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A NEW APPROACH FOR DERIVING C^2-BOUNDS ON THE EFFECTIVE CONDUCTIVITY OF RANDOM DISPERSIONS

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A new variational procedure for evaluating the effective conductivity of a dilute random dispersion of spheres is proposed. The classical variational principles are employed, in which a class of trial fields in the form of suitably truncated factorial series is introduced. In general, this class leads to a rigorous formula for the effective conductivity, which is correct to the order "square of sphere fraction," and makes use of the disturbance to the temperature field in an unbounded matrix, generated by two spherical inhomogeneities. The basic idea in the present study consists in replacing this "two-sphere" field by a superposition of disturbances, generated by the same two spheres, but considered as single already, together with the disturbance due to another single sphere, centered between them and radially inhomogeneous. In this way new variational bounds on the effective conductivity are derived and discussed in more detail for a special choice of the middle sphere's properties. The obtained bounds improve, in particular, on the known three-point bounds on the effective conductivity of the dispersion.

Keywords: random media, dispersions of spheres, variational bounds, effective conductivity

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1. INTRODUCTION

Consider a statistically homogeneous dispersion of equi-sized nonoverlapping spheres of conductivity κ_f and radii a, immersed at random into a matrix of conductivity κ_m. In the heat conductivity context and absence of body sources, the temperature field, $\theta(\mathbf{x})$, in the dispersion is governed by the equations

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

where $\kappa(\mathbf{x})$ is the random conductivity field of the medium, $\mathbf{q}(\mathbf{x})$ — the heat flux vector, $\mathbf{G}$ is the prescribed macroscopic value of the temperature gradient, and the brackets $\langle\cdot\rangle$ denote statistical averaging [1]. Since the field $\kappa(\mathbf{x})$ takes the values κ_f or κ_m depending on whether $\mathbf{x}$ lies in a sphere or in the matrix respectively, it allows the representation

$$\kappa(\mathbf{x}) = \langle\kappa\rangle + [\kappa]\int h(\mathbf{x}-\mathbf{y})\psi'(\mathbf{y})\,d^3\mathbf{y}, \tag{1.2}$$

where $[\kappa] = \kappa_f - \kappa_m$, $h(\mathbf{x})$ is the characteristic function of a single sphere of radius a located at the origin, and $\psi'(\mathbf{x})$ is the fluctuating part of the random density field

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

generated by the random field $\{\mathbf{x}_j\}$ of sphere's centers [2]. The integrals hereafter are over the whole $\mathbb{R}^3$ if the integration domain is not explicitly indicated.

The solution of Eq. (1.1) is understood in a statistical sense, so that one is to evaluate all multipoint moments (correlation functions) of $\theta(\mathbf{x})$ and the joint moments of $\kappa(\mathbf{x})$ and $\theta(\mathbf{x})$, see, e.g., [1]. Let c be the volume fraction of the spheres, then $n = c/V_a$ is their number density. As discussed in [3–5], the solution $\theta(\mathbf{x})$ of the random problem (1.1), asymptotically valid to the order c^2, can be found in the form of truncated functional series:

$$\theta(\mathbf{x}) = \mathbf{G}\cdot\mathbf{x} + \int T_1(\mathbf{x}-\mathbf{y})D_\psi^{(1)}(\mathbf{y})\,d^3\mathbf{y} + \iint T_2(\mathbf{x}-\mathbf{y}_1,\mathbf{x}-\mathbf{y}_2)D_\psi^{(2)}(\mathbf{y}_1,\mathbf{y}_2)\,d^3\mathbf{y}_1\,d^3\mathbf{y}_2, \tag{1.3}$$

where T_1 and T_2 are certain non-random kernels and the fields

$$D_\psi^{(0)} = 1, \quad D_\psi^{(1)}(\mathbf{y}) = \psi'(\mathbf{y}), \quad D_\psi^{(2)}(\mathbf{y}_1,\mathbf{y}_2) = \psi(\mathbf{y}_1)[\psi(\mathbf{y}_2) - \delta(\mathbf{y}_1-\mathbf{y}_2)] - ng_0(\mathbf{y}_1-\mathbf{y}_2)[D_\psi^{(1)}(\mathbf{y}_1) + D_\psi^{(1)}(\mathbf{y}_2)] - n^2 g_0(\mathbf{y}_1-\mathbf{y}_2) \tag{1.4}$$

are the first three terms in the c^2-orthogonal system, formed as a result of the appropriate virial orthogonalization, see again [3–5] for details and discussion. In Eq. (1.4) $g_0(r)$ is the leading part of the well-known radial distribution function $g(r) = f_2(r)/n^2$ for the dispersion in the dilute case $n \to 0$, i.e. $g(r) = g_0(r) + O(n)$; $f_2(r)$ denotes the two-point probability density for the set of sphere centers and $r = |\mathbf{y}_1 - \mathbf{y}_2|$.

The identification of the kernels T_1 and T_2 is performed in [4] and [5] by means of a procedure, proposed by Christov and Markov [6]. It consists in inserting the truncated series (1.3) into the random equation (1.1), multiplying the result by the fields $D_\psi^{(p)}$, $p = 0, 1, 2$, and averaging the results. In this way a certain system of integro-differential equations for the needed kernels of the truncated series can be straightforwardly derived. The solution is analytically obtained in [4] and hence the full statistical solution of the problem (1.1), asymptotically correct to the order c^2, is known. In particular, this solution allows one to derive the effective conductivity κ^* of the dispersion, to the same order c^2, through evaluating the one-point moment

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

As a result, the renormalized c^2-formula of Jeffrey [7] for the effective conductivity of the dispersion was rederived, but with rigorous justification of the integration mode in the appropriate conditionally convergent integrals.

As shown in [8], the same result is obtained when the truncated series (1.3) are employed as trial fields in the classical variational principle, corresponding to the problem (1.1):

$$W_A[\theta(\cdot)] = \left\langle \kappa(\mathbf{x}) |\nabla \theta(\mathbf{x})|^2 \right\rangle \longrightarrow \min, \quad \langle \nabla \theta(\mathbf{x}) \rangle = \mathbf{G}, \tag{1.5}$$

$\min W_A = \kappa^* G^2$, see, e.g., [1]. Moreover, the leading parts in the virial expansions

$$T_1(\mathbf{x}) = T_1(\mathbf{x}; n) = T_{1,0}(\mathbf{x}) + T_{1,1}(\mathbf{x}) n + \cdots, \tag{1.6}$$

$$T_2(\mathbf{x}, \mathbf{y}) = T_2(\mathbf{x}, \mathbf{y}; n) = T_{2,0}(\mathbf{x}, \mathbf{y}) + T_{2,1}(\mathbf{x}, \mathbf{y}) n + \cdots \tag{1.7}$$

of the optimal kernels T_1 and T_2 suffice to determine the effective conductivity κ^* to the order c^2. In this way the equations for the virial coefficients $T_{1,0}$ and $T_{2,0}$, already found in [4], have been rederived. It turned out that $T_{1,0}(\mathbf{x})$ coincides with the disturbance $T^{(1)}(\mathbf{x})$ to the temperature field $\mathbf{G} \cdot \mathbf{x}$ in an unbounded matrix, introduced by a single spherical inhomogeneity, located at the origin:

