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## A NOTE ON THE $C^2$ -TERM OF THE EFFECTIVE CONDUCTIVITY FOR RANDOM DISPERSIONS

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*Константин Марков, Керанка Илиева.* ЗАМЕЧАНИЕ О  $C^2$ -ЧЛЕНЕ ЭФФЕКТИВНОЙ  
ПРОВОДИМОСТИ СЛУЧАЙНОЙ ДИСПЕРСИИ СФЕР

Работа посвящена исследованию эффективной теплопроводности  $\kappa^*$  случайной разряженной суспензии сфер. Специальное внимание уделено  $C^2$ -коэффициенту  $a_2$  в разложении этой проводимости по степеням объемной концентрации сфер  $c$ . Пользуясь простыми соображениями показано, что  $a_2$  представляется суммой постоянной и линейного функционала от радиальной функции распределения сфер. В равнинном случае (материал армированный волокнами) найден аналитический вид этого ядра и выведены некоторые простые оценки для него.

*Konstantin Markov, Keranka Ilieva.* A NOTE ON THE  $C^2$ -TERM OF THE EFFECTIVE  
CONDUCTIVITY FOR RANDOM DISPERSIONS

The paper is devoted to the study of the effective conductivity  $\kappa^*$  of a random dilute dispersion of spheres. A special attention is paid to the  $C^2$ -coefficient  $a_2$  in the expansion of  $\kappa^*$  in powers of the volume fraction  $c$  of the spheres. The functional dependence of  $a_2$  upon the radial distribution function is discussed and it is shown, using simple arguments, that  $a_2$  is a sum of a constant and a linear functional of the said function. The analytical form and certain estimates for the kernel of this functional are obtained in the two-dimensional case (fiber-reinforced material).

## 1. INTRODUCTION

Consider a random dispersion of spheres in the three-dimensional case ( $3D$ ) or cylinders in the two-dimensional ( $2D$ ) case, i.e., an unbounded matrix of conductivity  $\kappa_m$ , containing an array of either spherical or parallel cylindrical inclusions, each one of radius  $a$  and conductivity  $\kappa_f$ . The centers of the inclusions, assumed nonoverlapping, are in the random points  $\mathbf{x}_j$ . The statistics of the dispersion is described by the multipoint distribution densities  $f_p(\mathbf{y}_1, \dots, \mathbf{y}_p)$  that give the probability of finding a center of an inclusion per each of the infinitesimal volumes  $\mathbf{y}_i < \mathbf{y} < \mathbf{y}_i + d\mathbf{y}_i$ ,  $i = 1, \dots, p$ . We assume, as usual, that the dispersion is statistically homogeneous and isotropic and  $f_p \sim n^p$  in the dilute limit  $n \rightarrow 0$ , where  $n$  is the number density of the inclusions. The classical problem consists in evaluating the effective (or overall) conductivity  $\kappa^*$  of the dispersion, making use of the known conductivities  $\kappa_m$  and  $\kappa_f$  of the constituents, and of the statistical information represented by the functions  $f_p$  (cf., e.g., [1-6]). The mathematical formulation of the problem reads

$$(1.1) \quad \nabla \cdot \{\kappa(\mathbf{x})\nabla\theta(\mathbf{x})\} = 0, \quad \langle \nabla\theta(\mathbf{x}) \rangle = \mathbf{G},$$

where  $\theta(\mathbf{x})$  is the random temperature field,  $\kappa(\mathbf{x})$ —the given conductivity field ( $\kappa(\mathbf{x}) = \kappa_f$  or  $\kappa_m$  depending on whether  $\mathbf{x}$  lies in an inclusion or in the matrix respectively),  $\mathbf{G}$ —the prescribed macroscopic gradient of the temperature, and  $\langle \cdot \rangle$  denotes ensemble averaging. Upon solving the random problem (1.1), one calculates the mean flux, which is proportional to the macrogradient  $\mathbf{G}$ :

$$(1.2) \quad \langle \kappa(\mathbf{x})\nabla\theta(\mathbf{x}) \rangle = \kappa^*\mathbf{G},$$

where  $\kappa^*$  is the effective conductivity of the medium. The difficulties in calculating  $\kappa^*$  are well acknowledged in the literature: they stem from the need to account properly for the multiparticle interactions in the dispersions and for the slow decay of the single-inclusion field [2,4,5]. A number of approximations for  $\kappa^*$  exist; one of the first and most famous of them has been proposed by J. Maxwell [7]. Though he dealt with dispersions of spheres, we give the respective result in a bit more general form in order to be able to cover both  $3D$  (dispersion of spheres) and  $2D$ -case (dispersions of aligned cylinders, i.e. fiber-reinforced materials) simultaneously:

$$(1.3a) \quad \frac{\kappa^*}{\kappa_m} = 1 + \frac{d\beta_d}{1 - \beta_d c} = 1 + d\beta_d c + d\beta_d^2 c^2 + \dots,$$

where

$$(1.3b) \quad \beta_d = \frac{[\kappa]}{\kappa_f + (d-1)\kappa_m}, \quad [\kappa] = \kappa_f - \kappa_m;$$

hereafter  $d = 3$  in  $3D$ -case and  $d = 2$  in  $2D$ -case,  $c$  is the volume fraction of the inclusions,  $c = nV_a$ ,  $V_a = \frac{4}{3}\pi a^3$  in  $3D$ -case, or  $c = nS_a$ ,  $S_a = \pi a^2$  in  $2D$ -case.

Let

$$(1.4) \quad \frac{\kappa^*}{\kappa_m} = 1 + a_1 c + a_2 c^2 + \dots$$

be the so-called virial (or density) expansion of  $\kappa^*$  in powers of the volume fraction  $c$  of the inclusions. As a matter of fact, the coefficient  $a_1$  is the only thing rigorously calculated by J. Maxwell (cf. [7]):  $a_1 = d\beta_d$ , while for the  $c^2$ -coefficient his formula yields only a certain approximation

$$(1.5) \quad a_2 = d\beta_d^2.$$

The rigorous evaluation of  $a_2$  has attracted the attention of many authors because this is the simplest case in which the multiparticle interaction shows up in a non-trivial way (see, e.g., the papers [4-6], [10]), where  $a_2$  has been expressed in a closed form, making use of the zero-density limit  $g_0(r)$  of the so-called radial distribution function for the spheres, and of the one- and two-inclusion fields for the conductivity problem under study. Let us point out also the paper [8], where certain bounds on  $a_2$  are derived in which the same function  $g_0(r)$  appears; the counterpart of these bounds in  $2D$ -case is given in [9].

