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# Conductivity of periodic composites made of matrix and polydispersed aggregates

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We present closed-form expressions of the effective conductivity of composites made of matrix and impenetrable polydispersed particles. The approach developed in a recent work has been generalized to treat periodic multiphase composite materials. Particular cases including orthorhombic arrangements of spherical or spheroidal inclusions and random distributions of spheres are considered where simple closed-form expressions are obtained. The case of random distributions allows us to recover Mori-Tanaka estimation after introducing partial structure factors as a statistical information on the microstructure.

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## I. INTRODUCTION

Determining the effective response of heterogeneous materials from their constituents is of both theoretical and practical interest. The so-called homogenization procedure which allows such a determination usually relies on solving the boundary value problem on a representative volume element (RVE) and finding the relation between the average quantities [1]. If the microstructure of the heterogeneous material is periodic, rigorous definitions of the overall properties were derived for many classes of problems [2]. Numerical homogenization techniques based on periodic RVEs have also been applied successfully to periodic structures [3,4] and to systems with random nature in many situations [5–7]. The convergence of these methods becomes a serious problem for high contrasts, and a refinement using the spectrum of the Green's operator can help to the convergence in these cases [8,9].

The present work presents a solution of the homogenization problem of periodic media composed of matrix and inclusions, where the effective conductivity is concerned. Most of the literature works on these types of microstructure deal with two phases composites, including the matrix material [7,10–16]. Despite their popularity and technological importance, materials made from particles of different sizes and properties received less consideration (see, e.g., Refs. [1,17–20] and the references therein). In this paper, we propose a general estimation of the effective conductivity of impenetrable polydispersed particles and treat the hard ellipsoidal and spheres as special cases. The approach developed by the authors is a generalization of the recent work on two-phases materials [21], and its origin can be traced back to Nemat-Nasser-Iwakuma-Hejazi (NIH) approximation in linear elasticity [22]. The latter has been shown to perform well for a large range of volume fractions.

From the governing integral equation, an analytical approximation is applied to each species $\alpha$ containing particles of the same size and material, and the average localization tensor of all $m$ species in the model can be determined by solving a linear system of $m$ equations. The coefficients of this linear system of equations depend obviously on the distribution of the different kinds of inclusions. In the case of composites containing only one kind of inclusions, the expression of effective properties is expressed by using the Fourier transform coefficients of the characteristic function which defines the domain containing the inclusions, while for composites containing different kinds of inclusions, the solution contains terms coupling the different domains related to different inclusions as shown in Ref. [1]. These terms, which do not appear in the case of monodispersion, must now be obtained, which is a different aspect compared to the original method [21]. Two different cases are treated in detail.

A first case corresponds to a distribution of inclusions which are centered at the points of lattices (cubic or orthorhombic). In this case, the effective properties can be obtained in a closed form, which can be effected by computing analytically the "coupling terms" corresponding to different kinds of inclusions. Closed form solutions have been obtained for cubic lattice or tetragonal arrangement of two families of spheres or ellipsoids. Application results are in a good agreement with numerical methods based on Fourier transform. The extension to ellipsoidal inclusions constitutes also an original extension of the method introduced in Ref. [21].

A second case is the one of a random distribution of polydispersed hard spheres. In the case of an isotropic random distributions of spheres, a statistical connection with partial structure factors has been established. It was shown that, in the infinite volume limit, simple expressions are obtained, depending only on the volume fractions and properties of the species. The final expression of the effective conductivity is identical to the Mori-Tanaka estimation for multicomponent composites [18], which is connected to the Clausius-Mossotti approximation [20]. This result is particularly interesting, because the classical way of expressing the Mori-Tanaka estimation, following the Benveniste reformulation [23], is to consider that the interaction between phases is approximated by subjecting each inclusion separately to the mean temperature gradient in the matrix phase. It is clear that this formulation does not rest on any statistical information, while our result rests explicitly on such information. Our result is therefore, for the Mori-Tanaka distribution, the equivalent of the "perfectly disordered distribution" associated with the well-known self-consistent model [24] and completes the relation between "ad hoc" estimations (self-consistent, Mori-Tanaka, etc.) and the statistical information to which it corresponds.

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Section II presents the general formalism for periodic materials, which introduces the coupling coefficients. This formalism is detailed in order to produce explicitly the effective conductivity in the case of a periodic cell containing three planes of symmetry. Section III presents the estimation of the conductivity of bimodal periodic arrangements of spheres or ellipsoids. Finally, Sec. IV presents the estimation of the conductivity of a random isotropic distribution of polydispersed bimodal hard spheres.

Regarding the notation found in the paper, we use bold characters to denote the vector and matrix quantities. The notation $*$ is used to denote the convolution integral, center dot $\cdot$ for vector products, and nothing for the action of a matrix on a vector.

## II. PERIODIC CONDUCTION PROBLEM AND ITS SOLUTION FOR SPECIFIC SYMMETRIES OF THE PERIODIC CELL

### A. Governing integral equation

Let us consider a periodic media made by repeating infinitely a parallepipedic cell of volume $V = a_1 \times a_2 \times a_3$ along the three directions $x_1$,$x_2$, and $x_3$. The cell has heterogeneous nature in terms of thermal conduction, and its thermal conductivity $\mathbf{K}$ is a function of position $\mathbf{x}$. To determine the effective conductivity $\mathbf{K}^{\text{eff}}$ of the material, we must solve the periodic problem in the unit cell [2] and find the relation between $\mathbf{Q}$, the average of the local flux $\mathbf{q}$, and $\mathbf{E}$, the average of the local (minus) temperature gradient $\mathbf{e}$. Alternatively, we can solve $\mathbf{e}^*(\mathbf{x}) = -\boldsymbol{\nabla} T$ from the integral equation [21]

$$(\mathbf{K}^0 - \mathbf{K}(\mathbf{x}))\left\{\mathbf{E} + \sum_{\boldsymbol{\xi}\neq\mathbf{0}} \boldsymbol{\Gamma}(\boldsymbol{\xi})\left[\frac{1}{V}\int_V \mathbf{e}^*(\mathbf{x}')e^{i\boldsymbol{\xi}\cdot(\mathbf{x}-\mathbf{x}')}\,d\mathbf{x}'\right]\right\} = \mathbf{K}^0\mathbf{e}^*(\mathbf{x}), \tag{1}$$

with $\mathbf{K}^0$ being any second order tensor, usually taken to be isotropic, and $\boldsymbol{\Gamma}(\boldsymbol{\xi})$ the second order tensor defined as

$$\boldsymbol{\Gamma}(\boldsymbol{\xi}) = \overline{\boldsymbol{\xi}} \otimes \overline{\boldsymbol{\xi}}, \quad \overline{\boldsymbol{\xi}} = \boldsymbol{\xi}/\xi, \quad \xi = |\boldsymbol{\xi}|. \tag{2}$$