$$T_{1,0}(\mathbf{x}) = T^{(1)}(\mathbf{x}) = 3\beta G \cdot \nabla \varphi(\mathbf{x}), \tag{1.8}$$

where $\varphi(\mathbf{x}) = \varphi(\mathbf{x}, a) = h * \dfrac{1}{4\pi|\mathbf{x}|}$ is the Newtonian potential for the single sphere of the radius a and $\beta = [\kappa]/(\kappa_f + 2\kappa_m)$. For the coefficient $T_{2,0}$ one has

$$2T_{2,0}(\mathbf{x}, \mathbf{x} - \mathbf{z}) = T^{(2)}(\mathbf{x}; \mathbf{z}) - T^{(1)}(\mathbf{x}) - T^{(1)}(\mathbf{x} - \mathbf{z}), \tag{1.9}$$

where $T^{(2)}(\mathbf{x}; \mathbf{z})$ is the disturbance to the temperature field $\mathbf{G} \cdot \mathbf{x}$ in an unbounded matrix of conductivity κ_m, generated by a pair of spherical inhomogeneities of conductivity κ_f, centered at the origin and at the point $\mathbf{z}$, $|\mathbf{z}| > 2a$.

It is important to point out that the variational derivation, involving the truncated series (1.3), leads to a c^2-formula for the effective conductivity that contains absolutely convergent integrals solely. Namely, let

$$\frac{\kappa^*}{\kappa_m} = 1 + 3\beta c + a_{2\kappa}c^2 + \cdots, \quad a_{2\kappa} = 3\beta^2 + a'_{2\kappa}, \tag{1.10}$$

be the virial expansion of κ^*. For the c^2-deviation $a'_{2\kappa}$. from the well-known Maxwell formula one has

$$a'_{2\kappa}G^2 = \frac{[\kappa]}{\kappa_m}\frac{1}{V_a^2}\int h(\mathbf{x})\,d^3\mathbf{x}\int g_0(\mathbf{y})\nabla_x T^{(1)}(\mathbf{x}-\mathbf{y})\cdot\nabla_x T^{(2)}(\mathbf{x};\mathbf{y})\,d^3\mathbf{y}, \tag{1.11}$$

where $V_a = \frac{4}{3}\pi a^3$. (See also [9,10], where the Hashin-Shtrikman variational principle was employed to derive the same formula (1.11).)

In order to calculate the c^2-coefficient $a_{2\kappa}$, one needs the field $T^{(2)}(\mathbf{x};\mathbf{z})$. The latter can be explicitly found, e.g. by means of the method of twin expansions. The calculations, based on this solution and the formula (1.11), however, will be not simpler than the ones in the well-known works [7] and [11], based on the "renormalized" formula of Jeffrey [7]. That is why our aim here is to look for an appropriate approximation for the field $T^{(2)}(\mathbf{x};\mathbf{z})$ which, when combined with (1.9), will produce a class of trial field in the form (1.3). However, this class will be narrower than (1.3) and as a result certain variational bounds on $a_{2\kappa}$ will follow only.

Consider first the simplest case when the kernel T_1 in (1.3) is adjustable and the kernel T_2 vanishes: $T_2 = 0$, i.e.

$$\theta(\mathbf{x}) = \mathbf{G}\cdot\mathbf{x} + \int T_1(\mathbf{x}-\mathbf{y})D_\psi^{(1)}(\mathbf{y})\,d^3\mathbf{y}. \tag{1.12}$$

This class has been introduced and discussed in detail by Markov in [12], where it is shown that minimizing the functional $W_A[\theta(\cdot)]$ over the class (1.12) gives the best three-point upper bound $\kappa^{(3)}$ on the effective conductivity κ^*, i.e. the most restrictive one which uses three-point statistical information for the medium. According to (1.9), this bound corresponds to the approximation

$$T^{(2)}(\mathbf{x};\mathbf{z}) \approx T^{(1)}(\mathbf{x}) + T^{(1)}(\mathbf{x}-\mathbf{z}) \tag{1.13}$$

of the disturbance $T^{(2)}(\mathbf{x};\mathbf{z})$. We will come back to the three-point bounds again in Section 2.1.

Obviously, the approximation (1.13) is appropriate when the two spheres are far away, i.e. $|\mathbf{z}| \gg 2a$. Here we propose an improvement of this approximation that consists in adding the disturbance $\widetilde{T}^{(1)}(\mathbf{x}-\mathbf{z}/2)$ to the adjustable temperature field $\mathbf{\Phi}(\mathbf{z})\cdot\mathbf{x}$, generated by a single radial inhomogeneous sphere, centered between two spheres, i.e. at the point $\mathbf{z}/2$. Thus, we assume the approximation

$$T^{(2)}(\mathbf{x};\mathbf{z}) \approx \widetilde{T}^{(1)}\left(\mathbf{x}-\frac{\mathbf{z}}{2}\right) + T^{(1)}(\mathbf{x}) + T^{(1)}(\mathbf{x}-\mathbf{z}). \tag{1.14}$$

This idea is suggested by some successful models in the theory of dispersions, see, e.g., [13] and [14], where, in fact, the interactions of the spheres are taken into account by introducing a single radial inhomogeneous sphere, immersed into effective medium. According to (1.9), the approximation (1.14) leads to the following choice of the kernel T_2 in (1.3):

$$T_2(\mathbf{x}-\mathbf{y}_1,\mathbf{x}-\mathbf{y}_2)=\frac{1}{2}\widetilde{T}^{(1)}\left(\mathbf{x}-\frac{\mathbf{y}_1+\mathbf{y}_2}{2}\right). \tag{1.15}$$

In Section 2.2 a new variational procedure will be considered. It is based on the possibility to vary both the field $\boldsymbol{\Phi}(\mathbf{z})$ and the conductivity distribution of the middle sphere. Its counterpart that yields lower bounds will be discussed in Section 2.3. Finally, in Section 3 a simple case will be considered, when the middle sphere is homogeneous and encompasses the other two spheres. This case allows us to obtain quite easily explicit results, which will be then compared with some of the known variational bounds.

2. THE VARIATIONAL PROCEDURE

The disturbance $\widetilde{T}^{(1)}(\mathbf{x}-\mathbf{z}/2)$ to the temperature field $\boldsymbol{\Phi}(\mathbf{z})\cdot\mathbf{x}$, generated by a single radial inhomogeneous sphere, centered at the point $\mathbf{z}/2$, has the form

$$\widetilde{T}^{(1)}\left(\mathbf{x}-\frac{\mathbf{z}}{2}\right)=\boldsymbol{\Phi}(\mathbf{z})\cdot\nabla f\left(\mathbf{x}-\frac{\mathbf{z}}{2},\mathbf{z}\right), \tag{2.1}$$

where $f(\mathbf{w},\mathbf{z})=f(|\mathbf{w}|,\mathbf{z})$ is a function, specified by the radial distribution of the conductivity coefficient of the sphere. The dependence of $f(\mathbf{w},\mathbf{z})$ on its second argument $\mathbf{z}$ indicates explicitly the possibility that the latter distribution is arbitrary for the moment. Hereafter the differentiation of the function $f(\mathbf{w},\mathbf{z})$ is with respect to its first argument, $\nabla=\nabla_w$.