In this paper we shall first concentrate on the functional dependence of  $a_2$  upon the above mentioned function  $g_0(r)$ . We shall show in §2, using the bounds of [8,9], that  $a_2$  is a sum of a constant and a linear functional of  $g_0(r)$  with a certain kernel  $\Phi_1$ , and estimates on  $\Phi_1$  will be then proposed (§3). In §4 we shall evaluate  $\Phi_1$  analytically in the  $2D$ -case, making use of a method originated by J. Peterson and J. Hermans [10]. In this way we avoid twin expansion technique of D. Jeffrey [4] and B. Felderhof *et al.* [5], needed in  $3D$ -case when solving the two-sphere problem, and get the eventual  $2D$ -case result for  $a_2$  in an explicit integral form. Moreover, for some simple but important particular cases the integration can be performed analytically employing certain well-known higher transcendental functions. Finally we consider some power series expansions for  $a_2$  which allow us to calculate the latter easily (§5).

## 2. FUNCTIONAL DEPENDENCE OF $a_2$ UPON THE RADIAL DISTRIBUTION FUNCTION

Due to the assumption  $f_p \sim n^p$ , the coefficient  $a_2$  could depend on the two-point distribution density  $f_2$  only. As usual, we represent the latter as  $f_2(\mathbf{y}_1, \mathbf{y}_2) = f_2(r) = n^2 g(r) = n^2 g_0(r) + o(n^2)$ , where  $g(r)$  is the radial distribution function and  $g_0(r)$  is its zero-density limit,  $r = |\mathbf{y}_1 - \mathbf{y}_2|$ . Obviously, only  $g_0(r)$  could influence  $a_2$ , so that

$$(2.1) \quad a_2 = \mathfrak{F}[g_0(\cdot)].$$

The functional  $\mathfrak{F}$  is defined on the space  $\tilde{C}$  of all bounded, piece-wise continuous functions on the interval  $[2, \infty)$ ,  $g_0(\lambda a)$ ,  $\lambda = r/a$  (due to the nonoverlapping assumption) and  $g_0(r) \rightarrow 1$  at  $r \rightarrow \infty$  (no long-range order in the dispersion). The continuity of this functional in the  $C$ -norm seems obvious so that, according to the general representation theorem of V. Volterra [11], we can write down  $a_2$  in the

form of a functional Volterra series:

$$(2.2) \quad a_2 = \Phi_0 + \int_2^\infty \Phi_1(\lambda) g_0(\lambda a) d\lambda \\ + \int_2^\infty \int_2^\infty \Phi_2(\lambda_1, \lambda_2) g_0(\lambda_1 a) g_0(\lambda_2 a) d\lambda_1 d\lambda_2 + \dots,$$

where  $\Phi_0 = \text{const}$  and  $\Phi_1(\lambda)$ ,  $\Phi_2(\lambda_1, \lambda_2)$ , etc., are certain kernels that vanish at infinity. These kernels do not depend on the statistics of the dispersions but only on the ratio  $\alpha = \kappa_f/\kappa_m$  of the constituent conductivities or, which is the same, on the parameters  $\beta_d$ , introduced in (1.3b); to emphasize this fact we shall use the notations  $\Phi_1 = \Phi_1(\lambda; \beta_d)$ , etc.

Let us recall now the bounds on  $a_2$ , derived in [8,9] in the 3D- and 2D-cases respectively:

$$(2.3a) \quad d\beta_d^2 \left( 1 + \frac{d\beta_d}{1 - (d-1)\beta_d} m_2 \right) \leq a_2 \leq d\beta_d^2 \left( 1 + \frac{d\beta_d}{1 + \beta_d} m_2 \right),$$

$$(2.3b) \quad m_2 = (d-1) \int_2^\infty \frac{\lambda^{d-1}}{(\lambda^2 - 1)^d} g_0(\lambda a) d\lambda, \quad d = 2, 3.$$

As a first consequence of (2.3) we shall show that the functional (2.1) has the form

$$(2.4) \quad a_2 = d\beta_d^2 + \int_2^\infty \Phi_1(\lambda; \beta_d) g_0(\lambda a) d\lambda,$$

i.e.

$$(2.4a) \quad \Phi_0 = d\beta_d^2$$

and

$$(2.4b) \quad \Phi_2 = \Phi_3 = \dots = 0.$$

The proof is based on the fact that (2.3) holds for all admissible functions  $g_0(r) \in \tilde{C}$ . Indeed, consider the class of functions  $g_0^A \in \tilde{C}$  such that  $g_0^A(r) = 0$  at  $r \leq A$  and  $g_0^A(r) = 1$  at  $r > A$ ,  $A > 2a$ . The statistical parameter in (2.3b) depends then on  $A$ ,  $m_2 = m_2^A$ , and it can be easily calculated in this case, but we need here only the obvious fact that

$$(2.5) \quad m_2^A \rightarrow 0 \quad \text{at} \quad A \rightarrow \infty.$$

On the other hand,

$$(2.6) \quad \begin{aligned} \phi_1^A &= \int_2^\infty \Phi_1(\lambda) g_0^A(\lambda a) d\lambda \rightarrow 0, \\ \phi_2^A &= \int_2^\infty \int_2^\infty \Phi_2(\lambda_1, \lambda_2) g_0^A(\lambda_1 a) g_0^A(\lambda_2 a) d\lambda_1 d\lambda_2 \rightarrow 0, \end{aligned}$$

etc., at  $A \rightarrow \infty$ . We employ now (2.3a) for the function  $g_0(r) = g_0^A(r)$ :

$$d\beta_d^2 \left( 1 + \frac{d\beta_d}{1 - (d-1)\beta_d} m_2^A \right) \leq \Phi_0 + \phi_1^A + \phi_2^A + \dots \leq d\beta_d^2 \left( 1 + \frac{d\beta_d}{1 + \beta_d} m_2^A \right).$$

Letting  $A \rightarrow \infty$  and recalling (2.5) and (2.6), we get from the last inequalities that  $\Phi_0 = d\beta_d^2$  which proves (2.4a).