Here the vector field $\mathbf{e}^*$, which may be called the *free temperature gradient*, is defined from the reference conductivity $\mathbf{K}^0$ and the local Fourier law

$$\mathbf{q}(\mathbf{x}) = \mathbf{K}^0[\mathbf{e}(\mathbf{x}) - \mathbf{e}^*(\mathbf{x})]. \tag{3}$$

The infinite sum in (1) involve all wave vector $\boldsymbol{\xi}$ with components $\xi_i$:

$$\xi_i = \frac{2\pi z_{i,n}}{a_i}, \quad z_{i,n},n \in \mathbb{Z}, \quad i = 1,2,3. \tag{4}$$

Since the solution $\mathbf{e}^*(\mathbf{x})$ is linear in $\mathbf{E}$, we can write

$$\frac{1}{V}\int_V \mathbf{e}^*(\mathbf{x}) = \mathbf{L}\mathbf{E} \tag{5}$$

and derive the effective conductivity of the material by the expression

$$\mathbf{K}^{\text{eff}} = \mathbf{K}^0\,(\mathbf{I} - \mathbf{L})\,. \tag{6}$$

It is worthwhile noticing that the reference material may be isotropic even if the overall material is anisotropic. The purpose of the paper is in closed form solutions of the previous integral equation. However, a classical way to obtain the solution of this integral equation is to use an iterative numerical solution. In the following, this numerical solution, whose description is shortly recalled in Appendix A, is used as a reference whose results are compared with the closed form solutions obtained thereafter.

### B. A formula for the effective conductivity of matrix-inclusion composites

We shall now use (1) to study the main problem where the composite is a mixture of hard particles embedded in a matrix. The particles are classified into $m$ groups of species (denoted thereafter species) according to their geometry and material. Assuming that the conductivity of each phase is isotropic and using the matrix conductivity $k_0$ as the reference value, Eq. (1) is then reduced to $m$ integral equations:

$$\mathbf{E} + \sum_{\boldsymbol{\xi}\neq\mathbf{0}} e^{i\boldsymbol{\xi}\cdot\mathbf{x}}\boldsymbol{\Gamma}(\boldsymbol{\xi})\left[\frac{1}{V}\int_V \mathbf{e}^*(\mathbf{x}')e^{-i\boldsymbol{\xi}\cdot\mathbf{x}'}\,d\mathbf{x}'\right] = \frac{k_0}{k_0 - k_\alpha}\mathbf{e}^*(\mathbf{x}), \quad \forall\mathbf{x}\in\Omega_\alpha, \quad i = 1,2,..,m, \tag{7}$$

for each group domain $\Omega_\alpha$ of conductivity $k_\alpha$ and volume fraction $f_\alpha$. Making use of the fact that, due to the choice of reference material, $\mathbf{e}^*$ vanishes in the matrix phase, we integrate both sides of (7) over domain $\Omega_\alpha$ and obtain

$$\Omega_\alpha\mathbf{E} + \frac{1}{V}\sum_{\boldsymbol{\xi}\neq\mathbf{0}}\boldsymbol{\Gamma}(\boldsymbol{\xi})\int_{\Omega_\alpha} e^{i\boldsymbol{\xi}\cdot\mathbf{x}}\,d\mathbf{x}\sum_\beta\int_{\Omega_\beta}\mathbf{e}^*(\mathbf{x}')e^{-i\boldsymbol{\xi}\cdot\mathbf{x}'}\,d\mathbf{x}' = \frac{k_0}{k_0 - k_\alpha}\int_{\Omega_\alpha}\mathbf{e}^*(\mathbf{x})\,d\mathbf{x}, \quad \alpha = 1,2,\ldots,m. \tag{8}$$

At this step, we evaluate the integral $\int_{\Omega_\alpha}\mathbf{e}^*(\mathbf{x})e^{i\boldsymbol{\xi}\cdot\mathbf{x}}\,d\mathbf{x}$ by using the approximation [21,22]

$$\int_{\Omega_\alpha}\mathbf{e}^*(\mathbf{x})e^{i\boldsymbol{\xi}\cdot\mathbf{x}}\,d\mathbf{x} \simeq I_\alpha(\boldsymbol{\xi})\mathbf{L}_\alpha\mathbf{E}, \tag{9}$$

with

$$I_\alpha(\boldsymbol{\xi}) = \int_{\Omega_\alpha} e^{i\boldsymbol{\xi}\cdot\mathbf{x}}\,d\mathbf{x}, \quad \frac{1}{\Omega_\alpha}\int_{\Omega_\alpha}\mathbf{e}^*(\mathbf{x})\,d\mathbf{x} = \mathbf{L}_\alpha\mathbf{E}. \tag{10}$$

In the case of dilute ellipsoidal inclusions, the approximation (9) is exact since $\mathbf{e}^*$ is uniform inside the inclusion [25–29]. For a moderate volume fraction range below the percolation threshold, the simplification (9) was shown to perform largely better [21] than in the dilute case. Indeed, it is based on average approximation and does not strictly require the uniformity of $\mathbf{e}^*$ contrarily to the dilute case. Obviously this procedure recovers the same results as those coming from the dilute case at low concentrations.