According to (1.15) and (2.1), we should employ the classical variation principle (1.5) over the class of trial field (1.3), provided the kernel T_2 has the form

$$T_2(\mathbf{x},\mathbf{x}-\mathbf{z})=\frac{1}{2}\boldsymbol{\Phi}(\mathbf{z})\cdot\nabla f\left(\mathbf{x}-\frac{\mathbf{z}}{2},\mathbf{z}\right),$$

i.e.

$$T_2(\mathbf{x}-\mathbf{y}_1,\mathbf{x}-\mathbf{y}_2)=\frac{1}{2}\boldsymbol{\Phi}(\mathbf{y}_2-\mathbf{y}_1)\cdot\nabla f\left(\mathbf{x}-\frac{\mathbf{y}_1+\mathbf{y}_2}{2},\mathbf{y}_2-\mathbf{y}_1\right). \tag{2.2}$$

Here the kernel $T_1(\mathbf{y})$, the functions $\boldsymbol{\Phi}$ and f are adjustable. To this end it is appropriate to remind briefly the variational procedure, connected with the derivation of the so-called optimal three-point bounds.

Making use of Eq. (1.2) and the formulae for the moments of the random density field $\psi(\mathbf{x})$, we find an expression for the restriction $W_A^{(1)}[T_1(\cdot)]$ of the functional W_A over the class (1.12), see [12] for details. The optimal kernel $T_1(\mathbf{x})$, i.e. the solution of the Euler-Lagrange equation for the functional $W_A^{(1)}$, is looked for in the virial form (1.6). This representation of $T_1(\mathbf{x})$ generates the appropriate virial expansion of the restriction $W_A^{(1)}[T_1(\cdot)]$, namely,

$$W_A^{(1)}[T_1(\cdot)] = \langle\kappa\rangle\, G^2 + W_A^{(1,1)}[T_{1,0}(\cdot)]n + W_A^{(1,2)}[T_{1,0}(\cdot), T_{1,1}(\cdot)]n^2 + \cdots, \quad (2.3)$$

see [15, Eqs. (4.2)-(4.5)]. An analysis of the coefficient $W_A^{(1,1)}$ shows that

$$\delta W_A^{(1,1)}[T_{1,0}(\cdot)] = 0 \Longleftrightarrow T_{1,0}(\mathbf{x}) = T^{(1)}(\mathbf{x}), \quad (2.4)$$

where $T^{(1)}(\mathbf{x})$ is the disturbance (1.8), generated by a single spherical inhomogeneity. It turns out, however, that at $T_{1,0}(\mathbf{x}) = T^{(1)}(\mathbf{x})$ the virial coefficient $W_A^{(1,2)}$ does not depend on $T_{1,1}(\mathbf{x})$, i.e.

$$W_A^{(1,2)}[T^{(1)}(\cdot), T_{1,1}(\cdot)] = \overline{W}_A^{(1,2)}[T^{(1)}(\cdot)] = 3\beta^2\kappa_m\left(1 + \frac{[\kappa]}{\kappa_m}m_2\right)V_a^2G^2, \quad (2.5)$$

where

$$m_2 = m_2[g_0(\cdot)] = 2\int_2^\infty \frac{\lambda^2}{(\lambda^2-1)^3}\, g_0(\lambda a)d\lambda, \quad \lambda = |\mathbf{y}|/a, \quad (2.6)$$

is a statistical parameter for the dispersion, introduced in [12]. Hence, according to Eq. (2.3), we have for the optimal upper tree-point bound $\kappa^{(3)}$

$$\kappa^* G^2 \le \kappa^{(3)}G^2 = \langle\kappa\rangle\, G^2 + \frac{1}{V_a}W_A^{(1,1)}[T^{(1)}(\cdot)]c + \frac{1}{V_a^2}\overline{W}_A^{(1,2)}[T^{(1)}(\cdot)]c^2 + o(c^2). \quad (2.7)$$

On the base of this analysis it is shown in [15] that the Beran's bounds [16] are c^2-optimal in the above explained sense. Eqs. (2.5) and (2.7) yield straightforwardly the following estimate for the c^2-coefficient $a_{2\kappa}$ in the virial expansion (1.10) of κ^* (see [12, 15]) :

$$a_{2\kappa} \le a_{2\kappa}^u, \quad a_{2\kappa}^u = 3\beta^2\left(1 + \frac{[\kappa]}{\kappa_m}m_2\right). \quad (2.8)$$

Let us note that the formula (2.8) for the upper bound $a_{2\kappa}^u$ can be obtained also if we insert (1.13) into (1.11), taking into account (1.10) and the identities

$$\int h(\mathbf{x})\, d^3\mathbf{x} \int g_0(\mathbf{y})\nabla T^{(1)}(\mathbf{x}-\mathbf{y})\cdot\nabla T^{(1)}(\mathbf{x})\, d^3\mathbf{x} = 0, \quad (2.9a)$$

$$\int h(\mathbf{x})\, d^3\mathbf{x} \int g_0(\mathbf{y})|\nabla T^{(1)}(\mathbf{x}-\mathbf{y})|^2\, d^3\mathbf{y} = 3\beta^2V_a^2m_2. \quad (2.9b)$$

It is interesting to point out that inserting (1.13) into the "renormalized" formula of Jeffrey [7] leads, however, to the Maxwell c^2-value $a_{2\kappa} = 3\beta^2$ that corresponds to the Hashin-Shtrikman bound.

2.2. NEW UPPER BOUND FOR THE DISPERSION

Using the formulae for the moments of the fields $D_\psi^{(1)}$ and $D_\psi^{(2)}$, see [4, Eqs. (3.4)], the restriction $W_A^{(2)}[T_1(\cdot), T_2(\cdot,\cdot)]$ of the functional W_A over the general class (1.3) becomes

$$W_A^{(2)}[T_1(\cdot), T_2(\cdot,\cdot)] = W_A^{(1)}[T_1(\cdot)] + \widetilde{W}_A^{(2)}[T_1(\cdot), T_2(\cdot,\cdot)], \tag{2.10}$$

where

$$\begin{aligned}\widetilde{W}_A^{(2)}[T_1(\cdot), T_2(\cdot,\cdot)] &= 2n^2\kappa_m \iint g_0(\mathbf{y}_1-\mathbf{y}_2)\, |\nabla_x T_2(\mathbf{x}-\mathbf{y}_1, \mathbf{x}-\mathbf{y}_2)|^2\, d^3\mathbf{y}_1\, d^3\mathbf{y}_2 \\ &+ 2n^2[\kappa] \iint g_0(\mathbf{y}_1-\mathbf{y}_2)\big[h(\mathbf{x}-\mathbf{y}_1) + h(\mathbf{x}-\mathbf{y}_2)\big]\, |\nabla_x T_2(\mathbf{x}-\mathbf{y}_1, \mathbf{x}-\mathbf{y}_2)|^2\, d^3\mathbf{y}_1\, d^3\mathbf{y}_2 \\ &+ 2n^2[\kappa] \iint g_0(\mathbf{y}_1-\mathbf{y}_2)\Big[h(\mathbf{x}-\mathbf{y}_1)\nabla T_1(\mathbf{x}-\mathbf{y}_2) + h(\mathbf{x}-\mathbf{y}_2)\nabla T_1(\mathbf{x}-\mathbf{y}_1)\Big] \\ &\cdot \nabla_x T_2(\mathbf{x}-\mathbf{y}_1, \mathbf{x}-\mathbf{y}_2)\, d^3\mathbf{y}_1\, d^3\mathbf{y}_2 + o(n^2),\end{aligned} \tag{2.11}$$

see [8, Section 3]; here we have used the fact that the kernel $T_2(\mathbf{y}_1, \mathbf{y}_2)$ is a symmetric function of its arguments.