The proof of (2.4b) is very simple if the functional series (2.2) is finite, containing  $N$  terms,  $N \geq 2$ . Let  $N = 2$  first. Consider the kernel  $\Phi_2$  and suppose that in the neighbourhood

$$\Lambda_\varepsilon = (\lambda_1^0 - \varepsilon, \lambda_1^0 + \varepsilon) \times (\lambda_2^0 - \varepsilon, \lambda_2^0 + \varepsilon)$$

of the point  $(\lambda_1^0, \lambda_2^0) \in \mathbb{R}^2$  we have, say,  $\Phi_2(\lambda_1, \lambda_2) > 0$ . We consider the class of step-constant functions  $g_0(r) \in \tilde{C}$ , such that  $g_0(r) = \mu$  if  $r \in (\lambda_1^0 - \varepsilon, \lambda_1^0 + \varepsilon) \cup (\lambda_2^0 - \varepsilon, \lambda_2^0 + \varepsilon)$ ;  $g_0(r) = 1$  at  $r \geq A$  and vanishes otherwise. In this case the parameter  $m_2$  is a linear function of  $\mu$ . On the other hand, the two-tuple term in (2.2) is a quadratic function of  $\mu$  with a positive multiplier of  $\mu^2$ . If  $\mu$  and  $A$  are big enough, the inequality (2.3a) will be violated, which proves that  $\Phi_2 = 0$ . The proof in the case when  $N > 2$  but is finite, is fully similar.

We should finally show that the series (2.2) for  $a_2$  is finite. To this end it suffices to recall the definition (1.2) and the representations

$$\begin{aligned} \kappa(\mathbf{x}) &= \langle \kappa \rangle + [\kappa] \int h(\mathbf{x} - \mathbf{y}) \omega'(\mathbf{y}) d^3 \mathbf{y}, \\ \theta(\mathbf{x}) &= \mathbf{G} \cdot \mathbf{x} + \int T_1(\mathbf{x} - \mathbf{y}) \omega'(\mathbf{y}) d^3 \mathbf{y} \\ &+ \iint T_2(\mathbf{x} - \mathbf{y}_1, \mathbf{x} - \mathbf{y}_2) D_\omega^{(2)}(\mathbf{y}_1, \mathbf{y}_2) d^3 \mathbf{y}_1 d^3 \mathbf{y}_2 + o(n^2), \end{aligned}$$

where  $\omega'(\mathbf{x}) = \omega(\mathbf{x}) - n$ ,

$$\omega(\mathbf{x}) = \sum_j \delta(\mathbf{x} - \mathbf{x}_j)$$

is the random density field for the dispersion and

$$D_\omega^{(2)}(\mathbf{y}_1, \mathbf{y}_2) = \omega(\mathbf{y}_1) [\omega(\mathbf{y}_2) - \delta(\mathbf{y}_{1,2}) - n g_0(\mathbf{y}_{1,2})] [\omega'(\mathbf{y}_1) + \omega'(\mathbf{y}_2)] - n^2 g_0(\mathbf{y}_{1,2}),$$

$\mathbf{y}_{1,2} = \mathbf{y}_1 - \mathbf{y}_2$ . The kernels  $T_1$  and  $T_2$  have been specified in [6], but we need here only the fact that the two- and three-point moments of  $\omega(\mathbf{x})$  depend linearly

on  $g_0(r)$ , to the needed order  $n^2$  [12], so that the series (2.2) should be finite and, moreover, should indeed have the form (2.2), truncated after the one-tuple term.

### 3. BOUNDS ON THE KERNEL $\Phi_1$

Let us denote by  $a'_2$  the  $c^2$ -deviation of  $a_2$  from its Maxwell value (1.5), i.e.  $a'_2 = a_2 - d\beta_d^2$ . From (2.3) and (2.4) we have

$$(3.1) \quad a'_2 = \int_2^\infty \Phi_1(\lambda; \beta_d) g_0(\lambda a) d\lambda,$$

$$(3.2) \quad \frac{d^2 \beta_d^3}{1 - (d-1)\beta_d} m_2 \leq a'_2 \leq \frac{d^2 \beta_d^3}{1 + \beta_d} m_2.$$

Since the statistical parameter  $m_2$  is a linear functional of  $g_0(\lambda a)$  and (3.2) should hold for all admissible functions  $g_0 \in \tilde{C}$ , we can conclude that

$$(3.3) \quad \begin{aligned} & \frac{d^2(d-1)\beta_d^3}{1 - (d-1)\beta_d} \frac{\lambda^{d-1}}{(\lambda^2 - 1)^d} \leq \Phi_1(\lambda; \beta_d) \\ & \leq \frac{d^2(d-1)\beta_d^3}{1 + \beta_d} \frac{\lambda^{d-1}}{(\lambda^2 - 1)^d}, \quad \lambda \in [2, \infty). \end{aligned}$$

The proof employs the arbitrariness of  $g_0(\lambda a)$  in the space  $\tilde{C}$  and is fully similar to that in §2.

Note that the estimates (3.3) imply that  $\Phi_1$  decays as  $\lambda^{-(d+1)}$  at  $\lambda \rightarrow \infty$  and

$$(3.4) \quad \Phi_1(\lambda; \beta_d) = d^2(d-1)\beta_d^3 \frac{\lambda^{d-1}}{(\lambda^2 - 1)^d} + o(\beta_d^3).$$

If  $\kappa_f/\kappa_m \rightarrow \infty$ , i.e.  $\beta_d \rightarrow 1$ , the upper bound (3.3) degenerates; if  $\kappa_f/\kappa_m \rightarrow 0$ , i.e.  $\beta_3 \rightarrow -\frac{1}{2}$  or  $\beta_2 \rightarrow -1$ , the lower bound (3.3) degenerates (cf. Fig. 2 below).

