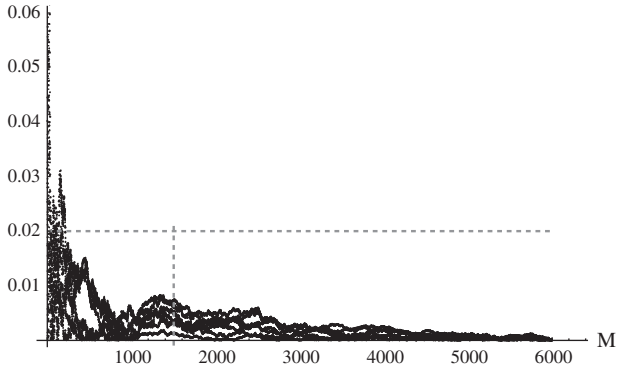

Fig. 5. Computational error in $\langle e_p \rangle$ when p runs over the set 22, 33, 44, 55, 2222, 2332, 3322 in case (b).

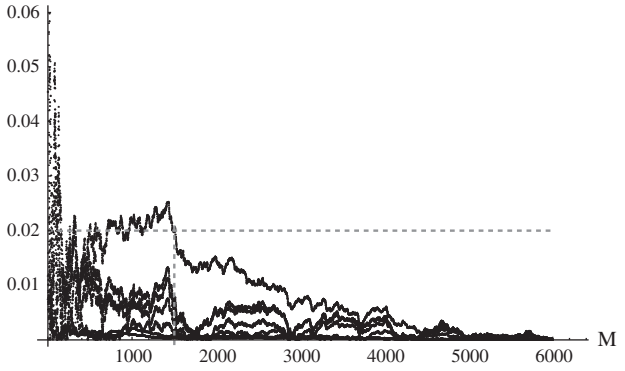

Fig. 6. Computational error in $\langle e_p \rangle$ when p runs over the set 22, 33, 44, 55, 2222, 2332, 3322 in case (c).

$$\hat{\lambda}(v,1) = 1 + 2v + 2v^2 + 3.9696v^3 + 11.3611v^4 - 26.528v^5 + 19.4411v^6 + O(v^7). \tag{20}$$

Padé approximation for (20) of order (2, 4) has the form

$$\hat{\lambda}(v,1) \approx \frac{3.299}{v - 1.241} - \frac{3.313}{v - 0.9095} + \frac{0.014v + 0.001}{v^2 + 0.26v + 0.076}. \tag{21}$$

One can see that the function (21) has a pole at the point $v = 0.9095$ which is close to $\frac{\pi}{\sqrt{12}} \approx 0.9069$ (the respective error is equal to 0.002867). The results of computations are shown in Fig. 11. The Padé approximation for (20) with the fixed pole at $\frac{\pi}{\sqrt{12}} \approx 0.9069$ yields

$$\hat{\lambda}(v,1) \approx \frac{3.223}{v - 1.247} - \frac{3.237}{v - 0.9069} + \frac{0.014v + 0.001}{v^2 + 0.261v + 0.076}. \tag{22}$$

Formulae (21) and (22) give numerically the same results.

We now proceed to compare our formula (19) with formula (20.77) by Torquato (see [29, p. 526]) written in our designations

$$\hat{\lambda} = 1 + \frac{2a_1\rho}{1-\rho} + \frac{4a_2\rho^2}{(1-\rho)^2} + \frac{8a_3\rho^3}{(1-\rho)^3} + \frac{16a_4\rho^4}{(1-\rho)^4}, \tag{23}$$

where

$$\begin{aligned} a_1 &= v, \\ a_2 &= -\frac{v - v^2}{2}, \\ a_3 &= \frac{v(1-v)}{4}(1 - v + \zeta), \\ a_4 &= \frac{1}{8}(-12a_3 + 2v + 8a_3v - 5v^2 + 4v^3 - v^4). \end{aligned} \tag{24}$$

The parameter ζ is given in Table 22.1 (see [29, p. 599], column "Identical hard cylinders"). Formulae (19) and (23) give the same results for $\rho < 0.45$. The difference between these formulae is demonstrated in Fig. 12.