Let us consider now the narrower class (1.3) when the kernel T_2 has the form (2.2). Then, according to Eqs. (2.3) and (2.10), for the restriction $W_A^{(2)}[T_1(\cdot), \boldsymbol{\Phi}(\cdot), f(\cdot,\cdot)]$ of the functional W_A over this class we get

$$\begin{aligned}W_A^{(2)}[T_1(\cdot), \boldsymbol{\Phi}(\cdot), f(\cdot,\cdot)] &= \langle\kappa\rangle G^2 + W_A^{(1,1)}[T_{1,0}(\cdot)]\, n \\ &+ W_A^{(2,2)}[T_{1,0}(\cdot), T_{1,1}(\cdot), \boldsymbol{\Phi}(\cdot), f(\cdot,\cdot)]\, n^2 + o(n^2),\end{aligned} \tag{2.12}$$

where

$$\begin{aligned}W_A^{(2,2)}&[T_{1,0}(\cdot), T_{1,1}(\cdot), \boldsymbol{\Phi}(\cdot), f(\cdot,\cdot)] \\ &= W_A^{(1,2)}[T_{1,0}(\cdot), T_{1,1}(\cdot)] + \widehat{W}_A^{(2)}[T_{1,0}(\cdot), \boldsymbol{\Phi}(\cdot), f(\cdot,\cdot)],\end{aligned} \tag{2.13a}$$

$$\widehat{W}_A^{(2)}[T_{1,0}(\cdot), \boldsymbol{\Phi}(\cdot), f(\cdot,\cdot)] = \frac{1}{n^2}\widetilde{W}_A^{(2)}\left[T_{1,0}(\cdot), \frac{1}{2}\boldsymbol{\Phi}(\cdot)\cdot\nabla f(\cdot,\cdot)\right]. \tag{2.13b}$$

Here $W_A^{(1,1)}$ and $W_A^{(1,2)}$ are the virial coefficients from Eq. (2.3) for which, let us recall, Eqs. (2.4) and (2.5) hold. Hence, the minimization of the functional $W_A^{(2)}$ is reduced to that of the functional

$$\widehat{W}_A^{(2)\dagger}[\boldsymbol{\Phi}(\cdot), f(\cdot,\cdot)] = \widehat{W}_A^{(2)}\left[T^{(1)}(\cdot), \boldsymbol{\Phi}(\cdot), f(\cdot,\cdot)\right]. \tag{2.14}$$

Taking into account Eqs. (1.8) and (2.13b), after an appropriate change of integrand variables in (2.11), we find the following form of functional (2.14):

$$\widehat{W}_A^{(2)\dagger}\left[\boldsymbol{\Phi}(\cdot), f(\cdot,\cdot)\right]$$

$$= \frac{1}{2}\iint g_0(\mathbf{z})\left[\kappa_m + 2[\kappa]h\left(\mathbf{w}-\frac{\mathbf{z}}{2}\right)\right]\left|\boldsymbol{\Phi}(\mathbf{z})\cdot\nabla\nabla f(\mathbf{w},\mathbf{z})\right|^2 d^3\mathbf{w}\, d^3\mathbf{z} \tag{2.15}$$

$$+ 6\beta[\kappa]\mathbf{G}\cdot\iint g_0(\mathbf{z})h\left(\mathbf{w}-\frac{\mathbf{z}}{2}\right)\nabla\nabla\varphi\left(\mathbf{w}+\frac{\mathbf{z}}{2}\right)\cdot\nabla\nabla f(\mathbf{w},\mathbf{z})\cdot\boldsymbol{\Phi}(\mathbf{z})\, d^3\mathbf{w}\, d^3\mathbf{z}\,.$$

The minimizing functions $\boldsymbol{\Phi}$ and f satisfy the Euler-Lagrange equations

$$\delta_\Phi \widehat{W}_A^{(2)\dagger} = 0, \qquad \delta_f \widehat{W}_A^{(2)\dagger} = 0\,. \tag{2.16}$$

The first of these equations yields straightforwardly

$$\boldsymbol{\Phi}(\mathbf{z})\cdot\int\left[\kappa_m + 2[\kappa]h\left(\mathbf{w}-\frac{\mathbf{z}}{2}\right)\right]\nabla\nabla f(\mathbf{w},\mathbf{z})\cdot\nabla\nabla f(\mathbf{w},\mathbf{z})\, d^3\mathbf{w}$$

$$= -6\beta[\kappa]\mathbf{G}\cdot\int h\left(\mathbf{w}-\frac{\mathbf{z}}{2}\right)\nabla\nabla\varphi\left(\mathbf{w}+\frac{\mathbf{z}}{2}\right)\cdot\nabla\nabla f(\mathbf{w},\mathbf{z})\, d^3\mathbf{w} \tag{2.17}$$

at $|\mathbf{z}| > 2a$, whose solution $\boldsymbol{\Phi}(\mathbf{z})$ can be easily found for a given function f. Taking into account that $f(\mathbf{w},\mathbf{z}) = f(|\mathbf{w}|,\mathbf{z})$, the second equation in (2.16) is recast as

$$\Phi_i(\mathbf{z})\Phi_j(\mathbf{z})\int_{\Omega_{1w}}\left\{\kappa_m\left(\Delta f(|\mathbf{w}|,z)\right)_{,ij} + 2[\kappa]\left(h\left(\mathbf{w}-\frac{\mathbf{z}}{2}\right)f_{,ik}(|\mathbf{w}|,\mathbf{z})\right)_{,kj}\right\} dS_w$$

$$= -6\beta[\kappa]G_i\Phi_j(\mathbf{z})\int_{\Omega_{1w}}\left(h\left(\mathbf{w}-\frac{\mathbf{z}}{2}\right)\varphi_{,ik}\left(\mathbf{w}+\frac{\mathbf{z}}{2}\right)\right)_{,kj} dS_w \tag{2.18}$$

at $|\mathbf{z}| > 2a$, where Ω_{1w} is the sphere $|\mathbf{w}| = 1$.

Eqs. (2.17) and (2.18) form a very complicated system of integro-differential equations for the optimal functions $\boldsymbol{\Phi}$ and f. That is why we shall consider a simpler procedure in which the function f is fixed.