The presence of the second order tensor $\mathbf{L}_\alpha$ associated with phase $\alpha$ in (10) is due to the linearity of the problem in $\mathbf{E}$. Substituting (9) into (7) yields a system of $m$ linear equations for $\mathbf{L}_\alpha$:

$$\frac{k_0\mathbf{L}_\alpha}{k_0 - k_\alpha} - \sum_\beta \mathbf{A}_{\alpha\beta}\mathbf{L}_\beta = \mathbf{I},$$

$$\mathbf{A}_{\alpha\beta} = \frac{1}{f_\alpha V^2}\sum_{\boldsymbol{\xi}\neq\mathbf{0}}\overline{\boldsymbol{\xi}}\otimes\overline{\boldsymbol{\xi}}\,I_\alpha(\boldsymbol{\xi})I_\beta(-\boldsymbol{\xi}), \tag{11}$$

where $\mathbf{I}$ is the second order identity tensor. After solving (11) to obtain all tensors $\mathbf{L}_\alpha$, the effective conductivity of the mixture can be computed from

$$\mathbf{K}^{\text{eff}} = k_0 \left( \mathbf{I} - \sum_\alpha f_\alpha \mathbf{L}_\alpha \right). \tag{12}$$

As an illustration, we consider a mixture of two species ($m = 2$). Inverting (11) yields the following expression for localization tensors $\mathbf{L}_1$ and $\mathbf{L}_2$:

$$\begin{aligned} \mathbf{L}_\alpha = & \left[ \mathbf{A}_{\alpha\beta}^{-1} \left( \frac{k_0\mathbf{I}}{k_0 - k_\alpha} - \mathbf{A}_{\alpha\alpha} \right) - \left( \frac{k_0\mathbf{I}}{k_0 - k_\beta} - \mathbf{A}_{\beta\beta} \right)^{-1} \mathbf{A}_{\beta\alpha} \right]^{-1} \\ & \times \left[ \mathbf{A}_{\beta\alpha}^{-1} + \left( \frac{k_0\mathbf{I}}{k_0 - k_\alpha} - \mathbf{A}_{\alpha\alpha} \right)^{-1} \right], \\ & \alpha \neq \beta, \quad \alpha,\beta = 1,2. \end{aligned} \tag{13}$$

### C. Effective conductivity for specific symmetries of the periodic cell

#### 1. Orthotropic cells

If the microstructure takes the three planes $x_i = 0$ ($i = 1,2,3$) as planes of symmetry, the material has the orthotropic symmetry. In this case, all tensors $\mathbf{A}_{\alpha\beta}$ and $\mathbf{L}_\alpha$ are diagonal in the current basis $Ox_1x_2x_3$, and mathematical operations with them are highly simplified. For example, the multiplication can be done without respecting their left-right order,

$$\mathbf{AB} = \mathbf{BA}, \tag{14}$$

and the fractional notation can be used without any ambiguity:

$$\mathbf{AB}^{-1} = \mathbf{B}^{-1}\mathbf{A} = \mathbf{A}/\mathbf{B} = \frac{\mathbf{A}}{\mathbf{B}}. \tag{15}$$

As a result, we can obtain $\mathbf{K}^{\text{eff}}$ from Eq. (12)

$$\frac{\mathbf{K}^{\text{eff}}}{k_0} = \mathbf{I} - \frac{\left(\frac{k_0\mathbf{I}}{k_0 - k_2} - \mathbf{A}_{22} + \mathbf{A}_{12}\right) f_1 + \left(\frac{k_0\mathbf{I}}{k_0 - k_1} - \mathbf{A}_{11} + \mathbf{A}_{21}\right) f_2}{\left(\frac{k_0\mathbf{I}}{k_0 - k_2} - \mathbf{A}_{22}\right)\left(\frac{k_0\mathbf{I}}{k_0 - k_1} - \mathbf{A}_{11}\right) - \mathbf{A}_{12}\mathbf{A}_{21}}. \tag{16}$$

#### 2. Cubic cells

In a more special case where the distribution of all heterogeneous phases are symmetrical with respect to nine planes $x_i = 0$ and $x_i \pm x_j = 0$ with $i,j = 1,2,3$, the overall conductivity is isotropic. Indeed, it can be shown that for all $\alpha$, $I_\alpha(\boldsymbol{\xi})$ is symmetrical with respect to nine planes $\xi_i = 0$ and $\xi_i \pm \xi_j = 0$ with $i,j = 1,2,3$. The following relation can be deduced immediately:

$$\mathbf{A}_{\alpha\beta} = A_{\alpha\beta}\mathbf{I}, \quad A_{\alpha\beta} = \frac{1}{3 f_\alpha V^2} \sum_{\boldsymbol{\xi} \neq \mathbf{0}} I_\alpha(\boldsymbol{\xi}) I_\beta(-\boldsymbol{\xi}). \tag{17}$$

Tensors $\mathbf{L}_\alpha$ are also scalar matrices, say, $\mathbf{L}_\alpha = L_\alpha \mathbf{I}$, with $L_\alpha$ being determined from

$$\begin{bmatrix} \frac{k_0}{k_0 - k_1} - A_{11} & \cdots & -A_{1m} \\ \cdots & \cdots & \cdots \\ -A_{m1} & \cdots & \frac{k_0}{k_0 - k_m} - A_{mm} \end{bmatrix} \begin{bmatrix} L_1 \\ \cdots \\ L_m \end{bmatrix} = \begin{bmatrix} 1 \\ \cdots \\ 1 \end{bmatrix}. \tag{18}$$

As a result, the effective conductivity is isotropic and equal to

$$\mathbf{K}^{\text{eff}} = k^{\text{eff}}\mathbf{I}, \quad \frac{k^{\text{eff}}}{k_0} = 1 - \sum_\alpha f_\alpha L_\alpha. \tag{19}$$

For a mixture of two species ($m = 2$), the effective conductivity finally reads

$$\frac{k^{\text{eff}}}{k_0} = 1 - \frac{\left(\frac{k_0}{k_0 - k_2} - A_{22} + A_{12}\right) f_1 + \left(\frac{k_0}{k_0 - k_1} - A_{11} + A_{21}\right) f_2}{\left(\frac{k_0}{k_0 - k_2} - A_{22}\right)\left(\frac{k_0}{k_0 - k_1} - A_{11}\right) - A_{12}A_{21}}. \tag{20}$$

In the following applications, we shall focus on materials possessing three or nine planes of symmetry and be limited to expressions (16) and (16) with the main concern being the evaluation of $\mathbf{A}_{\alpha\beta}$ and $A_{\alpha\beta}$.

## III. EXPLICIT SOLUTIONS FOR PERIODIC ARRAYS OF SPHERES AND ELLIPSOIDS

The heterogeneous material is constituted from an orthorhombic arrangement of polydispersed spheres or ellipsoids, in which the final effective conductivity can be either isotropic or anisotropic. For the sake of clarity, we consider only body-centered arrangements and treat separately the isotropy and anisotropy effect in different subsections.