6. Discussion

The main result of the present paper is extension of Torquato's formula (23) to high values of the contrast parameter ρ. We deduce formula (19) which is valid for the uniformly distributed non-overlapping disks. The introduced distribution $\mathcal{U}$ is a probabilistic distribution, i.e., an ideal mathematical object which can be realized by various statistical methods (see [29] and further discussion devoted to this point in [30]). In the present paper, we use two special methods (sequence location and random walk) which theoretically converge to the distribution $\mathcal{U}$. The number of computational experiments is optimally chosen and should not be increased. It is checked that it is sufficient in the framework of the described rules of simulation. But the rules can be changed and extended. Method II is based on random walks beginning from the hexagonal array. Other initial positions for the random walks such as rectangular ar-

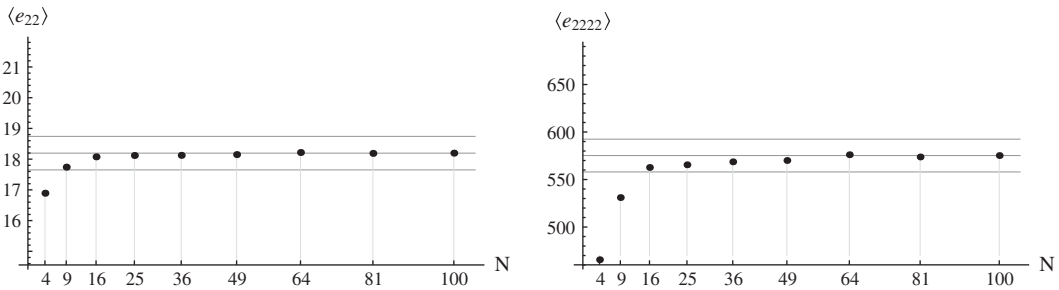

Fig. 7. Results for $\langle e_{22} \rangle$ *i* $\langle e_{2222} \rangle$ in case (a.1).

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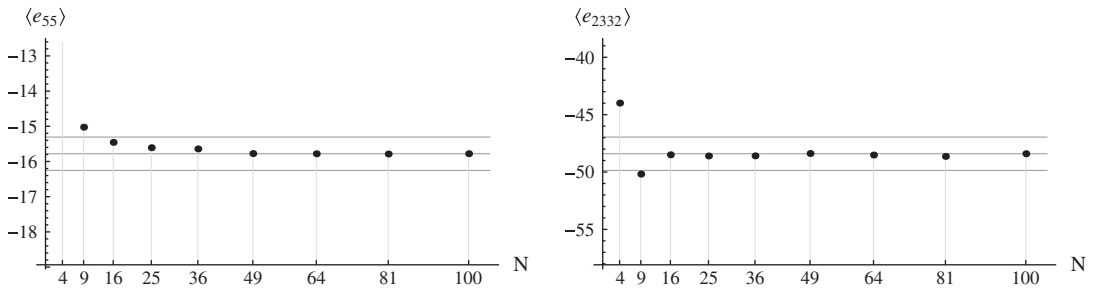

Fig. 8. Results for $\langle e_{55}\rangle$ *i* $\langle e_{2332}\rangle$ in case (a.2).

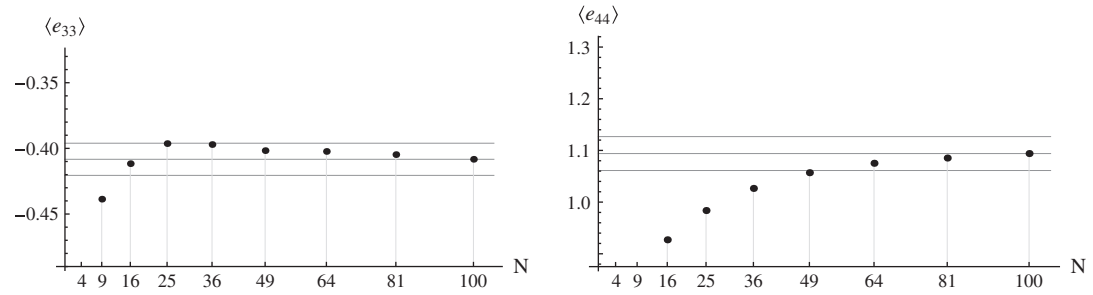

Fig. 9. Results for $\langle e_{33}\rangle$ *i* $\langle e_{44}\rangle$ in case (a.3).