Making use of Eq. (2.17), the minimum value of the functional $\widehat{W}_A^{(2)\dagger}$ can be recast now in the form in which the solution $\boldsymbol{\Phi}(\mathbf{z})$ of this equation enters linearly:

$$\min_{\Phi}\widehat{W}_A^{(2)\dagger}\left[\boldsymbol{\Phi}(\cdot), f(\cdot,\cdot)\right]$$

$$= 3\beta[\kappa]\mathbf{G}\cdot\iint g_0(\mathbf{z})h\left(\mathbf{w}-\frac{\mathbf{z}}{2}\right)\nabla\nabla\varphi\left(\mathbf{w}+\frac{\mathbf{z}}{2}\right)\cdot\nabla\nabla f(\mathbf{w},\mathbf{z})\cdot\boldsymbol{\Phi}(\mathbf{z})\, d^3\mathbf{w}\, d^3\mathbf{z}\,. \tag{2.19}$$

With the notations

$$\boldsymbol{\mathcal{R}}(\mathbf{z}) = \frac{1}{V_a}\int\left[1 + 2\frac{[\kappa]}{\kappa_m}h\left(\mathbf{w}-\frac{\mathbf{z}}{2}\right)\right]\nabla\nabla f(\mathbf{w},\mathbf{z})\cdot\nabla\nabla f(\mathbf{w},\mathbf{z})\, d^3\mathbf{w}, \tag{2.20a}$$

$$\mathcal{J}(\mathbf{z}) = \frac{1}{V_a}\int h\left(\mathbf{w}-\frac{\mathbf{z}}{2}\right)\nabla\nabla\varphi\left(\mathbf{w}+\frac{\mathbf{z}}{2}\right)\cdot\nabla\nabla f(\mathbf{w},\mathbf{z})\,d^3\mathbf{w}, \tag{2.20b}$$

Eqs. (2.17) and (2.19) can be written in the form

$$\boldsymbol{\Phi}(\mathbf{z})\cdot\mathcal{R}(\mathbf{z}) = -6\beta\frac{[\kappa]}{\kappa_m}\mathbf{G}\cdot\mathcal{J}(\mathbf{z}),$$

$$\min_{\Phi}\widehat{W}_A^{(2)\dagger}\left[\boldsymbol{\Phi}(\cdot), f(\cdot,\cdot)\right] = 3\beta[\kappa]V_a\mathbf{G}\cdot\int g_0(\mathbf{z})\mathcal{J}(\mathbf{z})\cdot\boldsymbol{\Phi}(\mathbf{z})\,d^3\mathbf{z}\,.$$

Thus the solution of Eq. (2.17) is

$$\boldsymbol{\Phi}(\mathbf{z}) = -6\beta\frac{[\kappa]}{\kappa_m}\mathbf{G}\cdot\mathcal{J}(\mathbf{z})\cdot\mathcal{R}^{-1}(\mathbf{z}) \tag{2.21}$$

and the minimum value of the functional $\widehat{W}_A^{(2)\dagger}$ is

$$\min_{\Phi}\widehat{W}_A^{(2)\dagger}\left[\boldsymbol{\Phi}(\cdot), f(\cdot,\cdot)\right] = -\,18\beta^2\frac{[\kappa]^2}{\kappa_m}V_a\mathbf{G}\cdot\int g_0(\mathbf{z})\mathcal{J}(\mathbf{z})\cdot\mathcal{R}^{-1}(\mathbf{z})\cdot\mathcal{J}(\mathbf{z})\,d^3\mathbf{z}\cdot\mathbf{G}\,. \tag{2.22}$$

Hence, according to Eqs. (2.4), (2.5), (2.7), (2.12)–(2.14), we obtain the following upper bound on the effective conductivity κ^*:

$$\kappa^* G^2 \le \kappa^\dagger G^2, \quad \kappa^\dagger G^2 = \langle\kappa\rangle G^2 + \frac{1}{V_a}W_A^{(1,1)}[T^{(1)}(\cdot)]\,c$$

$$+\frac{1}{V_a^2}\left\{\overline{W}_A^{(1,2)}[T^{(1)}(\cdot)] + \min\widehat{W}_A^{(2)\dagger}\right\}c^2 + o(c^2) = \kappa^{(3)} + \frac{1}{V_a}^2\min\widehat{W}_A^{(2)\dagger}\,c^2 + o(c^2). \tag{2.23}$$

In turn, Eqs. (2.5), (2.22) and (2.23) yield straightforwardly an upper bound for the c^2-coefficient $a_{2\kappa}$ in the virial expansion (1.10) of κ^*, namely,

$$a_{2\kappa} \le a_{2\kappa}^{u\dagger}, \quad a_{2\kappa}^{u\dagger} = 3\beta^2\left(1 + \frac{[\kappa]}{\kappa_m}m_2 - \left(\frac{[\kappa]}{\kappa_m}\right)^2\widetilde{m}_2^u\right), \tag{2.24}$$

where

$$\widetilde{m}_2^u = \widetilde{m}_2^u[g_0(\cdot), f(\cdot,\cdot),\alpha] = \frac{2}{V_a}\int g_0(\mathbf{z})\,\mathrm{tr}\left[\mathcal{J}(\mathbf{z})\cdot\mathcal{R}^{-1}(\mathbf{z})\cdot\mathcal{J}(\mathbf{z})\right]\,d^3\mathbf{z} \tag{2.25}$$

is a new statistical parameter for the dispersion, $\alpha = \kappa_f/\kappa_m$. This parameter depends not only on the leading part $g_0(r)$ of the radial distribution function g, but on the given function $f(\mathbf{w},\mathbf{z})$ and on the ratio α for the dispersion as well, see Eqs. (2.20).

In order to obtain a similar lower bound on κ^*, we shall employ the classical dual variational principle for the problem (1.1), formulated with respect to the heat flux $\mathbf{q}(\mathbf{x}) = \nabla \times \mathbf{U}(\mathbf{x})$,

$$W_B[\mathbf{U}(\cdot)] = \left\langle k(\mathbf{x})|\nabla \times \mathbf{U}(\mathbf{x})|^2 \right\rangle \longrightarrow \min, \quad \langle \mathbf{q}(\mathbf{x}) \rangle = \mathbf{Q}, \tag{2.26}$$

$\min W_B = k^* Q^2$, $k^* = 1/\kappa^*$. The compliance field $k(\mathbf{x}) = 1/\kappa(\mathbf{x})$ has the form (1.2), i.e.

$$k(\mathbf{x}) = \langle k \rangle + [k] \int h(\mathbf{x}-\mathbf{y})\psi'(\mathbf{y})\, d^3\mathbf{y}, \quad [k] = k_f - k_m. \tag{2.27}$$

Similarly to the above-performed analysis, consider the functional W_B over the class of trial field

$$\begin{aligned} \mathbf{U}(\mathbf{x}) = \frac{1}{2}\mathbf{Q} \times \mathbf{x} + \int \mathbf{S}_1(\mathbf{x}-\mathbf{y}) D_\psi^{(1)}(\mathbf{y})\, d^3\mathbf{y} \\ + \iint \mathbf{S}_2(\mathbf{x}-\mathbf{y}_1, \mathbf{x}-\mathbf{y}_2) D_\psi^{(2)}(\mathbf{y}_1, \mathbf{y}_2)\, d^3\mathbf{y}_1\, d^3\mathbf{y}_2 \end{aligned} \tag{2.28}$$