### 4. EVALUATION OF THE KERNEL $\Phi_1$ IN 2D-CASE

Let us recall first the formula for  $a'_2$ , derived in [4,10], see also [6], which in the 2D-case reads

$$(4.1) \quad a'_2 \mathbf{G} = \frac{[\kappa]}{\kappa_m} \frac{1}{S_a^2} \int_{S_a} d^2 \mathbf{x} \int_{Z_{2a}} g_0(\mathbf{z}) \left[ \nabla_x T^{(2)}(\mathbf{x}; \mathbf{z}) - \nabla T^{(1)}(\mathbf{x}) \right] d^2 \mathbf{z},$$

where  $Z_{2a} = \{ \mathbf{z} \mid |\mathbf{z}| \geq 2a \} \subset \mathbb{R}^2$ , and

$$(4.2) \quad T^{(1)}(\mathbf{x}) = -\beta \mathbf{G} \cdot \mathbf{x} \quad \text{at } |\mathbf{x}| \leq a; \quad \beta = \beta_2 = \frac{[\kappa]}{\kappa_f + \kappa_m},$$

is the solution of one-inclusion problem at  $|\mathbf{x}| \leq a$ ; the inclusion hereafter is the disc  $S_a = \{ \mathbf{x} \mid |\mathbf{x}| \leq a \}$  of radius  $a$ , located at the origin. The field  $T^{(2)}(\mathbf{x}; \mathbf{z})$  is the solution of the two-inclusion problem which represents the disturbance to the temperature field introduced by the pair of equal discs *1* and *2* centered at the origin and at the point  $\mathbf{z}$ , respectively, when the temperature gradient at infinity equals  $\mathbf{G}$ . The field  $T^{(2)}(\mathbf{x}; \mathbf{z})$  satisfies the equation

$$(4.3) \quad \kappa_m \Delta T^{(2)}(\mathbf{x}; \mathbf{z}) + [\kappa] \nabla \cdot \left\{ [h(\mathbf{x}) + h(\mathbf{x} - \mathbf{z})] [\mathbf{G} + \nabla T^{(2)}(\mathbf{x}; \mathbf{z})] \right\} = 0;$$

here  $\mathbf{z}$  plays the role of a parameter and  $\mathbf{z} \in Z_{2a}$ , since the discs are not allowed to overlap. The integral in (4.1) is conditionally convergent and is understood in the sense

$$(4.4) \quad \int \cdot d^2 \mathbf{z} = \lim_{R \rightarrow \infty} \int_{Z_{2a,R}} \cdot d^2 \mathbf{z}; \quad \int_{Z_{2a,R}} \cdot d^2 \mathbf{z} = \int_0^R r dr \int_{\Omega} \cdot d\Omega,$$

where  $Z_{2a,R} = \{ \mathbf{z} \mid 2a \leq |\mathbf{z}| \leq R \}$ . This means that in the integral over the region  $Z_{2a,R}$  we first integrate with respect to the angular coordinates, i.e. on the unit circle  $\Omega = \{ \mathbf{z} \mid |\mathbf{z}| = 1 \}$ , and then with respect to the radial coordinate  $r = |\mathbf{z}|$ , see [4,6].

We shall calculate here this integral by means of an obvious extension of the arguments of J. Peterson and J. Hermans [10], who tacitly considered only the well-stirred case  $g_0(r) = 1$ .

Let us introduce the tilted coordinate system  $(x'_1, x'_2)$  as shown in Fig. 1, where  $|O'O_1| = |O'O_2| = L$ , and the bipolar coordinate system  $(\sigma, \tau)$  for which

$$(4.5) \quad x'_1 = b \frac{\text{sh } \tau}{\text{ch } \tau - \cos \sigma} = b \left( 1 + 2 \sum_{p=1}^{\infty} e^{-p\tau} \cos p\sigma \right),$$

$$x'_2 = b \frac{\sin \sigma}{\text{ch } \tau - \cos \sigma} = 2b \sum_{p=1}^{\infty} e^{-p\tau} \sin p\sigma.$$

The boundaries of the two discs *1* and *2* correspond to the coordinate lines  $\tau = \pm \tau_0$ , where

$$(4.6) \quad a = \frac{b}{\text{sh } \tau_0}, \quad L = a \text{ch } \tau_0.$$

The solution of the problem (4.3), bounded at infinity can be obtained straightforwardly, making use of the bipolar coordinates  $(\sigma, \tau)$  (see, e.g., [10]). We shall need in what follows only the values of the solution at the boundary  $\tau = \tau_0$  of the disc *1*:

$$(4.7) \quad \left( \mathbf{G} \cdot \mathbf{x} + T^{(2)}(\mathbf{x}; \mathbf{z}) \right) \Big|_{\tau=\tau_0}$$

$$= bG'_1 \left( 1 + \sum_{p=0}^{\infty} \frac{2\kappa_m \cos p\sigma}{\kappa_f \text{sh } p\tau_0 + \kappa_m \text{ch } p\tau_0} \right) + bG'_2 \sum_{p=1}^{\infty} \frac{2\kappa_m \sin p\sigma}{\kappa_f \text{ch } p\tau_0 + \kappa_m \text{sh } p\tau_0},$$

where  $G'_1$  and  $G'_2$  are the projections of the temperature gradient at infinity  $\mathbf{G}$  on the axes  $x'_1$  and  $x'_2$ , respectively.

As it follows from (4.2),

$$(4.8) \quad T^{(1)}(\mathbf{x}) = -\beta \mathbf{G} \cdot \mathbf{x} = -\beta(G'_1 x'_1 + G'_2 x'_2) + \text{const},$$

so that the field  $W(\mathbf{x}; \mathbf{z}) = T^{(2)}(\mathbf{x}; \mathbf{z}) - T^{(1)}(\mathbf{x})$ , needed in (4.1), has the form

$$W(\mathbf{x}; \mathbf{z}) = G'_1 W'_1 + G'_2 W'_2,$$

$$(4.9) \quad W'_1 = \sum_{p=0}^{\infty} W'_{p1} \cos p\sigma, \quad W'_2 = \sum_{p=1}^{\infty} W'_{p2} \sin p\sigma,$$

$$W'_{p1} = \frac{2\kappa_m \beta b e^{-2p\tau_0}}{\kappa_f \text{sh } p\tau_0 + \kappa_m \text{ch } p\tau_0}, \quad W'_{p2} = -\frac{2\kappa_m \beta b e^{-2p\tau_0}}{\kappa_f \text{ch } p\tau_0 + \kappa_m \text{sh } p\tau_0}.$$

Let us change now the order of integration in (4.1) and then apply the Gauss theorem

$$(4.10) \quad a'_2 \mathbf{G} = \frac{[\kappa]}{\kappa_m} \frac{1}{S_a^2} \int_{Z_{2a}} g_0(\mathbf{z}) d^2 \mathbf{z} \int_{|x|=a} \mathbf{n} W(\mathbf{x}; \mathbf{z}) ds;$$