Table 8
Computed values $\langle e_{22},\rangle$, $\langle e_{33},\rangle$, $\langle e_{44},\rangle$ and $\langle e_{55},\rangle$ for various concentrations v. Computations are performed for the optimal choice of the parameters including $N = 64$.

v	$\langle e_{22}\rangle$	$\langle e_{33}\rangle$	$\langle e_{44}\rangle$	$\langle e_{55}\rangle$
0.05	54.4562	−382.333	4190.05	−50427.7
0.1	30.3341	−94.6858	554.766	−3416.08
0.15	22.346	−41.9403	174.956	−739.574
0.2	18.2159	−23.2581	76.7296	−252.174
0.25	15.7966	−14.6562	41.6043	−114.382
0.3	14.2628	−9.88543	25.2077	−61.0286
0.35	12.8822	−6.46391	15.3884	−33.213
0.4	12.1174	−4.69994	10.8078	−22.0623
0.45	11.5527	−3.49796	7.93768	−15.7805
0.5	11.0934	−2.57665	5.94659	−11.9023
0.55	10.766	−1.90511	4.49997	−9.38056
0.6	10.4837	−1.38498	3.38179	−7.58524
0.65	10.2755	−0.911354	2.37488	−6.06037
0.7	10.0566	−0.402342	1.07512	−3.85177
0.75	9.96082	−0.198966	0.52554	−2.16478
0.8	9.90916	−0.0839384	0.216754	−0.982389
0.85	9.8814	−0.0222027	0.0588841	−0.267471
0.9	9.86974	−0.00031838	0.000867687	−0.0038436

Table 9
Continuation of the previous table.

v	$\langle e_{2222}\rangle$	$Re\,(\langle e_{3322}\rangle)$	$\langle e_{2332}\rangle$
0.05	9643.33	−66675.3	−64221.9
0.1	2135.82	−5660.96	−5548.78
0.15	974.611	−1463.18	−1476.22
0.2	576.092	−573.461	−591.867
0.25	395.838	−286.93	−303.729
0.3	297.019	−163.827	−178.244
0.35	223.286	−91.9693	−102.209
0.4	186.765	−60.5965	−68.9144
0.45	161.768	−41.8831	−48.5197
0.5	142.964	−28.9071	−34.1006
0.55	129.983	−20.3632	−24.4439
0.6	119.44	−14.1216	−17.1713
0.65	111.759	−8.97626	−11.1578
0.7	103.82	−3.64061	−5.11471
0.75	100.567	−1.72926	−2.54225
0.8	98.7614	−0.71325	−1.07474
0.85	97.7665	−0.187318	−0.284677
0.9	97.414	−0.00267407	−0.0040697

rays were considered. It is established that after 80 cycles of steps the Eisenstein–Rayleigh sums do not practically change, hence, the inclusions are intimately stirred and their centers can be taken as representative results of the Monte Carlo experiments. Such a model certainly holds for random composites obtained from regular arrays. May be, other rules of simulations can yield other values of the Eisenstein–Rayleigh sums. It is interesting to investigate such configurations of disks that can be transformed into the hexagonal array by random walks with very small probabilities. If the probabilistic measure of such subsets of configurations is not small in $\mathcal{U}$, the Eisenstein–Rayleigh sums, hence formulae (21) and (22) can be changed. It is not clear now how such configurations can be simulated to get representative elements since the most appropriate candidates, disordered jammed packings are ill-defined and depend on the protocol used to produce the packings [11].

In the limit case of high contrast parameter, we deduce formulae (21) and (22) which are valid for all concentrations. The first formula (21) is obtained from (19) by substitution $\rho = 1$ and use of the Padé approximation of order (2,4). The singularity at the point $v = 0.9095$ confirms that the accuracy $O(v^7)$ is sufficient to get the theoretical singularity at $v = 0.9069$ and that the order of the Padé approximation is properly chosen. The second formula (22) is obtained by the Padé approximation with the fixed pole at $v = 0.9069$.