— the counterpart of the class (1.3). Similarly, if the kernels $\mathbf{S}_1$ and $\mathbf{S}_2$ are arbitrary adjustable functions, the class (2.28) leads to the exact c^2-value of the effective compliance k^*, as it was the case with the effective conductivity κ^*. For the restriction $W_B^{(2)}[\mathbf{S}_1(\cdot), \mathbf{S}_2(\cdot,\cdot)]$ of the functional W_B over this class one has

$$W_B^{(2)}[\mathbf{S}_1(\cdot), \mathbf{S}_2(\cdot,\cdot)] = W_B^{(1)}[\mathbf{S}_1(\cdot)] + \widetilde{W}_B^{(2)}[\mathbf{S}_1(\cdot), \mathbf{S}_2(\cdot,\cdot)], \tag{2.29}$$

where $W_B^{(1)}[\mathbf{S}_1(\cdot)]$ is the restriction of W_B over the class

$$\mathbf{U}(\mathbf{x}) = \frac{1}{2}\mathbf{Q} \times \mathbf{x} + \int \mathbf{S}_1(\mathbf{x}-\mathbf{y}) D_\psi^{(1)}(\mathbf{y})\, d^3\mathbf{y} \tag{2.30}$$

and

$$\begin{aligned} \widetilde{W}_B^{(2)}[\mathbf{S}_1(\cdot), \mathbf{S}_2(\cdot,\cdot)] &= 2n^2 k_m \iint g_0(\mathbf{y}_1-\mathbf{y}_2)\, |\nabla_x \times \mathbf{S}_2(\mathbf{x}-\mathbf{y}_1, \mathbf{x}-\mathbf{y}_2)|^2\, d^3\mathbf{y}_1\, d^3\mathbf{y}_2 \\ &+ 2n^2[k] \iint g_0(\mathbf{y}_1-\mathbf{y}_2)\big[h(\mathbf{x}-\mathbf{y}_1) + h(\mathbf{x}-\mathbf{y}_2)\big]\, |\nabla_x \times \mathbf{S}_2(\mathbf{x}-\mathbf{y}_1, \mathbf{x}-\mathbf{y}_2)|^2\, d^3\mathbf{y}_1\, d^3\mathbf{y}_2 \\ &+ 2n^2[k] \iint g_0(\mathbf{y}_1-\mathbf{y}_2)\Big[h(\mathbf{x}-\mathbf{y}_1)\nabla \times \mathbf{S}_1(\mathbf{x}-\mathbf{y}_2) + h(\mathbf{x}-\mathbf{y}_2)\nabla \times \mathbf{S}_1(\mathbf{x}-\mathbf{y}_1)\Big] \\ &\cdot \nabla_x \times \mathbf{S}_2(\mathbf{x}-\mathbf{y}_1, \mathbf{x}-\mathbf{y}_2)\, d^3\mathbf{y}_1\, d^3\mathbf{y}_2 + o(n^2). \end{aligned} \tag{2.31}$$

The class (2.30) is the counterpart of (1.12) and leads to the c^2-optimal three-point lower bound $1/k^{(3)}$ on the effective conductivity κ^*, see [12]. The solution of the Euler-Lagrange equation $\delta W_B^{(1)}[\mathbf{S}_1(\cdot)] = 0$ now has the form

$$\mathbf{S}_1(\mathbf{x}) = \mathbf{S}^{(1)}(\mathbf{x}) + O(c),$$

where $\mathbf{q}^{(1)}(\mathbf{x}) = \nabla \times \mathbf{S}^{(1)}(\mathbf{x})$ is the disturbance to the constant heat flux $\mathbf{Q}$ in an unbounded matrix, introduced by a single spherical inhomogeneity, located at the origin:

$$\mathbf{q}^{(1)}(\mathbf{x}) = 3\beta \mathbf{Q} \cdot \left[\nabla\nabla\varphi(\mathbf{x}) + h(\mathbf{x})\mathbf{I} \right], \quad \text{i.e.} \quad \mathbf{S}^{(1)}(\mathbf{x}) = -3\beta \mathbf{Q} \times \nabla\varphi(\mathbf{x}). \tag{2.32}$$

Then

$$\begin{aligned} k^* \leq k^{(3)}, \quad k^{(3)}Q^2 &= \min W_B^{(1)}[\mathbf{S}_1(\cdot)] = W_B^{(1)}[\mathbf{S}^{(1)}(\cdot)] + o(c^2) \\ &= k_m \left\{ 1 - 3\beta c + 3\beta^2 \left(2 + \frac{[k]}{k_m} m_2 \right) c^2 \right\} Q^2 + o(c^2), \end{aligned} \tag{2.33}$$

where m_2 is the statistical parameter (2.6). In virtue of these relations, the optimal three-point lower bounds for the c^2-coefficient $a_{2\kappa}$ in the virial expansion (1.10) of κ^* are straightforwardly obtained (see [12, 15]):

$$a_{2\kappa}^l \leq a_{2\kappa}, \quad a_{2\kappa}^l = 3\beta^2 \left(1 + \frac{[\kappa]}{\kappa_f} m_2 \right). \tag{2.34}$$

Eqs. (2.29) and (2.31) are the counterparts of Eqs. (2.10) and (2.11) respectively. A fully similar analysis shows in turn that the leading part $\mathbf{S}_{2,0}$ of the optimal kernel $\mathbf{S}_2$, $\mathbf{S}_2(\mathbf{x}, \mathbf{y}) = \mathbf{S}_{2,0}(\mathbf{x}, \mathbf{y}) + O(c)$, has now the form

$$2\mathbf{S}_{2,0}(\mathbf{x}, \mathbf{x} - \mathbf{z}) = \mathbf{S}^{(2)}(\mathbf{x}; \mathbf{z}) - \mathbf{S}^{(1)}(\mathbf{x}) - \mathbf{S}^{(1)}(\mathbf{x} - \mathbf{z}), \tag{2.35}$$

where $\mathbf{q}^{(2)}(\mathbf{x}; \mathbf{z}) = \nabla_x \times \mathbf{S}^{(2)}(\mathbf{x}; \mathbf{z})$ is the disturbance to the constant heat flux $\mathbf{Q}$ in an unbounded matrix of conductivity κ_m, generated by a pair of spherical inhomogeneities of conductivity κ_f, centered at the origin and at the point $\mathbf{z}$.

In order to improve on the optimal lower bound (2.34), similarly to Eqs. (1.8), (2.1)–(2.3) for the upper one and (2.32), we can make the following choice of the kernel $\mathbf{S}_2$ in (2.28):

$$\mathbf{S}_2(\mathbf{x}, \mathbf{x} - \mathbf{z}) = \frac{1}{2} \boldsymbol{\Phi}(\mathbf{z}) \times \nabla f \left(\mathbf{x} - \frac{\mathbf{z}}{2}, \mathbf{z} \right), \tag{2.36a}$$

where the functions $\boldsymbol{\Phi}$ and f can be again treated as adjustable. Let us note that now the field

$$\begin{aligned} \widetilde{\mathbf{q}}_2(\mathbf{x}, \mathbf{x} - \mathbf{z}) &= 2\nabla_x \times \mathbf{S}_2(\mathbf{x}, \mathbf{x} - \mathbf{z}) \\ &= -\boldsymbol{\Phi}(\mathbf{z}) \cdot \left[\nabla\nabla f \left(\mathbf{x} - \frac{\mathbf{z}}{2}, \mathbf{z} \right) - \Delta f \left(\mathbf{x} - \frac{\mathbf{z}}{2}, \mathbf{z} \right) \mathbf{I} \right], \end{aligned} \tag{2.36b}$$

in general, is *not* the disturbance to a certain heat flux in an unbounded matrix, introduced by a single radial inhomogeneous sphere, centered at the point $\mathbf{z}/2$. A simple check shows, however, that for a homogeneous middle sphere the field $\widetilde{\mathbf{q}}_2(\mathbf{x}, \mathbf{x} - \mathbf{z})$ is indeed such a disturbance, see Eqs. (2.32) and (2.36a). An example of this kind will be considered in Section 3.