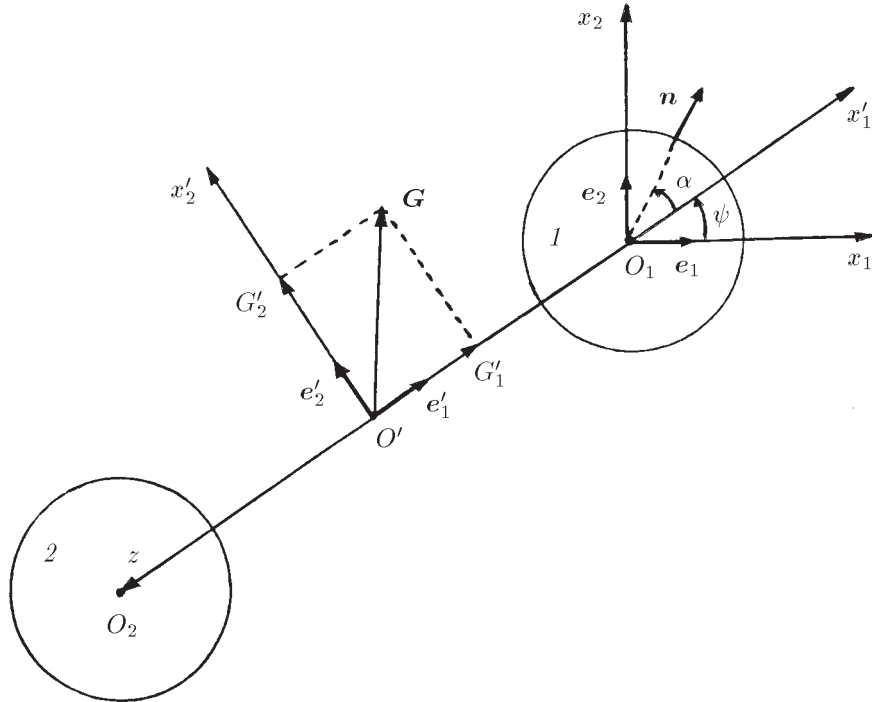


Fig. 1. Coordinate systems in the two-inclusion problem (2D-case).



here  $\mathbf{n}$  is the unit outward normal to the disk  $l$  and  $ds$  is its element of length

$$(4.11) \quad ds = \frac{d\sigma}{h}, \quad h = \frac{b}{\operatorname{ch} \tau_0 - \cos \sigma}.$$

Since the integral with respect to  $\mathbf{z}$  is to be understood in the sense (4.4) and  $g_0(\mathbf{z}) = g_0(|\mathbf{z}|)$ , we carry out the integration consecutively: first, at fixed  $|\mathbf{z}| = 2L = 2a \operatorname{ch} \tau_0$ , i.e. at fixed  $\tau_0$ , we integrate with respect to all orientations of the dumb-bell shaped figure (see Fig. 1), described by the angle  $\psi$ . Next we integrate with respect to all  $|\mathbf{z}|$ , i.e. to all  $\tau_0$ . This procedure is equivalent to a transition to the polar coordinates  $(\rho, \alpha)$  in the plane  $(x_1, x_2)$  with a center at the point  $O_1$ , so that  $\rho = |\mathbf{z}| = 2L$ , after which the integration is performed first with respect to  $\alpha$  and then with respect to  $\rho$  (cf. Fig. 1).

Consider first the integration with respect to  $\rho$ . Due to (4.9)<sub>1</sub> and (4.11), we have

$$(4.12) \quad \begin{aligned} \mathbf{K}(L) &= \int_{-\pi}^{\pi} d\alpha \int_{|x|=a} \mathbf{n} W(\mathbf{x}; a) ds \\ &= \int_{-\pi}^{\pi} d\alpha \int_{|x|=a} \mathbf{n} (G'_1 W'_1 + G'_2 W'_2) ds = \int_{-\pi}^{\pi} d\alpha \int_{|x|=a} (W'_1 \mathbf{n} \mathbf{e}'_1 + W'_2 \mathbf{n} \mathbf{e}'_2) ds \cdot \mathbf{G}. \end{aligned}$$

In this expression we should once integrate over the orientations of the pair of unit vectors  $\mathbf{e}'_1, \mathbf{e}'_2$  and once over the orientations of the normal  $\mathbf{n}$ . Instead, we first fix the angle  $\psi$  between  $\mathbf{n}$  and  $\mathbf{e}'_1$ :

$$(4.13) \quad \mathbf{n} \cdot \mathbf{e}'_1 = \cos \psi, \quad \mathbf{n} \cdot \mathbf{e}'_2 = \sin \psi,$$

and rotate rigidly the triad  $\mathbf{e}'_1, \mathbf{e}'_2, \mathbf{n}$ . The dyadics  $\mathbf{n} \mathbf{e}'_1, \mathbf{n} \mathbf{e}'_2$ , after such an integration become proportional to the unit second-rank tensor  $\mathbf{I}$ , so that, in virtue of (4.11) and (4.13), the integral in (4.12) becomes

$$(4.14) \quad \mathbf{K}(L) = \pi \mathbf{G} \int_{-\pi}^{\pi} (G'_1 W'_1 + G'_2 W'_2) \frac{d\sigma}{h}.$$

It remains to integrate with respect to the angle  $\psi$  only.

Let us recall now the formulas

$$(4.15) \quad \begin{aligned} \frac{\cos \psi}{h} &= b \frac{\operatorname{ch} \tau_0 \cos \sigma - 1}{(\operatorname{ch} \tau_0 - \cos \sigma)^2} = 2b \sum_{p=1}^{\infty} p e^{-p\tau_0} \cos p\sigma, \\ \frac{\sin \psi}{h} &= b \frac{\operatorname{sh} \tau_0 \sin \sigma}{(\operatorname{ch} \tau_0 - \cos \sigma)^2} = 2b \sum_{p=1}^{\infty} p e^{-p\tau_0} \sin p\sigma, \end{aligned}$$

which, when substituted into (4.14), together with (4.9) yield

$$\begin{aligned}
\mathbf{K}(L) &= \pi b \mathbf{G} \sum_{p=1}^{\infty} p e^{-p\tau_0} (W'_{p1} + W'_{p2}) \\
(4.16) \quad &= 16\pi^2 b^2 \beta^2 \frac{\kappa_m}{\kappa_m + \kappa_f} \mathbf{G} \sum_{p=1}^{\infty} \frac{p e^{-6p\tau_0}}{1 - \beta^2 e^{-4p\tau_0}}.
\end{aligned}$$