The n-point correlation functions are not formally used in our method. However, they are implicitly presented in (21) and (22) According to [29] the n-point correlation functions, as a pure geometric object, are used to solve the corresponding boundary value problem and to estimate the effective conductivity by an appropriate averaging. In our formulae one can observe these averaged correlation functions. The coefficients A_k are presented as linear

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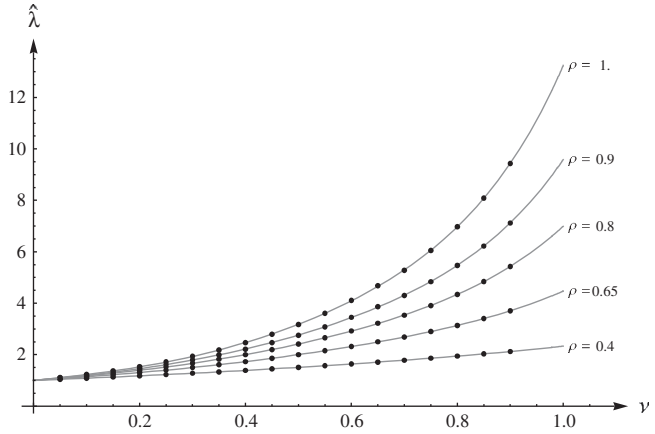

Fig. 10. Effective conductivity (19) for various ρ. Dots correspond to the values form Tables 8 and 9.

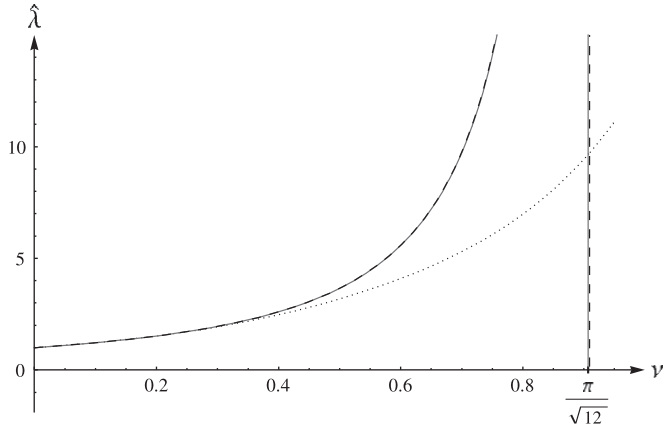

Fig. 11. Effective conductivity as function of ν. Data are for: dotted line (20); broken line: (21); solid line (22).

combinations of the Eisenstein–Rayleigh sums (33) with coefficients containing ρ. Every sum (33) depends only on the distances $|a_k - a_m|$. This is the result of the averaging of n-point correlation functions. Thus, the effective conductivity is decomposed onto the sum (21) in which the pure geometric terms (33), the contrast parameter ρ and the concentration ν are explicitly presented. In the present paper, the terms (33) are computed for the distribution $\mathcal{U}$ which describes the uniform non-overlapping location of inclusions. It is possible to define another distribution and to determine the effective conductivity tensor by (33).

One can consider the set $\{e_{m_1\ldots m_q}, m_j = 2, 3, \ldots\}$ as a basis in the space of the deterministic or random locations of inclusions. This observation was used in [24] to create a constructive theory of representative volume elements. The results presented in Tables 8 and 9 can be considered as the characteristic parameters of the distribution $\mathcal{U}$ in the following way. Let a set of the centers of inclusions be measured in a sample with two-dimensional geometry and the sums $e_{m_1\ldots m_q}$ are computed for the measured data. If $e_2 \approx \pi$, the considered composite is rather isotropic. This assertion is true at least for not high concentrations. If the rest sums $e_{m_1\ldots m_q}$ are closed to the values from Tables 8 and 9, one can assign this composite to the class of composites $\mathcal{U}$ obtained by stirring of the hard inclusions in a host. Let us consider another case when the values $e_{m_1\ldots m_q}$ coincide with the theoretical values from Tables 8 and 9 but for higher concentrations than in the sample. This means that the sample could be obtained from a periodic structure by perturbations modelled by random walks in Method II. These perturbations can be restricted, for instance in time. Hence, the hard inclusions are not intimately stirred. To the best of our knowledge no such experimental investigations have been carried out.