### A. Cubic lattice arrangement of spheres

The basic cell is a cube of side length $a$ containing a sphere of radius $R_1$ and conductivity $k_1$ located at the center and eight eighths of a sphere of radius $R_2$ and conductivity $k_2$ at the eight corners [see Fig. 1(a)]. In this case, the number of (inclusion) species is $m = 2$, and the characteristic functions of phases $I_1(\boldsymbol{\xi})$ and $I_2(\boldsymbol{\xi})$ are given by, for the sphere at the center,

$$I_1(\boldsymbol{\xi}) = 3V_{s1}h(\eta_1), \tag{21}$$

and for the spheres at the corner,

$$I_2(\boldsymbol{\xi}) = 3V_{s2}h(\eta_2)(-1)^{z_1+z_2+z_3} \tag{22}$$

with

$$h(\eta) = \frac{\sin\eta - \eta\cos\eta}{\eta^3}, \quad \eta_\alpha = \xi R_\alpha,$$
$$V_{s\alpha} = \frac{4\pi}{3} R_\alpha^3, \quad \alpha = 1,2, \tag{23}$$

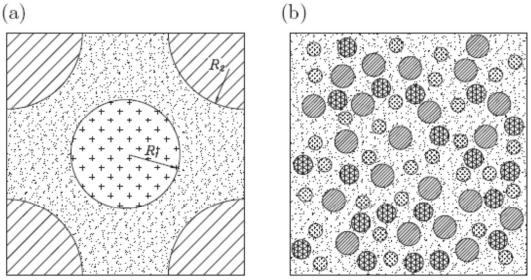


FIG. 1. 2D images of RVEs with polydispersed hard aggregates (left: orthorhombic body centered arrangement, right: random distribution).

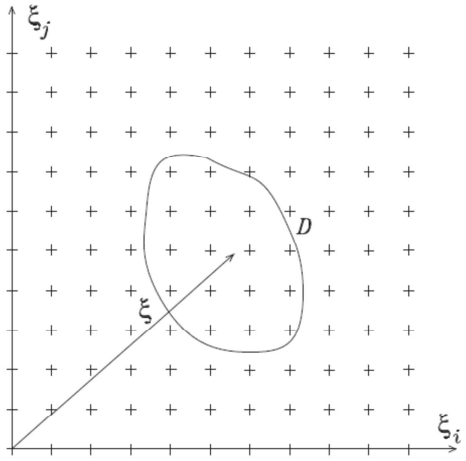


FIG. 2. Grid of $\xi$ in 2D. The lattice sum $\sum_{\xi\in D} I_\alpha(\xi)I_\beta(-\xi)$ in the volume element $D$ is approximated by $\rho\int_D P(\xi)\,dV$.

where $\xi$ is the modulus of $\boldsymbol{\xi}$.

Coefficients $A_{\alpha\beta}$ can be computed either numerically or analytically by using an extension of the method presented in Ref. [21]. The former involves a summation over all $\boldsymbol{\xi}\neq\mathbf{0}$ as defined in (17). In the latter approach, we make use of the fact that the $\boldsymbol{\xi}$ points form a regular lattice in $\boldsymbol{\xi}$ space with density $\rho=(2\pi/a)^3$ and $I_\alpha(\boldsymbol{\xi})I_\beta(-\boldsymbol{\xi})$ is a decaying function with $\boldsymbol{\xi}$.

Then, $I_\alpha(\boldsymbol{\xi})I_\beta(-\boldsymbol{\xi})$ can be written as $I_\alpha(\boldsymbol{\xi})I_\beta(-\boldsymbol{\xi})=h(\xi R_\alpha)h(\xi R_\beta)\Phi_{\alpha\beta}$ where $\Phi_{\alpha\beta}$ accounts for the fluctuating term $(-1)^{z_1+z_2+z_3}$ in $I_2$.

For a sufficiently remote and large domain $D$ in Fourier space, we estimate the infinite sum (see Fig. 2) by

$$\sum_{\xi\in D} I_\alpha(\boldsymbol{\xi})I_\beta(-\boldsymbol{\xi})\simeq\rho\int_D P_{\alpha\beta}(\boldsymbol{\xi})\,dV,$$
$$P_{\alpha\beta}(\boldsymbol{\xi})=h(\xi R_\alpha)h(\xi R_\alpha)\overline{\Phi}_{\alpha\beta}, \tag{24}$$

where $\overline{\Phi_{\alpha\beta}}$ is the average value of the fluctuating term $\Phi_{\alpha\beta}$ over $D$,which is equal to $\Phi_{\alpha\beta}=\delta_{\alpha\beta}$ for the centered cubic arrangement. As a result, we have

$$P_{11}(\boldsymbol{\xi})=9V_{s1}^2h^2(\eta_1),\quad P_{22}(\boldsymbol{\xi})=9V_{s2}^2h^2(\eta_2),$$
$$P_{12}(\boldsymbol{\xi})=P_{21}(\boldsymbol{\xi})=0. \tag{25}$$

Finally the computation of the remainder of the lattice sums for main terms $P_{\alpha\alpha}$ are similar to the ones obtained in the monodispersed case, and the computation of the coupling terms $P_{\alpha\beta},\alpha\neq\beta$ shows that the lattice sum is more rapidly convergent than for main terms, the complementary integral being null.

The value of $P_{\alpha\beta}(\boldsymbol{\xi})$ depends uniquely on $\xi$ and will be denoted as $P_{\alpha\beta}(\xi)$ afterwards. Certainly, the above approximation is not valid for $D$ close to the origin. Then the infinite sum $\sum_{\xi\neq\mathbf{0}} I_\alpha(\boldsymbol{\xi})I_\beta(-\boldsymbol{\xi})$ is best approximated by keeping the important leading terms, while estimating the remainder lattice sum by using a continuous integral in the $\boldsymbol{\xi}$ space:

$$\sum_{\xi\neq\mathbf{0}} I_\alpha(\boldsymbol{\xi})I_\beta(-\boldsymbol{\xi})\simeq\sum_{|\xi|<\xi_c} I_\alpha(\boldsymbol{\xi})I_\beta(-\boldsymbol{\xi})$$
$$+\frac{a^3}{2\pi^2}\int_{\xi_c}^{\infty} P_{\alpha\beta}(\xi)\xi^2\,d\xi. \tag{26}$$

In (26), the cutoff distance $\xi_c$ determines the number of leading terms kept in the series. From the values of $P_{\alpha\beta},\alpha\neq\beta$, it comes that, for the coupling terms, the contribution of the integral over domain $D$ to the evaluation of the lattice sum is strictly null. Therefore, the evaluation of the lattice sum can be confined to the terms close to the origin.