Since the radius  $a$  of the discs is fixed, the integration with respect to  $\rho = 2a \operatorname{ch} \tau_0$  is an integration over  $\tau_0 \in (0, \infty)$  and

$$\rho d\rho = 4a^2 \operatorname{ch} \tau_0 \operatorname{sh} \tau_0 d\tau_0.$$

Making use of (4.7) and (4.16), we thus get

$$\begin{aligned}
a'_2 \mathbf{G} &= \frac{[\kappa]}{\kappa_m} \frac{1}{S_a^2} \int_{2a}^{\infty} \mathbf{K}(L) g_0(\rho) \rho d\rho = 4\beta^3 M(\beta) \mathbf{G}, \\
(4.17) \quad & \\
M(\beta) &= 16 \sum_{p=1}^{\infty} p \int_0^{\infty} g_0(2a \operatorname{ch} \tau_0) \frac{\operatorname{ch} \tau_0 \operatorname{sh}^3 \tau_0}{1 - \beta^2 e^{-4p\tau_0}} e^{-6p\tau_0} d\tau_0.
\end{aligned}$$

Upon inserting (4.17) into (4.1) we easily obtain the eventual  $c^2$ -formula for the effective transverse conductivity  $\kappa^*$  of a fiber-reinforced material:

$$(4.18) \quad \frac{\kappa^*}{\kappa_m} = 1 + 2\beta c + 2\beta^2 (1 + 2\beta M(\beta)) c^2 + o(c^2),$$

with the function  $M(\beta)$  defined in (4.17). This function is obviously even, which implies the relation

$$(4.19) \quad a_2(\beta) + a_2(-\beta) = 4\beta^2$$

for the coefficient  $a_2$ , considered as a function of the parameter  $\beta$ . It is to be noted that (4.19) is a simple consequence of the Keller interchange formula [13], which reads

$$\kappa^*(\kappa_f, \kappa_m) \kappa^*(\kappa_m, \kappa_f) = \kappa_f \kappa_m;$$

here  $\kappa^*(\kappa_f, \kappa_m)$  denotes the effective transverse conductivity of the fiber material under study and  $\kappa^*(\kappa_m, \kappa_f)$  is the conductivity of the same material, but when the fibers are made of the matrix material and the matrix—of fiber's.

The comparison of (3.1) and (4.17) yields the analytical form of the kernel  $\Phi_1$ :

$$(4.20) \quad \Phi_1(\lambda; \beta) = 4\beta^3 \lambda (\lambda^2 - 4) \sum_{p=1}^{\infty} \frac{p \Lambda^{6p}}{1 - \beta^2 \Lambda^{6p}};$$

here  $\Lambda = e^{-\tau_0} = \frac{1}{2}(\lambda - \sqrt{\lambda^2 - 4})$ ,  $\lambda \geq 2$ .

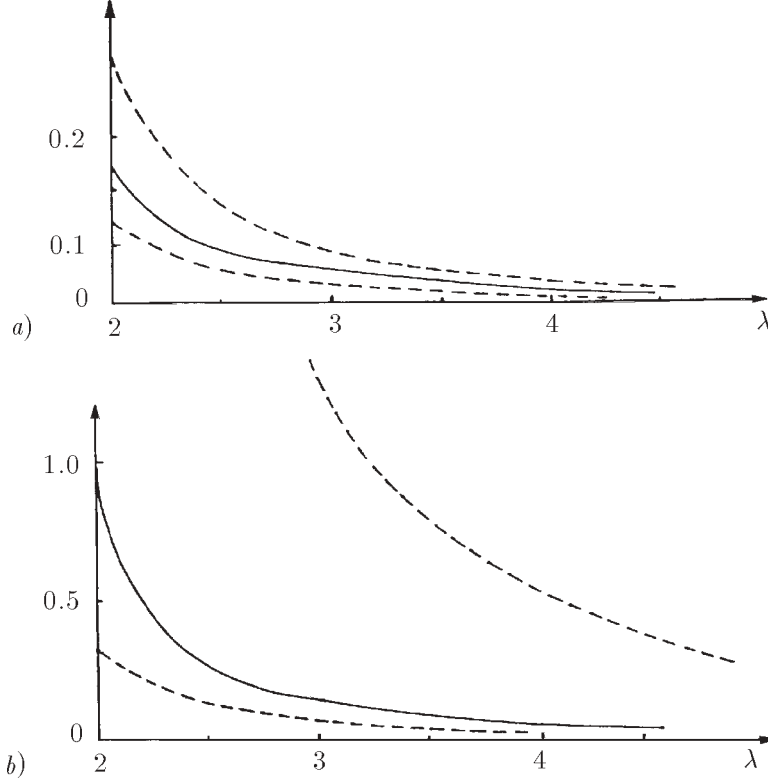


Fig. 2. Plots of the kernel  $\Phi_1$  (continuous line) and the bounds  $\Phi_1^L$  and  $\Phi_1^U$  (dashed lines) (2D-case); a)  $\beta = 0.5$ ; b)  $\beta = 0.9$

By means of (2.4) and (4.20) we can evaluate numerically the coefficient  $a_2$  for an arbitrary sphere statistics, represented here by the function  $g_0(r)$ . Thus in 2D-case under study we avoid twin expansion technique of D. Jeffrey [4] and B. Felderhof *et al.* [5], needed in 3D-case when solving the two-sphere problem, and get the eventual result for  $a_2$  as an explicit integral. Moreover, for some simple but important particular cases the integration can be performed analytically employing certain well-known higher transcendental functions, as we shall see in the next Section.

The bounds (3.3) in 2D-case under study have the form

$$\Phi_1^L(\lambda; \beta) \leq \Phi_1(\lambda; \beta) \leq \Phi_1^U(\lambda; \beta),$$

$$(4.21) \quad \Phi_1^L = \frac{4\beta^3}{1 + \beta} \frac{\lambda}{(\lambda^2 - 1)^2}, \quad \Phi_1^U = \frac{4\beta^3}{1 - \beta} \frac{\lambda}{(\lambda^2 - 1)^2}.$$

The exact values of the kernel  $\Phi_1$  together with the bounds  $\Phi_1^L$  and  $\Phi_1^U$  as functions of  $\lambda$  are shown in Fig. 2 in two cases:  $\beta = 0.5$  and  $\beta = 0.9$ .