Appendix A

Following [22,24] we present constructive formulae for the Einstein–Rayleigh sums S_m and the Einstein functions $E_m(z)$ corresponding to the lattice $\mathcal{Q}$.

The Einstein–Rayleigh lattice sums S_m are defined as

$$S_m = \sum_{m_1,m_2} (m_1 + im_2)^{-m}, \quad m = 2, 3, \ldots, \tag{25}$$

where m_1, m_2 run over all integers except $m_1 = m_2 = 0$. The sums (25) are slowly convergent. They can be easily calculated through the rapidly convergent series

$$S_2 = \left(\frac{\pi}{\omega_1}\right)^2 \left(\frac{1}{3} - 8\sum_{m=1}^{\infty} \frac{mq^{2m}}{1-q^{2m}}\right), \quad q = \exp\left(\pi i\, \frac{\omega_2}{\omega_1}\right), \tag{26}$$

$$S_4 = 60\left(\frac{\pi}{\omega_1}\right)^4 \left(\frac{4}{3} + 320\sum_{m=1}^{\infty} \frac{m^3q^{2m}}{1-q^{2m}}\right), \tag{27}$$

$$S_6 = 1400\left(\frac{\pi}{\omega_1}\right)^6 \left(\frac{8}{27} - \frac{448}{3}\sum_{m=1}^{\infty} \frac{m^5q^{2m}}{1-q^{2m}}\right). \tag{28}$$

S_{2n} $(n \geqslant 4)$ can be calculated by the recurrent formula

$$S_{2n} = \frac{3}{(2n+1)(2n-1)(n-3)} \sum_{m=2}^{n-2} (2m-1)(2n-2m-1) S_{2m} S_{2(n-m)}. \tag{29}$$

The rest sums vanish. For the hexagonal array, $S_2 = \pi$, $S_4 = 0$, $S_6 = 3.80815$.

The Einstein functions are related to the Weierstrass function $\wp(z)$ [4] by the identities [32]

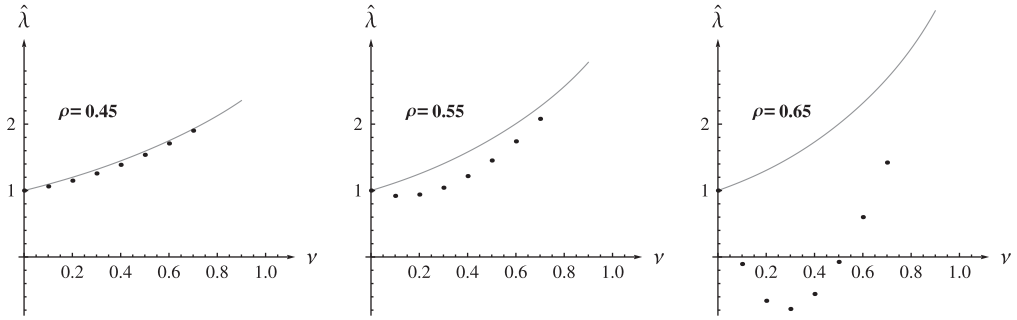

Fig. 12. Effective conductivity as function of ν. Data are for: solid line (19); dotted line (23).