Denoting $\epsilon_\alpha=2\pi R_\alpha/a$, $\alpha=1,2$ and keeping four leading terms, which has been found satisfactory, yields the following expressions:

$$A_{\alpha\alpha}\simeq f_\alpha A^*(\epsilon_\alpha)+\varphi(2\epsilon_\alpha),$$
$$\frac{A_{12}}{f_2}=\frac{A_{21}}{f_1}\simeq B^*(\epsilon_1,\epsilon_2), \tag{27}$$

in which $A^*$ and $B^*$ are given by

$$A^*(\epsilon)=18h^2(\epsilon)+36h^2(\sqrt{2}\epsilon)+24h^2(\sqrt{3}\epsilon)$$
$$+18h^2(\sqrt{3}\epsilon),$$
$$B^*(\epsilon_1,\epsilon_2)=-18h(\epsilon_1)h(\epsilon_2)+36h[\sqrt{2}\epsilon_1)h(\sqrt{2}\epsilon_2]$$
$$-24h(\sqrt{3}\epsilon_1)h(\sqrt{3}\epsilon_2)+18h(2\epsilon_1)h(2\epsilon_2), \tag{28}$$

and the function $\varphi$ is defined as

$$\varphi(\epsilon)=\frac{3-\cos 2\epsilon}{3\pi\epsilon}+\frac{2\sin^2\epsilon}{3\pi\epsilon^3}-\frac{2\sin 2\epsilon}{3\pi\epsilon^2}-\frac{2\mathrm{Si}(2\epsilon)}{3\pi}+\frac{1}{3}. \tag{29}$$

Finally, the effective conductivity is given by relation (20) in which coefficients $A_{\alpha\beta}$ are given by relations (27) and (28).

The results for the effective conductivity related to the previously defined polydispersed structure are shown in Fig. 3. The results show that the present estimation scheme agrees very well with the exact result issued from the complete Fourier

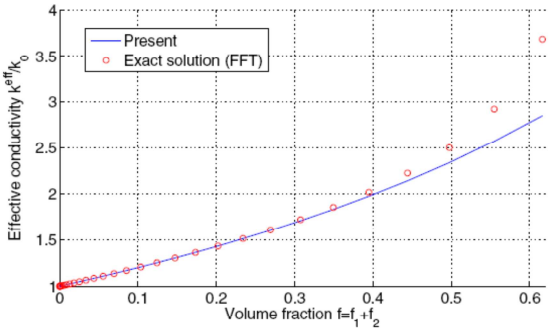


FIG. 3. (Color online) Comparison between present estimation [Eq. (16)] and exact solution (Fourier transform method with resolution $128\times128\times128$). The microstructure is body-centered cubic with $R_1=2R_2$, $k_1=10k_0$, and $k_2=0.1k_0$. The maximal volume fraction is 0.62 when the spheres touch each other.

transform-based method [7] described in Appendix A, for a large range of volume fractions, up to $f = 0.4$.

### B. Microstructures with anisotropy effects

We study two examples based on body-centered arrangements of orthorhombic type possessing three planes of symmetry. In these cases, the approximation of the lattice sum with continuous integral as done in the previous section yield closed form expressions for $\mathbf{A}_{\alpha\beta}$.

#### *1. Example 1 (spherical inclusions and parallelepipedic box)*

The unit cell is a parallelepipedic box of dimensions $a_1 \times a_2 \times a_3$ with spherical inclusions of radius $R_1$ and $R_2$ (two species). The expressions for $I_\alpha(\boldsymbol{\xi})$ and $P_{\alpha\beta}(\boldsymbol{\xi})$ are unchanged with respect to the cubic lattice case, the same as Eqs. (21)–(25). A similar lattice sum approximation as Eq. (24) and Fig. 2 is also applicable in this situation. The only difference is that we have a parallelepipedic grid with the density $\rho = a_1 a_2 a_3 / 8\pi^3$. The explicit expressions for $\mathbf{A}_{\alpha\beta}$ taking $\xi_c$ as a cutoff radius become

$$\mathbf{A}_{\alpha\beta} \simeq 9 f_\beta \sum_{|\boldsymbol{\xi}|<\xi_c} \overline{\boldsymbol{\xi}} \otimes \overline{\boldsymbol{\xi}} h(\eta_\alpha) h(\eta_\beta) (-1)^{z_1+z_2+z_3}, \quad \alpha \neq \beta,$$

$$\mathbf{A}_{\alpha\alpha} \simeq 9 f_\alpha \sum_{|\boldsymbol{\xi}|<\xi_c} \overline{\boldsymbol{\xi}} \otimes \overline{\boldsymbol{\xi}} h^2(\eta_\alpha) + \varphi(\xi_c R_\alpha)\mathbf{I}. \tag{30}$$

We remark that the nondiagonal terms of $\mathbf{A}_{\alpha\beta}$ are mainly due to the leading terms in the lattice sum, as for the isotropic case.

#### *2. Example 2 (spheroidal inclusions and parallelepipedic box)*

In the second example, the unit cell is a parallelepipedic box of dimensions $a_1 \times a_2 \times a_3$, but the inclusions are spheroids with the same semiaxes ratio and with $x_1$ being the revolution axis. Denoting $R_\alpha/\chi, R_\alpha$ and $R_\alpha$ as semiaxes of species $\alpha$, $\alpha = 1,2$, the shape functions of the two species admit the following expression:

$$I_1(\boldsymbol{\xi}) = 3V_{e1} h(\eta_1), \quad I_2(\boldsymbol{\xi}) = 3V_{e2} h(\eta_2)(-1)^{z_1+z_2+z_3},$$
$$\eta_\alpha = R_\alpha \sqrt{\xi_1^2/\chi^2 + \xi_2^2 + \xi_3^2}, \quad V_{e\alpha} = \frac{4\pi}{3\chi} R_\alpha^3. \tag{31}$$