## 5. SOME FORMULAS CONCERNING $a_2$ IN 2D-CASE

In order to make easier the numerical evaluation of  $a_2$  for the fiber-reinforced materials let us expand the function  $M(\beta)$  in (4.17) in powers of the parameter  $\beta$ :

$$(5.1) \quad M(\beta) = \sum_{k=0}^{\infty} M_k \beta^{2k}, \quad \beta = \frac{[\kappa]}{\kappa_f + \kappa_m},$$

$$(5.2) \quad M_k = 16 \sum_{j=1}^{\infty} j \int_0^{\infty} g_0(2a \operatorname{ch} \tau) \operatorname{ch} \tau \operatorname{sh}^3 \tau e^{-2j(3+2k)\tau} d\tau.$$

The estimates (3.2) for  $a_2$  now imply

$$(5.3) \quad \frac{2\beta}{1+\beta} m_2 \leq 2\beta M(\beta) \leq \frac{2\beta}{1-\beta} m_2,$$

so that

$$(5.4) \quad M_0 = m_2 = \int_2^{\infty} \frac{\lambda}{(\lambda^2 - 1)^2} g_0(\lambda a) d\lambda,$$

$$(5.5) \quad M(\beta) = m_2 + O(\beta), \quad \text{i.e. } a_2 = 2\beta^2(1 + 2\beta m_2) + o(\beta^3).$$

The formula (5.2) can be recast as

$$M_k = 4 \int_0^{\infty} g_0(2a \operatorname{ch} \tau) \frac{\operatorname{ch} \tau \operatorname{sh}^3 \tau}{\operatorname{sh}^2(3+2k)\tau} d\tau.$$

Having used the known formula for  $\operatorname{sh} n\tau$  and making the substitution  $\lambda = 2 \operatorname{ch} \tau$ , we get eventually

$$(5.6) \quad M_k = \int_2^{\infty} \lambda g_0(\lambda a) \left\{ \sum_{j=0}^{k+1} (-1)^j C_{2+2k-j}^j \lambda^{2(2k-j+1)} \right\}^{-1} d\lambda.$$

The formula (5.4) coincides with (5.6) at  $k = 0$ . At  $k = 1$  we have

$$(5.7) \quad M_1 = \int_2^{\infty} \frac{\lambda g_0(\lambda a)}{(\lambda^4 - 3\lambda^2 + 1)^2} d\lambda,$$

and this integral, as well as the integral in (5.4), can be easily evaluated in the most frequently used well-stirred approximation for which  $g(r) = g_0(r) = g_0(\lambda a) = 1$  at  $\lambda \geq 2$ , yielding

$$(5.8) \quad M_0 = M_0^{ws} = \frac{1}{6}, \quad M_1 = M_1^{ws} = \frac{1}{10} + \frac{\sqrt{5}}{25} \ln \frac{3 - \sqrt{5}}{2}.$$

However, the analytical evaluation of  $M_k$  by means of (5.6) at  $k \geq 2$  is not easy even in the well-stirred case. In the latter case we employ (5.2) which leads, after simple manipulations, to the following result:

$$(5.9a) \quad M_k^{ws} = c_k^2 \left\{ 2\psi(1+c_k) - 2\psi(1+2c_k) + \frac{1}{c_k} \left( \frac{2\pi c_k}{\sin 2\pi c_k} - 1 \right) \right\},$$

where

$$(5.9b) \quad c_k = \frac{1}{2k+3}, \quad \psi(x) = \frac{\Gamma'(x)}{\Gamma(x)},$$

so that  $\psi(x)$  is the logarithmic derivative of the Euler Gamma-function which is investigated in detail and tabulated [14,15]. As a matter of fact, the formula (5.9) is given in [10]. Note that since the arguments  $1+c_k$  and  $1+2c_k$  are rational, we can employ the formula for  $\psi(p/q)$ , cf. [15, p.722], which allows us to represent  $M_k^{ws}$  by means of elementary functions, namely

$$(5.10) \quad M_k^{ws} = c_k^2 \left\{ \frac{1}{2c_k} + 8 \sum_{j=1}^{k+1} \sin(j\pi c_k) \sin(3j\pi c_k) \ln \sin(j\pi c_k) \right\}.$$

Note also the asymptotic formula

$$(5.11) \quad M_k^{ws} = 6\zeta(3)c_k^4 + 30\zeta(5)c_k^6 + o(c_k^6),$$

where  $\zeta(3) = 1.2021$  and  $\zeta(5) = 1.0369$  are the respective values of the Riman  $\zeta$ -function. The formula (5.11) gives four correct decimal numbers for  $M_k^{ws}$  at  $k \geq 4$  and six at  $k \geq 6$ .

The formulas (5.10) and (5.11) make possible to evaluate  $a_2$  in the well-stirred case, having truncated the series (5.1) and replacing the remaining coefficients  $M_k^{ws}$  with their asymptotic values (5.11). In this way one easily finds, e.g.,

$$(5.12) \quad \begin{aligned} a_2^{ws} &= 2.7450 \quad \text{at } \beta = 1, \quad \text{i.e. } \kappa_f/\kappa_m = \infty, \\ a_2^{ws} &= 1.2550 \quad \text{at } \beta = -1, \quad \text{i.e. } \kappa_f/\kappa_m = 0. \end{aligned}$$

The dependence  $a_2 = a_2(\beta)$  is shown in Fig. 3 together with the bounds (2.3a), which in the well-stirred  $2D$ -case under study read

$$2\beta^2 \left( 1 + \frac{\beta}{3(1+\beta)} \right) \leq a_2^{ws} \leq 2\beta^2 \left( 1 + \frac{\beta}{3(1-\beta)} \right).$$