The further analysis is fully similar to the one, already performed in Section 2.2. That is way we shall present the basic results only. The explicit form of the functional

$$\widehat{W}_B^{(2)\dagger}\left[\boldsymbol{\Phi}(\cdot), f(\cdot,\cdot)\right] = \frac{1}{n^2}\widetilde{W}_B^{(2)}\left[\mathbf{S}^{(1)}(\cdot), \frac{1}{2}\boldsymbol{\Phi}(\cdot)\cdot\nabla f(\cdot,\cdot)\right] \tag{2.37}$$

is obtained straightforwardly by means of Eqs. (2.29), (2.31), (2.32) and (2.36); it is of the same form (2.15), provided we replace κ by k, $\mathbf{G}$ by $\mathbf{Q}$, $\nabla\nabla\varphi$ by $\nabla\nabla\varphi + h\mathbf{I}$ and $\nabla\nabla f$ by $-(\nabla\nabla f - \Delta f\mathbf{I})$.

With the notations

$$\mathfrak{R}(\mathbf{z}) = \frac{1}{V_a}\int\left[1 - 2\frac{[\kappa]}{\kappa_f}h\left(\mathbf{w}-\frac{\mathbf{z}}{2}\right)\right]\left[\nabla\nabla f(\mathbf{w},\mathbf{z}) - \Delta f(\mathbf{w},\mathbf{z})\mathbf{I}\right] \tag{2.38a}$$
$$\cdot\left[\nabla\nabla f(\mathbf{w},\mathbf{z}) - \Delta f(\mathbf{w},\mathbf{z})\mathbf{I}\right] d^3\mathbf{w},$$

$$\mathfrak{S}(\mathbf{z}) = \frac{1}{V_a}\int h\left(\mathbf{w}-\frac{\mathbf{z}}{2}\right)\left[\nabla\nabla\varphi\left(\mathbf{w}+\frac{\mathbf{z}}{2},\mathbf{z}\right) + h\left(\mathbf{w}+\frac{\mathbf{z}}{2}\right)\mathbf{I}\right] \tag{2.38b}$$
$$\cdot\left[\nabla\nabla f(\mathbf{w},\mathbf{z}) - \Delta f(\mathbf{w},\mathbf{z})\mathbf{I}\right] d^3\mathbf{w},$$

the Euler-Lagrange equation $\delta_{\Phi}\widehat{W}_B^{(2)\dagger} = 0$ reads

$$\boldsymbol{\Phi}(\mathbf{z})\cdot\mathfrak{R}(\mathbf{z}) = 6\beta\frac{[k]}{k_m}\mathbf{Q}\cdot\mathfrak{S}(\mathbf{z}),$$

whose solution is

$$\boldsymbol{\Phi}(\mathbf{z}) = 6\beta\frac{[k]}{\kappa_m}\mathbf{Q}\cdot\mathfrak{S}(\mathbf{z})\cdot\mathfrak{R}^{-1}(\mathbf{z}). \tag{2.39}$$

Then the minimum value of the functional $\widehat{W}_B^{(2)\dagger}$ is

$$\min_{\Phi}\widehat{W}_B^{(2)\dagger}\left[\boldsymbol{\Phi}(\cdot), f(\cdot,\cdot)\right] = -18\beta^2\frac{[k]^2}{k_m}V_a\mathbf{Q}\cdot\int g_0(\mathbf{z})\mathfrak{S}(\mathbf{z})\cdot\mathfrak{R}^{-1}(\mathbf{z})\cdot\mathfrak{S}(\mathbf{z})\,d^3\mathbf{z}\cdot\mathbf{Q}. \tag{2.40}$$

According to Eqs. (2.29), (2.33), (2.37) and (2.40), an upper bound $k^\dagger$ on the effective compliance k^* immediately follows

$$k^*Q^2 \le k^\dagger Q^2 = k^{(3)} + \frac{1}{V_a^2}\min\widehat{W}_B^{(2)\dagger}c^2 + o(c^2)$$

$$= k_m\left\{1 - 3\beta c + 3\beta^2\left(2 + \frac{[k]}{k_m}m_2 - \frac{[k]}{k_m}^2\widetilde{m}_2^l\right)c^2\right\}Q^2 + o(c^2).$$

Here

$$\widetilde{m}_2^l = \widetilde{m}_2^l[g_0(\cdot), f(\cdot,\cdot), \alpha] = \frac{2}{V_a}\int g_0(\mathbf{z})\,\mathrm{tr}\left[\mathfrak{S}(\mathbf{z})\cdot\mathfrak{R}^{-1}(\mathbf{z})\cdot\mathfrak{S}(\mathbf{z})\right]\,d^3\mathbf{z} \tag{2.41}$$

is the counterpart of the statistical parameter $\widetilde{m}_2^u$, see (2.25). In virtue of these relations we obtain straightforwardly the following lower bound for the c^2-coefficient $a_{2\kappa}$ in the virial expansion (1.10) of κ^*:

$$a_{2\kappa}^{l\dagger} \leq a_{2\kappa}, \quad a_{2\kappa}^{l\dagger} = 3\beta^2 \left(1 + \frac{[\kappa]}{\kappa_f} m_2 + \left(\frac{[\kappa]}{\kappa_f}\right)^2 \widetilde{m}_2^l\right). \tag{2.42}$$

Let us note that the bounds (2.24) and (2.42) are *five-point* bounds in the sense that they require knowledge of the first ℓ-point moments for the random density field $\psi(\mathbf{x})$ up to $\ell = 5$, see Eqs. (1.2)–(1.5), (2.26)–(2.28). To get explicitly the parameters $\widetilde{m}_2^l$ and $\widetilde{m}_2^u$ for a given function f, an analytical evaluation of the integrals (2.20) and (2.38) is needed however.

3. A SIMPLE EXAMPLE

Let us choose now the function $f(\mathbf{w}, \mathbf{z})$ in Eqs. (2.2) and (2.36) in the form

$$f(\mathbf{w}, \mathbf{z}) = \varphi\left(\mathbf{w}, \frac{|\mathbf{z}|}{2} + A\right)$$

at $|\mathbf{z}| \geq 2a$, i.e.

$$f\left(\mathbf{x} - \frac{\mathbf{z}}{2}, \mathbf{z}\right) = \varphi\left(\mathbf{x} - \frac{\mathbf{z}}{2}, \frac{|\mathbf{z}|}{2} + A\right), \tag{3.1}$$

where A is a scalar parameter, $A \geq a$, so that $|\mathbf{z}|/22 + A \geq 2a$.

According to the foregoing analysis, this choice means that the disturbance $T^{(2)}(\mathbf{x}; \mathbf{z})$, generated by two spheres centered at the origin and at the point $\mathbf{z}$, is approximated by the superposition of the disturbances $T^{(1)}(\mathbf{x})$ and $T^{(1)}(\mathbf{x} - \mathbf{z})$, generated by the same two spheres, but considered as singly, and the disturbance $T^{(1)}(\mathbf{x} - \mathbf{z}/2)$, generated by a single homogeneous sphere, centered exactly between them and encompassing the same spheres, see Eqs. (1.8), (1.14), (2.1) and (3.1). At that, let us recall, the middle sphere is immersed into adjustable temperature field $\boldsymbol{\Phi}(\mathbf{z})$ that has been varied in order to derive the best c^2-bounds on the effective conductivity. Now we shall obtain this bounds as functions of the parameter $s = A/a$, $s \geq 1$.