By making the following homothetic change:

$$\xi_1' = \xi_1/\chi, \quad \xi_2' = \xi_2, \quad \xi_3' = \xi_3, \tag{32}$$

the lattice sum in $\boldsymbol{\xi}$ space can be converted to $\boldsymbol{\xi}'$ space. With the help of Appendix B, the analytical approximations of the matrices $\mathbf{A}_{\alpha\alpha}$ and $\mathbf{A}_{\alpha\beta}$ lead to the following expressions:

$$(\mathbf{A}_{\alpha\alpha})_{11} \simeq 9 f_\alpha \sum_{\xi'<\xi_c'} \frac{\chi^2 \xi_1'^2 h^2(R_\alpha \xi')}{\chi^2 \xi_1'^2 + \xi_2'^2 + \xi_3'^2} + 3C_1 \varphi(R_\alpha \xi_c),$$

$$(\mathbf{A}_{\alpha\alpha})_{22} \simeq 9 f_\alpha \sum_{\xi'<\xi_c'} \frac{\xi_2'^2 h^2(R_\alpha \xi')}{\chi^2 \xi_1'^2 + \xi_2'^2 + \xi_3'^2} + 3C_2 \varphi(R_\alpha \xi_c),$$

$$(\mathbf{A}_{\alpha\alpha})_{33} \simeq 9 f_\alpha \sum_{\xi'<\xi_c'} \frac{\xi_3'^2 h^2(R_\alpha \xi')}{\chi^2 \xi_1'^2 + \xi_2'^2 + \xi_3'^2} + 3C_3 \varphi(R_\alpha \xi_c), \tag{33}$$

and

$$(\mathbf{A}_{\alpha\beta})_{11} \simeq 9 f_\beta \sum_{\xi'<\xi_c'} \frac{\chi^2 \xi_1'^2 h(R_\alpha \xi') h(R_\beta \xi')}{\chi^2 \xi_1'^2 + \xi_2'^2 + \xi_3'^2} (-1)^{z_1+z_2+z_3},$$

$$(\mathbf{A}_{\alpha\beta})_{22} \simeq 9 f_\beta \sum_{\xi'<\xi_c'} \frac{\xi_2'^2 h(R_\alpha \xi') h(R_\beta \xi')}{\chi^2 \xi_1'^2 + \xi_2'^2 + \xi_3'^2} (-1)^{z_1+z_2+z_3},$$

$$(\mathbf{A}_{\alpha\beta})_{33} \simeq 9 f_\beta \sum_{\xi'<\xi_c'} \frac{\xi_3'^2 h(R_\alpha \xi') h(R_\beta \xi')}{\chi^2 \xi_1'^2 + \xi_2'^2 + \xi_3'^2} (-1)^{z_1+z_2+z_3}. \tag{34}$$

The factors $C_1, C_2$, and $C_3$ depend on the shape of the inclusion.

For oblate inclusions ($\chi > 1$),

$$C_2 = C_3 = \frac{1}{2\gamma^3}(\lambda\gamma^2 + \lambda - \gamma), \quad C_1 = \frac{\chi^2}{\gamma^3}(\gamma - \lambda), \tag{35}$$

with $\gamma = \sqrt{\chi^2 - 1}$ and $\lambda = \arctan \gamma$.

For prolate inclusions ($\chi < 1$),

$$C_2 = C_3 = \frac{1}{2\delta^3}(\mu - \mu\delta^2 - \delta), \quad C_1 = \frac{\chi^2}{\delta^3}(\mu - \delta), \tag{36}$$

with $\delta = \sqrt{1 - \chi^2}$ and $\mu = \tanh^{-1} \delta$.

It is interesting to remark that $C_1$, $C_2$, and $C_3$ given by (35) and (36) are identical to depolarization factors along directions 1, 2, and 3 of spheroidal inclusions [30].

From results in Figs. 4 and 5, the present estimation recovers the anisotropy effect due to the material distribution in both cases and agrees well with the exact solutions issued from the FFT iterative scheme, the departure from the FFT solution appearing, however, at a lower concentration for the case of the spheres in a parallelepipedic box. For spherical inclusions, the anisotropy is weaker and becomes much more significant at high volume fraction. In this case, the direction dependent conduction is due to the arrangement of the inclusions and not to their shapes. In the case of ellipsoidal inclusions, the anisotropic effect is much stronger, and it is already considerable at small volume fraction.

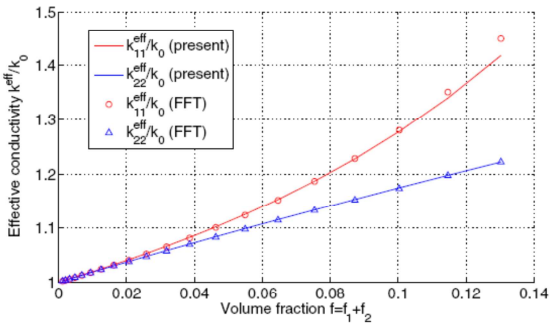


FIG. 4. (Color online) Example 1: Effective conductivity along two different directions $x_1, x_2$ estimated by Eq. (16). The unit cell is parallelepedic box of dimension $1 \times 2 \times 2$ with the spheres of properties $R_1 = 2R_2$, $k_1 = 10k_0$, and $k_2 = 0.1k_0$.

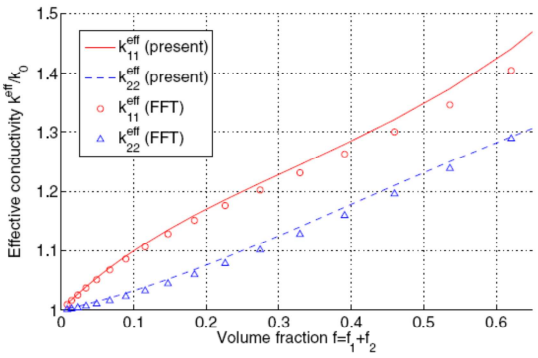


FIG. 5. (Color online) Example 2: Effective conductivity along two different directions $x_1$,$x_2$ estimated by Eq. (16). The unit cell is parallelepedic box of dimension $2 \times 1 \times 1$ with ellipsoidal inclusions. The ellipsoids are identical in size and shape with the semiaxes $2R$, $R$, and $R$ (i.e., $R_1 = R_2 = R$, $\chi = 0.5$) but different in properties $k_1 = 10k_0$ and $k_2 = 0.1k_0$.