It is instructive to consider as well the more general radial distribution function

$$(5.13) \quad g_0(r) = 1 + A_1 \frac{a}{r}, \quad r \geq 2a,$$

where  $A_1$  is a certain scalar parameter such that  $A_1 \geq -2$  (in order to have  $g_0(r) \geq 0$ ). The coefficients  $M_k$ , corresponding to the distribution function (5.13) can be easily evaluated by means of (5.2) and the final result is

$$(5.14) \quad M_k = M_k^{ws} + A_1 N_k,$$

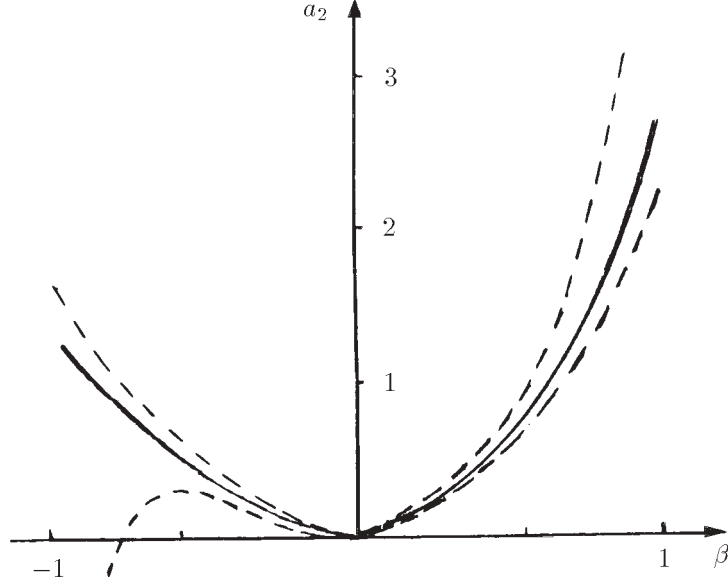


Fig. 3. The  $c^2$ -coefficient  $a_2$  in the well-stirred  $2D$ -case as a function of  $\beta$ .

$$(5.15) \quad N_k = \frac{3}{2}c_k^2 \left\{ 2\psi\left(1 + \frac{1}{2}c_k\right) - \psi\left(1 + \frac{3}{2}c_k\right) + \frac{\pi \cos(\pi c_k/2)}{\sin(3\pi c_k/2)} - \frac{2}{3c_k} \right\},$$

$k = 0, 1, \dots$ , where  $c_k$  are defined in (5.9b) and  $M_k^{ws}$  are the respective coefficients in (5.1) in the well-stirred case, cf. (5.10). Having applied the above mentioned formula for  $\psi(p/q)$  from [15, p. 722], we get

$$(5.16) \quad N_k = \frac{3}{2}c_k^2 \left\{ \frac{2}{3c_k} + 4 \sum_{j=1}^{2(k+1)} \sin(j\pi c_k) \sin(2j\pi c_k) \ln \sin \frac{j\pi c_k}{2} \right\},$$

so that  $N_k > 0$ ,  $k = 0, 1, \dots$

In particular,

$$N_0 = \frac{1}{3} - \frac{1}{4} \ln 3, \quad N_1 = \frac{1}{5} + \frac{9\sqrt{5}}{100} \ln \frac{3 - \sqrt{5}}{2}.$$

The asymptotic formula for  $N_k$  reads

$$N_k = 3\zeta(3)c_k^4 + o(c_k^4).$$

It gives four correct decimal digits at  $k \geq 2$  and six at  $k \geq 8$ .

Since  $A_1$  should only exceed  $-2$  and thus it can take arbitrarily big values, equation (5.14) suggests that the statistics of the dispersion affects very strongly the  $c^2$ -coefficient in the virial expansion (1.4) of the effective conductivity. This is illustrated in Fig. 4 for the radial distribution function (5.13) in the cases  $A_1 = -2$ ,  $A_1 = 0$  (well-stirred) and  $A_1 = 5$ .

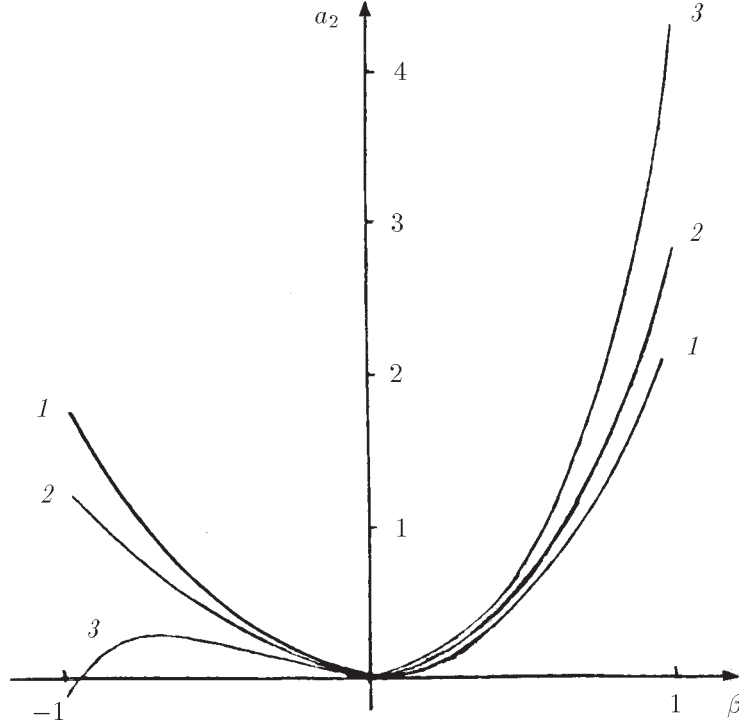


Fig. 4. The  $c^2$ -coefficient  $a_2$  in the well-stirred  $2D$ -case as a function of  $\beta$  for the distribution function  $g_0(r)$  given in (5.13);  
 1 —  $A_1 = -2$ ; 2 —  $A_1 = 0$  (well-stirred); 3 —  $A_1 = 5$

Let us note finally that  $M(\beta) > 0$  at  $\beta \in (-1, 1)$ , cf. (4.17), and it could take arbitrarily big values, e.g. for the distribution function (5.13). Then (4.19) implies the following sharp estimates for the coefficient  $a_2$  in  $2D$ -case:

$$(5.17) \quad \begin{aligned} 2\beta^2 < a_2 < \infty, & \text{ if } \beta > 0, \text{ i.e. } \kappa_f > \kappa_m, \\ -\infty < a_2 < 2\beta^2, & \text{ if } \beta < 1, \text{ i.e. } \kappa_f < \kappa_m, \end{aligned}$$

having taken  $\sup a_2$  and  $\inf a_2$  with respect to all admissible radial distribution functions  $g_0(r)$  (so that varying, in particular the parameter  $A_1$  in (5.13) from  $-2$  to infinity). We can thus conclude that there is no finite interval, independent of the statistics of the fibres, within which the  $c^2$ -coefficient  $a_2$  is to be always found. Note that similar to (5.17) estimates are to be expected to hold in the  $3D$ -case, i.e. for dispersions of spheres, with the only difference that the factor 2 should be replaced by 3, and again there will be no finite interval for the coefficient  $a_2$ , independent of the statistics of the dispersion.

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