After simple change of the integrand variable the fields $\mathcal{R}(\mathbf{z})$ and $\mathcal{J}(\mathbf{z})$ in (2.20) are recast as

$$\mathcal{R}(\mathbf{z}) = \frac{1}{V_a} \int \left[1 + 2\frac{[\kappa]}{\kappa_m} h(\mathbf{u})\right] \nabla\nabla f\left(\frac{\mathbf{z}}{2} - \mathbf{u}, \mathbf{z}\right) \cdot \nabla\nabla f\left(\frac{\mathbf{z}}{2} - \mathbf{u}, \mathbf{z}\right) d^3\mathbf{u},$$

$$\mathcal{J}(\mathbf{z}) = \frac{1}{V_a} \int h(\mathbf{u}) \nabla\nabla\varphi(\mathbf{z} - \mathbf{u}) \cdot \nabla\nabla f\left(\frac{\mathbf{z}}{2} - \mathbf{u}, \mathbf{z}\right) d^3\mathbf{u}. \tag{3.2}$$

Taking into account that $\nabla\nabla\varphi(\mathbf{u},\mathbf{z}) = -\frac{1}{3}\mathbf{I}$ at $|\mathbf{u}| < |\mathbf{z}|$ and the Eqs. (3.1) and (3.2), we get

$$\mathcal{R}(\mathbf{z}) = \frac{1}{9}\left\{3\left(\frac{|\mathbf{z}|}{2a}+s\right)^3 + 2\frac{[\kappa]}{\kappa_m}\right\}\mathbf{I},$$
$$\mathcal{J}(\mathbf{z}) = -\frac{1}{3}\boldsymbol{\omega}(\mathbf{z}), \quad \boldsymbol{\omega}(\mathbf{z}) = \frac{1}{V_a}\int h(u)\nabla\nabla\varphi(\mathbf{z}-\mathbf{u})\,d^3\mathbf{u}. \tag{3.3}$$

The field $\boldsymbol{\omega}(\mathbf{z})$ is the same one that appears in the variational procedure of Willis [17], see [9, 10] also, whose explicit form is

$$\boldsymbol{\omega}(\mathbf{z}) = \frac{1}{3}\left(\frac{a}{|\mathbf{z}|}\right)^3(3\mathbf{e}_r\mathbf{e}_r - \mathbf{I}), \quad \mathbf{e}_r = \mathbf{z}/|\mathbf{z}|. \tag{3.4}$$

In the same way one obtains for the fields $\mathfrak{R}(\mathbf{z})$ and $\mathfrak{I}(\mathbf{z})$ in (2.38) the following formulae:

$$\mathfrak{R}(\mathbf{z}) = \frac{2}{9}\left\{3\left(\frac{|\mathbf{z}|}{2a}+s\right)^3 - 4\frac{[\kappa]}{\kappa_f}\right\}\mathbf{I}, \quad \mathfrak{I}(\mathbf{z}) = \frac{2}{3}\boldsymbol{\omega}(\mathbf{z}). \tag{3.5}$$

After simple algebra, based on Eqs. (2.25), (2.41), (3.3)–(3.5), we get eventually the needed parameters $\widetilde{m}_2^u$ and $\widetilde{m}_2^l$:

$$\widetilde{m}_2^u = 32\int_0^{1/2} g_0\left(\frac{a}{\rho}\right)\frac{\rho^5}{3(1+2s\rho)^3 + 16\rho^3[\kappa]/\kappa_m}\,d\rho,$$
$$\widetilde{m}_2^l = 64\int_0^{1/2} g_0\left(\frac{a}{\rho}\right)\frac{\rho^5}{3(1+2s\rho)^3 - 32\rho^3[\kappa]/k_f}\,d\rho. \tag{3.6}$$

Thus, for the simple choice (3.1) of the function $f(\mathbf{w},\mathbf{z})$, we have obtained the c^2-bounds (2.24), (2.42) explicitly. A simple check shows that the integrands in (3.6) are always positive, and so are the parameters $\widetilde{m}_2^l$ and $\widetilde{m}_2^u$. Then, from (2.8), (2.24), (2.34) and (2.42), we can conclude that the obtained bounds always improve on the optimal three-point bounds. Moreover, it is immediately seen that the parameters $\widetilde{m}_2^l$ and $\widetilde{m}_2^u$ are decreasing functions of the parameter s, vanishing as $s \to \infty$. Therefore the obtained bounds are the best if $s = 1$, i.e. when the middle sphere, encompassing the other two ones, touches them. This fact suggests that the consideration of the case when the middle sphere overlaps the other two spheres could lead to better results. The calculations in this case, however, are more complicated. In the limiting case $s \to \infty$ our bounds coincide with the optimal three-point bounds.

The behaviour of our bounds is illustrated in the well-stirred case when $g(\mathbf{z}) = 1$ at $|\mathbf{z}| > 2a$, see Table 1. It is seen that the new lower bound (at $s = 1$) improves

TABLE 1. Comparison of various bounds on the c^2-coefficient a_2 for a well-stirred dispersion of spheres; the exact values are due to Felderhof *et al.* [11] and the value of the parameter m_2 is $m_2 \approx 0.14045$ [12]

β	α	Lower bounds			exact	Upper bounds		
		3-point (2.34)	present (2.42)	Willis [9, (7.21)]		present (2.24)	3-point (2.8)	Willis [9, (7.21)]
-0.5	0	$-\infty$	$-\infty$	–	0.588	0.641	0.645	0.659
-0.49	0.013	-6.715	-2.934	–		0.617	0.620	0.634
-0.4	0.143	0.076	0.165	–	0.399	0.421	0.422	0.433
-0.3	0.308	0.185	0.194	–	0.236	0.243	0.244	0.250
-0.2	0.500	0.103	0.104	–	0.110	0.111	0.112	0.114
-0.1	0.727	0.028	0.028	–	0.029	0.029	0.029	0.029
0	1	0	0	0	0	0	0	0
0.2	1.75	0.127	0.127	0.126	0.130	0.132	0.133	–
0.4	3.00	0.525	0.527	0.529	0.563	0.607	0.615	–
0.6	5.50	1.204	1.211	1.249	1.370	1.686	1.763	–
0.8	13	2.169	2.185	2.328	2.638	4.437	5.156	–
0.9	28	2.759	2.782	3.016	3.485	8.576	11.645	–
0.99	298	3.352	3.382	3.726		58.705	125.592	–
1.0	∞	3.420	3.450	3.811	4.506	∞	∞	–

considerably on the respective three-point bound when $\alpha \to 0$; a similar improvement takes place for the upper ones at $\alpha \to \infty$. In Table 1 the bound of Willis [9] is also given. Recall that it improves on the lower three-point bound, but the upper one is worse.

Finally, we shall note that the proposed approach to derive variational bounds can be employed on the base of the variational principle of Hashin-Shtrikman. In this case it can be easily shown, for example, that the bounds of Willis correspond to the approximation $\nabla T^{(2)}(\mathbf{x};\mathbf{z}) = \nabla T^{(1)}(\mathbf{x}) + \boldsymbol{\Phi}(\mathbf{z})$, see [9, 10]. This means that the bounds of Willis can be treated as the exact HS-counterpart of our bounds, derived in Section 3. More details will be given elsewhere.

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