## IV. RANDOM ISOTROPIC DISTRIBUTIONS OF POLYDISPERSED HARD SPHERES

The case of random distributions of polydispersed hard spheres represents an approximation of many situations of practical interest. Let us therefore consider, inside a sufficiently large cubic volume $V$ of size $a$, a set of $N$ hard spheres of different families $\alpha$, associated with particle numbers $N_\alpha$ and fraction numbers $n_\alpha$, which are randomly distributed [see Fig. 1(b)]. These quantities and the volume fractions $f_\alpha$ are interconnected via the relations

$$N = \sum_\alpha N_\alpha, \quad n_\alpha = \frac{N_\alpha}{N}, \quad f_\alpha = \frac{N_\alpha V_{s\alpha}}{V}. \tag{37}$$

First, we remark that the partial structure factor $S_{\alpha\beta}$ between two species $\alpha$ and $\beta$ is defined from the expression [31]

$$S_{\alpha\beta}(\boldsymbol{\xi}) = \frac{1}{N}\left\langle \sum_i^{N_\alpha} e^{i\boldsymbol{\xi}\cdot\mathbf{x}_i} \sum_j^{N_\beta} e^{-i\boldsymbol{\xi}\cdot\mathbf{x}_j} \right\rangle \tag{38}$$

and related to the partial pair distribution function $g_{\alpha\beta}$ as follows:

$$S_{\alpha\beta}(\boldsymbol{\xi}) = n_\alpha\delta_{\alpha\beta} + n_\alpha n_\beta \rho \int_V [g_{\alpha\beta}(\mathbf{r}) - 1] e^{-i\boldsymbol{\xi}\cdot\mathbf{r}}\, d\mathbf{r}. \tag{39}$$

In (38) and (39), $\langle..\rangle$ denotes the ensemble average, $\delta_{\alpha\beta}$ the Kronecker delta and $\mathbf{x}_i$ and $\mathbf{x}_j$ the sphere centers. Assuming that the distributions of the spherical inclusions are isotropic (for all species) and adopting the change of variable $\bar{r} = r/R_\beta$, we obtain

$$\frac{S_{\alpha\beta}(\xi)}{n_\alpha} = \delta_{\alpha\beta} + 3f_\beta \int_0^\infty [g_{\alpha\beta}(r) - 1] \frac{\sin\eta_\beta\bar{r}}{\eta_\beta} \bar{r}\, d\bar{r}. \tag{40}$$

For randomly heterogeneous materials, the terms $L_\alpha$ in (19) can be replaced by $\langle L_\alpha\rangle$. However, in the framework of ergodic media [20], it is reasonable to assume that $L_\alpha$ for each phase is stationary in the infinite volume limit and equal to $\langle L_\alpha\rangle$, which leads to the following approximation:

$$\langle L_\alpha\rangle \simeq L_\alpha, \quad \langle A_{\alpha\beta}L_\beta\rangle \simeq \langle A_{\alpha\beta}\rangle L_\beta, \quad \forall\alpha,\beta. \tag{41}$$

Now, the determination of $L_\alpha$ can be effected, based on (18) using $\langle A_{\alpha\beta}\rangle$ instead of $A_{\alpha\beta}$. From another point of view, the characteristic function $I_\alpha(\boldsymbol{\xi})$ of phase $\alpha$ in one sample takes the form

$$I_\alpha(\boldsymbol{\xi}) = 3V_{s\alpha} \frac{\sin\eta_\alpha - \eta_\alpha\cos\eta_\alpha}{\eta_\alpha^3} \sum_i^{N_\alpha} e^{i\boldsymbol{\xi}\cdot\mathbf{x}_i}. \tag{42}$$

Therefore, based on (38), the ensemble average of (17) can be rewritten as follows:

$$A_{\alpha\beta} \simeq \sum_{\xi\neq 0} \frac{3V_{s\beta}}{n_\alpha V} \frac{(\sin\eta_\alpha - \eta_\alpha\cos\eta_\alpha)}{\eta_\alpha^3} \times \frac{(\sin\eta_\beta - \eta_\beta\cos\eta_\beta)}{\eta_\beta^3} S_{\alpha\beta}(\xi). \tag{43}$$

In the infinite volume limit, (17) admits the integral form

$$\langle A_{\alpha\beta}\rangle \simeq \frac{2}{\pi}\int_0^\infty \frac{(\sin\eta_\alpha - \eta_\alpha\cos\eta_\alpha)}{\eta_\alpha^3} \times \frac{(\sin\eta_\beta - \eta_\beta\cos\eta_\beta)}{\eta_\beta} \frac{S_{\alpha\beta}(\xi)}{n_\alpha}\, d\eta_\beta. \tag{44}$$

It is clear that expression (44) could be derived from (43) as a limit case where the lattice point becomes infinitely dense and (26) holds true for all domain $D$. Since, from the hard sphere assumption, the spheres are nonoverlapping, $g_{\alpha\beta}(r)$ must vanish for $r \leqslant R_\alpha + R_\beta$.

Substituting (40) into (44) and making use of the fact that [32]

$$\int_0^\infty \frac{(\sin\eta_\alpha - \eta_\alpha\cos\eta_\alpha)}{\eta_\alpha^3} \frac{(\sin\eta_\beta - \eta_\beta\cos\eta_\beta)}{\eta_\beta^2} \sin\eta_\beta\bar{r}\, d\eta_\beta = 0, \quad \forall\bar{r} \geqslant 1 + \gamma_{\alpha\beta} \quad \text{and} \quad \gamma_{\alpha\beta} = R_\alpha/R_\beta = n_\alpha/n_\beta \tag{45}$$

we obtain finally a remarkably simple expression for $\langle A_{\alpha\beta}\rangle$:

$$\langle A_{\alpha\beta}\rangle = \frac{\delta_{\alpha\beta} - f_\beta}{3}. \tag{46}$$

It implies that, based on (18), (19), and (46), we obtain

$$\begin{bmatrix} \frac{k_0}{k_0-k_1} - \frac{1-f_1}{3} & \cdots & \frac{f_m}{3} \\ \cdots & \cdots & \cdots \\ \frac{f_1}{3} & \cdots & \frac{k_0}{k_0-k_m} - \frac{1-f_m}{3} \end{bmatrix} \begin{bmatrix} L_1 \\ \cdots \\ L_m \end{bmatrix} = \begin{bmatrix} 1 \\ \cdots \\ 1 \end{bmatrix}. \tag{47}$$

Combining (47) and (19) and after some algebraic manipulations, the effective conductivity can be recast in the following form:

$$\frac{k^{\text{eff}}}{k_0} = \frac{1 + 2\sum_\alpha \frac{k_\alpha/k_0 - 1}{2 + k_\alpha/k_0} f_\alpha}{1 - \sum_\alpha \frac{k_\alpha/k_0 - 1}{2 + k_\alpha/k_0} f_\alpha}. \tag{48}$$

This final formula is explicit, and it is interesting to remark that it depends only on volume fraction regardless the local structure of the particles $S_{\alpha\beta}(\xi)$ or $g_{\alpha\beta}(r)$.