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$$E_2(z) = \wp(z) + S_2, \quad E_m(z) = \frac{(-1)^m}{(m-1)!}\frac{d^{m-2}\wp(z)}{dz^{m-2}}, \quad m = 3, 4, \ldots. \tag{30}$$

Every function (30) is doubly periodic and has a pole of order m at $z = 0$. The Eisenstein functions of the even order $E_{2m}(z)$ can be presented in the form of the series [32]

$$E_{2m}(z) = \frac{1}{z^{2m}} + \sum_{k=1}^{\infty}\sigma_k^{(m)} z^{2(k-1)}, \tag{31}$$

where

$$\sigma_k^{(m)} = \frac{(2m+2k-3)!}{(2m-1)!(2k-2)!} S_{2(m+k-1)}. \tag{32}$$

We follow [24] to introduce the generalized Eisenstein–Rayleigh sums. Let a_k ($k = 1, 2, \ldots, N$) be a set of points. Let q be a positive integer; k_t runs over 1 to N; $m_j = 2, 3, \ldots$. Let $\mathbf{C}$ be the operator of complex conjugation. Introduce the following sum of multi-order $(m_1, \ldots, m_q)$

$$e_{m_1 \ldots m_q} := N^{-[1+\frac{1}{2}(m_1+\cdots+m_q)]} \sum_{k_0 k_1 \ldots k_q} E_{m_1}(a_{k_0} - a_{k_1}) \times \overline{E_{m_2}(a_{k_1} - a_{k_2})} \ldots \mathbf{C}^q E_{m_q}(a_{k_{q-1}} - a_{k_q}). \tag{33}$$

Here, it is assumed for convenience that

$$E_m(0) := S_m. \tag{34}$$

According to (33) and (34), e_m becomes the classical Eisenstein–Rayleigh sum S_m in the case $N = 1$.

The number of sums in formula (33) is equalt to $(q + 1)$ and can be reduced as follows. Introduce the function

$$F_p(z) = \frac{1}{N}\sum_{k=1}^{N} E_p(z - a_k). \tag{35}$$

Then the sum e_{pp} can be written in the form

$$e_{pp} = \frac{1}{N^p}\sum_{k_0, k_1} E_p(a_{k_0} - a_{k_1})\overline{F_p(a_{k_1})} = \frac{(-1)^p}{N^{p-1}}\sum_{k_1} F_p(a_{k_1})\overline{F_p(a_{k_1})} = \frac{(-1)^p}{N^{p+1}}\sum_{m=1}^{N}\left|\sum_{k=1}^{N} E_p(a_m - a_k)\right|^2. \tag{36}$$

Here, it is used that $E_p(-z) = (-1)^p E_p(z)$.

In order to transform e_{2222} introduce the function

$$G_2(z) = \frac{1}{N^2}\sum_{k_1, k_2} E_2(z - a_{k_1})\overline{E_2(a_{k_1} - a_{k_2})}. \tag{37}$$

Then the sum e_{2222} can be transformed similar to (36)

$$e_{2222} = \frac{1}{N}\sum_{k=1}^{N}|G_2(a_k)|^2. \tag{38}$$

Formulae (36) and (38) contain one sum on N less than the definition (33) that is important in computations.

Lemma. *Let* $\alpha = \sum_{j=1}^{q} m_j$. *Then*

$$e_{m_1 \ldots m_q} = (-1)^{\alpha}\mathbf{C}^q e_{m_q \ldots m_1}. \tag{39}$$

Proof. In view of (33) we have

$$e_{m_1 \ldots m_q} = N^{-(1+\frac{1}{2}\alpha)} \sum_{k_0 k_1 \ldots k_q} E_{m_1}(-(a_{k_1} - a_{k_0})) \times \overline{E_{m_2}(-(a_{k_2} - a_{k_1}))} \ldots \mathbf{C}^q E_{m_q}(-(a_{k_q} - a_{k_{q-1}})). \tag{40}$$

It follows from (36) that right-hand part of (40) is of the form

$$(-1)^{\alpha} N^{-(1+\frac{1}{2}\alpha)} \sum_{k_0 k_1 \ldots k_q} \mathbf{C}^q E_{m_q}(a_{k_q} - a_{k_{q-1}}) \ldots \overline{E_{m_2}(a_{k_2} - a_{k_1})} E_{m_1}(a_{k_1} - a_{k_0})$$

which is equal to $(-1)^{\alpha} e_{m_q \ldots m_1}$ or $(-1)^{\alpha}\overline{e_{m_q \ldots m_1}}$ since q is odd or even respectively. The lemma is proved. □

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