Another fundamental remark is that this last formula is strictly identical to the one obtained by using the Mori-Tanaka scheme for multicomponent composites [18], a generalization of the Clausius-Mossotti approach. However, in our derivation, there is no "ad hoc" assumption as the one described in the introduction: our derivation is based essentially on statistical assumptions.

## V. FINAL REMARKS AND DISCUSSION

In this paper, we have computed the effective conductivity of multicomponent composite materials. The proposed computation scheme is directly based on an integral equation for the *free temperature gradient* and has many points in common with the collocation resolution method. The present work has generalized the previous results concerning two phase composites [21,22] by obtaining closed form solutions related to polydispersed distributions in the case of periodic inclusion-matrix structures of orthotropic symmetry containing spherical or spheroidal inclusions and also in the case of isotropic random distributions. Compared to previous results obtained in Ref. [21], the paper has shown how the closed form approximation of the coupling terms can be effected. In addition, it provides closed form solutions in the case of spheroidal inclusions. Our results show that the closed-form solution agrees quite well with numerical solution obtained by the numerical FFT method which is used as a reference, being able to obtain the solution at any order of accuracy.

In the latter case of a random distribution, the final solution has been found by using the statistical information on the material, including radial distribution function and structure factor. In the homogenization limit, a very simple closed form is obtained and is found to be identical to Mori-Tanaka scheme for multicomponent composites [18,33] and independent of the local distribution of the particles $S_{\alpha\beta}(\xi)$ or $g_{\alpha\beta}(r)$. This result gives a strong basis to the Mori-Tanaka solution, which rests usually only on "ad hoc" assumptions.

This last result comes from the assumption of an isotropic distribution of hard sphere particles. Nevertheless, anisotropy effects can also be treated within the framework of the present method, using directly the ensemble average definition of $\langle \mathbf{A}_{\alpha\beta} \rangle$ in (11) and $S(\boldsymbol{\xi})$ in (38). Since realistic anisotropic ensembles can be generated by available atomistic simulation techniques [5] in suitable conditions, the determination of the effective conductivity is feasible and shall be addressed in a future work.


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## APPENDIX A: INTEGRAL EQUATION AND ITS CLASSICAL NUMERICAL SOLUTION USING AN ITERATIVE SCHEME

We note that in Fourier transform-based numerical methods, an equivalent equation to (1) is used:

$$\mathbf{e}(\mathbf{x}) = \mathbf{E} + \mathbf{G} * \delta\mathbf{K}\mathbf{e}(\mathbf{x}), \quad \delta\mathbf{K} = [\mathbf{K}^0 - \mathbf{K}(\mathbf{x})], \tag{A1}$$

where $\mathbf{G} = \boldsymbol{\Gamma}(\mathbf{K}^0)^{-1}$ The solution procedure is to use a fixed point recurrence for solving (A1) from the initial field $\mathbf{e}^0(\mathbf{x}) = \mathbf{E}$,

$$\mathbf{e}^N(\mathbf{x}) = \mathbf{E} + \mathbf{G} * \delta\mathbf{K}\mathbf{e}^{N-1}(\mathbf{x}), \tag{A2}$$

until convergence. At each step, the evaluation of $\boldsymbol{\Gamma}*$ (periodic convolution integral) is done using the fast Fourier transform (FFT) and its inverse [7]. The computation domains in the Fourier and real space are meshed with voxel elements related to a given resolution. The basic iterative scheme (A2) is not the unique way to solve (A1). It is well known that the convergence rate is sensitive to the contrast ratio and to the choice of the reference materials. Alternative schemes have been proposed to improve the basic scheme [15,34] in some aspects, but the convergence is always difficult in the case of high contrast between physical properties.

## APPENDIX B: EVALUATION OF INTEGRALS

The analytical approximation throughout the paper requires the evaluation of the following integral:

$$\begin{aligned} Z_1 &= 9 f_\alpha \rho' \int_{\xi' > \xi'_c} \frac{\chi^2 \xi_1'^2 h^2(R_\alpha \xi') d\boldsymbol{\xi}'}{\chi^2 \xi_1'^2 + \xi_2'^2 + \xi_3'^2}, \\ Z_k &= 9 f_\alpha \rho' \int_{\xi' > \xi'_c} \frac{\xi_k'^2 h^2(R_\alpha \xi') d\boldsymbol{\xi}'}{\chi^2 \xi_1'^2 + \xi_2'^2 + \xi_3'^2} \quad \text{with} \quad k = 2,3, \end{aligned} \tag{B1}$$

where $\rho'$ is the density of the wave vector $\boldsymbol{\xi}'$:

$$\rho' = \frac{\chi a_1 a_2 a_3}{8\pi^3}. \tag{B2}$$

Making use of the fact that

$$f_\alpha = \frac{4\pi R_\alpha^3}{3\chi a_1 a_2 a_3}, \tag{B3}$$

the integral $Z_k$ can be recast into the following form:

$$\begin{aligned} Z_k &= 3C_k \varphi(R_\alpha \xi_c), \\ C_1 &= \frac{1}{4\pi} \int_{|\xi'|=1} \frac{\chi^2 \xi_1'^2 dS}{\chi^2 \xi_1'^2 + \xi_2'^2 + \xi_3'^2}, \\ C_k &= \frac{1}{4\pi} \int_{|\xi'|=1} \frac{\xi_k'^2 dS}{\chi^2 \xi_1'^2 + \xi_2'^2 + \xi_3'^2} \quad \text{with} \quad k = 2,3, \end{aligned} \tag{B4}$$

where $C_k$, $k = 1,2,3$ are integrals over the surface of a unit sphere at the origin and $\varphi$ is the function defined in (29). The evaluation of $C_k$ can be done in spherical coordinate systems $(r,\theta,\phi)$, for example,

$$\begin{aligned} C_1 &= \frac{1}{2} \int_0^\pi \frac{\chi^2 \cos^2\theta \sin\theta d\theta}{\sin^2\theta + \chi^2 \cos^2\theta}, \\ C_2 = C_3 &= \frac{1}{4} \int_0^\pi \frac{\sin^3\theta d\theta}{\sin^2\theta + \chi^2 \cos^2\theta}. \end{aligned} \tag{B5}$$

The evaluation of $C_1, C_2$, and $C_3$ yields finally the closed form solutions (35)–(36).

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